A Microscopic Approach to Dynamical Heterogeneity in Supercooled Liquids

BY

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Abstract

Building on recently derived inhomogeneous Mode-Coupling Theory, we extend the Generalised Mode Coupling Theory (GMCT) of supercooled liquids to inhomogeneous environments. This provides a systematic and rigorous way of deriving high-point dynamical susceptibilities from variations of the many-body dynamic structure factors with respect to their conjugate field by treating the equations of motion as a Landau theory. This provides a novel and fully microscopic framework to probe for collective relaxation mechanisms in supercooled liquids near the glass transition. We provide an in-depth exploration of these higher-order susceptibilities in the case of a simplified schematic model by studying the behaviour and criticality of dynamical susceptibilities for different glass transition scenarios.

Acknowledgements

I would to express my earnest gratitude towards Prof. Liesbeth Janssen. Firstly for her Nico van Kampen colloquium in Utrecht on that cold and rainy January the 15th 2020, where we first met and my fascination for glassy systems was (re)ignited. Secondly for having accepted to take me as one of her students despite belonging to another institute. Dear Liesbeth, I am thrilled to have worked under your supervision, and I am excited at the idea of continuing this fruitful work in the coming years.

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Before moving onto the science, I would like to place a few words for my two dear friends A & L. Dear A, we started as neighbours and our Mediterranean spirits clicked instantly. Dear L. I am immensely happy to have met you during my time here as well. I am glad to count you as my close friends. I cannot wait to see you both again very soon over some 51s.

I, *Corentin Christophe Laurent Laudicina*, declare that this thesis titled, *"A Microscopic Approach to Dynamical Heterogeneity in Supercooled Liquids"* and the work presented in it are my own. I confirm that:

- This work was done wholly or mainly while in candidature for a research degree at this University.
- Where any part of this thesis has previously been submitted for a degree or any other qualification at this University or any other institution, this has been clearly stated.
- Where I have consulted the published work of others, this is always clearly attributed.
- Where I have quoted from the work of others, the source is always given. With the exception of such quotations, this thesis is entirely my own work.
- I have acknowledged all main sources of help.
- Where the thesis is based on work done by myself jointly with others, I have made clear exactly what was done by others and what I have contributed myself.

Signed:

Date: July 1st 2021

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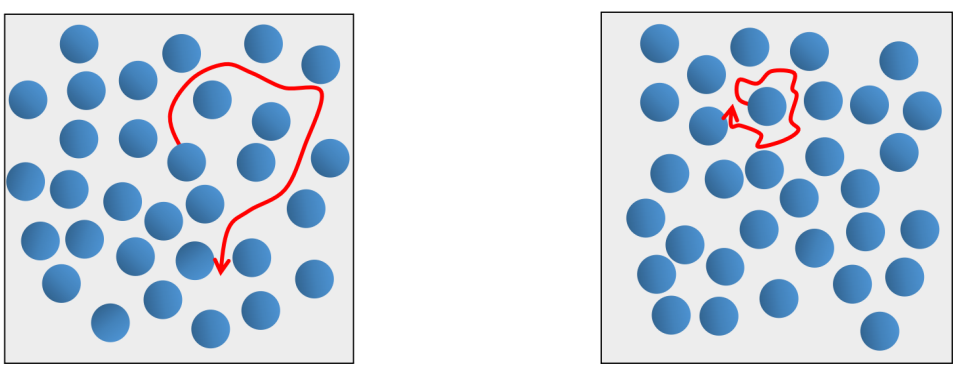
1 Introduction

1.1 The Glass Transition in a Nutshell

Glasses are ubiquitous in today's world, from mere kitchenware to the latest smartphones, we can find structural glasses everywhere. Glasses can be made out of an immense variety of base materials : silica, polymers or even metals. In recent years we have seen the advent of colloidal and granular glasses in our labs as prototypical glass-forming materials. It might thus come as a surprise that the nature of the glass transition is still very poorly understood to this day [4].

Under regular conditions, as a liquid is cooled to its melting temperature T_M, a first order phase transition occurs and the liquid crystallises via nucleation processes. Suppose instead that the liquid does not crystallise upon cooling it past T_M. For instance we could temperature quench it fast enough to simply forbid nucleation, or consider a complex material which does not admit any stable crystalline structure. The liquid then enters a supercooled regime, which is a metastable state of matter. As the temperature is further lowered, very little structural changes are observed, (see Fig.2 for a schematic illustration) yet there exists a small temperature window ΔT centred around T_G in which the viscosity increases dramatically (over 10 orders of magnitude) and full dynamical arrest, on experimental timescales, is reached. This is commonly referred to as a glassy state, and T_G is called the glass transition temperature. The dramatic increase in relaxation time within the space of minute changes in control parameters accompanied by an apparent decoupling between structure and dynamics was referred to as "deepest and most interesting unsolved problem in solid state theory" [4] by P.W. Anderson, the 1977 Nobel Laureate in Physics.



(a) (b)

Figure 2: (a) *Schematic illustration of liquid-like behaviour. Particles are in a ballistic / diffusive regime and the structural correlations are short-lived.* (b) *Schematic Illustration of deep supercooled / glass-like behaviour. Particles do not diffuse over experimental time-scales and vibrate randomly around some effective equilibrium point.*

We emphasise that the transition temperature is empirically defined. On general grounds, T_G is defined as the point where the viscosity $\nu \sim 10^{13}$ [P], in CGS units. We also stress that the supercooled state being a non-equilibrium state, T_G is highly protocol dependent. Furthermore, the glass transition also has tints of a regular thermodynamic transition of second-order. This is notable in the behaviour of the heat capacity, which has an abrupt but continuous jump across T_G. This leads to important properties of various macroscopic observables which may display hysteresis behaviour

across transition lines in the phase diagram.

It is then more appropriate to cast the glass transition as an ergodicity-breaking transition, rather than a phase transition. We may think of it as a purely dynamical phenomena (a dramatic increase in the kinetic coefficients) with some thermodynamic features. In the supercool regime, the free-energy landscape becomes increasingly rugged and our system gets effectively stuck in some deep local minima. It does not visit the neighbouring basins anymore, which breaks ergodicity. To detect this ergodicity breaking point, we use an order parameter that we denote $F_2(\mathbf{k}, t) \propto \langle \hat{\rho}_{\mathbf{k}}(0)^* \hat{\rho}_{\mathbf{k}}(t) \rangle$ where $\hat{\rho}_{\mathbf{k}}(t)$ is a time-dependent Fourier mode of density fluctuations. F_2 is then an autocorrelation function of these density modes. This quantity has the advantage that at long-times it behaves as an order parameter. We find that $F_2 = 0$ in the liquid regime as we can effectively see a liquid as a Markoffian system where the density field relaxes over short timescales, while $F_2 > 0$ in the glassy state since time-dependent structural correlation functions do not decay in solids. By long-time here we mean a time too long to be observed over experimental timescales. We will come back to this order parameter later on.

We briefly consider the thermodynamics of a deeply supercooled liquid. Once in the supercooled regime (past the red-point in Fig.3), one may imagine that a liquid can be cooled indefinitely while never turning into a glass using suitable protocols: the system remains on the blue curve. We can then imagine delaying the glass transition to lower temperatures until the excess entropy $S_{\text{exc}}(T)$ of the liquid with respect to the corresponding crystal state vanishes at some temperature T_K ($S_{\text{exc}}(T_K) = 0$, purple point in Fig.3). This point is known as the Kauzmann temperature [42, 43] and is the source of an interesting paradox : at T_K, we find that the total entropies of a crystal and that of an amorphous solid are equal ! To circumvent this paradox, a true thermodynamic phase transition has been historically postulated to take place at T_K, where spontaneous symmetry breaking into the true thermodynamically stable crystalline state is expected. Since still little is known regarding the nature of the very deep supercooled regime, we may not neglect other possible resolutions to this paradox.

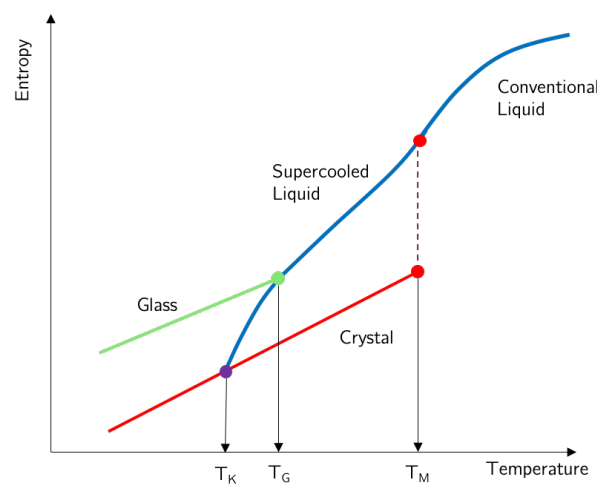


Figure 3: *Illustrated phase-diagram of a liquid at low temperatures and/or high packing densities. T_M is the melting temperature, T_G the glass transition temperature and T_K the Kauzmann temperature.*

1.2 Hopes for a Growing Lengthscale

1.2.1 Dynamical Heterogeneity in Supercooled Liquids

It has long been known that supercooled liquids display strongly heterogeneous dynamics, also often referred to as *dynamical heterogeneity* [72, 68, 13]. More precisely it is not uncommon to find persistent correlations between particle motion in "effective" clusters of size say ξ_{cl}. We show an example of dynamical heterogeneity in a system of binary hard-disks [56] in Fig.4. We clearly see that there are regions of fast-moving particles (in green & red) that coexist with regions of slower moving particles (in blue). This type of behaviour is actually common to a very wide variety of glass-forming materials. The existence of these correlated clusters raises important questions about the true nature of the glass transition and its connection with conventional thermodynamic phase transitions. We are entitled to ask about the mechanisms leading to the emergence these structures, what their sizes are and what quantities govern their size. We may also ask if there are particular structural patterns in these correlated clusters with recent answers from machine learning aided studies [19, 71]. Yet despite years of efforts, these questions are still central to the glass transition problem and are for mostly still unanswered and poorly understood.

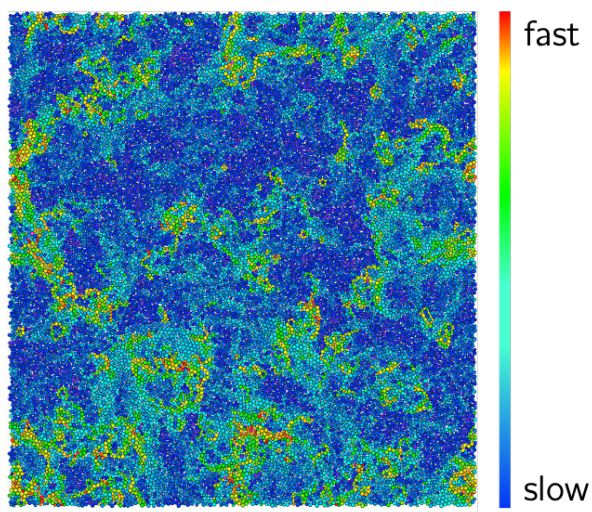


Figure 4: *Mobility field for a simulation of binary hard-disks with radii σ_S, σ_L for the small and large disks respectively. In blue : the slow particles and in red : fast particles. Fast particles have travelled mean-square distance $\langle r^2 \rangle > \sigma_L$ and slow particles $\langle r^2 \rangle < 0.01\sigma_L$ over the principal structural relaxation time τ_α.* Adapted from [56]

The existence of these correlated clusters is in line with the early phenomenological approaches to the glass transition of Adam-Gibbs [1], where the concept of *cooperatively rearranging regions* (CRRs) was first introduced. This was later more rigorously formalised in the context of Random First Order Transition Theory [17] (for a concise review), where it is argued that the size of these CRRs (sometimes referred to as glassites) scales as $\xi_{\text{cl}} \sim 1/S_{\text{exc}}$, the inverse excess configurational entropy previously introduced. As the Kauzmann point is approached, we see that $\xi_{\text{cl}} \to \infty$ as expected for second order phase transitions. However, liquids tend to vitrify at much higher temperatures than the Kauzmann point. So while the existence of a strictly diverging lengthscale at the structural glass transition, which is of interest here, remains a hot debate in the field, we shall nonetheless accept that

there exists at least an emerging and growing one from the combination of numerical and experimental results available. These provide a compelling basis for a viable critical phenomena-like description of glass formation. In this case, we would observe the growth of some sort of a long-ranged 'amorphous' order, hidden in high-order correlation functions indicating the onset of vitrification.

A gross measure of dynamical heterogeneity is the non-Gaussian parameter $\alpha_2(t)$ [54] which can be calculated from particle displacements. For 3D systems, the non-Gaussian parameter is defined as

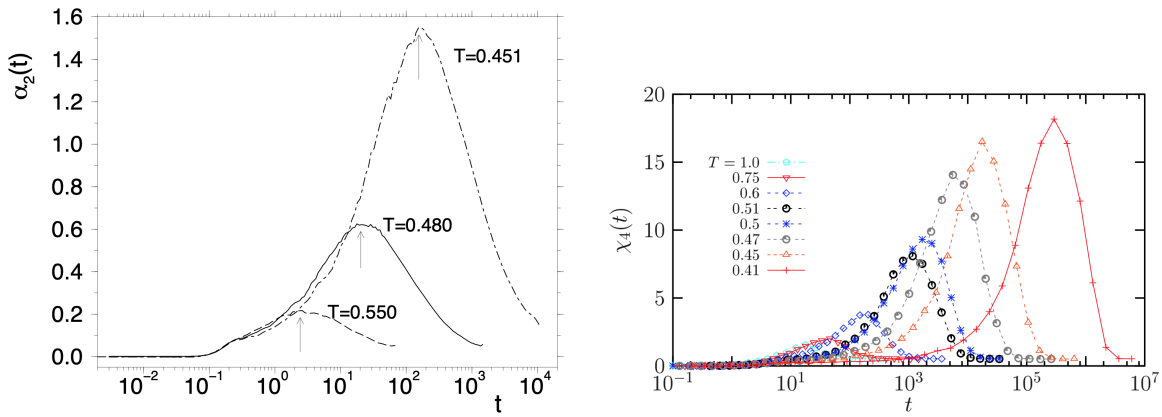
$$\alpha_2(t) = \frac{3}{5}\frac{\langle \Delta r(t)^4 \rangle}{\langle \Delta r(t)^2 \rangle^2} - 1 \tag{1}$$

The non-Gaussian parameter measures the deviation of the particle relaxation from the Gaussian process expected of conventional equilibrium liquids. We show in Fig.5a an example of the non-Gaussian parameter $\alpha_2(t)$ for a supercooled Lennard-Jones mixture displaying dynamical heterogeneities. We see that it has a peak-like behaviour and that this peak grows as the temperature is lowered. It is clear that by construction $\alpha_2(t)$ does capture dynamical heterogeneity in some way, as we observe growing deviations from Gaussian relaxation as the system descends in the supercooled regime. However, this quantity only provides evidence of such non-trivial relaxation processes which are generally assumed to be of cooperative nature, even though nothing in $\alpha_2(t)$ indicates that this might be the case. Extracting a meaningful growing length-scale associated with collective effects from this quantity is therefore impossible.

The established standard measure of dynamical heterogeneity takes the form of a 4-point dynamical susceptibility denoted $\chi_4(t)$ [13]. In resonance with critical phenomena, we may define $\chi_4(t)$ as the (spontaneous) fluctuations of some mobility field, say $\mu(\mathbf{r}, t, t')$ which quantifies the mobility of a particle located at $\mathbf{r}$ in time interval $t - t'$. We can imagine this mobility as the continuum limit of the one showed in Fig.4. Fluctuations in this mobility field indicate the presence of dynamical heterogeneities [30]. Mathematically, we write $\mathrm{Var}[\mu(\mathbf{r}, t, t')] \equiv G_4(\mathbf{r}, t, t')$, and the 4-point susceptibility would then be given by the spatial integration of the latter:

$$\chi_4(t, t') = \int d\mathbf{r} G_4(\mathbf{r}, t, t') \sim \int d\mathbf{r} B(\mathbf{r}, t, t') e^{-\frac{|\mathbf{r}|}{\xi_4(T)}} \tag{2}$$

where $\xi_4(T)$ is the length-scale that we seek to extract and $B(\mathbf{r}, t, t')$ some scaling function. Numerous studies (both experimental and numerical) [10, 46, 66, 11] demonstrate that $\chi_4(t)$ grows in a peak-like manner as the experimental glass transition is approached. This indicates a growing value of $\xi_4(T \to T_c)$ [63]. We provide an example of $\chi_4(t)$ for a simulated supercooled liquid in Fig.5b. We observe a similar behaviour as that of $\alpha_2(t)$: $\chi_4(t)$ displays a peak-like behaviour and as the temperature is lowered, we see that the peak grows and is shifted in time. The location of the peak in time is generally non-arbitrary and can be associated with structural relaxation timescales in the systems. Obtaining an accurate measure of $\chi_4(t)$ in the supercooled regime is a complicated task both numerically and experimentally. Measuring 4-point susceptibilities requires long temporal (several decades) and extensive spatial resolutions (sub-particle radii resolution over large length-scales) since we are probing for correlations of collective fluctuations.



(a) (b)

Figure 5: (a) *Non-Gaussian parameter* $\alpha_2(t)$ *for a supercooled Lennard-Jones liquid at dimensionless temperatures* $T = 0.550, 0.480, 0.451$. Reproduced from [45] (b) *4-point dynamic correlation function* $\chi_4(t)$ *for a supercooled interacting fluid.* Reproduced from [11]

1.2.2 Conventional Critical Phenomena

An important question is whether or not we may cast the problem of the glass transition in the conventional formalism of critical phenomena. Phase transitions are generally characterised by order parameters, whose sign or value correspond to a particular phase. The order parameter is generally a scalar, vector or tensor quantity. For instance the the density field $\rho(\mathbf{r})$ acts as the order parameter for the liquid-gas transition, or the magnetisation $\vec{M}(\mathbf{r})$ in a paramagnetic-ferromagnetic transition.

