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Transition Vitreuse: Théorie de Champ Moyen

Glass Transition: A Mean-Field Theory

Soutenu le 9 Octobre 2007 devant le jury composé de:

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Abstract

A typical liquid can be cooled down so as to avoid crystallisation resulting in what is called a supercooled liquid. Its static properties are mere continuations of those of a liquid. However several new phenomena emerges in dynamics. The most prominent of them is the tremendous slowing down of the dynamics, referred to as a glass transition. The global relaxation time grows from a picosecond scale at the crystallisation transition up to a macroscopic value for low temperatures. Understanding of the phenomena in the low temperature state of supercooled liquids, in particular the tremendous slowing down, is one of the longstanding open problems of the out of equilibrium statistical physics.

Different scenarios were proposed to explain the observed low-T behaviour of supercooled liquids. In this thesis we follow the one based on the results of generalised spin glass models (mean-field in nature) and the Mode-Coupling Theory (MCT). This scenario reproduces well the initial slow down of the dynamics but predicts a spurious transition to non-ergodic phase for low temperatures. Thus a problem naturally arises: how to enhance the model so as to get a correct behaviour at low temperatures and to eliminate the transition ?

The main aim of the thesis is to construct such a generalisation. The general expectation is that corrections to MCT modify the low-T theory. However the main drawback of the MCT-based scenario is the uncontrolled nature of the approximation lying at the heart of MCT what makes the computation of corrections within the original projector operator formalism an almost unsolvable problem. A promising way to circumvent this problem is a rederivation of MCT within a field theoretical context. Revision of previous attempts reveals their inconsistency caused by the violation of the time-reversal symmetry (TRS) in perturbation series. Resolving this issue results in a correct derivation of MCT within a field theoretical context and yields perturbation series respecting TRS. This provides a direct way to test a structural stability of MCT and, thus, provides an insight on the very important problem of whether the transition cutoff can be understood within some refined approximation or has more fundamental foundations. The result is that MCT is only a mean-field theory of the glass transition, so that perturbative corrections are irrelevant. Within this context we also explore the mapping via a Cole-Hopf transformation between the supercooled liquids and reaction-diffusion systems.

The above mentioned results apply to the supercooled liquids above the transition *i.e.* to equilibrium dynamics. The last chapter deals with their partial generalisation to low temperatures where the system never reaches equilibrium during the observation and the global relaxation time is, effectively, infinite. Analysis turns out to be more complicated in that case since a bunch of new phenomena like ageing of physical properties comes into play.

Abstrait

N'importe quel liquide peut être refroidi au-dessous de sa température de cristallisation sans se cristalliser en devenant un liquide surfondu. Ces propriétés statiques sont les continuations analytiques de celles d'un liquide ordinaire. Par contre la dynamique montre un ralentissement dramatique qu'on appelle la transition vitreuse: le temps de relaxation croit de picoseconds au voisinage de la transition de cristallisation au temps macroscopique pour les basses températures. Compréhension et explication de ce comportement des liquides surfondus à basses températures est l'un des problèmes ouverts de mécanique statistique hors d'équilibre.

Les scénarios divers ont été proposes pour expliquer ce comportement des liquides surfondus. Dans cette thèse nous adoptons celui basé sur l'analyse des modèles de verres de spins généralisés et la Théorie de Couplage de Modes (MCT). Ce scénario reproduit bien le ralentissement initial de la dynamique. Cependant il prédit une transition spurieuse vers une phase non-ergodique pour les basses températures. Ça pose un problème naturel: comment peut-t-on généraliser le modèle afin de reproduire le comportement aux basses températures observe et éliminer la transition ?

Le but principal de la thèse était de construire une telle généralisation. Il est suppose de façon générale que c'est les corrections aux MCT qui modifient la théorie aux basses températures. Cependant le défaut principal de MCT est la nature incontrôlée de l'approximation qui amène à MCT ce qui fait le calcul des corrections à MCT un problème quasiment insoluble. Un méthode promettant pour contourner cette difficulté est la rederivation de MCT dans le contexte d'une théorie de champs. La révision des essaies précédents a relevé leur inconsistance à cause d'une violation de la symétrie par rapport de renversement de temps (TRS) dans les séries perturbatives. La solution de ce problème amène à une rederivation correcte de MCT et donne des séries perturbatives respectantes TRS. Cette construction permet de vérifier directement la stabilité structurelle de MCT ce qui permet de répondre à une question principale de la théorie: Est ce que la transition est coupée par dans le cadre d'une approximation plus astucieuse où il y a des raisons plus fondamentales ? Le réponse est que MCT n'est qu'une théorie de champ moyen et, par conséquence, les corrections pertrubatives sont irrelevantes. Dans le meme contexte on étudie aussi le mapping des liquides surfondus sur les systèmes de reaction-diffusion.

Les résultats donnes au-dessus sont valables au-dessus de la transition vitreuse *i.e* pour les liquides en équilibre. Dans la dernière chapitre on étudie la généralisation de ces résultats aux basses températures ou le système n'atteint jamais l'équilibre pendant l'observation et le temps relaxation est, effectivement, infini. L'analyse est plus complique dans ce cas car des nouveaux phénomène, comme le vieillissement des propriétés physique, apparaissent.

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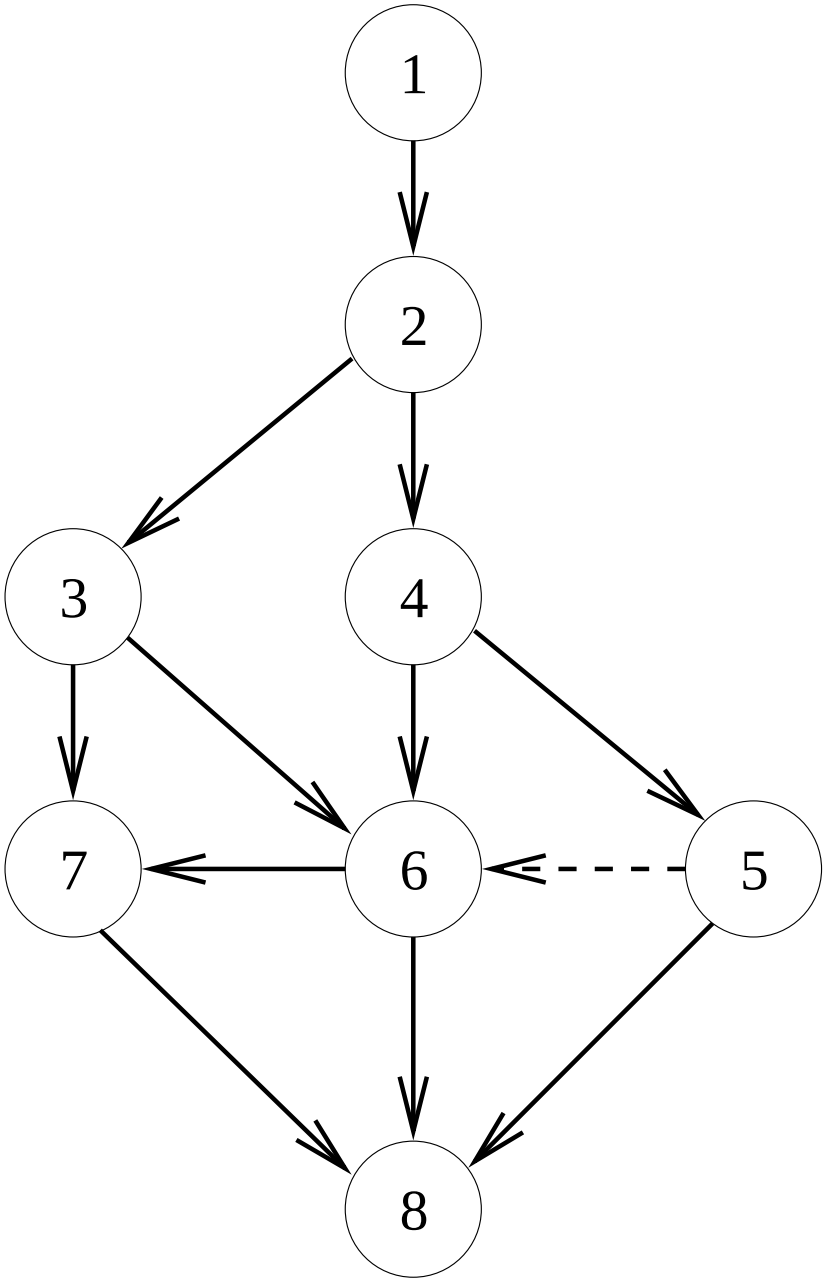


Figure 1: Mutual dependences of the chapters. The dotted line stands for "not yet understood, but supposed".

Introduction

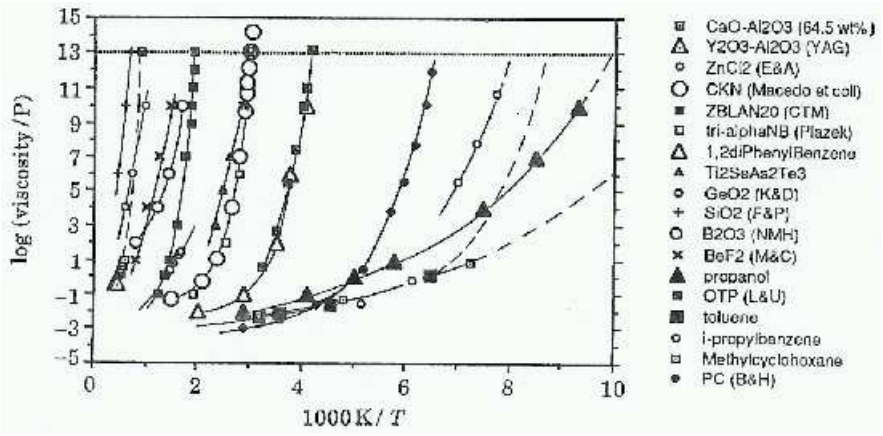
Les phénomènes hors d'équilibre sont largement représentés dans la nature. Ils ont attires beaucoup d'intérêt et ont été beaucoup étudiés ces dernières dizaines d'années. Contrairement à l'équilibre ou les méthodes puissantes ont été développés pendant la vingtième siècle les systèmes hors d'équilibre sont beaucoup moins étudiés. Ils peuvent être divises en deux classes:

- Systèmes qui relaxent vers l'équilibre (par exemple les problèmes de croissances des domaines tombent dans cette classe).
- Systèmes qui sont mis hors d'équilibre par une force externe (par exemple les liquides sous l'étirement ou la croissance des surfaces décrit par l'équation KPZ).

Les *phases vitreuses* tombent dans la première classe car ils relaxent vers l'équilibre. Néanmoins cette relaxation est caractérisée par un temps de relaxation extrêmement grand et par présence de plusieurs échelles de temps dans le système. Ça amène à une situation où le temps de relaxation dépasse le temps d'observation du système. Comme conséquence le système n'atteint jamais l'équilibre et des nouveaux phénomènes se présentent.

Verres: notions de base

Un verre se produit d'un liquide trempe. Donc, considérons un liquide. Le temps de relaxation τ_{REL} est d'ordre 10^{-12} ps pour les températures élevées; la viscosité η est d'ordre 10^{-3} Poise (C'est la viscosité de l'eau en conditions ambiantes.). Lorsque la température baisse une transition de phase se produise: le liquide devient un solide et se cristallise à une température bien précis T_m (température de cristallisation). Cependant n'importe quel liquid peut être refroidis au-dessous de T_m sans se cristalliser. Dans ce cas on parle d'un liquide surfondu c'est-à-dire un état métastable détruit éventuellement par une cristallisation spontanée. En principe il peut être maintenu jusqu'à la limite instable de stabilité, la où l'état devient instable par rapport aux fluctuations microscopiques. La question importante concernant les liquides surfondus est "Est ce que un liquide surfondu présente des propriétés d'un liquide ordinaire, c'est-à-dire ces propriétés se prolongent jusqu'à la limite instable ou il y a des particularités ?" L'analyse des propriétés statiques ne relève pas des particularités: ils

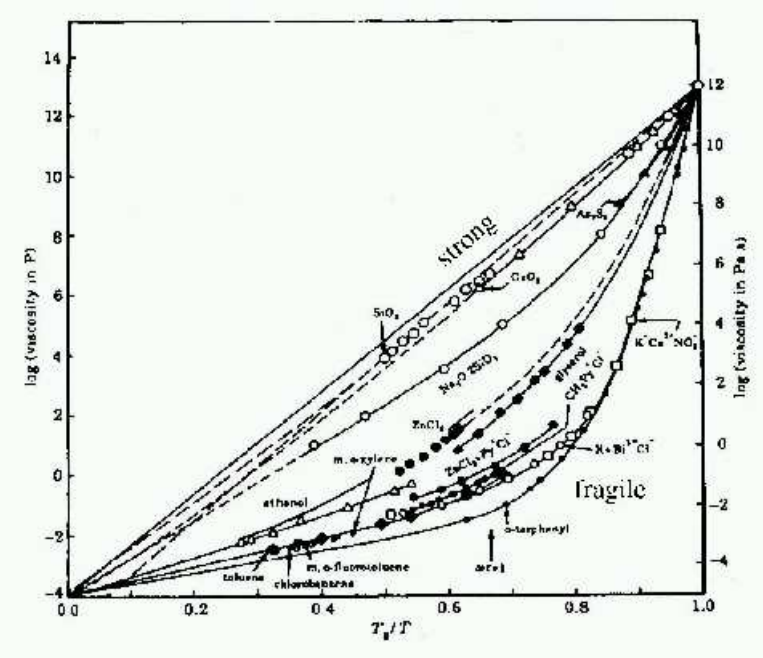


sont les prolongements continues de ceux d'un liquide ordinaire. Néanmoins la situations est complètement diffèrent en cas de propriétés dynamiques, comme la viscosité η, constant de la diffusion D ou le temps de relaxation τ_{REL}. Ils présentent une comportement très sensible aux variations des paramètres de contrôle comme la température. Les données expérimentales et les simulations montrent un ralentissement extrêmement rapide de la relaxation structurelle lorsque la température baisse: τ_{REL} surpasse le temps d'observation τ_{EXP} de façon qu'une inégalité forte $\tau_{REL} \gg \tau_{EXP}$ s'établit. Ça gèle la relaxation structurelle dans le liquide surfondu aux basse températures. Le dépendance en température de la viscosité η dans divers liquides surfondus est présentée sur Fig. [3]. Le ralentissement de la relaxation structurelle dans liquides surfondus porte le nom *transition vitreuse*. Le liquide surfondu devient un solide amorphe *i.e. un verre*.

Les quantités les plus simples associées avec la relaxation structurelle sont le temps de relaxation τ_{REL} et la viscosité η. Cependant, valeurs de η varient fortement d'un matériel à l'autre. Laughlin and Uhlmann [4] ont propose une échelle réduite des température afin pouvoir comparer des liquides différents. Ils ont introduit une température T_g telle que $\eta(T_g) = 10^3$ Poise. Leur idée était de tracer la viscosité en echelle reduite de temperature *i.e.* en fonction de T_g/T. Une telle représentation est appelée "Angell plot" (voir Fig.). On en déduit la dépendance relativement simple dite "activée" de η:

$$\eta(T) = \eta_0 \exp\left(\frac{E(T)}{T}\right).$$

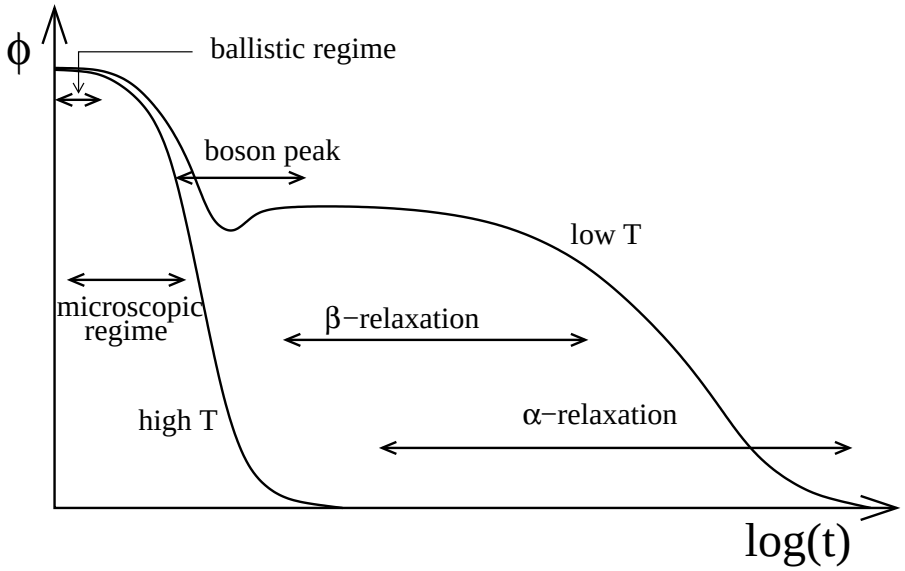
Certains liquides sont bien approximés par une lois d'Arrhenius simple tant que les autres montrent une dépendance plus compliquée avec $E(T)$ étant une fonction de T. Cette différence inspirait Angell [5] à introduire une classification des liquides surfondus et les divisés en deux classes: "forte" et "fragile". Comme E a un dimension d'énergie on peut imaginer naïvement que c'est une barrière énergétique à franchir par le système pour pouvoir relaxer. Alors le valeur



constant de E pour les verres dites "fortes" peut être associer avec la coupure d'une lien chimique. Pour les verres "fragiles" $E(T)$ croit avec la décroissance de T: la liquide sent des barrières des plus en plus élevés lorsque la température diminue. Ça fait penser à la relaxation coopérative. Malheureusement il manque encore la compréhension de la différence précise entre ces deux types de verres.

Plus d'information est fournie par l'analyse des fonctions à deux points qui possèdent une courbe de relaxation bien particulière présentée sur la Fig. . Pour les températures élevés on peut distinguer trois régimes différentes dans la relaxation: temps courts/balistique ou la relaxation est déterminée par le mouvement libre des particules; intermédiaire ou l'interaction entre particules gouverne la relaxation; la relaxation exponentielle finale *i.e.* relaxation de Debye [6]. Une courbe de relaxation simple exponentielle présentée pour les températures élevées se complique lorsque la température baisse. En baissant la température plus de régimes apparaissent en formant l'image dite "à deux pas":

- Temps courts. C'est la même régime balistique.
- Relaxation β. Pour les températures suffisamment basses un plateau apparaisse dans la courbe de relaxation. Ce régime comprend tous les temps où la fonction reste proche de plateau. Il peut propager sur plusieurs ordre de grandeur en temps (remarque l'échelle logarithmique sur Fig.).
- Relaxation α. Dans ce régime le corrélateur quitte le plateau et décroît à zéro. L'échelle de ce régime est τ_{REL}.



Ce motif de relaxation est souvent interprété dans le contexte de l'effet de cage. Le déplacement libre des particules aux stades initiales est supprime par leur voisins qui sont bloques eux-mêmes par leur environnement. Cette interprétation explique l'apparition du plateau dans le régime β. Les particules restent bloquer par leur environnement très long temps avant pouvoir se déplacer et oublier la configuration initiale. Ces actions représentent le régime α. Ce dernier régime présente une autre spécialité de la dynamique lente: la relaxation des fonction des corrélation est bien approximee par l'exponentielle simple pour les températures élevées et les temps longues. Au contraire la décroissance des correlateurs dans le régime α n'est plus une simple exponentielle. Ça forme exacte n'est pas connue, cependant la fonction de Kohlrausch-Williams-Watts (KWW) [7, 8] ou l'exponentielle étirée est souvent une bonne approximation:

$$\Phi(t) = \Phi_0 \exp\left[-(t/\tau_{REL})^{\beta}\right]$$

ou β est le paramètre KWW; $\beta < 1$ typiquement. *A priori* tous les paramètres de cette approximation dépendent de température des observables entrant dans le corrélateur. Néanmoins on trouve souvent qu'ils sont universels dans un certain fenêtre de temps et ne dépendent pas de température. Ce fait est souvent connue comme *le principe de superposition de temps et de température*. L'origine de cette décroissance en exponentielle étirée dans les liquides surfondus n'est pas bien expliquée en ce moment. Les deux scénarios possible sont discutées dans la littérature [6]:

- *Hétérogène.* La dynamique devient hétérogène et le système se décompose en régions "rapides" et "lentes" qui évoluent à des vitesses différentes.

Tous ces régions contribuent à la relaxation globale qui devient non-exponentielle suite à des différences entre les régions. Ce phénomène de décomposition spontanée porte le nom de *hétérogénéités dynamiques*.

- *Homogène.* La relaxation est compliquée et non-exponentielle suite à la dynamique complique de système.

Il est pas claire laquelle des deux possibilités est réalisée dans la nature. Il semble que tous les deux contribuent dans la dynamique de relaxation des liquides surfondus. L'analyse de ces scénarios avait inspirée l'étude des fonctions à plusieurs points dynamique telle que les fonctions à quatre points dynamique [9, 10, 11] qui permettent de tester les hétérogénéités dynamiques et étudier la coopérativité derrière la transition vitreuse. Cette coopérativité est implique par les hétérogénéités: la dynamique des particules à l'intérieur d'une domaine est forcement corrélée; la taille typique de domaines fixe la longueur de corrélation.

Le ralentissement rapide de la dynamique pose la question sur la mécanisme responsable de ce ralentissement. La coopérativité et le fait que l'arrêt structurelle arrive pour des variations de la température réduites suggèrent une transition de phases cachée derrière ce phénomène. Un ralentissement de la dynamique est observe au voisinage d'une transition de second dégrée: on observe une croissance en lois de puissance de temps de relaxation. Cependant les quantités statiques ne montrent aucune singularité ce qui élimine la possibilité d'une transition statique conventionnelle. La dépendance en température des observables dynamiques peut être approximée dans certain cas par la lois de Vogel-Fulcher(-Tamman) [12, 13, 14]:

$$\eta(T) = \eta_0 \exp\left[\frac{A}{T - T_0}\right],$$

où T_0 est le paramètre ajustable qu'on appelle "la température de Vogel" [6]. Cette une dépendance super-Arrhenius: $E(T) \sim T/(T - T_0)$. Néanmoins cette lois est une approximation. Les valeurs intermédiaires de la viscosité peuvent aussi être approximées par une lois de puissance:

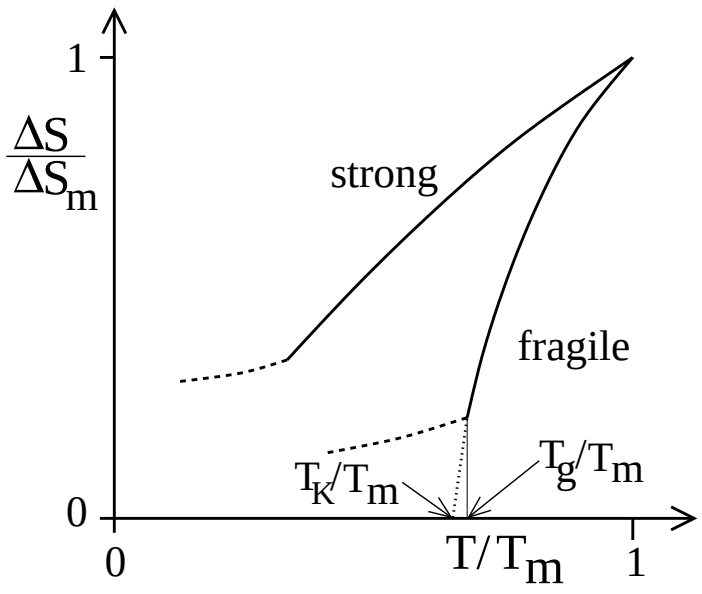
$$\eta(T) \sim \eta_0 (T - T_c)^{-\gamma},$$

où de façon générale $T_c < T_0$ et $T_0/T_c \approx 0.8$. Des approximations similaires existent pour τ_{REL}. Les deux approximation prédisent la divergence de la viscosité à une température finie. Deux hypothèses différentes ont été proposées pour expliquer la divergence:

- La transition statique non-conventionnelle à $T = T_K$.
- La transition dynamique à $T = T_d$.

La première hypothèse est basée sur l'extrapolation de l'entropie S du liquide surfondu aux basses températures. L'entropie est calculée par l'intégration

Figure 2: Un dessin schématique de la dépendance en température de la différence normalisée entre les entropies de liquide et de cristal. Reproduit de [6].



thermale [6] de chaleur spécifique:

$$S_\alpha(T_m) = S_\alpha(T) + \int_T^{T_m} \frac{dT}{T} C_\alpha \qquad \alpha \in \{liquide, cristal\}.$$

Pour un liquide surfondu on suggère que l'entropie S est composée de deux contributions:

$$S = S_{VIBR} + S_{CONF}$$

où S_{VIBR} est la contribution des modes de vibration dans un état métastable. Le deuxième terme, S_{CONF}, ou l'entropie configurationnelle vient de l'existence de plusieurs état métastables. L'hypothèse complémentaire impose que S_{VIBR} coïncide avec l'entropie vibrationnelle de cristal. Alors $S_{CONF} \approx S - S_{CRISTAL}$.

S_{CONF} dd́écroît avec la décroissance de température (voir Fig. 2). L'extrapolation de S_{CONF} pour les verres fragiles aux basses températures (voire la ligne en pointes sur Fig. 2) s'annule à une température T_K (température de Kauzmann). Les entropies de cristal et de liquide deviennent égales à $T = T_K$ (*paradoxe de Kauzmann*, (1948)). Au-dessous de T_K l'entropie du liquide est plus petite que l'entropie du cristal ce qui pose un certain nombre de problèmes [15]. La solution de paradoxe proposée par Kauzmann reposait sur la cristallisation spontanée. Cependant, la nucléation, qui est responsable de la cristallisation selon

Kauzmann, est hautement supprimée aux basses températures à cause de l'arrêt structurelle [6, 15]. Donc, la cristallisation ne peut pas être une solution universelle. L'autre solution de paradoxe a été proposée par Angell et Tucker [16]. Ils ont suppose que liquide n'existait plus au-dessous de T_K et que une transition vers une nouvelle phase, dite le *verre parfait* [16], arrivait à T_K. A nouveau ce n'était pas la solution universelle. D'abord la construction entière repose sur l'extrapolation de l'entropie aux basses températures. Dans ce domaine le temps de relaxation montre une croissance gigantesque et le liquide tombe hors d'équilibre bien avant on arrive à T_K. Aussi $S_{CONF} = 0$ implique un nombre sous-exponentiel d'état dans la phase de verre parfait. Un argument simple construit par Stillinger pour les systèmes en l'interaction à courte porte montre que l'existence d'un seul état vitreux implique immédiatement l'existence d'un nombre exponentiel d'états [17]. Ça semble donner une contradiction, cependant l'argument de Stillinger n'était pas une preuve rigoureuse. Selon Stillinger les états metastables sont simplement les minimas de l'énergie potentielle, la définition, qui semble être beaucoup trop restrictive [18, 19].

Finalement, les arguments pro et contra de l'existence d'un transition statique de nouveau type à T_K ne sont pas décisifs et l'existence de la transition est une question ouverte. Il est néanmoins remarquable que les valeurs numériques de la température de Vogel, introduite quelques paragraphes plus haut, sont souvent remarquablement proche de ceux de T_K.

La deuxième hypothèse suggère une transition purement dynamique vers une phase non-ergodique à une température $T = T_d > T_K$. Lorsque $T \to T_d$ l'espace de phases est de plus en plus dominé par les états marginalement stables. Il prend de plus en plus de temps pour le système de les explorer tous. Au-dessous de T_d l'espace de phases est domine par le vrai minimas avec des barrières infinis entre eux. L'espace de phases, donc, se décompose en domaines disjointes. Ce scénario de la transition est base sur l'étude de certains modèles de champ moyen de verres de spin généralisés. Le scénario est aussi supporte par des résultats de la théorie de Couplage de Modes (Mode-Coupling theory) [20, 21] présentée dans la Chapitre 2. Elle reproduit certaines propriétés de la dynamique des liquides surfondus, comme la relaxation à deux pas. D'autre part aucune transition dynamique n'est observée expérimentalement. L'explication standard de ce défaut de la théorie fait appel à l'image de l'espace de phase qui est domine par des états différents en fonction de la température. Comme on l'avait discuter quelques lignes plus haut, pour les températures suffisamment basses l'espace de phases est domine par les minimas sépares par des barrières infinies. Cependant pour les systèmes en dimension finis et avec des interaction de portée finie ces barrières ne peuvent pas l'être et ils sont toujours finies. Alors la transition est détruite par les évènements activées correspondantes à des transitions entre les minimas différents. Cette explication suggère, donc, que la transition vitreuse marque le passage de la relaxation de type flot à la relaxation de type activée [22, 23, 24, 27].

Les deux approches aussi différentes ont une image en commune: l'image d'un système bloque dans une configuration amorphe. Cette dernière agit comme un désordre induit dans le système, dite le *désordre auto-induit* [25, 26].

Ce phénomène relie les verres structuraux aux verres de spin où le désordre gèle est implicite.

La discussion précédente portait sur les propriétés d'équilibre des liquides surfondus. Le temps de relaxation τ_{REL} crois si extrêmement vite avec la dd́écroissance de la température que il n'est pas possible d'équilibrer le liquide à n'importe quelle basse température. Ça amène vite à une situation où τ_{REL} dépasse le temps d'observation et le système n'arrive plus à s'équilibrer pendant l'observation: il tombe hors d'équilibre. L'état hors d'équilibre du système fait apparaître des nouveaux phénomènes physique dont le vieillissement. L'impossibilité d'équilibrer le système à une température arbitrairement basse dans un temps accessible expérimentalement est l'obstacle principale pour la preuve expérimentale de la température de Kauzmann T_K. Les liquides tombent hors d'équilibre bien avant le voisinage de T_K.

Pour conclure, voici quelques remarques:

- Les basses températures sont définies par rapport à la température de cristallisation T_m qui, elle-même, peut être bien élevé pour un liquide particulier ($T_m = 1723^o$C pour le silice).
- Le temps de relaxation peut prendre des valeurs géologiques dans les verres pour les températures suffisamment basses ce qui permet de parler d'un verre ou d'un état vitreux dans le sense pratique.

Le plot

La thèse se concentre sur l'amélioration la théorie de couplage de modes pour les liquides surfondus. Bien qu'elle prédise la transition spurieuse, elle reproduit certains aspects de la dynamique lente comme la courbe de relaxation à deux pas. Ce succès partiel de MCT indique que on est sur la bonne piste et il motive pour l'amélioration de MCT qui prend en compte les évènements actives et la coupure de la transition. Dans la suite on se concentre sur 3 problèmes principales:

- **La mécanisme de coupure de la transition**. La rôle centrale des évènements activées discutée plus haut n'était pas qu'une hypothèse à prouver. Une mécanisme différente a été proposée par Das et Mazenko [28] avec le couplage aux courants étant responsable de coupure de la transition dynamique.
- **La stabilité de la MCT par rapport aux corrections**. Ce problème est relie au problème precedent: il n'est pas claire si la transition est coupée par les corrections à l'approximation CT ou il y a des raisons plus profondes pour son absence.
- **La régime des basses températures**. La version originale de MCT ne peut pas être étendu au-dessous de la transition. Une rederivation complète est nécessaire pour un tel extension.

La difficulté additionnelle est l'absence des méthodes directes pour calculer les corrections aux MCT dans le cadre de la formalisme d'opérateur de projection qui est à la coeur de la dérivation de MCT. Par conséquence, il est extrêmement difficile de tirer les conclusions sur la stabilité structurelle ou sur la mécanisme de la coupure. L'extension de MCT aux basses températures est possibles [29] mais il loin d'être trivial. La contrôle sur la symétrie par rapport aux renversement du temps (TRS) est d'un grand importance en cette régime [30, 31]. Cependant, dans MCT originelle TRS est imposée sans la prouver.

La résolution de ces problèmes est important pour la construction d'un scénario consistant de la transition vitreuse. La théorie de champs propose des solutions naturelles aux problèmes techniques mentionnes plus haut:

- Il existe toujours une méthode bien défini pour calculer les corrections à n'importe quelle approximation faite dans la théorie de champs.
- Les symétries physiques, y compris TRS, sont facile à contrôler dans une théorie de champs car ils correspondent à une certaine transformation de champs.
- Théorie de champs est bien définies pour toutes les valeurs de la température.

La contenue des chapitres qui suivent représente l'analyse détaillé de ces points.

Chapter 1

Introduction

Out of equilibrium situations are widely present in nature. They have attracted a lot of attention and have been widely studied in the last decades. Contrary to equilibrium systems where reliable methods were developed in the last century, non-equilibrium systems are much less understood. Roughly, the out of equilibrium systems can be divided into two main classes:

- Systems relaxing towards equilibrium. A well-known example is the domain-growth problem [1].
- Systems driven out of equilibrium by external force. An example is a liquid under shear or the surface growth modelled by a KPZ equation [2].

Glassy phases fall into the first class *i.e.* they relax towards equilibrium. However this relaxation is characterised by extremely large relaxation times and the presence of many timescales in the system. This leads to a situation when the relaxation time exceeds the observation time so that the system does not equilibrate during the experiment and a variety of new phenomena appears.

1.1 Basic facts about glasses

Let's consider a liquid. For high enough temperatures the relaxation time τ_{REL} is of order 10^{-12} ps(picosecond) and viscosity η is of order 10^{-2} Poise (viscosity of the water in ambient conditions). As the temperature is lowered the liquid undergoes a phase transition: it crystallises below a well-defined temperature T_m. However any liquid can be cooled below its melting temperature T_m avoiding crystallisation. It becomes a supercooled liquid *i.e* a metastable state unstable with respect to crystallisation. Such a state can be maintained in principle down to the unstable limit of stability where it becomes unstable with respect to any fluctuations. One might wonder if something particular happens as the liquid is cooled below the melting point; or if the liquid characteristics continue smoothly down to the unstable limit of stability. Analysis of static

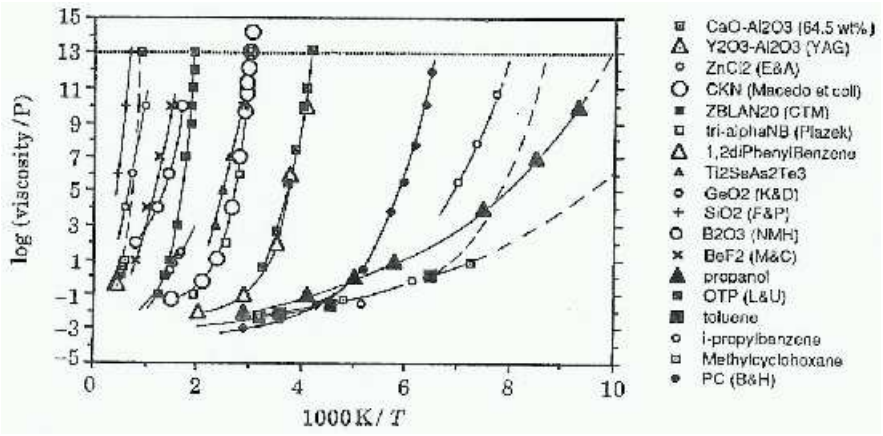


Figure 1.1: Temperature dependence of viscosity in several glass-forming liquids. Reproduced from [3].

properties reveals no singularities: they extrapolate smoothly below T_m. However the situation is completely different for dynamic quantities, like viscosity η, diffusion constant D or relaxation time τ_{REL}: they show a very pronounced T-dependence - extremely large variations for only a mild variations of control parameters *i.e.* temperature. Experiments and numerical simulations reveal an extremely fast slowing down of the structural relaxation as the temperature is lowered: τ_{REL} exceeds the observation time τ_{EXP} so that a strong inequality $\tau_{REL} \gg \tau_{EXP}$ holds. This leads to a structural arrest in supercooled liquids at low temperatures. Temperature dependence of η in various glass-formers is presented on Fig. 1.1 [3]. This structural arrest in supercooled liquids for low temperatures is referred to as a *glass transition*; a supercooled liquid becomes an amorphous solid which is referred to as *glass*.

The simplest quantitative measures of the slowing down are the structural relaxation time τ_{REL} and the viscosity η. However as one can see on Fig. 1.1, values of η vary widely due to different characteristics of materials. Laughlin and Uhlmann [4] proposed a reduced temperature scale in order to compare different glass-formers: they suggested to define a temperature T_g as the temperature where viscosity has the value of 10^13 Poise (which is somewhat arbitrary) and to plot the viscosity as a function of T_g/T. Such a representation ("Angell plot") is shown on Fig. 1.2. This plot shows a relatively simple activated-like temperature dependence of viscosity:

$$\eta(T) = \eta_0 \exp\left(\frac{E(T)}{T}\right).$$

For certain liquids η is fitted well by a simple Arrhenius law while for others the dependence is more complex and E depends on temperature. This inspired Angell [5] to classify supercooled liquids into "strong" and "fragile" respectively

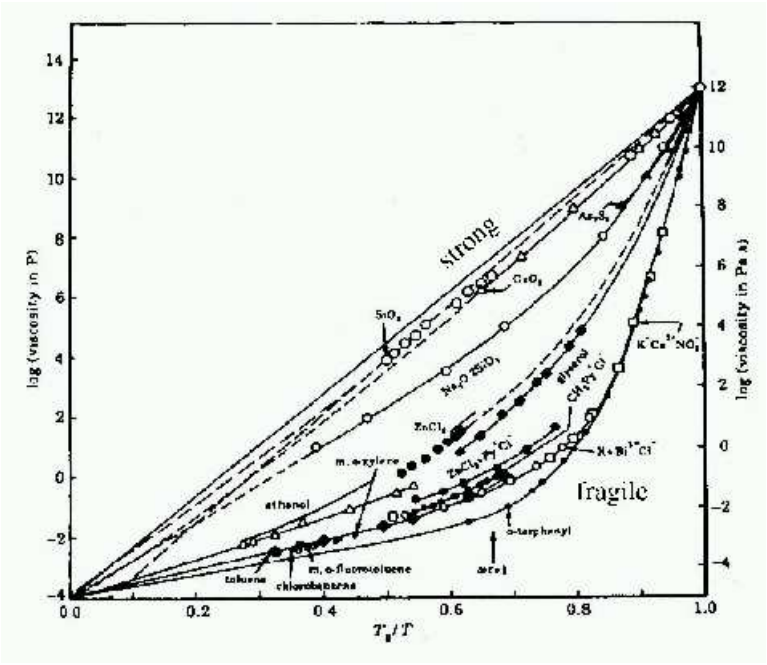


Figure 1.2: Temperature dependence of viscosity in several glass-forming liquids. Reproduced from [3].

to distinguish these two types of behaviour. The parameter E has a dimension of energy. Naively one may think that E is an energy barrier system has to overcome in order to relax. Then the constant value of E for "strong" glasses can be interpreted as an energy required to break a chemical bond. For "fragile" glass formers $E(T)$ increases as T is lowered, that is, the system "sees" higher barriers for lower temperatures: this suggests that relaxation becomes cooperative. However the exact understanding of the difference between "strong" and "fragile" behaviours is still missing.

More information is provided by the analysis of two-point quantities which show a particular two-step relaxation pattern like the one presented on Fig. 1.3. A simple relaxation pattern for high temperatures changes to a more complex pattern as the temperature is lowered. In high-temperature regime one can define three different regimes in the relaxation pattern: short time or ballistic where the relaxation is ruled by a free motion of the particles; intermediate which is governed by an interaction between particles and a final exponential decay *i.e* a Debye relaxation [6]. As the temperature is lowered more regimes emerges:

- Short times. This is the same ballistic regime of the high-T region.
- β-relaxation. For low temperatures correlator shows a plateau; β-relaxation

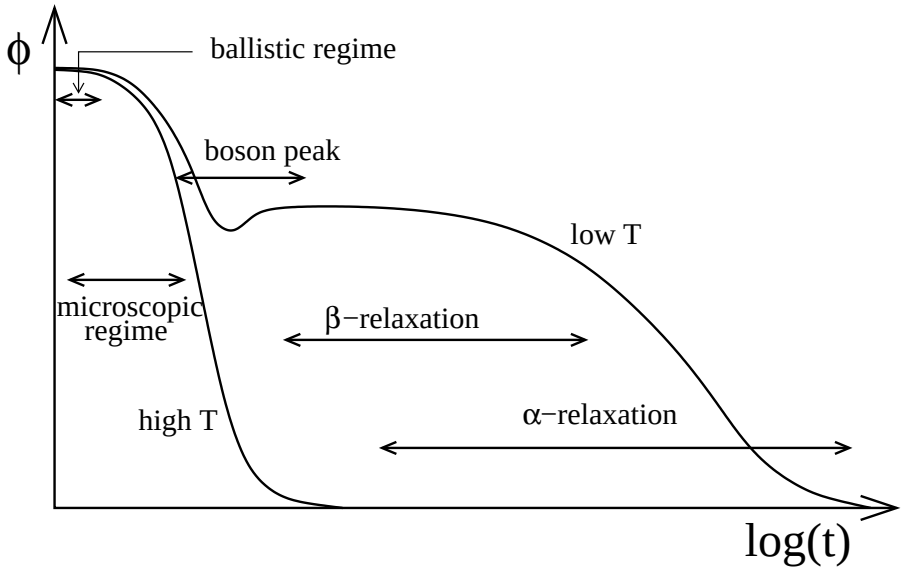


Figure 1.3: Schematic plot of the two-step relaxation of the correlators. Two curves describe the high and the low temperature relaxation. Reproduced from [6].

refers to a time window when the value of the correlator stays close to the plateau. This regime propagates over many decades in time, note the logarithmic scale on Fig. 1.3.

- α-relaxation. This regime refers to a time window when correlator leaves the plateau and decays to zero. This happens on the timescales of order τ_{REL} (global relaxation time).

The standard interpretation of the two-step relaxation pattern is provided by the so called "cage effect". After the initial free motion particles get blocked by their neighbours as the temperature is lowered, the neighbours are themselves blocked by the environment. This explains the appearance of the plateau in β regime. Only after a long time particles succeed to "escape" from the cage and information on initial configuration gets lost. This last regime corresponds to α-relaxation. This time regime shows another peculiarity of slow dynamics. Decay of correlation functions for long times is well approximated by exponential for high temperatures but the decay in α regime follows a different law for low temperatures. Its exact form is unknown but a good fit is provided by the Kohlrausch-Williams-Watts function (KWW) [7, 8] (stretched exponential):

$$\Phi(\tau) = \Phi_0 \exp\left[-(\tau/\tau_{REL})^{\beta}\right]$$

where β is the KWW exponent. Although all parameters in this fit should

depend on the temperature and observable it is often found that in a certain time window this law has a universal form: parameters do not depend on the temperature. This is often referred to as *time-temperature superposition principle*. Exponent β is usually lesser than 1. The physical origin of the stretched exponential decay in supercooled liquids is unclear. Two extreme scenarios are discussed in the literature [6]:

- *Heterogeneous*. Dynamics becomes heterogeneous and system splits into "fast" and "slow" regions evolving at different rates. Consequently the overall relaxation which results from contributions of many regions with different relaxation times becomes a stretched exponential. Such spontaneous decomposition is referred to as *dynamic heterogeneities*.

- *Homogeneous*. The system shows a non-exponential relaxation due to complicated dynamics.

It is unclear which scenario is realised and it seems that both the mosaic structure with "fast" and "slow" regions and complicated relaxation takes place in supercooled liquids. This issue inspired analysis of multi-point functions like four-point functions [9, 10, 11] which allow to probe dynamic heterogeneities and to look for cooperativity underlying the glass transition. Cooperativity is implied by the heterogeneities: dynamics of particles inside a region is correlated and typical size of the regions defines a correlation length.

Tremendous slowing down of the dynamics motivates a question: what is the mechanism responsible for the structural arrest ? The presence of cooperativity in dynamics together with the above remark that the slowing down happens for mild variations of temperature suggests an interpretation of the glass transition as a phase transition. It is well-known that dynamics slows down close to the second order phase transition: exponential relaxation changes to a power law. However as we pointed out static quantities show no singularities below T_m. This rules out a conventional static transition. Some insight is provided by the analysis of temperature dependence of dynamic quantities. For fragile glasses temperature dependence of the viscosity is well fitted in some cases by the Vogel-Fulcher(-Tamman) law [12, 13, 14]:

$$\eta(T) = \eta_0 \exp\left[\frac{A}{T - T_0}\right]$$

where T_0 is referred to as "Vogel temperature" [6]. This is a stronger than Arrhenius dependence with $E(T) \sim T/(T - T_0)$. However this law is only a fit. For intermediate values of viscosity a different, power-law fit is also compatible with data:

$$\eta(T) \approx \eta_0 (T - T_c)^{-\gamma}$$

and $T_c < T_0$ with $T_0/T_c \approx 0.8$. A similar fits exist for τ_{REL} [15, 6]. Both fits predict a divergence of viscosity at a finite value of the temperature. There are two different hypothesis explaining this issue:

- There is an unconventional static transition at $T = T_K$.
- There is a dynamic transition at $T = T_d > T_K$.

The first hypothesis is based on extrapolation of entropy $\mathcal{S}$ of supercooled liquids down to low temperatures. The entropy is computed by means of thermal integration [6] from the specific heat:

$$S_\alpha(T_m) = S_\alpha(T) + \int_T^{T_m} \frac{dT}{T} C_\alpha \qquad \alpha \in \{liquid, crystal\}$$

. For supercooled liquid one assumes that the entropy $\mathcal{S}$ is composed by two contributions:

$$\mathcal{S} = \mathcal{S}_{VIBR} + \mathcal{S}_{CONF}$$

where $\mathcal{S}_{VIBR}$ is the entropy related to vibrational motion inside a metastable state. The second term $\mathcal{S}_{CONF}$ is a *configurational entropy* which accounts for existence of multiple metastable states. It is further assumed that $\mathcal{S}_{VIBR}$ is equal to the entropy of a crystal, so that $\mathcal{S}_{CONF} = \mathcal{S} - \mathcal{S}_{CRYSTAL}$.

Thus defined $\mathcal{S}_{CONF}$ decays as the temperature decreases as one can see from Fig. 1.4; a reasonable extrapolation (see dotted lines on Fig. 1.4) for fragile glasses vanish at some value $T = T_K$ which is referred to as Kauzmann temperature. The entropies of a supercooled liquid and a crystal become equal at $T = T_K$ (*Kauzmann paradox*, (1948)). The entropy of a supercooled liquid becomes smaller that that of a crystal below T_K leading to problems [15]. The original solution to the paradox proposed by Kauzmann assumed that spontaneous crystallisation intervened below T_K. However nucleation required for crystallisation is highly suppressed for low temperatures because of the structural arrest [15, 6] and thus cannot be a universal solution. A different solution to the paradox was proposed by Angell and Tucker [16] who suggested that liquid cannot exist below T_K and a phase transition to a new phase referred to as *idealised glass phase* [16] takes place at T_K. However there are several problems related to this solution. First, the whole construction relies entirely on the extrapolation of the entropy to low temperatures. However in this region relaxation times increase fast and liquid falls out of equilibrium so that a direct test of this hypothesis is impossible. Second, $\mathcal{S}_{CONF} = 0$ implies a non-exponential number of states for an ideal glass phase. Stillinger has put forward a simple argument for systems with short ranged interactions that existence of a single glass state implies immediately existence of exponentially many of them [17]. However this is not a rigorous proof. Stillinger defined metastable states as minima of potential energy which seems to be too restrictive [18, 19]. Finally both the arguments in favour of and against the existence of a static transition at T_K are inconclusive and there is still no clear answer. It is worth noting though that the Vogel temperature T_0 is often found to be remarkably close to the Kauzmann temperature T_K.

The second hypothesis assumes a dynamic transition to a non-ergodic phase at $T = T_d$. As the temperature approaches T_d an increasing number of marginally

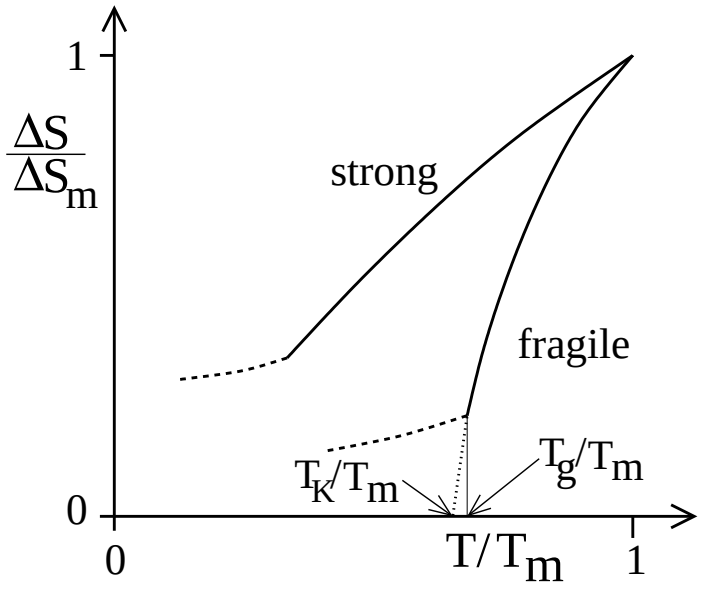


Figure 1.4: Schematic plot of the temperature dependence of the normalised entropy difference between the liquid and crystalline states. Solid and dashed lines represent the liquid state and the glass respectively. Two branches correspond to strong and fragile glasses. Dotted line represents a reasonable extrapolation of the entropy difference between the liquid and the crystal. Reproduced from [6].

stable states appear in phase space and it takes a long time to explore them all. Phase space splits into disconnected domains corresponding the states for $T < T_d$. The support to this hypothesis is provided by results of Mode-Coupling Theory [20, 21] presented in Chapter 2. It captures some of the properties listed above, like the two-step relaxation pattern of the two-point correlators. Unfortunately no transition was observed experimentally. This usual interpretation of the absence of the transition consists in pointing out that the transition requires appearance of states separated by infinite energy barriers for $T < T_d$. However such states does not exist in any finite dimensional system: energy barriers between any two states are always finite and therefore the transition is destroyed by an activated processes which provide an extra channel of relaxation. This lead to hypothesis that the glass transition marks a crossover from flow-like relaxation mechanism to an activated relaxation [22, 23, 27, 24].

Both approaches however share the same idea of a system stuck in some amorphous configuration. The latter acts as a dynamically induced disorder in the system *i.e. self-induced disorder* [25, 26] and links the glass transition problem with a much studied theory of *disordered systems* with explicit quenched disorder.

Previous discussion addressed the properties of the equilibrium state. However, the relaxation time τ_{REL} increases so fast with decreasing T that it is not possible to equilibrate the system at arbitrarily low temperatures; at some moment relaxation time exceeds the observation time τ_{EXP} and the system never relaxes towards equilibrium. Within this regime a variety of non-equilibrium phenomena is observed like ageing. The fall out of equilibrium is also the main reason which makes the direct experimental evidence for Kauzmann temperature problematic. Relaxation times in glasses increase extremely fast and exceed times accessible experimentally well before the vicinity of T_K.

It is worth noting finally that:

- The term *low temperatures* is defined with respect to the melting temperature T_m which can be quite elevated for a given material ($T_m = 1723^oC$ for silica SiO_2).
- Relaxation times in glasses for low temperatures can reach geological values, which allows one to speak of a glass as a state of matter in a practical sense.

1.2 The outline

In this work we focus on the analysis of the approach provided by the Mode-Coupling Theory. The central problem of MCT is the absence of the transition. Nevertheless as we stated and as we will see below it captures some aspects of the slow dynamics in supercooled liquids. This motivates the research for an improvement of MCT that accounts for the absence of the transition. Such analysis suggests consideration of three related problems:

- *Mechanism responsible for the cutoff of the transition.* The activated like relaxation mentioned above is only an assumption and should be justified. A different mechanism for the cutoff was proposed by Das and Mazenko [28]. They stated that it is the coupling to currents which was responsible for the cutoff. However their statement was not confirmed numerically.

- *Stability of MCT with respect to corrections.* This problem is related with the previous one. It is unclear whether the transition is cutoff by systematic corrections to MCT *i.e.* whether refining the approximation leading to MCT may cutoff the transition or not.

- *Low temperature regime.* As we will see the original derivation of MCT cannot be extended directly to low temperatures and requires a complete rederivation.

An additional difficulty is that the projection operator formalism used to derive MCT provides no way to compute systematic corrections around MCT. Therefore it is impossible neither to test the stability with respect to corrections nor to conclude anything about the cutoff mechanism. Extension to low temperatures is possible but it is far from trivial [29] and requires extra approximations with respect to the ones already done in the original MCT. However this is also a problem within MCT. As we shall see TRS is assumed to hold within MCT but it is not rigorously proven.

Solution to these problems is important because it allows one to construct a consistent scenario for the glass transition. The central statement of the thesis is that the natural context for analysis of these issues is a field theory:

- Any approximation within a field theory can be improved systematically.

- Physical symmetries are easy to control since they correspond to invariance under certain fields transformations.

- Field theory is valid for any value of the temperature.

The content of the following chapters represent a step by step analysis of the above three problems in the order they were presented.

Chapter 2

Mode-Coupling theory

The Mode-Coupling Theory (MCT) introduced in this chapter represents a microscopic dynamical approach to the glass transition. Derivation of MCT relies on the projection operator formalism [32] which exploits the idea of reduced description: separate the variables into "slow" and "fast" and integrate out the latter. This results into an effective theory in terms of "slow" variables only. However this formalism provides only a rearrangement of the initial theory and has the same complexity; approximations are required in order to obtain results.

The projection operator formalism was first applied to high temperature liquids [33] with density, currents and energy as a "slow" variables. This led to coupled integral equations for various correlators. These equations are not closed since all the complexity is encoded into memory kernels which are the key objects in the formalism and which rule the evolution. For high temperature liquids anzats based on the knowledge of short time and long time hydrodynamic behaviour of a system were used to approximate the kernels [33].

An important breakthrough was made by Götze [21] and Leutheusser [20] who applied the formalism to the case of supercooled liquids. They considered a simpler models with only one "slow" variable - density. They proposed the Mode-Coupling Factorisation [20, 21] (MCF)[1]: a rather successful first principle approximation of the memory kernel within the context of the projection operator formalism. The resulting theory, known as Mode-Coupling theory, has captured some aspects of the glass transition presented in the introduction. Models based on a greater set of "slow" variables: density + currents, were also introduced later [34, 28].

This chapter is dedicated to the detailed introduction of MCT: in Section 2.1 we introduce the projection operator formalism for a generic case; the derivation of MCT is considered in Section 2.2. In section 2.3 we present the results derived in the context of MCT for supercooled liquids. Section 2.4 is dedicated to the schematic version of MCT with no wavevector dependence; section 2.5

[1] We use a different name for what is usually known as the Mode-Coupling Approximation in MCT to avoid confusion with the Mode-Coupling Approximation of the critical dynamics which is presented later in the text.

is dedicated to unveiling of a diverging correlation length hidden within MCT. Finally Sections 2.6 and 2.7 deal with numerical/experimental evidences for MCT and criticism of MCT, and an extension of MCT which accounts for more slow variables respectively.

2.1 Projection operator formalism

Let's consider a classical system with a Hamiltonian $\mathcal{H}(X)$ where X is a phase space variable (which is a set of all positions and momenta $X = (\mathbf{r}, \mathbf{p})$ for a system of point interacting particles). Then the time evolution of any phase space function $g(t, X)$ is governed by a Liouville equation:

$$\frac{\partial g(t,X)}{\partial t} = i\mathcal{L}g(t,X) \tag{2.1}$$

where $\mathcal{L}g = \{\mathcal{H}, g\}$ and $\{,\}$ are the Poisson brackets.

The space of all functions $g(X)$ is transformed into a vector space by introduction of an inner product:

$$(f,g) \equiv \langle f \cdot g \rangle = \int dX P(X) f(X) g^*(X) \tag{2.2}$$

where star $*$ denotes a complex conjugate. The actual choice of the kernel $P(X)$ depends on many circumstances. One particular choice that we adopt henceforth is the canonical equilibrium distribution: $P(X) = \exp(-\beta H(X))/Z$ with the inverse temperature β although different definitions are possible [35]. This results into an infinite dimensional Hilbert space of phase space functions g.