To see how the formalism of conventional critical phenomena might be applicable to the glass transition, we recapitulate standard results for the (continuum) paramagnetic-ferromagnetic transition. The order parameter is the magnetisation $\vec{M}(\mathbf{r}, T)$, let us denote by T_C the critical temperature at which ferromagnetic ordering takes place (also known as the Curie temperature). Within the Landau formalism of second order phase transitions, we may expand the free-energy $\mathcal{F}$ in powers of the order parameter (and powers of its gradients) by simply requiring a $\mathbb{Z}_2$ invariant free-energy in M. This gives :

$$\mathcal{F}[M] = \int d\mathbf{r} \left(\gamma_1(T)(\vec{\nabla} \cdot \vec{M}(\mathbf{r}))^2 + \gamma_2(T)|\vec{M}(\mathbf{r})|^2 + \gamma_4(T)|\vec{M}(\mathbf{r})|^4 + \mu \vec{H}(\mathbf{r}) \cdot \vec{M}(\mathbf{r}) \right) \tag{3}$$

by truncating the expansion at $\mathcal{O}(M^4)$. The factors $\gamma_j(T)$ are continuous functions of the temperature, and they dictate the behaviour of the system. The last term represents the coupling with the conjugate field H, and we treat it as a source term to generate correlation functions in the partition function. We can determine equilibrium states by finding solutions to $\delta\mathcal{F}[M]/\delta M(\mathbf{r}') = 0$. We define susceptibilities as the variation of relevant order parameters with respect to their conjugate field. In the case of the paramagnetic-ferromagnetic transition, the magnetic susceptibility reads:

$$\chi_m(\mathbf{r}, \mathbf{r}', T) = \frac{\delta M(\mathbf{r})}{\delta H(\mathbf{r}')}$$

which, assuming an isotropic medium can be written in Fourier $\mathbf{q}$-space as:

$$\chi_m(\mathbf{q}) \propto \frac{\xi^2}{1+\xi^2|\mathbf{q}|^2} \tag{4}$$

where ξ is known as the *correlation length*, due to its physical dimension. It describes the length-scale over which the response of magnetisation (i.e. individual spins) is correlated. More intuitively, a low susceptibility implies a single-component response, while a large one implies a collective response to an external stimulus. Near the critical point and in a vanishing external field, both the susceptibilities and the correlation length diverge as:

$$\lim_{H\to 0; T\to T_C^{\pm}} \chi_m(T,H) \propto (1-T/T_C)^{\gamma_\pm} \qquad \lim_{H\to 0; T\to T_C^{\pm}} \xi(T,H) \propto (1-T/T_C)^{\nu_\pm} \tag{5}$$

where quantities $\gamma_\pm, \nu_\pm$ are known as critical exponents. They govern the scaling of these divergences at criticality. Calculating these critical exponents forms the basis of the modern approach to critical phenomena and their classification into *universality classes* from renormalisation group methods for instance.

1.2.3 Dynamical Non-Linear Susceptibilities

We have already mentioned in previous sections that $\chi_4(t)$ is a historically important probe and quantifier of dynamical heterogeneity. However we have also said that it was a very difficult quantity to calculate. We have in mind to construct an equation of motion for a quantity that is capable of capturing dynamical heterogeneity. We recall that we have an order parameter to detect the glassy phase : our auto-correlation function of density modes $F_2(\mathbf{k},t)$. By interpreting the equation of motion for this order parameter as a Landau theory (we need simply assume it exists for now), we would then be able to consider variations of this order parameter with respect to its conjugate field that we denote $U(\mathbf{q}_0)$. We should have in mind that $U(\mathbf{q}_0)$ is an external small pinning field, like a sine or a cosine wave with wave-number $\mathbf{q}_0$, as schematically depicted in Fig.6. We can then define a dynamic susceptibility, that we denote $\chi_3(t)$ analogously to the magnetic susceptibility in conventional critical phenomena :

$$\lim_{U\to 0} \frac{\delta F_2(\mathbf{k};\mathbf{k}',t)}{\delta U(\mathbf{q}_0)} = \chi_3(\mathbf{k};\mathbf{k}',\mathbf{q}_0,t) \tag{6}$$

where we have written $F_2(\mathbf{k};\mathbf{k}',t) \propto \langle \hat{\rho}^*_{\mathbf{k}} \hat{\rho}_{\mathbf{k}'}(t)\rangle$ with two explicit wave-number modes, because the presence of the pinning field U breaks translational invariance. This type of 3-point susceptibility was first proposed as an alternative to $\chi_4(t)$ in a series of important papers [15, 14]. The 4-point and 3-point susceptibilities are actually intimately linked. In the case where the conjugate field is assumed to be the density field, a simple combination of the Cauchy-Schwartz inequality and the Fluctuation Dissipation theorem leads to the conclusion that $\chi_4(t) \geq \rho_0 \kappa_T \beta^{-1} (\chi_3(t))^2$ [12, 15, 14], where ρ_0 is the bulk-fluid density, κ_T the isothermal compressibility and β the inverse temperature. That is, the 3-point susceptibility provides a strict lower bound to the 4-point one. Similar expressions also exist in the case of different conjugate fields [12, 15, 14]. The 3-point susceptibility has the advantage of being much easier to determine than the 4-point one. The reason is that obtaining a good-enough signal for F_2 and then taking a derivative with respect to the conjugate field is much easier (for both simulations and experiments) than waiting for spontaneous fluctuations in the mobility fields in the deep supercooled regime to determine a 4-point correlation function with sufficient accuracy.

Following the theoretical introduction of these 3-point susceptibilities in [12, 15, 14], several groups were able to extract this 3-point susceptibility from high-precision dielectric spectroscopy experiments

in supercooled glycerol [27, 9, 24]. More recently, the 5-point dielectric susceptibility $\chi_5(t)$ [2] was even extracted from supercooled glycerol and propylene carbonate. All these studies demonstrate that $\chi_3(t)$ (and $\chi_5(t)$) behave as 'conventional' critical susceptibilities near vitrification. Hence, the 3-point susceptibility (and its higher order counter-parts) could be very useful objects to probe for the putative growing lengthscale at the onset of the glassy-regime in supercooled liquids. In light of these promising experimental and theoretical results, our objective with this work is to derive a fully microscopic framework that enables us to obtain an equation of motion for the non-linear susceptibilities $\chi_{2n+1}(t)$ of any order. For simplicity, we will consider a conjugate field $U(\mathbf{q}_0) = g\hat{\rho}_{\mathbf{q}_0}$ that couples exclusively to single density modes where g is a coupling strength.

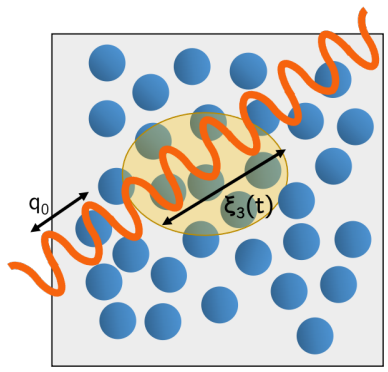


Figure 6: *Schematic illustration of the pinning field $U(\boldsymbol{q}_0)$ and the collective response it induces in a liquid in a supercooled regime. We denote $\xi_3(t)$ the lengthscale over which the 3-point correlated response extends.*

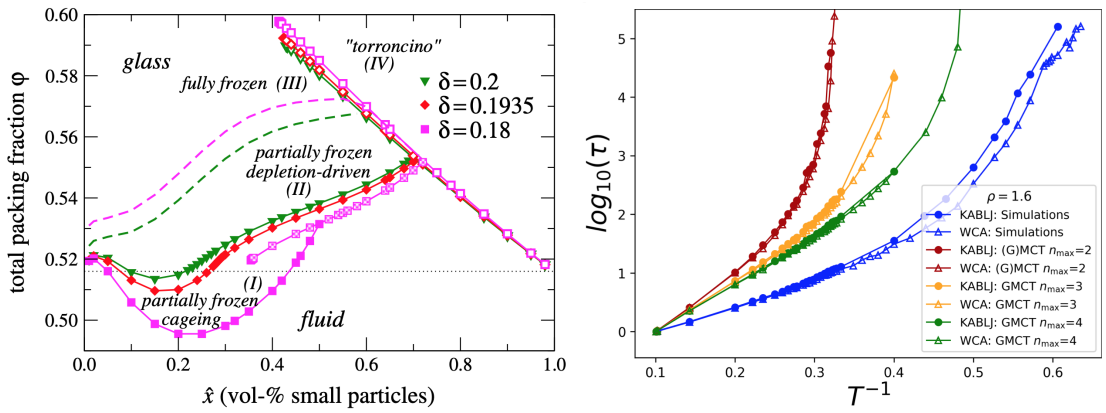
1.3 A Microscopic Framework

Despite decades of efforts, the Mode Coupling Theory (MCT) [20, 32] stands as the only fully microscopic approach to the glass transition problem. The MCT allows for the derivation a self-consistent equation of motion for the order parameter $F_2(\mathbf{k}, t)$ (also commonly referred to as an intermediate scattering function) which take as input structural information only. One of great successes of the MCT is the prediction of an ergodic-to-non-ergodic transition at temperature T_{MCT} from static structure factor input *only*. The theory is also capable of predicting qualitative phase-diagrams for rich glass formers as shown in Fig.7a [67]. Furthermore, the theory also makes important predictions relating power-law exponents of intermediate scattering functions at key physical timescales near the glass transition which are consistent with experimental findings.

However despite the qualitative and quantitative successes, the critical temperature T_{MCT} systematically overestimates the experimentally observed glass transition temperature T_G. It is thus not uncommon to find 'shifted' and rescaled MCT predictions in the literature to allow for direct comparison with experiments. We note that several attempts, commonly referred to as extended MCT (eMCT) [36, 60] introduce additional couplings to the theory with the hope of curing the spurious transition. This however poses serious problems in the case of Brownian dynamics, where no such couplings actually exist. Lastly, we mention that MCT also fails to report on the notorious breakdown of the Stokes-Einstein relation in the supercooled regime.

This caveat of the MCT can (amongst other things) be traced back to an *uncontrolled approx-*

imation made to obtain a self-consistent equation of motion for $F_2(\mathbf{k}, t)$. In 2003, an ambitious theoretical framework was developed by Szamel [61] which effectively delays this uncontrolled approximation by including physical higher-order density correlations in the picture. These higher-order density correlation functions are defined as straight generalisations of $F_2(\mathbf{k}, t)$: $F_{2n}(\mathbf{k}_1, ..., \mathbf{k}_n, t) \propto \langle \hat{\rho}^*_{\mathbf{k}_1} ... \hat{\rho}^*_{\mathbf{k}_n} \hat{\rho}_{\mathbf{k}_1}(t) ... \hat{\rho}_{\mathbf{k}_n}(t) \rangle$. This framework is now formally known as the Generalised Mode Coupling Theory (GMCT) [41]. By incorporating higher-order correlation functions, the hope was to cure MCT's overestimation of T_g. Initial results by Wu et al. [70] were promising in this light, and more recent results seem to show that a 'logarithmic convergence' $T_{\text{GMCT}} \to T_G$ is attained by increasing the number of considered many-density correlation functions in the theory [49, 50]. GMCT also provides quantitative improvements on the power-law exponents of the MCT. Other recent GMCT results [26] also show that it is capable of accurately predicting very different dynamics of interacting glass formers from essentially indistinguishable (for the human eye) structural input. In this case as well, we see from Fig.7b below that increasing the number $n_{\max}$ of many-body correlation functions in the theory seemingly leads to a convergence towards the simulation results.



(a) (b)

Figure 7: (a) *Phase diagram of binary hard-sphere system predicted by MCT. Parameter δ represent the radii ratio of the binary system [67].* - (b) *Comparison of relaxation time predicted from GMCT for multi-component interacting glass formers with simulation [26].*

In this work, we will make use of the fact that the GMCT of the glass transition can incorporate an arbitrary number of many-density correlation functions F_{2n} to obtain direct access to the higher-order dynamical susceptibilities defined as straight generalisations to (6). In a first instance, we will therefore extend the GMCT to inhomogeneous environments by appropriately considering a spatially varying external field $U(\mathbf{q}_0)$ that we recall is chosen to couple to the density modes. We stress that this is a completely novel result, and refer to this new framework as iGMCT. By treating the equations of motion for these F_{2n} as a Landau theory [5], their functional variations with respect to the external field $U(\mathbf{q}_0)$ then results in an equation of motion for the odd-point susceptibilities $\chi_{2n+1}(t) \equiv \delta F_{2n}(t)/\delta U$ of any order which can in turn be solved self-consistently. We note that progress in this line has already been made in recent years with the development of the inhomogeneous MCT from which the 3-point susceptibility has been derived [18, 52]. Additionally, the microscopic equation for the three-point susceptibility $\chi_3(t)$ has been solved in [64] with non-trivial results relating to the dependence on the wave-numbers. The authors report on the emergence of a lengthscale from the iMCT framework but more intriguingly on the absence of a dominant density mode $\hat{\rho}_{\mathbf{k}}$ that would drive these dynamical heterogeneities in time. We stress however that the development of these non-linear susceptibilities is

still in its infancy, and in-depth comparisons with the established $\chi_4(t)$ are still lacking.

2 A Microscopic Framework for the Glass Transition in Inhomogeneous Environments

2.1 The Mori-Zwanzig Formalism & Generalised Langevin Equations

We seek to construct equations of motion for a set of classical dynamical observables in the language of statistical mechanics. These observables will eventually turn out to be our order parameters F_{2n}, but we keep the discussion general for now. We demonstrate below how to construct a formal function space out of classical dynamical variables and how to derive Generalised Langevin Equations (GLEs) in this function space. This technique serves as the basis of mode-coupling theories.

Let $\mathcal{A}$ be a general dynamical variable of an arbitrary autonomous classical system with Hamiltonian $\mathcal{H}$. Clearly $\mathcal{A} \equiv \mathcal{A}(\{\mathbf{q}\}, \{\mathbf{p}\})$ is a function of the set of generalised positions and momenta $\{\mathbf{q}\}, \{\mathbf{p}\}$. This dynamical variable evolves in time according to Hamilton's equation of motion, which can be casted in the following compact form using Poisson brackets:

$$\frac{d\mathcal{A}(t)}{dt} = \{\mathcal{H}, \mathcal{A}\} = i\mathcal{L}\mathcal{A} \tag{7}$$

where the Liouvillian operator :

$$i\mathcal{L} = \sum_j \left[\frac{\partial \mathcal{H}}{\partial p_j} \frac{\partial}{\partial q_j} - \frac{\partial \mathcal{H}}{\partial q_j} \frac{\partial}{\partial p_j} \right] \tag{8}$$

has been defined. We note that (7) is a linear differential equation which admits solution

$$\begin{aligned} \mathcal{A}(t) &= e^{i\mathcal{L}t}\mathcal{A}(t=0) \\ &= \sum_{l=0}^{\infty} \frac{(i\mathcal{L}t)^l}{l!}\mathcal{A}(t=0) \end{aligned} \tag{9}$$

provided that $\mathcal{L}$ is bounded in the (function) space of dynamical variables. This demonstrates that the Liouvillian is the infinitesimal generator of the time-translation group in classical mechanics [34]. Since $i\mathcal{L}$ is a linear operator, we may think of the dynamical variables on which it acts as members of some vector space. To make this idea clearer, we introduce bra-ket notation : $\mathcal{A} \to |\mathcal{A}\rangle \in \mathbb{H}$, where $\mathbb{H}$ denotes the space of all dynamical variables. Additionally, we introduce an inner product on this vector space with the help of a Gibbsian probability weight $\psi(\Gamma) = \exp[-\beta\mathcal{H}(\Gamma)]$

$$\langle A|B\rangle \equiv \frac{1}{\mathcal{Z}} \int d\Gamma A^* B \psi(\Gamma) \tag{10}$$

where A^* denotes complex conjugation and $\mathcal{Z}$ the partition function. This choice of inner product is commonly referred to as Zwanzig's inner product [73]. Other suitable inner products also exist, see for instance Mori's inner product [38], which resembles a (classical) Kubo transform. A Euclidean metric can be easily defined as $d = \sqrt{\langle A|A\rangle}$. This completes the construction of our space of dynamical variables, which is formally a Hilbert function space.

The formal solution (9) for the time evolution of dynamical variables is not very useful as evaluating the exponential of a differential operator is generally a complex task. Instead, it is more judicious

to consider only a subset of dynamical variables $\{|\mathcal{A}_j\rangle\} \subset \mathbb{H}$, whose elements are referred to as slow variables and are generally of greater physical interest such as the energy density, number density, momentum density,or angular momentum density [58, 57, 51]. In our case, we will later restrict the slow subspace to (multiplets) of density modes and their associated currents. If necessary, the Gram-Schmidt procedure (or any other orthonormalisation method) can be employed to construct an orthonormal basis for the subspace spanned by this set. The denomination *slow* stems from the fact that these dynamical variables should evolve on macroscopic time-scales rather than microscopic.