The full equation (2.1) is extremely complicated for direct analysis and we are interested in a reduced description of the dynamics. For this purpose the variables of the problem are divided into two sets of "fast" and "slow" variables. The latter regroups all the variables of interest which we denote as $\{A_k\}_{1..n}$ or by a column vector $\mathbf{A}(\tau)$ for brevity. All the other variables which are considered as irrelevant are included in the "fast" set. To simplify the notation the time dependence is denoted by a subscript: $\mathbf{A}_\tau \equiv \mathbf{A}(\tau)$, so that $\mathbf{A}_0 = \mathbf{A}(0)$. Let's define the projection operator $\mathcal{P}$ on the subspace spanned by $\{A_k\}_{1..n}$ and the orthogonal projector $\mathcal{Q}$:

$$\mathcal{P}f = (\mathbf{A}_0, f)(\mathbf{A}_0, \mathbf{A}_0)^{-1}\mathbf{A}_0 = \sum_{ln} A_n(0)(A_l(0), A_n(0))^{-1}(A_l(0), f)$$

$$\mathcal{Q} = 1 - \mathcal{P}$$

where $\mathcal{Q}$ is the projection operator on the subspace orthogonal to $\{A_k\}$. It is straightforward to check that $\mathcal{P}, \mathcal{Q}$ have the usual properties: $\mathcal{P}^2 = \mathcal{P}$, $\mathcal{Q}^2 = \mathcal{Q}$ and $\mathcal{P}\mathcal{Q} = 0$. It is worth noting that we only need to specify the relevant

variables $\{A_k\}$ in order to construct the projectors and do not need to enumerate the irrelevant ones.

The starting point of the derivation of an effective theory is the Liouville equation (2.1). We would like to describe the evolution of $\mathbf{A}(\tau)$ in terms of "slow" variables only and to eliminate all the irrelevant or "fast" variables from the equation with the help of $\mathcal{P}$ and $\mathcal{Q}$. First we write the formal solution of (2.1): $\mathbf{A}_\tau = \exp(i\mathcal{L}\tau)\mathbf{A}_0$. Second, we insert this solution and the identity decomposition $1 = \mathcal{P} + \mathcal{Q}$ into (2.1):

$$\frac{d\mathbf{A}_\tau}{d\tau} = \exp(i\mathcal{L})[\mathcal{P}+\mathcal{Q}]i\mathcal{L}\mathbf{A}_0 = i\Omega\cdot\mathbf{A}_\tau + \exp(i\mathcal{L}\tau)\mathcal{Q}i\mathcal{L}\mathbf{A}_0 \tag{2.3}$$

$$i\Omega = (\mathbf{A}_0, i\mathcal{L}\mathbf{A}_0)\cdot(\mathbf{A}_0,\mathbf{A}_0)^{-1} \tag{2.4}$$

The second term is simplified with the help of the following operator identity:

$$\exp(i\mathcal{L}\tau) = \exp(i\mathcal{L}\tau)\mathcal{B}(\tau) + \exp(i\mathcal{Q}\mathcal{L}\tau) \tag{2.5}$$

where $\mathcal{B}$ is easily identified: indeed differentiating the identity with respect to t yields consequently:

$$\begin{aligned}
i\mathcal{L}e^{i\mathcal{L}\tau} &= i\mathcal{L}e^{i\mathcal{L}\tau}\mathcal{B}(\tau) + e^{i\mathcal{L}\tau}\dot{\mathcal{B}}(\tau) + i\mathcal{Q}\mathcal{L}e^{i\mathcal{Q}\mathcal{L}\tau}\\
i\mathcal{L}\left(e^{i\mathcal{L}\tau}\mathcal{B}(\tau) + \exp(i\mathcal{Q}\mathcal{L}\tau)\right) &= i\mathcal{L}e^{i\mathcal{L}\tau}\mathcal{B}(\tau) + e^{i\mathcal{L}\tau}\dot{\mathcal{B}}(\tau) + i\mathcal{Q}\mathcal{L}e^{i\mathcal{Q}\mathcal{L}\tau}\\
i\mathcal{L}e^{i\mathcal{Q}\mathcal{L}\tau} &= e^{i\mathcal{L}\tau}\dot{\mathcal{B}}(\tau) + i\mathcal{Q}\mathcal{L}e^{i\mathcal{Q}\mathcal{L}\tau}\\
\dot{\mathcal{B}}(\tau) &= ie^{-i\mathcal{L}\tau}\mathcal{P}\mathcal{L}e^{i\mathcal{Q}\mathcal{L}\tau}\\
\mathcal{B}(\tau) &= i\int_0^\tau du e^{-i\mathcal{L}u}\mathcal{P}\mathcal{L}e^{i\mathcal{Q}\mathcal{L}u}
\end{aligned}$$

and $\mathcal{B}(0) = 0$ from (2.5). Now applying (2.5) to the second term in the right hand side of (2.3) yields:

$$\exp(i\mathcal{L}\tau)\mathcal{Q}i\mathcal{L}\mathbf{A}_0 = \int_0^\tau e^{i\mathcal{L}(\tau-u)}i\mathcal{P}\mathcal{L}\mathbf{f}(u) + \mathbf{f}(\tau) \tag{2.6}$$

where we introduced a new variable $\mathbf{f}(t)$ - the *fluctuating force*:

$$\mathbf{f}(\tau) \equiv e^{i\mathcal{Q}\mathcal{L}\tau}i\mathcal{Q}\mathcal{L}\mathbf{A}_0 \tag{2.7}$$

We see that the evolution of $\mathbf{f}(\tau)$ from its initial value $i\mathcal{Q}\mathcal{L}\mathbf{A}_0$ (which is a "fast" variable) is ruled by an "orthogonal" dynamics $\exp(i\mathcal{Q}\mathcal{L}\tau)$. For this reason $\mathbf{f}(\tau)$ is expected to be a pure "fast" variable if $\mathbf{A}_\tau$ is really "slow". The definition of $\mathbf{f}$ implies that that it is always orthogonal to $\mathbf{A}_0$: $(\mathbf{f}(\tau),\mathbf{A}_0) = 0$ (indeed by definition $\mathbf{f}(\tau) = \mathcal{Q}[\cdots]$ belongs to a subspace orthogonal to the one spanned by $\mathbf{A}_0$).

Finally (2.3) reads (after substituting (2.6) in (2.3)):

$$\frac{d\mathbf{A}_\tau}{d\tau} = i\Omega\cdot\mathbf{A}_\tau - \int_0^\tau du\mathbf{M}(\tau-u)\mathbf{A}_u + \mathbf{f}(\tau) \tag{2.8}$$
$$\mathbf{M}(\tau) = -i(\mathbf{A}_0, \mathcal{L}\mathbf{f}(\tau))(\mathbf{A}_0,\mathbf{A}_0)^{-1}$$

This equation is called the generalised Langevin equation and $\mathbf{M}$ - the *memory function* or the *memory kernel*. Indeed the structure of (2.8) resembles that of a Langevin equation although $\mathbf{f}$ is not a noise. Using the following identity:

$$i(\mathbf{A}_0, \mathcal{L}\mathbf{f}(\tau)) = i(\mathcal{L}\mathbf{A}_0, \mathbf{f}(\tau)) = i(\mathcal{Q}\mathcal{L}\mathbf{A}_0, \mathbf{f}(\tau)) = -(\mathbf{f}(0), \mathbf{f}(\tau))$$

We can rewrite $\mathbf{M}$ as:

$$\mathbf{M}(\tau) = (\mathbf{f}(0), \mathbf{f}(\tau))(\mathbf{A}_0,\mathbf{A}_0)^{-1} \tag{2.9}$$

An equation on $\mathbf{C}(\tau) = (\mathbf{A}_\tau, \mathbf{A}_0)$, the matrix of the correlation functions, follows immediately from (2.8) by multiplying it with $\mathbf{A}_0$:

$$\frac{d\mathbf{C}(\tau)}{d\tau} = i\Omega\cdot\mathbf{C}(\tau) - \int_0^\tau du\mathbf{M}(\tau-u)\cdot\mathbf{C}(u) \tag{2.10}$$

Note that the fluctuating force dropped out.

It is important to realise that (2.8) is just a rearrangement of the initial Liouville equation (2.1) and has the same complexity. Therefore (2.10) is formally exact but the formal definition of $\mathbf{M}$ (2.9) is too complicated to be used directly for computation. Its advantage over the original Liouville equation is that it focuses on a reduced set of variables. However it needs to be supplied by an approximation for the kernel for any practical use. If $\mathbf{A}$ is a complete set of the slow variables of the system (in the above context) then there are only two possibilities for $\mathbf{M}$: either it is a fast variable and it can be averaged over short time scales to give a δ-function on a longer ("slow") timescales so that $\mathbf{M}(\tau) = M\delta(\tau)$, either it is a slow variable and it can be expanded in powers of $\mathbf{A}$.

Let's also note that the overall success relies crucially on the clever choice of the "slow"/relevant variables. A good choice may lead to fine results. At the same time the bad one while still being formally correct may lead to bad results. It is not always possible to distinguish choices in advance. Existence of different sharp separated timescales in a system can justify the separation in "slow" and "fast" variables for variables defined on different timescales.

2.2 Supercooled liquids

We would like to describe a supercooled liquid in the context of the above formalism with density as a slow variable. An extremely fast growth of relaxation

time τ_{REL} for low temperatures defines two sharp separated timescales in the problem: a microscopic time which describes processes at an atomic/molecular level and the relaxation time. This defines naturally fast and slow variables.

2.2.1 Theory of dynamic density fluctuations

A liquid is a system of N interacting point particles with a pairwise interaction $U(r)$. Its Hamiltonian $\mathcal{H}$ reads:

$$\mathcal{H} = \sum_l \frac{\mathbf{p}_l^2}{2m} + \frac{1}{2}\sum_{l\neq j} U(\mathbf{r}_l - \mathbf{r}_j)$$

where $\mathbf{r}$ and $\mathbf{p}$ are particles positions and momenta respectively, m is a particle mass. The phase space is generated by the set of all coordinates and momenta of the particles: $X = (\mathbf{r}, \mathbf{p})$.

Time evolution is governed by the Liouville equation (2.1). The operator $\mathcal{L}$ in this particular case reads:

$$i\mathcal{L} = \frac{1}{m}\sum_l \left(\mathbf{p}_l \frac{\partial}{\partial \mathbf{r}_l}\right) - \sum_{l\neq j}\left(\frac{\partial U(\mathbf{r}_l - \mathbf{r}_j)}{\partial \mathbf{r}_l}\cdot\frac{\partial}{\partial \mathbf{p}_l}\right)$$

We define the inner product as the canonical average *i.e.* the inner product kernel (see (2.2)) is $P(\mathbf{r}) = \exp(-\beta\mathcal{H})$ where β is the inverse temperature. The density ρ, density fluctuations $\delta\rho$ and the longitudinal current are defined as:

$$\begin{aligned}
\rho_{\mathbf{q}}(\tau) &= \sum_j \exp(i\mathbf{q}\cdot\mathbf{r}_j(\tau)) \\
\delta\rho_{\mathbf{q}}(\tau) &= \rho_{\mathbf{q}}(\tau) - (2\pi)^3\rho\delta(\mathbf{q}) \\
\dot{\rho}_{\mathbf{q}}(\tau) = i\mathbf{q}\cdot\sum_l \frac{\mathbf{p}_l(\tau)}{m} e^{i\mathbf{q}\cdot\mathbf{r}_l(\tau)} &= i\mathbf{q}\cdot j_{\mathbf{q}}(\tau) = i|\mathbf{q}|j^L_{\mathbf{q}}(\tau)
\end{aligned} \tag{2.11}$$

where $\rho \equiv (\rho, 1)$ is the average density. The last equation (2.11) is the density conservation law implied by Hamiltonian dynamics. It implies in particular that if ρ is a "slow" variable than $j^L \sim \dot{\rho}$ is also a "slow" variable. We take $\delta\rho_{\mathbf{q}}$ and $j^L_{\mathbf{q}}$ as relevant/"slow" variables and

$$\mathbf{A} = \begin{bmatrix} \delta\rho_{\mathbf{q}} \\ j^L_{\mathbf{q}} \end{bmatrix},$$

which is an assumption. The matrix of correlators $\mathbf{C}$ reads:

$$\mathbf{C}(\tau) = \begin{pmatrix} \langle\delta\rho_{-\mathbf{q}}(0)\delta\rho_{\mathbf{q}}(\tau)\rangle & \langle\delta\rho_{-\mathbf{q}}(0)j^L_{\mathbf{q}}(\tau)\rangle \\ \langle\delta j^L_{-\mathbf{q}}(0)\delta\rho_{\mathbf{q}}(\tau)\rangle & \langle j^L_{-\mathbf{q}}(0)j^L_{\mathbf{q}}(\tau)\rangle \end{pmatrix}$$

$$C(\mathbf{q},\tau) = \frac{1}{N}\langle\delta\rho(-\mathbf{q})\delta\rho(\mathbf{q},\tau)\rangle$$

The fluctuating force at zero time reads:

$$\mathbf{f}(0) = \mathcal{Q}i\mathcal{L}\mathbf{A} = \mathcal{Q}\frac{d\mathbf{A}_\tau}{d\tau}(\tau=0) = \begin{bmatrix} \frac{d\delta\rho_\mathbf{q}}{d\tau} \\ \frac{dj^L_\mathbf{q}}{d\tau} \end{bmatrix} - \begin{bmatrix} 0 & i\mathbf{q} \\ \frac{iqT}{mS_\mathbf{q}} & 0 \end{bmatrix} \cdot \begin{bmatrix} \delta\rho_\mathbf{q} \\ j^L_\mathbf{q} \end{bmatrix} = \begin{bmatrix} 0 \\ \frac{dj^L_\mathbf{q}}{d\tau} - \frac{iqT}{mS_\mathbf{q}}\delta\rho_\mathbf{q} \end{bmatrix} = \begin{bmatrix} 0 \\ R_\mathbf{q}(0) \end{bmatrix}$$

and $S_\mathbf{q} = C(\mathbf{q}, 0)$ is the static structure factor; recall that $R_\mathbf{q}(\tau)$ evolves under the projected dynamics $\exp(i\mathcal{Q}\mathcal{L}\tau)$. Applying the formal constructions of the previous section one derives the following equation (see Appendix A for details) for $\tau \geq 0$:

$$\frac{d^2C(\mathbf{q},\tau)}{d\tau^2} + \frac{\mathbf{q}^2T}{mS_\mathbf{q}}C(\mathbf{q},\tau) + \frac{m}{NT}\int_0^\tau duM(\mathbf{q},\tau-u)\frac{dC(\mathbf{q},u)}{du} = 0 \qquad (2.12)$$

$$R_\mathbf{q}(\tau) = e^{i\mathcal{Q}\mathcal{L}\tau}\left[\frac{dj^L_\mathbf{q}}{d\tau}(0) - \frac{iqT}{mS_\mathbf{q}}\delta\rho_\mathbf{q}(0)\right] \qquad (2.13)$$

$$M(\mathbf{q},\tau) = \langle R_{-\mathbf{q}}(0)R_\mathbf{q}(\tau)\rangle \qquad (2.14)$$

where M is the memory kernel and $R_\mathbf{q}$ is the current associated component of the fluctuating force $\mathbf{f}$ (see Appendix A).

2.2.2 Mode-Coupling Factorisation

As we stated above equation (2.12) is a rearrangement of the Liouville equation (2.1) and has equivalent complexity. As such it requires some approximation. As we pointed out in the end of Section 2.1 the general form of M is

$$M(\tau) = M_0\delta(\tau) + M_1[\rho, j^L]$$

where $M_1[\rho, j^L]$ is a functional of the correlation of slow variables: density and longitudinal current. M_1 cannot be zero: in that case solution of (2.12) presents exponential relaxation for all temperatures and no slow dynamics emerges. Thus $M_1 \neq 0$, and consequently the fluctuating force $\mathbf{f}$ is not a pure "fast" variable: its variance has a slow contribution. We are forced to conclude that the fluctuating force has "fast" and "slow" components. The "fast" component $M_{fast}(\mathbf{q},\tau)$ is well approximated by a δ-function: $M_{fast}(\mathbf{q},\tau) = M_0(\mathbf{q})\delta(\tau)$. This is the part of the memory kernel responsible for the usual relaxation dynamics in liquids but not important to describe the slow dynamics. Computation of its exact functional form is a problem of the liquid state theory.

The original idea of Götze and Leuthesser [21, 20] was to expand the "slow" component $M_{slow}(\mathbf{q},\tau)$ in powers of $\delta\rho$ and j^L. The fluctuating force is orthogonal to $\delta\rho$ and j^L by definition. The next possible term in the expansion is

quadratic in slow variables; an inspection of $R_{\mathbf{q}}$ supports this idea: recalling that

$$R_{\mathbf{q}}(0) = \frac{dj^L_{\mathbf{q}}}{d\tau}(0) - i\frac{qT}{mS_{\mathbf{q}}}\delta\rho_{\mathbf{q}}(0)$$

and ($\hat{\mathbf{q}} = \mathbf{q}/q$) leads to:

$$\frac{dj^L_{\mathbf{q}}}{d\tau} = \frac{1}{m}\frac{d}{d\tau}\left\{\sum_l (\hat{\mathbf{q}}\cdot\mathbf{p}_l)e^{i\mathbf{q}\cdot\mathbf{r}_l}\right\} = \frac{1}{m}\sum_l\left(\hat{\mathbf{q}}\cdot\frac{d\mathbf{p}_l}{d\tau}\right)e^{i\mathbf{q}\cdot\mathbf{r}_l} + \frac{i}{m^2}\sum_l(\hat{\mathbf{q}}\cdot\mathbf{p}_l)^2\,e^{i\mathbf{q}\cdot\mathbf{r}_l}$$

The first term is easily evaluated recalling that $d\mathbf{p}/d\tau$ is a force:

$$\sum_l\left(\hat{\mathbf{q}}\cdot\frac{d\mathbf{p}_l}{d\tau}\right)e^{i\mathbf{q}\cdot\mathbf{r}_l} = -\hat{\mathbf{q}}\cdot\sum_{j\neq l}\nabla U(|\mathbf{r}_l-\mathbf{r}_j|)e^{i\mathbf{q}\cdot\mathbf{r}_l} =$$
$$-\hat{\mathbf{q}}\cdot\iint dxdy e^{i\mathbf{q}\cdot x}\rho(x)\rho(y)\nabla U(x-y) \sim i\sum_k(\hat{\mathbf{q}}\cdot\mathbf{k})\,U_{\mathbf{k}}\rho_{\mathbf{k}}\rho_{-\mathbf{k}}$$

Clearly a product of slow modes $\delta\rho_{-\mathbf{q}}\delta\rho_{\mathbf{q}}$ appears in $R_{\mathbf{q}}(0)$ [36]. For $\tau > 0$ $R_{\mathbf{q}}(\tau)$ evolves under the projected dynamics and a similar evaluation is highly non-trivial. However following Götze [21] and Leuthesser [20] we assume that this contribution, $\delta\rho_{-\mathbf{q}}\delta\rho_{\mathbf{q}}$, is dominant for all τ: $R_{\mathbf{q}}(\tau) \sim V_{\mathbf{q}}\delta\rho_{\mathbf{q}}\delta\rho_{-\mathbf{q}}$. This hypothesis is the base of the Mode-Coupling Factorisation. Below we present briefly the steps of the approximation which are detailed afterwards (some details are presented in Appendix A). The first step is to use the "dominant contribution" hypothesis and to approximate M as:

$$M_{slow}(\mathbf{q},\tau) = \langle R_{-\mathbf{q}}\exp(i\mathcal{Q}\mathcal{L}\tau)R_{\mathbf{q}}\rangle \approx \int V^2\langle\delta\rho(0)\delta\rho(0)e^{i\mathcal{Q}\mathcal{L}\tau}\delta\rho(0)\delta\rho(0)\rangle$$

The wavevector dependence of the right hand side is detailed later. Still the computation of the four-point density correlator in the right hand side of the equation is complicated because of the presence of the "projected" propagator. We substitute it by the normal propagator $e^{i\mathcal{L}\tau}$:

$$\begin{aligned} V^2\langle\delta\rho(0)\delta\rho(0)e^{i\mathcal{Q}\mathcal{L}\tau}\delta\rho(0)\delta\rho(0)\rangle \quad &\rightarrow \quad V^2\langle\delta\rho(0)\delta\rho(0)e^{i\mathcal{L}\tau}\left[\delta\rho(0)\delta\rho(0)\right] \\ &\rightarrow \quad V^2\langle\delta\rho(0)\delta\rho(0)\delta\rho(\tau)\delta\rho(\tau)\rangle \quad (2.15)\end{aligned}$$

The resulting four-point function in Eq. (2.15) is then factorised into a product of two-point correlators so that

$$M_{slow}(\mathbf{q},\tau) \sim \int_{\mathbf{k}} V^2(\mathbf{q},\mathbf{k})C(\mathbf{q}-\mathbf{k},\tau)C(\mathbf{k},\tau).$$

The above neglection of "projected" dynamics is not justified at all but it is the only way to compute M. There is an associated subtlety indicating that

the substitution should be done with care. Indeed if one neglects the projected dynamics directly in the definition of $R_{\mathbf{q}}$ (see Eq. (2.13)) then one can express $R_{\mathbf{q}}(\tau)$ via $j^L_{\mathbf{q}}(\tau)$ and $\delta\rho_{\mathbf{q}}(\tau)$. Consequently M can be evaluated immediately:

$$M_{slow}(\mathbf{q},\tau) \sim C(\mathbf{q},\tau) + \text{Time derivatives of } C$$

which provides completely wrong results.

Let's now introduce more details to the above scheme. The computation of V is done via introduction of a projection operator $\mathcal{P}_2$ that projects $R_{\mathbf{q}}$ onto its (presumably) dominant contribution [36]: $\delta\rho\delta\rho$:

$$\mathcal{P}_2 = \sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} \delta\rho_{\mathbf{k}_1}\delta\rho_{\mathbf{k}_2}\langle\delta\rho^*_{\mathbf{k}_3}\delta\rho^*_{\mathbf{k}_4}\cdots\rangle\langle\delta\rho^*_{\mathbf{k}_1}\delta\rho^*_{\mathbf{k}_2}\delta\rho_{\mathbf{k}_3}\delta\rho_{\mathbf{k}_4}\rangle^{-1}$$

where a star denotes a complex conjugation. First, the fluctuating force is projected onto its dominant contribution *i.e.* we replace $e^{i\mathcal{Q}\mathcal{L}\tau} \to \mathcal{P}_2 e^{i\mathcal{Q}\mathcal{L}\tau}\mathcal{P}_2$ in the expression for M:

$$M(\mathbf{q},\tau) = \langle R_{\mathbf{q}}(0)e^{i\mathcal{Q}\mathcal{L}\tau}R_{\mathbf{q}}(0)\rangle \approx \langle \mathcal{P}_2 R_{\mathbf{q}}(0)e^{i\mathcal{Q}\mathcal{L}\tau}\mathcal{P}_2 R_{\mathbf{q}}(0)\rangle.$$

The projection $\mathcal{P}_2 R_{\mathbf{q}}$ reads (all quantities are evaluated at $\tau = 0$):

$$\mathcal{P}_2 R_{\mathbf{q}} = \sum_{\mathbf{k}_1,\mathbf{k}_2} V_{\mathbf{q}}(\mathbf{k}_1,\mathbf{k}_2)\delta\rho_{\mathbf{k}_1}\delta\rho_{\mathbf{k}_2}$$
$$V_{\mathbf{q}}(\mathbf{k}_1,\mathbf{k}_2) = \sum_{\mathbf{k}_3,\mathbf{k}_4} \langle\delta\rho_{\mathbf{k}_1}\delta\rho_{\mathbf{k}_2}R_{\mathbf{q}}\rangle\cdot\langle\delta\rho_{\mathbf{k}_1}\delta\rho_{\mathbf{k}_2}\delta\rho_{\mathbf{k}_3}\delta\rho_{\mathbf{k}_4}\rangle^{-1}$$

Second, we neglect the projected dynamics of $\mathcal{P}_2 R_{\mathbf{q}}(\tau)$: we replace $e^{i\mathcal{Q}\mathcal{L}\tau} \to e^{i\mathcal{L}\tau}$ so that $\delta\rho$ in $\mathcal{P}_2 R_{\mathbf{q}}(\tau)$ evolves under complete dynamics and not the projected one:

$$e^{i\mathcal{L}\tau}\mathcal{P}_2 R_{\mathbf{q}} = \sum_{\mathbf{k}_1,\mathbf{k}_2} V_{\mathbf{q}}(\mathbf{k}_1,\mathbf{k}_2)\delta\rho_{\mathbf{k}_1}(\tau)\delta\rho_{\mathbf{k}_2}(\tau).$$

We have already discussed the justification of such simplification. It is worth pointing that the "projected" dynamics is neglected only after projecting on the dominant contribution. Now, the approximation for $M_{slow}(\mathbf{q},\tau)$ reads:

$$M(\mathbf{q},\tau) \approx \sum_{\mathbf{k}_1,\mathbf{k}_2} |V^*(\mathbf{k}_1,\mathbf{q}-\mathbf{k}_1)V(\mathbf{k}_2,\mathbf{q}-\mathbf{k}_2)|\langle\delta\rho_{-\mathbf{k}_2}\delta\rho_{\mathbf{k}_2-\mathbf{q}}\delta\rho_{\mathbf{k}_1}(\tau)\delta\rho_{\mathbf{q}-\mathbf{k}_1}(\tau)\rangle.$$

The four-point density correlator is factorised into products of two-point functions by virtue of Wick's theorem:

$$\langle\delta\rho_{-\mathbf{k}_2}\delta\rho_{\mathbf{k}_2-\mathbf{q}}\delta\rho_{\mathbf{k}_1}(\tau)\delta\rho_{\mathbf{q}-\mathbf{k}_1}(\tau)\rangle \approx$$
$$N^2\delta_{\mathbf{k}_1,\mathbf{k}_1-\mathbf{q}}\delta_{\mathbf{k}_2,\mathbf{k}_2-\mathbf{q}}C^2(\mathbf{q}/2,0) + N^2\left[\delta_{\mathbf{k}_1,\mathbf{k}_2} + \delta_{\mathbf{k}_2,\mathbf{q}-\mathbf{k}_1}\right]C(\mathbf{k}_1,\tau)C(\mathbf{q}-\mathbf{k}_1,\tau).$$

The first term is a static contribution and therefore can be neglected. Plugging this into the above expression for the memory kernel yields:

$$M_slow(\mathbf{q},\tau) \sim \sum_{\mathbf{k}} |V(\mathbf{k},\mathbf{q}-\mathbf{k})|^2 C(\mathbf{k},\tau)C(\mathbf{q}-\mathbf{k},\tau).$$

The computation of V is presented in Appendix B.

Finally, the derived approximation for M is the famous Mode-Coupling factorisation which closes Eq. (2.12):

$$\begin{aligned} M(\mathbf{q},\tau) &= M_{slow}(\mathbf{q},\tau) + M_{fast}(\mathbf{q},\tau) \\ M_{fast}(\mathbf{q},\tau) &= M_0(\mathbf{q})\delta(\tau) \\ M_{slow}(\mathbf{q},\tau) &= \frac{\rho T}{16\pi^3 m}\int \frac{d^3\mathbf{k}}{(2\pi)^3}|V(\mathbf{q}-\mathbf{k},\mathbf{k})|^2 C(\mathbf{q},\tau)C(\mathbf{q}-\mathbf{k},\tau) \\ V(\mathbf{q}-\mathbf{k},\mathbf{k}) &= \{(\hat{\mathbf{q}}\cdot\mathbf{k})c(k) + \hat{\mathbf{q}}\cdot(\mathbf{q}-\mathbf{k})c(|\mathbf{q}-\mathbf{k}|)\} \\ c(q) &= \frac{1}{\rho}\left(1-\frac{1}{S(q)}\right) \end{aligned} \tag{2.16}$$

Equation (2.12) with the total memory kernel $M(\mathbf{q},\tau)$ is known as Full MCT equation:

$$\frac{d^2C(\mathbf{q},\tau)}{d\tau^2} + \frac{mM_0(\mathbf{q})}{NT}\frac{dC(\mathbf{q},\tau)}{d\tau} + \frac{\mathbf{q}^2T}{mS_{\mathbf{q}}}C(\mathbf{q},\tau) + \tag{2.17}$$

$$+ \int_0^\tau du M_{slow}(\mathbf{q},\tau-u)\frac{dC(\mathbf{q},u)}{du} = 0 \tag{2.18}$$

As we pointed out earlier the regular relaxation kernel M_0 is not important for the slow dynamics and can be dropped. Furthermore the second derivative term can be replaced by the first derivative in Eq. (2.17) without qualitative change of the results [6]:

$$\frac{dC(\mathbf{q},\tau)}{d\tau} + \Omega_{\mathbf{q}}^2 C(\mathbf{q},\tau) + \kappa_{\mathbf{q}}\int_0^\tau du M_{sl}(\mathbf{q},\tau-u)\frac{dC(\mathbf{q},u)}{du} = 0 \tag{2.19}$$

$$\begin{aligned} M_{sl}(\mathbf{q},\tau) &= \int \frac{d^3\mathbf{k}}{(2\pi)^3}|V(\mathbf{q}-\mathbf{k},\mathbf{k})|^2 C(\mathbf{k},\tau)C(\mathbf{q}-\mathbf{k},\tau) \\ \Omega_{\mathbf{q}}^2 &= \frac{\mathbf{q}^2T}{mS(\mathbf{q})} \quad \text{and} \quad \kappa_{\mathbf{q}} = \frac{\rho T}{15\pi^3 m} \end{aligned}$$

Or after Laplace transform: $\hat{C}(\mathbf{q},z) = \mathcal{L}C = \int_0^\infty d\tau \exp(-z\tau)C(\mathbf{q},\tau)$

$$\frac{T\hat{C}(\mathbf{q},z)}{S(\mathbf{q}) - z\hat{C}(\mathbf{q},z)} = \frac{\kappa_{\mathbf{q}}}{\Omega_{\mathbf{q}}^2}\hat{M}_{sl}(\mathbf{q},z). \tag{2.20}$$

2.3 Ideal glass transition

The main MCT equation (2.19) is an equation of an over-damped oscillator with self-consistent time-dependent damping. The non-linear feedback effect leads to a strong dependence of the relaxation dynamics on the input parameters and provides a qualitative explanation of the extremely fast slowing down of the dynamics in glass-forming liquids for a mild variation of parameters. The only input to (2.19) is the static structure factor $S(q)$ and the temperature T.

Equation (2.20) is still too complicated for an analytical solution. Numerical solutions of (2.20) provide an insight into its properties [37]: the relaxation time grows beyond any bound as the temperature approaches a limit value $T = T_d$ and the density-density correlator fails to decay to zero at infinite times. This defines a non-ergodic parameter $f_{\mathbf{q}}$ as an infinite time limit of the correlator:

$$f_{\mathbf{q}} = \frac{1}{S_{\mathbf{q}}} \lim_{\tau\to\infty} C(\mathbf{q},\tau).$$

The limit $\tau \to \infty$ of (2.20) yields an equation on $f_{\mathbf{q}}$ within MCT:

$$\frac{f_{\mathbf{q}}}{1-f_{\mathbf{q}}} = \frac{\kappa_{\mathbf{q}} S_{\mathbf{q}}}{\Omega_{\mathbf{q}}^2} \int \frac{d^3\mathbf{k}}{(2\pi)^3} |V(\mathbf{q}-\mathbf{k},\mathbf{k})|^2 S_{\mathbf{q}} S_{\mathbf{q}-\mathbf{k}} f_{\mathbf{q}} f_{\mathbf{q}-\mathbf{k}} \tag{2.21}$$

This equations was analysed for certain class of potentials U, like hard spheres or Lennard-Jones interaction. This analysis revealed that for high enough temperatures there is a unique solution $f_{\mathbf{q}} = 0$. As the temperature is lowered a non zero solution $f_{\mathbf{q}}$ appears signaling a structural arrest: the liquid freezes in an amorphous configuration which breaks translation invariance since $f_{\mathbf{q}} \neq 0$ implies $\langle \delta\rho_{\mathbf{q}} \rangle \neq 0$. This is not a liquid-solid transition because $f(\mathbf{q}) \equiv 0$ in solids but a transition to a non-ergodic phase when the phase space splits into disconnected domains at $T = T_d$.

The relaxation of $C(\mathbf{q},\tau)$ for $T = T_d(1+\epsilon)$ where $\epsilon \ll 1$ obtained from numerical simulations and theoretical analysis is described in the context of a two-step relaxation [6, 21, 39, 38]: a simple exponential decay for high temperatures is replaced by a complex relaxation with three timescales, a plateau $f_{\mathbf{q}}$ emerges in the relaxation pattern of $C(\mathbf{q},\tau)$ which extends to infinity as $T \to T_d$:

- Short times $\tau \sim O(1)$: $C(\mathbf{q},\tau)$ relaxes from its initial value $S_{\mathbf{q}}$ to the plateau $f_{\mathbf{q}}$.
- β-relaxation $\tau \sim \epsilon^{-1/2a}$: describes the relaxation close to the plateau; $C(\mathbf{q},\tau)$ acquires a scaling form:

$$\begin{aligned} C(\mathbf{q},\tau) &= S_{\mathbf{q}} f_{\mathbf{q}} + \delta C(\mathbf{q},\tau) \\ &\sim S_{\mathbf{q}}(f_{\mathbf{q}} + \sqrt{\epsilon}(1-f_{\mathbf{q}})^2 G_1(\mathbf{q},\tau\epsilon^{1/2a}) + \epsilon(1-f_{\mathbf{q}})^2 G_2(\mathbf{q},\tau\epsilon^{1/2a})\cdots) \end{aligned}$$

  and $G_1(\mathbf{q},s)$ ($s = \tau\epsilon^{1/2a}$) has singular asymptotics at $s = 0$ and $s = \infty$ where C deviates from the plateau and this anzats fails:

$$G(\mathbf{q}, s\to 0) \sim s^{-a} \quad \text{and} \quad G(\mathbf{q}, s\to\infty) \sim s^{b}.$$

$G_1(\mathbf{q}, s)$ factorises into a product of a wave-vector dependent amplitude and a time dependent part:

$$G_1(\mathbf{q}, s) = H_1(\mathbf{q})g(s).$$

This property is referred to as a "factorisation property".

- α-relaxation $\tau \sim \epsilon^{-\gamma}$: describes the final decay to zero and τ_α is the relaxation time; $C(\mathbf{q}, \tau)$ takes a scaling form $C_\alpha(\mathbf{q}, \tau\epsilon^\gamma)$. This scaling form is referred to as the time-temperature superposition [6, 40] since the master function C_α does not depend on T.

- The scaling function g satisfies a non-linear equation:

$$\frac{1}{z} + \frac{z}{\lambda}\hat{g}^2 = L[g^2] \tag{2.22}$$

where λ is a numerical constant fixed by Eq. (2.19); g has singular asymptotics: $g(s \to 0) \sim s^{-a}$ and $g(s \to \infty) \sim -s^b$ inherited from G_1.

- The values of exponents a, b are fixed by an equation derived from (2.22):

$$\frac{\Gamma^2(1-a)}{\Gamma(1-2a)} = \frac{\Gamma^2(1+b)}{\Gamma(1+2b)} = \lambda \tag{2.23}$$

and possible values of a are limited to a range $[0, 0.5]$.

In the α-regime the solution of the MCT equation is well approximated by a stretched exponential: $C_\alpha(\mathbf{q}, s) \sim f_\mathbf{q} \exp(-As^b)$ [41]. This is an empirical function used often to fit the data. It is supported by the asymptotics of the late β-regime:

$$\begin{aligned} C(\mathbf{q}, \tau) &\sim f_\mathbf{q} - \sqrt{\epsilon}(1-f_\mathbf{q})^2 \left(\frac{\tau}{\tau_\beta}\right)^b \sim f_\mathbf{q} - (1-f_\mathbf{q})^2 \left(\frac{\tau}{\tau_\beta \epsilon^{-1/2b}}\right)^b \\ &\sim f_\mathbf{q} \exp\left[-\frac{(1-f_\mathbf{q})^2}{f_\mathbf{q}} \left(\frac{\tau}{\tau_\beta \epsilon^{-1/2b}}\right)^b\right] \end{aligned}$$

However while MCT predicts a transition it provides little information about the low temperature phase because the derivation presented in Sec. 2.2 is no more valid in this phase. It is expected that the system falls out-of-equilibrium for low temperatures so that time-translation invariance and time-reversal symmetry assumed within MCT no more hold. It is worth noting however that an attempt to extend the derivation to low temperatures was made by Lätz [29].

2.4 Schematic MCT

Equations (2.17) and (2.19) are very complicated for the analysis so that no analytical solution is known. Numerical solution is also a formidable task due to an extremely fast increase of relaxation time for low temperatures. This reasons motivated to look for a simplification of MCT that would comply with the following requirements:

- Essential results derived within MCT are preserved.
- Analysis of the theory should be simpler.

Such a simplification was first proposed by Bengtzelius, Götze *et al.* [37]. Analysing the numerical solution of (2.17) they found that the main contribution to the kernel $M_{sl}(\mathbf{q},\tau)$ comes from wave-vectors $\mathbf{q} \approx \mathbf{q}_0$ where $\mathbf{q}_0$ is the first peak of the static structure factor $S_{\mathbf{q}}$. They suggested to insert the structure factor: $S_{\mathbf{q}} = 1 + S_0\delta(\mathbf{q}-\mathbf{q}_0)$ in M_{sl} and evaluate $C(\mathbf{q},\tau) \sim C(\tau)\delta(\mathbf{q}-\mathbf{q}_0)$. This eliminates the wave-vector dependence from (2.19):

$$\frac{dC(\tau)}{d\tau} + \Omega^2 C(\tau) + \kappa \int_0^\tau du C^2(\tau-u)\frac{dC(u)}{du} = 0 \tag{2.24}$$

This version of the MCT equation is referred to as schematic MCT [37]. As we shall see in Chapters 3 and 6 the existence of the schematic MCT preserving the essential properties of the full MCT has deep underlying reasons.

2.5 Diverging lengthscale

Let's come back to the full wave-vector dependent theory. The interpretation of the transition predicted within MCT as a transition to a non ergodic phase suggests that the slowing down of the dynamics close to the transition is caused by increasing cooperativity as $T \to T_d$ when more and more particles have to move simultaneously. This is associated with a growing correlation length which links to the notion of the dynamic heterogeneities when the liquid splits into regions evolving differently; this also suggests an analogy with critical phenomena which could probably shed more light on the subtleties of the transition. The idea of cooperativity behind the glass transition pervaded the literature for years [42]. Surprisingly, MCT provides a different image at a first glance. Equations (2.17), (2.19) present no short wave-vector singularity and consequently no correlation length can be extracted. The authors of MCT insisted on a local, short lengthscale nature of the glass transition [34] caused by the self-blockage of particles inside a cages formed by their neighbours. Particles rattle in independent cages before they finally escape. Because of assumed independence cages were considered as short scale phenomena. The fact that sMCT given by Eq. (2.24) which lacks any spatial dependence retains all the essential properties of the full MCT provided an additional argument in favour of the absence of diverging lengthscale in MCT.

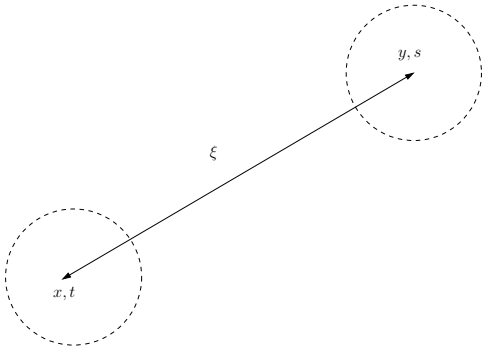


Figure 2.1: Four-point function measures how the dynamics at a point y at a time s correlates with a dynamics at a different point x at some different time t. The lengthscale ξ then defines the distance at wich fluctuations at both points are still correlated.

It is only recently that the proper measure of the cooperativity was identified in experiments [43] and numerical simulations [44, 45, 46, 47, 48]: the idea is to measure how events are correlated in space and time. The idea resembles that used to define the correlation length in critical phenomena. Precisely if we perturb the system at around a given point $\mathbf{x}$ at time t then we are interested how this influences the dynamics at a different point $\mathbf{y}$ at some later time s. The region affected by a perturbation defines a correlation length ξ. In the context of supercooled liquids this amounts to an analysis of a divergence in a multi-point correlation function [49].

The lengthscale also exists within MCT and it was identified 3 years ago [50]. The computation of the lengthscale was carried out in a context of field-theories derived earlier in [51, 28] and was based on a resummation of a certain class of diagrams in the expansion of the four-point correlation function. Recently the growing lengthscale was also identified in the context of Inhomogeneous MCT (IMCT) derived via the projector operator formalism [52] and directly related to standard MCT. Inhomogeneous MCT extends MCT to spatially inhomogeneous situations and provides an access to the lengthscale via an analysis of the influence of a perturbation at point $\mathbf{x}$ on the dynamics at a point $\mathbf{y}$ at a later time (see Fig. 2.5).

Let's reconsider a liquid introduced in 2.2 perturbed by an external potential $U_{EXT}(\mathbf{x})$ that breaks the space translation invariance (STI). The derivation follows that of section 2.2 although one should account for the broken space translation invariance:

$$\rho(\mathbf{q}) = \frac{1}{N}\sum_{l=1}^{N}\langle \exp(i\mathbf{q}\cdot \mathbf{r}_l)\rangle$$
$$\delta\rho_{\mathbf{q}} = \sum_{l=1}^{N}\exp(i\mathbf{q}\cdot \mathbf{r}_l) - \rho_{\mathbf{q}}$$
$$C(\mathbf{q}_1, \mathbf{q}_2; \tau) = \frac{1}{N}\langle \delta\rho_{-\mathbf{q}_1}(0)\delta\rho_{\mathbf{q}_2}(\tau)\rangle$$

Repeating steps of the Mori-Zwanzig formalism from 2.1 yields an equation:

$$\frac{\partial^2 C(\mathbf{q}_1, \mathbf{q}_2; \tau)}{\partial \tau^2} + \int d\mathbf{k}\Omega^2(\mathbf{q}_1, \mathbf{k})C(\mathbf{k}, \mathbf{q}_2; \tau) \tag{2.25}$$
$$+ \int d\mathbf{k}\int_0^{\tau} du M(\mathbf{q}_1, \mathbf{k}; \tau - u)\frac{\partial}{\partial u}C(\mathbf{k}, \mathbf{q}_2; u) = 0$$

where $\Omega^2(\mathbf{q}, \mathbf{k}) = \frac{T}{m}\mathbf{q}\cdot \mathbf{k}\rho(\mathbf{q}-\mathbf{k})S^{-1}(\mathbf{q}, \mathbf{k})$ and $S(\mathbf{q}, \mathbf{k})$ is the static structure factor. $M(\mathbf{q}_1, \mathbf{q}_2; \tau)$ is a memory kernel generalising the memory kernel from Sec. 2.2 which is expressed in terms of the fluctuating force. A Mode-Coupling approximation analogous to that of usual MCT can be done to close this equation but the final equation is rather cumbersome[53]. Fortunately one can consider a weakly inhomogeneous situation $U_{EXT}(\mathbf{x}) \ll T$ to access the correlation length. The aim is to compute the susceptibility $\chi_{\mathbf{p}}(\mathbf{q}, \tau)$ associated with the response of the structure factor to a small external perturbation of an arbitrary structure, in particular a localised perturbation:

$$\frac{\delta C(\mathbf{x}, \mathbf{y}; \tau)}{\delta U_{EXT}(\mathbf{z})}|_{U_{EXT}=0} = \int \frac{d^3\mathbf{p}}{(2\pi)^3}\int \frac{d^3\mathbf{q}}{(2\pi)^3} e^{-i\mathbf{q}\cdot(\mathbf{x}-\mathbf{y})+i\mathbf{p}\cdot(\mathbf{y}-\mathbf{z})}\chi_{\mathbf{p}}(\mathbf{q}, \tau).$$

For a perturbation localised at the origin $U_{EXT}(\mathbf{x}) = U_0\delta(\mathbf{x})$ one finds:

$$\delta C(\mathbf{q}_1, \mathbf{y}, \tau) = \int d\mathbf{p} e^{i\mathbf{q}\cdot\mathbf{y}}\chi_{\mathbf{p}}(\mathbf{q}, \tau).$$

The susceptibility $\chi_{\mathbf{x}}(\mathbf{y}, \tau)$ probes the dynamic response for perturbations and measures the retarded response of the dynamics in a point $\mathbf{y}$ to a perturbation in a point $\mathbf{x}$ after time τ. This function has the same critical properties as the previously considered four-point function from [50, 52, 53].

Assuming that U_{EXT} is small one can expand all the quantities to the first order in U_{EXT}. An equation on χ is derived by differentiating (2.25) with respect to U_{EXT} [52, 53]:

$$\frac{\partial^2 \chi_{\mathbf{p}}(\mathbf{q},\tau)}{\partial^2 \tau} + \frac{T\mathbf{q}^2}{mS(q)}\chi_{\mathbf{p}}(\mathbf{q},\tau) + \int_0^\tau du M_{slow}(\mathbf{q},\tau-u)\frac{\partial \chi_{\mathbf{p}}(\mathbf{q},u)}{\partial u} \qquad (2.26)$$

$$+ \int_0^\tau du \frac{Tn^2\rho q}{m|\mathbf{q}+\mathbf{p}|}\int \frac{d^3\mathbf{k}}{(2\pi)^3} V(\mathbf{q};\mathbf{k},\mathbf{q}-\mathbf{k})V(\mathbf{q}+\mathbf{p};\mathbf{q}-\mathbf{k},\mathbf{p}+\mathbf{k}) \times$$

$$\times \chi_{\mathbf{p}}(\mathbf{k},\tau-u)C_0(\mathbf{q}-\mathbf{k},\tau-u)\frac{\partial C_0(\mathbf{q}+\mathbf{p},u)}{\partial u} = m_{\mathbf{p}}(\mathbf{q},\tau)$$

where V is the usual MCT vertex (2.16): $V(\mathbf{q};\mathbf{k}_1,\mathbf{k}_2) = \hat{\mathbf{q}}\cdot\mathbf{k}_1 c(k_1) + \hat{\mathbf{q}}\cdot\mathbf{k}_2 c(k_2)$, M_{slow} is the MCT memory kernel (2.16). The source term $m_{\mathbf{p}}(\mathbf{q},\tau)$ contains C_0 and static multi-point correlations but it does not depend on χ. Then (2.26) is a linear equation of the form $\mathcal{L}_{\mathbf{p}}\chi_{\mathbf{p}} = m_{\mathbf{p}}$ where $\mathcal{L}$ is a certain operator. The lowest eigenvalues of $\mathcal{L}_{\mathbf{p}}$ gives access to the diverging lengthscale.

An involved computation [52, 53] shows the following behaviour of $\chi_{\mathbf{p}}(\mathbf{q},\tau)$ on different timescales (we use below the results presented in 2.2):

- β regime:
$$\chi_{\mathbf{p}}(\mathbf{q},\tau) = \frac{S(q)H_1(q)}{\Gamma\mathbf{p}^2+\sqrt{\epsilon}} g_\beta\left(\frac{\mathbf{p}^2}{\sqrt{\epsilon}}, \tau\epsilon^{1/2a}\right).$$
- α regime:
$$\chi_{\mathbf{p}}(\mathbf{q},\tau) = \frac{\Theta(\Gamma\mathbf{p}^2/\sqrt{\epsilon})}{\sqrt{\epsilon}(\Gamma\mathbf{p}^2+\sqrt{\epsilon})} g_\alpha(\mathbf{q},\tau\epsilon^\gamma)$$
where $\Theta(0) \neq 0$ and $\Theta(x \gg 1) \sim 1/x$, $g_\alpha(\mathbf{q},x) \sim S(q)H_1(q)x^b$ to match β regime and $g_\alpha(\mathbf{q},x \gg 1) \to 0$.

These results imply a diverging lengthscale $\xi \sim \sqrt{\Gamma}\epsilon^{-1/4}$ that governs the dynamics on β and α regimes. This shows that the interpretation of the β regime as a rattling of particles in independent cages is misleading: close to the transition the cages become more and more correlated.

2.6 Experimental evidences, numerics and problems with MCT

The success of the projection operator formalism and hence MCT relies on assumption used to choose the slow variables and assumptions lying at the heart of the Mode-Coupling Factorisation approximation. Up to now we focused on analysis of mathematical properties of MCT equations and neglected whether the theory applies to real systems. As we will see MCT is exact for mean-field systems and it is expected to be only an approximation for short ranged systems.

The applicability of MCT has been tested for a wide class of systems like molecular glasses, colloids, polymers [54], gels, $\cdots$ The general conclusion is

that MCT does a remarkably good job in capturing dynamical properties qualitatively or even quantitatively for the first decades of slowing down [55]. Predictions presented in Sec. 2.3 hold only in the immediate vicinity of the transition *i.e.* they are *asymptotic*, and cannot be reliable for deviations $T - T_d/T_d$ or order one. A reliable method to check MCT predictions against experimental data or numerical simulations is to compare them with a numerical solution of (2.17). However this is a difficult task and is not often done. Usually data are fitted by MCT so that parameters like λ/T_d from (2.23) are computed from data fits.

Predictions of MCT have been tested both numerically and experimentally using different approaches [38, 55, 6]. Experimental techniques include neutron or lightscattering, dielectric spectroscopy and analysis of a mechanical response to external perturbations [55]. Numerical simulations divide into two classes: molecular dynamics simulations and Monte-Carlo simulations. In the first case on tries to solve directly the dynamic equation (2.1) governing the evolution of a system. This allows in principle to compute various characteristics of the system from first principles and it is the greatest advantage of this approach. At the same time such simulations require immense computational resources because a reliable solution of equation (2.1) over extended time ranges is requires. The second method, Monte-Carlo simulations, does not require a solution of dynamic equations but a different problem arises: how to equilibrate a system effectively for low temperatures if one is interested in equilibrium dynamics ?

Typical tests aim at computing certain quantities or establishing some properties of a system:

- Position of the transition *i.e.* identify T_d from data analysis. As anticipated this is done by data fit.
- Direct measurement of $f_{\mathbf{q}}$ [38, 55].
- Check of the β-scaling, factorisation property and relation (2.23). This is achieved by tracing curves of correlators at various wave-vectors and collapsing them on a single master curve [55, 6].
- Check of the α-relaxation time τ_α divergence and time-temperature superposition principle [55].
- Self-diffusion: the average $\langle r^2(t)\rangle$ presents a diffusive behaviour for high temperatures but deviates from Dt plot for low temperatures where a plateau emerges indicating caging. This is a direct evidence for cage effect. A distribution of displacements δr is also studied via measurement of the van Hove function defined as:

  $$G(r,t) = \frac{1}{N}\sum_k \langle \delta(r - |r_k(t) - r_k(0)|)\rangle.$$

  A useful information is also provided by a non-Gaussian parameter

  $$\alpha_2 = \frac{3\langle \delta r^4\rangle}{5\langle \delta r^2\rangle^2} - 1$$

which is zero identically for Gaussian distributions.

- Multi-point correlations: a direct experimental test of the growing correlation length was performed in [9].

Results of these tests lead to the following classification of successes and failures of MCT:
Successes:

- MCT captures the two step relaxation pattern.
- The scaling predicted in β-regime and time-temperature superposition in α-regime work well. The value of exponents a and b is qualitatively correct: $0 < a < 0.5$ and $b < 1$.
- Wave-vector dependence of the non-ergodic parameter $f_{\mathbf{q}}$ predicted by MCT is reproduced experimentally and numerically [55]. Fig. 2.1 shows a curve $f_{\mathbf{q}}$ *vs* $\mathbf{q}$ resulting from molecular dynamics simulations for silica [56].
- The growing lengthscale is extracted from the four-point function [52, 50, 9, 46, 57, 58, 59] although it shows only a mild increase.

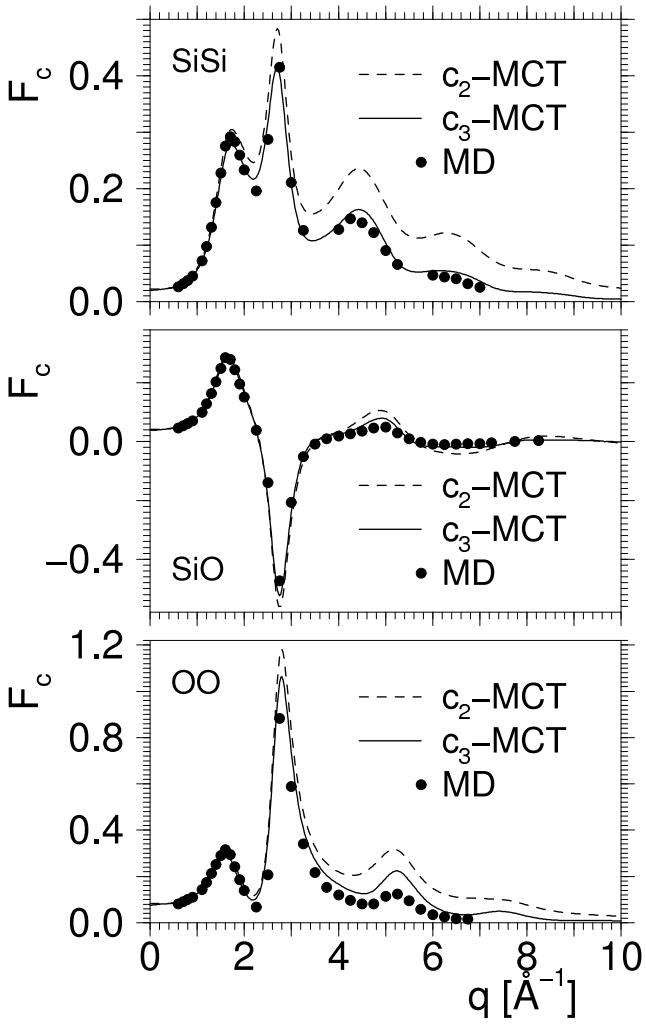
Failures:

- The absence of the dynamic transition in real systems; first principle computation of T_d and data fits (like the above mentioned T_g) differs by a factor 2.
- Self-diffusion described by $\langle r^2 \rangle$ fits well into the MCT image, but the distribution provided by the van Hove function $G(r,t)$ does not. A peak predicted for $\alpha_2(t)$ for intermediate times within MCT [60] show a less pronounced growth than observed experimentally and in numerical simulations [61, 62, 63].
- Decoupling of self-diffusion coefficient D and viscosity η. In a high temperature liquid they are related by the Stokes-Einstein relation stating that $D\eta/T$ is a constant. However this ratio shows a fast growth for temperatures below the onset of slow dynamics [43, 64]. Standard MCT fails to account for this sharp increase [6, 65]. A possible explanation is the arrival of critical fluctuations [66] which are completely neglected within MCT as we will see later.

In general there is a time window where data fit quite well with MCT predictions if the temperature is not too close to T_d *i.e.* for finite ϵ. This is controversial since the predictions are only valid asymptotically as $\epsilon \to 0$.

The cut off of the dynamic transition is usually attributed to activated events. The transition is interpreted as a transition to a non-ergodic phase where the systems gets blocked in an amorphous configuration. However it cannot freeze in such a configuration forever: finally a rearrangement arrives that

Figure 2.2: Non-ergodic parameter for silica, comparison of molecular dynamic simulations (points) and MCT predictions (c_2 - a basic version, c_3 - a more refined version in which static three-point function is accounted for in the factorisation approximation). Reproduced from [56].



brings the system into a different state. This reflects the fact that configurations are separated by a finite energy barriers. Now if a flow-like relaxation is no more possible it is substituted by the relaxation via rearrangements or activated events (because the system has to overcome a barrier). These events smooth the transition into a crossover between different relaxation mechanisms.

2.7 Extending MCT

The problems with interpretation of experimental data and numerical simulations within MCT led to the introduction of several extensions of MCT that tried to address the difficulties and take into account the activated events.

2.7.1 Conserved quantities and Extended MCT

A possible origin of breakdown of MCT is that we have not taken into account all the slow variables in the system. Recall that in Sec. 2.2 we assumed that the only slow variables were the density and the longitudinal current. As we noted in Sec. 2.2 the longitudinal current j^L has been related to density by a conservation law. A careful inspection tells that the same statement holds for all conserved quantities coupled to density. Then we should treat all these quantities as "slow" variables. Practically this means that all conserved quantities should be treated as a "slow" variables because of the non-linear couplings between them.

A model extending MCT by considering a larger set of conserved/slow variables was first introduced by Das and Mazenko [28] in the context of a field theory based on the Non-linear Fluctuating Hydrodynamics (FNH) which we will consider later in Chapter 4. Das and Mazenko argued that FNH reproduced MCT in the lowest order of perturbation theory but corrections coming from coupling to transverse currents (neglected in the derivation of Sec. 2.2) which appear in higher orders of perturbation cut off the transition [28, 40, 38]. Therefore they stated that coupling to currents was responsible for cut off of the transition.

These results were later reproduced in the context of the projection operator formalism and the theory known as extended MCT (eMCT) [34, 67]. The derivation follows closely that presented in Sec. 2.2 but the set of "slow" variables is enhanced to include two transverse currents $\mathbf{j}^T$ in addition to the density and longitudinal current j^L. The derivation simplifies if one considers the phase space density function which generates all these quantities instead:

$$\phi(\mathbf{r},\mathbf{p},\tau) = \sum_l \delta(\mathbf{r}-\mathbf{r}_l(\tau))\delta(\mathbf{p}-\mathbf{p}_l(\tau)) \tag{2.27}$$

$$\phi(\mathbf{q},\mathbf{p},\tau) = \sum_l \exp(i\mathbf{q}\cdot\mathbf{r}_l(\tau))\delta(\mathbf{p}-\mathbf{p}_l(\tau)) \tag{2.28}$$

Now density and the currents read:

$$\rho(\mathbf{r},t) = \frac{1}{N}\int \frac{d^3\mathbf{p}}{(2\pi)^3}\phi(\mathbf{r},\mathbf{p},t)$$
$$\mathbf{j} = \frac{1}{N}\int d^d\mathbf{r}\,\mathbf{p}\,\phi(\mathbf{r},\mathbf{p},t)$$

Repeating the steps of Sec. 2.1 an equation (2.10) for the phase space density correlator is derived:

$$C_{\mathbf{q}}(\mathbf{p}_1,\mathbf{p}_2;\tau) = \langle\phi_{-\mathbf{q}}(\mathbf{p}_1,\mathbf{p}_2;\tau)\phi_{\mathbf{q}}(\mathbf{p}_1,\mathbf{p}_2;\tau)\rangle \quad (2.29)$$
$$\partial_\tau C_{\mathbf{q}}(\mathbf{p}_1,\mathbf{p}_2;\tau) = \int \frac{d^3\mathbf{k}}{(2\pi)^3}\Omega_{\mathbf{q}}(\mathbf{p}_1,\mathbf{k})C_{\mathbf{q}}(\mathbf{k},\mathbf{p}_2;\tau) - \quad (2.30)$$
$$- \int_0^\tau du\int \frac{d^3\mathbf{k}}{(2\pi)^3}M_{\mathbf{q}}(\mathbf{p}_1,\mathbf{k};\tau-u)\partial_u C_{\mathbf{q}}(\mathbf{k},\mathbf{p}_2;u)$$

The phase space density correlator $C_{\mathbf{q}}(\mathbf{p}_1,\mathbf{p}_2;\tau)$ encodes extra information we are not interested in, like energy correlations, *etc*: our final goal is to derive equations on density and current correlators only. To simplify the above equation and to separate explicitly density and current fluctuations, the momentum distribution $\phi_{\mathbf{q}}(\mathbf{q},\tau)$ is substituted by its irreducible moments [34]: $\phi_{\mathbf{q}}(\mathbf{p},\tau) \to \phi^\alpha_{\mathbf{q}}(\tau)$ labelled by index $\alpha = 0,1,2,\cdots$. The first four moments is the density and the three components of the current correspondingly, the fifth moment is the energy. Now Eq. (2.30) transforms into a set of equations for irreducible moments:

$$C_{\alpha\beta}(\mathbf{q},\tau) = \langle\phi^\alpha_{-\mathbf{q}}\phi^\beta_{\mathbf{q}}(\tau)\rangle \quad (2.31)$$
$$\partial_\tau C_{\alpha\beta}(\mathbf{q},\tau) = \sum_\gamma \Omega_{\alpha\gamma}(\mathbf{q})C_{\gamma\beta}(\mathbf{q},\tau) - \sum_\gamma\int_0^\tau M_{\alpha\gamma}(\mathbf{q},\tau-u)\partial_u C_{\gamma\beta}(\mathbf{q},u) \quad (2.32)$$

This is an infinite set of equations that is too hard to analyse. We assume several simplifications. First, only the first four tensors are retained; the others are neglected *i.e.* they are assumed to be irrelevant. This limits the indices to the range $[0,1,2,3]$ and leaves us only with 16 equations. Second, a Mode-Coupling factorisation similar the one presented in Sec. 2.2 is used to compute the kernels $M_{\alpha\beta}$:

$$M_{\alpha\beta}(\mathbf{q},\tau) = \sum_{\alpha_1\beta_1\alpha_2\beta_2}\sum_{\mathbf{k}_1,\mathbf{k}_2} V^{\alpha_1\beta_1\alpha_2\beta_2}_{\alpha\beta}(\mathbf{q},\mathbf{k}_1,\mathbf{k}_2;\tau)C_{\alpha_1\beta_1}(\mathbf{k}_1,\tau)C_{\alpha_2\beta_2}(\mathbf{k}_2,\tau) \quad (2.33)$$

Furthermore, if one neglects all the vertices V except $V^{0000}_{\alpha\beta}$ then (2.32) reduces to (2.19) and reproduces MCT plus a set of decoupled equations ruling the evolution of other correlators. The authors of [34, 67] conjectured that the

vertices V can be ordered with respect to the decay of the corresponding correlators. Namely they assumed that only density-density correlator $C_{00}(\mathbf{q},\tau)$ has a non-trivial two-step relaxation pattern which extends over decades on timescale while the other correlators which contains transverse currents were assumed to show a faster decay: algebraic or exponential [67]. The leading order is obtained by retaining $V^{0000}_{\alpha\beta}$ only. The next to leading order is obtained by retaining $V^{0\gamma 0\delta}_{\alpha\beta}$ and $V^{000\gamma}_{\alpha\beta}$. After some algebra [34, 67] a unique equation on density-density correlator is derived which generalises Eq. (2.20):

$$\frac{T\hat{C}(\mathbf{q},z)}{S(\mathbf{q})-[z+\Delta(\mathbf{q})]\hat{C}(\mathbf{q},z)}=\frac{\kappa_{\mathbf{q}}}{\Omega^2_{\mathbf{q}}}\hat{M}_{sl}(\mathbf{q},z) \tag{2.34}$$

where Δ has contributions from the vertices $V^{0\gamma 0\delta}_{\alpha\beta}$, $V^{000\gamma}_{\alpha\beta}$. Therefore it results from the coupling to transverse currents. Results obtained in the framework of FNH have a similar form [28]. A straightforward check shows that for $\Delta(\mathbf{q})\neq 0$ the equation implies $f_{\mathbf{q}}=C(\mathbf{q},\infty)=0$ for any temperature. Therefore no transition is present. Analysis of (2.34) shows that $\Delta(\mathbf{q})$ introduces an extra timescale into the problem which delimits the region where MCT is valid: for $z\ll 1/\Delta(\mathbf{q})$ one recovers MCT [34, 67]; for $z\gg 1/\Delta(\mathbf{q})$ $\hat{C}(\mathbf{q},z)$ decays from the plateau to zero.

2.7.2 Criticism of eMCT

Nevertheless eMCT does not solve the problem. First, the validity of eMCT has been criticised recently [68, 69]. We will return to this point later in Chapter 4. Second, the eMCT scenario assumes that the mechanism responsible for the cut off of the transition is different for Newtonian dynamics where momentum and energy are conserved and Brownian dynamics where only density is conserved. However numerical simulations indicate no difference with respect to dynamics [61, 71, 70].

2.8 Summary

In the previous sections we introduced the Mode-Coupling theory and presented its main predictions. We also discussed its successes and failures in the description of the slowing down in supercooled liquids. The problem of the spurious dynamic transition predicted within MCT motivated a research for extensions of MCT. Therefore we considered an extension taking into account a larger set of slow variables as compared to MCT. The results derived within eMCT indicate the cut off of the transition. However this assumes that different mechanisms smooth the transition for Newtonian and Brownian dynamics. This conclusion is not confirmed neither experimentally nor numerically. This together with several other issues forms a (incomplete) list of problems that we try to address in the following chapters:

- The Mode-Coupling Factorisation lacks a small parameter and represents a non-perturbative approximation. It is unclear how to improve it and to generate systematic corrections. Existence of a model within which this factorisation is perturbative would shed some light on the approximation. Precisely this would allow to analyse the stability of MCT with respect to corrections. This analysis would indicate if the transition is cut off by higher order corrections to MCF or by a different mechanism.

- The problem with eMCT indicates that something is wrong with it. As we will see this is related to the violation of time reversal symmetry assumed within eMCT. It is not clear how to control the preservation of the symmetry within projection operator formalism. This requires a development of a different formalism which allows to control the preservation of TRS perturbatively.

- Time reversal symmetry lies at the origin of a different problem with standard MCT: the description presented in Sec. 2.2 fails below the transition. Time reversal symmetry and fluctuation-dissipation theorem assumed within MCT no more hold in the low temperature phase if one follows the non-ergodic interpretation of the transition. This requires a complete rederivation of MCT without assumption of equilibrium [29].

Appendix A. Derivation of the full MCT equation

We present below the detailed derivation of (2.12) that follows the guidelines of Section 2.1. We are interested in deriving equation (2.10) in the framework of MCT of density fluctuations. It is a matrix equation however we concentrate on the lower left element since it will lead to an equation on density-density correlator (2.12). The matrix of the correlations reads:

$$\mathbf{C}(\tau) = \begin{pmatrix} \langle \delta\rho_{-\mathbf{q}} \delta\rho_{\mathbf{q}}(\tau)\rangle & \langle \delta\rho_{-\mathbf{q}} j^L_{\mathbf{q}}(\tau)\rangle \\ \langle \delta \mathrm{j}^L_{-\mathbf{q}} \delta\rho_{\mathbf{q}}(\tau)\rangle & \langle j^L_{-\mathbf{q}} j^L_{\mathbf{q}}(\tau)\rangle \end{pmatrix}$$

At zero times it simplifies to:

$$\mathbf{C} = \begin{pmatrix} NS(q) & 0 \\ 0 & \frac{NT}{m} \end{pmatrix}$$

Below we compute (2.10) term by term. We start from $i\Sigma$:

$$i\Omega = \langle \mathbf{A}^* \dot{\mathbf{A}}\rangle \cdot \langle \mathbf{A}^* \mathbf{A}\rangle^{-1} = \begin{pmatrix} \langle \delta\rho_{-\mathbf{q}} \delta\dot{\rho}_{\mathbf{q}}\rangle & \langle \delta\rho_{-\mathbf{q}} \frac{dj^L_{\mathbf{q}}}{d\tau}\rangle \\ \langle j^L_{-\mathbf{q}} \delta\dot{\rho}_{\mathbf{q}}\rangle & \langle \frac{dj^L_{-\mathbf{q}}}{d\tau} \delta\dot{\rho}_{\mathbf{q}}\rangle \end{pmatrix} \cdot \begin{pmatrix} \frac{1}{NS(q)} & 0 \\ 0 & \frac{m}{NT} \end{pmatrix} =$$
$$\begin{pmatrix} 0 & i\frac{NqT}{m} \\ i\frac{NqT}{m} & 0 \end{pmatrix} \cdot \begin{pmatrix} \frac{1}{NS(q)} & 0 \\ 0 & \frac{m}{NT} \end{pmatrix} = \begin{pmatrix} 0 & iq \\ i\frac{qT}{mS(q)} & 0 \end{pmatrix}$$

where we used the fact that $\langle A\dot{A}\rangle = 0$, integration by parts and the identity:

$$\langle j^L_{-\mathbf{q}}\delta\dot{\rho}_{\mathbf{q}}\rangle = \frac{i}{m^2}\sum_{l,n}\left\langle(\hat{\mathbf{q}}\cdot\mathbf{p}_l)e^{-i\mathbf{q}\cdot\mathbf{r}_l}(\mathbf{q}\cdot\mathbf{p}_n)e^{-i\mathbf{q}\cdot\mathbf{r}_n}\right\rangle = \frac{iq}{m}\sum_l\langle\mathbf{p}_l^2\rangle = i\frac{NqT}{m}$$

The fluctuating force at zero time reads:

$$\mathbf{f}(0) = i\mathcal{Q}\mathcal{L}\mathbf{A} = (1-\mathcal{P})\dot{\mathbf{A}} = \begin{pmatrix}\delta\dot{\rho}_{\mathbf{q}}\\ \frac{dj^L_{\mathbf{q}}}{du}\end{pmatrix} - \begin{pmatrix}0 & iq\\ i\frac{qT}{mS(q)} & 0\end{pmatrix}\cdot\begin{pmatrix}\delta\rho_{\mathbf{q}}\\ j^L_{\mathbf{q}}\end{pmatrix} = \begin{pmatrix}0\\ \frac{dj^L_{\mathbf{q}}}{du} - i\frac{qT}{mS(q)}\delta\rho_{\mathbf{q}}\end{pmatrix} \equiv \begin{pmatrix}0\\ R_{\mathbf{q}}\end{pmatrix}$$

Then:

$$i\Omega\cdot\mathbf{C}(\tau) = \begin{pmatrix}0 & iq\\ i\frac{qT}{mS(q)} & 0\end{pmatrix}\cdot\begin{pmatrix}\langle\delta\rho_{-\mathbf{q}}\delta\rho_{\mathbf{q}}(\tau)\rangle & \langle\delta\rho_{-\mathbf{q}}j^L_{\mathbf{q}}(\tau)\rangle\\ \langle\delta\mathrm{j}^L_{-\mathbf{q}}\delta\rho_{\mathbf{q}}(\tau)\rangle & \langle j^L_{-\mathbf{q}}j^L_{\mathbf{q}}(\tau)\rangle\end{pmatrix}$$

The memory matrix reads (see the definition (2.9)):

$$M(\mathbf{q},\tau) = \left\langle\begin{pmatrix}0\\ R_{\mathbf{q}}\end{pmatrix}\cdot\begin{pmatrix}0 & R_{\mathbf{q}}(\tau)\end{pmatrix}\right\rangle\cdot\begin{pmatrix}\frac{1}{NS(q)} & 0\\ 0 & \frac{m}{NT}\end{pmatrix} = \begin{pmatrix}0 & 0\\ 0 & \frac{m}{NT}\langle R_{-\mathbf{q}}R_{\mathbf{q}}(\tau)\rangle\end{pmatrix}$$

Now taking the lower left element of (2.10) we arrive at (2.12):

$$\frac{d^2C(\mathbf{q},\tau)}{d\tau^2} + \frac{\mathbf{q}^2T}{mS_{\mathbf{q}}}C(\mathbf{q},\tau) + \frac{m}{NT}\int_0^\tau du\langle R_{-\mathbf{q}}R_{\mathbf{q}}(\tau-u)\rangle\frac{dC(\mathbf{q},u)}{du} = 0$$

Appendix B. Factorisation approximation

We should compute:

$$V_{\mathbf{q}}(\mathbf{k}_1,\mathbf{k}_2) = \sum_{\mathbf{k}_3,\mathbf{k}_4}\langle\delta\rho_{\mathbf{k}_1}\delta\rho_{\mathbf{k}_2}R_{\mathbf{q}}\rangle\cdot\langle\delta\rho_{\mathbf{k}_1}\delta\rho_{\mathbf{k}_2}\delta\rho_{\mathbf{k}_3}\delta\rho_{\mathbf{k}_4}\rangle^{-1}$$

The four-point function in the denominator is factorised in a product of two-point functions. To compute the nominator we recall the expression for $R_{\mathbf{q}}$:

$$R_{\mathbf{q}} = \frac{dj^L_{\mathbf{q}}}{d\tau} - i\frac{qT}{mS_{\mathbf{q}}}\delta\rho_{\mathbf{q}}$$

Then the following terms appear in the numerator (where we have used the integration by parts):

$$\left\langle \delta\rho_{-\mathbf{k}}\delta\rho_{\mathbf{k}-\mathbf{q}}\frac{dj^L_{\mathbf{q}}}{d\tau}\right\rangle = -\langle\delta\dot{\rho}_{-\mathbf{k}}\delta\rho_{\mathbf{k}-\mathbf{q}}j^L_{\mathbf{q}}\rangle - \langle\delta\rho_{-\mathbf{k}}\delta\dot{\rho}_{\mathbf{k}-\mathbf{q}}j^L_{\mathbf{q}}\rangle$$
$$-\frac{iqT}{mS(q)}\langle\delta\rho_{-\mathbf{k}}\delta\rho_{\mathbf{k}-\mathbf{q}}\delta\rho_{\mathbf{q}}\rangle$$

The first two terms can be computed exactly:

$$-\langle\delta\dot{\rho}_{-\mathbf{k}}\delta\rho_{\mathbf{k}-\mathbf{q}}j^L_{\mathbf{q}}\rangle = i\langle\sum_{jln}(\mathbf{k}\cdot\mathbf{p}_j)e^{-i\mathbf{k}\cdot\mathbf{r}_j}e^{i(\mathbf{k}-\mathbf{q})\cdot\mathbf{r}_l}(\hat{\mathbf{q}}\cdot\mathbf{p}_n)e^{i\mathbf{q}\cdot\mathbf{r}_n}\rangle =$$
$$= i(\mathbf{k}\cdot\hat{\mathbf{q}})\frac{T}{m}\sum_{jl}\left\langle e^{i(\mathbf{k}-\mathbf{q})\cdot\mathbf{r}_j}e^{i(\mathbf{q}-\mathbf{k})\cdot\mathbf{r}_l}\right\rangle = i(\mathbf{k}\cdot\hat{\mathbf{q}})\frac{TN}{m}S(\mathbf{k}-\mathbf{q})$$

The second terms is computed similarly to give:

$$-\langle\delta\rho_{-\mathbf{k}}\delta\dot{\rho}_{\mathbf{k}-\mathbf{q}}j^L_{\mathbf{q}}\rangle = i(\hat{\mathbf{q}}\cdot(\mathbf{q}-\mathbf{k}))\frac{TN}{m}S(\mathbf{k}--\mathbf{q})$$

The three point density fluctuation term is factorised within the convolution approximation [33]:

$$\langle\delta\rho_{-\mathbf{k}}\delta\rho_{\mathbf{k}-\mathbf{q}}\delta\rho_{\mathbf{q}}\rangle \approx NS(k)S(q)S(|\mathbf{k}-\mathbf{q}|)$$

and $V_{\mathbf{q}}(\mathbf{k}_1,\mathbf{k}_2)$ simplifies to $V(\mathbf{k},\mathbf{q}-\mathbf{k})$ as a consequence of translation invariance. Combining together the above results yields:

$$V(\mathbf{k},\mathbf{q}-\mathbf{k}) = \frac{i\rho T}{2mN}\left[(\hat{\mathbf{q}}\cdot\mathbf{k})c(k) + \hat{\mathbf{q}}\cdot(\mathbf{q}-\mathbf{k})c(|\mathbf{k}-\mathbf{q}|)\right]$$

where we introduced a direct correlation function $c(k) = \frac{1}{\rho}\left(1-\frac{1}{S_k}\right)$.

Chapter 3

Connection with disordered systems

In previous chapter we introduced the Mode-Coupling theory that has captured some aspects of the slow dynamics in supercooled liquids. However the nature of the Mode-Coupling factorisation remained unclear. A reliable guide for this approximation is necessary for several reasons:

- An uncontrolled closure leading to MCT might violate some physical constraints or symmetries. This will be discussed in relation with standard MCT and eMCT in the next chapters. Such a possibility might also suggest that results derived within MCT/eMCT are artifacts of the approximation.
- Insights into the Mode-Coupling Factorisation would shed more light on the physical origins of the success of MCT.
- The guide would also provide an idea on how to generate a systematic perturbation around MCT which would allow the structural stability test of MCT with respect to corrections.

These points lead to an intensive research for models whose dynamics is exactly described by MCT. Surprisingly such models exist [72, 73, 74]: examples are Potts glasses with number of states greater than 4 and spherical p-spin models on fully connected nets. Their high temperature dynamics are governed by a schematic MCT equation (2.24). In this chapter we introduce the spherical 3-spin model and develop a field theory formalism for this model. The latter allows us to derive dynamic equations and establish the results of 2.3 for the 3-spin model. Finally Sec. 3.4 presents a physical interpretation of the transition in the model.

3.1 The spherical 3-spin model

The model is defined as a set of N continuous spins interacting by triplets (or by p-multiplets in case of p-spin models) on a fully connected network. The bonds have a Gaussian distribution of zero mean. The Hamiltonian reads:

$$H = -\frac{1}{3!}\sum_{k,l,m=1}^{N} J_{klm}\phi_k\phi_l\phi_m \tag{3.1}$$

$$\overline{J_{klm}} = 0, \quad \overline{J_{klm}^2} = \frac{3!}{2N^2}$$

The spins evolve under Langevin dynamics with the Gaussian white noise of zero mean:

$$\partial_t\phi_k = -\frac{\partial H}{\partial\phi_k} - z\phi_k + \xi_k \tag{3.2}$$

$$\langle\xi_k\rangle = 0, \quad \langle\xi_k(t)\xi_l(s)\rangle = 2T\delta_{kl}\delta(t-s) \tag{3.3}$$

Spins are subject to the spherical constraint: $\sum_k \phi_k^2 = N$ that is enforced by a Lagrangian multiplier $z(t)$. Initial conditions are fixed as follows: the system is at equilibrium at $T = \infty$ at $t = 0$ and is quenched to a finite T for $t > 0$. One could start from an equilibrated initial condition but the procedure is more involved [75]. Henceforth thermal average is denoted by angular brackets $\langle\cdots\rangle$ and the average over the disorder is denoted by an overline $\overline{\cdots}$.

A field theory is constructed from the Langevin equation (3.2) by the virtue of the Martin-Siggia-Rose [76] formalism which is a standard method[77]. The derivation is completely standard and yields a field theory for fixed disorder:

$$S_J[\phi,\hat{\phi}] = \int_0^\infty dt\left[\sum_{k=1}^{N}\left(T\hat{\phi}_k^2 + \hat{\phi}_k(\partial_t + z(t))\phi_k\right) - \sum_{jkl} J_{jkl}\hat{\phi}_j\phi_k\phi_l(t)\right] \tag{3.4}$$

We would like to carry out the average over disorder J. This average is very delicate if one computes static quantities where it requires introduction of replicas [78]. In the dynamic case this is not always necessary [30, 79]. The average of a quantity $A[\hat{\phi},\phi]$ reads:

$$\overline{\langle A\rangle} = \frac{\int dJ\mathcal{P}(J)\int D\hat{\phi}D\phi A[\hat{\phi},\phi]e^{-S_J}}{\int dJ\mathcal{P}(J)\int D\hat{\phi}D\phi e^{-S_J}}.$$

If initial conditions are uncorrelated with disorder there is no need to introduce replicas: the generating functional which leads to the above expression

for averages is constructed from a path integral which is independent of disorder [30]. Therefore the denominator is constant and can be absorbed in the normalisation. Averages can be simply computed from

$$\overline{Z} = \int DJ\mathcal{P}(J) \int D\hat{\phi}D\phi e^{-S_J}.$$

But if the initial conditions are correlated with the disorder, then one has to introduce replicas and work with replicated dynamic quantities. Our protocol provides initial conditions uncorrelated with disorder so that the average over J is just a Gaussian integration over the couplings J_{ijk}. The action S for a field theory averaged over disorder reads:

$$\begin{aligned} S[\phi,\hat{\phi}] = S_2 + S_{INT} = \sum_{k=1}^{N} \int_0^\infty dt[T\hat{\phi}_k^2 + \hat{\phi}_k(\partial_t + z(t))\phi_k] - \\ -\frac{3}{4N^2}\sum_{klm}\int_0^\infty dt \int_0^\infty ds\,(\hat{\phi}_k\phi_l\phi_m)(t)(\hat{\phi}_k\phi_l\phi_m)(s) - \\ -\frac{3}{2N^2}\sum_{klm}\int_0^\infty dt \int_0^\infty ds\,(\hat{\phi}_k\phi_l\phi_m)(t)(\hat{\phi}_l\phi_k\phi_m)(s) \end{aligned} \tag{3.5}$$

$$S_2 = \sum_{k=1}^{N} \int_0^\infty dt[T\hat{\phi}_k^2 + \hat{\phi}_k(\partial_t + z(t))\phi_k] \tag{3.6}$$

$$\begin{aligned} S_{INT} = -\frac{3}{4N^2}\sum_{klm}\int_0^\infty dt \int_0^\infty ds\,(\hat{\phi}_k\phi_l\phi_m)(t)(\hat{\phi}_k\phi_l\phi_m)(s) - \\ -\frac{3}{2N^2}\sum_{klm}\int_0^\infty dt \int_0^\infty ds\,(\hat{\phi}_k\phi_l\phi_m)(t)(\hat{\phi}_l\phi_k\phi_m)(s) \end{aligned} \tag{3.7}$$

where we introduced a perturbation expansion in powers of $1/N$ where N is the total number of spins: the quadratic terms in $\hat{\phi}$,ϕ define the Gaussian part S_2 and the other terms compose an interaction S_{INT}. Note that the average over the disorder leads to an interaction non-local in time. Let's present some properties of the field theory. First, Eq. (3.2) is invariant under a local gauge symmetry [80]: changing the sign of a single bond J_{klm} in (3.2) is compensated by a flip of the spin ϕ_k associated to that bond. This implies that the one-point quantities $\overline{\langle\phi_k\rangle}$ and $\overline{\langle\hat{\phi}_k\rangle}$ are zero identically; the two-point functions $\overline{\langle\phi_k\phi_l\rangle}$ and $\overline{\langle\phi_k\hat{\phi}_l\rangle}$ have a diagonal index structure:

$$\begin{aligned} \overline{\langle\phi_k\phi_l\rangle} &= \delta_{kl}\overline{\langle\phi_k\phi_k\rangle} \\ \overline{\langle\phi_k\hat{\phi}_l\rangle} &= \delta_{kl}\overline{\langle\phi_k\hat{\phi}_k\rangle} \end{aligned}$$

and the diagonal elements do not depend on the index value. These identities hold in perturbation theory order by order. First, let's note that the Gaussian theory given by S_2 has a diagonal index structure. Second, an attempt to construct a diagram contributing to $\langle\overline{\phi_k\phi_l}\rangle$ or $\langle\overline{\phi_k\hat{\phi}_l}\rangle$ with $k \neq l$ leads to the appearance of bare lines $\langle\overline{\phi_m\phi_n}\rangle_0$ or $\langle\overline{\phi_m\hat{\phi}_n}\rangle_0$ with $m \neq n$ within a diagram (Here index 0 stands for a Gaussian average with S_2). This is ensured by the vertices of the interaction S_{INT}. Then, we can define the following two-point functions:

$$\begin{aligned} C(t,s) &= \langle\overline{\phi_k(t)\phi_k(s)}\rangle \\ R(t,s) &= \langle\overline{\phi_k(t)\hat{\phi}_k(s)}\rangle \\ \overline{R}(t,s) &= \langle\overline{\hat{\phi}_k(t)\phi_k(s)}\rangle \\ Q(t,s) &= \langle\overline{\hat{\phi}_k(t)\hat{\phi}_k(s)}\rangle \end{aligned} \tag{3.8}$$

where C is the spin autocorrelation function and R is the response function (This is easily verified by adding a magnetic field to (3.1)) and by definition $\overline{R}(t,s) = R(s,t)$). The response functions have the usual causal structure: $R(t,s) \equiv 0 \quad t < s$, $\overline{R}(t,s) \equiv 0 \quad t > s$ and $Q(t,s) \equiv 0$. Clearly this is true for the Gaussian theory given by S_2. This together with the causal structure of the interaction S_{INT} *i.e.* the fact that all vertices in Eq. (3.7) contain at least one field $\hat{\phi}$ ensures that causality is preserved order by order in perturbation theory.

3.2 Self-consistent perturbation theory

The bare correlators implied by Eq. (3.6) show a simple exponential relaxation at any temperature. The non-trivial dynamics appearing as the temperature is lowered requires resummation of the perturbation series for $C(t,s)$ and $R(t,s)$ in order to analyse their behaviour. This can be done in the context of the Schwinger-Dyson equation which is derived in the context of the standard perturbation theory generated by the splitting of the action S into interaction S_{INT} (3.7) and Gaussian part S_2 (3.6) [77]:

$$G_0^{-1}G = \mathbb{I} + \Sigma[G_0]$$

where G_0 denotes the matrix of bare correlators and G denotes the matrix of dressed correlators; Σ is a self-energy composed of all vacuum diagrams built from the bare correlators with one line amputated. However, this is not sufficient for our purpose since further resummation within Σ is required to access the non-trivial dynamics of C and R. We would like to derive equations which fixe C and R self-consistently in the spirit of the Mode-Coupling Theory. Such equations are provided in the context of the self-consistent perturbation theory where the bare correlator C_0 and response function R_0 are substituted by their

dressed counterparts C and R in perturbation series. The latter are fixed self-consistently by the Schwinger-Dyson (SD) equations:

$$G_0^{-1} G = \mathbb{I} + \Sigma[G]$$

where $\Sigma[G]$ is a self-energy given by the sum of all 2 particle irreducible vacuum diagrams built with dressed lines with one line amputated. A 2 particle irreducible diagram remains connected under amputation of any two lines. The construction of the perturbation theory is presented in Appendix as well as the derivation of SD equations. The SD equations for the 3-spin case read:

$$\begin{aligned}
(\partial_t + z(t))C(t,s) &= \int_0^t du \Sigma_{\hat{\phi}\phi}(t,u)C(u,s) + \int_0^s du \Sigma_{\hat{\phi}\hat{\phi}}(t,u)R(s,u) \\
(\partial_t + z(t))R(t,s) &= \int_s^t du \Sigma_{\hat{\phi}\phi}(t,u)R(u,s)
\end{aligned} \tag{3.9}$$

where we set $t > s$ so that the δ-function terms dropped out; the self-energies $\Sigma_{\hat{\phi}\phi}$ and $\Sigma_{\hat{\phi}\hat{\phi}}$ are given by a sum of all 2PI vacuum diagrams with one line cut off ($\overline{R}$ and Q respectively, see Appendix for details). The vertices of the interaction are still given by S_{INT}. It is worth mentioning here that the full SD equations (with no constraints on the values of t, s) admit non-causal solutions with $Q(t,s) \neq 0$ [19] although none is known so far.

The structure of Eq. (3.7) suggests a perturbation expansion in powers of $1/N$. Naively, the order of a diagram in $1/N$ is determined by the number of vertices since each vertex contributes a factor $1/N^2$. However the index structure of the vertices also contributes. Two point correlators have diagonal index structure and do not depend on indices. This fixes certain indices and summation over the other indices gives a factor N^f where f is the number of unfixed indices per vertex. It turns out that by an appropriate choice of the topology of a diagram one can compensate these two contributions and construct a diagram of a given order in $1/N$ with arbitrarily large number of vertices. Therefore any order in $1/N$ contains infinitely many diagrams so that a partial resummation is necessary to get a useful expansion in $1/N$. This is a difficult problem in general. The only exception is the lowest order in $1/N$: there are only two diagrams of order N and they are constructed from a single vertex (see Fig. 3.2).

For high enough temperatures, time translation invariance (TTI) and time reversal symmetry (TRS) (and consequently fluctuation-dissipation theorem (FDT)) hold: $R(t,s) = 1/T \partial_s C(t,s)$ [31]. Therefore there is a single independent two-point function. A low temperature phase with different properties will be considered later in Chapter 7. A limit $t, s \to \infty$ with fixed $\tau = t - s$ should be taken in perturbation expansions in order to forget the initial conditions at $t = 0$. Under this assumptions the SD equations (3.9) reduce to a single equation:

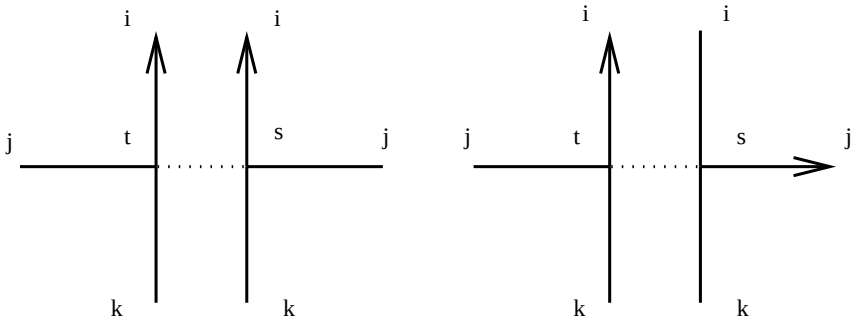


Figure 3.1: Vertices of the 3-spin model. Arrow lines correspond to the field $\hat{\phi}$, the simple lines correspond to the field ϕ; t and s denote times.

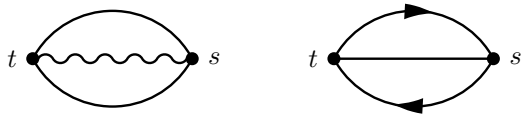


Figure 3.2: The lowest order diagrams in perturbation theory. The wiggly line denotes Q, simple line denotes C and directed lines stand for R.

$$(\partial_\tau + z(\infty))C(\tau) = \frac{1}{T}\left[\Sigma(0)C(\tau) - \int_0^\tau du\Sigma(\tau-u)\partial_u C(u)\right] \tag{3.10}$$

where we introduced $\Sigma \equiv \Sigma_{\hat{\phi}\hat{\phi}}$[1]. The constant $z(\infty)$ is eliminated with the help of the $\tau \to 0$ limit of the above equation: $z(\infty) = T + \Sigma(0)/T$. Then (3.10) reads:

$$(\partial_\tau + T)C(\tau) = -\frac{1}{T}\int_0^\tau du\Sigma(\tau-u)\partial_u C(u) \tag{3.11}$$

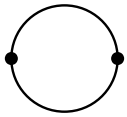
or after Laplace transform:

$$\hat{f}(z) = \mathcal{L}[f] = \int_0^\infty d\tau f(\tau)\exp(-z\tau)\frac{T^2\hat{C}(z)}{1-z\hat{C}(z)} = T + \hat{\Sigma}(z) \tag{3.12}$$

Note that it looks remarkably similar to the schematic equation (2.24). However this time the kernel $\Sigma(\tau)$ admits a systematic expansion in C contrary to the sMCT case.

3.3 Local self-energy approximation.

The thermodynamic limit $N \to \infty$ suppresses all the diagrams in the expansion of Σ except a single one - the left diagram on Fig. 3.2:

$\Sigma =$  $= \frac{3}{2}C^2(\tau)$ (3.13)

Then (3.11) reads:

$$(\partial_\tau + T)C(\tau) = -\frac{3}{2T}\int_0^\tau du C^2(\tau-u)\partial_u C(u) \tag{3.14}$$

$$\frac{T^2\hat{C}(z)}{1-z\hat{C}(z)} = T + \frac{3}{2}\mathcal{L}\left[C^2\right] \tag{3.15}$$

This is the main result of the section: equations (3.14)/(3.15) are mathematically identical to (2.24) up to a redefinition of constants. Thus, the thermodynamic limit $N \to \infty$ of the 3-spin model exactly reproduces sMCT. The same statement holds for a generic polynomial expression of Σ:

[1] This is different from [30, 31] where the same quantity is denoted as D

$$\Sigma(\tau) = \Sigma[C(\tau)] = \sum_{p>1} \frac{p}{2} H_p^2 C^p(\tau) \tag{3.16}$$

where the underlying microscopic model is a generalised p-spin model:

$$\mathcal{H} = -\sum_{p>2} \frac{H_p}{p!} \sum_{k_i=1, i=1..p}^{N} J_{k_1 k_2 \cdots k_p} \phi_{k_1} \cdots \phi_{k_p}.$$

3.4 Free-energy landscape interpretation

Analysis of the statics and the free energy landscape of the 3-spin model provided an insight on physics underlying the MCT transition [74]. A careful analysis of the 3-spin model revealed existence of two distinct transitions: a dynamic one at $T = T_d$ which is the schematic MCT transition and a static one at $T = T_c$. The slowing down of the dynamics close to T_d is caused by the appearance of a multiplicity of marginally stable states of the free energy $\mathcal{F}$; it takes more and more time for the system to explore the phase space landscape. Finally below $T < T_d$ the barriers between different metastable states diverge and phase space splits into disconnected domains: the system gets trapped in a particular state. The value of the non-ergodic parameter $f = \lim_{\tau\to\infty} C(\tau)$ (Edwards-Anderson parameter in the spin glass terminology) corresponds to the average overlap between the microscopic states inside the same domain. The static transition at T_c is usually associated with the Kauzmann temperature T_K.

3.5 Summary

The mapping between spin glasses and structural glasses represented by MCT has several implications:

- Existence of models for which MCT is exact is a good sign. It explains the partial success of MCT in capturing the slow dynamics and suggests that MCT does not violate any physical constraints.
- The Mode-Coupling Factorisation is just the Mode-Coupling approximation (MCA) in the context of the 3-spin model. The former is well-known in critical dynamics [81]. Thus, one may hope that the full MCT can also be derived as an MCA within some density fluctuations theory.
- We only considered the lowest order of perturbation theory for the 3-spin model. The self-consistent perturbation theory provides a systematic way to compute higher order corrections to Eq. (2.24). Therefore it provides a framework for the structural stability test of MCT.
- The absence of a similar mapping for eMCT suggests that something is wrong with eMCT. As we will show in the next chapter, eMCT [28] violates time reversal symmetry.

- MCT is not valid at low temperatures *i.e.* below a transition where time translation invariance and time reversal symmetry are broken. The mapping provides an insight into possible extensions of MCT to the low-T phase. The 3-spin model considered above is well-defined for all temperatures but as we will see in Chapter 7 the above analysis should be completely reconsidered.

We cover these points one by one in the following chapters.

Appendix. Second Legendre Transform and self-consistent perturbation theory

The derivation of the dynamical equations which lead to sMCT require the introduction of the Second Legendre Transform [82, 83]. The latter allows to construct the correct perturbation theory in terms of the dressed correlators and it is in this sense self-consistent. It is worth noticing that higher order Legendre Transforms are possible leading to a more elaborated perturbation theories including dressed vertices. The main idea is to reformulate the problem using the variational approach. In this subsection we present in detail the Second Legendre Transform and construct the self-consistent perturbation theory for the 3-spin model.

To simplify the notations we introduce the column vector $\Psi(t) = (\phi(t) \quad \hat{\phi}(t))^T$ and general vertices $A_k(t_1, \dots t_k), k = 1, 2, \dots$ which are supposed to be non-zero. We also introduce:

$$\mathbf{m}(t) = \langle \Psi(t) \rangle = \begin{pmatrix} \overline{\langle \phi \rangle} & \overline{\langle \hat{\phi} \rangle} \end{pmatrix}^T$$
$$\mathbf{G}(t,s) = \langle \Psi(t)\Psi(s) \rangle = \begin{pmatrix} C(t,s) & R(t,s) \\ \overline{R}(t,s) & Q(t,s) \end{pmatrix}$$

Then the action (3.5) reads:

$$S = -\sum_k \frac{1}{k!} \int \left[\prod_{l=1}^{k} dt_l \right] A_k(t_1, \cdots t_k) \prod_{n=1}^{k} \Psi(t_n),$$
$$Z[A] = \int D\Psi e^{-S}, \quad W[A] = \log Z[A]$$

Let's use the definition of $W[A]$ to write the following equations which fix $\mathbf{m}$ and $\mathbf{G}$:

$$\frac{\delta W}{\delta A_1(t)} = \mathbf{m}(t), \quad \frac{\delta W}{\delta A_2(t,s)} = \frac{1}{2}(\mathbf{G}(t,s) - \mathbf{m}(t)\mathbf{m}(s)) \tag{3.17}$$

The main problem is to find the values of $\mathbf{m}$ and $\mathbf{G}$ given the vertices $\{A_k\}$. To reformulate it as a variational principle we introduce Γ as the functional of $\mathbf{m}$ and $\mathbf{G}$:

$$\Gamma[A] = W[A] - \int dt\ \mathbf{m}(t)A_1(t) - \iint dt\,ds\ \frac{1}{2}(\mathbf{G}(t,s) - \mathbf{m}(t)\mathbf{m}(s))A_2(t,s) \quad (3.18)$$

where we inverted the equations (3.17) to define A_1, A_2 as functions of $\mathbf{m}, \mathbf{G}$. This transformation corresponds to the passage from the bare averages and correlators to the dressed averages $\mathbf{m}$ and correlators $\mathbf{G}$ as independent variables. To fix the latter we derive Γ with respect to $\mathbf{m}, \mathbf{G}$. This yields [82, 83]:

$$\frac{\delta\Gamma}{\delta\mathbf{m}} - 2\mathbf{m}\frac{\delta\Gamma}{\delta\mathbf{G}} = -A_1 \quad (3.19)$$

$$2\frac{\delta\Gamma}{\delta\mathbf{G}} = \mathbf{G}^{-1} + \mathbf{\Sigma} = -A_2 = \mathbf{G}_0^{-1} \quad (3.20)$$

where we have denoted the bare propagator as $\mathbf{G}_0$. We also introduced a new quantity $\mathbf{\Sigma}$ usually called self-energy. In these equations A_1, A_2 should be considered as the external sources. This can be reformulated as the variational problem [82, 83] of finding the stationary points of the functional $\Gamma[\mathbf{m}, \mathbf{G}] + \mathbf{m}A_1 + \frac{1}{2}(\mathbf{G} - \mathbf{m}\mathbf{m})A_2$. In order to do that we need some method of reconstructing the functional Γ: for this we can solve the equations (3.19),(3.20) iteratively in the powers of the vertices $\{A_k\}$. This yields for Γ:

$$\Gamma = \frac{1}{2}\mathrm{tr}\log\mathbf{G} + \begin{array}{c}\text{sum of all 2-particle irreducible}\\ \text{diagrams with } \mathbf{G}\text{-line and } \mathbf{G}^{-1}\mathbf{m}\\ \text{tail}\end{array}$$

The $\mathbf{G}^{-1}\mathbf{m}$ tail means that $\mathbf{m}$ is attached directly to the vertex. As it was mentioned above for the 3-spin model gauge invariance imply $\mathbf{m} = 0$ and the above expression simplifies to:

$$\Gamma = \frac{1}{2}\mathrm{tr}\log\mathbf{G} + \begin{array}{c}\text{sum of all 2-particle irreducible}\\ \text{diagrams with } \mathbf{G}\text{-line}\end{array} \quad (3.21)$$

If we denote $\Phi[\mathbf{G}]$ as the set of all 2-particle irreducible (2PI) diagrams then equation (3.20) reads:

$$\int du\mathbf{G_0}^{-1}(t,u)\mathbf{G}(u,s) = \mathbb{I}\times\delta(t-s) + \int du\mathbf{\Sigma}(t,u)\mathbf{G}(u,s) \quad (3.22)$$

$$\mathbf{G}_0(t,s) = \begin{pmatrix} 0 & -\partial_t + \mu(t) \\ \partial_t + \mu(t) & 2T \end{pmatrix} \times \delta(t-s)$$

$$\mathbf{\Sigma}(t,s) = \frac{\delta\Phi[\mathbf{G}]}{\delta\mathbf{G}(t,s)} = \begin{pmatrix} \Sigma_{\phi\phi}(t,s) & \Sigma_{\phi\hat{\phi}}(t,s) \\ \Sigma_{\hat{\phi}\phi}(t,s) & \Sigma_{\hat{\phi}\hat{\phi}}(t,s) \end{pmatrix} \quad (3.23)$$

Each component of $\mathbf{\Sigma}$ is obtained by cutting one of the lines from the diagrams of Γ we get the following expressions for these components:

$$\begin{aligned}
\Sigma_{\phi\phi} &= \text{sum of all 2-particle irreducible diagrams with amputated } C\text{-line} \\
\Sigma_{\phi\hat{\phi}} &= \text{sum of all 2-particle irreducible diagrams with amputated } R\text{-line} \\
\Sigma_{\hat{\phi}\phi} &= \text{sum of all 2-particle irreducible diagrams with amputated } \overline{R}\text{-line} \\
\Sigma_{\hat{\phi}\hat{\phi}} &= \text{sum of all 2-particle irreducible diagrams with amputated } Q\text{-line}
\end{aligned} \tag{3.24}$$

It is easy to show using the causality of theory (3.5) that each 2PI diagram contains either a Q-line either a loop of $R/\overline{R}$-lines. This implies immediately that $\Sigma_{\phi\phi}(t,s) \equiv 0$ in all orders of perturbation theory. For the same reason i.e causality we have: $\Sigma_{\phi\hat{\phi}}(t,s) \equiv 0 \quad t < s$ and $\Sigma_{\hat{\phi}\phi}(t,s) \equiv 0 \quad t > s$. Now there are only two *a priori* independent correlators $C(t,s)$ and $R(t,s)$ since $Q(t,s) = 0$ and $\overline{R}(t,s) = R(s,t)$ as was argued in the end of the previous subsection. This suggests that there are only two independent components of Σ namely $\Sigma_{\hat{\phi}\hat{\phi}}$ and $\Sigma_{\hat{\phi}\phi}$ and consequently only two independent equations out of four (3.20) (we impose $t > s$):

$$\begin{aligned}
(\partial_t + \mu(t))C(t,s) &= \int_{-\infty}^{t} du \Sigma_{\hat{\phi}\phi}(t,u)C(u,s) + \int_{-\infty}^{s} du \Sigma_{\hat{\phi}\hat{\phi}}(t,u)R(s,u) \\
(\partial_t + \mu(t))R(t,s) &= \int_{s}^{t} du \Sigma_{\hat{\phi}\phi}(t,u)R(u,s)
\end{aligned} \tag{3.25}$$

These equations provide the dynamic evolution of the correlators of the spherical 3-spin model.

Finally, let's note that the generic SD equation (3.22) can be solved by iterations in powers of the interaction vertices. This solution yields the standard perturbation theory for the correlator $\mathbf{G}$. This shows that the self-consistent perturbation theory introduced above is just a partial resummation of the simple perturbation theory.

Chapter 4

Dynamical field theories for glass-forming liquids

The problems with MCT considered in previous chapters inspired a research aimed at the theory into there are related directions:

- Generation of systematic corrections. Analysis of the corrections would allow to test the structural stability and shed more light on the problem of the spurious transition and its cut off.
- Cut off of the transition: search for the mechanism responsible for smoothing the transition.
- Extension to low temperatures where the liquid falls out of equilibrium.

Experiments and numerical simulations [55, 6] indicate that density fluctuations are an adequate set of slow variables for describing the dynamics of supercooled liquids. Field theory provides a convenient context for analysis of the above three points. First, perturbation series which provide systematic corrections are easy to obtain within a field theory and there exist developed tools for dealing with them. This makes the structural stability analysis possible. Second, a field theory description extends naturally to glassy phases where liquid falls out-of-equilibrium and relaxation times become macroscopic. A similar derivation within the projection operator formalism is still out of reach despite the efforts [29]. Third, reconstruction of MCT within such a theory would shed more light on its physical background. Fourth, as we will see the dynamic equations derived within the field theory also contain static equations. This provides a possibility of simultaneous analysis of both static and dynamics properties contrary to the projection operator formalism where statics is merely an input for dynamics. Next, multi-point functions which are essential for probing dynamic heterogeneities are easily computed within this context. Finally, the results of 2.5 which identify the growing correlation length show that the glass transition

is not a short scale phenomenon as it was stressed [34, 84]. This validates application of a field theory description which is a long and intermediate scale formalism to the problem of the glass transition.

The derivation of MCT within a field theory received a lot of attention and several attempts were made [85, 28, 86, 38]. However the derivation seemed to be problematic: analysis of these works shows that the main obstacle is the preservation of the fluctuation-dissipation theorem (FDT) and therefore the underlying time-reversal symmetry (TRS) [87]. The equations of motions preserve TRS but an arbitrary approximation scheme does not necessarily. Within the derivations FDT was either assumed [85, 28] or some important simplifications had to be made [86] which as we will see, oversimplify the problem and lead to a model which cannot have a transition. Our attempt to reanalyse these works revealed a violation of TRS within the approximations used to construct MCT. Time reversal symmetry is a key ingredient of equilibrium dynamics and should not be violated: as we will see below in this chapter this may lead to incorrect results. On the other hand, low-T analysis of glassy phases also relies on time-reversal symmetry [31, 30, 88] and requires a theory that respects FDT in the first place.

The resolution of these issues is the main content of the present chapter. It is split into two big parts. In the first part we focus on formal symmetry related properties of the field theories. Our aim is to control the symmetry in perturbation expansions which would guarantee its preservation. We do that on the examples of field theories used in [85, 28]: Brownian dynamics which parallels MCT of 2.2 and Fluctuating Non-linear Hydrodynamics (eMCT). Time-reversal symmetry has a non-trivial expression within these field theories and requires much care. Neglecting this issue may lead to problems illustrated in Sec. 4.3. We show how to resolve this problem and construct correct perturbation expansions that automatically preserve TRS. These results are used in the second part where we highlight the problems with FDT appearing in [85, 28] and apply the developed formalism to recover MCT within the field theories.

4.1 Field theories of density fluctuations

We start by introducing two field theories used to study glass-forming liquids: dense colloidal systems which are represented well by Brownian dynamics and supercooled liquids which are studied in the context of Fluctuating Non-linear Hydrodynamics (FNH). FNH describes a compressible liquid which evolves under Newton dynamics with several conserved quantities on long- and intermediate timescales. We do not include energy for simplicity but it can be incorporated easily.

4.1.1 Brownian dynamics and the Dean equation

The starting point is a set of Langevin equations for a system of N Brownian particles in a 3d Euclidean space interacting via a pair potential U (lower case

Greek letters label coordinates, lower case Roman letters label the particles):

$$\partial_t \mathbf{x}_\alpha = -\sum_{\alpha<\beta} \nabla U(\mathbf{x}_\alpha - \mathbf{x}_\beta) + \xi_\alpha \tag{4.1}$$

where ξ_α is a Gaussian white noise of zero mean and with the variance:

$$\langle \xi_{\alpha,i}(\mathbf{x},t)\xi_{\beta,j}(\mathbf{y},s)\rangle = 2T\delta_{\alpha\beta}\delta_{ij}\ \delta(\mathbf{x}-\mathbf{y})\ \delta(t-s) \tag{4.2}$$

Using Itô calculus Dean showed that the local density $\rho(\mathbf{x},t) = \sum_\alpha \delta(\mathbf{x} - \mathbf{x}_\alpha(t))$ of the system evolving under (4.1) obeys the following Langevin equation [89]:

$$\partial_t \rho(\mathbf{x},t) = \nabla\cdot\left(\rho(\mathbf{x},t)\nabla\frac{\delta\mathcal{F}}{\delta\rho(\mathbf{x},t)}\right) + \eta(\mathbf{x},t) \tag{4.3}$$

This time, η is a Gaussian multiplicative noise with zero mean. Its variance reads:

$$\langle \eta_{\alpha,i}(\mathbf{x},t)\eta_{\beta,j}(\mathbf{y},s)\rangle = 2T\rho(\mathbf{x},t)\nabla_{\mathbf{x}}\nabla_{\mathbf{y}}\delta(\mathbf{x}-\mathbf{y})\delta(t-s)$$

Appearance of the multiplicative noise is not surprising: density cannot fluctuate in empty regions. The density functional $\mathcal{F}$ reads (for a fluid of average density ρ_0):

$$\begin{aligned}\mathcal{F}[\rho(\mathbf{x})] = T\int d^3\mathbf{x} \quad \rho(\mathbf{x})\left(\ln\tfrac{\rho(\mathbf{x})}{\rho_0} - 1\right) + \tfrac{1}{2}\int d^3\mathbf{x}\int d^3\mathbf{y}\ \rho(\mathbf{x})U(\mathbf{x}-\mathbf{y})\rho(\mathbf{y}) \\ = -T\mathcal{S}[\rho] + \mathcal{F}_{int}[\rho]\end{aligned} \tag{4.4}$$

Now the dynamic average of an observable $\mathcal{A}$ over realisations of the noise η can be expressed via a functional integral:

$$\begin{aligned}\langle\mathcal{A}\rangle = \int \mathcal{D}\rho\mathcal{A}[\rho]\langle\delta(\rho-\rho(\mathbf{x},t)))\rangle_\eta \\ = \int \mathcal{D}\rho\mathcal{A}[\rho]\left\langle\delta\left[\partial_t\rho(\mathbf{x},t) - \nabla\cdot\left(\rho(\mathbf{x},t)\nabla\frac{\delta\mathcal{F}}{\delta\rho(\mathbf{x},t)}\right) - \eta(\mathbf{x},t)\right]\right\rangle_\eta\end{aligned}$$

This is a standard procedure to derive a field theory from a stochastic equation (see [77]). The Jacobian arising from the transformation of δ-functions is constant and is absorbed into the definition of the integral. This is possible since (4.3) is defined in Itô sense and therefore has a Markov property.

The integral representation of the functional Dirac distribution through a conjugated field $\hat\rho$ allows one to carry out the average over the noise η. Then the average of $\mathcal{A}$ reads:

$$\langle\mathcal{A}\rangle = \iint\int \mathcal{D}\rho\int\mathcal{D}\hat\rho\ \mathcal{A}[\rho]e^{S[\rho,\hat\rho]}$$

with

$$S[\rho,\hat{\rho}] = \int d^3\mathbf{x} \int dt \left\{ \quad \hat{\rho}(\mathbf{x},t) \left[-\partial_t \rho(\mathbf{x},t) + \nabla \cdot \left(\rho(\mathbf{x},t) \nabla \frac{\delta \mathcal{F}[\rho]}{\delta \rho(\mathbf{x},t)} \right) \right] \right.$$
$$\left. + T\rho(\mathbf{x},t)(\nabla \hat{\rho}(\mathbf{x},t))^2 \right\} \tag{4.5}$$

or explicitly

$$\begin{aligned} S[\rho,\hat{\rho}] &= \int d^3\mathbf{x} \int dt \left\{ \hat{\rho}(\mathbf{x},t) \left[-\partial_t \rho(\mathbf{x},t) + T\nabla^2 \rho(\mathbf{x},t) \right.\right. \qquad (4.6) \\ &+ \left.\left. \nabla \cdot \left(\rho(\mathbf{x},t) \int d^3\mathbf{y} \; \nabla U(\mathbf{x}-\mathbf{y}) \rho(\mathbf{y},t) \right) \right] + T\rho(\mathbf{x},t)(\nabla \hat{\rho}(\mathbf{x},t))^2 \right\} \end{aligned}$$

For brevity we denote $x = (\mathbf{x}, t)$.

4.1.2 Fluctuating Non-linear Hydrodynamics

The equations of FNH are not derived from first principles contrary to the Dean equation (4.3). They are meant to be a generalisation of the hydrodynamics equations to intermediate time- and lengthscales. An accurate description of the short time- and length scales is not expected. Hence they describe the dynamics of a slow variables subject to a thermal noise generated by integrating the fast degrees of freedom.