We abuse notation and write $|\mathcal{A}(t=0)\rangle \equiv |\mathcal{A}\rangle$ for quantities evaluated at zero time from now on and for the rest of this work. We may define a projector $\mathcal{P}$, which projects any dynamical variable onto the subspace spanned by our chosen subset $\{|\mathcal{A}_j\rangle\}$. The physical motivation behind this is that the equations of motion for a set of macroscopic 'slow' observables are generally self-determined in physics. Formally, we write

$$\mathcal{P} = \sum_{l,l'} |\mathcal{A}_l\rangle\, g_{ll'}\, \langle \mathcal{A}_{l'}| \tag{11}$$

where $g_{ll'} \equiv \langle A_l|A_{l'}\rangle^{-1}$ is such that $\mathcal{P}^2 = \mathcal{P}$, as for any projector. Let us denote the complement to this projector by $\mathcal{Q} = \mathbb{1} - \mathcal{P}$. We may re-write the Liouville equation:

$$\begin{aligned}
\frac{d\,|\mathcal{A}_j(t)\rangle}{dt} &= \frac{d}{dt} e^{i\mathcal{L}t}\,|\mathcal{A}_j\rangle \\
&= e^{i\mathcal{L}t}(\mathcal{P}+\mathcal{Q})i\mathcal{L}\,|\mathcal{A}_j\rangle \\
&= \sum_{l,l'} \langle \mathcal{A}_j|\mathcal{A}_l\rangle^{\dagger} \langle \mathcal{A}_l|\mathcal{A}_{l'}\rangle^{-1} e^{i\mathcal{L}t}\,|\mathcal{A}_l\rangle + e^{i\mathcal{L}t}\mathcal{Q}i\mathcal{L}\,|\mathcal{A}_j\rangle \\
&= \sum_{l} i\Omega_{jl}\,|\mathcal{A}_l(t)\rangle + e^{i\mathcal{L}t}\mathcal{Q}i\mathcal{L}\,|\mathcal{A}_j\rangle
\end{aligned} \tag{12}$$

where the quantities $i\Omega_{ij}$ are elements of a matrix $i\mathbf{\Omega}$ which we call a *frequency* matrix since it has the dimension of inverse time. We now decompose the time-evolution operator in a non-trivial manner using the Dyson identity:

$$e^{i\mathcal{L}t} = e^{i\mathcal{L}t}\mathcal{O}(t) + e^{i\mathcal{Q}\mathcal{L}t}$$

where it can be shown that [55]

$$\mathcal{O}(t) = i\int_0^t d\tau e^{-i\mathcal{L}\tau}\mathcal{P}\mathcal{L}e^{i\mathcal{Q}\mathcal{L}t}$$

Upon substitution into (12), we obtain

$$\frac{d\,|\mathcal{A}_j(t)\rangle}{dt} = \sum_l i\Omega_{jl}\,|\mathcal{A}_l(t)\rangle + \int_0^t d\tau e^{i\mathcal{L}(t-\tau)}i\mathcal{P}\mathcal{L}\,|f_j(\tau)\rangle + |f_j(t)\rangle \tag{13}$$

where the fluctuating force $|f_j(t)\rangle \equiv e^{i\mathcal{Q}\mathcal{L}t}i\mathcal{Q}\mathcal{L}\,|\mathcal{A}_j\rangle$ has been defined. At time zero, we can write

$$\begin{aligned}
|f_j\rangle &= (1-\mathcal{P})|\dot{\mathcal{A}}_j\rangle \\
&= |\dot{\mathcal{A}}_j\rangle - \sum_l i\Omega_{jl}\,|\mathcal{A}_l\rangle
\end{aligned} \tag{14}$$

where we have used the following result: $\mathcal{P}|\dot{\mathcal{A}}_j\rangle = \sum_l i\Omega_{jl}\,|\mathcal{A}_l\rangle$. Formally, the fast-fluctuating force is composed of the time-derivative of the slow-variables, projected in the orthonormal (fast) subspace and propagated in that same fast-subspace [55]. By construction we have that the fluctuating force is orthogonal to the set of slow dynamical variables : $\langle f_i(t)|\mathcal{A}_j\rangle = 0\ \forall\ j$ and at all times, which motivates its nomenclature. Next we need to determine $i\mathcal{P}\mathcal{L}\,|f_j(t)\rangle$:

$$\begin{aligned} i\mathcal{P}\mathcal{L}\,|f_j(t)\rangle &= \sum_{l,l'} \langle i\mathcal{L}f_j(t)|\mathcal{A}_l\rangle^\dagger \langle\mathcal{A}_l|\mathcal{A}_{l'}\rangle^{-1}\,|\mathcal{A}_{l'}\rangle \\ &= i\sum_{l,l'} \langle f_j(t)|\mathcal{L}\mathcal{A}_l\rangle^\dagger \langle\mathcal{A}_l|\mathcal{A}_{l'}\rangle^{-1}\,|\mathcal{A}_{l'}\rangle \\ &= i\sum_{l,l'} \langle f_j(t)|\mathcal{Q}\mathcal{L}\mathcal{A}_l\rangle^\dagger \langle\mathcal{A}_l|\mathcal{A}_{l'}\rangle^{-1}\,|\mathcal{A}_{l'}\rangle \\ &= -\sum_{l,l'} \langle f_j(t)|f_l(0)\rangle^\dagger \langle\mathcal{A}_l|\mathcal{A}_{l'}\rangle^{-1}\,|\mathcal{A}_{l'}\rangle \\ &= -\sum_{l} K_{jl}(t)\,|\mathcal{A}_l\rangle \end{aligned} \tag{15}$$

where in the third line we have made use of orthogonality of the fluctuating force $|f_j\rangle$ with the slow subset $\{|\mathcal{A}_l\rangle\}$. By substitution into (13), we obtain:

$$\frac{d\,|\mathcal{A}_j(t)\rangle}{dt} = \sum_l i\Omega_{jl}\,|\mathcal{A}_l(t)\rangle - \sum_l \int_0^t d\tau K_{jl}(\tau)\,|\mathcal{A}_l(t-\tau)\rangle + |f_j(t)\rangle \tag{16}$$

Since we are interested in correlation functions of slow variables $C_{ij}(t) = \langle\mathcal{A}_i|\mathcal{A}_j(t)\rangle$, we close the above expression to obtain, using matrix notation (and changing variables in the convolution integral):

$$\boxed{\frac{d\mathbf{C}(t)}{dt} = i\mathbf{\Omega}\cdot\mathbf{C}(t) - \int_0^t d\tau\mathbf{K}(t-\tau)\cdot\mathbf{C}(\tau)} \tag{17}$$

which is known as the Mori-Zwanzig (M-Z) equation. It is a type of Generalised Langevin Equation due to its resemblance to the phenomenological equation for a Brownian colloid in a solvent if we associate the integral term with random stochastic forces. The M-Z equation differs from conventional Fokker-Planck approaches of out-of-equilibrium statistical mechanics [65] by its delayed-feedback nature encoded in the memory kernel $\mathbf{K}$ and the absence of explicit random processes. The M-Z equation is microscopically exact and fully deterministic. As we will see, once the slow variables of a system have been identified, the main difficulty lies in finding an approximate closed form for the memory kernel $\mathbf{K}$. We stress that while the memory kernel is proportional to the auto-correlation of the random force, it does not necessarily imply that it is a fast-variable itself. Lastly, we emphasise that no approximations have been made, and that (17) is a formally exact equation for the time evolution of correlation functions.

The construction leading to (17) helps us understand the behaviour of the solutions in an abstract manner. Since time evolution takes the form of a unitary operator, we may imagine the time evolution of the vector of slow variables $|\vec{\mathcal{A}}\rangle$ as a rotation over the surface of a classical 'Bloch hypersphere'. At short times, $|\vec{\mathcal{A}}\rangle$ remains in the sub-space spanned by its elements, but at longer-times the time-evolution will make $|\vec{\mathcal{A}}\rangle$ pick up character from the fast-subspace [73], which we associate with random noise and non-trivial hydrodynamic effects in applications to liquid behaviour. It is this 'interaction' of $|\vec{\mathcal{A}}\rangle$ with the fast subspace that leads to the successes of mode-coupling theories in predicting

the behaviour of supercooled liquids for instance. We additionally mention that the above formalism is directly translatable to quantum systems where the language of Hilbert spaces is more common [53].

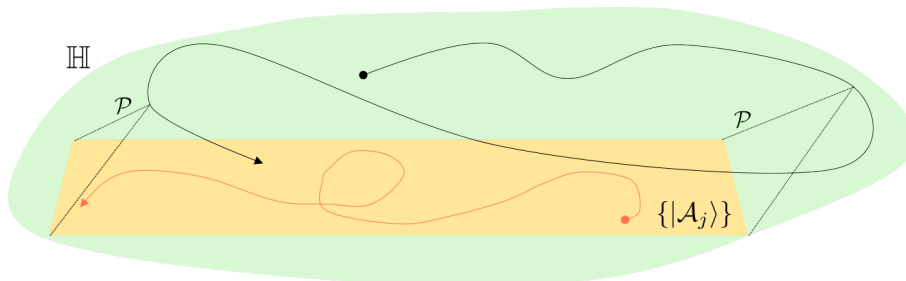


Figure 8: *Schematic illustration of the Mori-Zwanzig projection operator formalism. The green blob represents our functional space of all dynamical variables. In orange we represent the slow-subspace. The exact solution to the slow variables (black trajectory) is projected onto the slow subspace (red trajectory) with the help of projection operator $\mathcal{P}$.*

2.2 Inhomogeneous General Langevin Equations

We extend the formalism of the previous section to inhomogeneous environments. We consider a classical fluid of N-particles in a spatially varying external field $U(\mathbf{q}_0)$ which couples to density fluctuations. The Hamiltonian reads : $\mathcal{H} = \mathcal{H}_0 + U(\mathbf{q}_0)$ where $\mathcal{H}_0$ contains the usual kinetic and interaction potential contributions. In addition to the matrix structure of the equation of motion we introduce a *-product, which takes into account integration over the relevant shared degrees of freedom (momenta in our case below). This is due to translational-symmetry breaking from the external field. We should think of it as a continuous matrix product as is common in Green's function methods in field theories. Using Einstein summation convention for repeated (matrix) Greek indices, (17) now reads:

$$\dot{C}_{\alpha\beta}(t) - i\Omega_{\alpha\gamma} * C_{\gamma\beta}(t) + \int_0^t d\tau K_{\alpha\gamma}(t-\tau) * C_{\gamma\beta}(\tau) = 0 \tag{18}$$

where for instance, the frequency matrix now admits the following form : $i\Omega_{\alpha\beta} = \langle \dot{A}_\alpha | A_\gamma \rangle * \langle A_\gamma | A_\beta \rangle^{-1}$. Down the line we will also make use of a timescale separation approximation, which decomposes the integral kernel into $K_{\alpha\beta}(t) = \nu_{\alpha\beta}\delta(t) + M_{\alpha\beta}(t)$. The fast decaying quantities are all concatenated in an effective instantaneous friction parameter $\nu_{\alpha\beta}$ while the slow modes are kept in the memory term $M_{\alpha\beta}(t)$.

To obtain the final form of the equations of motion, we will have to invert correlation-functions, which is a non-trivial exercise from the given 'operator'-like structure of the latter. Accounting for the fact that classical quantities commute with each other with an appropriate symmetry factor $1/m!$, we define the inverse of an arbitrary equal-time correlation matrix $C^{(2m)}_{\alpha\beta}$ of dimension $2m$ in Fourier space as follows [3, 62]:

$$\begin{aligned}\left(C^{(2m)}\right)_{\alpha\gamma} * \left(C^{(2m)}\right)^{-1}_{\gamma\beta} &= \frac{1}{m!}\sum_{\{\mathbf{k}''_j\}}\left(C^{(2m)}\right)_{\alpha\gamma}(\mathbf{k}_1,...,\mathbf{k}_m;\mathbf{k}''_1,...,\mathbf{k}''_m)\left(C^{(2m)}\right)^{-1}_{\gamma\beta}(\mathbf{k}''_1,...,\mathbf{k}''_m;\mathbf{k}'_1,...,\mathbf{k}'_m)\\ &= \delta_{\alpha\beta}\sum_{\sigma(\{\mathbf{k}'_j\})}\delta_{\mathbf{k}_1,\mathbf{k}'_1}...\delta_{\mathbf{k}_m,\mathbf{k}'_m}\\ &= \delta_{\alpha\beta}\quad \mathrm{Id}_{2m}(\mathbf{k}_1,...,\mathbf{k}_m;\mathbf{k}'_1,...,\mathbf{k}'_m)\end{aligned} \tag{19}$$

where the sum $\sum_\sigma$ runs over all permutations of the elements of the set $\{\mathbf{k}'_j\}$. We have defined a general identity that we denote $\mathrm{Id}_{2m}(\mathbf{k}_1,...,\mathbf{k}_m;\mathbf{k}'_1,...,\mathbf{k}'_m) \equiv \sum_{\sigma(\{\mathbf{k}'_j\})}\delta_{\mathbf{k}_1,\mathbf{k}'_1}...\delta_{\mathbf{k}_m,\mathbf{k}'_m}$ which is sometimes referred to as a symmetrised generalised Kronecker symbol. We note that we must pay careful attention to algebraic manipulations in theories with an inhomogeneous environment as the nature of the considered external field could breaks the hermicity of the Hamiltonian ($\mathcal{H} \neq \mathcal{H}^\dagger$) and thus that of the Liouvillian as well ($i\mathcal{L} \neq -i\mathcal{L}^\dagger$).

In the present work, we will always have a subset of slow variables of dimension two, so that the MZ equation is a 2×2 matrix integro-differential equation. Ultimately, only the 2-1 element of the system will be of interest to us. Explicitly, we can always write (for reasons that will become clear later on)

$$\dot{C}_{11}(t) = i\Omega_{12} * C_{21}(t)$$

and

$$\dot{C}_{21}(t) = i\Omega_{21} * C_{11}(t) - \int d\tau K_{22}(t-\tau) * C_{21}(\tau)$$

We invert the first equation to close the second one in favour of $C_{11}(t)$, leading to :

$$\boxed{\ddot{C}_{11}(t) - i\Omega_{12} * i\Omega_{21} * C_{11}(t) + \nu\,\dot{C}_{11}(t) + \int d\tau\; i\Omega_{12} * M_{22}(t-\tau) * (i\Omega_{12})^{-1} * \dot{C}_{11}(\tau) = 0} \tag{20}$$

which forms the basis of inhomogeneous mode coupling theories for the glass transition.

2.3 Inhomogeneous Mode Coupling Theory

We are now in a position to choose our set of dynamical variables and derive an inhomogeneous Mode-Coupling Theory (iMCT). Since we seek to derive a theory for dynamical correlations of density fields, a density fluctuation mode and its associated (longitudinal) current seems an appropriate starting point. In Fourier space, these quantities take the following form :

$$\begin{aligned}|A^{(1)}_{\mathbf{k}}(t)\rangle = |\hat{\rho}_{\mathbf{k}}(t)\rangle &= \sum_{j=1}^{N} e^{i\mathbf{k}\cdot\mathbf{r}_j(t)} - \left\langle \sum_{j=1}^{N} e^{i\mathbf{k}\cdot\mathbf{r}_j(t)}\right\rangle\\ |A^{(2)}_{\mathbf{k}}(t)\rangle = |j^L_{\mathbf{k}}(t)\rangle &= \sum_{j=1}^{N}\frac{\hat{\mathbf{k}}\cdot\mathbf{p}_j}{m}e^{i\mathbf{k}\cdot\mathbf{r}_j(t)}\end{aligned} \tag{21}$$

where the canonical average is performed with respect to the perturbed Hamiltonian $\mathcal{H}$ defined in the previous section. The dynamic correlation matrix is thus $C_{\alpha\beta}(\mathbf{k};\mathbf{k}',t) \equiv \langle A^{(\alpha)}_{\mathbf{k}}|A^{(\beta)}_{\mathbf{k}'}(t)\rangle$ where the

semi-column separates the vectors in the space of dynamical variables (to the right of the semi-column) from those belonging to its dual (to the left of the semi-colomn). By convention, we will always assume that time-dependent quantities belong to the space of dynamical variables and are thus to the right of the semi-column. It is easy to see that for our choice of dynamical variables, the zero-time correlation matrix is diagonal: $C_{\alpha\beta} \propto \delta_{\alpha\beta}$, as for any equal time variable x, $\langle x|\dot{x}\rangle = -\langle x|\dot{x}\rangle = 0$ by integrating by parts. We thus have

$$\mathbf{C}(\mathbf{k}_1;\mathbf{k}_2,t=0) = \begin{pmatrix} NS(\mathbf{k}_1;\mathbf{k}_2) & 0 \\ 0 & \frac{Nk_BT}{m}(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_2)\phi(\mathbf{k}_1-\mathbf{k}_2) \end{pmatrix} \tag{22}$$

The first entry $S(\mathbf{k}_1;\mathbf{k}_2) \equiv N^{-1}\langle\hat{\rho}_{\mathbf{k}_1}|\hat{\rho}_{\mathbf{k}_2}\rangle$ is known as the (inhomogeneous) *static-structure factor*. We have also defined the average of a single density mode $\phi(\mathbf{k}) = N^{-1}\langle\sum_{j=1}^{N} e^{i\mathbf{k}\cdot\mathbf{r}_j}\rangle$. Along with their higher-order equivalents, many-body structure factors serves as sole initial conditions to the equations of motion we derive. The 2-body static structure factor $S(\mathbf{k}_1;\mathbf{k}_2)$ is related to the well known pair-distribution function $g(\mathbf{r}_1,\mathbf{r}_2)$, which can be calculated from integral equation theory [39]. Its time dependent version will be denoted $F_2(\mathbf{k}_1;\mathbf{k}_2,t) \equiv N^{-1}\langle\hat{\rho}_{\mathbf{k}_1}|\hat{\rho}_{\mathbf{k}_2}(t)\rangle$ which we recall serves as an order parameter to detect the glass phase. F_2 is often referred to as the *dynamical structure factor*. To obtain the second non-zero entry, we make use of the following result :

$$\begin{aligned}
\langle A^{(2)}_{\mathbf{k}}|A^{(2)}_{\mathbf{k}'}\rangle &= \left\langle \sum_{l,l'=1}^{N} \frac{\hat{\mathbf{k}}\cdot\mathbf{p}_l}{m} e^{-i\mathbf{k}\cdot\mathbf{r}_l} \frac{\hat{\mathbf{k}'}\cdot\mathbf{p}_{l'}}{m} e^{i\mathbf{k}'\cdot\mathbf{r}_{l'}} \right\rangle \\
&\approx \frac{k_BT}{m} \sum_{l,l'=1}^{N} \delta_{ll'} \left\langle (\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')e^{-i\mathbf{k}\cdot\mathbf{r}_l} e^{i\mathbf{k}'\cdot\mathbf{r}_{l'}} \right\rangle \\
&= \frac{Nk_BT}{m}(\hat{\mathbf{k}}\cdot\hat{\mathbf{k}}')\phi(\mathbf{k}-\mathbf{k}')
\end{aligned}$$