A phenomenological derivation and discussion can be found in [28, 90]. In what follows we concentrate on FNH as presented by Das and Mazenko [28] in their analysis of a glass transition in compressible liquids. Their equations read:

$$\partial_t \rho_x = \int d^3\mathbf{y} \{\rho_x, g_{i,y}\} \frac{\delta \mathcal{F}}{\delta g_{i,y}} \tag{4.7}$$

$$\begin{aligned} \partial_t g_{i,x} &= \int d^3\mathbf{x}' \{g_{i,x}, \rho_y\} \frac{\delta \mathcal{F}}{\delta \rho_y} + \int d^3\mathbf{x}' \sum_j \{g_{i,x}, g_{j,y}\} \frac{\delta \mathcal{F}}{\delta g_{j,y}} \\ &+ \sum_j \int d^3\mathbf{y} \Gamma_{ij}(\mathbf{x}-\mathbf{y}) \frac{\delta \mathcal{F}}{\delta g_{j,y}} + \eta_{i,x} \end{aligned} \tag{4.8}$$

where ρ and g_i are density and the ith component of the momentum respectively. η_i is a Gaussian white noise with variance: $\langle \eta_i(\mathbf{x},t)\eta_j(\mathbf{y},s)\rangle = 2T\Gamma_{ij}(\mathbf{x}-\mathbf{y})\delta(t-s)$. The effective free energy functional $\mathcal{F}$ reads:

$$\mathcal{F} = \mathcal{F}_{kin} + \mathcal{F}_{int}$$
$$\mathcal{F}_{kin}[\rho, \mathbf{g}] = \frac{1}{2}\int d^3\mathbf{x}\frac{\mathbf{g}^2(\mathbf{x})}{\rho(\mathbf{x})},$$
$$\mathcal{F}_{int}[\rho, \mathbf{g}] = \frac{T}{m}\int d^3\mathbf{x}\rho(\mathbf{x})(\log[\rho(\mathbf{x})/\rho_0] - 1)$$
$$-\frac{T}{2m^2}\int d^3\mathbf{x}d^3\mathbf{y}c(\mathbf{x}-\mathbf{y})(\rho(\mathbf{x})-\rho_0)(\rho(\mathbf{y})-\rho_0)$$

where ρ_0 is the average density, m is the particles mass and $c(\mathbf{x})$ is the direct correlation function. The functional $\mathcal{F}_{int}$ is the Ramakrishnan-Youssouff functional. The Poisson brackets $\{\cdot,\cdot\}$ and Γ_{ij} are defined so that the continuity equation is satisfied and the linearised versions of equations (4.7) and (4.8) coincides with usual linear hydrodynamics equations:

$$\Gamma_{ij}(\mathbf{x}-\mathbf{y}) = \left[-\eta_0\left(\frac{1}{3}\nabla_i\nabla_j + \delta_{i,j}\nabla^2\right) - \zeta_0\nabla_i\nabla_j\right]\delta(\mathbf{x}-\mathbf{y}) = L_{ij}\delta(\mathbf{x}-\mathbf{y})$$
$$\{\rho(\mathbf{x}), g_i(\mathbf{x}')\} = -\nabla^i_x\delta(\mathbf{x}-\mathbf{x}')\rho(\mathbf{x})$$
$$\{g_i(\mathbf{x}), \rho(\mathbf{x}')\} = -\rho(\mathbf{x})\nabla^i_x\delta(\mathbf{x}-\mathbf{x}')$$
$$\{g_i(\mathbf{x}), g_j(\mathbf{x}')\} = -\nabla^j_x\delta(\mathbf{x}-\mathbf{x}')g_i(\mathbf{x}') - g_j(\mathbf{x})\nabla^i_x\delta(\mathbf{x}-\mathbf{x}')$$

η_0 and ζ_0 are bare shear and bulk viscosity. Equations (4.7),(4.8) can be written in a more explicit way:

$$\begin{aligned}\partial_t\rho_x &= -\nabla\cdot\mathbf{g}_x \\ \partial_t g_{i,x} &= -\rho_x\nabla_i\frac{\delta\mathcal{F}_{int}}{\delta\rho_x} - \sum_j\nabla_j\left(\frac{g_{i,x}g_{j,x}}{\rho_x}\right) - \sum_j L_{ij}\left(\frac{g_{j,x}}{\rho_x}\right) + \eta_{i,x}\end{aligned} \tag{4.9}$$

Note the difference with standard MCT where the longitudinal current was also considered in the derivation: here all the three components of the current are presented and coupled to density, like in EMCT considered in Sec. 2.7.

Starting from these equations and applying the same procedure as the one used for Dean's equation in Sec. 4.1.1 a field theory for ρ, $\mathbf{g}$ and conjugated fields $\hat\rho$ and $\hat{\mathbf{g}}$ is derived. The action reads: $S = \int d^3\mathbf{x}\int dt\, s_x$

$$\begin{aligned}s_x &= -\hat\rho_x\left[\partial_t\rho_x + \nabla_i\left(\rho_x\frac{\delta\mathcal{F}}{\delta g_{i,x}}\right)\right] + T\hat g_{i,x}L_{ij}\hat g_{j,x} \\ &\quad -\hat g_{i,x}\left[\partial_t g_{i,x} + \rho_x\nabla_i\frac{\delta\mathcal{F}}{\delta\rho_x} + \nabla_j\left(g_{i,x}\frac{\delta\mathcal{F}}{\delta g_{j,x}}\right) + g_{j,x}\nabla_i\frac{\delta\mathcal{F}}{\delta g_{j,x}} + L_{ij}\frac{\delta\mathcal{F}}{\delta g_{j,x}}\right]\end{aligned} \tag{4.10}$$

Note that Brownian dynamics has a multiplicative noise, but FNH does not. However in both case the transformations corresponding to time-reversal

symmetry are non-linear in fields which is due to the multiplicative noise for BD and the coupling to currents for FNH. This is the origin of the problems with preservation of time reversal symmetry in approximations [87]. In the following we focus on the field theory for Brownian dynamics. The analysis of FNH follows closely the one for Brownian dynamics and will be presented separately in Sec. 4.5. We would refer to the field theory generated by Dean's equation as Brownian dynamics (BD) and to field theory generated by FNH equations as FNH.

4.2 Time-reversal symmetry and fluctuation-dissipation theorem

Time-reversal symmetry (TRS) relates the probabilities of a path in configurational space and its time reversal counterpart. It plays an important role in equilibrium dynamics: all the systems in equilibrium obey this symmetry. All physical correlation functions are invariant under time reversal. For two point functions this results into a well-known fluctuation dissipation theorem. In context of field theory this symmetry is represented by a transformation of the fields which leaves the action invariant.

In this section we derive the transformations associated to TRS, identify the response function and derive the fluctuation-dissipation theorem (FDT) within Brownian dynamics. To highlight the subtleties related to TRS for dynamical theories of liquids we use the 3-spin model as a reference because it has a simple transformation associated to TRS so that no subtleties appear.

4.2.1 Spherical 3-spin model

A straightforward check shows that the following transformation leaves the action (3.5) invariant under time reversal $t \to -t$:

$$\begin{bmatrix} \phi(-t) \\ \hat{\phi}(-t) \end{bmatrix} \quad \to \quad \begin{pmatrix} 1 & 0 \\ -\frac{1}{T}\partial_t & 1 \end{pmatrix} \begin{bmatrix} \phi(t) \\ \hat{\phi}(t) \end{bmatrix} = \mathbf{M} \begin{bmatrix} \phi(t) \\ \hat{\phi}(t) \end{bmatrix}$$

Note that this transformation is linear in fields. As we have seen in Sec. 3.1 conjugate fields $\hat{\phi}_l$ are also the response fields so that the response function reads:

$$R(t,s) = \left\langle \overline{\frac{\delta \phi_k(t)}{\delta h(s)}} \right\rangle_{h=0} = \overline{\langle \phi_k(t) \hat{\phi}_k(s) \rangle}.$$

The above transformation implies Ward-Tahakashi identities for various correlators. In particular for R and $\overline{R}$ (see (3.8)) it gives the fluctuation-dissipation theorem:

$$R(s-t) = R(t-s) - \frac{1}{T}\partial_s C(t-s) \quad \rightarrow \quad R(t-s) = \frac{\theta(t-s)}{T}\partial_s C(t-s)$$
$$\overline{R}(s-t) = \overline{R}(t-s) - \frac{1}{T}\partial_t C(t-s) \quad \rightarrow \quad \overline{R}(t-s) = \frac{\theta(s-t)}{T}\partial_t C(t-s)$$

where we have used causality of R and $\overline{R}$. This identities is the standard form of FDT.

4.2.2 Brownian dynamics

Let's now repeat the analysis for Brownian dynamics. It is easy to see that the transformations which leaves the action (4.5) invariant under time-reversal $t \to -t$ should transform $\hat{\rho}$ in a non-trivial way $\hat{\rho}(t, \mathbf{x}) \to \hat{\rho}(-t, \mathbf{x}) + \cdots$ and should not affect the density field: $\rho(t, \mathbf{x}) \to \rho(-t, \mathbf{x})$. An explicit computation shows that there are two different transformations that leave the action unchanged (up to a border term $-1/T \int_x \partial_t \mathcal{F}[\rho_x]$):

- First transformation:

$$\mathcal{T} : \begin{cases} t & \to -t \\ \hat{\rho}_x & \to \hat{\rho}_x + f_x, \end{cases} \tag{4.11}$$

  where f verifies:

$$\nabla \cdot (\rho_x \nabla f_x) = -\frac{1}{T}\partial_t \rho_x. \tag{4.12}$$

  and plays a role similar to the longitudinal part of a current for the density field (Indeed the above equation is the usual density conservation equation with the "current" $\rho \nabla f$).

- Second transformation:

$$\mathcal{U} : \begin{cases} t & \to -t \\ \hat{\rho}_x & \to -\hat{\rho}_x + \frac{1}{T}\frac{\delta \mathcal{F}}{\delta \rho_x}. \end{cases} \tag{4.13}$$

The above transformations ensure that any average of the type $\langle \prod_i \rho(\mathbf{x}_i, t_i) \rangle$ is invariant under time reversal. Although TRS has two different associated transformations the latter provide the same identities when applied to correlation functions. The most important fact is that these transformations are non-linear in the fields. The origin of this issue is easily tracked down: multiplicative noise in Eq. (4.3).

We now would like to derive FDT. This requires a definition of the response function which describes a response of density fluctuations to a perturbation by an external potential. The field $\hat{\rho}$ *is not* a response field contrary to naive expectations: applying $\mathcal{U}$ to a naive response function $\mathcal{G}_{xy}(t, s) = \langle \rho(\mathbf{x}, t)\hat{\rho}(\mathbf{y}, s) \rangle$ one gets:

$$\frac{1}{T}\left\langle \rho(\mathbf{x},t)\frac{\delta\mathcal{F}[\rho]}{\delta\rho(\mathbf{y},s)}\right\rangle = \Theta(t-s)C_{\rho\hat{\rho},xy}(t-s) + \Theta(s-t)C_{\rho\hat{\rho},yx}(s-t) \quad (4.14)$$

Clearly $C_{\rho\hat{\rho}}$ cannot be a response function since it is not related by a correct FDT to a density correlator. A correct method to identify the response function is to perturb density by adding a source term $\mathcal{F}_{ext}[\rho] = -\int_x \rho_x\mu_x$ to the free energy $\mathcal{F}$ in Eq. (4.4). The response $R_{xy}(t,s)$ at time t and position $\mathbf{x}$ to an infinitesimal external force switched on at time u and position $\mathbf{y}$ is defined as

$$\langle\rho(\mathbf{x},t)\rangle_\mu = \langle\rho(\mathbf{x},t)\rangle_{\mu=0} + \int d^3\mathbf{y}\int_s^t du\ R_{xy}(t,u)\mu(\mathbf{y},u) + o(\mu), \quad (4.15)$$

where $\langle\cdot\rangle_\mu$ is the average taken with the free energy functional $\mathcal{F}+\mathcal{F}_{ext}$. Expanding the path integral to first order in μ, one gets:

$$\langle\rho(\mathbf{x},t)\rangle_\mu = \langle\rho(\mathbf{x},t)\rangle_{\mu=0} + \int d^3\mathbf{y}\int_s^\infty du\ \left\langle\rho(\mathbf{x},t)\hat{\rho}(\mathbf{y},u)\nabla\cdot(\rho(\mathbf{y},u)\nabla\mu(\mathbf{y},u))\right\rangle.$$

Due to causality time u in the integral runs until t only, and integrating twice by parts, one obtains the response function:

$$R_{xy}(t,s) = -\langle\rho(\mathbf{x},t)\nabla\cdot(\rho(\mathbf{y},s)\nabla\hat{\rho}(\mathbf{y},s))\rangle. \quad (4.16)$$

This is the same response function as the one studied in [87].

Applying any of the transformations $\mathcal{T}$ or $\mathcal{U}$ to (4.16) yields FDT:

$$\frac{1}{T}\partial_s C_{xy}(t-s) = R_{xy}(t-s) - R_{xy}(s-t) \quad (4.17)$$

where $C_{\rho\rho,xy}(t-s) = \langle\delta\rho(\mathbf{x},t)\delta\rho(\mathbf{y},s)\rangle$ and $C_{ab,xy}(t,s) = \langle a(x,t)b(y,s)\rangle$[1]. Contrary to the 3-spin model the response function in Brownian dynamics looks like a three-point function but it is still a two-point function; the correct response field is $\nabla\cdot(\rho(\mathbf{y},s)\nabla\hat{\rho}(\mathbf{y},s))$ as implied by Eq. (4.16).

4.3 Non-linear symmetry and perturbation theory

The fact that time reversal symmetry for BD is associated to non-linear transformations of the fields has several important consequences. First, it makes Mode-Coupling Approximations violate the symmetry and hence fluctuation-dissipation theorem. Second, TRS plays an important role in field theory: it provides a number of identities between correlation functions known as Ward-Takahashi identities. As we will see below these identities hold perturbatively

[1] We adopt this notation in what follows for two point quantities

for the 3-spin model and simplify greatly the perturbation theory. On the contrary these identities are hard to use in Brownian dynamics and bring almost no simplification to perturbation theory. Furthermore as will be illustrated in Sec. 4.3.2 much care should be taken when considering a self-consistent perturbation theory which is of interest to us in order to preserve TRS: a careless treatment leads to violation of FDT. The illustration of these problems is the main focus of the section.

4.3.1 Linear symmetry

Let's consider again the 3-spin model and analyse the implications of TRS as given by Eq. (4.11) for perturbation series introduced in Sec. 3.1 which are generated by the decomposition of the action S (Eq. (3.5)) into a Gaussian part S_2 (Eq. (3.6)) and an interaction S_{INT} (Eq. (3.7)). For convenience we introduce the vector $\Psi = (\Psi_1 \Psi_2)^T$ with components $\Psi = \phi$ and $\Psi = \hat{\phi}$ (here and forth ^{T} is a transpose of a vector or a matrix) so that the transformation given by Eq. (4.11) reads in these notations as $\Psi_k(-t) = M_{kl}\Psi_l(t)$. The integration measure $D\Psi_1 D\Psi_2$ is invariant under the time reversal transformation. Therefore applied to a correlation function TRS generates Ward-Takahashi (WT) identities:

$$\langle \tilde{\Psi}_{k_1} \cdots \tilde{\Psi}_{k_n} \rangle = \sum_{l_1 \cdots l_n} M_{k_1 l_1} \cdots M_{k_n l_n} \langle \Psi_{l_1} \cdots \Psi_{l_n} \rangle$$

where $\tilde{\Psi}(t) = \Psi(-t)$ denotes a time-reversed quantity. These identities hold non-perturbatively. However perturbation expansions of both sides of the identity generate terms with arbitrary powers m:

$$\langle \Psi_{k_1} \cdots \Psi_{k_n} \left(S_{INT}[\Psi]\right)^m \rangle_0$$

averaged with e^{S_2}. It is straightforward to check that S_2 and S_{INT} are separately invariant under the transformation (4.11). Therefore every term in the perturbative expansion is also invariant under TRS and the WT identities hold perturbatively order by order. In particular the following identity holds perturbatively for two-point quantities:

$$\tilde{G} = M \cdot G \cdot M^T \tag{4.18}$$

and $\tilde{G} = \langle \tilde{\Psi} \tilde{\Psi} \rangle$. Generally this result means that time reversal symmetry is respected order by order in perturbation series.

This reasoning applied to the standard perturbation theory. As we have seen in Sec. 3.1 analysis of dynamical properties requires a self-consistent perturbation theory. This theory also preserves the symmetry order by order: the proof is based on the invariance of the functional $\Gamma[G]$ which generates the self-consistent series under time reversal transformation. This is expected because a physical symmetry should not change the value of a functional. The invariance is proved using the definition of Γ given by Eq. (3.18). One should pay attention to the time independence of the vertices A_k: explicit time dependence of say, A_1 is equivalent to time dependent external field and breaks TRS explicitly.

The invariance of Γ has several consequences:

- Any time-reverted solution $\tilde{G}$ of $\delta\Gamma/\delta G = 0$ (see (3.20)) is also a solution. If time-reversal symmetry is unbroken which is certainly true at equilibrium, then there is a unique solution and FDT holds: $G = MGM^T$.
- Any expansion of Γ in powers of its parameters is also invariant *i.e.* coefficients of the expansions are invariant. Therefore the diagrammatic representation (3.21) is invariant under time-reversal because it is an expansion of Γ in powers of interaction vertices.

This together with the definition of self-energy Σ given by Eq. (3.24) allows to derive the transformation rule for the self-energy under time-reversal:

$$\begin{aligned} \tilde{\Sigma} &= M^{-T} \cdot \Sigma \cdot M^{-1} \\ \text{which gives explicitly:} & \\ \Sigma_{\phi\hat{\phi}}(t-s) &= \frac{\theta(s-t)}{T} \partial_t \Sigma_{\hat{\phi}\hat{\phi}}(t-s) \\ \Sigma_{\hat{\phi}\phi}(t-s) &= \frac{\theta(t-s)}{T} \partial_s \Sigma_{\hat{\phi}\hat{\phi}}(t-s) \\ \Sigma_{\hat{\phi}\hat{\phi}}(s-t) &= \Sigma_{\hat{\phi}\hat{\phi}}(t-s) \\ \Sigma_{\phi\phi} &= 0 \end{aligned} \tag{4.19}$$

Summarising, TRS is respected in perturbative expansions either simple or self-consistent order by order. A careful analysis of the above reasoning reveals that the proof is based on the linear structure of the transformation (4.11). The linearity assures that different powers of the fields do not mix under transformation so that the quadratic action S_2 has no chances to mix with the interaction S_{INT} under TRS. To make this point even more clear let's imagine that the transformation (4.11) is;

$$\Psi_k = M_{kl} \Psi_l^2.$$

Now S_2 and S_{INT} would be no more separately invariant under TRS since different powers of the fields are mixed. Therefore WT identities would then read:

$$\langle \tilde{\Psi}_{k_1} \cdots \tilde{\Psi}_{k_n} \rangle = M_{k_1 l_1} \cdots M_{k_n l_n} \langle \Psi_{l_1}^2 \cdots \Psi_{l_n}^2 \rangle.$$

They would mix correlation functions with different number of fields and would no more hold perturbatively. Self-consistent perturbation theory would no more preserve the symmetry order by order (in powers of the vertex) and the Mode-Coupling approximation would violate the time reversal symmetry.

4.3.2 Violation of the fluctuation dissipation theorem in the self-consistent perturbation theory for Brownian dynamics

Brownian dynamics with the non-linear transformation falls under this case. In this section we focus on the BD field theory and highlight the difficulties and the

failures of perturbation theories *vis à vis* FDT. The case of FNH is conceptually identical but practically more clumsy because of the larger number of fields.

Let us start with a bare perturbation theory. In order to do a perturbative analysis it is convenient to separate the Gaussian zero mean part of the local density field from the interacting one by introducing density fluctuations in (4.5) $\delta\rho_x = \rho_x - \rho_0$. This gives:

$$S = \int_x \left(s_{2,x} + s_{INT,x} \right), \tag{4.20}$$

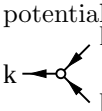
with:

$$\begin{aligned} s_{2,x} &= \hat{\rho}_x \left(-\partial_t \delta\rho_x + T\nabla^2 \delta\rho_x + \rho_0 \int_y U(\mathbf{x}-\mathbf{y}) \nabla^2 \delta\rho_y \right) + T\rho_0 \left(\nabla\hat{\rho}_x \right)^2 \\ s_{INT,x} &= T\delta\rho_x (\nabla\hat{\rho}_x)^2 + \hat{\rho}_x \, \nabla \cdot \left(\delta\rho_x \int_y \nabla U(\mathbf{x}-\mathbf{y}) \, \delta\rho_y \right). \end{aligned} \tag{4.21}$$

This may be written in a more compact form through the bidimensional vector field $(\delta\rho, \hat{\rho})^\dagger$. In Fourier space, the inverse of the propagator of this field is:

$$\tilde{G}_0^{-1} = \begin{pmatrix} 0 & i\omega + T\mathbf{q}^2 \left(1 + \beta\rho_0 U(\mathbf{q}) \right) \\ -i\omega + T\mathbf{q}^2 \left(1 + \beta\rho_0 U(\mathbf{q}) \right) & -2T\rho_0 \mathbf{q}^2 \end{pmatrix}. \tag{4.22}$$

This gives the following Feynman rules:

- bare density correlator:
   $C_0(\mathbf{q}, \omega)$
- bare naive response:
   $\mathcal{G}_0(\mathbf{q}, \omega)$
- potential vertex:
  

  $\frac{1}{2} \left(\mathbf{k} \cdot \mathbf{k}' U(\mathbf{k}') + \mathbf{k} \cdot \mathbf{k}'' U(\mathbf{k}'') \right) = \mathcal{V}(\mathbf{k}, \mathbf{k}', \mathbf{k}'')$
- noise vertex:
  

  $-T\mathbf{k}' \cdot \mathbf{k}'$

The bare density correlator is

$$C_0(\mathbf{q}, \omega) = \frac{2T\rho_0 \mathbf{q}^2}{\omega^2 + \left(T\mathbf{q}^2 \right)^2 \left(1 + \beta\rho_0 U(\mathbf{q}) \right)^2}, \tag{4.23}$$

and the bare naive response (as we have already seen this is not really a response to an external perturbation)

$$\mathcal{G}_0(\mathbf{q},\omega) = \frac{1}{-i\omega + T\mathbf{q}^2\left(1+\beta\rho_0 U(\mathbf{q})\right)} \tag{4.24}$$

Due to the form of the vertices, diagrams with tadpoles and hence corrections to the average density ρ_0 vanish to all orders (the momentum at the entrance into the tadpole is zero). This is expected since density is a conserved quantity.

In terms of density fluctuations, the response (4.16) is

$$\begin{aligned} R_{xy}(t,s) &= -\langle \delta\rho(\mathbf{x},t)\nabla\cdot(\delta\rho(\mathbf{y},s)\nabla\hat{\rho}(\mathbf{y},s))\rangle - \rho_0\nabla^2\langle\delta\rho(\mathbf{x},t)\hat{\rho}(\mathbf{y},s)\rangle \\ &= \chi_{xy}(t,s) - \rho_0\ \nabla^2\mathcal{G}_{xy}(t,s), \end{aligned} \tag{4.25}$$

where $\mathcal{G}$ is the naive response introduced earlier and χ is an "anomalous" response. Having a look at (4.25), one sees that part of the non-triviality of the FDT arises from the anomalous response χ, which itself comes from the multiplicative aspect of the noise, or equivalently from the nonlinearity of the transformation of the fields associated with time-reversal. Consequently both transformations $\mathcal{T}$ and $\mathcal{U}$ mix quadratic action S_2 and interaction S_{INT}. Therefore time-reversal symmetry is not preserved in the perturbation theory order by order unlike in the 3-spin model. Indeed applying transformation to diagrams of a fixed order would produce diagrams of higher orders. If one would like to enforce the preservation then one has to take into account interaction vertices non-perturbatively. This discussion makes it clear that any approximation which neglects vertex renormalisation is expected to be in contradiction with FDT. This is indeed what happens in self-consistent approximations as we shall show below.

Let us now focus on a self-consistent perturbation theory and in particular on a mode-coupling approximation introduced by Kawasaki [81] that consists in neglecting vertex renormalisation.

First, we write the Schwinger-Dyson (SD) equations (3.20)

$$G_0^{-1}\cdot G = 1 + \Sigma\cdot G, \tag{4.26}$$

where Σ is the self energy,

$$\Sigma(\mathbf{k},\omega) = \begin{pmatrix} \Sigma_{\rho\rho}(\mathbf{k},\omega) & \Sigma_{\rho\hat{\rho}}(\mathbf{k},\omega) \\ \Sigma_{\hat{\rho}\rho}(\mathbf{k},\omega) & \Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{k},\omega) \end{pmatrix}, \tag{4.27}$$

and the associative product $(\cdot)$ is defined as follows:

$$(A\cdot B)(\mathbf{q},\tau) = \int_{-\infty}^{\infty} du\, A(\mathbf{q},\tau-u)B(\mathbf{q},u).$$

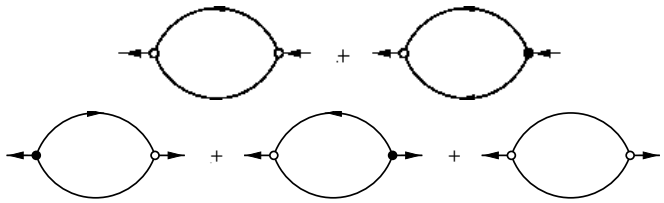
assuming that initial conditions are set at $\tau = -\infty$. Causality and reality of the density auto-correlator imply that the self-energies verify:

$$
\begin{aligned}
\Sigma_{\rho\hat{\rho}}(\mathbf{q},\tau) &= \Sigma_{\hat{\rho}\rho}(\mathbf{q},-\tau) \\
\Sigma_{\rho\hat{\rho}}(\mathbf{q},\tau) &= 0 \qquad \text{for} \quad \tau < 0 \\
\Sigma_{\rho\rho}(\mathbf{q},\tau) &= 0
\end{aligned}
\tag{4.28}
$$

The first diagrams contributing to the self-energies are

$\Sigma^{(2)}_{\hat{\rho}\rho}$ =

$\Sigma^{(2)}_{\hat{\rho}\hat{\rho}}$ =



Diagrams of higher orders all contain vertex renormalisation. Hence, if one neglects renormalisation of both vertices, the SD equations (4.26) become the Mode-Coupling equations for (4.3) (for $\tau > 0$):

$$
\begin{aligned}
\partial_\tau \mathcal{G}(\mathbf{q},\tau) &= -\rho_0 T\mathbf{q}^2 \ (1+\beta\, U(\mathbf{q}))\, \mathcal{G}(\mathbf{q},\tau) + \textstyle\int_0^\tau du\ \Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau-u)\ \mathcal{G}(\mathbf{q},u) \\
\partial_\tau C(\mathbf{q},\tau) &= -\rho_0 T\mathbf{q}^2 \ (1+\beta\, U(\mathbf{q}))\, C(\mathbf{q},\tau) + \textstyle\int_{-\infty}^0 du\ \Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u)\ \mathcal{G}(\mathbf{q},u) \\
&\quad + \textstyle\int_{-\infty}^\tau du\ \Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau-u)\ C(\mathbf{q},u)
\end{aligned}
\tag{4.29}
$$

with:

$$
\begin{aligned}
\Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau) &= 4\int \frac{d^3\mathbf{k}}{(2\pi)^3}\ \mathcal{G}(\mathbf{q},\tau)\ C(\mathbf{q}-\mathbf{k},\tau)\mathcal{V}(\mathbf{k},\mathbf{q},\mathbf{q}-\mathbf{k})\mathcal{V}(\mathbf{q},\mathbf{k},\mathbf{q}-\mathbf{k}) \\
&\quad -2\int \frac{d^3\mathbf{k}}{(2\pi)^3}\ \mathcal{G}(\mathbf{q},\tau)\ \mathcal{G}(\mathbf{q}-\mathbf{k},\tau)\mathbf{k}\cdot(\mathbf{q}-\mathbf{k})\mathcal{V}(\mathbf{k},\mathbf{q},\mathbf{q}-\mathbf{k}) \\
\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},t) &= 2\int \frac{d^3\mathbf{k}}{(2\pi)^3}\ C(\mathbf{q},\tau)\ C^2(\mathbf{q}-\mathbf{k},\tau) \\
&\quad -8\int \frac{d^3\mathbf{k}}{(2\pi)^3}\ \Re\mathcal{G}(\mathbf{q},\tau)\ C(\mathbf{q}-\mathbf{k},\tau)\mathbf{k}\cdot\mathbf{q}\mathcal{V}(\mathbf{k},\mathbf{q},\mathbf{q}-\mathbf{k})
\end{aligned}
\tag{4.30}
$$

These equations are not compatible with FDT as can be seen from the perturbative solution of SD equations in powers of the potential and noise vertices [87]. FDT is trivially verified at order zero. At order one the MC equations are exact and hence they are automatically compatible with FDT. Incompatibilities appear at order two, where diagrams such as those shown in Fig. 4.1, the first diagrams contributing to vertex renormalisation, have to be taken into account in the non self-consistent perturbation theory and therefore also in the self-consistent one (as discussed previously, in order to preserve FDT one has

always to take into account the contributions at all orders of the vertices). This suggests that if one wants to improve the approximation by keeping for instance the first vertex corrections, one has to include at least all the diagrams of order two in the self-energy. In that case, the incompatibility with FDT would be an effect of order three. However, nothing guaranties that the violation of FDT by the self-consistent approximation is attenuated when the order of the approximation is increased. That is, the self-consistent perturbation theory is not consistent with FDT and any approximation within this theory would also violate TRS. Another consequence of practical importance is that times in the integrals are not restricted in $[0, \tau]$. On the contrary, when time-reversal - and thus FDT - is preserved, times in the integrals run from 0 to τ as we will see later in this chapter and in Chapter 6 where this property is referred to as time ordering.

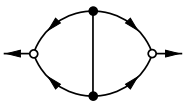


Figure 4.1: Example of diagram which contribute at order U^2 to FDT but is absent in the MC equations.

To conclude this paragraph, we remark that incompatibilities with FDT have arisen from an explicit breaking of the time-reversal symmetry by the decomposition $S = \int_x (s_{2,x} + s_{INT,x})$ which is due to the nonlinearity of the field transformations related to time-reversal symmetry.

4.3.3 Quadratic density functional

Let's also illustrate another important subtlety related with the response function: even if the transformations $\mathcal{T}$ or $\mathcal{U}$ were linear so that the decomposition $S = \int_x (s_{2,x} + s_{INT,x})$ does not break TRS, the response function may still be expressed via a multi-point (more than two) quantity.

Let's consider a quadratic density functional $\mathcal{F}$:

$$\mathcal{F}[\rho] = \frac{1}{2} \int_{x,y} \rho_x W(x-y) \rho_y \tag{4.31}$$

This can be achieved by truncation of the expansion of the entropic part $\mathcal{S}$ up to the order two:

$$\mathcal{S}[\rho] \approx \mathcal{S}[\rho_0] - T \int_x \frac{\delta \rho_x^2}{2\rho_0} \text{ So that } \quad W(\mathbf{x}) = U(\mathbf{x}) + \frac{T}{\rho_0} \delta \mathbf{x}$$

Then the equilibrium measure is also Gaussian:

$$\mathcal{P}[\rho] \sim \exp\left[-\frac{1}{2}\int \frac{d^3\mathbf{q}}{(2\pi)^3}\frac{\rho(-\mathbf{q})\rho(\mathbf{q})}{S(\mathbf{q})}\right]$$
$$S(\mathbf{q}) = \langle\rho(-\mathbf{q})\rho(\mathbf{q})\rangle = T/W(\mathbf{q})$$

where we use the fact that the potential V is defined up to a constant (see (4.1)) and fixed this constant so that $\int d^3\mathbf{x}U(x) = -T/\rho_0$ and $\int_x W(x) = 0$.

Now $\delta\mathcal{F}/\delta\rho$ is linear in ρ; therefore the transformation $\mathcal{U}$ given by Eq. (4.13) becomes linear:

$$\mathcal{U}: \begin{cases} t \to -t \\ \hat{\rho}_x \to -\hat{\rho}_x + \frac{1}{T}\int_y W(x-y)\rho_y \end{cases} \tag{4.32}$$

The linearity of $\mathcal{U}$ ensures that TRS is preserved in perturbation theory (both standard and self-consistent). Relation (4.14) simplifies to:

$$\frac{W_\mathbf{q}}{T}C(\mathbf{q},\tau) = \Theta(\tau)\mathcal{G}(\mathbf{q},\tau) + \Theta(-\tau)\mathcal{G}(\mathbf{q},\tau) \tag{4.33}$$

and holds perturbatively. Similarly self-energies (4.27) are now related by Eq. (4.19) or explicitly:

$$\Sigma_{\hat{\rho}\rho}(\mathbf{q},t) = -S(\mathbf{q})\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},t) \qquad t > 0 \tag{4.34}$$

This makes (4.33) compatible with the SD equations (4.29) which reduce to a single equation:

$$\partial_\tau C(\mathbf{q},\tau) = -\rho_0 T\mathbf{q}^2\ (1+\beta\ U(\mathbf{q}))\, C(\mathbf{q},\tau) - S(\mathbf{q})\int_0^\tau du\ \Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u)\ C(\mathbf{q},u)$$

But the response function is still given by Eq. (4.25) and still looks like a three-point quantity. Equivalently, the response field is a complicated two-field quantity.

4.4 Restoration of time-reversal symmetry in perturbative expansion

The violation of TRS within the self-consistent perturbation theory which is vital for reconstructing MCT is an important problem. Another related problem is the expression of the response function via a three-point quantity so that FDT is complicated and relates two-point quantity to a three-point one. Therefore it is hard to control. In this section we show how to resolve these problems and restore the symmetry in loop expansions.

4.4.1 How to linearise the symmetry and FDT ?

The example of 3-spin model and quadratic density functional considered above indicate that there are no symmetry related problems in perturbative expansions if the associated transformation is linear. This suggests to linearise the transformations $\mathcal{T}$ or $\mathcal{U}$ associated to TRS. The other point is to make the response function R given by Eq. (4.16) a two-point quantity.

The solution to these problems is to introduce auxiliary fields associated with the non-linear terms in the transformations $\mathcal{T}$ or $\mathcal{U}$ and define a response field which makes the response function a two-point quantity explicitly. This solves the problem at the price of a more complicated field theory with more fields.The 2 different transformations associated to TRS imply that one can construct three different field theories by linearising either both transformations simultaneously either only one of them. Below we present the latter case. The simultaneous linearisation of $\mathcal{T}$ and $\mathcal{U}$ is presented in [91].

Let's consider the time-reversal transformation $\mathcal{T}$ first: we start from the identity:

$$\langle \mathcal{A} \rangle = \int \mathcal{D}\rho \mathcal{A}[\rho] \left\langle \delta \left(\partial_t \rho(\mathbf{x},t) - \nabla \cdot \left(\rho(\mathbf{x},t) \nabla \frac{\delta \mathcal{F}}{\delta \rho(\mathbf{x},t)} \right) + \eta(\mathbf{x},t) \right) \right\rangle \tag{4.35}$$

We now plug into the functional integral the representation of the identity to introduce the field f from $\mathcal{T}$ (see (4.11))

$$\int \mathcal{D} \prod_{x,t} \delta \left(\nabla \cdot (\rho_x \nabla f_x) + \frac{1}{T} \partial_t \rho_x \right) \left(\det \left[-\nabla \cdot (\rho_x \nabla) \right] \right) = 1. \tag{4.36}$$

The minus sign is set in the determinant to assure the positivity of the operator so that we do not have to take the absolute value of the determinant. We exponentiate the delta function using an auxiliary field $\hat{f}$ and we introduce fermionic fields ϕ and $\overline{\phi}$ to exponentiate the determinant. As a consequence there are new terms to add to the previous action (4.5) that read:

$$\int_x \hat{f}_x \left(\nabla(\rho_x \nabla f_x) + \frac{1}{T} \partial_t \rho_x \right) - \int_x \rho_x \nabla \phi_x \cdot \nabla \overline{\phi}_x. \tag{4.37}$$

Furthermore we introduce also the response field $\psi = \nabla \cdot (\rho \nabla \hat{\rho})$ which makes the response function a two-point quantity. This leads to introduce a conjugated field $\hat{\psi}$ for the Fourier representation of the delta function related to ψ. The final action is the integral of

$$s_x = -\hat{\rho}_x \partial_t \rho_x + \psi_x \tfrac{\delta \mathcal{F}}{\delta \rho_x} - T \hat{\rho}_x \psi_x + \hat{\psi}_x (\psi_x - \nabla_x \cdot (\rho_x \nabla_x \hat{\rho}_x)) \tag{4.38}$$

$$+ \hat{f}_x \left(\tfrac{1}{T} \partial_t \rho_x + \nabla_x \cdot (\rho_x \nabla_x f_x) \right) - \rho_x \nabla_x \cdot \left(\phi_x \nabla_x \overline{\phi}_x \right). \tag{4.39}$$

This action now remains invariant up to the boundary terms under the following linear transformation $\mathcal{T}_1$: first invert the time $t \to -t$ then transform the fields in as follows:

$$\mathcal{T}_1 : \left\{ \begin{array}{lcl} \hat{\rho}_x & \to & \hat{\rho}_x + f_x \\ \psi_x & \to & \psi_x + \frac{1}{T}\partial_t \rho_x \\ \hat{\psi}_x & \to & \hat{\psi}_x + T f_x \\ \hat{f}_x & \to & -\hat{f}_x + T f_x + \hat{\psi}_x + T\hat{\rho}_x \\ f_x & \to & -f_x \end{array} \right. \tag{4.40}$$

Formally we can write this as $\tilde{\Psi} = M \cdot \Psi$, where $\Psi = (\rho, \hat{\rho}, \psi, \hat{\psi}, f, \hat{f}, \phi, \overline{\phi})^T$ and $\tilde{\Psi}(\mathbf{x}, t) = \Psi(-\mathbf{x}, t)$. Now the field transformation corresponding to TRS is linear. As we already know the propagators transform following the rule:

$$\langle \tilde{\Psi\Psi} \rangle = M \cdot \langle \Psi\Psi \rangle \cdot M^T \tag{4.41}$$

This transformation has a determinant of modulus one, as a product of simple transformations with this property.

Let's now show that this transformation implies FDT. Consider $R_{xy}(s - t) = -\langle \rho(\mathbf{x}, -t)\psi(\mathbf{y}, -s) \rangle$. Under the transformation $\mathcal{T}_1$ this transforms into $R_{xy}(t - s) - \frac{1}{T}\partial_s C_{xy}(t - s)$. Thus the equality (4.41) implies in particular $R_{xy}(t - s) = R_{xy}(s - t) + \frac{1}{T}\partial_s C_{xy}(t - s)$ which is the fluctuation-dissipation relation given by Eq. (4.17).

We now show how to linearise the second transformation $\mathcal{U}$. Let's introduce the field $\theta = \delta\mathcal{F}/\delta\rho$ and the conjugate one $\hat{\theta}$ to exponentiate the delta function[2]. The action S is then transformed into an integral of

$$s_x = -\hat{\rho}_x \partial_t \rho_x + T\rho_x (\nabla_x \hat{\rho}_x)^2 + \hat{\theta}_x \left(\theta_x - \frac{\delta\mathcal{F}}{\delta\rho_x} \right) - \rho_x (\nabla_x \hat{\rho}_x)(\nabla_x \theta_x). \tag{4.42}$$

The corresponding linear transformation $\mathcal{U}_1$ reads

$$\mathcal{U}_1 : \left\{ \begin{array}{lcl} \hat{\rho}_x & \to & -\hat{\rho}_x + \frac{1}{T}\theta_x \\ \hat{\theta}_x & \to & \hat{\theta}_x - \frac{1}{T}\partial_t \rho_x, \end{array} \right. \tag{4.43}$$

As before we write it as $\tilde{\Psi} = M \cdot \Psi$, where $\Psi = (\rho, \hat{\rho}, \theta, \hat{\theta})^T$ this time and $\tilde{\Psi}(\mathbf{x}, t) = \Psi(-\mathbf{x}, t)$. The same identity (4.41) holds for time-reverted and usual correlators. Again the transformation has a determinant of modulus one. In the context of $\mathcal{U}_1$ the response function expresses as $R_{xy}(t-s) = \langle \rho(\mathbf{x}, t)\hat{\theta}(\mathbf{y}, s) \rangle$ so that there is no need to introduce a special response field. Applying the transformation $\mathcal{U}_1$ to $R_{xy}(t - s)$ we recover FDT

$$\langle \rho(\mathbf{x}, -t)\hat{\theta}(\mathbf{y}, -s) \rangle = R_{xy}(t - s) - \frac{1}{T}\partial_s C_{xy}(t - s)$$

[2]The usefulness of introducing these two fields when dealing with the BD field theory was noticed by Chamon and Cugliandolo from a slightly different perspective [92]

One might wonder how final results depend on a particular implementation of the linearised field theory. As far as self-consistent perturbation theory and MCT are concerned we derived dynamical equations within

a) completely linearised theory where both symmetries are made linear.

b) theory where only transformation $\mathcal{T}$ is made linear.

c) theory where only transformation $\mathcal{U}$ is made linear.

At the order of one loop, we have found the same sets of equations for correlation and response function at long times in all cases. This is not surprising, since the different transformations do not affect the physical fields and change the response fields in the same way. We thus expect this to be valid at all orders. Indeed, as we will show below, one gets closed equations for the dynamical evolutions of correlators involving only the fields ρ and θ. In addition, this suggests that FDT makes the results robust with respect to the choice of extra dynamical variables. This remark allows to focus in the following on the simplest theory written above in terms of ρ, $\hat{\rho}$, θ and $\hat{\theta}$ only, the choice of the fields to work with being merely a matter of taste. For brevity we will refer to it as a minimal theory.

4.4.2 Minimal theory and Ward-Tahakashi identities

Let's consider the minimal theory and its properties. Our aim is to derive explicitly the Ward-Tahakashi identities for two-point correlators and introduce the perturbation theory. As the symmetry is preserved we expect a considerable simplifications in computations related with perturbation theory. Indeed as we shall see there are only 3 independent correlators although 4 fields suggests 16 correlators in general case.

The minimal theory is defined by the action (4.42); TRS transformation is $\mathcal{U}_1$. Standard perturbation theory defined similarly to that considered in Sec. 4.3.2: we consider density fluctuations $\delta\rho$ around the constant average value ρ_0. This decomposes the action into Gaussian part $s_{2,x}$ and interaction $s_{INT,x}$:

$$s_{2,x} = -\hat{\rho}_x \partial_t \delta\rho_x - T\rho_0 \hat{\rho}_x \nabla^2 \hat{\rho}_x + \hat{\theta}_x \theta - \hat{\theta}_x \int_y W(x-y)\rho_y + \rho_0 \hat{\rho}_x \nabla^2 \theta_x \quad (4.44)$$

$$s_{INT,x} = T\delta\rho_x (\nabla\hat{\rho}_x)^2 - \delta\rho_x (\nabla\hat{\rho}_x)(\nabla\theta_x) + T\hat{\theta}_x \sum_{n>1} \frac{1}{n} \left[-\frac{\delta\rho_x}{\rho_0} \right]^n \quad (4.45)$$

where W is given by (4.31) and satisfies the constraint $\int d^3\mathbf{x} W(x) = 0$. The physical background of such perturbation will be discussed later. Both the Gaussian part s_2 and the interaction s_{INT} are separately invariant under $\mathcal{U}_1$. Furthermore every term in the sum over n in $s_{INT,x}$ is also separately invariant.

Recalling the form of $\mathcal{U}_1$ (4.43) and the generic transformation rule for two-point quantities (4.18) the following WD identities are derived:

$$
\begin{aligned}
C_{\rho\hat{\rho}}(\mathbf{q},\tau) &= \frac{\Theta(\tau)}{T}C_{\rho\theta}(\mathbf{q},\tau) \\
C_{\theta\hat{\rho}}(\mathbf{q},\tau) &= \frac{\Theta(\tau)}{T}C_{\theta\theta}(\mathbf{q},\tau) \\
C_{\theta\hat{\theta}}(\mathbf{q},\tau) &= -\frac{1}{T}\frac{\partial}{\partial\tau}\left[\Theta(\tau)C_{\rho\theta}(\mathbf{q},\tau)\right] \\
C_{\rho\hat{\theta}}(\mathbf{q},\tau) &= -\frac{\Theta(\tau)}{T}\frac{\partial}{\partial\tau}C_{\rho\rho}(\mathbf{q},\tau) = R(\mathbf{q},\tau)
\end{aligned}
\tag{4.46}
$$

where equilibrium and time translation invariance are supposed. Note the explicit causality of the correlators expressed by Θ-functions. It is worth noting one more time that FDT appears among these identities. There are only three independent correlators: $C_{\rho\theta}$,$C_{\theta\theta}$ and $C_{\rho\rho,xy}$ while all the rest are either related to these via (4.46) or vanish identically.

Bare propagators $G_0(\mathbf{q},\omega)$ as implied by (4.44) read (rows and columns are arranged in the following order $\rho,\hat{\rho},\theta,\hat{\theta}$):

$$
\begin{pmatrix}
\frac{2T\rho_0 q^2}{(\omega^2+\rho_0^2 q^4 W(\mathbf{q})^2)} & -\frac{1}{i\omega+\rho_0 q^2 W(\mathbf{q})} & \frac{2T\rho_0 q^2 W(\mathbf{q})}{(\omega^2+\rho_0^2 q^4 W(\mathbf{q})^2)} & \frac{\rho_0 q^2}{i\omega+\rho_0 q^2 W(\mathbf{q})} \\
-\frac{1}{-i\omega+\rho_0 q^2 W(\mathbf{q})} & 0 & -\frac{W(\mathbf{q})}{-i\omega+\rho_0 q^2 W(\mathbf{q})} & 0 \\
-\frac{2T\rho_0 q^2 W(\mathbf{q})}{(\omega^2+\rho_0^2 q^4 W(\mathbf{q})^2)} & -\frac{W(\mathbf{q})}{i\omega+\rho_0 q^2 W(\mathbf{q})} & -\frac{2T\rho_0 q^2 W(\mathbf{q})^2}{(\omega^2+\rho_0^2 q^4 W(\mathbf{q})^2)} & -\frac{i\omega}{i\omega+\rho_0 q^2 W(\mathbf{q})} \\
\frac{\rho_0 q^2}{-i\omega+\rho_0 q^2 W(\mathbf{q})} & 0 & \frac{i\omega}{i\omega+\rho_0 q^2 W(\mathbf{q})} & 0
\end{pmatrix}
\tag{4.47}
$$

Bare propagators are also causal. This together with the structure of the interaction s_{INT} preserves causality in perturbation theory. The structure of the bare propagator $C_{0,\theta\hat{\theta}}$ explains the anomaly in the third identity in Eq. (4.46) where time derivative of the Heaviside function appears. By definition $C_{0,\theta\hat{\theta}}$ reads:

$$C_{0,\theta\hat{\theta}}(\mathbf{k},\omega) = -1 + \text{function of } (\mathbf{k},\omega).$$

It is easy to prove using (4.44) and (4.45) that there are no diagrammatic corrections to the -1 term so that it persists perturbatively. Then we can write:

$$C_{\theta\hat{\theta},xy}(\tau) = -\frac{\Theta(\tau)}{T}\frac{\partial}{\partial\tau}C_{\rho\theta,xy}(\tau) - \delta(\tau)$$

Since $C_{\rho\theta,xy}(0) = T$ (see Sec. 4.4.4) the last equation transforms into the anomalous identity in Eq. (4.46).

4.4.3 The Schwinger-Dyson equations for minimal theory

The self-consistent perturbation theory constructed by standard methods from s_2 and s_{INT} respects FDT and also the other WD identities considered above because of the linearity of $\mathcal{U}_1$. The Schwinger-Dyson equations read: $\mathbf{G}^{-1}(\mathbf{k},\omega) = \mathbf{G}_0^{-1}(\mathbf{k},\omega) - \mathbf{\Sigma}(\mathbf{k},\omega)$ (note that this is a matrix equation) or

$$\mathbf{G}_0^{-1}\cdot\mathbf{G}(\mathbf{q},\tau)=\mathbb{I}(\mathbf{q})\delta(\tau)+\mathbf{\Sigma}\cdot G(\mathbf{q},\tau) \tag{4.48}$$

and the self-energy is a matrix

$$\mathbf{\Sigma}=\begin{pmatrix} 0 & \Sigma_{\rho\hat{\rho}} & 0 & \Sigma_{\rho\hat{\theta}} \\ \Sigma_{\hat{\rho}\rho} & \Sigma_{\hat{\rho}\hat{\rho}} & \Sigma_{\hat{\rho}\theta} & \Sigma_{\hat{\rho}\hat{\theta}} \\ 0 & \Sigma_{\theta\hat{\rho}} & 0 & \Sigma_{\theta\hat{\theta}} \\ \Sigma_{\hat{\theta}\rho} & \Sigma_{\hat{\theta}\hat{\rho}} & \Sigma_{\hat{\theta}\theta} & \Sigma_{\hat{\theta}\hat{\theta}} \end{pmatrix}$$

where $\Sigma_{ab}=\delta\Gamma/\delta C_{ab}$ and we used the identity $\Sigma_{ab}=0$ for $a,b\in\{\rho,\theta\}$: all diagrams contributing to these self-energies vanish because of causality of the correlators.

The WD identities for the self-energies given by Eq. (4.19) read:

$$\begin{aligned}
\Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau) &= -\frac{1}{T}\frac{\partial}{\partial\tau}\left[\Theta(\tau)\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau)\right] \\
\Sigma_{\hat{\theta}\rho}(\mathbf{q},\tau) &= -\frac{\Theta(\tau)}{T}\frac{\partial}{\partial\tau}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau) \\
\Sigma_{\hat{\rho}\theta}(\mathbf{q},\tau) &= -\frac{\Theta(\tau)}{T}\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau) \\
\Sigma_{\hat{\theta}\theta}(\mathbf{q},\tau) &= -\frac{\Theta(\tau)}{T}\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau)
\end{aligned} \tag{4.49}$$

The anomaly in the first identity on $\Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau)$ has the same diagrammatic origin as the one we considered before for $C_{0,\theta\hat{\theta}}$: there are diagrams contributing to $\Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau)$ which produce a δ-function term. However this times their direct resummation is cumbersome and we used the SD equations to fix the coefficient of the δ-function.

Consistently with the case of correlators, these identities imply that there are only three independent self-energies. Since there are only three independent correlators one expects only three independent SD equations. It is easy to prove that there are only 4 independent SD equations among the 16 given by Eq. (4.48), the other being related to them by $\mathcal{U}_1$. Below we present these four equations while their derivation is detailed in Appendix A. The proof that one of the four equations is not independent is trickier and is left for Appendix B. We take $\tau>0$ in Eq. (4.48) so that the δ-function terms drops out resulting in equations valid only for strictly positive times. The $\tau=0$ version of (4.48) provides static equations and is discussed Sec. 4.4.4.

We first consider $(G_0^{-1}\cdot G-\Sigma\cdot G)_{\hat{\rho}\rho}=0$ which results into:

$$\begin{aligned}
\partial_\tau C_{\rho\rho}(\mathbf{q},\tau)+\rho_0\mathbf{q}^2C_{\rho\theta}(\mathbf{q},\tau) = &-\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u)C_{\rho\theta}(\mathbf{q},u) \\
&-\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u)
\end{aligned} \tag{4.50}$$

Next consider $(G_0^{-1}\cdot G-\Sigma\cdot G)_{\hat{\rho}\theta}=0$:

$$\partial_\tau C_{\rho\theta}(\mathbf{q},\tau)+\rho_0\mathbf{q}^2C_{\theta\theta}(\mathbf{q},\tau)=\frac{1}{T}\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau)C_{\rho\theta}(\mathbf{q},0) \quad (4.51)$$
$$-\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u)C_{\theta\theta}(\mathbf{q},u)-\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\theta}(\mathbf{q},u)$$

and $(G_0^{-1}\cdot G-\Sigma\cdot G)_{\hat{\theta}\rho}=0$:

$$W(\mathbf{q})C_{\rho\rho}(\mathbf{q},\tau)-C_{\rho\theta}(\mathbf{q},\tau)=\frac{1}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0)C_{\rho\rho}(\mathbf{q},\tau) \quad (4.52)$$
$$-\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)C_{\rho\theta}(\mathbf{q},u)-\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u)$$

Finally $(G_0^{-1}\cdot G-\Sigma\cdot G)_{\hat{\theta}\theta}=0$:

$$W(\mathbf{q})C_{\rho\theta}(\mathbf{q},\tau)-C_{\theta\theta}(\mathbf{q},\tau)=\frac{1}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0)C_{\rho\theta}(\mathbf{q},\tau)-\frac{1}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau)C_{\rho\theta}(\mathbf{q},0)$$
$$-\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)C_{\theta\theta}(\mathbf{q},u)-\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\theta}(\mathbf{q},u) \quad (4.53)$$

As was pointed out above this system is redundant and the last equation is related to the others (see Appendix B).

Finally the non-trivial result is that equations (4.50),(4.51),(4.52) are *exact* self-consistent equations closed by self-consistent expansions for self-energies which respect time-reversal symmetry and FDT. Therefore approximations made within these equations would preserve TRS and FDT automatically.

4.4.4 Static limit

The dynamic equation (4.50),(4.51),(4.52) derived in the previous subsection require initial conditions to fix their solutions. These conditions are fixed by the limit $\tau=0$ of (4.48) which provides static equations (The derivation is detailed in Appendix A):

$$\begin{aligned}
C_{\rho\theta}(\mathbf{q},0) &= T\\
\dot{C}_{\rho\rho}(\mathbf{q},0^+) &= -\rho_0\mathbf{q}^2C_{\rho\theta}(\mathbf{q},0)=-T\rho_0\mathbf{q}^2\\
C_{\theta\theta}(\mathbf{q},0) &= W(\mathbf{q})C_{\rho\theta}(\mathbf{q},0)=TW(\mathbf{k})\\
\dot{C}_{\rho\theta}(\mathbf{q},0^+) &= -W(\mathbf{q})T\rho_0\mathbf{q}^2-T\Sigma_{\hat{\theta}\theta}(\mathbf{q},0)\\
C_{\rho\rho}(\mathbf{q},0) &= \frac{T}{W(\mathbf{q})}+\frac{1}{TW(\mathbf{q})}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0)C_{\rho\rho}(\mathbf{q},0)
\end{aligned} \quad (4.54)$$

where $\dot{C}$ stands for $\partial_\tau C$. The last equation was derived as a $\tau=0$ limit of (4.52) where we have use the zero time value of $C_{\rho\theta}(\mathbf{q},0)$. This is the advantage of the

field theory approach where the statics is contained in the dynamics contrary to the projection operator formalism. We verified that the same equations are obtained within the context of the static theory implied by the free energy $\mathcal{F}$ and the perturbation theory generated by the decomposition of ρ into fluctuations $\delta\rho$ and an average ρ_0.

4.5 Fluctuating Nonlinear Hydrodynamics

Before analysing the self-consistent equations for Brownian dynamics we consider the Fluctuating Non-Linear Hydrodynamics and repeat the above derivations. Since the latter follow closely those for BD, we just give the results.

4.5.1 Time-reversal symmetry and fluctuation-dissipation theorem

This time there are four response functions produced by the extra term $\mathcal{F}_{ext} = -\int_x(\rho_x\mu_x + \mathbf{g}_x\cdot\mathbf{p}_x)$

$$\begin{aligned}
\langle\rho(\mathbf{x},t)\rangle_\mu = &\quad \langle\rho(\mathbf{x},t)\rangle_{\mu=0} + \int d^3y\int_s^t du R_{\rho\rho,xy}(t-u)\mu(\mathbf{y},u) && (4.55)\\
&+\int d^3y\int_s^t du R^k_{\rho g,xy}(t-u)p_k(\mathbf{y},u) + o(\mu,\mathbf{p}) && (4.56)\\
\langle g_i(\mathbf{x},t)\rangle_\mu = &\quad \langle g_i(\mathbf{x},t)\rangle_{\mu=0} + \int d^3y\int_s^t du R^{ij}_{gg,xy}(t-u)p_j(\mathbf{y},u) && (4.57)\\
&+\int d^3y\int_s^t du R^i_{g\rho,xy}(t-u)\mu(\mathbf{y},u) + o(\mu,\mathbf{p}) && (4.58)
\end{aligned}$$

which gives:

$$\begin{aligned}
R_{\rho\rho,xy}(t-s) &= \langle\rho(\mathbf{x},t)\nabla\cdot(\rho\hat{\mathbf{g}})(\mathbf{y},s)\rangle && (4.59)\\
R^k_{g\rho,xy}(t-s) &= \langle g_k(\mathbf{x},t)\nabla\cdot(\rho\hat{\mathbf{g}})(\mathbf{y},s)\rangle && (4.60)\\
R^k_{\rho g,xy}(t-s) &= \langle\rho(\mathbf{x},t)\rho(\mathbf{y},s)\nabla_k\hat{\rho}(\mathbf{y},s)\rangle + \langle\rho(\mathbf{x},t)g_i(\mathbf{y},s)\nabla_k\ \hat{g}_i(\mathbf{y},s)\rangle\\
&\quad +\langle\rho(\mathbf{x},t)\nabla_i(g_k\hat{g}_i)(\mathbf{y},s)\rangle - \langle\rho(\mathbf{x},t)L_{ki}\hat{g}_i(\mathbf{y},s)\rangle && (4.61)\\
R^{kl}_{gg,xy}(t-s) &= \langle g_k(\mathbf{x},t)\rho(\mathbf{y},s)\nabla_l\hat{\rho}(\mathbf{y},s)\rangle + \langle g_k(\mathbf{x},t)g_i(\mathbf{y},s)\nabla_l\ \hat{g}_i(\mathbf{y},s)\rangle\\
&\quad +\langle g_k(\mathbf{x},t)\nabla_i(g_l\hat{g}_i)(\mathbf{y},s)\rangle - \langle g_k(\mathbf{x},t)L_{li}\hat{g}_i(\mathbf{y},s)\rangle && (4.62)
\end{aligned}$$

Similarly to the BD case there are two different transformations corresponding to TRS which leave the action invariant and provide FDT. However due to larger number of fields we only consider the simpler transformation which is the analogy of the $\mathcal{U}$ in the case of BD. Derivation within the context of the other transformation becomes very involved technically due to a large number of fields.