In the absence of an external field, we have $\phi(\mathbf{k})\delta_{\mathbf{k},\mathbf{0}} = 1$. We note that we make use of energy conservation to write $\langle v_l^{\alpha} v_{l'}^{\alpha'}\rangle = \delta_{\alpha\alpha'}k_BT/m$, as the direction vector $\hat{\mathbf{k}}$ only picks up a single arbitrary direction α. The next quantity to be evaluated is the frequency matrix $i\Omega_{\alpha\beta}(\mathbf{k}_1,\mathbf{k}_2) \equiv \sum_{\mathbf{k}}\langle A^{(\alpha)}_{\mathbf{k}_1}|A^{(\gamma)}_{\mathbf{k}}\rangle\langle A^{(\gamma)}_{\mathbf{k}}|A^{(\beta)}_{\mathbf{k}_2}\rangle^{-1}$. It can be put into form

$$i\mathbf{\Omega}(\mathbf{k}_1;\mathbf{k}_2) = \begin{pmatrix} 0 & i|\mathbf{k}_1|\delta_{\mathbf{k}_1,\mathbf{k}_2} \\ \sum_{\mathbf{k}} \frac{k_BT}{m}\hat{\mathbf{k}}_1\cdot(i\mathbf{k})\phi(\mathbf{k}_1-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}_2) & 0 \end{pmatrix} \tag{23}$$

At last the integral kernel needs to be evaluated. We recall the definition of the fluctuating force (14) which in our case is a two-component vector where the first element is zero:

$$|\mathbf{f}_{\mathbf{k}}\rangle = \begin{pmatrix} 0 \\ |R_{\mathbf{k}}\rangle \end{pmatrix}$$

and

$$|R_{\mathbf{k}}\rangle = |\dot{A}^{(2)}_{\mathbf{k}}\rangle - \sum_{\mathbf{k}'} i\Omega_{21}(\mathbf{k},\mathbf{k}')\,|A^{(1)}_{\mathbf{k}'}\rangle \tag{24}$$

which can then be inserted in the definition of the memory kernel (15):

$$\begin{aligned}\mathbf{K}(\mathbf{k}_1;\mathbf{k}_2,t) &= \sum_{\mathbf{k}} \langle \mathbf{f}_{\mathbf{k}_1}|e^{i\mathcal{Q}\mathcal{L}t}|\mathbf{f}_{\mathbf{k}}\rangle\langle \mathbf{A}_{\mathbf{k}}|\mathbf{A}_{\mathbf{k}_2}\rangle^{-1} \\ &= \sum_{\mathbf{k}} \begin{pmatrix} 0 & 0 \\ 0 & \langle R_{\mathbf{k}_1}|e^{i\mathcal{Q}\mathcal{L}t}|R_{\mathbf{k}}\rangle \end{pmatrix} \begin{pmatrix} NS(\mathbf{k};\mathbf{k}_2) & 0 \\ 0 & J_2(\mathbf{k};\mathbf{k}_2) \end{pmatrix}^{-1} \\ &= \begin{pmatrix} 0 & 0 \\ 0 & \sum_{\mathbf{k}} \langle R_{\mathbf{k}_1}|e^{i\mathcal{Q}\mathcal{L}t}|R_{\mathbf{k}}\rangle J_2^{-1}(\mathbf{k};\mathbf{k}_2) \end{pmatrix}\end{aligned}$$

This form justifies the inversion leading to (20), as all matrix elements of $\mathbf{K}$ are zero expect for the lower-right corner. We note that we have defined $J_2^{-1}(\mathbf{k};\mathbf{k}') \equiv \langle A_{\mathbf{k}}^{(2)}|A_{\mathbf{k}'}^{(2)}\rangle^{-1}$ to ease notation. In full, we may now write the inhomogeneous GLE for the dynamic structure factor:

$$\begin{aligned}0 = \ddot{F}_2(\mathbf{k}_1;\mathbf{k}_2,t) + \nu\dot{F}_2(\mathbf{k}_1;\mathbf{k}_2,t) + \frac{k_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}(\mathbf{k}_1\cdot\mathbf{k})\phi(\mathbf{k}_1-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}')F_2(\mathbf{k}';\mathbf{k}_2,t) \\ + \sum_{\mathbf{k},\mathbf{k}'}\int_0^t d\tau \frac{|\mathbf{k}_1|}{|\mathbf{k}'|}\langle R_{\mathbf{k}_1}|e^{i\mathcal{Q}\mathcal{L}t}|R_{\mathbf{k}}\rangle J_2^{-1}(\mathbf{k};\mathbf{k}')\dot{F}_2(\mathbf{k}';\mathbf{k}_2,\tau)\end{aligned} \tag{25}$$

The principal difficulty lies in the calculation of the propagation of the fluctuating force in the fast subspace : $\langle R_{\mathbf{k}_1}|e^{i\mathcal{Q}\mathcal{L}t}|R_{\mathbf{k}}\rangle$, as a closed form for $\mathcal{Q}$ is not directly obtainable, let alone its exponential. We look for a first order approximation to this correlation-function. We remark that $|R_{\mathbf{k}}\rangle \propto |\dot{A}_{\mathbf{k}}^{(2)}\rangle \propto |\frac{d}{dt}\mathbf{p}\rangle$ is an impulse term. Since any force can be expressed as the gradient of a potential, we heuristically have, neglecting the external field contributions:

$$\frac{d}{dt}\mathbf{p} = -\sum_{ll'}\vec{\nabla}\cdot\Phi(|\mathbf{r}_l-\mathbf{r}_{l'}|) \propto \sum_{\mathbf{q}} i\mathbf{q}\tilde{\Phi}(\mathbf{q})\hat{\rho}_{\mathbf{q}}^*\hat{\rho}_{\mathbf{q}}$$

which contains a pair of fluctuating density modes. Moreover, a pair of density modes $|\hat{\rho}\hat{\rho}\rangle$ is the simplest combination of dynamical variables at hand that satisfies the even-parity of $|R\rangle$ under time-reversal. Surprisingly then, we find a slow variable hidden in the memory function which we thought was governed by fast variables ! This result suggests that a pair of density modes is an appropriate candidate to construct a subspace which we can restrict the fast dynamics to. We replace the propagation in the fast-subspace by a time-propagation in the entire space of dynamical variables, but projected onto pairs of density modes. More formally, the following approximation is made $e^{i\mathcal{Q}\mathcal{L}t} \to \mathcal{P}_2 e^{i\mathcal{L}t}\mathcal{P}_2$ [55, 34] where $\mathcal{P}_2$ is a projection operator onto the subspace spanned by pairs of density modes $|\hat{\rho}_{\mathbf{q}}\hat{\rho}_{\mathbf{q}'}\rangle$:

$$\mathcal{P}_2 = \frac{1}{(2!)^2}\sum_{\mathbf{q},\ldots,\mathbf{q}'''} |\hat{\rho}_{\mathbf{q}}\hat{\rho}_{\mathbf{q}'}\rangle \langle\hat{\rho}_{\mathbf{q}}\hat{\rho}_{\mathbf{q}'} \mid \hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle^{-1} \langle\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}| \tag{26}$$

It is easy to show that $\mathcal{P}_2\mathcal{P}_2 = \mathcal{P}_2$ is indeed a projector by using the definition of the inverse (19). We emphasise that this approximation is *uncontrolled* at best, as important other important contributions from the fast-subspace could be neglected. Denoting by $G(\mathbf{q},\mathbf{q}';\mathbf{k},\mathbf{k}') \equiv \langle\hat{\rho}_{\mathbf{q}}\hat{\rho}_{\mathbf{q}'}|\hat{\rho}_{\mathbf{k}}\hat{\rho}_{\mathbf{k}'}\rangle$ static four-point structure factors and $F_4(\mathbf{q},\mathbf{q}';\mathbf{k},\mathbf{k}',t) \propto \langle\hat{\rho}_{\mathbf{q}}\hat{\rho}_{\mathbf{q}'}|\hat{\rho}_{\mathbf{k}}(t)\hat{\rho}_{\mathbf{k}'}(t)\rangle$ the associated time-dependent quantity, we may write:

$$\begin{aligned}\langle R_{\mathbf{k}_1}|\mathcal{P}_2 e^{i\mathcal{L}t}\mathcal{P}_2|R_{\mathbf{k}}\rangle =& \frac{1}{(2!)^4}\sum_{\mathbf{q}_1,\ldots,\mathbf{q}_4}\sum_{\mathbf{q}'_1,\ldots,\mathbf{q}'_4}\langle R_{\mathbf{k}_1}|\hat{\rho}_{\mathbf{q}_1}\hat{\rho}_{\mathbf{q}_2}\rangle G^{-1}(\mathbf{q}_1,\mathbf{q}_2;\mathbf{q}_3,\mathbf{q}_4)F_4(\mathbf{q}_3,\mathbf{q}_4;\mathbf{q}'_1,\mathbf{q}'_2;t)\\ &\times G^{-1}(\mathbf{q}'_1,\mathbf{q}'_2;\mathbf{q}'_3,\mathbf{q}'_4)\langle\hat{\rho}_{\mathbf{q}'_3}\hat{\rho}_{\mathbf{q}'_4}|R_{\mathbf{k}}\rangle\\ =& \frac{1}{(2!)^2}\sum_{\mathbf{q},\mathbf{q}'}\sum_{\mathbf{q}'',\mathbf{q}'''}V_{\mathbf{q}_0}(\mathbf{k}_1;\mathbf{q},\mathbf{q}')F_4(\mathbf{q},\mathbf{q}';\mathbf{q}'',\mathbf{q}''',t)V^{\dagger}_{\mathbf{q}_0}(\mathbf{k};\mathbf{q}'',\mathbf{q}''')\end{aligned} \tag{27}$$

where the following inhomogeneous vertices $V_{\mathbf{q}_0}$ have been defined:

$$\begin{aligned}V_{\mathbf{q}_0}(\mathbf{k}_1;\mathbf{q},\mathbf{q}') &= \frac{1}{2!}\sum_{\mathbf{q}'',\mathbf{q}'''}\langle R_{\mathbf{k}_1}|\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle\, G^{-1}(\mathbf{q}'',\mathbf{q}''';\mathbf{q},\mathbf{q}')\\ &\approx \frac{1}{2!N^2}\sum_{\mathbf{q}'',\mathbf{q}'''}\langle R_{\mathbf{k}_1}|\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle\left[S^{-1}(\mathbf{q}'';\mathbf{q})S^{-1}(\mathbf{q}''';\mathbf{q}')+S^{-1}(\mathbf{q}'';\mathbf{q}')S^{-1}(\mathbf{q}''';\mathbf{q})\right]\\ &= \frac{1}{N^2}\sum_{\mathbf{q}'',\mathbf{q}'''}\langle R_{\mathbf{k}_1}|\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle\left[S^{-1}(\mathbf{q}'';\mathbf{q})S^{-1}(\mathbf{q}''';\mathbf{q}')\right]\end{aligned}$$

In the second line above, we made use of *Gaussian factorisation* of the four-point static correlation functions [69]: $G(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4)\approx G_D(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4)\equiv S(\mathbf{k}_1;\mathbf{k}_3)S(\mathbf{k}_2;\mathbf{k}_4)+S(\mathbf{k}_1;\mathbf{k}_4)S(\mathbf{k}_2;\mathbf{k}_3)$. The form of the inverse G_D^{-1} follows from the definition given in (19). We can simplify it further by writing down explicitly the fluctuating force term :

$$\begin{aligned}\langle R_{\mathbf{k}_1}\mid\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle =& \frac{-iNk_BT}{m}\left[(\hat{\mathbf{k}}_1\cdot\mathbf{q}'')S^{-1}(\mathbf{k}_1-\mathbf{q}'';\mathbf{q}''')+(\mathbf{q}''\leftrightarrow\mathbf{q}''')\right]\\ &+\frac{iNk_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}\hat{\mathbf{k}}_1\cdot\mathbf{k}\phi(\mathbf{k}_1-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}')S_3(\mathbf{k}';\mathbf{q}'',\mathbf{q}''')\end{aligned} \tag{28}$$

where $S_3(\mathbf{k};\mathbf{k}',\mathbf{k}'')\equiv N^{-1}\langle\hat{\rho}_{\mathbf{k}}|\hat{\rho}_{\mathbf{k}'}\hat{\rho}_{\mathbf{k}''}\rangle$ is a 3-point static correlator. Hence, the full inhomogeneous mode coupling vertex can be written as

$$\begin{aligned}V_{\mathbf{q}_0}(\mathbf{k};\mathbf{q},\mathbf{q}') =& -\frac{ik_BT}{Nm}\sum_{\mathbf{q}'',\mathbf{q}'''}\left[(\hat{\mathbf{k}}_1\cdot\mathbf{q}'')S^{-1}(\mathbf{k}_1-\mathbf{q}'';\mathbf{q}''')+(\mathbf{q}''\leftrightarrow\mathbf{q}''')\right]S^{-1}(\mathbf{q}'';\mathbf{q})S^{-1}(\mathbf{q}''';\mathbf{q}')\\ &+\frac{ik_BT}{Nm}\sum_{\mathbf{q}'',\mathbf{q}'''}\sum_{\mathbf{k},\mathbf{k}'}\hat{\mathbf{k}}_1\cdot\mathbf{k}\phi(\mathbf{k}_1-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}')S_3(\mathbf{k}';\mathbf{q}'',\mathbf{q}''')S^{-1}(\mathbf{q}'';\mathbf{q})S^{-1}(\mathbf{q}''';\mathbf{q}')\end{aligned} \tag{29}$$

By defining a reduced vertex function :

$$\bar{V}_{\mathbf{q}_0}(\mathbf{k};\mathbf{q},\mathbf{q}') = \frac{1}{2}V_{\mathbf{q}_0}(\mathbf{k};\mathbf{q},\mathbf{q}')$$

we may write down the full e.o.m. for dynamic structure factors F_2 :

$$\boxed{\begin{aligned}0 &= \ddot{F}_2(\mathbf{k}_1;\mathbf{k}_2,t) + \nu\dot{F}_2(\mathbf{k}_1;\mathbf{k}_2,t) + \frac{k_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}(\mathbf{k}_1\cdot\mathbf{k})\phi(\mathbf{k}_1-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}')F_2(\mathbf{k}';\mathbf{k}_2,t)\\ &+\frac{k_BT}{m}\int_0^t d\tau\sum_{\mathbf{k},\mathbf{k}'}\sum_{\mathbf{q},\mathbf{q}'}\sum_{\mathbf{q}'',\mathbf{q}'''}\frac{k_1}{k'}\bar{V}_{\mathbf{q}_0}(\mathbf{k}_1;\mathbf{q},\mathbf{q}')F_4(\mathbf{q},\mathbf{q}';\mathbf{q}'',\mathbf{q}''',t-\tau)\bar{V}^{\dagger}_{\mathbf{q}_0}(\mathbf{k};\mathbf{q}'',\mathbf{q}''')J^{-1}(\mathbf{k};\mathbf{k}')\dot{F}_2(\mathbf{k}';\mathbf{k}_2,\tau)\end{aligned}} \tag{30}$$

where momentum conservation is implied : $\mathbf{k}_1 = \mathbf{k}_2 + \mathbf{q}_0$ in the statistically averaged quantities. We observe that the effect of the external field is entirely implicit in (30), which has a very similar form to that of homogeneous mode-coupling theory [20]. Indeed, all necessary information is contained within the (many-body) static structure factors which serve as initial conditions to the equation of motion. The reason for this is rooted in density functional theory, which tells us that the density profile $\rho(\mathbf{r})$ and higher order equivalents $\rho^{(n)}(\{\mathbf{r}_j\})$ (from which many-body structure factors can be derived) are *uniquely* determined by the interaction potentials between particles, and the external field. Furthermore, the reader familiar with statistical field-theoretic ideas may recognise the similarity of (30) with the Dyson equation : $F = F_0 + F * \Sigma * F_0$. The similarity with a phonon propagator in the random phase approximation (RPA) is even more striking when (30) is expressed in Laplace frequency-space, which heuristically gives $F(z) \propto [z - M(z)]^{-1}$ and where the memory kernel acts as self-energy corrections. While there exists a large number of field-theoretic approaches to the problem of glass formation, the equivalence of equations of motion obtained from truncated perturbative expansions and that obtained via projection operator techniques remains unclear [6, 5, 28, 29]

The next step to finalise the mode-coupling theory would be to render (30) self-consistent by approximating the dynamic four-point structure factor

$$F_4(\mathbf{q},\mathbf{q}';\mathbf{q}'',\mathbf{q}''',t-\tau) \approx F_2(\mathbf{q},;\mathbf{q}'',t-\tau)F_2(\mathbf{q}',;\mathbf{q}''',t-\tau) + (\mathbf{q}'' \leftrightarrow \mathbf{q}''')$$

By applying this last factorisation approximation, we recover known results previously derived by Biroli et al. [18]. However, we seek to derive an inhomogeneous Generalised Mode-Coupling theory (iGMCT), in the spirit of [61, 41]. The idea is to delay the self-consistent approximation in order to incorporate higher-order correlation functions in our framework. The incorporation and treatment of higher-order correlation functions has been central to the understanding of glass physics in the recent years. We can then follow the Mori-Zwanzig formalism once again to obtain a formally exact equation of motion for the 4-point correlation function $F_4(\mathbf{q},\mathbf{q}';\mathbf{q}'',\mathbf{q}''',t)$ by considering doublets of density modes $|\hat{\rho}_{\mathbf{q}}\hat{\rho}_{\mathbf{q}'}\rangle$ as slow dynamical variables. The equation of motion for F_4 will in turn have a memory function of its own, which to first order can be shown to be governed by 6-point correlation functions F_6 as we will see. This 6-point function will in turn be governed by an 8-point function F_8 etc. We then effectively obtain a hierarchy of coupled equations of motion for the set of even dynamic many-body density correlation functions F_{2n}. We may then imagine delaying the self consistent approximation to infinity, which would effectively wash it away. With these ideas in mind, we turn ourselves to the derivation of equations of motion for higher-order dynamic structure factors $F_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n},t)$ in the presence of a spatially varying external field from the Mori-Zwanzig formalism. We note that the derivation of an inhomogeneous GMCT is a completely novel result.

2.4 Inhomogeneous Generalised Mode Coupling Theory

2.4.1 An Explicit Equation of motion for F_4

We derive below an explicit equation of motion for the 4-point inhomogeneous dynamic correlation function: $F_4(\mathbf{k}_1, \mathbf{k}_2; \mathbf{k}_3, \mathbf{k}_4, t) = N^{-1}\langle \hat{\rho}_{\mathbf{k}_1}\hat{\rho}_{\mathbf{k}_2}|\hat{\rho}_{\mathbf{k}_3}(t)\hat{\rho}_{\mathbf{k}_4}(t)\rangle$. From the Mori-Zwanzig formalism developed for arbitrary dynamical variables, we know that (18) is correct for the following choice of dynamical variables: a density mode doublet $|A^{(1)}_{\mathbf{k},\mathbf{k}'}\rangle = |\hat{\rho}_{\mathbf{k}}\hat{\rho}_{\mathbf{k}'}\rangle$ and the associated generalised current $|A^{(2)}_{\mathbf{k}\mathbf{k}'}\rangle = -i|\dot{A}^{(1)}_{\mathbf{k},\mathbf{k}'}\rangle$. By extracting the two necessary equations:

$$\dot{C}_{11}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4; t) = \frac{1}{2!}\sum_{\mathbf{k},\mathbf{k}'} i\Omega_{12}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}, \mathbf{k}')C_{21}(\mathbf{k}, \mathbf{k}', \mathbf{k}_3, \mathbf{k}_4; t)$$

and

$$\begin{aligned}\dot{C}_{21}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4; t) &= \frac{1}{2!}\sum_{\mathbf{k},\mathbf{k}'} i\Omega_{21}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}, \mathbf{k}')C_{11}(\mathbf{k}, \mathbf{k}', \mathbf{k}_3, \mathbf{k}_4; t)\\ &- \frac{1}{2!}\sum_{\mathbf{k},\mathbf{k}'}\int dt' K_{22}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}, \mathbf{k}'; t')C_{21}(\mathbf{k}, \mathbf{k}', \mathbf{k}_3, \mathbf{k}_4; t-t')\end{aligned}$$

we can then close this last one in favour of C_{11} to eventually give

$$\begin{aligned}\ddot{C}_{11}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4; t) = &\frac{1}{(2!)^2}\sum_{\mathbf{k},\ldots,\mathbf{k}'''} i\Omega_{12}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}, \mathbf{k}')i\Omega_{21}(\mathbf{k}, \mathbf{k}', \mathbf{k}'', \mathbf{k}''')C_{11}(\mathbf{k}'', \mathbf{k}''', \mathbf{k}_3, \mathbf{k}_4; t)\\ &- \frac{1}{(2!)^3}\int dt' \sum_{\mathbf{k},\ldots,\mathbf{k}'''''} i\Omega_{12}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}, \mathbf{k}')K_{22}(\mathbf{k}, \mathbf{k}', \mathbf{k}'', \mathbf{k}'''; t')\\ &\times (i\Omega_{12})^{-1}(\mathbf{k}'', \mathbf{k}''', \mathbf{k}'''', \mathbf{k}''''')\dot{C}_{11}(\mathbf{k}'''', \mathbf{k}''''', \mathbf{k}_3, \mathbf{k}_4; t-t')\end{aligned} \tag{31}$$

From our choice of dynamical variables, we immediately have at zero time $C_{\alpha\beta} \propto \delta_{\alpha\beta}$. where we can write the matrix

$$\mathbf{C}(\mathbf{k}, \mathbf{k}'; \mathbf{q}, \mathbf{q}') = \begin{pmatrix} NG(\mathbf{k}, \mathbf{k}'; \mathbf{q}, \mathbf{q}') & 0 \\ 0 & J_4(\mathbf{k}, \mathbf{k}'; \mathbf{q}, \mathbf{q}') \end{pmatrix} \tag{32}$$

where G is the static 4-point structure factor introduced in the previous section. It will be Gaussian decomposed as custom ($G \approx G_D$). The quantity $J_4 \equiv \langle A^{(2)}|A^{(2)}\rangle$ is evaluated using similar tricks as for J_2. This essentially consists of :

$$\begin{aligned}J_4(\mathbf{k}, \mathbf{k}'; \mathbf{q}, \mathbf{q}') = \frac{1}{m\beta}\Big[&(\mathbf{k}\cdot\mathbf{q})\langle\hat{\rho}^*_{\mathbf{k}'}\hat{\rho}_{\mathbf{q}'}\hat{\rho}_{\mathbf{q}-\mathbf{k}}\rangle + (\mathbf{k}\cdot\mathbf{q}')\langle\hat{\rho}^*_{\mathbf{k}'}\hat{\rho}_{\mathbf{q}}\hat{\rho}_{\mathbf{q}'-\mathbf{k}}\rangle\\ &+ (\mathbf{k}'\cdot\mathbf{q})\langle\hat{\rho}^*_{\mathbf{k}}\hat{\rho}_{\mathbf{q}'}\hat{\rho}_{\mathbf{q}-\mathbf{k}'}\rangle + (\mathbf{k}'\cdot\mathbf{q}')\langle\hat{\rho}^*_{\mathbf{k}}\hat{\rho}_{\mathbf{q}}\hat{\rho}_{\mathbf{q}'-\mathbf{k}'}\rangle\Big]\end{aligned} \tag{33}$$

where we remark that all 3-point correlators in (33) above are of the form $\langle\hat{\rho}_{\mathbf{a}}\hat{\rho}_{\mathbf{b}}\hat{\rho}_{\mathbf{c}+\mathbf{d}}\rangle$. We factorise the terms as : $\langle\hat{\rho}_{\mathbf{a}}\hat{\rho}_{\mathbf{b}}\hat{\rho}_{\mathbf{c}+\mathbf{d}}\rangle \approx \langle\hat{\rho}_{\mathbf{c}+\mathbf{d}}\rangle\langle\hat{\rho}_{\mathbf{a}}\hat{\rho}_{\mathbf{b}}\rangle$, which is of $\mathcal{O}(N^2)$. We note that we omit terms from the decomposition that go as $\langle\hat{\rho}_{\mathbf{a}}\rangle\langle\hat{\rho}_{\mathbf{b}}\hat{\rho}_{\mathbf{c}+\mathbf{d}}\rangle$. This is due to the fact that in the zero field limit, such terms

go $\propto \delta_{\mathbf{a},0}$ which will be neglected as our theoretical development ignores all zero wave-vector modes. This is an illustration of the N-ordering argument that is often used in mode-coupling theories [57]. Hence we may write:

$$\begin{aligned} J_4(\mathbf{k},\mathbf{k}';\mathbf{q},\mathbf{q}') \approx \frac{N^2}{m\beta}\Big[&(\mathbf{k}\cdot\mathbf{q})S(\mathbf{k}';\mathbf{q}')\phi(\mathbf{k}-\mathbf{q}) + (\mathbf{k}\cdot\mathbf{q}')S(\mathbf{k}';\mathbf{q})\phi(\mathbf{k}-\mathbf{q}') \\ &+ (\mathbf{k}'\cdot\mathbf{q})S(\mathbf{k};\mathbf{q}')\phi(\mathbf{k}'-\mathbf{q}) + (\mathbf{k}'\cdot\mathbf{q}')S(\mathbf{k};\mathbf{q})\phi(\mathbf{k}'-\mathbf{q}')\Big] \end{aligned} \tag{34}$$

In turn, the frequency matrix also goes as $i\Omega_{\alpha\beta} \propto (1-\delta_{\alpha\beta})$ where: $i\Omega_{12}(\mathbf{k},\mathbf{k}';\mathbf{q},\mathbf{q}') = i\,\mathrm{Id}_2(\mathbf{k},\mathbf{k}';\mathbf{q},\mathbf{q}')$, and

$$i\Omega_{21}(\mathbf{k},\mathbf{k}';\mathbf{q},\mathbf{q}') = \frac{i}{m\beta}\sum_{\mathbf{n}}\Big((\mathbf{k}\cdot\mathbf{n})\phi(\mathbf{k}-\mathbf{n})\big(S^{-1}(\mathbf{n};\mathbf{q})\delta_{\mathbf{k}',\mathbf{q}'} + S^{-1}(\mathbf{n};\mathbf{q}')\delta_{\mathbf{k}',\mathbf{q}}\big) + (\mathbf{k}\leftrightarrow\mathbf{k}')\Big)$$

such that we may write the frequency term in (31) as

$$\begin{aligned} \frac{i^2}{2!}\sum_{\mathbf{k}'',\mathbf{k}'''}&\Omega_{21}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}'',\mathbf{k}''')C_{11}(\mathbf{k}'',\mathbf{k}''',\mathbf{k}_3,\mathbf{k}_4;t) = \\ &= -\frac{1}{m\beta}\sum_{\mathbf{n},\mathbf{q}}\Big((\mathbf{k}_1\cdot\mathbf{n})\phi(\mathbf{k}_1-\mathbf{n})S^{-1}(\mathbf{n};\mathbf{q})C_{11}(\mathbf{q},\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t) \\ &\quad + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2) \end{aligned} \tag{35}$$

We notice the important similarity of the frequency term above with that of homogeneous GMCT where it is found that $i\Omega_{21}^{(4)} \sim i\Omega_{21}^{(2)} + i\Omega_{21}^{(2)}$ [41]. Additionally, while the term seems divergent we must keep in mind that momentum conservation must be enforced on each observable. This adds strong constraints over the summations.

We now turn ourselves to the integral term. After insertion of the two $i\Omega_{12}$ in the third term of (31) on the right hand side, we eventually have

$$\frac{1}{2!}\int dt'\sum_{\mathbf{k},\mathbf{k}'}K_{22}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k},\mathbf{k}';t')\frac{\partial}{\partial t}C_{11}(\mathbf{k},\mathbf{k}',\mathbf{k}_3,\mathbf{k}_4;t-t')$$

where

$$K_{22}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4;t) = \frac{1}{2!}\sum_{\mathbf{k},\mathbf{k}'}\langle R_{\mathbf{k}_1,\mathbf{k}_2}|e^{i\mathcal{Q}\mathcal{L}t}|R_{\mathbf{k},\mathbf{k}'}\rangle\langle A^{(2)}_{\mathbf{k},\mathbf{k}'}|A^{(2)}_{\mathbf{k}_3,\mathbf{k}_4}\rangle^{-1} \tag{36}$$

with

$$|R_{\mathbf{k}_1,\mathbf{k}_2}\rangle = |\dot{A}^{(2)}_{\mathbf{k}_1,\mathbf{k}_2}\rangle - \frac{i}{(2!)^2}\sum_{\mathbf{k},\ldots,\mathbf{k}'''}\langle A^{(2)}_{\mathbf{k}_1,\mathbf{k}_2}|A^{(2)}_{\mathbf{k},\mathbf{k}'}\rangle\langle A^{(1)}_{\mathbf{k},\mathbf{k}'}|A^{(1)}_{\mathbf{k}'',\mathbf{k}'''}\rangle^{-1}|A^{(1)}_{\mathbf{k}'',\mathbf{k}'''}\rangle \tag{37}$$

the fluctuating force term associated with doublets of density modes. For analogous reasons as (24), the propagator in the fast-subspace cannot be evaluated exactly. In the spirit of MCT we look for the dominant terms in the fluctuating force. It is not hard to see that similar impulse terms arise and

that this time to leading order, $|R\rangle \sim |\hat{\rho}\hat{\rho}\hat{\rho}\rangle$: triplets of density modes. We then decide to restrict the dynamics of the 4-point dynamic correlator onto the subsapce spanned by 6-point correlators. Formally, we make the following approximation:

$$\langle R|e^{i\mathcal{Q}\mathcal{L}\mathcal{Q}t}|R\rangle \to \langle R|\mathcal{P}_3 e^{i\mathcal{L}t}\mathcal{P}_3|R\rangle \tag{38}$$

with $\mathcal{P}_3$ a projector on density triplets. Contrary to homogeneous GMCT where the projection is made on a set of two dominant density triplets denoted $|\alpha_{\mathbf{n}}\rangle, |\beta_{\mathbf{n}}\rangle$ [41], we consider a projection onto the subspace of **all** density triplets $|\gamma_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3}\rangle \equiv |\hat{\rho}_{\mathbf{n}_1}\hat{\rho}_{\mathbf{n}_2}\hat{\rho}_{\mathbf{n}_3}\rangle$. We show in Sec-2.3 that $\mathcal{P}_3$ reduces to the homogeneous triplet projection in an appropriate vanishing field limit. The projector then reads:

$$\mathcal{P}_3 = \frac{1}{(3!)^2}\sum_{\{\mathbf{n}_j\},\{\mathbf{n}'_j\}} |\gamma_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3}\rangle \left\langle\gamma_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3}\middle|\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}\right\rangle^{-1} \left\langle\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}\right| \tag{39}$$

where we naturally have $\mathcal{P}_3\mathcal{P}_3 = \mathcal{P}_3$. We may then write the projected integral kernel:

$$\langle R_{\mathbf{k}_1,\mathbf{k}_2}|\mathcal{P}_3 e^{i\mathcal{L}t}\mathcal{P}_3|R_{\mathbf{k},\mathbf{k}'}\rangle = \frac{1}{(3!)^2}\sum_{\{\mathbf{n}_j\},\{\mathbf{n}'_j\}} \mathcal{V}_{\mathbf{q}_0}(\mathbf{k}_1,\mathbf{k}_2;\{\mathbf{n}_j\}) \left\langle\gamma_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3}\middle|\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}(t)\right\rangle \mathcal{V}^{\dagger}_{\mathbf{q}_0}(\mathbf{k},\mathbf{k}';\{\mathbf{n}'_j\}) \tag{40}$$

where a 'generalised' vertex analogous to the iMCT case can be defined:

$$\begin{aligned}
\mathcal{V}_{\mathbf{q}_0}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3) &= \frac{1}{3!}\sum_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3} \langle R_{\mathbf{k}_1,\mathbf{k}_2}|\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}\rangle\langle\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}|\gamma_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3}\rangle^{-1} \\
&= \frac{1}{3!}\sum_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3} \langle \dot{A}^{(2)}_{\mathbf{k}_1,\mathbf{k}_2}|\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}\rangle\langle\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}|\gamma_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3}\rangle^{-1} \\
&\quad + \frac{1}{3!}\sum_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3} \frac{ik_BT}{m}\sum_{\mathbf{k}'',\mathbf{k}'''}\sum_{\mathbf{n}} \big((\mathbf{k}_1\cdot\mathbf{n})\phi(\mathbf{k}_1-\mathbf{n})S^{-1}(\mathbf{n};\mathbf{k}'')\delta_{\mathbf{k}_2,\mathbf{k}'''} + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2)\big) \\
&\qquad\qquad \times \langle A^{(1)}_{\mathbf{k}'',\mathbf{k}'''}|\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}\rangle\langle\gamma_{\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3}|\gamma_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3}\rangle^{-1}
\end{aligned} \tag{41}$$

We notice that the quantity $\langle\gamma|\gamma\rangle$ is a 6-point static density correlation function, which we Gaussian factorise into 6 terms : $\langle\gamma|\gamma\rangle \sim \sum_\sigma N^3SSS$. We also apply the usual set of approximations on the other static correlation functions. Lengthy algebraic manipulations eventually lead to

$$\begin{aligned}
\mathcal{V}_{\mathbf{q}_0}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3) &\approx \frac{-6ik_BT}{mN}\sum_{\mathbf{n}}(\mathbf{k}_1\cdot\mathbf{n})S^{-1}(\mathbf{n};\mathbf{n}_1)\delta_{\mathbf{k}_1-\mathbf{n},\mathbf{n}_2}\delta_{\mathbf{k}_2,\mathbf{n}_3} \\
&\quad + \frac{ik_BT}{mN^2}\sum_{\mathbf{n}'_1,\ldots,\mathbf{n}'_3}\sum_{\mathbf{k}'',\mathbf{n}}\Big((\mathbf{k}_1\cdot\mathbf{n})\phi(\mathbf{k}_1-\mathbf{n})S^{-1}(\mathbf{n};\mathbf{k}'')S_5(\mathbf{k}'',\mathbf{k}_2;\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3) \\
&\qquad\qquad \times S^{-1}(\mathbf{n}'_1;\mathbf{n}_1)S^{-1}(\mathbf{n}'_2;\mathbf{n}_2)S^{-1}(\mathbf{n}'_3;\mathbf{n}_3)\Big) \\
&\quad + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2)
\end{aligned} \tag{42}$$

While the first and second terms of (42) above seem to differ by a factor of $1/N$, both go as $\mathcal{O}(1)$ in the thermodynamic limit, and thus must be regarded on equal footings. Hence, the full equation of motion for the 4-point dynamic structure factor F_4 can be written in full as:

$$\boxed{\begin{aligned}0 &= \ddot{F}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t) + \nu\dot{F}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t) + \frac{k_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}(\mathbf{k}_1\cdot\mathbf{k})\phi(\mathbf{k}_1-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}')F_4(\mathbf{k}',\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)\\ &+ \frac{k_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}(\mathbf{k}_2\cdot\mathbf{k})\phi(\mathbf{k}_2-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}')F_4(\mathbf{k}_1,\mathbf{k}';\mathbf{k}_3,\mathbf{k}_4,t)\\ &+ \frac{N}{(2!)^2(3!)^2}\sum_{\{\mathbf{n}_j\},\{\mathbf{n}'_j\}}\sum_{\mathbf{k},\ldots,\mathbf{k}'''}\int_0^\tau \mathcal{V}_{\mathbf{q}_0}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)F_6(\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3;\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3,t-\tau)\mathcal{V}^\dagger_{\mathbf{q}_0}(\mathbf{k},\mathbf{k}';\mathbf{n}'_1,\mathbf{n}'_2,\mathbf{n}'_3)\\ &\qquad\times J_4^{-1}(\mathbf{k},\mathbf{k}';\mathbf{k}'',\mathbf{k}''')\dot{F}_4(\mathbf{k}'',\mathbf{k}''';\mathbf{k}_3,\mathbf{k}_4,\tau)\end{aligned}} \tag{43}$$

We would like to emphasise that this is the first fully microscopic equation of motion for a dynamic 4-point correlation function of density modes in the presence of a spatially varying external field, and provides way to go beyond iMCT which is ultimately still cluttered by the same issues as its homogeneous counterpart. This is an important first step towards the development of an iGMCT. We will see later that a variation of (43) with respect to the conjugate field will in turn give an equation of motion for the 5-point susceptibility χ_5.