The transformation reads:

$$\mathcal{V}: \begin{cases} t & \to -t \\ \mathbf{g}_x & \to -\mathbf{g}_x \\ \hat{\rho}_x & \to -\hat{\rho}_x + \frac{1}{T}\dfrac{\delta \mathcal{F}}{\delta \rho_x} \\ \hat{\mathbf{g}}_x & \to \hat{\mathbf{g}}_x - \frac{1}{T}\dfrac{\delta \mathcal{F}}{\delta \mathbf{g}_x} \end{cases} \tag{4.63}$$

It leaves the action invariant up to boundary terms:

$$\int_x \left[\frac{\delta \mathcal{F}}{\delta \mathbf{g}_x} \cdot \partial_t \mathbf{g}_x + \frac{\delta \mathcal{F}}{\delta \rho_x} \partial_t \rho_x + \nabla_i \left(g_{j,x} \frac{\delta \mathcal{F}}{\delta g_{i,x}} \frac{\delta \mathcal{F}}{\delta g_{j,x}} \right) \right] \tag{4.64}$$

Following the procedure used for BD field theory one can see that density correlation functions are invariant under time-reversal and derive FDT. The naive self-consistent perturbation theory for FNH violates time-reversal symmetry in the same way as it does in case of the BD (see Sec. 4.3.2). The solution to this problem is exactly the same: one should introduce extra fields to linearise the symmetry and to include a response field.

4.5.2 Restoration of time-reversal symmetry in perturbative expansions

In order to make the transformation $\mathcal{V}$ linear we introduce two additional fields $\theta = \frac{\delta \mathcal{F}}{\delta \rho}$ and $\mathbf{v} = \frac{\delta \mathcal{F}}{\delta \mathbf{g}}$. We are lead to add

$$-\int_x \hat{\theta}_x \left[\theta_x - \frac{\delta \mathcal{F}}{\delta \rho_x} \right] - \int_x \hat{\mathbf{v}}_x \cdot \left[\mathbf{v}_x - \frac{\delta \mathcal{F}}{\delta \mathbf{g}_x} \right] \tag{4.65}$$

to the action, which becomes the integral of

$$\begin{aligned} s_x \quad = \quad & \Big\{ -\hat{\rho}_x \left[\partial_t \rho_x + \nabla_i \left(\rho_x v_i \right) \right] + T \hat{g}_{i,x} L_{ij} \hat{g}_{j,x} \\ & -\hat{g}_{i,x} \left[\partial_t g_{i,x} + \rho_x \nabla_i \theta + \nabla_j \left(g_{i,x} v_j \right) + g_{j,x} \nabla_i v_j + L_{ij} v_j \right] \\ & -\hat{\theta}_x \left[\theta_x - \frac{\delta \mathcal{F}}{\delta \rho_x} \right] - \hat{\mathbf{v}}_x \cdot \left[\mathbf{v}_x - \frac{\delta \mathcal{F}}{\delta \mathbf{g}_x} \right] \Big\} \end{aligned} \tag{4.66}$$

The corresponding linear time-reversal transformation reads:

$$\mathcal{V}_1: \begin{cases} t & \to -t \\ \mathbf{g}_x & \to -\mathbf{g}_x \\ \mathbf{v}_x & \to -\mathbf{v}_x \\ \hat{\rho}_x & \to -\hat{\rho}_x + \frac{1}{T}\theta_x \\ \hat{\mathbf{g}}_x & \to \hat{\mathbf{g}}_x - \frac{1}{T}\mathbf{v}_x \\ \hat{\theta}_x & \to \hat{\theta}_x + \frac{1}{T}\partial_t \rho_x \\ \hat{\mathbf{v}}_x & \to -\hat{\mathbf{v}}_x - \frac{1}{T}\partial_t \mathbf{g}_x \end{cases} \tag{4.67}$$

The response functions derived above now read:

$$
\begin{aligned}
R_{\rho\rho,xy}(t-s) &= \langle \rho(\mathbf{x},t)\hat{\theta}(\mathbf{y},s)\rangle \\
R^k_{g\rho,xy}(t-s) &= \langle g_k(\mathbf{x},t)\hat{\theta}(\mathbf{y},s)\rangle \\
R^k_{\rho g,xy}(t-s) &= \langle \rho(\mathbf{x},t)\hat{v}_k(\mathbf{y},s)\rangle \\
R^{kl}_{gg,xy}(t-s) &= \langle g_k(\mathbf{x},t)\hat{v}_l(\mathbf{y},s)\rangle
\end{aligned}
$$

As before, the action is split into a Gaussian and an interaction parts by introducing density fluctuations around the average $\rho = \rho_0 + \delta\rho$: we expand all the terms in powers of $\delta\rho/\rho_0$:

$$
\begin{aligned}
s_{2,x} &= -\hat{\rho}_x(\partial_t\delta\rho_x + \rho_0\nabla\cdot\mathbf{v}_x) - \hat{g}_{i,x}\left[\partial_t g_{i,x} + \rho_0\nabla_i\theta_x + L_{ij}v_{j,x}\right] \\
&\quad + T\hat{g}_{i,x}L_{ij}\hat{g}_{j,x} - \hat{\theta}_x\theta_x + \hat{\theta}_x(W^{FNH}\star\delta\rho)_x - \hat{\mathbf{v}}_x\cdot\mathbf{v}_x + \frac{1}{\rho_0}\hat{\mathbf{v}}_x\cdot\mathbf{g}_x
\end{aligned}
\tag{4.68}
$$

$$
\begin{aligned}
s_{INT,x} &= -\hat{\rho}_x\nabla\cdot(\delta\rho_x\mathbf{v}_x) - \delta\rho_x\hat{\mathbf{g}}_x\cdot\nabla\theta_x - \hat{g}_{i,x}\nabla_j(g_{i,x}v_{j,x}) \\
&\quad - \hat{g}_{i,x}g_{j,x}\nabla_i v_{j,x} - \sum_{p>1}\frac{(-1)^p}{p}\frac{T}{m}\hat{\theta}_x\frac{\delta\rho_x^p}{\rho_0^p} \\
&\quad + \sum_{n>0}(-1)^n\left[(\hat{\mathbf{v}}_x\cdot\mathbf{g}_x)\delta\rho_x + n\hat{\theta}_x\frac{{\mathbf{g}_x}^2}{2}\right]\frac{\delta\rho_x^{n-1}}{\rho_0^{n+1}}
\end{aligned}
\tag{4.69}
$$

with

$$
W^{FNH}(\mathbf{x}) = \frac{T}{m}\left[\frac{1}{\rho_0}\delta(\mathbf{x}) - \frac{c(\mathbf{x})}{m}\right] \tag{4.70}
$$

This expansion produces two series of vertices, each vertex being separately invariant under the transformation $\mathcal{V}_1$. Applying $\mathcal{V}_1$ to two-point quantities generates the Ward-Tahakashi identities for correlators. In particular, we recover FDT relating the response functions and the correlators. The full set of WD identities and dynamical equations is presented in Appendix C.

4.5.3 Static limit

The analysis of the singularities of the SD equations at short time difference gives:

$$C_{\rho\theta}(\mathbf{q},0) = T \tag{4.71}$$
$$C_{gv}(\mathbf{q},0) = T \tag{4.72}$$
$$W(\mathbf{k})C_{\rho\rho}(\mathbf{q},0) = T + \frac{1}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0)C_{\rho\rho}(\mathbf{q},0) \tag{4.73}$$
$$C_{\theta\theta}(\mathbf{q},0) = TW(\mathbf{q}) \tag{4.74}$$
$$\frac{1}{\rho_0}C_{gg}(\mathbf{q},0) = T + \frac{1}{T}\Sigma_{\hat{v}\hat{v}}(\mathbf{q},0)C_{gg}(\mathbf{q},0) \tag{4.75}$$
$$C_{vv}(\mathbf{q},0) = \frac{T}{\rho_0} \tag{4.76}$$

4.6 Large density perturbation theory respecting TRS and the glass transition

This section represents the second major part of the chapter. Previous sections considered the formal symmetry related properties of the field theories for liquids. Their main result is the self-consistent perturbation theories derived in Sec. 4.4.2 and the similar theory for FNH derived in Sec. 4.5. These theories respect time-reversal symmetry and provide a set of closed dynamic equations. In the following we analyse the problem of glass transition and slowing of the dynamics within this formalism aiming at reproducing MCT. Again the case of Brownian dynamics is considered in detail while the analysis for FNH is only sketched. We also compare our results to previous works [81, 28] and unveil the violation of TRS within their context.

4.6.1 BD: The transition

As we have seen in Sec. 2.3 the MCT dynamic transition is characterised by a non-zero limit of the non-ergodic parameter which signalises appearance of an infinite time correlations. The non-ergodic parameter was defined in Sec. 2.3 as:

$$f_{\mathbf{q}} = \frac{1}{S_{\mathbf{q}}} \lim_{\tau\to\infty} C_{\rho\rho}(\mathbf{q},\tau)$$

where $S_{\mathbf{q}} = C_{\rho\rho}(\mathbf{q},0)$ is the static structure factor. Inspection of the limit $\tau \to \infty$ of equations (4.50)-(4.52) implies that only $C_{\rho\rho}$ can have a non-zero limit, the other correlators ($C_{\rho\theta}$,$C_{\theta\theta}$) have strictly zero limits at $\tau \to \infty$ for all values of the temperature. This result has a physical interpretation provided by an idea of a phase space decomposition into multiple ergodic components below the MCT transition (if there is no transition there is only one such component). Indeed $C_{\rho\theta}$,$C_{\theta\theta}$ can be written using their definitions as:

$$C_{\rho\theta}(\mathbf{q},\infty) = \left\langle \rho(-\mathbf{q},0)\frac{\delta\mathcal{F}}{\delta\rho(\mathbf{q},\infty)} \right\rangle = \sum_\alpha \mathcal{P}_\alpha \langle\rho(-\mathbf{q},0)\rangle_\alpha \left\langle \frac{\delta\mathcal{F}}{\delta\rho(\mathbf{q},\infty)} \right\rangle_\alpha$$

$$C_{\theta\theta}(\mathbf{q},\infty) = \left\langle \frac{\delta\mathcal{F}}{\delta\rho(-\mathbf{q},0)}\frac{\delta\mathcal{F}}{\delta\rho(\mathbf{q},\infty)} \right\rangle = \sum_\alpha \mathcal{P}_\alpha \left\langle \frac{\delta\mathcal{F}}{\delta\rho(-\mathbf{q},0)} \right\rangle_\alpha \left\langle \frac{\delta\mathcal{F}}{\delta\rho(\mathbf{q},\infty)} \right\rangle_\alpha$$

where we used the fact that averages decorrelate completely at long times within a fixed ergodic component labelled by α and replaced averages of products by products of averages; the weight of a component is equal to $\mathcal{P}_\alpha$. The average force $\langle\delta\mathcal{F}/\delta\rho(\mathbf{q},\infty)\rangle_\alpha$ should vanish at infinity and therefore $C_{\rho\theta}(\mathbf{q},\infty)$ and $C_{\theta\theta}(\mathbf{q},\infty)$ should vanish too. Note that expressing infinite time averages via static quantities is only possible because our dynamic equations are consistent with TRS which assures the Gibbsian measure in the limit $\tau \to \infty$. Similarly all the self-energies except $\Sigma_{\hat\theta\hat\theta}$ vanish in the limit $\tau \to \infty$. Then there is only one non-ergodic parameter $f_{\mathbf{q}}$ which satisfies an exact equation:

$$\frac{f(\mathbf{q})}{1-f(\mathbf{q})} = \frac{S(\mathbf{q})}{T^2}\Sigma_{\hat\theta\hat\theta}(\mathbf{q},\infty) \tag{4.77}$$

This equation is of course to hard to analyse directly because $\Sigma_{\hat\theta\hat\theta}(\mathbf{q},\infty)$ contains an infinite number of terms which cannot be resummed. One should resort to approximation schemes in order to analyse (4.77).

4.6.2 BD: The Mode-Coupling approximation

The simplest approximation consist in truncating the expansion of self-energies to the lowest order in diagrammatic series. By construction this approximation is consistent with FDT. However there is a difficulty: the term $\int_x \hat\theta_x \log(1+\delta\rho_x/\rho_0)$ which comes from the entropic part of $\mathcal{F}$ gives a non-polynomial interaction. One may expand it in powers of $\delta\rho/\rho_0$. This leads to a series of vertices separately invariant under TRS. Since we are interested in one-loop theory we consider that all these vertices are of the same order. Then there are two possibilities: either we retain all the vertices or we truncate the expansion at the lowest non-trivial order. The first possibility requires a partial resummation the meaning of which is not clear due to the presence of an infinity of tadpoles which contribute to the static vertex renormalisation. For this reason we truncate the expansion at order two which leaves us only one vertex $\hat\theta\delta\rho^2$ but the computation is similar for any other truncation.

As we have pointed out within the field theory statics is consistent with dynamics. It is instructive to check that the above approximation when applied to static equations (4.54) derived from dynamic equations (4.50)-(4.52) coincides with the static theory where the entropic part of $\mathcal{F}$ is truncated at order three in $\delta\rho$:

$$C_{\rho\rho,xy} = \frac{\int \mathcal{D}\delta\rho\, \rho(\mathbf{x})\rho(\mathbf{y}) e^{-\beta\mathcal{F}_3[\delta\rho]}}{\int \mathcal{D}\delta\rho e^{-\beta\mathcal{F}_3[\delta\rho]}}$$

$$\text{with} \quad \mathcal{F}_3[\delta\rho] = \frac{1}{6}\int d^3\mathbf{x}\frac{\delta\rho(\mathbf{x})^3}{\rho_0^3} - \beta\int d^3\mathbf{x}\int d^3\mathbf{x}' W(\mathbf{x}-\mathbf{x}')\delta\rho(\mathbf{x})\delta\rho(\mathbf{x}')$$

where we used $\int_x W(x) = 0$. A direct computation shows that $C_{\rho\rho}(\mathbf{q})$ satisfies the equation (4.54).

The self-energies appearing in Eqs. (4.50)-(4.52) read within the approximation:

$$\Sigma_{\hat{\theta}\theta}(\mathbf{q},t) = \int \frac{d^3\mathbf{k}}{(2\pi)^3}\frac{(\mathbf{q}\cdot\mathbf{k})}{\rho_0^2} C_{\rho\theta}(\mathbf{k},t) C_{\rho\rho}(\mathbf{q}-\mathbf{k},t) \tag{4.78}$$

$$\Sigma_{\hat{\rho}\theta}(\mathbf{q},t) = \frac{1}{2T}\int \frac{d^3\mathbf{k}}{(2\pi)^3}\Big\{ (\mathbf{q}\cdot\mathbf{k})^2 C_{\theta\theta}(\mathbf{k},t) C_{\rho\rho}(\mathbf{q}-\mathbf{k},t) + (\mathbf{q}\cdot\mathbf{k})[\mathbf{q}\cdot(\mathbf{q}-\mathbf{k})] C_{\rho\theta}(\mathbf{k},t) C_{\rho\theta}(\mathbf{q}-\mathbf{k},t)\Big\} \tag{4.79}$$

$$\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},t) = \frac{T^2}{4\rho_0^4}\int \frac{d^3\mathbf{k}}{(2\pi)^3} C_{\rho\rho}(\mathbf{k},t) C_{\rho\rho}(\mathbf{q}-\mathbf{k},t) \tag{4.80}$$

4.6.3 BD: Non-ergodicity parameter

The one-loop expressions for self-energies substituted in (4.77) yield a closed equation on $f_{\mathbf{q}}$:

$$\frac{f(\mathbf{q})}{1-f(\mathbf{q})} = \frac{1}{2\rho_0^4}\int \frac{d^3\mathbf{k}}{(2\pi)^3} f(\mathbf{k}) f(\mathbf{q}-\mathbf{k}) S(\mathbf{q}) S(\mathbf{q}-\mathbf{k}) S(\mathbf{k}) \tag{4.81}$$

This equation looks very much like the corresponding MCT equation (2.21). However it admits only two solutions $f_{\mathbf{q}} = 0$ and $f_{\mathbf{q}} = 1$. The first solution corresponds to the liquid phase where ergodicity is not broken. The second solution suffers from several important problems:

- $f_{\mathbf{q}} = 1$ implies $C_{\rho\rho}(\mathbf{q},\tau) = S(\mathbf{q})$ for all times. This is clearly unphysical and should be rejected.
- The integral in the right hand side of Eq. (4.81) is divergent. The wave-vector dependence of the vertex $V(\mathbf{k},\mathbf{q}-\mathbf{k})$ given by Eq. (2.16) have ensured the convergence within MCT. However this vertex is absent in Eq. (4.81). Furthermore Eq. (4.54) for the structure factor $S(\mathbf{q})$ is also ill-defined for the same reason.

The numerical solution of Eq. (4.81) with an implied cut-off for large $\mathbf{k}$ is remarkably similar to that of MCT. As the cut-off is sent to infinity the solution

seems to converge to $f_{\mathbf{q}} = 1$ [91]. Although there is a physical cut-off for the description in terms of Langevin dynamics providing Eq. (4.3) which is the foundation of Brownian dynamics this cut-off dependence is clearly unphysical. We will come back to this problem at the end of the Chapter.

4.6.4 FNH: Equation for the non-ergodicity parameter

In the following we repeat the above analysis for the Fluctuating Non-linear Hydrodynamics. The only difference with respect to BD is the larger set of dynamic equations (see Appendix C), but their formal structure is very similar to the BD dynamic equations (4.50)-(4.52).

We focus on the equation for the non-ergodicity parameter. Due to long time decorrelation inside ergodic components, only $C_{\rho\rho}$, $C_{\rho g^\perp}$ and $C_{g^\perp g^\perp}$ where $\mathbf{g}^\perp$ is a transverse current, can have a non-zero limit as $\tau \to \infty$. This is also confirmed by the analysis of limit $\tau \to \infty$ of the dynamical equations given in Appendix C. However frozen currents are not expected to exist in glasses; therefore we assume that $C_{\rho g}(\mathbf{q}, \infty)$ and $C_{gg}(\mathbf{q}, \infty)$ also vanish. Since at least one of these appear in any diagram contributing to $\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q}, \infty)$, $\Sigma_{\hat{\rho}\hat{v}}(\mathbf{q}, \infty)$, $\Sigma_{\hat{g}\hat{\theta}}(\mathbf{q}, \infty)$, $\Sigma_{\hat{g}\hat{v}}(\mathbf{q}, \infty)$ and $\Sigma_{\hat{\theta}\hat{v}}(\mathbf{q}, \infty)$ these self-energies also vanish. Consequently only $\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q}, \infty)$ may have a non zero value. The in the limit $\tau \to \infty$ one obtains the exact equation for non-ergodicity parameter using (4.159) and its static limit (4.73):

$$\frac{f(\mathbf{q})}{1 - f(\mathbf{q})} = \frac{S_{\mathbf{q}}}{T^2} \Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q}, \infty) \tag{4.82}$$

This equation looks identical to Eq. (4.77). Any general approximation (one loop, two loops, etc) for the self-energy on the right hand side, leads to a non-linear equation on $f(\mathbf{q})$. As we will discuss later previous works have obtained very different structures because the time-reversal symmetry was violated. This may be very dangerous because it can generate spurious results. For example in the analysis of [28] this modified strongly the general structure of the Schwinger-Dyson equations and implied that the non-ergodic parameter had to vanish. However, this has nothing to do with the physical mechanism that cuts off the MCT transition; it is just an artifact of the violation of the time-reversal symmetry.

4.6.5 FNH: Mode-Coupling approximation

We restrict ourselves to the approximation similar to the one used for BD, truncating the series in $\delta\rho/\rho_0$ in Eq. (4.69) to the lowest order:

$$\begin{aligned} s_{INT,x} &= -\hat{\rho}_x \nabla \cdot (\delta\rho_x \mathbf{v}_x) - \delta\rho_x \hat{\mathbf{g}}_x \cdot \nabla\theta_x - \hat{g}_{i,x} \nabla_j (g_{i,x} v_{j,x}) \\ &\quad -\hat{g}_{i,x} g_{j,x} \nabla_i v_{j,x} - \hat{\mathbf{v}}_x \cdot \mathbf{g}_x \frac{\delta\rho_x}{\rho_0^2} - \hat{\theta}_x \frac{{\mathbf{g}_x}^2}{2\rho_0^2} - \frac{T}{2m} \hat{\theta}_x \frac{\delta\rho_x^2}{\rho_0^2} \end{aligned} \tag{4.83}$$

We do not write the set of all equations at one-loop level. Again the static correlation functions can be obtained from dynamical equations and coincide

with the ones obtained within the same approximation in static theory. At one-loop, the expression of $\Sigma_{\hat{\theta}\hat{\theta}}$ coincides with the one-loop expression obtained within BD (up to a multiplication by the mass). However this is a coincidence which is absent when higher order corrections are considered. Finally we get the same equation (4.81) on non-ergodic parameter which suffers from the same problems. However contrary to BD there is a natural large wave-vector cutoff within FNH which regularises the integrals over $\mathbf{k}$ because FNH is a long- and intermediate lengthscales description which is not valid on short lengthscales.

4.6.6 Relation with previous works

In this section we would like to put our work in the context of field theory derivations of MCT [85, 28] and discuss what can be learnt from the non-perturbative structure of the equations which we derived, and from the resulting mode-coupling equations.

The first issue we would like to discuss is derivations of the original mode-coupling equation (2.19) within a context of a field theory that one can find in the literature [85, 28]. They are all inconsistent with time-reversal symmetry and FDT because some additional identities related to time-reversal symmetry were assumed in the derivation. But these identities are incompatible with the same mode-coupling equations as used to derive MCT.

First, Kawasaki and Miyazima [85] considered Brownian dynamics with the potential U replaced by the direct correlation function c and the original perturbation theory introduced in Sec. 4.3.2. In their derivation they computed $\Sigma_{\hat{\rho}\hat{\rho}}$ and assumed that $\Sigma_{\hat{\rho}\rho}$ is related to it by time reversal symmetry:

$$\Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau) = -\frac{\Theta(\tau)}{T}\partial_\tau \Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau).$$

However the direct one-loop computation of both sides of this equations shows that this is not true. Furthermore such an identity together with the structure of Eqs. (4.29) imply FDT between $C_{\rho\rho}$ and the naive response $\mathcal{G}$ but as we have seen $\mathcal{G}$ is not a valid response function. The field theory which respects TRS in perturbation that we developed in Sec. 4.4.2 also predicts a different identity for $\Sigma_{\hat{\rho}\rho}$:

$$\Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau) = -\frac{1}{T}\frac{\partial}{\partial\tau}\left[\Theta(\tau)\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau)\right].$$

The second derivation by Das and Mazenko [28] focused on FNH. A similar inconsistency is present in their analysis where a linear relation is forced between $C_{\rho\rho}$ and $C_{\rho\hat{\rho}}$ to close the equations:

$$C_{\rho\hat{\rho}}(\mathbf{q},\tau) \sim \frac{\Theta(\tau)}{T} C_{\rho\rho}(\mathbf{q},\tau) \tag{4.84}$$

which is only valid in the hydrodynamic limit. However it holds only in the hydrodynamic limit *i.e.* for long time- and lengthscales. As we have seen time-

reversal symmetry implies a different identity derived in 4.5 for $C_{\rho\hat{\rho}}$:

$$\frac{1}{T}\langle\rho(\mathbf{x},t)\frac{\delta\mathcal{F}[\rho,\mathbf{g}]}{\delta\rho(\mathbf{y},s)}\rangle = \Theta(t-s)C_{\rho\hat{\rho},xy}(t-s) + \Theta(s-t)C_{\rho\hat{\rho},yx}(s-t)$$

where $\mathcal{F}$ is the free energy function of FNH (see 4.1.2).

Forcing a relation is useful to close the equations but it has several drawbacks:

- This can lead to inconsistencies and spurious results.
- This breaks time-reversal symmetry: we have seen that it predicted different identities in both cases.

In contrast the formalism developed in Sec. 4.4.2 and Sec. 4.5 provides dynamic equations closed by self-consistent series that automatically respect time-reversal symmetry and FDT. This is important for the consistency of the theory and for the analysis of out-of-equilibrium situation as we have pointed out earlier.

There is another issue related to results of Das and Mazenko. Based on results derived within FNH they insisted that corrections coming from the coupling to currents cut off the transition. We have also discussed the projection operator counterpart of this theory in Sec. 2.7 which predicted the same result. This implies that the cut off mechanism is different for Brownian particles described by BD and Newtonian particles described by FNH since no currents is present in the first case. However as we have discussed at the end of Sec. 2.7 numerical simulations do not confirm this conclusion and indicate that the mechanism is the same for both types of dynamics [71, 70, 61]. The result of [28] is an artifact of the forced relation (4.84) which violates time-reversal symmetry. Indeed forcing this relation in dynamic equations for FNH consistent with TRS (presented in Appendix C) implies the zero non-ergodic parameter for all temperatures. Let's consider one of the SD equations (from Appendix C):

$$\begin{aligned}
&\partial_\tau C_{\rho\rho}(\mathbf{q},\tau) - i\rho_0\mathbf{q}C_{v\rho}(\mathbf{q},\tau) = \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u)\\
+\quad &\frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\rho}(\mathbf{q},u) - \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u)C_{\theta\rho}(\mathbf{q},u)\\
-\quad &\frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{g}}(\mathbf{q},\tau-u)C_{v\rho}(\mathbf{q},u)
\end{aligned} \tag{4.85}$$

If $C_{\rho\hat{\rho}}$ is related to $C_{\rho\rho}$ by (4.84) and $f_{\mathbf{q}} \neq 0$ then $C_{\rho\hat{\rho}}$ also has a non-zero infinite value. Recalling an identity for $C_{\rho\hat{\rho}}$ implied by TRS (see Appendix C):

$$C_{\rho\rho}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T}C_{\rho\theta}(\mathbf{q},\tau)$$

we see that a contradiction appears: in the limit $\tau \to \infty$ the left hand side of Eq. (4.85) tends to zero while the right hand side remains non-zero. Therefore we should conclude that $f_{\mathbf{q}} \equiv 0$ always. However this is only due to the enforced relation (4.84) which is incompatible with TRS. Consequently the cut off of the transition is spurious.

Finally the authors of [86] considered quadratic density functional and also predicted the absence of the transition. We considered this case in Sec. 4.3.3. Indeed within this theory the straightforward perturbation theory respects TRS; the limit $\tau \to \infty$ of the dynamic equation (4.35) implies $f_{\mathbf{q}} \equiv 0$. However the transition is absent because of a too simple structure of the theory: $\Sigma_{\hat{\theta}\hat{\theta}} \equiv 0$ in this theory. If one considers non-quadratic functionals then this is no more true.

4.6.7 Discussion of the ultraviolet divergence

We now come back to the discussion of the large wave-vector divergence presented in Sec. 4.6.3. This problem was addressed recently in a new work by Kim and Kawasaki [93]. They reconsidered the field theory for Brownian dynamics given by Eq. (4.42) but used a different definition for the field θ: $\theta_{KK} = \theta - W\delta\rho$ *i.e.* they extracted the linear part of $\delta\mathcal{F}/\delta\rho$. They suggested an approximation scheme which have led to the following equation for the non-ergodic parameter at one-loop order:

$$\frac{f(\mathbf{q})}{1-f(\mathbf{q})} = \frac{1}{2\rho_0 T^2 \mathbf{q}^4} \int \frac{d^3\mathbf{k}}{(2\pi)^3} |V(\mathbf{q},\mathbf{k})|^2 S(\mathbf{q}) S(\mathbf{k}) S_(\mathbf{q}-\mathbf{k}) f(\mathbf{k}) f(\mathbf{q}-\mathbf{k})$$
$$V(\mathbf{q},\mathbf{k}) = (\mathbf{q}\cdot\mathbf{k})U(\mathbf{k}) + (\mathbf{q}\cdot(\mathbf{q}-\mathbf{k})U(\mathbf{q}-\mathbf{k}))$$

which is similar to Eq. (2.21) and does not suffer from the divergence problem. However there are several problems in their derivation of this result:

- The derivation employed an identity:

$$\nabla\left(\delta\rho\nabla\frac{\delta\mathcal{S}}{\delta\rho}\right) = T\nabla^2\delta\rho = \nabla\left(\rho\nabla(\theta - U\rho)\right)$$

  where $\mathcal{S}$ is given by Eq. (4.4). This non-perturbative identity implies a relation between the terms of the action (4.42) and is only valid as an insertion in perturbation theory. However Kim and Kawasaki used it to eliminate certain vertices at one loop. But the identity mixes Gaussian and interaction terms and therefore violates the time-reversal symmetry. To restore TRS one should either include all the terms in the Gaussian part and therefore treat the non-linearities non-perturbatively, or include all the terms in interaction where quadratic terms become line insertions. In both cases the new decomposition of the action should respect TRS because as we have seen throughout this Chapter the preservation of FDT in perturbation theory is a key component of a correct field theory.

- The results of Kim and Kawasaki are one-loop only and extension to higher orders is not trivial. However analysis of systematic corrections is one the primary aims of the introduction of the field theory.
- Actually the problem of the divergence is not solved because the vertices $\theta\delta\rho^n$ are still present. They do not appear at one loop, but they intervene at higher orders leading to the same divergences.

In this context the results of Kim and Kawasaki do not solve the UV divergence problem and even reintroduce the TRS preservation issue. Nevertheless the results are promising since they provide a correct equation, and require further analysis.

4.7 Summary

In this Chapter we focused on the rederivation of MCT within field theories considered in earlier works [85, 28] and revealed problems within previous attempts of rederivation. We found that straightforward perturbation theories usually violate time-reversal symmetry. This violation is caused by the non-linear structure of the transformations corresponding to the symmetry. Our approach lead to self-consistent perturbation theories respecting TRS automatically and dynamic equations for BD and FNH. This allows to analyse various self-consistent approximations without worrying about the preservation of the symmetry. Another advantage of the approach is that dynamics and statics are automatically connected.

Both theories predict the same equation for the non-ergodic parameter:

$$\frac{f_{\mathbf{q}}}{1-f_{\mathbf{q}}} = \frac{S_{\mathbf{q}}}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\infty).$$

Any approximation used to compute $\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\infty)$ would result in a non-linear functional of $f_{\mathbf{q}}$ in the right hand side of the above equation. There are no reasons which suppress the existence of a solution different from $f_{\mathbf{q}} \equiv 0$. The persistence of a transition for higher orders was verified for a toy model [94] such as a Langevin particle in a double well potential. This sets a problem of analysis of a higher loop corrections to the one-loop results within BD and FNH. This will be the main content of Chapter 6 where we will prove that the transition persists at any order. This suggests that the coupling to currents is not responsible for the cut off of the transition, and the cut off mechanism is non-perturbative and cannot be captured within any order of self-consistent perturbation theory. This was already found in a different context [69].

However there is a problem within our formalism: the ill-defined vertex which causes a divergence for large wave-vectors $\mathbf{k}$. This is an important issue because the structure of MCT vertex is responsible for the quantitative success of MCT. This divergence also requires to work with a cut off which regularises the integrals and is not well justified for BD. Convergence of integrals involving

the MCT vertex (2.16) is ensured by the direct correlation function appearing in the vertex. Our vertex is different and does not contain any potential related factor. This motivates to look for an interpretation of the perturbation theory generated by the decomposition given by Eqs. (4.44)-(4.45) (BD) or Eqs. (4.68)-(4.69) (FNH). In fact this perturbation is identified with the large density nature expansion. The decomposition $\rho = \rho_0 + \delta\rho$ together with the expansion of the logarithm $\log(1 + \delta\rho/\rho_0)$ is valid for large densities ρ; the vertices $\hat{\theta}\delta\rho^n$ in the interaction s_{INT} (4.45) are generated by the expansion of entropic part of the free energy $\mathcal{F}$(4.4) which dominates for large densities. Description of liquids for large densities is a problem of its own which is far from being solved. Therefore perturbing around this state should be done with much care.

We will address this problem in the next Chapter where we consider different perturbation theories which also respect TRS and try to fix the problem. Before concluding let's mention a possible alternative that we do not consider in the present work. The above perturbation scheme perturbs around a uniform density profile ρ_0. However we expect that liquid is trapped in an amorphous configuration close to the glass transition. Such configurations are characterised by a non-uniform density profile. This suggests to analyse perturbation around a static non-uniform profile $\rho_0(\mathbf{x})$ and repeat the derivations of this chapter within this context.

Appendix A. Derivation of dynamical and static equations

In this appendix we sketch the derivation of equations (4.51) and (4.54) (the fourth one) as an example. Other dynamical or static equations can be obtain following the same routes. We start from the SD equation

$$(G_0^{-1} \cdot G - \Sigma \cdot G)_{\hat{\rho}\hat{\rho}}(\mathbf{q}, \tau) = \delta(\tau) \tag{4.86}$$

for any value of τ. We have

$$(\Sigma \cdot G)_{\hat{\rho}\hat{\rho}}(\mathbf{q}, \tau) = (\Sigma_{\hat{\rho}\rho} \cdot C_{\rho\hat{\rho}})(\mathbf{q}, \tau) + (\Sigma_{\hat{\rho}\theta} \cdot C_{\theta\hat{\rho}})(\mathbf{q}, \tau) \tag{4.87}$$

Indeed $C_{\hat{\rho}\hat{\rho}}$ and $C_{\hat{\theta}\hat{\rho}}$ vanish by causality. We then get:

$$\begin{aligned}(\Sigma \cdot G)_{\hat{\rho}\hat{\rho}}(\mathbf{q}, \tau) = \; & \tfrac{\Theta(\tau)}{T} \int_0^\tau du \, \left[\partial_\tau \Sigma_{\hat{\theta}\theta}(\mathbf{q}, \tau - u) C_{\rho\theta}(\mathbf{q}, u)\right. \\ & \left. + \Sigma_{\hat{\rho}\theta}(\mathbf{q}, \tau - t) C_{\theta\theta}(\mathbf{q}, t)\right]. \end{aligned} \tag{4.88}$$

Integrating by parts, one gets

$$\begin{aligned}(\Sigma \cdot G)_{\hat{\rho}\hat{\rho}}(\mathbf{q}, \tau) = \tfrac{\Sigma_{\hat{\theta}\theta}(\mathbf{q}, \tau)}{T} C_{\rho\theta}(\mathbf{q}, \tau = 0) \\ + \tfrac{\Theta(\tau)}{T} \int_0^\tau du \, \left[\Sigma_{\hat{\theta}\theta}(\mathbf{q}, \tau - u) \partial_u C_{\rho\theta}(\mathbf{q}, u) + \Sigma_{\hat{\rho}\theta}(\mathbf{q}, \tau - u) C_{\theta\theta}(\mathbf{q}, u)\right] \end{aligned} \tag{4.89}$$

In addition we have

$$\begin{aligned}(G_0^{-1}\cdot G)_{\hat\rho\hat\rho}(\mathbf{q},\tau) &= \partial_\tau C_{\rho\hat\rho}(\mathbf{q},\tau)+\rho_0\mathbf{q}^2 C_{\theta\hat\rho}(\mathbf{q},\tau)\\ &= \tfrac{1}{T}\partial_\tau\left(\Theta(\tau^+)C_{\rho\theta}(\mathbf{q},\tau)\right)+\tfrac{\rho_0\mathbf{q}^2}{T}C_{\theta\theta}(\mathbf{q},\tau)\end{aligned}\qquad(4.90)$$

Equating the terms proportional to $\delta(\tau)$ in (4.86) one gets

$$\frac{1}{T}C_{\rho\theta}(\mathbf{q},0)=1,\qquad(4.91)$$

and taking the limit $\tau\to 0^+$, gives (4.54). Finally (4.51) is obtained by taking $\tau>0$. All other equations for the dynamical evolutions and the statics can be derived in the same way. When causality is not enough to restrict explicitly time integrals between 0 and τ, one can verify that in all cases FDT makes it possible to combine together different contributions of the same equations to finally end up with integrals between 0 and τ.

Let us add here that careful analysis of the self-energies shows that $\Sigma_{\hat\theta\rho}$ has a tadpole contribution. However this tadpole can be eliminated by adding a linear term $-A\int d^3\mathbf{x}\,\delta\rho(\mathbf{x})$ to the entropic part of the free energy and A to the potential, with a suitable value of the constant A.

Appendix B. Proof of the linear dependence of the Schwinger-Dyson equations

As we have already mentioned, it may appear unnatural to describe the evolution of 3 correlators with 4 dynamical equations. A series expansions at low $\tau>0$ of these equations makes this clearer. When expanded in series, they become a cascade of equations for the successive derivatives of the correlators at zero time difference. We start by expanding at first order, from which it is easy to guess what is going on at higher orders. At order 1 in τ, (4.50)-(4.53) read respectively:

$$\dot{C}_{\rho\rho}(\mathbf{q},0) + \rho_0\mathbf{q}^2 C_{\rho\theta}(\mathbf{q},0) + \tau\left[\ddot{C}_{\rho\rho}(\mathbf{q},0) + \rho_0\mathbf{q}^2\dot{C}_{\rho\theta}(\mathbf{q},0)\right] =$$
$$\tau\left[\Sigma_{\hat{\theta}\theta}(\mathbf{q},0^+)\dot{C}_{\rho\rho}(\mathbf{q},0) + \Sigma_{\hat{\rho}\theta}(\mathbf{q},0^+)C_{\rho\theta}(\mathbf{q},0)\right] \quad (4.92)$$

$$\dot{C}_{\rho\theta}(\mathbf{q},0) + \rho_0\mathbf{q}^2 C_{\theta\theta}(\mathbf{q},0) + \tau\left[\ddot{C}_{\rho\theta}(\mathbf{q},0) + \rho_0\mathbf{q}^2\dot{C}_{\theta\theta}(\mathbf{q},0)\right]$$
$$-\Sigma_{\hat{\theta}\theta}(\mathbf{q},0^+)C_{\rho\theta}(\mathbf{q},0) + \tau\left[\Sigma_{\hat{\rho}\theta}(\mathbf{q},0)C_{\theta\theta}(\mathbf{q},0) - \dot{\Sigma}_{\hat{\theta}\theta}(\mathbf{q},0^+)C_{\rho\theta}(\mathbf{q},0)\right] \quad (4.93)$$

$$W(\mathbf{q})C_{\rho\rho}(\mathbf{q},0) - C_{\rho\theta}(\mathbf{q},0) + \tau\left[W(\mathbf{q})\dot{C}_{\rho\rho}(\mathbf{q},0) - \dot{C}_{\rho\theta}(\mathbf{q},0)\right] =$$
$$\frac{1}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0)C_{\rho\rho}(\mathbf{q},0) + \tau\,\Sigma_{\hat{\theta}\theta}(\mathbf{q},0^+)C_{\rho\theta}(\mathbf{q},0) \quad (4.94)$$

$$W(\mathbf{q})C_{\rho\theta}(\mathbf{q},0) - C_{\theta\theta}(\mathbf{q},0) + \tau\left[W(\mathbf{q})\dot{C}_{\rho\theta}(\mathbf{q},0) - \dot{C}_{\theta\theta}(\mathbf{q},0)\right] =$$
$$\tau\left[\Sigma_{\hat{\theta}\theta}(\mathbf{q},0^+)C_{\theta\theta}(\mathbf{q},0) - \frac{1}{T}\dot{\Sigma}_{\hat{\theta}\hat{\theta}}(\mathbf{q},0)C_{\rho\theta}(\mathbf{q},0)\right] \quad (4.95)$$

In addition, the SD equations have an apparent singularity at $\tau = 0$ which comes from the $\delta(\tau)$ in the RHS of (4.48). This gives an initial condition: $C_{\rho\theta}(\mathbf{q},0^+) = T$. Thus there are 5 equations at order 0, which fix the values of $C_{\rho\rho}(\mathbf{q},0)$, $C_{\rho\theta}(\mathbf{q},0)$, $C_{\theta\theta}(\mathbf{q},0)$, $\dot{C}_{\rho\rho}(\mathbf{q},0)$ and $\dot{C}_{\rho\theta}(\mathbf{q},0)$. At order 1, there are 4 equations but 3 quantities only to be determined, namely $\dot{C}_{\theta\theta}(\mathbf{q},0)$, $\ddot{C}_{\rho\rho}(\mathbf{q},0)$ and $\ddot{C}_{\rho\theta}(\mathbf{q},0)$. Remark that the self-energies and their first derivatives appear in the equations. However, as they can be expressed in terms of the correlators, it can be checked that the successive derivatives of the self-energies can be expressed in terms of the quantities already computed at previous orders. This guarantees that at every order self-energies do not give extra variables to be determined. Now we show that in fact one of the equations obtained by identifying the terms of order τ is trivially satisfied by the solution of the equations at order 0. We focus on the term proportional to τ in the LHS of (4.94). We then express this term by using a linear combination of the terms of order 0 of (4.92) and (4.93):

$$W(\mathbf{q})\dot{C}_{\rho\rho}(\mathbf{q},0) - \dot{C}_{\rho\theta}(\mathbf{q},0) = -\rho_0\mathbf{q}^2\left[W(\mathbf{q})C_{\rho\rho}(\mathbf{q},0) - C_{\rho\theta}(\mathbf{q},0)\right] \quad (4.96)$$
$$+\Sigma_{\hat{\theta}\theta}(\mathbf{q},0^+)C_{\theta\theta}(\mathbf{q},0)$$

From order 0 of (4.94), the terms in brackets vanishes, and then we get:

$$W(\mathbf{q})\dot{C}_{\rho\rho}(\mathbf{q},t) - \dot{C}_{\rho\theta}(\mathbf{q},t)\Sigma_{\hat{\theta}\theta}(\mathbf{q},0^+)C_{\rho\theta}(\mathbf{q},0), \quad (4.97)$$

which corresponds to the terms proportional to τ in (4.94). Therefore the number of equations obtained at order τ is equal to the number of variables to be determined at this order.

The non perturbative generalisation of the previous approach comes from the following remark: the SD equations form a linear system of equations which

unknown variables are the correlators and coefficients are the components of G_0^{-1} and Σ. The solution of this system of equations is found easily using the Laplace transform. The SD equations read in Laplace transform:

$$C_{\rho\rho}(\mathbf{q},0^+)\left(1+\frac{\hat{\Sigma}_{\hat{\rho}\hat{\theta}}(\mathbf{q},z)}{T}\right)z\left(1+\frac{\hat{\Sigma}_{\hat{\rho}\hat{\theta}}(\mathbf{q},z)}{T}\right)\hat{C}_{\rho\rho}(\mathbf{q},z) + (\rho_0\mathbf{q}^2+\frac{\hat{\Sigma}_{\hat{\rho}\hat{\rho}}(\mathbf{q},z)}{T})\hat{C}_{\rho\theta}(\mathbf{q},z) \tag{4.98}$$

$$z\left(1+\frac{\hat{\Sigma}_{\hat{\rho}\hat{\theta}}(\mathbf{q},z)}{T}\right)\hat{C}_{\rho\theta}(\mathbf{q},z)+(\rho_0\mathbf{q}^2+\frac{\hat{\Sigma}_{\hat{\rho}\hat{\rho}}(\mathbf{q},z)}{T})\hat{C}_{\theta\theta}(\mathbf{q},z)=T \tag{4.99}$$

$$\begin{aligned}\frac{1}{T}\hat{\Sigma}_{\hat{\theta}\hat{\theta}}(\mathbf{q},z)C_{\rho\rho}(\mathbf{q},0^+) &= -\left(1+\frac{\hat{\Sigma}_{\hat{\rho}\hat{\theta}}(\mathbf{q},z)}{T}\right)\hat{C}_{\rho\theta}(\mathbf{q},z)\\ &\quad+\left(W(\mathbf{q})+\frac{z\hat{\Sigma}_{\hat{\theta}\hat{\theta}}(\mathbf{q},z)-\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0^+)}{T}\right)\hat{C}_{\rho\rho}(\mathbf{q},z)\end{aligned} \tag{4.100}$$

$$W(\mathbf{q})\ \hat{C}_{\rho\theta}(\mathbf{q},z)=\left(1+\frac{\hat{\Sigma}_{\hat{\rho}\hat{\theta}}(\mathbf{q},z)}{T}\right)\hat{C}_{\theta\theta}(\mathbf{q},z) - \frac{z\hat{\Sigma}_{\hat{\theta}\hat{\theta}}(\mathbf{q},z)-\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau=0)}{T}\ \hat{C}_{\rho\theta}(\mathbf{q},z) \tag{4.101}$$

For better clarity, we write formally this system as follows:

$$\begin{aligned}AC_{\rho\rho}(\mathbf{q},0) &= zA\hat{C}_{\rho\rho}(\mathbf{q},z)+B\hat{C}_{\rho\theta}(\mathbf{q},z) && (4.102)\\ T &= zA\hat{C}_{\rho\theta}(\mathbf{q},z)+B\hat{C}_{\theta\theta}(\mathbf{q},z) && (4.103)\\ DC_{\rho\rho}(\mathbf{q},0) &= E\hat{C}_{\rho\rho}(\mathbf{q},z)-A\hat{C}_{\rho\theta}(\mathbf{q},z) && (4.104)\\ 0 &= E\hat{C}_{\rho\theta}(\mathbf{q},z)-A\hat{C}_{\theta\theta}(\mathbf{q},z) && (4.105)\end{aligned}$$

The identity $E\ (RHS)_1 - A\ (RHS)_2 - zA\ (RHS)_3 - B\ (RHS)_4 = 0$, where $(RHS)_i$ stands for the RHS of the ith equation above, is trivially verified. It remains to prove that the LHS are linked by the same relation. Gathering the terms of $E\ (LHS)_1 - A\ (LHS)_2 - zA\ (LHS)_3 - B\ (LHS)_4 = 0$ (with obvious notation), one gets:

$$\left[W(\mathbf{q})-\frac{1}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0^+)\right]C_{\rho\rho}(\mathbf{q},0)=T \tag{4.106}$$

This is precisely the static equation (4.54), and the proof is complete.

Appendix C. Dynamical equations for Fluctuating Nonlinear Hydrodynamics

In this appendix we give the derivation of the dynamic equations for fluctuating nonlinear hydrodynamics. The calculus and the ideas behind are the same as the corresponding for BD although somewhat more cumbersome due to a larger number of fields. We start with the Schwinger-Dyson equations and use time-reversal to simplify them.

This time (4.18) applied to the transformation $\mathcal{V}_1$ gives the following equations for correlators:

$$C_{\rho\hat{\rho}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} C_{\rho\theta}(\mathbf{q},\tau) \tag{4.107}$$

$$C_{\rho\hat{g}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} C_{\rho v}(\mathbf{q},\tau) \tag{4.108}$$

$$C_{\rho\hat{\theta}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} \partial_\tau C_{\rho\rho}(\mathbf{q},\tau) \tag{4.109}$$

$$C_{\rho\hat{v}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} \partial_\tau C_{\rho g}(\mathbf{q},\tau) \tag{4.110}$$

$$C_{g\hat{\rho}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} C_{g\theta}(\mathbf{q},\tau) \tag{4.111}$$

$$C_{g\hat{g}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} C_{gv}(\mathbf{q},\tau) \tag{4.112}$$

$$C_{g\hat{\theta}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} \partial_\tau C_{g\rho}(\mathbf{q},\tau) \tag{4.113}$$

$$C_{g\hat{v}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} \partial_\tau C_{gg}(\mathbf{q},\tau) \tag{4.114}$$

$$C_{\theta\hat{\rho}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} C_{\theta\theta}(\mathbf{q},\tau) \tag{4.115}$$

$$C_{\theta\hat{g}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} C_{\theta v}(\mathbf{q},\tau) \tag{4.116}$$

$$C_{\theta\hat{\theta}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} \partial_\tau C_{\theta\rho}(\mathbf{q},\tau) \tag{4.117}$$

$$C_{\theta\hat{v}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T} \partial_\tau C_{\theta g}(\mathbf{q},\tau) \tag{4.118}$$

$$C_{v\hat{\rho}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T}C_{v\theta}(\mathbf{q},\tau) \quad (4.119)$$

$$C_{v\hat{g}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T}C_{vv}(\mathbf{q},\tau) \quad (4.120)$$

$$C_{v\hat{\theta}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T}\partial_\tau C_{v\rho}(\mathbf{q},\tau) \quad (4.121)$$

$$C_{v\hat{v}}(\mathbf{q},\tau) = \frac{\Theta(\tau)}{T}\partial_\tau C_{vg}(\mathbf{q},\tau) \quad (4.122)$$

and (4.19) yields the following identities for self-energies:

$$\Sigma_{\hat{\rho}\rho}(\mathbf{q},\tau) = \frac{1}{T}\partial_\tau\left[\Theta(\tau)\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau)\right] \quad (4.123)$$

$$\Sigma_{\hat{\rho}g}(\mathbf{q},\tau) = \frac{1}{T}\partial_\tau\left[\Theta(\tau)\Sigma_{\hat{\rho}\hat{v}}(\mathbf{q},\tau)\right] \quad (4.124)$$

$$\Sigma_{\hat{\rho}\theta}(\mathbf{q},\tau) = -\frac{1}{T}\Theta(\tau)\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau) \quad (4.125)$$

$$\Sigma_{\hat{\rho}v}(\mathbf{q},\tau) = -\frac{1}{T}\Theta(\tau)\Sigma_{\hat{\rho}\hat{g}}(\mathbf{q},\tau) \quad (4.126)$$

$$\Sigma_{\hat{g}\rho}(\mathbf{q},\tau) = \frac{1}{T}\partial_\tau\left[\Theta(\tau)\Sigma_{\hat{g}\hat{\theta}}(\mathbf{q},\tau)\right] \quad (4.127)$$

$$\Sigma_{\hat{g}g}(\mathbf{q},\tau) = \frac{1}{T}\partial_\tau\left[\Theta(\tau)\Sigma_{\hat{g}\hat{v}}(\mathbf{q},\tau)\right] \quad (4.128)$$

$$\Sigma_{\hat{g}\theta}(\mathbf{q},\tau) = -\frac{1}{T}\Theta(\tau)\Sigma_{\hat{g}\hat{\rho}}(\mathbf{q},\tau) \quad (4.129)$$

$$\Sigma_{\hat{g}v}(\mathbf{q},\tau) = -\frac{1}{T}\Theta(\tau)\Sigma_{\hat{g}\hat{g}}(\mathbf{q},\tau) \quad (4.130)$$

$$\Sigma_{\hat{\theta}\rho}(\mathbf{q},\tau) = \frac{1}{T}\Theta(\tau)\partial_\tau\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau) \quad (4.131)$$

$$\Sigma_{\hat{\theta}g}(\mathbf{q},\tau) = \frac{1}{T}\Theta(\tau)\partial_\tau\Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},\tau) \quad (4.132)$$

$$\Sigma_{\hat{\theta}\theta}(\mathbf{q},\tau) = -\frac{1}{T}\Theta(\tau)\Sigma_{\hat{\theta}\hat{\rho}}(\mathbf{q},\tau) \quad (4.133)$$

$$\Sigma_{\hat{\theta}v}(\mathbf{q},\tau) = -\frac{1}{T}\Theta(\tau)\Sigma_{\hat{\theta}\hat{g}}(\mathbf{q},\tau) \quad (4.134)$$

$$\Sigma_{\hat{v}\rho}(\mathbf{q},\tau) = \frac{1}{T}\Theta(\tau)\partial_\tau\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},\tau) \quad (4.135)$$

$$\Sigma_{\hat{v}g}(\mathbf{q},\tau) = \frac{1}{T}\Theta(\tau)\partial_\tau\Sigma_{\hat{v}\hat{v}}(\mathbf{q},\tau) \quad (4.136)$$

$$\Sigma_{\hat{v}\theta}(\mathbf{q},\tau) = -\frac{1}{T}\Theta(\tau)\Sigma_{\hat{v}\hat{\rho}}(\mathbf{q},\tau) \quad (4.137)$$

$$\Sigma_{\hat{v}v}(\mathbf{q},\tau) = -\frac{1}{T}\Theta(\tau)\Sigma_{\hat{v}\hat{g}}(\mathbf{q},\tau). \quad (4.138)$$

One can get some additional identities:

$$\begin{aligned}
\Sigma_{\hat{g}\hat{\rho}}(\mathbf{q},\tau) &= \Sigma_{\hat{\rho}\hat{g}}(\mathbf{q},\tau) && (4.139)\\
\Sigma_{\hat{\theta}\hat{\rho}}(\mathbf{q},\tau) &= -\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau) && (4.140)\\
\Sigma_{\hat{\theta}\hat{g}}(\mathbf{q},\tau) &= -\Sigma_{\hat{g}\hat{\theta}}(\mathbf{q},\tau) && (4.141)\\
\Sigma_{\hat{v}\hat{\rho}}(\mathbf{q},\tau) &= -\Sigma_{\hat{\rho}\hat{v}}(\mathbf{q},\tau) && (4.142)\\
\Sigma_{\hat{v}\hat{g}}(\mathbf{q},\tau) &= -\Sigma_{\hat{g}\hat{v}}(\mathbf{q},\tau) && (4.143)\\
\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},\tau) &= \Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},\tau), && (4.144)
\end{aligned}$$

and similar ones for correlators:

$$\begin{aligned}
C_{g\rho}(\mathbf{q},\tau) &= C_{\rho g}(\mathbf{q},\tau) && (4.145)\\
C_{\theta\rho}(\mathbf{q},\tau) &= C_{\rho\theta}(\mathbf{q},\tau) && (4.146)\\
C_{\theta g}(\mathbf{q},\tau) &= C_{g\theta}(\mathbf{q},\tau) && (4.147)\\
C_{v\rho}(\mathbf{q},\tau) &= C_{\rho v}(\mathbf{q},\tau) && (4.148)\\
C_{vg}(\mathbf{q},\tau) &= C_{gv}(\mathbf{q},\tau) && (4.149)\\
C_{v\theta}(\mathbf{q},\tau) &= C_{\theta v}(\mathbf{q},\tau). && (4.150)
\end{aligned}$$

All these identities reduce the number of independent correlators to ten, which are $C_{\rho\rho}$, $C_{\rho g}$, $C_{\rho\theta}$, $C_{\rho v}$, C_{gg},$C_{g\theta}$, C_{gv}, $C_{\theta\theta}$, $C_{\theta v}$ and C_{vv}.