2.4.2 Generalisation to Arbitrary Order

Having derived the first two levels of the hierarchy explicitly, we are in position to write down the iGMCT equation of motion at *any* hierarchy level provided that : a) the chosen slow variables are always density multiplets and their associated currents, b) the memory kernel is projected onto the $(n+1)$-th density multiplet. Indeed it is not hard to show inductively that the memory kernel in the equation of motion for the $2n$-th dynamic correlator is always proportional to $2(n+1)$-th collective modes analogously to [41]. Let us adopt the general notation $|A^{(1)}_{\mathbf{k}_1,\ldots,\mathbf{k}_n}\rangle \equiv |\hat{\rho}_{\mathbf{k}_1}\ldots\hat{\rho}_{\mathbf{k}_n}\rangle$ for density multiplets and $|A^{(2)}_{\mathbf{k}_1,\ldots,\mathbf{k}_n}\rangle \equiv -i|\dot{A}^{(1)}_{\mathbf{k}_1,\ldots,\mathbf{k}_n}\rangle$ for the associated current. Then, the equation of motion for the many body dynamic structure factor $F_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n},t) \equiv N^{-1}\langle A^{(1)}_{\mathbf{k}_1,\ldots,\mathbf{k}_n}|A^{(1)}_{\mathbf{k}_{n+1},\ldots,\mathbf{k}_{2n}}(t)\rangle$ reads:

$$\boxed{\begin{aligned}0 =& \ddot{F}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n},t) + \frac{1}{n!}\sum_{\{\mathbf{k}'_j\}}\Omega_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}'_1,...,\mathbf{k}'_n)F_{2n}(\mathbf{k}'_1,...,\mathbf{k}'_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n})\\ &+\nu_{2n}\dot{F}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n})\\ &+\frac{1}{(n!)^2}\sum_{\{\mathbf{k}'_j\},\{\mathbf{k}''_j\}}\int_0^t d\tau\mathcal{M}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}'_1,...,\mathbf{k}'_n,t-\tau)J^{-1}_{2n}(\mathbf{k}'_1,...,\mathbf{k}'_n;\mathbf{k}''_1,...,\mathbf{k}''_n)\\ &\qquad\times\dot{F}_{2n}(\mathbf{k}''_1,...,\mathbf{k}''_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n},\tau)\end{aligned}} \tag{44}$$

where the following constraint must be satisfied $\sum_{j=1}^n \mathbf{k}_j - \sum_{j=n+1}^{2n}\mathbf{k}_j - \mathbf{q}_0 = \mathbf{0}$. The bare frequency term may be written as:

$$\begin{aligned}\Omega_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{q}_1,...,\mathbf{q}_n) &= \frac{1}{n!}\sum_{\{\mathbf{p}_j\}}\langle A^{(2)}_{\mathbf{k}_1,...,\mathbf{k}_n}|\dot{A}^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_n}\rangle\langle A^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_n}|A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_n}\rangle^{-1}\\ &\approx \frac{1}{n!}\sum_{\{\mathbf{p}_j\}}\langle \dot{A}^{(1)}_{\mathbf{k}_1,...,\mathbf{k}_n}|\dot{A}^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_n}\rangle\langle A^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_n}|A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_n}\rangle^{-1}\\ &= \frac{k_BT}{n!m}\sum_{\{\mathbf{p}_j\}}\big((\mathbf{k}_1\cdot\mathbf{p}_1)\langle\hat{\rho}_{\mathbf{k}_1-\mathbf{p}_1}\hat{\rho}_{\mathbf{k}_2}...\hat{\rho}_{\mathbf{k}_n}|\hat{\rho}_{\mathbf{p}_2}...\hat{\rho}_{\mathbf{p}_n}\rangle + (\mathbf{p}_1\leftrightarrow\mathbf{p}_2) + ... + (\mathbf{p}_1\leftrightarrow\mathbf{p}_n)\big)\\ &\qquad\times\langle\hat{\rho}_{\mathbf{p}_1}...\hat{\rho}_{\mathbf{p}_n}|\hat{\rho}_{\mathbf{q}_1}...\hat{\rho}_{\mathbf{q}_n}\rangle^{-1} + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2) + ... + (\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\end{aligned}$$

We now Gaussian factorise the inverse n-point static correlator as usual, and employ an N-ordering argument for the static $(n-1)$-th dimension correlator. More precisely, we keep only $S_{n-1}\approx\phi S...S\sim\mathcal{O}(N^n)$ in its factorisation. It is easy to see that factorisation into products of many-body structure factors (>2) leads to a quantity of order $\mathcal{O}(N^m)$, $m<n$. Then, writing explicitly the $\mathbf{k}_1$-term only and using n-fold permutation symmetries over dummy summation variables $\{\mathbf{p}_j\}$, we find

$$\begin{aligned}&\approx \frac{k_BTn(n-1)!n!NN^{n-1}}{n!mN^n}\sum_{\{\mathbf{p}_j\}}(\mathbf{k}_1\cdot\mathbf{p}_1)\phi(\mathbf{k}_1-\mathbf{p}_1)S(\mathbf{k}_2;\mathbf{p}_2)\times...\times S(\mathbf{k}_n;\mathbf{p}_n)S^{-1}(\mathbf{p}_1;\mathbf{q}_1)\times...\times S^{-1}(\mathbf{p}_n;\mathbf{q}_n)\\ &\qquad + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2) + ... + (\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\\ &= \frac{k_BTn!}{m}\sum_{\mathbf{p}_1}(\mathbf{k}_1\cdot\mathbf{p}_1)\phi(\mathbf{k}_1-\mathbf{p}_1)S^{-1}(\mathbf{p}_1;\mathbf{q}_1)\delta_{\mathbf{k}_2,\mathbf{q}_2}\delta_{\mathbf{k}_3,\mathbf{q}_3}\times...\times\delta_{\mathbf{k}_n,\mathbf{q}_n} + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2) + ... + (\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\end{aligned}\tag{45}$$

Whence we can write in full that

$$\begin{aligned}&\frac{1}{n!}\sum_{\{\mathbf{k}'_j\}}\Omega_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}'_1,...,\mathbf{k}'_n)F_{2n}(\mathbf{k}'_1,...,\mathbf{k}'_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n}) =\\ &\qquad = \frac{k_BT}{m}\sum_{\mathbf{p},\mathbf{k}'}(\mathbf{k}_1\cdot\mathbf{p})\phi(\mathbf{k}_1-\mathbf{p})S^{-1}(\mathbf{p};\mathbf{k}')F_{2n}(\mathbf{k}',\mathbf{k}_2,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n})\\ &\qquad + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2) + ... + (\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\end{aligned}\tag{46}$$

We then see that analogously to the homogeneous GMCT bare frequencies, the inhomogeneous GMCT has a bare frequency term of arbitrary order $2n$ that can essentially be written as a sum of n bare frequencies of order $2:\ \Omega_{2n}\sim\Omega_2+...+\Omega_2$ [41]. Moving onto the memory term $\mathcal{M}$, it can be written as:

$$\begin{aligned}\mathcal{M}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}'_1,...,\mathbf{k}'_n,t) &= \langle R_{\mathbf{k}_1,...,\mathbf{k}_n}|e^{i\mathcal{Q}\mathcal{L}t}|R_{\mathbf{k}'_1,...,\mathbf{k}'_n}\rangle\\ &\approx \langle R_{\mathbf{k}_1,...,\mathbf{k}_n}|\mathcal{P}_{n+1}e^{i\mathcal{L}t}\mathcal{P}_{n+1}|R_{\mathbf{k}'_1,...,\mathbf{k}'_n}\rangle\end{aligned}\tag{47}$$

where we have defined the projector on the $(n+1)$-th set of density modes:

$$\begin{aligned}\mathcal{P}_{n+1} &= \frac{1}{((n+1)!)^2} \sum_{\{\mathbf{p}_j\},\{\mathbf{p}'_j\}} |A^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_{n+1}}\rangle\langle A^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_{n+1}}|A^{(1)}_{\mathbf{p}'_1,...,\mathbf{p}'_{n+1}}\rangle^{-1}\langle A^{(1)}_{\mathbf{p}'_1,...,\mathbf{p}'_{n+1}}| \\ &\approx \frac{1}{(n+1)!N^{n+1}} \sum_{\{\mathbf{p}_j\},\{\mathbf{p}'_j\}} |A^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_{n+1}}\rangle S^{-1}(\mathbf{p}_1;\mathbf{p}'_1) \times ... \times S^{-1}(\mathbf{p}_{n+1};\mathbf{p}'_{n+1})\langle A^{(1)}_{\mathbf{p}'_1,...,\mathbf{p}'_{n+1}}|\end{aligned}$$

We recall from the generalised Langevin equation (12) the definition of the arbitrary second element of the fluctuating force vector:

$$\begin{aligned}|R_{\mathbf{k}_1,...,\mathbf{k}_n}\rangle &= |\dot{A}^{(2)}_{\mathbf{k}_1,...,\mathbf{k}_n}\rangle - \frac{1}{n!}\sum_{\{\mathbf{q}_j\}} i\Omega_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{q}_1,...,\mathbf{q}_n)|A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_n}\rangle \\ &= |R_1(\mathbf{k}_1,...,\mathbf{k}_n)\rangle + |R_2(\mathbf{k}_1,...,\mathbf{k}_n)\rangle\end{aligned} \tag{48}$$

Hence, the memory term can be written as

$$\begin{aligned}&\mathcal{M}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}'_1,...,\mathbf{k}'_n,t) \\ &= \frac{N}{((n+1)!)^2} \sum_{\{\mathbf{p}_j\},\{\mathbf{p}'_j\}} \mathcal{V}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{p}_1,...,\mathbf{p}_{n+1})F_{2(n+1)}(\mathbf{p}_1,...,\mathbf{p}_{n+1};\mathbf{p}'_1,...,\mathbf{p}'_{n+1},t) \\ &\qquad \times \mathcal{V}^{\dagger}_{2n}(\mathbf{k}'_1,...,\mathbf{k}'_n;\mathbf{p}'_1,...,\mathbf{p}'_{n+1})\end{aligned} \tag{49}$$

where the generalised vertex function reads

$$\begin{aligned}\mathcal{V}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{p}_1,...,\mathbf{p}_{n+1}) &= \frac{1}{(n+1)!} \sum_{\{\mathbf{q}_j\}} \langle R_{\mathbf{k}_1,...,\mathbf{k}_n}|A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_{n+1}}\rangle\langle A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_{n+1}}|A^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_{n+1}}\rangle^{-1} \\ &\approx \frac{1}{N^{n+1}} \sum_{\{\mathbf{q}_j\}} \langle R_{\mathbf{k}_1,...,\mathbf{k}_n}|A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_{n+1}}\rangle S^{-1}(\mathbf{q}_1;\mathbf{p}_1) \times ... \times S^{-1}(\mathbf{q}_{n+1};\mathbf{p}_{n+1})\end{aligned} \tag{50}$$

We can evaluate the term $\langle R|A\rangle \equiv \langle R_1|A\rangle + \langle R_2|A\rangle$ by construction using the same set of tricks used in prior parts of this work. For the first term $\langle R_1|A\rangle$, we only write the $\mathbf{k}_1$ element, the others can be written by mere variable exchange. This gives:

$$
\begin{aligned}
&\frac{1}{(n+1)!}\sum_{\{\mathbf{q}_j\}}\langle \dot{A}^{(2)}_{\mathbf{k}_1,...,\mathbf{k}_n}|A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_{n+1}}\rangle\langle A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_{n+1}}|A^{(1)}_{\mathbf{p}_1,...,\mathbf{p}_{n+1}}\rangle^{-1}\\
=&\frac{1}{N^{n+1}}\sum_{\{\mathbf{q}_j\}}\langle \dot{A}^{(2)}_{\mathbf{k}_1,...,\mathbf{k}_n}|A^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_{n+1}}\rangle S^{-1}(\mathbf{q}_1;\mathbf{p}_1)\times ...\times S^{-1}(\mathbf{q}_{n+1};\mathbf{p}_{n+1})\\
=&\frac{-i}{N^{n+1}}\sum_{\{\mathbf{q}_j\}}\langle \dot{A}^{(1)}_{\mathbf{k}_1,...,\mathbf{k}_n}|\dot{A}^{(1)}_{\mathbf{q}_1,...,\mathbf{q}_{n+1}}\rangle S^{-1}(\mathbf{q}_1;\mathbf{p}_1)\times ...\times S^{-1}(\mathbf{q}_{n+1};\mathbf{p}_{n+1})\\
=&\frac{-ik_BT}{mN^{n+1}}\sum_{\{\mathbf{q}_j\}}\Big((\mathbf{k}_1\cdot\mathbf{q}_1)\langle\hat{\rho}_{\mathbf{k}_1-\mathbf{q}_1}\hat{\rho}_{\mathbf{k}_2}...\hat{\rho}_{\mathbf{k}_n}|\hat{\rho}_{\mathbf{q}_2}...\hat{\rho}_{\mathbf{q}_{n+1}}\rangle+(\mathbf{q}_1\leftrightarrow\mathbf{q}_2)+...+(\mathbf{q}_1\leftrightarrow\mathbf{q}_{n+1})\Big)\\
&\qquad\times S^{-1}(\mathbf{q}_1;\mathbf{p}_1)\times ...\times S^{-1}(\mathbf{q}_{n+1};\mathbf{p}_{n+1})+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...+(\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\\
=&\frac{-ik_BT(n+1)}{mN^{n+1}}\sum_{\{\mathbf{q}_j\}}(\mathbf{k}_1\cdot\mathbf{q}_1)\langle\hat{\rho}_{\mathbf{k}_1-\mathbf{q}_1}\hat{\rho}_{\mathbf{k}_2}...\hat{\rho}_{\mathbf{k}_n}|\hat{\rho}_{\mathbf{q}_2}...\hat{\rho}_{\mathbf{q}_{n+1}}\rangle S^{-1}(\mathbf{q}_1;\mathbf{p}_1)\times ...\times S^{-1}(\mathbf{q}_{n+1};\mathbf{p}_{n+1})+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...\\
\approx&\frac{-ik_BT(n+1)n!N^n}{mN^{n+1}}\sum_{\{\mathbf{q}_j\}}(\mathbf{k}_1\cdot\mathbf{q}_1)S(\mathbf{k}_1-\mathbf{q}_1;\mathbf{q}_2)S(\mathbf{k}_2;\mathbf{q}_3)\times ...\times S(\mathbf{k}_n;\mathbf{k}_{n+1})S^{-1}(\mathbf{q}_1;\mathbf{p}_1)\times ...\\
&\qquad\times S^{-1}(\mathbf{q}_{n+1};\mathbf{p}_{n+1})+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...+(\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\\
=&\frac{-ik_BT(n+1)!}{mN}\sum_{\mathbf{q}_1}(\mathbf{k}_1\cdot\mathbf{q}_1)S^{-1}(\mathbf{q}_1;\mathbf{p}_1)\delta_{\mathbf{k}_1-\mathbf{q}_1,\mathbf{p}_2}\delta_{\mathbf{k}_2,\mathbf{p}_3}\times ...\times\delta_{\mathbf{k}_n,\mathbf{p}_{n+1}}+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...+(\mathbf{k}_1\leftrightarrow\mathbf{k}_n)
\end{aligned}
\tag{51}
$$

where in the 6-th line we have Gaussian decomposed the $2n$-point static structure factor, we then systematically apply the identity (19) for the static structure factors. For the second term $\langle R_2|A\rangle$, making use of the bare frequency result (46) we may write

$$
\begin{aligned}
&\frac{ik_BT}{m(n+1)!}\sum_{\{\mathbf{t}'_j\}}\sum_{\mathbf{p},\mathbf{q}}\Big((\mathbf{k}_1\cdot\mathbf{p})\phi(\mathbf{k}_1-\mathbf{p})S^{-1}(\mathbf{p};\mathbf{q})\langle A^{(1)}_{\mathbf{q},\mathbf{k}_2...\mathbf{k}_n}|A^{(1)}_{\mathbf{t}'_1,...,\mathbf{t}'_{n+1}}\rangle\langle A^{(1)}_{\mathbf{t}'_1,...,\mathbf{t}'_{n+1}}|A^{(1)}_{\mathbf{t}_1,...,\mathbf{t}_{n+1}}\rangle^{-1}\Big)\\
&\qquad+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...+(\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\\
=&\frac{ik_BT}{mN^n}\sum_{\{\mathbf{t}'_j\}}\sum_{\mathbf{p},\mathbf{q}}(\mathbf{k}_1\cdot\mathbf{p})\phi(\mathbf{k}_1-\mathbf{p})S^{-1}(\mathbf{p};\mathbf{q})S_{2n+1}(\mathbf{q},\mathbf{k}_2,...,\mathbf{k}_n;\mathbf{t}'_1,...,\mathbf{t}'_{n+1})S^{-1}(\mathbf{t}'_1;\mathbf{t}_1)\times ...\times S^{-1}(\mathbf{t}'_{n+1};\mathbf{t}_{n+1})\\
&\qquad+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...+(\mathbf{k}_1\leftrightarrow\mathbf{k}_n)
\end{aligned}
\tag{52}
$$