In the case of FNH, there are in principle 64 Schwinger-Dyson equation. We write 16 of these equations, the other being trivially linear dependent on these:

$$\begin{aligned}
&\partial_\tau C_{\rho\rho}(\mathbf{q},\tau) - i\rho_0\mathbf{q}C_{v\rho}(\mathbf{q},\tau) = \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u)\\
+\;& \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\rho}(\mathbf{q},u) - \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u)C_{\theta\rho}(\mathbf{q},u)\\
-\;& \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{g}}(\mathbf{q},\tau-u)C_{v\rho}(\mathbf{q},u) \qquad (4.151)
\end{aligned}$$

$$\begin{aligned}
&\partial_\tau C_{\rho g}(\mathbf{q},\tau) - i\rho_0\mathbf{q}C_{vg}(\mathbf{q},\tau) = \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho g}(\mathbf{q},u)\\
+\;& \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{gg}(\mathbf{q},u) - \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u)C_{\theta g}(\mathbf{q},u)\\
-\;& \frac{1}{T}\int_0^\tau du\Sigma_{\hat{\rho}\hat{g}}(\mathbf{q},\tau-u)C_{vg}(\mathbf{q},u) \qquad (4.152)
\end{aligned}$$

$$\begin{aligned}
&\partial_\tau C_{\rho\theta}(\mathbf{q},\tau) - i\rho_0\mathbf{q}C_{v\theta}(\mathbf{q},\tau) = \Sigma_{\hat{\rho}\hat{v}}(\mathbf{q},\tau)\\
+\;& \frac{1}{T}\int_0^\tau du\Big[\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\theta}(\mathbf{q},u) + \Sigma_{\rho\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\theta}(\mathbf{q},u)\\
-\;& \Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u)C_{\theta\theta}(\mathbf{q},u) - \Sigma_{\hat{\rho}\hat{g}}(\mathbf{q},\tau-u)C_{v\theta}(\mathbf{q},u)\Big] \qquad (4.153)
\end{aligned}$$

$$
\begin{aligned}
&\partial_\tau C_{\rho v}(\mathbf{q},\tau) - i\rho_0 \mathbf{q} C_{vv}(\mathbf{q},\tau) = \Sigma_{\hat{\rho}\hat{v}}(\mathbf{q},\tau) \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho v}(\mathbf{q},u) + \Sigma_{\hat{\rho}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{gv}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta v}(\mathbf{q},u) - \Sigma_{\hat{\rho}\hat{g}}(\mathbf{q},\tau-u) C_{vv}(\mathbf{q},u) \Big]
\end{aligned} \tag{4.154}
$$

$$
\begin{aligned}
&\partial_\tau C_{g\rho}(\mathbf{q},\tau) - i\rho_0 \mathbf{q} C_{\theta\rho}(\mathbf{q},\tau) + L C_{v\rho}(\mathbf{q},\tau) = \frac{1}{T}\int_0^\tau du \Sigma_{\hat{g}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u) \\
+ \quad & \frac{1}{T}\int_0^\tau du \Sigma_{\hat{g}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\rho}(\mathbf{q},u) - \frac{1}{T}\int_0^\tau du \Sigma_{\hat{g}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta\rho}(\mathbf{q},u) \\
- \quad & \frac{1}{T}\int_0^\tau du \Sigma_{\hat{g}\hat{g}}(\mathbf{q},\tau-u) C_{v\rho}(\mathbf{q},u)
\end{aligned} \tag{4.155}
$$

$$
\begin{aligned}
&\partial_\tau C_{gg}(\mathbf{q},\tau) - i\rho_0 \mathbf{q} C_{\theta g}(\mathbf{q},\tau) + L C_{vg}(\mathbf{q},\tau) = +\frac{1}{T}\int_0^\tau du \Sigma_{\hat{g}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho g}(\mathbf{q},u) \\
+ \quad & \frac{1}{T}\int_0^\tau du \Sigma_{\hat{g}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{gg}(\mathbf{q},u) - \frac{1}{T}\int_0^\tau du \Sigma_{\hat{g}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta g}(\mathbf{q},u) \\
- \quad & \frac{1}{T}\int_0^\tau du \Sigma_{\hat{g}\hat{g}}(\mathbf{q},\tau-u) C_{vg}(\mathbf{q},u)
\end{aligned} \tag{4.156}
$$

$$
\begin{aligned}
&\partial_\tau C_{g\theta}(\mathbf{q},\tau) - i\rho_0 \mathbf{q} C_{\theta\theta}(\mathbf{q},\tau) + L C_{v\theta}(\mathbf{q},\tau) = \Sigma_{\hat{g}\hat{\theta}}(\mathbf{q},\tau) \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{g}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\theta}(\mathbf{q},u) - \Sigma_{\hat{g}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\theta}(\mathbf{q},u) \\
- \quad & \Big[\Sigma_{\hat{g}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta\theta}(\mathbf{q},u) + \Sigma_{\hat{g}\hat{g}}(\mathbf{q},\tau-u) C_{v\theta}(\mathbf{q},u) \Big]
\end{aligned} \tag{4.157}
$$

$$
\begin{aligned}
&\partial_\tau C_{gv}(\mathbf{q},\tau) - i\rho_0 \mathbf{q} C_{\theta v}(\mathbf{q},\tau) + L C_{vv}(\mathbf{q},\tau) = \Sigma_{\hat{g}\hat{v}}(\mathbf{q},\tau) \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{g}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho v}(\mathbf{q},u) + \Sigma_{\hat{g}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{gv}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{g}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta v}(\mathbf{q},u) - \Sigma_{\hat{g}\hat{g}}(\mathbf{q},\tau-u) C_{vv}(\mathbf{q},u) \Big]
\end{aligned} \tag{4.158}
$$

$$
\begin{aligned}
&C_{\theta\rho}(\mathbf{q},\tau) - W(\mathbf{q}) C_{\rho\rho}(\mathbf{q},\tau) = -\frac{1}{T}\left[\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0) C_{\rho\rho}(\mathbf{q},\tau) + \Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},0) C_{g\rho}(\mathbf{q},\tau) \right] \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u) + \Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\rho}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{\theta}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta\rho}(\mathbf{q},u) - \Sigma_{\hat{\theta}\hat{g}}(\mathbf{q},\tau-u) C_{v\rho}(\mathbf{q},u) \Big]
\end{aligned} \tag{4.159}
$$

$$
\begin{aligned}
& C_{\theta g}(\mathbf{q},\tau) - W(\mathbf{q}) C_{\rho g}(\mathbf{q},\tau) = -\frac{1}{T}\left[\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0) C_{\rho g}(\mathbf{q},\tau) + \Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},0) C_{gg}(\mathbf{q},\tau)\right] \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho g}(\mathbf{q},u) + \Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{gg}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{\theta}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta g}(\mathbf{q},u) - \Sigma_{\hat{\theta}\hat{g}}(\mathbf{q},\tau-u) C_{vg}(\mathbf{q},u)\Big]
\end{aligned} \tag{4.160}
$$

$$
\begin{aligned}
& C_{\theta\theta}(\mathbf{q},\tau) - W(\mathbf{q}) C_{\rho\theta}(\mathbf{q},\tau) = \frac{1}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau) \\
- \quad & \frac{1}{T}\left[\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0) C_{\rho\theta}(\mathbf{q},\tau) + \Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},0) C_{g\theta}(\mathbf{q},\tau)\right] \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\theta}(\mathbf{q},u) + \Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\theta}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{\theta}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta\theta}(\mathbf{q},u) - \Sigma_{\hat{\theta}\hat{g}}(\mathbf{q},\tau-u) C_{v\theta}(\mathbf{q},u)\Big]
\end{aligned} \tag{4.161}
$$

$$
\begin{aligned}
& C_{\theta v}(\mathbf{q},\tau) - W(\mathbf{q}) C_{\rho v}(\mathbf{q},\tau) = \frac{1}{T}\Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},\tau) \\
- \quad & \frac{1}{T}\left[\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0) C_{\rho v}(\mathbf{q},\tau) + \Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},0) C_{gv}(\mathbf{q},\tau)\right] \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho v}(\mathbf{q},u) + \Sigma_{\hat{\theta}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{gv}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{\theta}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta v}(\mathbf{q},u) - \Sigma_{\hat{\theta}\hat{g}}(\mathbf{q},\tau-u) C_{vv}(\mathbf{q},u)\Big]
\end{aligned} \tag{4.162}
$$

$$
\begin{aligned}
& C_{v\rho}(\mathbf{q},\tau) - \frac{1}{\rho_0} C_{g\rho}(\mathbf{q},\tau) = -\frac{1}{T}\left[\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},0) C_{\rho\rho}(\mathbf{q},\tau) + \Sigma_{\hat{v}\hat{v}}(\mathbf{q},0) C_{g\rho}(\mathbf{q},\tau)\right] \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u) + \Sigma_{\hat{v}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\rho}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{v}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta\rho}(\mathbf{q},u) - \Sigma_{\hat{v}\hat{g}}(\mathbf{q},\tau-u) C_{v\rho}(\mathbf{q},u)\Big]
\end{aligned} \tag{4.163}
$$

$$
\begin{aligned}
& C_{vg}(\mathbf{q},\tau) - \frac{1}{\rho_0} C_{gg}(\mathbf{q},\tau) = -\frac{1}{T}[\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},0) C_{\rho g}(\mathbf{q},\tau) + \Sigma_{\hat{v}\hat{v}}(\mathbf{q},0) C_{gg}(\mathbf{q},\tau)] \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho g}(\mathbf{q},u) + \Sigma_{\hat{v}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{gg}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{v}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta\rho}(\mathbf{q},u) - \Sigma_{\hat{v}\hat{g}}(\mathbf{q},\tau-u) C_{vg}(\mathbf{q},u)\Big]
\end{aligned} \tag{4.164}
$$

$$
\begin{aligned}
& C_{v\theta}(\mathbf{q},\tau) - \frac{1}{\rho_0} C_{g\theta}(\mathbf{q},\tau) = \frac{1}{T}\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},\tau) \\
- \quad & \frac{1}{T}\left[\Sigma_{\hat{v}\theta}(\mathbf{q},0) C_{\rho\hat{\theta}}(\mathbf{q},\tau) + \Sigma_{\hat{v}\hat{v}}(\mathbf{q},0) C_{g\theta}(\mathbf{q},\tau)\right] \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{v}\theta}(\mathbf{q},\tau-u)\partial_u C_{\rho\theta}(\mathbf{q},u) - \Sigma_{\hat{v}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{g\theta}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{v}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta\rho}(\mathbf{q},u) + \Sigma_{\hat{v}\hat{g}}(\mathbf{q},\tau-u) C_{v\theta}(\mathbf{q},u)\Big]
\end{aligned}
\tag{4.165}
$$

$$
\begin{aligned}
& C_{vv}(\mathbf{q},\tau) - \frac{1}{\rho_0} C_{gv}(\mathbf{q},\tau) = \frac{1}{T}\Sigma_{\hat{v}\hat{v}}(\mathbf{q},\tau) \\
- \quad & \frac{1}{T}\left[\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},0) C_{\rho v}(\mathbf{q},\tau) + \Sigma_{\hat{v}\hat{v}}(\mathbf{q},0) C_{gv}(\mathbf{q},\tau)\right] \\
+ \quad & \frac{1}{T}\int_0^\tau du \Big[\Sigma_{\hat{v}\hat{\theta}}(\mathbf{q},\tau-u)\partial_u C_{\rho v}(\mathbf{q},u) - \Sigma_{\hat{v}\hat{v}}(\mathbf{q},\tau-u)\partial_u C_{gv}(\mathbf{q},u) \\
- \quad & \Sigma_{\hat{v}\hat{\rho}}(\mathbf{q},\tau-u) C_{\theta\rho}(\mathbf{q},u) + \Sigma_{\hat{v}\hat{g}}(\mathbf{q},\tau-u) C_{vv}(\mathbf{q},u)\Big]
\end{aligned}
\tag{4.166}
$$

As for BD, the number of independent correlators is smaller than the number of equations, and there are here 6 redundant equations. The extension of the proof of appendix B to the present equations is straightforward but very painful.

These are the exact non-perturbative dynamical equations preserving FDT. One can then use different approximation schemes for self-energies to close the equations. It is worth noting that whatever the approximation, the FDT is always verified due to the way the equations were derived.

Chapter 5

Interacting particles and stochastic equations

Derivation of MCT within a field theory is a non-trivial problem. Previous attempts suffered from violation of time-reversal symmetry. Our approach presented in the previous Chapter resolved this problem. However we found equations that looked similar to MCT but were not exactly the equations that we were looking for: they suffered from a divergence and had to be regularised by a large wave-vector cut off. In this chapter we address this problem on the example of Brownian dynamics.

The divergence motivates to look for a different perturbation theory. As we have discussed the perturbation theory considered in Sec. 4.4.2 is a large density expansion which is not well defined. In static liquid theory a well-defined perturbation series are provided by a low density expansion [33]. However its derivation relies on a purely static methods and does not extend to dynamics easily. Let's note that BD is a theory with a fixed number of particles and conserved density. Derivation of a low density expansion requires a theory with varying number of particles and, therefore, density. Therefore we should couple the system to a particle reservoir or, equivalently, include the processes of particles creation-annihilation in the description. *A priori* it is not clear how to do that in the context of BD. However this is easily done in the context of reaction-diffusion systems [95]. This is a widely studied class of systems that consider dynamics of interacting diffusing particles. Their interaction is usually described by a chemical reaction like rules, for example $A + A \to \emptyset$. Then creation-annihilation of particles within BD can be approached by the following rules: $A \to \emptyset$ and $A \to A + A$. Reaction-diffusion systems are analysed within a context of Doï-Peliti field theory [96, 97, 98] derived from "second quantised" ladder operator representation of master equations via coherent state representation. However this requires to work with field different from ρ that do not have an immediate physical meaning; density correlations and response function which are of primary interest to us have complicated multi-point representation

in these fields. This sets a problem of construction of a description in terms of density for such systems that we consider in this chapter.

As we will see the solution to this problem is related to a different perturbation theory that has the required properties: it respects TRS and has a better vertex which ensures the convergence for large wave-vectors. This theory is based on the remark that the potential vertex in the straightforward perturbation theory of Sec. 4.3.2 decreases to zero large wave-vectors and makes integrals converge. This suggests to look for a perturbation in powers of the potential U *i.e.* for a different decomposition of S (4.5). Recalling the conclusions of the previous chapter we also imply that every part of the decomposition is separately invariant under TRS. Indeed, we have seen that a decomposition $S = S_2 + S_{INT}$ (see Sec. 4.3.2) violates time-reversal symmetry but we have not analysed if a different decomposition is possible such that its parts are separately invariant even under non-linear transformation. Surprisingly such a decomposition exists and it is exactly the expansion in powers of the potential U as we will see. However the resulting theory is inconvenient for direct application because both parts of the decomposition are not quadratic and the conventional perturbation techniques cannot be used. Solution of this technical problem brings us back to reaction-diffusion systems as we will show.

The overall aim of this chapter is to demonstrate how this two approaches can be unified within the context of reaction-diffusion systems and provide technical details of this demonstration. We start from the introduction of the expansion in the powers of the potential within BD and discuss its symmetry related properties; then we show how the resulting perturbation theory reduces to a reaction-diffusion system in Sec. 5.1. In the following section the Doï-Peliti derivation is presented in detail for BD with variable number of particles in Sec. 5.2. We then show how a reaction-diffusion system can be described by a stochastic equation on density. This allows us to use the standard method *à la* Martin-Siggia-Rose [76] instead of a more elaborated Doï-Peliti formalism.

5.1 Brownian dynamics and expansion in powers of the potential

In this section we present the different decomposition for BD action (4.5) which generates a perturbation expansion in powers of the potential U and preserves TRS. We also show how to carry out computations within this theory.

Let's recall the original field theory for interacting Brownian particles (4.5) derived from Dean equation in 4.1.1:

$$\begin{aligned} S[\rho,\hat{\rho}] &= \int d^3\mathbf{x} \int dt \left\{ \hat{\rho}(\mathbf{x},t) \left[-\partial_t \rho(\mathbf{x},t) + T\nabla^2 \rho(\mathbf{x},t) \right.\right. \\ &+ \left.\left. \nabla \cdot \left(\rho(\mathbf{x},t) \int d^3\mathbf{y}\ \nabla V(\mathbf{x}-\mathbf{y}) \rho(\mathbf{y},t) \right) \right] + T\rho(\mathbf{x},t) (\nabla \hat{\rho}(\mathbf{x},t))^2 \right\} \end{aligned} \tag{5.1}$$

As was pointed out above the decomposition of the action into quadratic part $s_{0,x}$ and interaction $s_{INT,x}$ is not invariant under non-linear transformations corresponding to time-reversal symmetry (TRS). However a perturbation expansion in powers of the potential U preserves the symmetry. Indeed the dynamical average of any function $\mathcal{A}[\rho,\hat{\rho}]$ may be written as

$$\begin{aligned}
\langle \mathcal{A}[\rho,\hat{\rho}]\rangle &= \int \mathcal{D}\rho \int \mathcal{D}\hat{\rho}\ \mathcal{A}[\rho,\hat{\rho}]\ e^{S_{FREE}[\rho,\hat{\rho}]+S_U[\rho,\hat{\rho}]} \\
&= \sum_{p=1}^{\infty} \int \mathcal{D}\rho \int \mathcal{D}\hat{\rho}\ \mathcal{A}[\rho,\hat{\rho}] \frac{(S_U[\rho,\hat{\rho}])^p}{p!} e^{S_{FREE}[\rho,\hat{\rho}]} \\
S_{FREE} &= \int d^3\mathbf{x}\ dt\hat{\rho}(\mathbf{x},t)\left[-\partial_t\rho(\mathbf{x},t)+T\nabla^2\rho(\mathbf{x},t)\right]+T\rho(\mathbf{x},t)(\nabla\hat{\rho}(\mathbf{x},t))^2 \\
S_U &= \int d^3\mathbf{x}\int d^3\mathbf{y}\ \hat{\rho}(\mathbf{x},t)\nabla\left(\rho(\mathbf{x},t)\left(\nabla U(\mathbf{x}-\mathbf{y})\right)\rho(\mathbf{y},t)\right) \qquad (5.2)
\end{aligned}$$

where S_{FREE} regroups the terms of the action which do not contain the potential. The key point is that both S_{FREE} and S_U are separately invariant under transformation $\mathcal{T}$ given by Eq. (4.11):

$$\mathcal{T}:\left\{\begin{array}{ll} t & \to -t \\ \hat{\rho}_x & \to \hat{\rho}_x + f_x, \end{array}\right. \qquad (5.3)$$

where f verifies:

$$\nabla\cdot(\rho_x\nabla f_x) = -\frac{1}{T}\partial_t\rho_x \qquad (5.4)$$

Hence the symmetry is verified to any finite order in the expansion in powers of S_U. Note that transformation $\mathcal{U}$ given by Eq. (4.13) still mixes S_{FREE} with S_U. Therefore the perturbation theory is not invariant under this transformation. This reveals a difference between these two transformations whose physical origin is unclear.

There are however several problems within such perturbation theory. First, the transformation $\mathcal{T}$ given by Eq. (4.11) is non-linear. This leads to problems with the Ward-Tahakashi (WD) identities. Indeed, the naive response $\mathcal{G}(\mathbf{q},t-s)=\langle\rho(-\mathbf{q},t)\hat{\rho}(\mathbf{q},s)\rangle$ transforms under $\mathcal{T}$ as:

$$\mathcal{G}(\mathbf{q},s-t) = \mathcal{G}(\mathbf{q},t-s) + \langle\rho(-\mathbf{q},t)f(\mathbf{q},s)\rangle$$

where the function f is fixed by Eq.(5.4). The latter is only defined within an average *i.e*:

$$T\langle\nabla(\rho(\mathbf{x},t)\nabla f(\mathbf{x},t))\cdots\rangle = -\partial_t\langle\rho(\mathbf{x},t)\cdots\rangle$$

and one should introduce at least extra fields $f,\hat{f}$ and $\phi,\hat{\phi}$ (see Sec. 4.4.1) to compute $\mathcal{G}$. However the naive response is not the function we are very interested in. Second, the response function R is still a three-point quantity:

$$R_{xy}(t,s) = -\langle\rho(\mathbf{x},t)\nabla\cdot(\rho(\mathbf{y},s)\nabla\hat{\rho}(\mathbf{y},s))\rangle.$$

Finally S_{FREE} *is not quadratic in the fields*. The nonlinearity of the fields transformation relate the noise cubic term to quadratic terms. Thus this vertex must be taken into account non-perturbatively. In other words, to compute the correlators at a given order in the power series expansion in S_U, one has to include contributions at all orders in the noise vertex. This is a difficult but not impossible task because, at a given order p in S_U, the diagrammatic expansion contains a finite number of diagrams, due to the absence of the propagator connecting two $\hat{\rho}$'s. Indeed, if one considers a diagram with p potential vertices and q noise vertices contributing to a correlation function of r ρ's and s $\hat{\rho}$'s, one must have $q + s \leq p + r$. Hence such a diagram must have less than $p + r - s$ noise vertices. Thus a bare perturbation theory which preserves the nonlinear symmetry can be set up but is considerably more complicated than the usual one.

Let's note however that the action S_{FREE} (5.2) describes the case of non-interacting Brownian particles particles. This follows from Eqs. (4.1) with $U = 0$ which are equivalent to the field theory. Therefore there should be a possibility to compute the averages with S_{FREE} exactly. This is implemented via a non-linear transformation of fields ρ and $\hat{\rho}$:

$$\begin{aligned} \rho(\mathbf{q}, t) &= \hat{\phi}(\mathbf{q}, t)\phi(\mathbf{q}, t-) \\ \hat{\rho}(\mathbf{q}, t) &= \log\left[\hat{\phi}(\mathbf{q}, t)\right] \end{aligned} \tag{5.5}$$

The action (5.1) reads

$$\begin{aligned} S[\hat{\phi}, \phi] &= S_2 + S_{INT} \\ S_2 &= \int d^3\mathbf{x} \int dt \hat{\phi}(\mathbf{x}, t) \left[-\partial_t \phi(\mathbf{x}, t) + T\nabla_x^2 \phi(\mathbf{x}, t)\right] \\ S_{INT} &= -\int d^3\mathbf{x}\, d^3\mathbf{y}\, dt (\nabla_x \hat{\phi}(\mathbf{x}, t))\phi(\mathbf{x}, t)(\nabla_y U(\mathbf{x} - \mathbf{y}))\hat{\phi}(\mathbf{y}, s)\phi(\mathbf{y}, s) \end{aligned} \tag{5.6}$$

Note that the Jacobian of the transformation has a modulus one and that S_{FREE} and S_U do not mix under this transformation also. This ensures that the theory (5.6) also respects time-reversal symmetry in expansion in powers of U. Now S_{FREE} became S_2 which is Gaussian and expansions in powers of the potential becomes a standard perturbation theory. But physical quantities of primary interest, the response function and the density-density correlator, became *multi-point quantities*:

$$C_{\rho\rho}(\mathbf{x} - \mathbf{y}, t - s) = \langle \hat{\phi}\phi(\mathbf{x}, t)\hat{\phi}\phi(\mathbf{y}, s)\rangle$$

The response function R given by 4.16 reads:

$$\begin{aligned} R(\mathbf{x} - \mathbf{y}, t - s) &= -\left\langle \hat{\phi}\phi(\mathbf{x}, t)\nabla\left(\hat{\phi}\phi(\mathbf{y}, s)\nabla \log \hat{\phi}(\mathbf{y}, s)\right)\right\rangle \\ &= -\left\langle \hat{\phi}\phi(\mathbf{x}, t)\nabla\left(\phi(\mathbf{y}, s)\nabla\hat{\phi}(\mathbf{y}, s)\right)\right\rangle \end{aligned}$$

And both the density-density correlator and the response function can be expressed through the same *four-point* function:

$$\gamma(\mathbf{x},\mathbf{y},\mathbf{z},\mathbf{r};t,s,u,v) = \langle\hat{\phi}(\mathbf{x},t)\hat{\phi}(\mathbf{y},s)\phi(\mathbf{z},u)\phi(\mathbf{r},v)\rangle$$

The TRS transformation $\mathcal{T}$ remains non-linear:

$$\begin{aligned} \hat{\phi} &\to \hat{\phi}\, e^{f} \\ \phi &\to \phi\, e^{-f} \\ T\nabla(\hat{\phi}\phi\nabla f) &= -\partial_t(\hat{\phi}\phi) \end{aligned}$$

The transformation given by Eq. (5.5) is not accidental and has a simple interpretation related to reaction-diffusion systems as we will see below.

5.2 Grand canonical dynamics

The main aim of this section is to derive a low density expansion. This is done in the context of the Doï-Peliti (DP) field theory which allow to take into account variable number of particles in BD and will lead to such expansion for Brownian dynamics. Another consequence is a better understanding of the origin of the transformation (5.5).

5.2.1 Master equation

Let's consider interacting diffusing particles on some regular lattice in a d dimensional space. The hopping rate W_{ij} which describes the diffusive motion and defines the probability for a particle to jump from site $\imath$ to site $\jmath$ is fixed by an on site potential H_j where $\jmath$ labels the sites:

$$W_{ij} = W_0 \exp\left[\frac{H_i - H_j}{2T}\right] \qquad H_i = \sum_l U(x_i - x_l) n_l.$$

We also suppose that detailed balance is satisfied. The interaction between particles is defined by the description of reactions between particles on the same site: $A + A \to \emptyset$, $A \to 2A$, *etc.* These formulae are supplied by the probability rates at which these reactions happen. Let's note that BD fits well into this class of systems: particles only diffuse and there are no on site reactions.

We consider a particular system with particles interacting following two rules: $A \to \emptyset$ and $A \to A + A$ with rates λ and μ respectively. That is particles can either decay, or branch into two. The evolution of this system is described by a master equation for the probability $\mathcal{P}(n,t)$ of realisation of a particular repartition of particles $\{n_i\}$ at time t:

$$
\begin{aligned}
\partial_t \mathcal{P}(n,t) &= \sum_i \sum_{j\in v(i)} \Big[(n_j+1) W_{ji} \mathcal{P}(n : n_i - 1, n_j + 1, t) \\
&+ (n_i+1) W_{ij} \mathcal{P}(n : n_i + 1, n_j - 1, t) - (n_i + n_j) W_{ij} \mathcal{P}(n : n_i, n_j, t) \Big] \\
&+ \lambda \sum_i \left[(n_i+1)\mathcal{P}(n : n_i + 1, t) - n_i \mathcal{P}(n : n_i, t) \right] \\
&+ \mu \sum_i \left[(n_i-1)\mathcal{P}(n : n_i - 1, t) - n_i \mathcal{P}(n : n_i, t) \right] \qquad (5.7)
\end{aligned}
$$

where the hopping is limited to nearest neighbours only; $v(i)$ is a set of all the neighbours of a site i. This equation is supplemented by an initial distribution $\mathcal{P}(n,0)$. For a uniform initial distribution with an average number n_0 of particles per site it equals:

$$
\mathcal{P}(n,0) = \prod_i \left(\frac{n_0^{n_i}}{n_i!} \exp(-n_0) \right).
$$

The master equation in this form is complicated and is hard to analyse because one has to work in the space of all configurations.

5.2.2 Second quantisation representation

The master equation (5.7) admits a second quantisation representation in terms of a creation-annihilation operators [96, 98]. This is possible because site occupation numbers change by an integer number only.

Let's introduce bosonic operators $a_i, a_i^\dagger$ for every site i. Their definition is pretty straightforward: $a, a^\dagger$ are defined by their action on a state

$$
\begin{aligned}
a_i |n_i\rangle &= n_i |n_i - 1\rangle \\
a_i^\dagger |n_i\rangle &= |n_i + 1\rangle
\end{aligned}
$$

where $|n_i\rangle$ denotes a configuration with n_i particles on site i following the quantum mechanics tradition. Note a different normalisation with respect to the standard definition. The operators satisfy "bosonic" commutation relations

$$
[a_i, a_j^\dagger] = \delta_{ij}, \qquad [a_i, a_j] = [a_i^\dagger, a_j^\dagger] = 0.
$$

The vacuum is defined by the requirement $a_i|0\rangle = 0 \quad \forall i$ (empty lattice). Excited ket states *i.e.* lattice with-non zero occupation numbers, is generated by a successive application of creation operators $a^\dagger$ to a vacuum:

$$
|n_i\rangle = \left(a_i^\dagger\right)^{n_i} |0\rangle, \qquad \text{and bra states} \qquad \langle n_i| = \langle 0| \frac{a^{n_i}}{n_i!}.
$$

Then a given repartition $n = \{n_i\}$ reads as $|n\rangle = \prod_i \left(a_i^\dagger\right)^{n_i} |0\rangle$. By definition excited states are orthogonal: $\langle n||m\rangle = \delta_{nm}$ and form a complete basis:

$$1 = \sum_n |n\rangle\langle n|.$$

The total probability $\mathcal{P}(n,t)$ is represented by a ket-vector

$$|\mathcal{P}(t)\rangle = \sum_n \mathcal{P}(n,t)|n\rangle$$

Within these notations the master equation (5.7) for $|\mathcal{P}(t)\rangle$ reads:

$$\begin{aligned} \partial_t|\mathcal{P}(t)\rangle &= \mathcal{H}|\mathcal{P}(t)\rangle & (5.8)\\ \mathcal{H} &= \sum_i \sum_{j\in v(i)} \left[W_{ji}a_i^\dagger a_j - W_{ij}a_i^\dagger a_i + W_{ij}a_j^\dagger a_i - W_{ij}a_j^\dagger a_j\right] & (5.9)\\ &\quad +\lambda\sum_i (1-a_i^\dagger)a_i + \mu\sum_i (a_i^\dagger - 1)a_i^\dagger a_i \end{aligned}$$

This equation admits a formal solution $|\mathcal{P}(t)\rangle = \exp(\mathcal{H}t)|\mathcal{P}(0)\rangle$. The average of a quantity $A(n)$ reads

$$\langle A(n)\rangle = \sum_n A(n)\mathcal{P}(n,t) = \langle O|\hat{A}|\mathcal{P}\rangle$$

where $\langle O|$ is the projection state $\langle O| = \sum_n \langle n| = \langle 0|e^a$; operator $\hat{A}$ is defined by its action on a state $\hat{A}|n\rangle = A(n)|n\rangle$. Note that all operators are diagonal in the basis of $|n\rangle$ within this formalism. Also all averages are linear in $|\mathcal{P}\rangle$ unlike in quantum mechanics where they are quadratic in wavefunction.

5.2.3 Coherent states representation

It is possible to continue the analysis in the context of the second quantisation formalism but as illustrated by quantum field theory computation in terms of operators is more involved technically and is less convenient than a field theory framework. Derivation of a field theory from a second quantisation representation is a standard procedure presented in textbooks [99, 100]. A particular presentation of derivation via a coherent state formalism that we use below is presented in [96].

Coherent states are defined as eigenvectors of the operator a: $a|\phi\rangle = \phi|\phi\rangle$ labelled by a complex number ϕ. An eigenstate for $a^\dagger$ is $\langle\phi|a^\dagger = \overline{\phi}\langle\phi|$. That is the eigenvalue of $a^\dagger$ is a complex conjugate of the eigenvalue of a. After some algebra one derives their expression in the basis of $|n\rangle$:

$$|\phi\rangle = \exp\left(-\frac{|\phi|^2}{2}\right)\sum_{n\geq 0}\frac{\phi^n}{n!}|n\rangle = \exp\left(-\frac{|\phi|^2}{2}+\phi a^\dagger\right)|0\rangle$$

$$\langle\phi| = \exp\left(-\frac{|\phi|^2}{2}\right)\sum_{n\geq 0}\frac{\overline{\phi}^n}{n!}\langle n| = \langle 0|\exp\left(-\frac{|\phi|^2}{2}+\hat{\phi}a\right)$$

For obvious reasons $\langle\phi|$ is an eigenvector of $a^\dagger$with an eigenvalue $\hat{\phi} = \overline{\phi}$ (a complex conjugate of ϕ). A generic coherent state is given by a product of coherent states for every site. Coherent states form a complete basis like $|n\rangle$:

$$1 = \sum_n |n\rangle\langle n| = \int \frac{d^2\phi}{2\pi}|\phi\rangle\langle\phi|$$

where $d\phi = d\Re\phi\, d\Im\phi$. However coherent states are not orthogonal:

$$\langle\psi||\phi\rangle = \exp\left(-\frac{|\psi|^2}{2}-\frac{|\phi|^2}{2}+\hat{\psi}\phi\right)$$

and this basis is overcomplete. The projection state $\langle O|$ is proportional to the coherent state with all $\phi = 1$ that is the coherent state $\langle 1|$.

Computation becomes especially simple in the basis of coherent states with the introduction of normal ordering. For a given operator $\hat{A}(a^\dagger, a)$ its normal ordered counterpart $\tilde{A}(a^\dagger, a)$ is computed by commuting all the $a^\dagger$ in $\hat{A}(a^\dagger, a)$ to the left and all a to the right. Then the bracket $\langle\psi|\hat{A}|\phi\rangle$ is computed immediately: $\langle\psi|\hat{A}|\phi\rangle = \tilde{A}(\psi,\phi)\langle\psi||\phi\rangle$.

5.2.4 Field theory *à la* Doï-Peliti

The derivation is fairly standard and is only sketched below. The average of an operator $\hat{A}$ reads:

$$\langle\hat{A}(t)\rangle = \langle O|\hat{A}|\mathcal{P}(t)\rangle = \langle O|\hat{A}e^{\mathcal{H}t}|\mathcal{P}(0)\rangle \tag{5.10}$$

Let's now slice the time interval $[0,t]$ into N parts so that $t = N\epsilon$. Then in the limit $N\to\infty$ factorisation takes place:

$$\exp[\mathcal{H}t] \approx \prod \exp[\mathcal{H}\epsilon].$$

We use this identity in (5.10) and plug in representations of unity:

$$\prod_i\prod_{s=0}^{t}\frac{d^2\phi_i(s)}{\pi}\langle 1|\hat{A}|\phi_i(t)\rangle\prod_s\langle\phi_i(s)|\exp[\mathcal{H}\epsilon]|\phi_i(s-\epsilon)\rangle\langle\phi_i(0)||\mathcal{P}(0)\rangle \tag{5.11}$$

The product $\langle 1|\hat{A}|\phi_i(t)\rangle$ is computed by normal ordering $\hat{A}$. Then

$$\langle 1|\hat{A}|\phi_i(t)\rangle = \tilde{A}(1,\phi_i(t))\langle 1||\phi_i(t)\rangle \sim \tilde{A}(1,\phi_i(t))\exp(-|\phi_i(t)|^2/2+\hat{\phi}_i(t))$$

and $\tilde{A}(1,\phi_(t)) = \tilde{A}(a^\dagger \to 1, a \to \phi(t))$. The initial distribution term $\langle \phi_i(0)||\mathcal{P}(0)\rangle$ can be computed exactly if $|\mathcal{P}(0)\rangle$ is a coherent state.

To evaluate the rest we use the identity

$$\langle \phi(s)| \exp(\mathcal{H}\epsilon)|\phi(s-\epsilon)\rangle = \langle \phi(s)||\phi(s-\epsilon)\rangle \exp\left(\epsilon\tilde{\mathcal{H}}(a^\dagger \to \hat{\phi}, a \to \phi) + O(\epsilon^2)\right)$$

where $\tilde{\mathcal{H}}$ is the normal ordering of $\mathcal{H}$. This yields:

$$\langle A(t)\rangle \sim \prod_i \prod_{s=0}^{t} \frac{d^2\phi_i(s)}{\pi} \tilde{A}(1,\phi_i(t)) \exp(-|\phi_i(t)|^2/2 + \hat{\phi}_i(t)) \langle \phi_i(0)||\mathcal{P}(0)\rangle$$

$$\times \prod_s \exp\left[-\hat{\phi}_i(s)(\phi_i(s) - \phi_i(s-\epsilon)) + \frac{|\phi_i(s)|^2}{2} - \frac{|\phi_i(s-\epsilon)|^2}{2} + \epsilon\tilde{\mathcal{H}}(\hat{\phi}_i(s), \phi_i(s-\epsilon))\right]$$

Quadratic terms in the exponential drop out when the product over s is taken except at $s = 0$ and $s = t$ resulting into:

$$\langle A(t)\rangle \sim \prod_i \prod_{s=0}^{t} \frac{d^2\phi_i(s)}{\pi} \tilde{A}(1,\phi_i(t)) e^{\hat{\phi}_i(t)} \langle \phi_i(0)||\mathcal{P}(0)\rangle$$

$$\times \exp\left[-\sum_s \hat{\phi}_i(s)(\phi_i(s) - \phi_i(s-\epsilon)) + \epsilon\sum_s \tilde{\mathcal{H}}(\hat{\phi}_i(s), \phi_i(s-\epsilon)) - \frac{|\phi_i(0)|^2}{2}\right]$$

Now the limit $\epsilon \to 0$ should be taken to yield a final theory. This limit can hide a problem of a choice between Itô or Stratonovich calculus related to time ordering of the fields if one works with a continuous theory. However in our case normal ordering implies the Itô calculus:

$$\langle A(t)\rangle = \mathcal{N}^{-1} \int \prod_i D\hat{\phi}_i D\phi_i \langle \phi_i(0)||\mathcal{P}(0)\rangle e^S$$

$$S = \sum_i \left\{ \phi_i(t) + \sum_i \left[-\int_0^t du \hat{\phi}_i \partial_u \phi_i + \tilde{\mathcal{H}}(\hat{\phi}_i, \phi_i)\right] - \frac{|\phi_i(0)|^2}{2}\right\} \tag{5.12}$$

Now ϕ and $\hat{\phi}$ are treated as independent variables. Taking the continuum limit we get:

$$S = \int d^3\mathbf{x}dt \left[\phi(\mathbf{x},t) - \hat{\phi}(\mathbf{x},t)\partial_t\phi(\mathbf{x},t) + \tilde{\mathcal{H}}(\hat{\phi},\phi) - \frac{|\phi(x,0)|^2}{2}\right] \tag{5.13}$$

Now let's write the explicit expression for $\tilde{\mathcal{H}}$ derived from (5.9):

$$\tilde{\mathcal{H}}(\hat{\phi},\phi) = \hat{\phi}(\mathbf{x},t)\nabla^2\phi(\mathbf{x},t) - \lambda(\hat{\phi}(\mathbf{x},t) - 1)\phi(\mathbf{x},t) + \mu(\hat{\phi}(\mathbf{x},t) - 1)\hat{\phi}\phi(\mathbf{x},t)$$
$$-T(\nabla\hat{\phi}(\mathbf{x},t))\phi(\mathbf{x},t)\int d^3\mathbf{y}(\nabla U(x-y))\hat{\phi}(y,t)\phi(y,t) \tag{5.14}$$

5.2.5 Field theory for the density

The goal of the above derivation was to account for creation and destruction of particles in Brownian dynamics that are represented by λ and μ related terms in the action (5.13). However we are interested in a field theory for the density. The field ϕ is not density: while the first moment $\langle\phi\rangle$ is equal to an average density, higher moments of ϕ do not coincide with moments of density, for example: $\langle\rho\rho\rangle = \langle\phi\phi\rangle + \langle\phi\rangle$.

For $\lambda = \mu = 0$ in (5.13) we recover the action $S[\hat{\phi}, \phi]$ (5.6) derived in Sec. 5.1 for interacting Brownian particles. This sheds some light on the origin of the transformation (5.5). Indeed now the fields ϕ and $\hat{\phi}$ from Sec. 5.1 can be identified with the fields of the Doï-Peliti field theory. And the transformation relates field theory in terms of $\phi, \hat{\phi}$ and field theory in terms of $\rho, \hat{\rho}$. This provides a method to get a density field theory for $\lambda, \mu \neq 0$: we should just use the inverted transformation. There are however two different ways to proceed:

- Apply the transformation directly to the action (5.13). This is a simple and efficient way to transform $S[\phi, \hat{\phi}]$ into $S[\rho, \hat{\rho}]$. However this transformation neglects completely the normal ordering of the fields.
- Another method is to make a canonical transformation of the operators $a, a^\dagger$: $a = \exp[-\rho^\dagger]\rho$, $a^\dagger = \exp[\rho^\dagger]$ such that $[\rho, \rho^\dagger] = 1$ and ρ is a density operator. This gives a different representation of the master equation. Repeating the steps of Sec. 5.2.3 and 5.2.4 gives a density field theory. The advantage of this method is that the normal ordering of the fields is conserved in the final action.

Finally the resulting action reads:

$$\begin{aligned} S[\rho, \hat{\rho}] &= \int d^3\mathbf{x} \int dt \left\{ \hat{\rho}(\mathbf{x}, t) \left[-\partial_t \rho(\mathbf{x}, t) + T\nabla^2 \rho(\mathbf{x}, t) \right.\right. \\ &+ \left.\left. \nabla \cdot \left(\rho(\mathbf{x}, t) \int d^3\mathbf{y}\ \nabla V(\mathbf{x} - \mathbf{y}) \rho(\mathbf{y}, t) \right) \right] + T\rho(\mathbf{x}, t) (\nabla \hat{\rho}(\mathbf{x}, t))^2 \right. \\ &+ \left. \mu \left(e^{\hat{\rho}(\mathbf{x}, t)} - 1 \right) \rho(\mathbf{x}, t) - \lambda (1 - e^{-\hat{\rho}(\mathbf{x}, t)}) \rho(\mathbf{x}, t) \right\} \end{aligned} \tag{5.15}$$

Before we proceed to analysis of this field theory we present a direct method to derive (5.15) from a stochastic equation.

5.2.6 Stochastic equations with Poisonnian noise

The Doï-Peliti formalism gives a field theory which is then transformed into a density field theory but the derivation is quite involved in the general case. After some practice one can skip most of the steps of Sec. 5.2.2, 5.2.3 and

5.2.4 and write down directly the result in terms of $\phi, \hat{\phi}$. One should rewrite the action in terms of density and a conjugated field yet. A direct derivation of a density field theory from stochastic equations *á la* Martin-Siggia-Rose is more convenient. However there is no recipe for how to construct a stochastic equation for ϕ in general case [101]. The structure of the action (5.13) together with (5.14) resembles an action that is usually derived in the context of Martin-Siggia-Rose formalism from a Langevin equation (on ϕ). We can assume that such equation exists and try to reconstruct it by identifying terms in the action. For simplicity we set $\lambda = \mu = 0$. Then the identification suggests the following stochastic equation:

$$\begin{aligned} \partial_t \phi(\mathbf{x}, t) &= T\nabla^2 \phi(\mathbf{x}, t) + \eta(\mathbf{x}, t) \\ \langle \eta(\mathbf{x}, t)\eta(\mathbf{y}, s)\rangle &= 2\nabla_x[\phi(\mathbf{x}, t)](\nabla_x U)(\mathbf{x} - \mathbf{y})\phi(\mathbf{y}, s)] \end{aligned} \tag{5.16}$$

where η is a multiplicative Gaussian noise with zero mean and an Itô calculus is adopted. However if we take a uniform ϕ and an attractive U, like $U(x) \sim -\exp(-x^2/2)$, the noise variance acquires a negative part which signalise a non-zero imaginary part of η. This makes the physical meaning of the noise η and of the whole equation (5.16) unclear. The situation becomes even worse for a system defined by reaction $A + A \to \emptyset$: the noise η is purely imaginary in that case.

On the contrary well-behaved stochastic equations exist if one starts from density ρ and allow a direct derivation of the field theory for density. Processes of creation and destruction of particles are well described by Poissonian jump processes [102]. Let's consider a single site with fluctuating occupation number n. Then the variation dn of n between t and $t + dt$ is given by $dJ = 0, \pm 1$:

$$dn = dJ \tag{5.17}$$

$$dJ = \begin{cases} 1 & n\mu dt \\ 0 & 1 - n(\lambda + \mu)dt \\ -1 & n\lambda dt \end{cases} \tag{5.18}$$

While dJ is certainly not a small quantity it has a vanishing probability to be non-zero as $dt \to 0$. This description is very close in spirit to the one provided by the master equation (5.7). Then the generating functional $Z[n, \hat{n}]$ *à la* MSR reads:

$$Z[n, \hat{n}] = \langle \prod_t \int Dn(t) \int D\hat{n}(t) \exp\left[\hat{n}(t)(dJ(t) - dn(t)\right]\rangle$$

where the angles denote the average over realisations of Poissonian jump processes. From the above definitions the average $\langle \exp[\hat{n}dJ]\rangle$ reads:

$$\begin{aligned} \langle \exp(\hat{n}dJ)\rangle &= \left(1 - n(\lambda + \mu)dt + e^{\hat{n}} n\mu dt + e^{-\hat{n}} n\lambda dt\right) \\ &= \left[1 + n\mu\left(e^{\hat{n}} - 1\right)dt + n\lambda\left(e^{-\hat{n}} - 1\right)dt\right] \\ &\approx \exp\left[n\lambda\left(e^{-\hat{n}} - 1\right)dt + n\mu\left(e^{\hat{n}} - 1\right)dt\right] \end{aligned}$$

Then Z is equal to:

$$\begin{aligned} Z[n,\hat{n}] &= \int D\hat{n}Dn \exp\left[\int dt S\right] \\ S[n,\hat{n}] &= -\hat{n}\partial_t n + n\lambda\left(e^{-\hat{n}}-1\right) + n\mu\left(e^{\hat{n}}-1\right) \end{aligned} \tag{5.19}$$

The above action has a λ- and μ- terms remarkably similar to those of (5.15).

Generalisation of the above construction to the model presented in Sec. 5.2.1 is straightforward by adding hopping rates W_{ij}. This gives a set of equations on variations of occupation numbers:

$$dn_i = dJ_i \tag{5.20}$$

$$dJ_i = \begin{cases} 1 & \left(n_i\mu + \sum\limits_{j\in v(i)} n_j W_{ji}\right) dt \\ 0 & 1 - \left[n_i(\lambda+\mu) - \sum\limits_{j\in v(j)} (n_i W_{ij} - n_j W_{ji})\right] dt \\ -1 & n_i\left(\lambda + \sum\limits_{j\in v(i)} W_{ij}\right) dt \end{cases} \tag{5.21}$$

Repeating the steps made for a single site yields the action:

$$S[n,\hat{n}] = -\sum_i \left[\hat{n}\partial_t n + n\lambda\left(e^{-\hat{n}}-1\right) + n\mu\left(e^{\hat{n}}-1\right)\right] + \sum_{<ij>} n_i W_{ij}\left(e^{\hat{n}_j-\hat{n}_i}-1\right) \tag{5.22}$$

Taking the continuum limit when the lattice spacing a tends to zero gives:

$$\begin{aligned} S[\rho,\hat{\rho}] &= \int d^3\mathbf{x}\int dt\, \left\{\hat{\rho}(\mathbf{x},t)\left[-\partial_t\rho(\mathbf{x},t) + T\nabla^2\rho(\mathbf{x},t)\right.\right. \\ &+ \left.\nabla\cdot\left(\rho(\mathbf{x},t)\int d^3\mathbf{y}\ \nabla V(\mathbf{x}-\mathbf{y})\rho(\mathbf{y},t)\right)\right] + T\rho(\mathbf{x},t)(\nabla\hat{\rho}(\mathbf{x},t))^2 \\ &+ \left.\mu\left(e^{\hat{\rho}(\mathbf{x},t)}-1\right)\rho(\mathbf{x},t) - \lambda(1-e^{-\hat{\rho}(\mathbf{x},t)})\rho(\mathbf{x},t)\right\} \end{aligned} \tag{5.23}$$

where we defined the local density field $\rho(x,t) = n_i(t)/a^d$ and the conjugated field $\hat{\rho}(x,t) = \hat{n}_i(t)$. Thus stochastic equations with Poissonian jump processes provide a direct derivation of a density field theory with variable number of particles. Let's also note that in some cases the scaling limit transforms Poissonian noise to a Gaussian noise, for example for interacting Brownian particles we considered in Sec. 4.1.1 and 5.1.

5.2.7 Low density expansion

The ability to express any creation-annihilation processes for the particles in terms of density field ρ and a conjugated field $\hat{\rho}$ suggests a method to derive the low density expansion for Brownian dynamics. Indeed, the λ and μ terms in the action given by Eq. (5.23) fix the average density ρ_0 so that it does not drop out like in Chapter 4. Varying λ and μ one can fix ρ_0 to arbitrary value. However the resulting theory has two difficulties:

- λ and μ terms are not invariant under transformations $\mathcal{T}$ and $\mathcal{U}$. This is straightforward to check by applying the transformations given by Eqs. (4.11) and (4.13). This suggests that the dynamics with variable number of particles has a different transformation associated to time-reversal symmetry that has not been identified so far.

- The field theory has a complicated perturbation theory with exponential vertices. Indeed perturbation in powers of λ and μ leads to the appearance of extra terms like $n\hat{\rho}$ in the Gaussian action as the $\exp\exp(\hat{\rho})$ terms are developed in powers of $\exp(\hat{\rho})$. Therefore the Gaussian action is different from order to order in perturbation theory.

These issues prevents application of the theory to the problem of glass transition where preservation of TRS is of primary importance.

5.3 Summary

In this chapter we aimed at fixing the divergence problem we discussed in Chapter 4. We focused on two different approaches to the problem. First, a different decomposition of the action (4.5) was considered. This decomposition lead to the perturbation theory in powers of the potential which respected TRS and lacked of ultraviolet divergence. We introduced the transformation (5.5) which provided a standard decomposition into Gaussian part and interaction at the price of a complicated expressions for density-density correlator $C_{\rho\rho}$ and response function R. The latter became four-point quantities. However now both of them can be expressed via a single function γ. The primary aim within this theory would be to construct self-consistent perturbation theory and analyse the 1-loop approximation in analogy with Sec. 4.6. Note that this time it is necessary to construct a self-consistent perturbation theory not only with dressed lines but also dressed vertices with 4 legs in order to derive a self-consistent equation on γ.

The other part of the Chapter was focused on the attempt to derive a low density expansion for Brownian dynamics. This required to derive a density field theory with variable number of particles. Such a generalisation of BD was implemented within a context of Doï-Peliti formalism. Although it has not resulted into a correct field theory because of the explicit time-reversal violation it provided some interesting side results. First, our analysis related

the expansion in powers of the potential and the Doï-Peliti formalism and shed light onto the origin of the transformation (5.5) which made quadratic the action S_{FREE}. Second, we developed a direct way to construct density field theories from well-behaved stochastic equations avoiding a somewhat involved coherent state representation.

Chapter 6

Landau theory for the glass transition

As was pointed out earlier in Sec. 2.6 MCT describes quite well the first decades of the slowing of dynamics close to the glass transition and captures the two step relaxation. It also predicts an ideal transition at $T = T_d$ which is not observed in simulations nor experimentally; data deviate systematically from MCT as T_d is approached. As was discussed in Sec. 2.3 MCT transition describes freezing in an amorphous configuration. In finite dimensions barriers between different configurations are always finite so that rearrangement will take place for any $T > 0$ albeit on very long scales. That's why the absence of the transition is usually explained by the appearance of activated events that come into play as the temperature is lowered and provide an extra relaxation channel destroying the transition. These facts imply that MCT is an incomplete theory which is only valid in a certain time and temperature window. As such it needs to be corrected and completed. This motivates to analyse several problems related with MCT:

- Corrections and structural stability: MCT does not provide any systematic method to compute corrections because the Mode-Coupling Factorisation (MCF) is uncontrolled. However it is of crucial importance to be able to compute them in order to improve MCT and to test the structural stability of MCT with respect to neglected contributions. Clearly there is something MCT misses which is responsible for the absence of the transition. But it is *a priori* unclear whether this factor is due to corrections or is a non-perturbative effect. To put it another way: whether the transition can be removed by refining the MCF or the procedure is more subtle. Some steps in this direction have already been done in [69] where higher-order correlations were incorporated in schematic MCT. The authors found that once the correlations are truncated one recovered MCT while no transition is present in the exact solution.

- The problem of corrections can be understood in a broader sense. We discussed earlier in Sec. 2.7 and 4.6 a possible mechanism for the cut off of the transition which relied on inclusion of extra conserved quantities in the dynamics. This is a fundamentally different type of corrections: within the Brownian dynamics of Sec. 4.4.2 one cannot account for transverse currents considered in FNH. At the same time as we have seen the glass transition is actually insensitive to these details: coupling to currents does not smear the transition.

- The growing lengthscale present within MCT which is accessed via a four-point function: it suggests to consider the main MCT Eq. (2.19) as a mean-field equation on an order parameter which is rather unconventional: it is a two-point function rather than a one-point one. This is supported by the fact that schematic MCT which lacks spatial dependence shows all the essential properties of the full MCT. Further analysis shows that spatial fluctuations become infinitely long-ranged at $T = T_d$ which is signaled by a diverging lengthscale like in standard critical phenomena. These fluctuations are expected to change the critical behaviour below the upper critical dimension d_u which is $d_u = 6$ or $d_u = 8$ for no conserved quantities and for dynamics with conserved quantities respectively (BD,FNH) [58, 59]. For $d < d_u$ the Ginzburg criterion delimits a region where critical fluctuations dominate from a region where they can be neglected. However, above d_u one should also worry about the stability of the refined theory (MCT + corrections) with respect to spatial fluctuations. Such test was carried out for the standard MCT within the context of Inhomogeneous MCT [52, 53].

In this Chapter we propose a Landau theory for the glass transition. Within critical phenomena this is a mean-field theory which is stable with respect to corrections and with respect to fluctuations. The main statement that we will prove throughout the whole Chapter is that MCT is a Landau theory for the glass transition. However, we only prove the structural stability; stability with respect to fluctuations is left for a future work.

6.1 Critical phenomena

We start by recalling the definition of a Landau theory for critical phenomena.

6.1.1 Ising model

The Landau theory is a general phenomenological theory for the equilibrium phase transitions [103, 104]. It relies on a number of natural hypotheses:

- There exists an order parameter $\mathbf{m}$ that distinguishes different phases under variation of an external thermodynamic parameter (temperature, magnetic or electric field, pressure) *i.e.* it takes distinct values in different phases. The order parameter varies from system to system: magnetisation for ferromagnets, director for liquid crystals, *etc.*

- The free energy of the system F is analytic in $\mathbf{m}$ and can be expanded in powers of the order parameter. Symmetries of the system can make certain terms in the expansion vanish. This important hypothesis is known to be violated below the upper critical dimension. The series are usually truncated to the lowest non trivial term.
- The coefficients of the series are assumed to be regular functions of the external thermodynamic parameters like temperature, magnetic/electric field, pressure, *etc.* The critical point is defined as the point where one of the coefficients vanishes. In some cases several coefficients can vanish simultaneously like for a tricritical point. Such a coefficient is approximated by a linear function of the external parameter with a zero at a critical point. All the other coefficients are approximated by their values at the critical point. This reduces the validity of the theory to the immediate vicinity of the transition.

In the classic example of the Ising model the order parameter is the magnetisation $\mathbf{m}(\mathbf{x})$. In the absence of magnetic field the system is invariant under $\mathbf{m} \to -\mathbf{m}$. Then the expansion of the free energy $\mathcal{F}$in the homogeneous case $\mathbf{m}(\mathbf{x}) = m$ reads (we also included an external field related term $-mh$):

$$\mathcal{F}[m] = \mathcal{F}_0 + \frac{b}{2}(T - T_c)m^2 + \frac{g}{4!}m^4 - mh \tag{6.1}$$

The extremum of $\mathcal{F}$ with respect to m defines an equilibrium magnetisation $m(T)$:

$$\frac{\delta F}{\delta m} = 0 \quad \longrightarrow \quad \left(b(T - T_c) + \frac{g}{6}m^2\right) m = h \tag{6.2}$$

In zero field $h = 0$, the paramagnetic solution $m = 0$ is stable for $T > T_c$; a non-zero solution $m = \sqrt{-6b(T - T_c)/g}$ only exists for $T < T_c$ and is stable in this region. The linear susceptibility $\chi = dm/dh$ diverges as $(T - T_c)^{-1/2}$ for $T \to T_c+$; right at $T = T_c$, m behaves like $h^{1/3}$ for small fields. These are the predictions of the Landau theory for an homogeneous case. One can go a step further and treat an inhomogeneous case by including a gradient expansion of $\mathbf{m}(\mathbf{x})$ in $\mathcal{F}$:

$$F[\mathbf{m}(\mathbf{x})] = \int d^3x \left[-\mathbf{m}(\mathbf{x})\mathbf{h}(\mathbf{x}) + \frac{a}{2}(\nabla\mathbf{m}(\mathbf{x}))^2 + \frac{b}{2}(T - T_c)\mathbf{m}^2(\mathbf{x}) + \frac{g}{4!}\mathbf{m}^4(\mathbf{x})\right]$$

Minimising the free energy with respect to $\mathbf{m}(\mathbf{x})$ yields in Fourier space:

$$\frac{\delta F}{\delta \mathbf{m}(\mathbf{x})} = 0 \quad \longrightarrow \quad \left(-a\Delta_x + b(T - T_c) + \frac{g}{6}\mathbf{m}^2(\mathbf{x})\right) \mathbf{m}(\mathbf{x}) = \mathbf{h}(\mathbf{x})$$

$$\chi(\mathbf{q}) = \frac{1}{a\mathbf{q}^2 + b(T - T_c) + \frac{g}{6}\mathbf{m}^2} = \frac{\xi^2}{1 + (\xi\mathbf{q})^2} \qquad \xi \sim (T - T_c)^{-1/2}$$

and the divergence of the susceptibility is associated with infinite range correlation of the magnetisation at T_c.

6.1.2 Breakdown of the Landau theory

It is well-established that the Landau theory breaks down below the upper critical dimension. It is instructive to classify the origins of this failure:

- Higher order terms in the expansion of $\mathcal{F}[\mathbf{m}]$ could alter the predictions. This happens, for example, if $g \to 0$ produces a tricritical point or if neglected terms induce a first order transition (this would be the case of a cubic term in the above expansion). But if the transition remains of second order than the neglected terms are irrelevant as $|T - T_c| \to 0$ because $m \sim \sqrt{T - T_c} \to 0$.

- Critical fluctuations: the Landau theory completely neglects the non-linear feedback from spatial fluctuations which changes the critical exponents below d_u. On the other hand one can prove, for example using diagrammatic theory, that for $d > d_u$ the effect of spatial fluctuations is negligible and the predictions of the Landau theory are correct.

Studies on growing correlation length and related multi-point functions provided an approach to MCT in a sense equivalent to that of previous section. The aim of the following analysis is to show that in this complicated case corrections do not modify the predictions and MCT can be understood as the Landau theory for glass transition. The complicated proof of the structural stability will be the scope of the following sections. The technical difficulty is that the order parameter for MCT transition is a time-dependent, two-point correlator $C(\mathbf{q}, \tau)$. To complete the construction of the Landau theory we should also test the stability with respect to spatial fluctuations for $d > d_u$. However we will completely disregard this issue and focus on the proof of the structural stability only. We also consider only the high temperature phase where time reversal symmetry (TRS) hold and hence fluctuation-dissipation theorem (FDT) hold. Although there are indications that the Landau theory can be generalised to a low-T phase where these symmetries are broken this is an even more complicated problem as we will see in Chapter 7.

6.2 Glass transition

The proof of structural stability is carried out in two steps. First, we provide a general proof for structural stability of MCT. Since several important subtleties appear in the derivation, we highlight them in a detailed proof of the Landau theory within perturbative expansions of particular models.

6.2.1 The background

The case of the glass transition is quite different from critical phenomena. Several formal differences prevent direct analogy. First, the glass transition is a purely dynamical phenomenon: static, thermodynamic properties do not present

any peculiarities as a liquid freezes into a glass. Indeed, the static structure factor $\langle \delta\rho(\mathbf{q},t)\rangle$ presents no peculiarities as the dynamics slows down. Second, the standard construction with the free energy as a main object simply does not exist due to the absence of a clear analogy to the free energy in the dynamical case. The results of the previous chapters allow to assume that the slowing down of the dynamics in a supercooled liquid is governed by density fluctuations; energy/momentum fluctuations are less important and can be neglected [91, 68, 105]. Density fluctuations are defined as $\delta\rho(\mathbf{q},t) = \rho(\mathbf{q},t) - \langle\rho\rangle$ where angular brackets denote the thermal average and $\mathbf{q}$ is a wave-vector. Contrary to critical phenomena where the order parameter is a static one point function order parameter in glasses is a dynamic two point correlators: the main objects are dynamic structural correlator $C(\mathbf{q},\tau) = \langle\delta\rho(\mathbf{q},t)\delta\rho(-\mathbf{q},s)\rangle$ and response function $R(\mathbf{q},\tau) = \langle\delta\rho(\mathbf{q},t)/\delta U(-\mathbf{q},s)\rangle$ at $U = 0$ (where $\tau = t - s$) which describes the reaction of the system on the external potential U.

Let's recall briefly the phenomenology of the transition (presented also as MCT results 2.3). The slowing down of the dynamics is accompanied by the appearance of a plateau $f_{\mathbf{q}}$ in the relaxation pattern of $C(\mathbf{q},\tau)$ and of two diverging timescales: one governing the approach to the plateau (β regime) and the other controlling the decay off the plateau and corresponding to a structural relaxation of the system (And which was denoted as α regime in the context of MCT). Within MCT both timescales diverge at $T = T_d$ and $C(\mathbf{q},\tau)$ acquires a non-zero limit at infinity

$$f_{\mathbf{q}} = \lim_{\tau\to\infty} C(\mathbf{q},\tau) \qquad T = T_d.$$

The non-ergodic parameter $f_{\mathbf{q}}$ does not provide access to important details of the transition like diverging timescales. As we have seen in 2.3 as the timescales increase the correlator $C(\mathbf{q},\tau)$ can be described as

$$C(\mathbf{q},\tau) \approx f_{\mathbf{q}} + \delta C(\mathbf{q},\tau)$$

in an increasing time window as T approaches T_d. MCT provided some explicit predictions about $\delta C(\mathbf{q},\tau)$ (see 2.3) and various scaling laws. The underlying idea of our Landau theory is to consider $\delta C(\mathbf{q},\tau)$ as the generalisation of the order parameter to the case of the glass transition and to derive a structurally stable dynamic equation for it which generalises (2.22).

Such a derivation is possible in the framework of the field theories (Brownian dynamics and FNH) presented in 4 or 3-spin model presented in 3.1 which provide exact dynamical equations for C and R and hence in principle, for δC. These theories give a very different sets of equations however one can prove that close to the transition they can all be reduced to a single equation:

$$\partial_\tau C(\mathbf{q},\tau) + TC(\mathbf{q},\tau) + \kappa \int_0^\tau du\Sigma(\mathbf{q},\tau-u)\partial_u C(\mathbf{q},u) = 0 \tag{6.3}$$

with initial conditions: $C(\mathbf{q},0) = S_{\mathbf{q}}$. The Laplace transform reads:

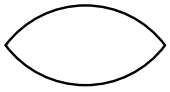
$$\frac{T\hat{C}(\mathbf{q},z)}{S_{\mathbf{q}}-z\hat{C}(\mathbf{q},z)}=1-\kappa\hat{\Sigma}(\mathbf{q},z) \tag{6.4}$$

The self-consistent self-energy $\Sigma(\mathbf{q},\tau)$ (or memory kernel in MCT terminology) is given by a sum of amputated 2 particle irreducible (2-PI) self-consistent diagrams built with C and R lines (see 4.4,4.5,3.1). We do not specify the details of the field theory underlying this equation, nor the Feynman rules for the diagrams contributing to Σ: we just need that such a theory exists. As we specified above we consider the high-T region so that the system is at equilibrium: both R and C are time-translation invariant and FDT holds at a diagrammatic level: $R(\mathbf{q},\tau)=-1/T\partial_\tau C(\mathbf{q},\tau)$.

Note that (6.3) resembles structurally (2.19) derived in the context of MCT. However, we have already stressed that within MCT there is no well defined prescription on how to build consistent approximations for Σ. In this sense MCT is a non-controlled approximation hard to improve: systematic corrections would be hard to identify and to account for. The field-theoretic approach provides approximations for Σ on a systematic basis (like the loop expansion of Σ).

6.2.2 1-loop approximation

In order to identify a candidate for a Landau theory one should find an approximation for Σ that is stable structurally *i.e.* stable with respect to corrections. Our main statement is that MCT is the Landau theory and further corrections bring no qualitative changes. One should find an approximation for Σ that reproduces (2.19). Fortunately this is an easy task as we have seen in 4.6: MCT is reproduced in the context of (6.3) within the 1-loop approximation for Σ when one retains only 1-loop diagram in the expansion of Σ:

$\Sigma(\mathbf{q},t-s)=$ t  s $=$ (6.5)

$$=\int\frac{d^3\mathbf{k}}{(2\pi)^3}V(\mathbf{q},\mathbf{k})C(\mathbf{q}-\mathbf{k},t-s)C(\mathbf{k},t-s)$$

where $V(\mathbf{q},\mathbf{k})$ is the effective vertex that makes the integral over $\mathbf{k}$ convergent. As we have seen earlier in Chapter 4 a problem of ultraviolet divergence related to an ill-behaved $V(\mathbf{q},\mathbf{k})$ appears within field theories. This divergence is regularised by a large wave-vector cutoff which we imply henceforth. Note that the cutoff should not be relevant for the proof of structural stability.

Now (6.4) reads:

$$\frac{T\hat{C}(\mathbf{q},z)}{S_{\mathbf{q}}-z\hat{C}(\mathbf{q},z)}=1-\kappa\int\frac{d^3\mathbf{k}}{(2\pi)^3}V(\mathbf{q},\mathbf{k})\mathcal{L}\left[C(\mathbf{q}-\mathbf{k},\tau)C(\mathbf{k},\tau)\right] \tag{6.6}$$

The resulting equation is identical to (2.19) up to a redefinition of the coefficients and an appropriate choice of the kernel $V(\mathbf{q},\mathbf{k})$. The analysis of this equation reproduces the results presented in 2.3.

6.2.3 The general case

Thus we have accomplished the first step and identified a candidate for the Landau theory. The next step is to prove that extra terms in the expansion of Σ do not modify this result. Let's consider (6.3) with Σ given by a finite sum of 2-PI diagrams including the one loop diagram. An important complication appears compared to the one-loop case: beyond 1-loop, diagrams become non-local in time. The proof is done in a self-consistent manner: we assume some properties of the solution of (6.3) and demonstrate that they are consistent with the results. Our assumptions about the solution of (6.3) parallel the results found within MCT (Sec. 2.3), observed experimentally and in simulations:

- Structural arrest at a certain temperature T_d with broken ergodicity below this temperature. The correlator $C(\mathbf{q},\tau)$ tends asymptotically to a non-zero value $f_{\mathbf{q}}$ at $\tau=\infty$.
- Appearance of a two-step relaxation pattern with three well separated time scales for $\epsilon\ll 1$ where $\epsilon=(T-T_d)/T_d>0$:
  - Short times $\tau\sim 1$ where $C(\mathbf{q},\tau)=C_0(\mathbf{q},\tau)$ and $C_0(\mathbf{q},\tau\to\infty)\to f_{\mathbf{q}}$.
  - β-relaxation $\tau=s\tau_\beta(\epsilon)$ with $s=O(1)$: $\delta C(\mathbf{q},s\tau_\beta)=C(\mathbf{q},\tau)-f_{\mathbf{q}}=S_{\mathbf{q}}m(\epsilon)(1-f_{\mathbf{q}})^2G(\mathbf{q},s))$ where $m(\epsilon)$ measures a deviation from the plateau and vanishes as $\epsilon\to 0$.
  - α-relaxation $\tau=x\tau_\alpha(\epsilon)$ with $x=O(1)$: $C(\mathbf{q},\tau)=C_\alpha(\mathbf{q},x)$ $(C_\alpha(0)-f_{\mathbf{q}})$ describes a final fall off the relaxation.
- We further assume that $G(\mathbf{q},s)$ can be expanded in powers of $m(\epsilon)$:

$$G(\mathbf{q},s)=\sum_{n>0}m^{n-1}(\epsilon)G_n(\mathbf{q},s)$$

  All the functions G_n are *a priori* singular at $s=0$ and at $s=\infty$ since they should match short times and α relaxation respectively where the deviation from the plateau δC ceases to be small. A crucial remark for what follows is that any function G_n is accompanied by a prefactor $m^n(\epsilon)$.

These hypotheses turn out to be sufficient to generalise MCT (1 loop) results in the β regime. First, the expansion of $C(\mathbf{k}, \tau/\tau_\beta)$ implies a similar expansion of the self-energy $\Sigma(\mathbf{k}, \tau/\tau_\beta)$ valid in the β-regime:

$$\begin{aligned}\Sigma(\mathbf{q}, s\tau_\beta) &= \Sigma_0(\mathbf{q}, s) + m(\epsilon)\Sigma_1(\mathbf{q}, s) + m^2(\epsilon)\Sigma_2(\mathbf{q}, s) + \cdots \\ \hat{\Sigma}(\mathbf{q}, z/\tau_\beta) &= \hat{\Sigma}_0(\mathbf{q}, z) + m(\epsilon)\hat{\Sigma}_1(\mathbf{q}, z) + m^2(\epsilon)\hat{\Sigma}_2(\mathbf{q}, z) + \cdots\end{aligned} \tag{6.7}$$

The coefficients Σ_n are defined as:

$$\Sigma_n(\mathbf{q}, s) = \left.\frac{d^n \Sigma(\mathbf{q}, s\tau_\beta(\epsilon))}{dm(\epsilon)^n}\right|_{m(\epsilon)=0}$$

and therefore do not depend on ϵ, but are some functionals of $C(\mathbf{q}, \tau)$. Namely, their most general functional form *a priori* includes contributions from all the scaling regimes introduced above:

$$\Sigma_n(\mathbf{q}, s) = \Sigma_n\Big[C_0(\mathbf{p}, s\tau_\beta), G(\mathbf{p}, s), C_\alpha(\mathbf{p}, s\tau_\beta/\tau_\alpha)\Big].$$

However, the independence from ϵ puts constraints on the actual functional form of Σ_n. We will illustrate this on the first three coefficients Σ_0, Σ_1 and Σ_2. The knowledge of these coefficients allows to fix G_1 and the scaling laws; higher order coefficients are actually irrelevant.

The zeroth order term $\Sigma_0(\mathbf{q}, s)$ can only be a pure wave-vector function $\Sigma_0(\mathbf{q})$. Since $\Sigma_0 = \lim_{\epsilon\to 0} \Sigma(\mathbf{q}, s\tau_\beta)$ and the time scales separate the general functional form for Σ_n simplifies for $n = 0$ to

$$\Sigma_0(\mathbf{q}) = \Sigma_0[C_0(\mathbf{q}, \infty), 0, C_\alpha(\mathbf{p}, 0)] = \Sigma_0[f_\mathbf{p}]$$

and all the s-dependence drops out. Thus this term is irrelevant for the Landau theory.

The first order term $\Sigma_1(\mathbf{q}, s)$ has two contributions: an s-independent term and an s-dependent term. The first one is irrelevant: it only renormalises the temperature (which is not computed exactly in the context of the Landau theory anyway). The second term admits a single representation

$$\Sigma_1(\mathbf{q}, s) = \int_0^\infty du \int \frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_1(\mathbf{q}, \mathbf{k}; s, u)G_1(\mathbf{k}, u).$$

Any other combination containing higher G_n's would produce an extra $m^n(\epsilon)$: this is a higher order contribution which can be safely neglected. The kernel σ_1 has a regular shape in the original time variables with a span fixed by a microscopic timescale. Hence it should be local in the rescaled variables s, u. To lowest order σ_1 is a δ-function in s, u, derivatives of the δ-function corresponding to higher order contributions. Finally:

$$\Sigma_1(\mathbf{q}, s) = \int \frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_1(\mathbf{q}, \mathbf{k})G_1(\mathbf{k}, s)$$

The second order term $\Sigma_2(\mathbf{q},s)$ has a richer structure. First, there is a term similar to the first order term Σ_1 with G_2: $\int \frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_{21}(\mathbf{q},\mathbf{k})G_2(\mathbf{k},s)$. The reason for its local time structure is exactly the same as for Σ_1. Second, there are terms quadratic in G_1:

$$\int \frac{d^3\mathbf{k}}{(2\pi)^3}\int \frac{d^3\mathbf{p}}{(2\pi)^3}\int\limits_0^\infty du\int\limits_0^\infty dv\sigma_2(\mathbf{q},\mathbf{k},\mathbf{p};s,u,v)G_1(\mathbf{k},u)G_1(\mathbf{p},v).$$

The time dependent part of $\sigma_2(\mathbf{q},\mathbf{k},\mathbf{p};s,u,v)$ is composed of δ-functions and their derivatives (for simplicity the wave-vector dependence is dropped):

$$\sigma_2(s,u,v)=\sigma_{2,loc}\delta(s-u)\delta(s-v)+\sigma_2\delta(v+u-s)(\partial_u+\partial_v)+\tilde{\sigma}_2(\partial_u+\partial_v)\delta(v+u-s)$$

The fact that only the $u+v-s$ combination enters the above expression is a direct consequence of the equilibrium. Another consequence is that in the above time integrations the above limit is not an infinity but s. The full justification is provided in Appendices $A1$ and $A2$. The first local term generalises the usual MCT, "1-loop like" contribution, but the other terms do not appear within a standard MCT (at least at a first glance). The third term is related to the second one by integration by parts:

$$\begin{aligned}\int\limits_0^s du[\partial_u G_1(\mathbf{k},u-s)]G_1(\mathbf{p},u) &= G_1(\mathbf{k},s)G_1(\mathbf{p},s)-G_1(\mathbf{k},0)G_1(\mathbf{p},0)- \\ &- \int\limits_0^s du\partial_u G_1(\mathbf{p},u)G_1(\mathbf{k},u-s) \qquad (6.8)\end{aligned}$$

The term $G_1(\mathbf{k},0)G_1(\mathbf{p},0)$ looks divergent because of the singular asymptotics of $G_1(\mathbf{q},s)$ at $s=0$. However this divergence can be regularised as we demonstrate below.

Finally (6.7) reads:

$$\begin{aligned}\Sigma(\mathbf{q},s\tau_\beta)=\Sigma_0(\mathbf{q})+m(\epsilon)\int\frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_1(\mathbf{q},\mathbf{k})G_1(\mathbf{k},s)+ \\ +m^2(\epsilon)\int\frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_2(\mathbf{q},\mathbf{k})G_2(\mathbf{k},s)+ \\ +m^2(\epsilon)\int\frac{d^3\mathbf{k}}{(2\pi)^3}\int\frac{d^3\mathbf{p}}{(2\pi)^3}\sigma_{21,loc}(\mathbf{q},\mathbf{k},\mathbf{p})G_1(\mathbf{k},s)G_1(\mathbf{p},s)+ \qquad (6.9)\end{aligned}$$

$$+m^2(\epsilon)\int\frac{d^3\mathbf{k}}{(2\pi)^3}\int\frac{d^3\mathbf{p}}{(2\pi)^3}\sigma_{21,nl}(\mathbf{q},\mathbf{k},\mathbf{p})\int\limits_0^s du[\partial_u G_1(\mathbf{k},u)]G_1(\mathbf{p},s-u) \qquad (6.10)$$

Or after Laplace transform

$$\hat{\Sigma}(\mathbf{q}, z/\tau_\beta) = \frac{\Sigma_0(\mathbf{q})}{z} + m(\epsilon)\int \frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_1(\mathbf{q},\mathbf{k})\hat{G}_1(\mathbf{k},z) + \\ +m^2(\epsilon)\int \frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_2(\mathbf{q},\mathbf{k})\hat{G}_2(\mathbf{k},z) + \\ +m^2(\epsilon)\int \frac{d^3\mathbf{k}}{(2\pi)^3}\int \frac{d^3\mathbf{p}}{(2\pi)^3}\sigma_{21,loc}(\mathbf{q},\mathbf{k},\mathbf{p})\mathcal{L}[G_1(\mathbf{k},s)G_1(\mathbf{p},s)](z) + \\ +m^2(\epsilon)\int \frac{d^3\mathbf{k}}{(2\pi)^3}\int \frac{d^3\mathbf{p}}{(2\pi)^3}\sigma_{21,nl}(\mathbf{q},\mathbf{k},\mathbf{p})(z\hat{G}_1(\mathbf{k},z) - G_1(\mathbf{k},0))\hat{G}_1(\mathbf{p},z)$$

where $\sigma_{21,nl}$ accounts for all non-local contributions.

This expansion is the central result in the construction of the Landau theory. It allows to expand (6.3) in β regime in powers of $m(\epsilon)$ using (6.9) and expansion of C. After the Laplace transform expansion of (6.4) reads:

$$\frac{T_d f_{\mathbf{q}}\epsilon}{z(1-f_{\mathbf{q}})} + m(\epsilon)\left[T_d(1+\epsilon)\hat{G}_1(\mathbf{q},z) + \kappa\int \frac{d^3\mathbf{k}}{(2\pi)^3}\Sigma_1(\mathbf{q},\mathbf{k})\hat{G}_1(\mathbf{k},z)\right] + \\ +T_d(1+\epsilon)m^2(\epsilon)\left[\hat{G}_2(\mathbf{q},z) + \kappa\int \frac{d^3\mathbf{k}}{(2\pi)^3}\Sigma_2(\mathbf{q},\mathbf{k})\hat{G}_2(\mathbf{k},z)\right] + \\ m^2(\epsilon)T_d(1+\epsilon)(1-f_{\mathbf{q}})z\hat{G}_1^2(\mathbf{q},z) = -m^2(\epsilon)\kappa\int \frac{d^3\mathbf{k}}{(2\pi)^3}\Sigma_{2,loc}(\mathbf{q},\mathbf{k})\hat{G}_2(\mathbf{k},z) - \\ -m^2(\epsilon)\kappa\int \frac{d^3\mathbf{k}}{(2\pi)^3}\int \frac{d^3\mathbf{p}}{(2\pi)^3}\Sigma_{21,loc}(\mathbf{q},\mathbf{k},\mathbf{p}) * L[G_1(\mathbf{k},\tau)G_1(\mathbf{p},\tau)](z) - \\ -m^2(\epsilon)\kappa\int \frac{d^3\mathbf{k}}{(2\pi)^3}\int \frac{d^3\mathbf{p}}{(2\pi)^3}\Sigma_{21,nl}(\mathbf{q},\mathbf{k},\mathbf{p})z\hat{G}_1(\mathbf{k},z)\hat{G}_1(\mathbf{p},z) \quad (6.11)$$

where we dropped the terms independent of ϵ which fix the non-ergodic parameter $f_{\mathbf{q}}$:

$$\frac{T_d f_{\mathbf{q}}}{1-f_{\mathbf{q}}} = -\kappa\Sigma_0(\mathbf{q}) \quad (6.12)$$

and $\Sigma_0(\mathbf{q})$ is a functional of f as we have proved above.

Identifying the coefficients of the expansion to zero produces series of equations. The first order fixes the yet unknown function $m(\epsilon)$: expansion (6.3) has terms with explicit powers of ϵ. They should be matched with powers of $m(\epsilon)$. Inspection of (6.11) shows that there are two possibilities: either $m(\epsilon) = \epsilon$, either $m(\epsilon) = \sqrt(\epsilon)$. The first choice yields a trivial solution for G_1 which is in contradiction the assumption of a two-step relaxation pattern with diverging timescales. Namely if $m(\epsilon) = \epsilon$ in (6.3) yields:

$$\frac{T_d f_{\mathbf{q}}}{z(1-f_{\mathbf{q}})} + \int \frac{d^3\mathbf{k}}{(2\pi)^3}\left[T_d\delta(\mathbf{q}-\mathbf{k}) + \kappa\sigma_1(\mathbf{q},\mathbf{k})\hat{G}_1(\mathbf{k},z)\right]\hat{G}_1(\mathbf{k},z) = 0 \quad (6.13)$$

$$\frac{T_d f_{\mathbf{q}}}{z(1-f_{\mathbf{q}})} + \mathbf{K}\hat{G}_1(\mathbf{q},z) = 0$$

where we introduced an operator $\mathbf{K}$ with the kernel $K(\mathbf{q},\mathbf{k}) = T_d\delta(\mathbf{q}-\mathbf{k}) + \kappa\sigma_1(\mathbf{q},\mathbf{k})$. Let's suppose that we know the eigenvectors $\hat{G}_1^{\Lambda}(\mathbf{q},z)$ and the eigenvalues Λ of $\mathbf{K}$. Then the above equation reads for any given eigenvalue Λ:

$$\frac{T_d f_{\mathbf{q}}}{z(1-f_{\mathbf{q}})} + \Lambda\hat{G}_1^{\Lambda}(\mathbf{q},z) = 0 \quad (6.14)$$

This equations are immediately solved with respect to z implying that $G_1^{\Lambda}(\mathbf{q},s)$ do not actually depend on s. Therefore $G_1(\mathbf{q})$ contributes to the height of the plateau $f_{\mathbf{q}}$ but do not describe a non-trivial dynamics which imposes singularities at $s=0$ and at $s=\infty$, in contradiction with our hypothesis. We are then forced to conclude that $m(\epsilon) = \sqrt{(}\epsilon)$ like at 1-loop and corrections do not renormalise this result. It is worth to mention that different solutions referred to as A_l singularities are possible for $m(\epsilon)$ [106, 107, 108].

The equation of order $\sqrt{\epsilon}$ reads:

$$T_d\hat{G}_1(\mathbf{q},z) = -\kappa\int\frac{d^3\mathbf{k}}{(2\pi)^3}\Sigma_1(\mathbf{q},\mathbf{k})\hat{G}_1(\mathbf{q},z) \quad (6.15)$$

This is a classical eigenvalue problem within MCT (see 2.3). It shows that G_1 is a product of wave-vector dependent and time dependent amplitudes thus reproducing the well-know MCT "factorisation property": $\hat{G}_1(\mathbf{q},z) = \hat{g}(z)H_1(\mathbf{q})$ where H_1 is the eigenvector of $-\kappa\sigma_1$ with the largest eigenvalue T_d. But the scaling function $\hat{g}(z)$ remains unfixed at this order and higher orders in $\sqrt{\epsilon}$ must be considered.

The second order equation is trickier:

$$\begin{aligned} T_d\hat{G}_2(\mathbf{q},z) + \kappa\int\frac{d^3\mathbf{k}}{(2\pi)^3}\Sigma_2(\mathbf{q},\mathbf{k})\hat{G}_2(\mathbf{k},z) = - \\ -\frac{T_d f_{\mathbf{q}}}{z(1-f_{\mathbf{q}})} - T_d(1-f_{\mathbf{q}})z\hat{g}^2(z)H_1^2(\mathbf{q}) - \\ -\kappa\int\frac{d^3\mathbf{k}}{(2\pi)^3}\int\frac{d^3\mathbf{p}}{(2\pi)^3}\Sigma_{21,loc}(\mathbf{q},\mathbf{k},\mathbf{p},z)L[g^2]H_1(\mathbf{k})H_1(\mathbf{p}) - \\ -\kappa z\hat{g}^2(z)\int\frac{d^3\mathbf{k}}{(2\pi)^3}\int\frac{d^3\mathbf{p}}{(2\pi)^3}\Sigma_{21,nl}(\mathbf{q},\mathbf{k},\mathbf{p})H_1(\mathbf{k})H_1(\mathbf{p}) \end{aligned} \quad (6.16)$$

An important remark is that the linear operator acting on $\hat{G}_2$ is exactly the same as the one in (6.15). This can be seen from the following argument: expanding

C as $C(s\tau_\beta) = S_{\mathbf{q}}(f_{\mathbf{q}} + \sqrt{\epsilon}(1-f_{\mathbf{q}})^2 G(\mathbf{q},s))$, $\Sigma(s\tau_\beta)$ can itself be expanded in powers of $\sqrt{\epsilon}G(\mathbf{q},s)$:

$$\Sigma(\mathbf{q}, s\tau_\beta) = \Sigma_0(\mathbf{q}) + \sqrt{\epsilon}\int \frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_1^\epsilon(\mathbf{q},\mathbf{k})G(\mathbf{k},s) + \cdots$$

where the superscript ϵ of σ_1 denotes that it depends on ϵ contrary to previously defined σ_n. The linear terms read:

$$\Sigma(\mathbf{q}, s\tau_\beta) = \Sigma_0(\mathbf{q}) + \sqrt{\epsilon}\int \frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_1(\mathbf{q},\mathbf{k})G_1(\mathbf{k},s) + \epsilon\int \frac{d^3\mathbf{k}}{(2\pi)^3}\sigma_1(\mathbf{q},\mathbf{k})G_2(\mathbf{k},s) + \cdots$$

proving the statement.

Following [21] we now multiply (6.16) by $H_1(\mathbf{q})$ and integrate over $\mathbf{q}$. The G_2 part of the equation vanishes (G_2 is also an eigenvector with the eigenvalue T_d like G_1) and the remainder yields an equation on g. After some algebra and a rescaling of z and $\hat{g}$ one finds:

$$\frac{1}{z} + \frac{z}{\lambda}\hat{g}_1^2 = L[g_1^2] \tag{6.17}$$

where λ is a certain numerical constant including the non-local contributions. Up to a shift of λ we recovered (2.22) found within MCT previously. We therefore know immediately its solution and properties: g has a singular power law asymptotics at $z \to \infty$: $\hat{g}(z) \sim z^{a-1}$ and $z \to 0$: $\hat{g}(z) \sim z^{-1-b}$ in accordance with our assumptions. The small time exponent a and long time exponent b characterise the decay to and off the plateau $f_{\mathbf{q}}$ and are related by a celebrated equation (2.23):

$$\frac{\Gamma^2(1-a)}{\Gamma(1-2a)} = \frac{\Gamma^2(1+b)}{\Gamma(1+2b)} = \lambda \tag{6.18}$$

which is a genuinely non-trivial MCT result. This result is thus quite general and survives the introduction of an arbitrary number of loop corrections. On the other hand the numerical values of a and b are not universal since they are determined by λ which is clearly renormalised by corrections.

The fact that the form of g is the same as at 1-loop (MCT) has 2 consequences. First, it fixes functional dependence of the time scales

$$\tau_\beta \sim \epsilon^{-1/2a} \qquad \tau_\alpha \sim \epsilon^{-\gamma}$$

with $\gamma = 1/2a + 1/2b$. This stems from the matching of $C(\mathbf{q}, s\tau_\beta)$ at both ends of β regime with the solutions at other regimes. Second, this allows to regularise the above divergences by introducing a small time cut-off in the integrals (6.8). in terms of the function $g(s)$ one finds:

$$\sqrt{\epsilon}\int_{\delta\epsilon^{1/2a}}^{s} dug'(u)g(s-u) = \sqrt{\epsilon}\int_{\delta\epsilon^{1/2a}}^{s} dug'(u)[g(s-u)-g(s)] + \\ + g^2(s) - \sqrt{\epsilon}g(s)g(\delta\epsilon^{1/2a})$$

Now the first term is regular as $\epsilon \to 0$. The second term contributes to the standard MCT while the last term should be computed in the limit $\epsilon \to 0$ followed by $\delta \to \infty$ (the order is important): it behaves like $\sim \delta^{-a}$ and tends to zero as $\delta \to \infty$. This regularisation renormalises the parameters only and is not relevant (See also Appendix C for details).

The conclusion is that the only extra contribution that can be constructed at second order in $g(s)$, namely the non-local term proportional to $\int du g'(u) g(s-u)$, leaves the basic MCT equation, Eq. (6.17), unchanged. Since higher order terms in ϵ (cubic terms in $g(s)$, etc.) are completely irrelevant to obtain Eq. (6.17), there is a strong degree of universality in the above derivation, and in the final MCT prediction, Eq. (6.18). This universality with respect to higher order *local* (in time) corrections was of course already shown by Götze long ago [109, 106]; here we have proven that this result is robust with respect to non-local corrections as well.

6.2.4 Diverging lengthscale

We have seen in 2.5 how MCT equations are generalised in the presence of spatial inhomogeneities, where the correlation function C becomes space dependent: $C(\mathbf{q}, \mathbf{r})$. When the scale of the inhomogeneities is large, one can establish a gradient expansion of the MCT equations [52] which parallels the derivation of 2.5. In the schematic limit where all wave-vector dependence is discarded, the self-energy reads:

$$\Sigma[C](s) = C(\mathbf{r}, s)^2 + w_1 C(\mathbf{r}, s) \nabla^2 C(\mathbf{r}, s) + w_2 \nabla C(\mathbf{r}, s) \cdot \nabla C(\mathbf{r}, s) \qquad (6.19)$$

where w_1 and w_2 are some coefficients [52]. These gradient terms are very important because they show how the MCT transition is in fact associated with a diverging correlation length, which corresponds to the scale over which a localised perturbation affects the surrounding dynamics. The corresponding susceptibility $\chi(\mathbf{p}, s)$ was computed in details in [52] ($\mathbf{p}$ is here the wavevector conjugated to $\mathbf{r}$ and should not be confused with the wavevectors $\mathbf{q}, \mathbf{k}$ used above). The correlation length ξ diverges as $\epsilon^{-1/4}$, and the long-ranged critical fluctuations renormalise the value of the MCT exponents in $d < d_u$ [58, 59]. The above analysis, which was done in the homogeneous limit $\nabla \to 0$, should be repeated in the inhomogeneous case. We expect that the same conclusion will hold, namely that the results obtained within inhomogeneous MCT are stable against the addition of higher order corrections. This would complete the proof of the structural stability of MCT and the validity of neglecting the spatial fluctuations for $d > d_u$.

6.3 Brownian dynamics

The scope of the following sections is the illustration of the generic recipes presented above in the context of particular microscopic models of MCT transition.

The main focus will the derivation of the kernel expansion (6.9) from diagrammatic perturbation expansions for $\Sigma(\mathbf{q},\tau)$. We consider Brownian dynamics of interacting particles as presented in 4 (see 4.4.2) and the schematic 3-spin model. Derivation for the Fluctuating Hydrodynamics (Newtonian dynamics) follows closely that of Brownian dynamics but is more involved technically because of greater number of equations (see Appendix in chapter 4).

6.3.1 Reduction of the Schwinger-Dyson equations

Let's first show how equations (4.50)-(4.52) of 4.4.3 reduce to a single equation (6.3). Lets' recall them first:

$$\partial_\tau C_{\rho\rho}(\mathbf{q},\tau) + \rho_0\mathbf{q}^2 C_{\rho\theta}(\mathbf{q},\tau) = -\frac{1}{T}\int_0^\tau du\,\Sigma_{\hat\rho\hat\rho}(\mathbf{q},\tau-u)C_{\rho\theta}(\mathbf{q},u) - \frac{1}{T}\int_0^\tau du\,\Sigma_{\hat\rho\hat\theta}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u) \tag{6.20}$$

$$\partial_\tau C_{\rho\theta}(\mathbf{q},\tau) + \rho_0\mathbf{q}^2 C_{\theta\theta}(\mathbf{q},\tau) = \frac{1}{T}\Sigma_{\hat\rho\hat\theta}(\mathbf{q},\tau)C_{\rho\theta}(\mathbf{q},0) - \frac{1}{T}\int_0^\tau du\,\Sigma_{\hat\rho\hat\rho}(\mathbf{q},\tau-u)C_{\theta\theta}(\mathbf{q},u) - \frac{1}{T}\int_0^\tau du\,\Sigma_{\hat\rho\hat\theta}(\mathbf{q},\tau-u)\partial_u C_{\rho\theta}(\mathbf{q},u) \tag{6.21}$$

$$W(\mathbf{q})C_{\rho\rho}(\mathbf{q},\tau) - C_{\rho\theta}(\mathbf{q},\tau) = \frac{1}{T}\Sigma_{\hat\theta\hat\theta}(\mathbf{q},0)C_{\rho\rho}(\mathbf{q},\tau) - \frac{1}{T}\int_0^\tau du\,\Sigma_{\hat\rho\hat\theta}(\mathbf{q},\tau-u)C_{\rho\theta}(\mathbf{q},u) - \frac{1}{T}\int_0^\tau du\,\Sigma_{\hat\theta\hat\theta}(\mathbf{q},\tau-u)\partial_u C_{\rho\rho}(\mathbf{q},u) \tag{6.22}$$

These exact equations are closed by self-consistent series for self-energies based on the large density expansion presented in 4.4.2. We assume large wavevector cut-off that regularises the expansion. Structural stability of MCT with respect to high order corrections is sensitive to the time dependence structure of the perturbative series. This is confirmed by results of sMCT. Wavevector dependence becomes important for the proof of stability with respect to fluctuations.

Only the density correlator $C_{\rho\rho}(\mathbf{q},\tau)$ is expected to exhibit slow dynamics characterised by a two-step relaxation pattern while the other correlators are expected to be irrelevant: as we showed in 4.6 these correlators are related to averages of the force; one does not expect to have frozen forces at the transition. It is worth recalling that equation (6.22) fixes the non-ergodic parameter. It is natural to assume that it is the most important; we only need to neglect somehow the terms related to irrelevant quantities *i.e.* decouple it from the rest of equations. The reduction to a single equation on $C_{\rho\rho}$ is made self-consistently: we assume the 1-loop properties for $C_{\rho\rho}(\mathbf{q},\tau)$ and $\Sigma_{\hat\theta\hat\theta}$, and prove that other contributions are negligible; a similar hypothesis is assumed for self-energies where only $\Sigma_{\hat\theta\hat\theta}$ is relevant. Let's fix time behaviour of the $C_{\rho\theta}$ and $C_{\theta\theta}$ as $T =$

$T_d(1+\epsilon)$ and $\epsilon \ll 1$. The limit $\tau \to \infty$ of (6.20)-(6.22) implies a zero infinite time limit of $C_{\rho\theta}$ and $C_{\theta\theta}$ [91] because they are integrable: $\int_0^\infty d\tau C_{\rho\theta,\theta\theta}(\mathbf{q},\tau) < \infty$. The same statement holds for self-energies: $\int_0^\infty d\tau \Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q},\tau) < \infty, \int_0^\infty d\tau \Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q},\tau) < \infty$. Landau theory applies to the β-relaxation regime. Integrability imposes the following scaling for these correlators and self-energies:

$$
\begin{aligned}
C_{\rho\theta}(\mathbf{q}, s\tau_\beta) &\lesssim \frac{1}{\tau_\beta} C^{(\beta)}_{\rho\theta}(\mathbf{q}, s) \\
C_{\theta\theta}(\mathbf{q}, s\tau_\beta) &\lesssim \frac{1}{\tau_\beta} C^{(\beta)}_{\theta\theta}(\mathbf{q}, s) \\
\Sigma_{\hat{\rho}\hat{\rho}}(\mathbf{q}, s\tau_\beta) &\lesssim \frac{1}{\tau_\beta} \Sigma^{(\beta)}_{\hat{\rho}\hat{\rho}}(\mathbf{q}, s) \\
\Sigma_{\hat{\rho}\hat{\theta}}(\mathbf{q}, s\tau_\beta) &\lesssim \frac{1}{\tau_\beta} \Sigma^{(\beta)}_{\hat{\rho}\hat{\theta}}(\mathbf{q}, s)
\end{aligned}
$$

This scaling is different from that of $C_{\rho\rho}(\mathbf{q}, s\tau_\alpha)$ for $\epsilon \ll 1$; it introduces an extra small quantity $\tau_\beta^{-1}(\epsilon)$ because τ_β diverges at T_d. This introduces a problem of comparison of powers of $m(\epsilon) = \sqrt{\epsilon}$ with powers of $\tau_\beta^{-1} = \epsilon^{1/2a}$. Assuming $\tau_\beta^{-1} \ll \epsilon$ as it is the case if a verifies the equation on MCT exponents (2.23) all the non-$C_{\rho\rho}$ and non-$\Sigma_{\hat{\theta}\hat{\theta}}$ related terms in (6.22) can be neglected and it becomes (6.4):

$$
\frac{W_q - \frac{1}{T}\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q}, 0)}{S_q - z\hat{C}_{\rho\rho}(\mathbf{q}, z/\tau_\beta)} \hat{C}_{\rho\rho}(\mathbf{q}, z/\tau_\beta) = \frac{1}{T}\hat{\Sigma}_{\hat{\theta}\hat{\theta}}(\mathbf{q}, z/\tau_\beta) \tag{6.23}
$$

Extra correlators and self-energies contribute to higher orders. This is indeed the case because of (6.18) which implies $0 < a < 1/2$; therefore $\tau_\beta^{-1} = \epsilon^{1/2a} \sim o(\epsilon)$. Remark however that $\Sigma_{\hat{\theta}\hat{\theta}}$ still has numerical contributions from $C_{\rho\theta}$ and $C_{\theta\theta}$ with no explicit dependence on τ like $\int_0^\infty du C_{\rho\theta}(u)$.

Finally, (6.23) holds only if the following assumptions are satisfied:

- Landau theory: this equation is a self-consistent assumption in the derivation of the Landau theory. It is validated by derivation of the $\sqrt{\epsilon}$ expansion of $\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q}, s\tau_\beta)$ (6.9).
- Equation (6.23) is only valid on the β-scale: on different scales neglecting terms in (6.22) is not justified.

6.3.2 Kernel expansion

Let's write an explicit expression for (6.23) in the β regime:

$$\frac{T_d\tilde{W}_{\mathbf{q}}f_{\mathbf{q}}}{z(1-f_{\mathbf{q}})}+m(\epsilon)T_d\tilde{W}_{\mathbf{q}}\hat{G}_1(\mathbf{q},z)+m^2(\epsilon)T_d\tilde{W}_{\mathbf{q}}(1-f_{\mathbf{q}})z\hat{G}^2(\mathbf{q},z)+$$
$$+m^2(\epsilon)T_d\tilde{W}_{\mathbf{q}}(1-f_{\mathbf{q}})^2\hat{G}_2(\mathbf{q},z)+\epsilon\frac{T_d\tilde{W}_{\mathbf{q}}f_{\mathbf{q}}}{z(1-f_{\mathbf{q}})}=\hat{\Sigma}_{\hat{\theta}\hat{\theta}}(\mathbf{q},z\epsilon^{1/2a}) \quad (6.24)$$

where $\tilde{W}_q = W_q - \Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},0)/T$. The equation on the non-ergodic parameter $f_{\mathbf{q}}$ is given by a zero order in $m(\epsilon)$ and it reproduces equation of $f_{\mathbf{q}}$ that was derived in 6.2.3:

$$\frac{T_d^2 f_{\mathbf{q}}}{1-f_{\mathbf{q}}} = \Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},\infty) \quad (6.25)$$

To close the expansion (6.24) one should compute the expansion $\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},z/\tau_\beta)$ in $m(\epsilon)$. In the context of Brownian dynamic $\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},z/\tau_\beta)$ admits a diagrammatic representation discussed in 4.4.2. The arguments of 6.2.3 give the series (6.9) for $\Sigma_{\hat{\theta}\hat{\theta}}(\mathbf{q},z/\tau_\beta)$ on general grounds. It is instructive to justify this result by a direct derivation from perturbative series. This derivation can be accomplished for Brownian dynamics. Nevertheless it is cumbersome because of the wavevector dependence which obscures the derivation with irrelevant technical details. A schematic model is better adapted for this purpose: we demonstrate the direct computation on the example of the 3-spin model below for this reason.

6.4 Spherical 3-spin model

This section focuses on an explicit derivation of the expansion (6.9) for $\Sigma(\tau)$ within the 3-spin model. The main point is to highlight the subtle technical details in the derivation from perturbation theories. Let's recall briefly the basic results for the model. The evolution of the spin-spin correlator $\hat{C}(z)$ is given by:

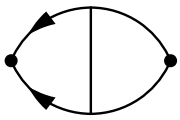
$$\frac{T^2\hat{C}(z)}{1-z\hat{C}(z)} = T + \hat{\Sigma}(z) \quad (6.26)$$

where $\hat{\Sigma}(z)$ is represented by a sum of 2 particle irreducible diagrams. Before we present the derivation of (6.7) for $\Sigma(s\tau_\beta)$ from the diagrammatic expansion.

6.4.1 Perturbative corrections to 1-loop

The lowest 1-loop order approximation for Σ was discussed in 3.3. Corrections from higher orders of perturbation have two characteristic properties:

- $\Sigma(\tau)$ is no more local in time i.e. it is no more a polynomial in $C(\tau)$. A typical $1/N$ diagram reads:



$$= \frac{18}{NT^2} \iint_0^\infty dxdy C'(x)C'(y)C(x-y)C(\tau-x)C(\tau-y)$$

- We discussed in 3.2 that perturbation theory in $1/N$ is not very suitable for analysis because each order in $1/N$ has an infinite number of diagrams and requires a preliminary partial resummation which is out of reach.

The first property breaks the derivation used for the local case and which was used in (3.3). The second issue prevents from using the $1/N$ perturbation theory. Instead we consider an approximation for Σ which consists in retaining the 1-loop diagram and any finite set of higher order diagrams; this way the resummation is avoided.

6.4.2 Time ordering in perturbation theory

A generic diagram of the self-consistent expansion contributing to $\Sigma(\tau)$ reads:

$$\int du_1 \cdots \int du_n R_1(u_1) \cdots R_n(u_n) \times \begin{array}{l}\text{Product of } C \text{ lines depending on linear} \\ \text{combinations of } \{u_k\} \text{ and } \tau\end{array}$$

where R is the response function. *A priori* internal times $\{u_k\}$ are contained in $[0,\infty]$. This can be further restricted to $[0,\tau]$. Furthermore all arguments of correlators inside a diagram can be restricted to $[0,\tau]$. This property of perturbation series which we refer to as time ordering, is a direct consequence of equilibrium. A detailed proof is presented in Appendices $A1$ and $A2$. We will see that neglecting this property of perturbation series may lead to an artificial break down of the Landau theory.

Remark also a particular choice of internal times $\{u_k\}$: they are defined so that any u_k appears once as $du_k R(u_k)$ in a diagram *i.e.* u_k span all the independent internal times in a diagram. This is a direct consequence of the structure of the interaction (3.7): for a given time u there is a single $\hat{\phi}$ line. Therefore for any vertex there is always a single incoming and a single outgoing R-line.

6.4.3 Diagrammatic derivation

In what follows we substitute the generic quantities $m(\epsilon)$, τ_α and τ_β by their MCT values for simplicity. The validity of such substitution is verified self-consistently.

We are interested in an expansion of $\Sigma(\tau = s\epsilon^{1/2a})$ in powers of $\sqrt{\epsilon}$ up to the second order. As we stated above the difficulty of the diagrams is the non-local time dependence that mixes correlators at different times. This leads to a coupling of different scaling regimes - short times, β and α which was absent for

the 1-loop approximation where only local terms were present. This coupling prevents from simple substitution $C(s\tau_\beta + \cdots) \to C(s\tau_\beta)$ because $\cdots$ can also be of order $O(\tau_\beta)$. To account for this extra contributions (as compared to the local term case) it is convenient to split the integration domain of a given internal time u into two disjoint intervals:

$$u \sim O(1) \quad u \sim O(\tau_\beta)$$

which define on what timescale u is. Applying this procedure to all internal times generates an expansion of the original diagram into a finite series:

$$\int_0^{s\tau_\beta} du = \int_0^{\lambda} du + \int_{\lambda}^{s\tau_\beta} du$$

where the α-regime is absent because of the time ordering. Now all the internal times are ordered in the following sense: for a given term every time u belongs to a particular time scale.

This idea is illustrated for a particular diagram in Appendix B; however it is quite cumbersome. For simplicity we illustrate it on a toy expression which is simple enough but still non-trivial:

$$\begin{aligned}\int_0^{s\epsilon^{-1/2a}} du R(u)C(s\epsilon^{-1/2a}-u) &= \int_0^{\lambda} du R(u)C(s\epsilon^{-1/2a}-u) \\ &+ \epsilon^{-1/2a}\int_{\lambda\epsilon^{1/2a}}^{s} du R(u)C((s-u)\epsilon^{-1/2a})\end{aligned}$$

where we introduced the scale λ which delimits the limit of applicability of the short time regime. It should be sent to infinity at the end after the $\epsilon \to 0$ is taken.

Assuming a sharp timescale separation for $\epsilon \ll 1$ substitution of R and C lines by their asymptotic values is justified:

$$\begin{array}{rcc} u,v \sim O(1) & C(u+v) \sim C_0(u+v) & R(u+v) \sim R_0(u+v) \\ u \sim O(\tau_\beta), v \sim O(1) & C(u+v) \sim C_\beta(u) & R(u+v) \sim \epsilon^{1/2a} R_\beta(u) \\ u,v \sim O(\tau_\beta) & C(u+v) \sim C_\beta(u+v) & R(u+v) \sim \epsilon^{1/2a} R_\beta(u+v) \end{array}$$

The the above splitting reads:

$$\begin{aligned}\int_0^{s\epsilon^{-1/2a}} du R(u)C(s\epsilon^{-1/2a}-u) = &\int_0^{\lambda} du R_0(u)C(s\epsilon^{-1/2a}-u) \\ &+\sqrt{\epsilon}(1-q)^2 \int_{\lambda\epsilon^{1/2a}}^{s} du (g'(u)+\sqrt{\epsilon}g_2'(u))C((s-u)\epsilon^{-1/2a})\end{aligned}$$

Note that the prefactor $\epsilon^{1/2a}$ of R_β was absorbed in du due to rescaling of u by $\epsilon^{-1/2a}$ in the second term. From the above expression one derives the following rules:

- Integration over short time gives only a constant contribution. Indeed, due to supposed sharp timescales separation short times decouple from times of order τ_β (including s).
- Integration over β-scale contributes to orders $\sqrt{\epsilon}$ and ϵ:

$$\begin{aligned}\int\limits_{\lambda}^{s\epsilon^{-1/2a}} du\partial_u C(u) &= \int\limits_{\lambda\epsilon^{1/2a}}^{s} du\partial_u C(u\epsilon^{-1/2a}) \\ &= \sqrt{\epsilon}(1-q)^2 \int\limits_{\lambda\epsilon^{1/2a}}^{s} du\left[g'(u)+\sqrt{\epsilon}g_2'(u)+\cdots\right]\end{aligned}$$

The second rule implies that a diagram which contributes to at most the order ϵ has at most two internal times of order τ_β; otherwise it contributes to an order which is higher than ϵ. This remark leads to a case by case analysis: one internal time and two internal times of order τ_β in a diagram. Two times case admits two different realisation on diagrammatic level: either integration domains are independent either one of them contains the other. Indeed all the integration domains are just a subsets of $[0, s\epsilon^{-1/2a}]$ because of the time ordering and they are all of the form $[0, x]$ i.e. they start from zero. Then any two of them are either disjoint (case 1), either one is a subset of the other (case 2).

1. Integration domains when the times are independent:

$$\epsilon(1-q)^4 \int\limits_{\lambda\epsilon^{1/2a}}^{s} du \int\limits_{\lambda\epsilon^{1/2a}}^{s} dv g'(u)g'(v) \times \text{const} =$$
$$\epsilon(1-q)^4 \left(g(s)-g(\lambda\epsilon^{1/2a})\right)^2 \times \text{const}$$

2. One integration domain is a subset of the other:

$$\epsilon(1-q)^4 \int\limits_{\lambda\epsilon^{1/2a}}^{s} du \int\limits_{\lambda\epsilon^{1/2a}}^{u} dv g'(u)g'(v) \times \text{const} =$$
$$\epsilon(1-q)^4 \left[\frac{1}{2}\left(g^2(s)-g^2(\lambda\epsilon^{1/2a})\right) - g(\lambda\epsilon^{1/2a})\left(g(s)-\lambda\epsilon^{1/2a}\right)\right] \times \text{const}$$

Where *const* is a numerical contribution from the rest of the diagram: short times integrations and C-lines depending on times u,v substituted by q.

The one time case also admits two distinct representations; there is one R-line and one C-line. This resembles the toy expression considered above:

- The C-line depends on s:

$$\sqrt{\epsilon}(1-q)^2 \int\limits_{\lambda\epsilon^{1/2a}}^{s} du(g'(u)+\sqrt{\epsilon}g_2'(u))C((s-u)\epsilon^{-1/2a}) =$$
$$\sqrt{\epsilon}q(1-q)^2\left(g(s)-g(\lambda\epsilon^{1/2a})\right)+\epsilon q(1-q)^2\left(g_2(s)-g_2(\lambda\epsilon^{1/2a})\right)+$$
$$+\epsilon(1-q)^4\int\limits_{\lambda\epsilon^{1/2a}}^{s} dug'(u)g(s-u)$$

- The C-line is independent of s

$$\sqrt{\epsilon}(1-q)^2 \int\limits_{\lambda\epsilon^{1/2a}}^{s} du(g'(u)+\sqrt{\epsilon}g_2'(u))C(u\epsilon^{-1/2a}) =$$
$$\sqrt{\epsilon}q(1-q)^2\left(g(s)-g(\lambda\epsilon^{1/2a})\right)+\epsilon q(1-q)^2\left(g_2(s)-g_2(\lambda\epsilon^{1/2a})\right)+$$
$$+\frac{1}{2}\epsilon(1-q)^4\left(g^2(s)-g^2(\lambda\epsilon^{1/2a})\right)$$

Finally if there is no internal times of order $\epsilon^{-1/2a}$ then this is a zero order contribution which fixes the equation on f.

Let's now eliminate the arbitrary scale λ. The final results should not depend on its value: a limit $\lambda\to\infty$ should be taken after $\epsilon\to 0$:

$$\lim_{\lambda\to\infty}\lim_{\epsilon\to 0}\epsilon g(\lambda\epsilon^{1/2a}) \sim \lim_{\lambda\to\infty}\lambda^{-a}=0$$

This means that these terms can be substituted by zero directly. However the order of the limits is important: higher orders produce vanishing corrections for the right sequence and lead to divergences for the inverted order (see Appendix C for more details).

This concludes the proof of (6.9) for the 3-spin spherical model:

$$\Sigma(s\epsilon^{-1/2a}) = \Sigma_0+\sqrt{\epsilon}\Sigma_1 g(s)+\epsilon\Sigma_2 g_2(s)+\epsilon\Sigma_{2,loc}g^2(s)+ \tag{6.27}$$
$$+\epsilon\Sigma_{2,nl}\int\limits_0^s dug'(u)[g(s-u)-g(s)] \tag{6.28}$$

and justifies the Landau theory as well as the assumptions made during the derivation, like the scaling of extra correlators for Brownian dynamics, substitution of generic functions by their Landau theory counterparts, *etc.*

6.5 Summary

Studies on growing correlation length within MCT and related analysis of multi-point functions suggested that MCT equations are only mean-field equations since they missed the correlation length completely. However this was an analogy only; a rigorous proof required analysis of corrections to MCT which in turn, required a systematic perturbation around MCT. Such context was provided by the field theories derived in the previous chapters. In this chapter we presented the proof of the mean-field character of the Mode-Coupling Theory and identified it as the Landau theory for the glass transition. Structural stability analysis also provided an insight on universal properties of MCT which persists against corrections:

- The factorisation property is universal.
- The exponents a and b are not universal, but equation (2.23) fixing the scaling function g is.
- The $\sqrt{\epsilon}$ dependence survives.

The mean-field nature of MCT explains also the discrepancy in the value of T_d as predicted by MCT and by data fits: this is a usual drawback of any Landau theory. However two important issues remained uncovered in this chapter. First, we neglected critical fluctuations, only some initial steps were presented in the analysis of stability with respect to critical fluctuations. Second, we focused on the high temperature phase where the system reaches equilibrium during observation so that equilibrium dynamics can be studied. This is no more true for low temperatures as we discussed in the introduction and as we will see in the next chapter.

A1. Time ordering: generic proof.

In this appendix we present a general proof of time ordering in diagrams of perturbation theory for the 3-spin model. For that let's consider the functional integral which generates the perturbation series: $\int D\phi D\hat{\phi} \exp(-S)$. Here there is a boundary condition on $\phi(t)$ at $t = 0$ (and we denoted by ϕ the whole ensemble of N fields). Let's split the integral into a product of two integrals:

$$\int_{\phi(0)} D\phi D\hat{\phi} \exp(-S) = \int d\phi(s) \int_{\phi(0)}^{\phi(s)} D\phi D\hat{\phi} \exp(-S) \int_{\phi(s)} D\phi D\hat{\phi} \exp(-S) \quad (6.29)$$

In fact the first integral is exactly the time dependent probability distribution $P(\phi, s) = \langle \delta(\phi - \phi(s)) \rangle$ where $\phi(s)$ is the solution of (3.2) and $\langle\rangle$ denotes the average over the thermal and quenched disorder (See [77, 110] for detailed proofs. The only subtlety is the presence of an extra average over the quenched disorder

i.e. the couplings J). This distribution is known to satisfy the Fokker-Planck equation and it tends to the equilibrium distribution $P_{eq}(\phi)$ as $s \to \infty$. Since we are considering the high-T regime the equilibration time t_{eq} is finite meaning that we can replace $P(\phi, s)$ by $P_{eq}(\phi) = \exp(-H/T)$ for $s \gg t_{eq}$.

The second integral in the left part of (6.29) already generates the diagrams with desired properties. Indeed the action reads $S = \int_s^\infty du L(u)$ and all the internal times in the diagrams are limited to $[s, \infty)$. The upper bound for the internal times of the diagrams is induced once we consider the diagrams for the two point functions. Namely if we consider the diagrams for $C(t, s)$ with $t > s$ then this upper bound would be t. This is related to the casual structure of the theory: the R-lines organise a tree structure in the diagram. The only possible position of the root is the time t. The other possibility would be an end point with time s and would have given a zero contribution.

Then the final expression reads:

$$\int d\phi(s) \int\limits_{\phi(s)} D\phi D\hat{\phi} \exp\left[- \int\limits_s^\infty L(u) - \frac{H(\phi(s))}{T} \right]$$

The perturbation series generated by this functional are indeed time ordered in the sense that all the internal times run from s. The upper bound is t for the expansion of $C(t, s)$.

Unfortunately there is one problem in the proof: the time decomposition used in (6.29) is not valid for the action (3.5). The main reason is that this is the field theory averaged over the disorder. This results into the non-local terms in (3.5) which make the above decomposition impossible.

In order to avoid this problem one should start from a field theory for a fixed realisation of a disorder. Then the action reads (3.5):

$$S_{eff}[\phi, i\hat{\phi}] = \sum_{k=1}^{N} \int\limits_0^\infty dt [T\hat{\phi}_k^2 + i\hat{\phi}_k(\partial_t + \mu)\phi_k] - \\ - \sum_{klm} J_{klm} \int\limits_0^\infty dt\, i\hat{\phi}_k \phi_l \phi_m(t) \qquad (6.30)$$

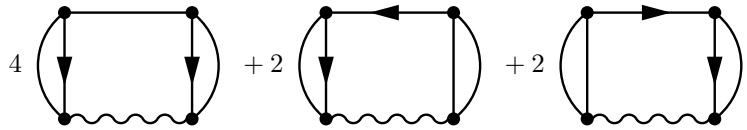
Now that there are no non-local terms the above proof applies and there is a simple *i.e.* non self-consistent, time-ordered perturbation theory.

All one has to do is transform it into a self-consistent disorder averaged one. Already one can carry out the average over the disorder for this perturbation theory: due to the Gaussian nature of the integration over J in (6.30) the resulting expansion coincides with the simple perturbation theory generated by (3.5) with the exception that now it is time ordered *i.e.* all the internal times are limited by s and t.