Whence, in full the arbitrary vertex can be written as :

$$\mathcal{V}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{p}_1,...,\mathbf{p}_{n+1}) = \frac{-ik_BT}{m}\Bigg[\frac{(n+1)!}{N}\sum_{\mathbf{q}}(\mathbf{k}_1\cdot\mathbf{q})S^{-1}(\mathbf{q};\mathbf{p}_1)\delta_{\mathbf{k}_1-\mathbf{q},\mathbf{p}_2}\delta_{\mathbf{k}_2,\mathbf{p}_3}\times...\times\delta_{\mathbf{k}_n,\mathbf{p}_{n+1}}$$
$$-\frac{1}{N^n}\sum_{\{\mathbf{t}'_j\}}\sum_{\mathbf{p}',\mathbf{q}'}(\mathbf{k}_1\cdot\mathbf{p}')\phi(\mathbf{k}_1-\mathbf{p}')S^{-1}(\mathbf{p}';\mathbf{q}')S_{2n+1}(\mathbf{q}',\mathbf{k}_2,...,\mathbf{k}_n;\mathbf{t}'_1,...,\mathbf{t}'_{n+1})$$
$$\times S^{-1}(\mathbf{t}'_1;\mathbf{p}_1)\times...\times S^{-1}(\mathbf{t}'_{n+1};\mathbf{p}_{n+1})\Bigg]$$
$$+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...+(\mathbf{k}_1\leftrightarrow\mathbf{k}_n) \tag{53}$$

The last term to be evaluated is the inverse current $J^{-1}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n}) \equiv \langle A^{(2)}_{\mathbf{k}_1,...,\mathbf{k}_n}|A^{(2)}_{\mathbf{k}_{n+1},...,\mathbf{k}_{2n}}\rangle^{-1}$. Due to the number of terms arising from both the use of equipartition and that of factorisation ansatz, a closed form satisfying (19) could not be found. We suggest instead to find the inverse numerically if one desires to solve these equations.

The general coupled hierarchy of equations of motions that (44) defines for the many-body dynamic structure factors F_{2n} is to our knowledge the most general inhomogeneous GMCT that can be written. Indeed, the inclusion of a spatially varying external field forces the framework to consider all possible couplings between the modes which in this case presents itself with the large number of running summations over **k**-space. This hierarchy also provides a good starting point to develop a GMCT for geometrically constrained systems, along the lines of [47, 59]. We point out however that in the case of extreme geometric confinements it could be useful to decompose the set of slow variables into longitudinal and transverse Fourier density modes.

2.5 Off-Diagonal Homogeneous Generalised Mode Coupling Theory

In order to make sure that the inhomogeneous theory developed in the previous sections is coherent and consistent, we take the zero-field limit $U(\mathbf{q}_0)\to 0$ from which we should recover standard (diagonal) GMCT [41] within a suitable set of approximations. Furthermore, this zero field limit also provides a consistent way of deriving a more complete version of homogeneous GMCT which is referred to as off-diagonal GMCT. The GMCT derived in [41] indeed only develops equations of motion for the diagonalised many-density correlation functions $F_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_1,...,\mathbf{k}_n,t)$, and does not include the incorporation of off-diagonal terms. The most general many-density correlation function is given by $F_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n},t)$ with the only requirement of momentum conservation: $\sum_{j=1}^{n}\mathbf{k}_j - \sum_{j=n+1}^{2n}\mathbf{k}_j = 0$. We see that taking the zero-field limit of (44) and simply enforcing momentum conservation gives direct access to this off-diagonal GMCT. We mention that to our knowledge only one attempt to the derivation of this off-diagonal GMCT exists for the 4-point dynamic correlation function F_4 [25]. We note however that their approach is different than ours.

2.5.1 Recovering Mode-Coupling Theory

We consider the zero-field limit of the inhomogenous MCT given by (30). We first note that the two derivative terms are trivial since $\lim_{U\to 0}F_2(\mathbf{k};\mathbf{k}',t) = F_2(\mathbf{k},t)\delta_{\mathbf{k},\mathbf{k}'}$ where we abuse notation slightly. The frequency term reads

$$\begin{aligned}
&\lim_{U\to 0}\frac{k_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}(\mathbf{k}_1\cdot\mathbf{k})\,\phi\left(\mathbf{k}_1-\mathbf{k}\right)S^{-1}\left(\mathbf{k};\mathbf{k}'\right)F_2\left(-\mathbf{k}';\mathbf{k}_2,t\right)\\
&=\frac{k_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}(\mathbf{k}_1\cdot\mathbf{k})\,\phi\left(\mathbf{k}_1-\mathbf{k}\right)S^{-1}\left(\mathbf{k};\mathbf{k}'\right)F_2\left(\mathbf{k}';\mathbf{k}_2,t\right)\delta_{\mathbf{k},\mathbf{k}'}\delta_{\mathbf{k}_1,\mathbf{k}}\delta_{\mathbf{k}_2,\mathbf{k}'}\\
&=\frac{k_BT}{m}\frac{|\mathbf{k}_1|^2}{S\left(\mathbf{k}_1\right)}F_2\left(\mathbf{k}_1,t\right)
\end{aligned}\tag{54}$$

which is the standard MCT-result. We then look at the individual terms of the vertices individually. The zero-field vertices read :

$$\begin{aligned}
\lim_{U\to 0}V_{\mathbf{q}_0}\left(\mathbf{k};\mathbf{q},\mathbf{q}'\right)&=\lim_{U\to 0}\frac{1}{2!}\sum_{\mathbf{q}'',\mathbf{q}'''}\langle R_{\mathbf{k}_1}\mid\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle G_D^{-1}\left(\mathbf{q}'',\mathbf{q}''';\mathbf{q},\mathbf{q}'\right)\\
&=\lim_{U\to 0}\frac{1}{2!}\sum_{\mathbf{q}'',\mathbf{q}'''}\langle\dot{A}^{(2)}_{\mathbf{k}_1}\mid\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle G_D^{-1}\left(\mathbf{q}'',\mathbf{q}''';\mathbf{q},\mathbf{q}'\right)\\
&\quad-\lim_{U\to 0}\frac{1}{2!}\sum_{\mathbf{q}'',\mathbf{q}'''}\sum_{\mathbf{k}}i\Omega_{21}\left(\mathbf{k}_1,\mathbf{k}\right)\langle A^{(1)}_{\mathbf{k}}\mid\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle G_D^{-1}\left(\mathbf{q}'',\mathbf{q}''';\mathbf{q},\mathbf{q}'\right)
\end{aligned}$$

where it is possible to show that

$$\lim_{U\to 0}\frac{1}{2!}\sum_{\mathbf{q}'',\mathbf{q}'''}\langle\dot{A}^{(2)}_{\mathbf{k}_1}\mid\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle G_D^{-1}\left(\mathbf{q}'',\mathbf{q}''';\mathbf{q},\mathbf{q}'\right)=-\frac{ik_BT}{Nm}\left[\frac{\hat{\mathbf{k}}_1\cdot\mathbf{q}}{S(\mathbf{q})}\delta_{\mathbf{k}_1-\mathbf{q},\mathbf{q}'}+\frac{\hat{\mathbf{k}}_1\cdot\mathbf{q}'}{S\left(\mathbf{q}'\right)}\delta_{\mathbf{k}_1-\mathbf{q}',\mathbf{q}}\right]$$

as well as

$$\lim_{U\to 0}\frac{1}{2!}\sum_{\mathbf{q}'',\mathbf{q}'''}\sum_{\mathbf{k}}i\Omega_{21}\left(\mathbf{k}_1,\mathbf{k}\right)\langle A^{(1)}_{\mathbf{k}}\mid\hat{\rho}_{\mathbf{q}''}\hat{\rho}_{\mathbf{q}'''}\rangle G_D^{-1}\left(\mathbf{q}'',\mathbf{q}''';\mathbf{q},\mathbf{q}'\right)=\frac{ik_BT}{Nm}\left|\mathbf{k}_1\right|$$

where we have used the convolution approximation for S_3 [8]. We then find that

$$\lim_{U\to 0}V_{\mathbf{q}_0}\left(\mathbf{k}_1;\mathbf{q},\mathbf{q}'\right)=V\left(\mathbf{k}_1;\mathbf{q},\mathbf{q}'\right)=-\frac{ik_BT}{Nm}\left[\frac{\hat{\mathbf{k}}_1\cdot\mathbf{q}}{S(|\mathbf{q}|)}+\frac{\hat{\mathbf{k}}_1\cdot\mathbf{q}'}{S\left(|\mathbf{q}'|\right)}-|\mathbf{k}_1|\right]\delta_{\mathbf{k}_1,\mathbf{q}+\mathbf{q}'}\tag{55}$$

which corresponds to the well-known MCT-vertex function [34] (up to a factor of 1/2 which is present in front of the time-integral as a symmetry factor). Focusing now on the integral term as a whole, we write:

$$\begin{aligned}
&\frac{1}{(2!)^2}\int_0^t d\tau\sum_{\mathbf{k},\mathbf{k}'}\sum_{\mathbf{q},\ldots,\mathbf{q}'''}\frac{k_1}{k'}V(\mathbf{k}_1;\mathbf{q},\mathbf{q}')\delta_{\mathbf{k}_1,\mathbf{q}+\mathbf{q}'}F_4(\mathbf{q},\mathbf{q}';\mathbf{q}'',\mathbf{q}''',\tau)\delta_{\mathbf{q}+\mathbf{q}',\mathbf{q}''+\mathbf{q}'''}V^\dagger(\mathbf{k}';\mathbf{q}'',\mathbf{q}''')\delta_{\mathbf{k}',\mathbf{q}''+\mathbf{q}'''}\\
&\qquad\times J^{-1}(\mathbf{k})\delta_{\mathbf{k},\mathbf{k}'}\dot{F}_2(\mathbf{k}_2,t-\tau)\delta_{\mathbf{k}',\mathbf{k}_2}\\
&=\frac{1}{4}\int_0^t d\tau\sum_{\mathbf{q},\mathbf{q}''}\frac{k_1}{k_2}V(\mathbf{k}_1;\mathbf{q},\mathbf{k}_1-\mathbf{q})F_4(\mathbf{q},\mathbf{k}_1-\mathbf{q};\mathbf{q}'',\mathbf{k}_2-\mathbf{q}'',\tau)\delta_{\mathbf{k}_1,\mathbf{k}_2}V^\dagger(\mathbf{k}_2;\mathbf{q}'',\mathbf{k}_2-\mathbf{q}'')\\
&\qquad\times J^{-1}(\mathbf{k}_2)\dot{F}_2(\mathbf{k}_2,t-\tau)
\end{aligned}$$

Whence, the full equation of motion in the vanishing field limit, where we slightly abuse notation writing $F_2(\mathbf{k}_1;\mathbf{k}_1,t) \equiv F_2(\mathbf{k}_1,t)$:

$$\begin{aligned}&\ddot{F}_2(\mathbf{k}_1,t) + \frac{k_BT|\mathbf{k}_1|^2}{mS(\mathbf{k}_1)}F_2(\mathbf{k}_1,t) + \nu\dot{F}_2(\mathbf{k}_1,t)\\&+\frac{1}{4}\int_0^t d\tau \sum_{\mathbf{q},\mathbf{q}'} V(\mathbf{k}_1;\mathbf{q},\mathbf{k}_1-\mathbf{q})F_4(\mathbf{q},\mathbf{k}_1-\mathbf{q};\mathbf{q}',\mathbf{k}_1-\mathbf{q}',\tau)V^\dagger(\mathbf{k}_1;\mathbf{q}',\mathbf{k}_1-\mathbf{q}')J^{-1}(\mathbf{k}_1)\dot{F}_2(\mathbf{k}_1,t-\tau) = 0\end{aligned} \tag{56}$$

To recover standard diagonal MCT, we use the diagonal approximation [40] which enforces $\mathbf{q} = \mathbf{q}'$. Lastly noting that $J^{-1}(\mathbf{k}) = m/Nk_BT$, we eventually obtain after absorbing the 1/4 factor inside the vertices:

$$\boxed{\begin{aligned}&\ddot{F}_2(\mathbf{k}_1,t) + \frac{k_BT|\mathbf{k}_1|^2}{mS(\mathbf{k}_1)}F_2(\mathbf{k}_1,t)\\&+\frac{m}{Nk_BT}\int_0^t d\tau \sum_{\mathbf{q}} |V(\mathbf{k}_1;\mathbf{q},\mathbf{k}_1-\mathbf{q})|^2 F_4(\mathbf{q},\mathbf{k}_1-\mathbf{q};\mathbf{q},\mathbf{k}_1-\mathbf{q},\tau)\dot{F}_2(\mathbf{k}_1,t-\tau) = 0\end{aligned}} \tag{57}$$

which is the standard result before the self-consistent approximation $F_4(\mathbf{q},\mathbf{k}_1-\mathbf{q};\mathbf{q},\mathbf{k}_1-\mathbf{q},t) \approx F_2(\mathbf{k}_1-\mathbf{q},t)F_2(\mathbf{q},t)$ to recover the homogeneous Mode Coupling Theory.

2.5.2 Recovering Generalised-Mode-Coupling Theory

In this section we look at the zero field limit of the iGMCT hierarchy developed earlier. We begin with the equation of motion for $F_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)$. Clearly the terms in $\dot{F}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)$, $\ddot{F}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)$ are trivial, as we only require momentum conservation:

$$\lim_{U\to 0} F_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t) = F_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}$$

Moving onto the frequency term, we find that

$$\begin{aligned}&\lim_{U\to 0}\frac{1}{2!}\sum_{\mathbf{k},\mathbf{k}'}\Omega_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k},\mathbf{k}')F_4(\mathbf{k},\mathbf{k}';\mathbf{k}_3,\mathbf{k}_4,t)\\&\quad= \lim_{U\to 0}\frac{k_BT}{2m}\sum_{\mathbf{k},\mathbf{k}'}\sum_{\mathbf{n}}\left((\mathbf{k}_1\cdot\mathbf{n})\phi(\mathbf{k}_1-\mathbf{n})\left(S^{-1}(\mathbf{n};\mathbf{k})\delta_{\mathbf{k}_2,\mathbf{k}'} + S^{-1}(\mathbf{n};\mathbf{k}')\delta_{\mathbf{k}_2,\mathbf{k}}\right) + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2)\right)F_4(\mathbf{k},\mathbf{k}';\mathbf{k}_3,\mathbf{k}_4,t)\\&\quad= \frac{k_BT}{2m}\sum_{\mathbf{k},\mathbf{k}'}\sum_{\mathbf{n}}\left((\mathbf{k}_1\cdot\mathbf{n}\delta_{\mathbf{k}_1,\mathbf{n}})\left(\frac{2\delta_{\mathbf{k}_2,\mathbf{k}'}\delta_{\mathbf{n},\mathbf{k}}}{S(\mathbf{n})}\right) + (\mathbf{k}_1\leftrightarrow\mathbf{k}_2)\right)F_4(\mathbf{k},\mathbf{k}';\mathbf{k}_3,\mathbf{k}_4,t)\\&\quad= \frac{k_BT}{m}\left(\frac{|\mathbf{k}_1|^2}{S(\mathbf{k}_1)} + \frac{|\mathbf{k}_2|^2}{S(\mathbf{k}_2)}\right)F_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4}\end{aligned} \tag{58}$$

as expected. Now regarding the integral term, enforcing momentum conservation leads to :

$$\frac{N}{(3!)^4(2!)^2}\int_0^t d\tau \sum_{\mathbf{n}_1,\mathbf{n}_2}\sum_{\mathbf{n}_1',\mathbf{n}_2'}\sum_{\mathbf{k},\mathbf{k}'}\mathcal{V}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_1-\mathbf{n}_1,\mathbf{k}_2+\mathbf{n}_2,\mathbf{n}_1-\mathbf{n}_2)$$
$$\times F_6(\mathbf{k}_1-\mathbf{n}_1,\mathbf{k}_2+\mathbf{n}_2,\mathbf{n}_1-\mathbf{n}_2;\mathbf{k}_1-\mathbf{n}_1',\mathbf{k}_2+\mathbf{n}_2',\mathbf{n}_1'-\mathbf{n}_2')\mathcal{V}^\dagger(\mathbf{k},\mathbf{k}_1+\mathbf{k}_2-\mathbf{k};\mathbf{k}_1-\mathbf{n}_1',\mathbf{k}_2+\mathbf{n}_2',\mathbf{n}_1'-\mathbf{n}_2')$$
$$\times J_4^{-1}(\mathbf{k},\mathbf{k}_1+\mathbf{k}_2-\mathbf{k};\mathbf{k}',\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}')\dot{F}_4(\mathbf{k}',\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}';\mathbf{k}_3,\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3,t-\tau)$$

from which we immediately see the form of the two conserved modes have the same form as the modes $\{|\alpha_\mathbf{n}\rangle,|\beta_\mathbf{n}\rangle\}$ [41] upon a shift in wave-vector summations and diagonal approximation ($\mathbf{n}_1=\mathbf{n}_2$). It is possible to show, following the simple simplification ansatze used in this work that :

$$\lim_{U\to 0}\sum_{\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3}\mathcal{V}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)F_4(\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3;...)\sim\sum_\mathbf{n}\frac{k_BT}{mN}\left(\frac{\mathbf{k}_1\cdot(\mathbf{k}_1-\mathbf{n})}{S(\mathbf{k}_1-\mathbf{n})}+\frac{\mathbf{k}_1\cdot\mathbf{n}}{S(\mathbf{n})}-|\mathbf{k}_1|^2\right)F_4(\mathbf{k}_1-\mathbf{n},\mathbf{n},\mathbf{k}_2;...)$$
$$+\sum_\mathbf{n}\frac{k_BT}{mN}\left(\frac{\mathbf{k}_2\cdot(\mathbf{k}_2-\mathbf{n})}{S(\mathbf{k}_2-\mathbf{n})}+\frac{\mathbf{k}_2\cdot\mathbf{n}}{S(\mathbf{n})}-|\mathbf{k}_2|^2\right)F_4(\mathbf{k}_1,\mathbf{n},\mathbf{k}_2-\mathbf{n};...) \tag{59}$$

which is equivalent to [41]. The homogeneous equation of motion for the four-point dynamic correlator can now be written down and is analogous to the one found in [41]. We omit it here for brevity however. We emphasise once more that off-diagonal GMCT has the potential of solving some caveats of the diagonal GMCT and that our very general framework provides a consistent approach to obtain off-diagonal GMCT. We believe that the off-diagonal contributions could be important to the theory, giving possible insights into what the diagonal GMCT is still missing. Especially relating to the break-down of the Stokes-Einstein relation in the deep supercooled regime.