The next step is to construct the self-consistent expansion. The generic procedure is presented in Appendix A1. The crucial note is that the self-consistent Dyson equation presented below can be solved by iterations: it generates exactly the simple perturbation theory. Thus the self-consistent perturbation theory is in fact a partial resummation of the perturbation series. Reverting this argument and applying it to our case we restore the self-consistent Dyson equation with time ordered diagrams since the integration in the Dyson equation are also limited by s and t.

A2. Time ordering: explicit resummation.

This appendix is dedicated to an explicit demonstration of how a general construction of time ordering presented above works. Precisely one might ask how the time ordered perturbation theory presented above is related to the standard perturbation theory. The answer is quite simple: the new expansion is just a partial resummation of the ordinary perturbation series. The latter transforms into time ordered series under resummation of all the diagrams with fixed topology in the graph theory sense. To highlight this transformation let's consider a class of one loop diagrams of order $1/N$ with a fixed topology (Here undirected links correspond to C-line, directed ones - to R lines and wiggly links - to Q lines. We also omitted the common prefactor):



Here topology is defined as an invariance under exchange of directed and undirected links. Then the position of the wiggly link define the topology.

The final diagrams are generated from the above diagrammatic sum by differentiating with respect to the Q-line *i.e.* by cutting the wiggly links. The result expressed via conventional integrals reads:

$$\frac{1}{T^2}\int_0^t du \int_0^s dv \partial_u C^2(t,u) C(u,v) \partial_v C^2(s,v) +$$
$$+\frac{1}{T}\int_0^t du \int_0^u dv \partial_u C^2(t,u) R(u,v) C^2(s,v) +$$
$$+\frac{1}{T}\int_0^s dv \int_0^v du \partial_v C^2(s,v) R(v,u) C^2(t,u)$$

We now should compute the limit $t, s \to \infty$ with $\tau = t - s$ fixed. A simple but tedious computation shows that this expression boils down in the limit to:

$$\frac{1}{T}\int_s^t du \int_s^u dv \partial_u C^2(t-u) R(u-v) C^2(v-s) +$$

$$+\frac{1}{T^2}\int_s^t du \partial_u C^2(t-u) C(u-s) + \frac{C^2(t-s)}{T^2} \sim$$

$$C^2(\tau) - \int_0^\tau du \partial_u C^2(u) C(\tau-u) + \int_0^\tau du \int_0^{\tau-u} dv \partial_u C^2(u) \partial_v C(v) C^2(\tau-u-v)$$

B. Diagrammatic proof: example.

Using the results of the previous appendices we present below a very detailed computation of the $\sqrt{\epsilon}$-expansion of the self-energy Σ on the example of the diagram presented in Eq. (6.27).

First we rewrite the above expression integrating by parts the second term:

$$C(\tau) - \int_0^\tau du \partial_u C(u) C^2(\tau-u) + \int_0^\tau du \int_0^{\tau-u} dv \partial_u C^2(u) \partial_v C(v) C^2(\tau-u-v)$$

Let's proceed to a term by term analysis. The very first term is local and it is treated in exactly the same way as in Section 3.3. The second term is non-local; following the guidelines of 6.4.3 it is split into integrals over the timescales and reads:

$$\int_0^{s\epsilon^{-1/2a}} duC'(u)C^2(s\epsilon^{-1/2a}-u) = \int_0^{\lambda} duC_0'(u)\Big[f^2+2\sqrt{\epsilon}f(1-f)^2g(s-u\epsilon^{1/2a})$$
$$+\epsilon(1-f)^4g^2(s-u\epsilon^{1/2a})+2\epsilon f(1-f)^2g_2(s-u\epsilon^{1/2a})\Big]$$
$$+\sqrt{\epsilon}(1-f)^2\int_{\lambda\epsilon^{1/2a}}^{s} du\left(g'(u)+\sqrt{\epsilon}g_2'(u)\right)\left[f^2+2\sqrt{\epsilon}f(1-f)^2g(s-u)+\cdots\right]$$
$$=\left[f^2+2\sqrt{\epsilon}f(1-f)^2g(s)+\epsilon(1-f)^4g^2(s)+2\epsilon f(1-f)^2g_2(s)\right]\int_0^{\lambda} duC_0'(u)$$
$$+\sqrt{\epsilon}(1-f)^2\int_{\lambda\epsilon^{1/2a}}^{s} du\left(g'(u)+\sqrt{\epsilon}g_2'(u)\right)\left[f^2+2\sqrt{\epsilon}f(1-f)^2g(s-u)+\cdots\right]\to$$
$$\to\left[f^2+2\sqrt{\epsilon}f(1-f)^2g(s)+\epsilon(1-f)^4g^2(s)+2\epsilon f(1-f)^2g_2(s)\right](1-f)$$
$$+\sqrt{\epsilon}f(1-f)^2(g(s)+\sqrt{\epsilon}g_2(s))$$
$$+2\epsilon f(1-f)^4\int_0^{s} g'(u)[g(s-u)-g(s)]+2\epsilon f(1-f)^4g^2(s)$$

The third term is the most complicated because of the double integration:

$$\int_0^{s\epsilon^{-1/2a}} du\int_0^{s\epsilon^{-1/2a}-u} dvC^{2\prime}(u)C'(v)C^2(s\epsilon^{-1/2a}-u-v)=$$
$$\left(\int_0^{\lambda}du\int_0^{\lambda}dv+\int_0^{\lambda}du\int_{\lambda}^{s\epsilon^{-1/2a}}dv+\int_{\lambda}^{s\epsilon^{-1/2a}}du\int_0^{\lambda}dv+\int_{\lambda}^{s\epsilon^{-1/2a}}du\int_{\lambda}^{s\epsilon^{-1/2a}}dv\right)\cdots$$

There are 4 different cases possible:

1.

$$\int\limits_0^\lambda du \int\limits_0^\lambda dv C_0^{2\prime}(u) C_0'(v) C^2(s\epsilon^{-1/2a} - u - v) =$$

$$\int\limits_0^\lambda du \int\limits_0^\lambda dv C_0^{2\prime}(u) C_0'(v) \Big[f^2 + 2\sqrt{\epsilon} f g(s - (u+v)\epsilon^{1/2a}) +$$

$$+ 2\epsilon f g_2(s - (u+v)\epsilon^{1/2a}) + \epsilon(1-f)^4 g^2(s - (u+v)\epsilon^{1/2a}) \Big] \to$$

$$\to (1-f)(1-f^2) \Big[f^2 + 2\sqrt{\epsilon} f (1-f)^2 g(s)$$

$$+ 2\epsilon f (1-f)^2 g_2(s) + \epsilon (1-f)^4 g^2(s) \Big]$$

2.

$$\int\limits_0^\lambda du \int\limits_\lambda^{s\epsilon^{-1/2a}} dv C^{2\prime}(u) C'(v) C^2(s\epsilon^{-1/2a} - u - v) =$$

$$\sqrt{\epsilon}(1-f)^2 \int\limits_0^\lambda du \int\limits_{\lambda\epsilon^{1/2a}}^s dv C_0^{2\prime}(u) \left[g'(v) + \sqrt{\epsilon} g_2'(v) \right] \times$$

$$\Big[f^2 + 2\sqrt{\epsilon} f (1-f)^2 g(s - v - u\epsilon^{1/2a}) \Big] \to$$

$$\to \sqrt{\epsilon} f^2 (1-f)^3 (1+f) \left(g(s) + \sqrt{\epsilon} g_2(s) \right) +$$

$$+ 2\epsilon f (1-f)^5 (1+f) \left[g^2(s) + \int\limits_0^s dv g'(v) (g(s-v) - g(s)) \right]$$

3. Due to symmetry $u \equiv v$ this case reduces to the previous one.

$$\int\limits_\lambda^{s\epsilon^{-1/2a}} du \int\limits_0^\lambda dv C^{2\prime}(u) C'(v) C^2(s\epsilon^{-1/2a} - u - v)$$

4.

$$\int\limits_\lambda^{s\epsilon^{-1/2a}} du \int\limits_\lambda^{s\epsilon^{-1/2a}} dv C^{2\prime}(u) C'(v) C^2(s\epsilon^{-1/2a} - u - v) =$$

$$\epsilon f^2 \int\limits_{\lambda\epsilon^{1/2a}}^s \int\limits_{\lambda\epsilon^{1/2a}}^s du dv g'(u) g'(v) \to \epsilon f^2 g^2(s)$$

It is worth noting that in this particular case analysis via the Laplace transform is easier due to the convolutive structure of the expression. Namely under Laplace transform the second and the third terms of the time ordered diagram read:

$$L\left[\int_0^\tau du\partial_u C(u)C^2(\tau-u)\right] = (z\hat{C}(z)-1)L\left[C^2\right]$$

$$L\left[\int_0^\tau du\int_0^{\tau-u} dv\partial_u C^2(u)\partial_v C(v)C^2(\tau-u-v)\right] = \hat{C}(z)\left(z\hat{C}(z)-1\right)\left(zL[C^2]-1\right)$$

Apparently there is only local terms and the analysis reduces to the local case considered in 3.3. Note however that this is a peculiarity of the diagram; this does not extend to a generic diagram.

C. Divergences.

The limit taken in the previous appendices - $\lambda \to \infty$ has some subtle points we would like to highlight in this appendix. Namely we present a part of the computation with more focus on the limit $\lambda \to \infty$. For the sake of simplicity we consider the case of the second term (more precisely only the λ dependent part)

$$\sqrt{\epsilon}(1-q)^2\int_{\lambda\epsilon^{1/2a}}^{s} du\left(g'(u)+\sqrt{\epsilon}(1-q)^2 g_2'(u)\right)\left[q^2+2\sqrt{\epsilon}q(1-q)^2 g(s-u)+\cdots\right] =$$
$$\sqrt{\epsilon}(1-q)^2q^2\left(g(s)-g(\lambda\epsilon^{1/2a})\right)+\epsilon q^2(1-q)^4\left(g_2(s)-g_2(\lambda\epsilon^{1/2a})\right)+\cdots$$

The scaling functions $g(s)$ and $g_2(s)$ have singular asymptotics as $s \to 0$: they behave as s^{-a} and s^{-2a} respectively. This reflects the fact that for very short times the scaling function description fails and we get into short times regime. The the terms $g(\lambda\epsilon^{1/2a})$ and $g_2(\lambda\epsilon^{1/2a})$ are potentially divergent. However a detailed inspection shows that this is not the case. Indeed:

$$\lim_{\lambda\to\infty}\lim_{\epsilon\to 0}\sqrt{\epsilon}g(\lambda\epsilon^{1/2a}) \sim \lim_{\lambda\to\infty}\lim_{\epsilon\to 0}\sqrt{\epsilon}(\lambda\epsilon^{1/2a})^{-a} \sim \lim_{\lambda\to\infty}\lambda^{-a}=0$$

That is, the possible divergence in ϵ is cancelled by a $\sqrt{\epsilon}$ prefactor. The origin of this cancellation is again in the breakdown of the description. For $s \to 0$ (more precisely for $s \sim o(\epsilon^{1/2a})$) the difference $C(s\epsilon^{-1/2a}) - q$ is no more of order $\sqrt{\epsilon}$ but rather finite.

Let's discuss the possible inversion of the limits $\lambda \to \infty$ and $\epsilon \to 0$. In that case λ is sent to zero rather than to infinity because it delimits short times

from β regime; the limit of $\sqrt{\epsilon}g(\lambda\epsilon^{1/2a})$ becomes divergent. However one can prove that this divergence is cancelled by divergences produced by short time contributions.

Chapter 7

Glassy phases

Throughout the previous Chapters we assumed time that translation invariance and time reversal symmetry hold. This is true for high temperatures where observation time can be always made greater than equilibration time so that system equilibrates after a transient regime and equilibrium dynamics can be observed. For very high temperatures the transient regime is so short that it is almost impossible to observe. However as we stated in the introduction the relaxation time grows tremendously for supercooled liquids as the temperature is lowered. It is worth recalling that this growth happens for mild variations of the temperature. The relaxation time becomes exceedingly long for low temperatures so that equilibration is avoided on macroscopic timescales; the equilibration time can reach geological values. Therefore time-reversal symmetry is broken and there is no more time translation invariance. In what follows we refer to such states as *glassy phases*.

The absence of equilibrium changes the dynamic properties. Glassy phases are characterised by an extremely slow relaxation processes and history dependent phenomena. One example is ageing: dynamic properties depend explicitly on the system's age; the more the system is aged, the slower is the relaxation. Non-equilibrium dynamics has been widely studied for systems with quenched disorder *i.e.* spin glasses [88, 111, 31] which we already mentioned in Chapter 3. The studies revealed some degree of universality in low-T dynamics and a particular scaling form for ageing instead of the time difference which is a standard for equilibrium.

Within the Mode-Coupling Theory scenario of the glass transition the fall out of equilibrium at low temperatures is approached by a dynamic transition to a non-ergodic phase; the equilibration time is finite above the transition, diverges at the transition and remains infinite below. That is, a supercooled liquid never equilibrates below the transition. However the derivation presented in 2.2 is not valid in the low-T phase since it relies on the assumption of equilibrium. An extension of MCT to low temperature within the projector operator formalism has been derived [29] and can be used for analysis. However field theories developed previously in Chapter 4 or Sec. 5.1 provide a more convenient method

to extend MCT to low temperatures. The dynamical equations derived in this context are valid for any temperature unless one uses FDT to simplify them. Therefore equations presented in Sec. 4.4.3 should be rewritten only in their general two time form if one is interested in low temperature regime.

Analysis of the full wave-vector dependent case has not yet been done because of the involved derivation as we will see, and is a subject of future work. Below we consider a simpler schematic case with no wavevector dependence. Schematic MCT maps on the spherical p-spin model which has a field theory representation valid at any temperature so that the general dynamic equations (3.9) describe dynamics at any temperature. It is a reliable assumption that results derived within the 3-spin model hold also for a low-T extension of schematic MCT.

The main aim of this chapter is twofold. First, we want to extend the Landau theory to low temperatures. As we will see this problem requires to define the analogy of the scaling function g (see Eq. (2.22)) and is quite involved computationally mainly due to the presence of many timescales in the system. For this reason we limit our analysis to the lowest order *i.e.* $N \to \infty$ in the context of the 3-spin model. Second, construction of the Landau theory for low temperatures is tightly connected with the study of ageing in disordered systems.

In the next section we present the phenomenology of glassy phases and highlight the problems we will consider in detail in the following sections.

7.1 Phenomenology

Let's introduce the phenomenology used to analyse glassy phases and mentioned in the introduction. As we have pointed out we focus on ageing. For glassy phases the characteristics of dynamics evolve in time: system ages. This is easily seen in experiments measuring time evolution. Typical example is the measurements of a.c. magnetic susceptibility χ at a certain frequency and 'Thermo-Remanent Magnetisation' (TRM) $m(t, t_w)$ for magnetic materials when a sample is quenched at $t = 0$ from a high temperature phase to low value of T subject to an external magnetic field during cooling and from $t = 0$ up to $t = t_w$. Experimental data clearly indicate that in both cases relaxation of the observed quantities depends explicitly on the waiting time t_w besides the frequency or time t. The general trend is that the longer t_w, the slower the relaxation. Analysis of experimental data shows that quantities can be split into a stationary part independent of t_w and an ageing component which depends on t_w:

$$\chi(\omega, t_w) = \chi_{st}(\omega) + \chi_{ag}(\omega, t_w)$$
$$m(t, t_w) = m_{st}(t - t_w) + m_{ag}(t, t_w)$$

The time dependence of the ageing parts is not arbitrary and is to some degree universal. Experimental data collapse on a master curve for a particular scaling

$m_{ag}(t,t_w) = \tilde{m}_{ag}(h(t_w)/h(t))$ where $h(t)$ is referred to as an *ageing function*. It is an increasing function of its argument. Several particular choices of h have their own references: $h(t) = t$ is referred to as simple or full ageing, $h(t) = exp(t^{1-\mu})$ - as sub-ageing.

Ageing effects are also observed for correlation functions. A widely studied example is a correlation function $C(t,s)$, $t > s$. Like one-point quantities it splits into stationary and ageing parts:

$$C(t,s) = C_{st}(t-s) + C_{ag}(t,s)$$

in glassy phases. In equilibrium correlation is related to response by the Fluctuation-Dissipation Theorem, but this is no longer true out of equilibrium: in general correlation and response provide different information. Nevertheless one can still relate them by a generalised relation [31]:

$$R(t,s) = \frac{\theta(t-s)X(t,s)}{T}\partial_s C(t,s) \tag{7.1}$$

where $X(t,s)$ is the so called *violation factor*.

The decomposition of quantities into several parts reflects the appearance of many regimes in the dynamics referred to as *time sectors* [88, 31], like stationary and ageing in the above examples. This is directly related to the existence of many timescales in the problem. Let's remind that on top of the scale imposed by t_w there are, in general, at least two more timescales present in the system: the microscopic time t_0 that determines the scale of the microscopic events and the equilibration time τ_{REL} providing the timescale on which the system equilibrates completely. The latter is always finite for finite systems although it can reach geological values. One can imagine that different physical mechanisms act on these timescales and that there is a hierarchy of timescales: $t_0 < t_1 < t_2 < \cdots < \tau_{REL}$. A certain degree of universality like independence of microscopic details emerges for times $t_0 \ll t, t_w \ll \tau_{REL}$. The hierarchy of timescales implies a complex decomposition of observables like a correlation function $C(t,s)$ with different scaling forms for different sectors [88]:

$$C(t,s) = C_{st}(t-s) + \sum_n \mathcal{C}_n\left(\frac{h_n(s)}{h_n(t)}\right) \tag{7.2}$$

$$t - s \sim O(1), \quad \frac{h_1(s)}{h_1(t)} \sim O(1), \quad \frac{h_2(s)}{h_2(t)} \sim O(1), \quad \text{etc} \tag{7.3}$$

and the functions h_i are monotonically increasing and satisfy the constraints:

$$0 < \frac{h_i(s)}{h_i(t)} < 1 \quad \text{and} \quad \frac{h_j(s)}{h_i(t)} = 1 \quad \text{for } j < i \tag{7.4}$$

If $\tau_{REL} = \infty$, that is the system never equilibrates, then various possible asymptotic regimes exist as $t, t_w \to \infty$ depending on the way t and t_w are taken to infinity. Note that for $\tau_{REL} = \infty$ there can be systems with infinitely many

timescales. A particlar example is provided by the Sherrington-Kirkpatrick model [112]. Glassy phases often have very large relaxation times so that $\tau_{REL} = \infty$ is a reliable assumption.

This decomposition was proved to hold for mean-field spin glasses [31] where $\tau_{REL} = \infty$ for low temperatures. However only two realisations of the above hierarchies are known for mean-field models: two scales, $t_0 < t_1$, and infinitely many scales. Consequently there are only two time sectors in the former case. Systems with infinitely many timescales also show two sectors in the dynamics but the ageing one is more complicated than for two scale case. This is connected to the type of dynamic transition that the system exhibits at low temperatures: discontinuous or continuous respectively. The difference is the behaviour of the order parameter at the transition. In the first case the order parameter jumps to a non-zero value at the transition like in the 3-spin model (which provides an example of discontinuous transition). In the second case the order parameter increases from zero at the transition like in the Sherrington-Kirkpatrick model. No models with a finite number (but greater than 2) of sectors are known so far[1]. One might wonder whether this classification exhausts all possibilities.

More details about non-equilibrium behaviour have been found analytically for mean-field models due to their simplicity compared to realistic systems. Namely it was shown that the violation factor $X(t,s)$ depends on time only through the correlator C: $X(t,s) = X[C(t,s)]$ in the limit of long times t, s [30] *i.e.* $t, s \gg t_0$. Furthermore X is constant within a given sector [31]. This allows one to introduce an effective temperature T_* within a sector through the generalised FDT given by Eq. (7.1):

$$R(t,s) = \frac{\theta(t-s)}{T/X_\infty}\partial_s C(t,s)$$

The system "sees" a different temperature from that of a thermal bath within a sector.

These results are based on the analysis of dynamic equations of various mean-field models like the 3-spin model. Their solutions support the multi timescale scenario and fix the functions $\mathcal{C}_n$ in the decomposition. However the ageing functions $h_n(t)$ are left unfixed. More precisely their exact functional form drops out in the derivation of the equations for $\mathcal{C}_n$ in the limit of long times $t \to \infty$. The final equations are invariant with respect to the choice of h, what is referred to as *time reparametrisation invariance* [88, 30]. However this is not true since experimental and numerical data collapse on a single curve for a particular choice of h [111]. This is also in contradiction with causality of the full dynamic equations which admit a unique solution for fixed initial conditions. A generic recipe for fixing h has remained an open problem for a long time. This invariance is an artifact of the derivation and of the long time limit $t \to \infty$. This invites one to reanalyse the derivation and inspect the finite-t corrections to the scaling equations.

[1]Right at the moment of editing of this thesis a class of models presenting 2RSB phase was identified by Crisanti and Leuzzi [113]

At this point the problem of the spurious symmetry crosses with the problem of the low-T extension of Landau theory. Indeed, as we will see the analysis of finite-t corrections leads to the introduction of a scaling function which is exactly the generalisation of the scaling function g from the high temperature regime. In this context we will analyse the spherical p-spin model which is an obvious generalisation of the 3-spin model introduced earlier in Chapter 3.1. As we stated earlier we only consider the limit $N \to \infty$ where the dynamical equations take a particularly simple form; extension to a general case is more involved technically. The starting point of our analysis is the Schwinger-Dyson equations (3.9) generalised to an arbitrary value of p.

7.2 Low-T phase of the spherical p-spin model

The dynamics of the p-spin model is ruled by the Dyson equations (3.9) with minor modifications. The spherical p-spin model is defined similarly to the 3-spin model:

$$H = -\frac{1}{p!}\sum_{k_1\cdots k_p} J_{k_1\cdots k_p}\phi_{k_1}\cdots\phi_{k_p}\,, \qquad \overline{J^2} = \frac{p!}{2N^{p-1}}.$$

Other definitions reflect those for the 3-spin model and lead to the following Schwinger-Dyson equations:

$$\begin{aligned}
(\partial_t + z(t))C(t,s) &= \int_0^t du D(t,u)C(u,s) + \int_0^s du\Sigma(t,u)R(s,u)\\
(\partial_t + z(t))R(t,s) &= \int_s^t du D(t,u)R(u,s)\\
z(t) &= T + \frac{p^2}{2}\int_0^t du C^{p-1}(t,u)R(t,u)\\
D(t,s) &= \frac{p}{2}C^{p-1}(t,s) \qquad \Sigma(t,s) = \frac{p(p-1)}{2}C^{p-2}(t,s)
\end{aligned}$$

They are valid for any value of the temperature T. The model exhibits a dynamical transition at temperature T_d. The transition is of the continuous type for $p = 2$ and of discontinuous type for $p > 2$ [31]. In what follows we only consider $p > 2$. The transition is signalised by the appearance of a non-zero limit of the spin-spin correlator $C(t,s)$:

$$f = \lim_{t-s\to\infty}\lim_{t,s\to\infty} C(t,s)$$

which is an order parameter.

We keep the generic two time dependence in the equations since we expect the time-reversal symmetry to be broken for $T < T_d$. For convenience we rewrite (3.9) in terms of the correlator $C(t,s)$ and the integrated response $F(t,s) = \int_s^t du R(t,u)$:

$$\begin{aligned}(\partial_t + z(t))F(t,s) &= 1 - \int_s^t du D(t,u)F(u,s) \qquad (7.5)\\ (\partial_t + z(t))C(t,s) &= -\int_0^t D(t,u)C(u,s) - \int_0^s du\Sigma(t,u)\partial_u F(s,u)\end{aligned}$$

and $\Sigma(t,s) = \frac{p}{2}C^{p-1}(t,s)$, $D(t,s) = \frac{p(p-1)}{2}C^{p-2}(t,s)\partial_s F(t,s)$ in the limit $N \to \infty$. The Lagrange multiplier $z(t)$ reads:

$$z(t) = T - \frac{p^2}{2}\int_0^t du C^{p-1}(t,u)\partial_u F(t,u).$$

This system cannot be solved analytically (except for $p = 2$) and numerical solutions are non trivial to obtain. The source of complexity is the two time dependence (instead of the one time like in equilibrium) that reflects the richer dynamics contained in these equations. Let's now analyse the different regimes of these equations.

7.3 Time sectors.

The spherical p-spin is a mean-field model; below the transition the relaxation time is infinite $\tau_{REL} = \infty$. Therefore the different regimes of Eqs. (7.5) are probed by various limits $t, s \to \infty$. As we have stated one can enumerate exactly all asymptotic regimes for $T < T_d$. There are exactly 2 time sectors for the p-spin model as $t, s \to \infty$ $(t > s)$ [31]:

- $t, s \to \infty$ and $\tau = t - s$ fixed: stationary regime which resembles equilibrium; time-reversal symmetry is unbroken. Correlator and integrated response are related by FDT: $C(t,s) = C_{ST}(\tau)$ and $F(t,s) = F_{ST}(\tau) = (1 - C_{ST}(\tau))/T$.
- $t, s \to \infty$ and $\lambda = h(s)/h(t)$ fixed: ageing regime. Time-reversal symmetry is broken and FDT is substituted by Eq. (7.1). Correlator and integrated response read:

$$\begin{aligned}C(t,s) &= \mathcal{C}(\lambda)\\ F(t,s) = \mathcal{F}(\lambda) &= \frac{1-f}{T} + \frac{f - \mathcal{C}(\lambda)}{T_*}\\ \frac{1}{T_*} &= (p-2)\frac{1-f}{fT}\end{aligned}$$

where T_* is an effective temperature.

Assuming a sharp timescale separation one can derive dynamic equations for the correlator and the integrated response within each sector from the full equations. Due to FDT in the stationary regime and generalised FDT with effective temperature T_* in the ageing regime the two equations (7.5) reduce to a single equation:

- Lagrange multiplier $z(\infty)$:

$$z(\infty) = T + \frac{p}{2T} - \frac{p}{2T} f^p + \frac{p}{2T_*} f^p \tag{7.6}$$

- Stationary: the equation is supplemented by boundary conditions $C_{ST}(0) = 1$ and $C_{ST}(\infty) = f$

$$\begin{aligned}(\partial_\tau + T) C_{ST}(\tau) = \frac{p f^p}{2} \left[\frac{1}{T_*} - \frac{1}{T}\right] (1 - C_{ST}(\tau)) \\ - \frac{p}{2T} \int_0^\tau du C_{ST}^{p-1}(\tau - u) \partial_u C_{ST}(u)\end{aligned} \tag{7.7}$$

- Ageing: the boundary conditions are $\mathcal{C}(0) = 0$ and $\mathcal{C}(1) = f$

$$\begin{aligned}T\mathcal{C}(\lambda) = \frac{p f^{p-1}}{2}(1-f) \left[\frac{1}{T_*} - \frac{1}{T}\right] \mathcal{C}(\lambda) + \frac{p(1-f)}{2T} \mathcal{C}^{p-1}(\lambda) \\ - \frac{p}{2T_*} \int_\lambda^1 dy \mathcal{C}^{p-1}(y) \partial_x \mathcal{C}\left(\frac{\lambda}{y}\right)\end{aligned} \tag{7.8}$$

  This equation can be transformed into a form similar to that of Eq. (7.7) if one makes a change of variables $x = -\log \lambda$:

$$\begin{aligned}T\mathcal{C}(x) = \frac{p f^{p-1}(1-f)}{2} \left[\frac{1}{T_*} - \frac{1}{T}\right] \mathcal{C}(x) + \frac{p(1-f)}{2T} \mathcal{C}^{p-1}(x) \\ - \frac{p}{2T_*} \int_0^x dy \mathcal{C}^{p-1}(y) \partial_y \mathcal{C}(x - y)\end{aligned} \tag{7.9}$$

The Edward-Anderson parameter f is fixed by both equations: either taking the limit $\tau \to \infty$ of Eq. (7.7), or taking the limit $\lambda \to 1$ of Eq. (7.8). Both limits yield the same equation:

$$\frac{T^2 f^2}{(1-f)^2} = \frac{p(p-1)}{2} f^p \tag{7.10}$$

The sectors have the following physical interpretation: the system is in equilibrium for short times but falls out of equilibrium on longer times. Equation (7.7)

resembles closely (3.11) for equilibrium dynamics and has similar properties but $C_{ST}(\tau)$ decays from the initial value to f. For very long times $\tau \gg 1$ this decay is described by a power law with exponent a:

$$C_{ST}(\tau \to \infty) \sim f + A_1\tau^{-a} + A_2\tau^{-2a} + \cdots$$

The behaviour of $\mathcal{C}(\lambda)$ for $\lambda \approx 1$ is described by a power law with exponent b:

$$\mathcal{C}(1-\epsilon) \sim f - B_1\epsilon^b - B_2\epsilon^{2b} + \cdots$$

In fact Eq. (7.8) has an exact solution:

$$\mathcal{C}(\lambda) = q\lambda^\nu \tag{7.11}$$

where exponent ν is left unfixed by the solution. This solution gives immediately $b = 1$ independently of the temperature.

These definitions together with Eqs. (7.7),(7.8) fix the values of the exponents a and b:

$$\frac{1}{T}\frac{\Gamma^2(1-a)}{\Gamma(1-2a)} = \frac{1}{T_*}\frac{\Gamma^2(1+b)}{\Gamma(1+2b)} = \frac{1}{2T_*}$$

where we used the fact that $b = 1$. This equation like the definitions for a,b resembles highly Eq. (2.23) which holds in the high-T regime. This motivates to look for a scaling function that generalises the scaling function g of a high-T phase and inspect a crossover regime that interpolates between stationary and ageing regimes acting as a generalisation of the β-regime of the equilibrium phase.

Another reason to identify the crossover regime is the problem of the computation of the ageing function $h(t)$. Clearly (7.8) is invariant under reparametrisation $t \to h(t)$ as was anticipated from the discussion of the generic scenario. The only non-reparametrisation invariant term in (7.5) namely the derivative term dropped out in the ageing limit $t, s \to \infty$:

$$\partial_t C(t,s) \to \partial_t \mathcal{C}\left(\frac{h(s)}{h(t)}\right) = -\frac{h'(t)}{h(t)}\frac{h(s)}{h(t)}\mathcal{C}\left(\frac{h(s)}{h(t)}\right) \to 0$$

Apparently an analysis of the regime that interpolates between the stationary and the ageing regimes for small and large arguments correspondingly should fix $h(t)$ since there is a unique solution in the stationary phase on one hand and a multiplicity of solutions connected by time reparametrisations on the other hand.

7.4 Crossover regime.

To identify a crossover regime we add an extra scale to the above picture. Namely, we assume the existence of an exponent β that governs the position of the plateau q with t (or equivalently s): $C(t, t - t^\beta) \sim q$. Time differences $t - s$ of order t^β interpolate between stationary and ageing regimes and thus, $\beta < 1$.

The thickness of the plateau with t or s is described by another exponent α. The existence of this regime is confirmed numerically[114]. These definitions generate expansions of the correlator and the integrated response:

$$\begin{aligned} C(t,s) &= f + t^{-\alpha} g_1\left(\frac{t-s}{t^\beta}\right) + t^{-2\alpha} g_2\left(\frac{t-s}{t^\beta}\right) + \cdots \\ F(t,s) &= \frac{1-f}{T} + t^{-\alpha} w_1\left(\frac{t-s}{t^\beta}\right) + t^{-2\alpha} w_2\left(\frac{t-s}{t^\beta}\right) + \cdots \\ R(t,s) &= t^{-\gamma} r_1\left(\frac{t-s}{t^\beta}\right) + t^{-\gamma-\alpha} r_2\left(\frac{t-s}{t^\beta}\right) + \cdots \end{aligned} \tag{7.12}$$

where $\gamma = \alpha + \beta$ is a consequence of (7.1); $r_l(x) = \partial_x w_l(x)$. Note the formal similarity of the expansions to those of the high-T phase in Sec. 6.4. One also introduces a "temperature" $T(x)$ that interpolates between T at $x = 0$ and T_* at $x = \infty$ and generalises the temperature in the modified FDT (7.1) connecting $w_l(x)$ and $g_l(x)$:

$$g_l'(x) = -T(x) w_l'(x).$$

The finiteness of $T(x)$ explains the same value of exponent α for C and F. The scaling regime interpolates between the stationary and the ageing regime for small and large arguments respectively. This gives a relation between the different exponents and scaling laws via matching with other regimes. We start from matching with the stationary regime: consider $\tau = t - s \sim x t^\beta$ while $t \to \infty$ followed by $x \ll 1$ (the order is important here and plays a similar role to suppress diverging terms as in Sec. 6.4). Using the power law series expansion of the late stationary regime yields:

$$\begin{aligned} t^{-l\alpha} g_l(x) &\sim_{x\to 0} A_l x^{-a} t^{l\beta a} \\ t^{-l\alpha} w_l(x) &\sim_{x\to 0} -\frac{A_l}{T} x^{-a} t^{l\beta a} \end{aligned}$$

so that

$$\begin{aligned} g_l(x \to 0) &\sim A_l x^{-a} \\ w_l(x) &\sim -\frac{A_l}{T} x^{-a} \end{aligned} \tag{7.13}$$

and $\alpha = \beta a$. Also quite naturally for small arguments w_l and g_l are related by a "real" FDT with temperature T. this analysis is quite similar to that of the high temperature phase [106].

The matching with the ageing regime is more subtle: let's take $\tau = x t^\beta$ but this time $x \gg 1$. On one hand the early ageing regime expansion reads:

$$C(t,s) = \mathcal{C}\left(\frac{h(t - x t^\beta)}{h(t)}\right) = f - B_1 (x t^\beta \phi(t))^b + \cdots$$

with $\phi(t) = \frac{d \log h(t)}{dt}$. Comparing with the expansion (7.12) for $C(t,s)$ yields:

$$B_l x^{bl} t^{l\beta b} \phi^b(t) \sim t^{-\alpha} g_l(x) \tag{7.14}$$

This defines an asymptotics for g_l, w_l and fixes $\phi(t)$:

$$g_l(x) \sim -B_l (Dx)^{bl}$$

$$w_l(x) \sim \frac{B_l}{T_*} (Dx)^{bl} \tag{7.15}$$

$$\phi(t) \sim \frac{t^{-\mu}}{D} \qquad \mu = \beta + \frac{\alpha}{b} \tag{7.16}$$

$$\mu = \beta + \frac{\alpha}{b} \tag{7.17}$$

The last identity implies

$$h(t) = \exp\left[\frac{t^{1-\mu}}{D(1-\mu)}\right] \tag{7.18}$$

if $\phi(t) - t^{-\mu} \sim o(1/t)$. Corrections to $t^{-\mu}$ are provided by a subleading singular terms in the expansion of $g_l, l > 1$. Indeed for a given $l > 1$ the above matching establishes only a leading term in the expansion of $g_l(x)$ around $x = \infty$. Since the leading term is of order x^{bl} one expects that $w_l(x)$ decomposes in this limit into singular and regular parts:

$$g_l(x \to \infty) = g_l^{\infty}(x^b) + g_l^{reg}(x)$$

where $g_l^{\infty}(x)$ is a polynomial of degree l in x and $g_l^{reg}(x \to \infty) \to 0$. A similar decomposition holds for w_l. Then Eqs. (7.14), (7.16) should be refined to include these corrections; the result reads:

$$\phi(t) \sim D t^{-\mu} \left(1 + \frac{1}{D^{lb}} \sum_{k>1} t^{-k\alpha} g_{l+k,l}^{\infty}\right)^{1/lb}$$

Then the first correction to the leading term is $\phi(t) - t^{-\mu} \sim O(t^{-\mu-\alpha})$. Because $h(t)$ is an increasing function of t the only relevant terms in this expansion are the terms which have divergent integrals in t as $t \to \infty$. That is, all the terms with exponents greater than 1 (in absolute value) can be neglected: they give a finite contribution after integration. Therefore only a finite number of terms really contributes because $\mu + k\alpha$ becomes greater than 1 for some k. This also imposes some constraints on possibles values of $g_{l,k}^{\infty}$ since the resulting function $h(t)$ should be independent of l. The the general form of h reads:

$$h(t) = \exp\left(\frac{t^{1-\mu}}{D(1-\mu)} P(t^{-\alpha})\right) \tag{7.19}$$

where $P(x)$ is a polynomial of x and $P(0) \neq 0$. There is a numerical evidence in case of p-spin model that $\mu + \alpha > 1$ so that Eq. (7.18) is correct. The

increasing character of h imposes $0 < \mu < 1$ because infinity should be mapped onto infinity. The constant D is set to unity by transformation $D \to D_1$ which is equivalent to transformation $\mathcal{C}(\lambda) \to \mathcal{C}\left(\lambda^{D/D_1}\right)$ *i.e.* the invariance with respect to exponent ν in the exact solution in the ageing regime (see Eq. (7.11)). This freedom to choose ν (or D) reflects the fact that the microscopic time scale t_0 is forgotten in the ageing regime. Indeed, $t^{1-\mu}/D$ should be dimensionless and hence D encodes some timescale. The only possibility is the microscopic timescale $t_0 \sim D^{1/1-\mu}$. This provides further support to the absence of relevant corrections to (7.18). Indeed $\log h(t) = t^{1-\mu} + D_2 t^{\delta}$ with $\delta > 0$ would fix the microscopic time scale t_0 through $D_2 \sim t_0^{-\delta}$. The independence of $C(t,s)$ from the microscopic timescale t_0 in the ageing regime can be used to fix directly the form of $h(t)$. Assuming $\mathcal{C}(\lambda) = q\lambda^{\nu(t_0)}$, $h = \exp\phi(t/t_0)$ and $\partial_{t_0} C(t,s) = 0$ in the ageing regime yields:

$$\partial_{t_0}\left[\exp\nu(t_0)\left(\phi\left(\frac{s}{t_0}\right) - \phi\left(\frac{t}{t_0}\right)\right)\right] = 0$$

$$t_0(\log\nu(t_0))'\phi\left(\frac{s}{t_0}\right) + \frac{s}{t_0}\phi'\left(\frac{s}{t_0}\right) = t_0(\log\nu(t_0))'\phi\left(\frac{t}{t_0}\right) + \frac{t}{t_0}\phi'\left(\frac{t}{t_0}\right)$$

$$\text{hence} \qquad A(t_0)\phi(x) + x\phi'(x) = B(t_0)$$

where $A(t_0) = t_0(\log\nu(t_0))'$ and $x = t/t_0$. The last equation is easily integrated to give $\phi(x) = b(t_0)x^{-\mu} + a(t_0)$. Thus h is either a stretched exponential either a power law while other possibilities are ruled out. Note finally that matching different regimes to fix h we integrated the large-time behaviour of $\phi(t)$ and (7.18) holds only approximately for large t. However the ageing function h is only defined for large values of t and in this sense Eq. (7.18) is exact.

To conclude let's note that a similar decomposition into singular and regular parts holds for $g_l(x)$ and $w_l(x)$ around $x = 0$. Again Eq. (7.13) identifies only leading terms. By analogy with the $x = \infty$ case we write:

$$g_l(x \to 0) = g_l^0(x^{-a}) + g_l^{reg}(x)$$

where g_l^0 is a polynomial of degree l in x and $g_l^{reg}(x \to 0) \to 0$. A similar decomposition holds for w_l.

Finally it is worth noting that these results hold provided that a late stationary and an early ageing expansions are valid. Also remark that similar matching can be done in the high temperature phase for short time, β- and α regimes and reproduces results of Sec. 3.1. In that case the appearance of power laws is justified by the underlying criticality. For low temperatures power law scaling in the crossover regime yields a self-consistent picture but a rigorous justification is still missing.

Everything is now ready for derivation of scaling equations which fix the functions g_l and w_l. Taking into account their definitions we expect these equations to be generalisation of the main Landau theory equation (6.17). Therefore we only derive the equations for $l = 1$.

7.5 Scaling equations.

The scaling functions g_1 and w_1 are fixed by Eqs. (7.5). The derivation gives equations on g_1 and w_1 but the exponents α, β drop out and stay unfixed. Due to a rich structure of the equations (7.5) where several distinct time sectors are present the derivation is quite involved. The idea is to set $t - s = xt^\beta$ in (7.5) and decompose integrals into parts corresponding to time sectors in the same manner that was used in diagrammatic derivation of Landau theory (see Sec. 6.4.3):

$$\int_0^t = \int_0^{\eta t} + \int_{\eta t}^{t-\Lambda t^\beta} + \int_{t-\Lambda t^\beta}^{t-\epsilon t^\beta} + \int_{t-\epsilon t^\beta}^{t}$$

$$\int_s^t = \int_s^{s+\epsilon_1 t^\beta} + \int_{s+\epsilon_1 t^\beta}^{t-\epsilon_2 t^\beta} + \int_{t-\epsilon_2 t^\beta}^{t}$$

where the first integral is separated into very short times, ageing, scaling and stationary regimes. A similar decomposition is made for the second integral. Assuming sharp time scale separation every integral is expressed via scaling functions of the corresponding regime. One should also take into account time dependence of the Lagrangian multiplier $z(t)$:

$$z(t) = z(\infty) - z_1 t^{-\alpha} - z_2 t^{-2\alpha} - z_3 t^{-3\alpha} - \cdots \tag{7.20}$$

Finally one should take a limit $t, s \to \infty$ followed by $\eta \to 0$, $\Lambda \to \infty$ and $\epsilon \to 0$ *afterwards*. The ordering of the limits is important.

These decompositions generate expansions of (7.5) in powers of $t^{-\alpha}$, $t^{-\beta}$ (the latter comes from derivative terms). Obviously higher orders are more complicated to compute because of the increasing number and complexity of terms. An extra source of complexity is the complex asymptotics of $g_l(x)$, $w_l(x)$ at $x = 0$ and at $x = \infty$ since these terms also appear in the expansions. All this makes computation of the equations for g_1 and w_1 lengthy and tedious. Higher order equations are expected to be even more painful. Finally one should identify all orders in $t^{-\alpha}$ in the expansions of (7.5) to fix g_l, w_l. The initial parts of expansions (at least up to the order 2) is in powers of $t^{-\alpha}$ only. Derivative terms generate a different powers of t. The order t^0 of both equations (7.5) reproduces Eq. (7.6) for $z(\infty)$. The order $t^{-\alpha}$ reduces to a system of linear equations:

$$\left(z(\infty) - T - \frac{p}{2T} + \frac{p}{2T}q^p - \frac{p}{2T_*}q^p\right) g_1(x) = 0$$

$$\left(z(\infty) - T - \frac{p}{2T} + \frac{p}{2T}q^p - \frac{p}{2T_*}q^p\right) w_1(x) = 0$$

and leaves g_1, w_1 unfixed because of Eq. (7.6). This provides an extra argument to the consistency of the picture with the extra scaling regime in addition to the stationary and ageing regimes. Indeed, otherwise the order $t^{-\alpha}$ would have fixed g_1 and w_1 to an explicit expressions with wrong asymptotics at $x = 0$ and at $x = \infty$. The order $t^{-2\alpha}$ provides equations for g_1, w_1:

$$\int_{x_0}^{\infty} dy[(p-1)g(y)(w'(x+y) - w'(y)) + w'(y)(g(x+y) - g(y))] +$$

$$(p-2)\int_0^{\infty} dy w'(x+y)(g(x+y) - g(y)) +$$

$$\int_{x_0}^{x} dy w'(y)(g(x-y) + (p-2)g(y)) +$$

$$\int_0^{x_0} dy[(p-1)g(y)w'(x+y) + w'(y)(g(x+y) + g(x-y) - 2g(x))] +$$

$$\frac{g^2(x)}{2T_*} + 2w(x_0)g(x) - \frac{(p-1)\nu}{T_*} q g_\infty x + \frac{(p-2)q^2\nu^2}{2T_*} x^2 = c_g \quad (7.21)$$

$$w_1^2(x) + \int_{x_0}^{x} dy w'(y)\frac{g(y)}{T_*} + \int_0^{x} dy w'(y)(w(x-y) - w(x)) = c_w \quad (7.22)$$

where x_0 is such that $g(x_0) = 0$, c_g and c_w contain all the contributions independent of x; g_∞ is a numerical constant. In the derivation of these equations we had to assume that $C(t,s)$ decays faster than $t^{-2\alpha}$ in the very short times regime where s is fixed and $t \to \infty$. Again this decay is consistent with the stretched exponential form of $h(t)$. The behaviour of $C(t,s)$ in the very short time regime can be estimated from the late ageing regime where $C(t,s) \sim q(h(s)/h(t))^\nu$. Clearly decay of $C(t,s)$ in the very short times regime is not slower than a stretched exponential. This hypothesis was also checked numerically.

Derivation of (7.21),(7.22) also fixed the leading order asymptotics of g_2 and w_2 and even some subleading terms. We were also able to conclude that $z_1 = 0$ in the large time expansion of the multiplier $z(t)$. It is easy to check that the solution of Eq. (7.22) has a compatible asymptotic behaviours at $x = 0$ and at $x = \infty$ with those obtained by matching with other regimes. A similar check for Eq. (7.21) is harder.

We have not been able so far to provide any analytic or numerical solution to Eqs. (7.21),(7.22). Neither were we able to prove the existence or uniqueness of their solutions but the existence of solution is supported by numerical checks. Also Eqs. (7.21),(7.22) are invariant under rescaling

$$\nu \to \Delta\nu, \quad (g_1, w_1) \to (\Delta g_1, \Delta w_1).$$

A priori this is a peculiarity of the p-spin model where the solution (7.11) of Eq. (7.8) leaves ν unfixed. Thus, Eqs. (7.21),(7.22) have a one parameter family of solutions. Singular asymptotics of g_1,w_1 at $x = 0$ and at $x = \infty$ discussed above provide also boundary conditions for these equations. The condition at $x = \infty$ (early ageing regime) is compatible with this invariance but the condition at $x = 0$ (short-time or stationary regime) is not. Hence the rescaling invariance is broken and ν is fixed to a particular value. This is not surprising since as it was discussed above the invariance under the change of ν is related to the fact the microscopic time scale is forgotten in the ageing regime. But this timescale is still present in the stationary regime, and the crossover regime relates both regimes.

Finally Eqs. (7.21),(7.22) is a low temperature generalisation of Eq. (6.17) which is one of the central results of the Landau theory.

7.6 Numerical check.

Since many conclusions of the previous section relied on conjectures and one of the exponents has left unknown we performed a series of numerical simulation to test them. Namely the system (7.5) was integrated numerically using the code provided to us by the authors of [52]. As was already discussed above the main technical complexity of numerical integration is that one should treat the two-time quantities. Taking into account that the timescales of interest (ageing regime) extend over several orders in time, usual methods should be modified and a variable time step in the integration procedure should be used.

We started from a direct measurement of α and β. The large time expansion of the multiplier $z(t)$ (7.20) provides a useful estimate for α. Since $z(t)$ is a one time quantity it is easy to compute numerically. The analysis of the corrections to $z(\infty)$ for $p = 3$ at $T = 0.5$ ($T_d = \sqrt{3/8} \approx 0.6125$) yielded $\alpha \approx 0.3$. The exponent β can be estimated directly from its definition as a timescale of a plateau: $C(s + t_\beta(s), s) = q$. This gave $\beta \approx 0.66$ for the same $p = 3$ model at $T = 0.5$. This is in a very good agreement with $\alpha = \beta a$ ($a \approx 0.448$) and yields ((7.17)) $\mu \approx 0.96$, which provides a good collapse of the data of the ageing regime, as found empirically in [114].

This test was repeated for several values of T and $p = 3, 4, 10$ providing good agreement with our analytic description. In Fig. 7.1 the exponents α, β and μ are shown. The observed tendency is that they decrease with p and T. In addition β and μ seem to saturate respectively, to $2/3$ and 1 at $T = 0$. The value $\mu(T = 0) = 1$ corresponds to approximate simple ageing slightly above $T = 0$. It is also clear from Fig. 7.1 that the working assumption $\mu + \alpha > 1$ that assures the validity (7.18) is reliable.

Finally we have also checked numerically the collapse of the data when we use the scaling for the crossover regime: in Fig. 7.2 $-T\partial F/\partial C$ is plotted vs. $(C(s + \tau, s) - q)s^\alpha$ ($\tau = t - s$) parametrised by τ for several values of waiting time s, for $p = 3$ and $T = 0.5$. as a consequence of our scaling hypothesis data on this plot should collapse onto a curve $T/T(x)$ vs $g_1(x)$. Indeed we obtained a

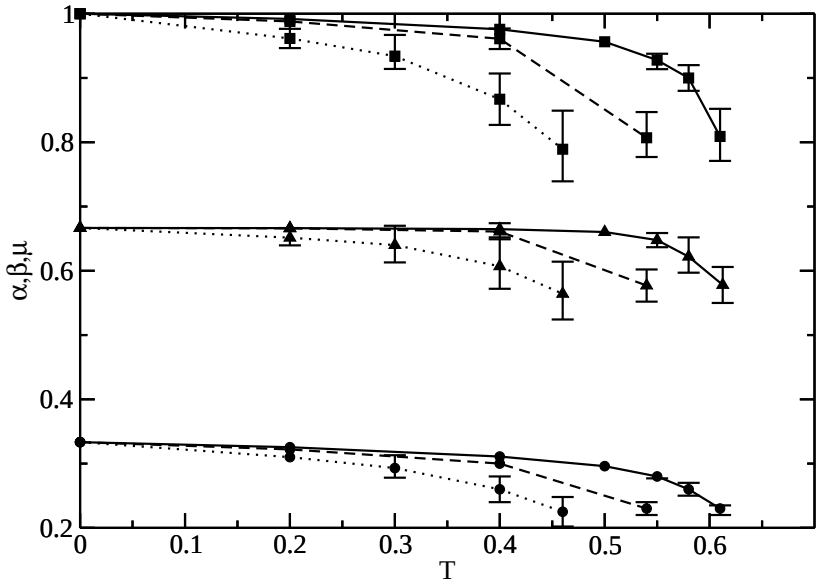


Figure 7.1: Exponents α (circles), β (triangles) and μ (squares) *vs.* temperature for $p = 3$ (solid lines), $p = 4$ (dashes lines) and $p = 10$ (dotted lines).

master curve which represented an onset of FDT violation during the crossover.

7.7 Summary

Construction of the Landau theory for the glass transition for low temperatures is much more complicated than in the high temperature phase because of the absence of equilibrium and the presence of many timescales. We presented only the first step in the construction and identified the candidate for the Landau theory. In the spirit of the high-T phase we had to identify the intermediate scale between the stationary and ageing regimes. For the moment the existence of such a timescale is a conjecture which is confirmed by results of numerical simulations. In order to confirm the existence analytically one has to compute the exponent β within the model or equivalently prove of the existence of solutions for Eqs. (7.21),(7.22). Another difficulty is the extension of the results to higher orders of perturbation. The complexity of the scaling equations which generalise Eq. (2.22) and the complexity of their derivation make such extension quite involved.

Finally, the low temperature Landau theory has been related to studies of ageing in spin glass models. Introduction of an extra scaling regime fixed the problem of spurious time reparametrisation invariance because the regime interpolated explicitly between the stationary and ageing regimes; it also highlighted

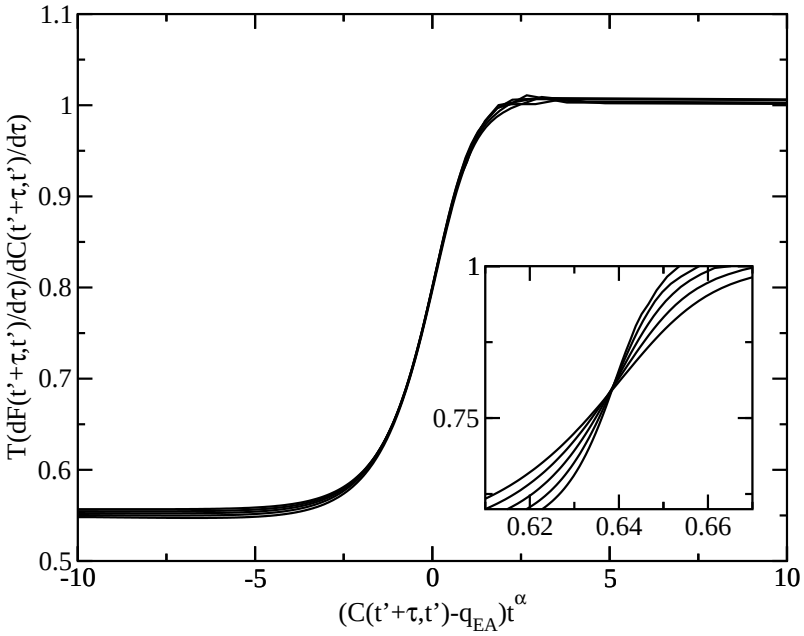


Figure 7.2: Test of the scaling hypothesis for $p = 3$ and $T = 0.5$. Inset: same as main, but with $C(t' + \tau, t')$ as horizontal coordinate ; the crossing occurs at q and corresponds to $\tau = x_0 t^\beta$, with $g(x_0) = 0$. Waiting times are $t' = 2197$, $t' = 4295$, $t' = 8590$, $t' = 17180$ and $t' = 34359$.

the origin of the particular scaling forms (sub-ageing) used to fit data in experiments and simulations. Simple ageing wiht exponent $\mu = 1$ is often used to fit the data. However the 3-spin model provides an example where data can be fitted with simple ageing but $\mu < 1$ exactly. Let's note that imposing $\mu = 1$ would fix the value of β to $1/(1+a(T))$. However a computation shows that this is an increasing function of T while our numerical results suggest the contrary.

Chapter 8

Conclusions and perspectives

The present work considered the problem of slowing down of the dynamics in supercooled liquids. The main focus was the analysis of the Mode-Coupling Theory based scenario of the glass transition. We aimed at resolving several problems related to MCT and extending it within the context of dynamical field theories:

- The uncontrolled nature of the Mode-Coupling Factorisation. It is vital to recover MCT within a field theory to check the consistency of the approximation with physical constraints.
- Analysis of corrections to MCT and test of structural stability of MCT with respect to corrections.
- Mechanism responsible for the cut off of the MCT transition.
- Extending MCT to low temperatures where the liquid falls out of equilibrium.

The results presented in the previous chapters provided solutions to these problems. The analysis of previous attempts to rederive MCT within a field theory context revealed that the importance of time-reversal symmetry for these derivations was overlooked and that these theories actually violated this symmetry. We rebuilt the theories from the ground taking into account the preservation of TRS and constructed perturbation theories that preserved TRS automatically order by order. MCT was identified as a Mode-Coupling approximation in this context. The perturbation series provided systematic corrections around MCT and we carried out a structural stability test of the theory. The latter revealed that MCT persists in all orders of perturbation theory. Thus the cut off mechanism should be identified with non-perturbative effects within the field theories. We also analysed the low temperature regime of a schematic model in order to

extend MCT to low temperatures. Our main result was that it was likely that the Landau theory generalised to this regime also.

Our analysis suggested some directions for a future research which we listed briefly:

- *Stability of MCT with respect to critical fluctuations.* This is a problem we discarded in Chapter 6: we need to show that critical fluctuations are not important for $d > d_u$. As we discussed this requires a generalisation of the proof presented in Chapter 6 to a non-uniform case. Initial steps in that direction that provide the context in which the proof should be generalised were presented in Sec. 6.2.4.

- *MCT below the upper critical dimension.* This problem is motivated by the analogy of MCT with critical phenomena: how critical fluctuations modify MCT predictions below d_u ? The answer is not expected to be simple nor is the computation which can be more complicated compared to critical phenomena. In general we expect that fluctuations modify the exponents a, b and the related exponents for timescales or the $\sqrt{\epsilon}$-dependence. It is unclear whether the cutoff mechanism is related to the fluctuations or not; that is, whether the transition is smoothed by critical fluctuations ?

- *Generalisation of the Landau theory to low temperatures.* As we have seen in Chapter 7 the computations at 1-loop order are already quite involved mainly because of the presence of many timescales in the system and the breakdown of time-reversal symmetry. Adding more loops will complicate the derivation even more. The scaling equations derived in Sec. 7.5 are cumbersome and their generalisation to higher orders is clumsy.

- *The analysis of the expansion in powers of the potential.* This mainly aims at the solution of the ultraviolet divergence problem discussed in Chapter 4. The expansion in powers of the potential was introduced in Sec. 5.1 without any analysis. Much care should be taken when dealing with this theory: although time-reversal symmetry is preserved in perturbation, the corresponding transformation is still non-linear. A first step would be to reproduce MCT within this theory. Once this is accomplished the next step is to construct the Landau theory and analyse the low temperature regime.

List of Figures

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