3 Equations of Motion for the Dynamical Susceptibilities

3.1 Linear Response Theory

Having developed a hierarchy of equation of motion for dynamic multi-point correlators in the presence of an external field and having verified that the zero field limits gave consistent results, we now move onto a linear response expansion of the order parameters F_{2n}. In the spirit of [18], we define a generalised multi-point dynamic susceptibility as

$$\chi_{2n+1}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n},\mathbf{q}_0,t)\equiv\lim_{U\to 0}\frac{\delta F_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n},t)}{\delta U(\mathbf{q}_0)} \tag{60}$$

We must also look at the response of the many-body static structure factors S_{2n}, the inverses S^{-1} and the generalised inverse currents J_{2n}^{-1}, since variations of static quantities do not necessarily vanish. We recall that our external field couples to density modes, and thus acts as a source term in the partition function $\mathcal{Z}$:

$$\mathcal{Z}\propto\int d\Gamma e^{-\beta\mathcal{H}(\Gamma)-\beta U(\mathbf{q}_0)}$$

where $U(\mathbf{q}_0)=g\hat{\rho}_{\mathbf{q}_0}$, with g an infinitesimal coupling constant that fixes the units. Then, for any static observable $\mathcal{O}_n(\mathbf{k}_1,...,\mathbf{k}_n)\propto\langle\hat{\rho}_{\mathbf{k}_1}...\hat{\rho}_{\mathbf{k}_n}\rangle$ (which may also contain time-derivatives, omitted here for simplicity), we find that

$$\begin{aligned}\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\mathcal{O}_n(\mathbf{k}_1,...,\mathbf{k}_n) &= \lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\int d\Gamma\,\hat{\rho}_{\mathbf{k}_1}...\hat{\rho}_{\mathbf{k}_n}e^{-\beta\mathcal{H}(\Gamma)-\beta U(\mathbf{q}_0)}\\ &= -\beta\int d\Gamma\,\hat{\rho}_{\mathbf{k}_1}...\hat{\rho}_{\mathbf{k}_n}\hat{\rho}_{\mathbf{q}_0}e^{-\beta\mathcal{H}(\Gamma)}\\ &= -\beta\tilde{\mathcal{O}}_{n+1}(\mathbf{k}_1,...,\mathbf{k}_n,\mathbf{q}_0)\delta_{\mathbf{k}_1+...+\mathbf{k}_n+\mathbf{q}_0,\mathbf{0}}\end{aligned} \tag{61}$$

where in the last line we enforce momentum conservation explicitly. $\tilde{\mathcal{O}}_{n+1}$ is then the response of the static quantity observable $\mathcal{O}_n$. We see that the response of an object with n degrees of freedom has $(n+1)$ degrees of freedom. We summarise the response of the necessary static quantities below:

- The statistical average of single density modes

$$\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\phi(\mathbf{k}) = -\beta S(\mathbf{k};\mathbf{q}_0)\delta_{\mathbf{k},\mathbf{q}_0}$$

- The $2n$-many body symmetric static structure factors

$$\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}S_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n}) = -\beta S_{2n+1}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{k}_{n+1},...,\mathbf{k}_{2n},\mathbf{q}_0)\delta_{\mathbf{k}_1+...+\mathbf{k}_n,\mathbf{k}_{n+1}+...+\mathbf{k}_{2n}+\mathbf{q}_0}$$

  which may then be simplified using the convolution approximations in Appendix A.

- The inverse 2-point correlation, obtained from functional differentiation of identity (19)

$$\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}S^{-1}(\mathbf{k}_1;\mathbf{k}_2) = \beta\frac{S_3(\mathbf{k}_1;\mathbf{k}_2,\mathbf{q}_0)}{S(\mathbf{k}_1)S(\mathbf{k}_2)}\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0} \approx \beta S(\mathbf{q}_0)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}$$

  Other inverse quantities should be obtainable via analogous methods.

We then start by taking $\lim_{U\to 0}\delta/\delta U(\mathbf{q}_0)$ of (30), which we re-write below :

$$\begin{aligned}0 = &\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\ddot{F}_2(\mathbf{k}_1;\mathbf{k}_2,t) + \lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\nu\dot{F}_2(\mathbf{k}_1;\mathbf{k}_2,t)\\ &+\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\frac{k_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}(\mathbf{k}_1\cdot\mathbf{k})\phi(\mathbf{k}_1-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}')F_2(\mathbf{k}';\mathbf{k}_2,t)\\ &+\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\frac{k_BT}{m}\int_0^t d\tau\sum_{\mathbf{k},\mathbf{k}'}\sum_{\mathbf{q},...,\mathbf{q}'''}\frac{k_1}{k'}\tilde{V}_{\mathbf{q}_0}(\mathbf{k}_1;\mathbf{q},\mathbf{q}')F_4(\mathbf{q},\mathbf{q}';\mathbf{q}'',\mathbf{q}''',t-\tau)\tilde{V}^{\dagger}_{\mathbf{q}_0}(\mathbf{k};\mathbf{q}'',\mathbf{q}''')J^{-1}(\mathbf{k};\mathbf{k}')\dot{F}_2(\mathbf{k}';\mathbf{k}_2,\tau)\end{aligned}$$

3.1.1 3-Point Dynamical Susceptibilities

The principal task is to evaluate the response of the static coefficients. We find that for the bare-frequency term

$$
\begin{aligned}
\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\frac{k_BT}{m}\sum_{\mathbf{k},\mathbf{k}'}&(\mathbf{k}_1\cdot\mathbf{k})\phi(\mathbf{k}_1-\mathbf{k})S^{-1}(\mathbf{k};\mathbf{k}')F_2(\mathbf{k}';\mathbf{k}_2,t)\\
&=\frac{S(|\mathbf{q}_0|)}{m}\left(|\mathbf{k}_1|^2-\frac{\mathbf{k}_1\cdot(\mathbf{k}_1-\mathbf{q}_0)}{S(|\mathbf{k}_1-\mathbf{q}_0|)}\right)F_2(\mathbf{k}_1-\mathbf{q}_0,t)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}\\
&-\frac{|\mathbf{k}_1|^2}{mS(|\mathbf{k}_1|)}\chi_3(\mathbf{k}_1;\mathbf{k}_2+\mathbf{q}_0)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}
\end{aligned}
\tag{62}
$$

Moving onto the integral term. The variation of the vertex

$$
\begin{aligned}
\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}V_{\mathbf{q}_0}(\mathbf{k}_1;\mathbf{q},\mathbf{q}')&=\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\frac{-ik_BT}{2mN}\left(\hat{\mathbf{k}}_1\cdot(\mathbf{k}_1-\mathbf{q})\,S^{-1}\left(\mathbf{k}_1-\mathbf{q};\mathbf{q}'\right)+(\mathbf{q}\leftrightarrow\mathbf{q}')\right)\\
&+\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\frac{ik_BT}{2mN}\sum_{\mathbf{q}'',\mathbf{q}'''}\sum_{\mathbf{k},\mathbf{k}'}\hat{\mathbf{k}}_1\cdot\mathbf{k}\phi\left(\mathbf{k}_1-\mathbf{k}\right)S^{-1}\left(\mathbf{k};\mathbf{k}'\right)S_3\left(\mathbf{k}';\mathbf{q}'',\mathbf{q}'''\right)\\
&\times S^{-1}\left(\mathbf{q}'';\mathbf{q}\right)S^{-1}\left(\mathbf{q}''';\mathbf{q}'\right)\\
&\approx\frac{-i}{2mN}S(\mathbf{q}_0)\left[\hat{\mathbf{k}}_1\cdot(\mathbf{k}_1-\mathbf{q})+\hat{\mathbf{k}}_1\cdot(\mathbf{k}_1-\mathbf{q}')+\hat{\mathbf{k}}_1\cdot(\mathbf{k}_1-\mathbf{q}_0)-2|\mathbf{k}_1|\right]\delta_{\mathbf{k}_1,\mathbf{q}+\mathbf{q}'+\mathbf{q}_0}\\
&=0
\end{aligned}
\tag{63}
$$

This means that the term $\delta/\delta U(VV^\dagger J^{-1})=VV^\dagger\delta/\delta U J^{-1}$ in the integral term. Hence, the equation of motion for the three-point susceptibility χ_3 reads

$$
\boxed{
\begin{aligned}
&\ddot{\chi}_3(\mathbf{k}_1;\mathbf{k}_2,t)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}+\frac{S(|\mathbf{q}_0|)}{m}\left(|\mathbf{k}_1|^2-\frac{\mathbf{k}_1\cdot(\mathbf{k}_1-\mathbf{q}_0)}{S(|\mathbf{k}_1-\mathbf{q}_0|)}\right)F_2(\mathbf{k}_1-\mathbf{q}_0,t)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}-\frac{|\mathbf{k}_1|^2}{mS(|\mathbf{k}_1|)}\chi_3(\mathbf{k}_1;\mathbf{k}_2,t)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}\\
&+\frac{m}{Nk_BT}\int_0^t d\tau\sum_{\mathbf{q},\mathbf{q}'}\frac{k_1}{k_2}V(\mathbf{k}_1;\mathbf{q},\mathbf{k}_1-\mathbf{q})F_4(-\mathbf{q},\mathbf{q}-\mathbf{k}_1;\mathbf{q}'',\mathbf{k}_1-\mathbf{q}'',\tau)V^\dagger(\mathbf{k}_1;\mathbf{q}'',\mathbf{k}_1-\mathbf{q}'')\\
&\qquad\times(\hat{\mathbf{k}}_1\cdot\hat{\mathbf{k}}_1-\hat{\mathbf{q}}_0)S(q_0)\dot{F}_2(\mathbf{k}_2,t-\tau)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}\\
&+\frac{m}{Nk_BT}\int d\tau\sum_{\mathbf{q},\mathbf{q}'}\frac{k_1}{k_2}V(\mathbf{k}_1;\mathbf{q},\mathbf{k}_1-\mathbf{q})\chi_5(\mathbf{q},\mathbf{k}_1-\mathbf{q};\mathbf{q}',\mathbf{k}_2-\mathbf{q}',\tau)V^\dagger(\mathbf{k}_2;\mathbf{q}',\mathbf{k}_2-\mathbf{q}')\dot{F}_2(\mathbf{k}_2,t-\tau)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}\\
&+\frac{m}{Nk_BT}\int d\tau\sum_{\mathbf{q},\mathbf{q}'}\frac{k_1}{|\mathbf{k}_2+\mathbf{q}_0|}V(\mathbf{k}_1;\mathbf{q},\mathbf{k}_1-\mathbf{q})F_4(-\mathbf{q},\mathbf{q}-\mathbf{k}_1;\mathbf{q}',\mathbf{k}_2+\mathbf{q}_0-\mathbf{q}',\tau)V^\dagger(\mathbf{k}_2+\mathbf{q}_0;\mathbf{q}',\mathbf{k}_2+\mathbf{q}_0-\mathbf{q}')\\
&\times\dot{\chi}_3(\mathbf{k}_2+\mathbf{q}_0;\mathbf{k}_2,t-\tau)\delta_{\mathbf{k}_1,\mathbf{k}_2+\mathbf{q}_0}=0
\end{aligned}
}
\tag{64}
$$

We remark that if we were to truncate the hierarchy in the Mode Coupling Approximation, the integral containing χ_5 would instead go as $\sim 2\chi_3F_2$, as found in [18, 52]. This is an important result, as we have derived an explicit and first-principles based equation for the three-point dynamical susceptibility of a supercooled liquid. As mentioned, we could in principle use the Mode Coupling Approximation here, but given the generality of the framework developed in the previous sections, we can derive similar microscopic equations for all the other odd-point susceptibilities.

3.2 Inclusion of Higher-Order Susceptibilities

3.2.1 5-point Dynamical Susceptibility

We apply the same methods as for the previous section on (43), the equation of motion for the 4-th point dynamic structure factor. Calculations for the time-derivative terms and the frequency terms are identical to the ones presented in the previous section, we write them down directly:

$$\begin{aligned}\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}&\left(\ddot{F}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)+\nu\dot{F}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)\right)\\&=-\beta\ddot{\chi}_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,\mathbf{q}_0,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0}-\beta\nu\dot{\chi}_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,\mathbf{q}_0,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0}\end{aligned}$$

and

$$\begin{aligned}\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\frac{1}{m\beta}\sum_{\mathbf{n},\mathbf{q}}&\left((\mathbf{k}_1\cdot\mathbf{n})\phi(\mathbf{k}_1-\mathbf{n})S^{-1}(\mathbf{n};\mathbf{q})F_4(\mathbf{q},\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)\right)\\&=\frac{S(\mathbf{q}_0)}{m}\left[|\mathbf{k}_1|^2-\frac{\mathbf{k}_1\cdot(\mathbf{k}_1-\mathbf{q}_0)}{S(\mathbf{k}_1-\mathbf{q}_0)}\right]F_4(\mathbf{k}_1-\mathbf{q}_0,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0}\\&+\frac{S(\mathbf{q}_0)}{m}\left[|\mathbf{k}_2|^2-\frac{\mathbf{k}_2\cdot(\mathbf{k}_2-\mathbf{q}_0)}{S(\mathbf{k}_2-\mathbf{q}_0)}\right]F_4(\mathbf{k}_1,\mathbf{k}_2-\mathbf{q}_0;\mathbf{k}_3,\mathbf{k}_4,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0}\\&-\left(\frac{|\mathbf{k}_1|^2}{S(\mathbf{k}_1)}+\frac{|\mathbf{k}_2|^2}{S(\mathbf{k}_2)}\right)\chi_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,\mathbf{q}_0,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0}\end{aligned}\tag{65}$$

from which we will be later able extract inductively all higher-order variations of the bare-frequency terms. Moving onto the integral term, we write from (42)

$$\begin{aligned}\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\mathcal{V}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)=&\frac{6i}{Nm}\mathbf{k}_1\cdot(\mathbf{n}_1+\mathbf{q}_0)S(\mathbf{q}_0)\delta_{\mathbf{k}_1-\mathbf{n}_1-\mathbf{q}_0,\mathbf{n}_2}\delta_{\mathbf{k}_2,\mathbf{n}_3}\\&-\frac{i}{N^2m}\frac{\mathbf{k}_1\cdot(\mathbf{k}_1-\mathbf{q}_0)S(\mathbf{q}_0)S_5(\mathbf{k}_1-\mathbf{q}_0,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)}{S(\mathbf{k}_1-\mathbf{q}_0)S(\mathbf{n}_1)S(\mathbf{n}_2)S(\mathbf{n}_3)}\delta_{\mathbf{k}_1-\mathbf{q}_0+\mathbf{k}_2,\mathbf{n}_1+\mathbf{n}_2+\mathbf{n}_3}\\&+\frac{i}{N^2m}\frac{|\mathbf{k}_1|^2S(\mathbf{q}_0)S_5(\mathbf{k}_1-\mathbf{q}_0,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3)}{S(\mathbf{n}_1)S(\mathbf{n}_2)S(\mathbf{n}_3)}\delta_{\mathbf{k}_1-\mathbf{q}_0+\mathbf{k}_2,\mathbf{n}_1+\mathbf{n}_2+\mathbf{n}_3}\\&-\frac{i}{N^2m}\frac{|\mathbf{k}_1|^2S_6(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3,\mathbf{q}_0)}{S(\mathbf{k}_1)S(\mathbf{n}_1)S(\mathbf{n}_2)S(\mathbf{n}_3)}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{n}_1+\mathbf{n}_2+\mathbf{n}_3+\mathbf{q}_0}\\&+\frac{i}{N^2m}\frac{|\mathbf{k}_1|^2S(\mathbf{q}_0)S_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1+\mathbf{q}_0,\mathbf{n}_2,\mathbf{n}_3)}{S(\mathbf{k}_1)S(\mathbf{n}_2)S(\mathbf{n}_3)}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{n}_1+\mathbf{n}_2+\mathbf{n}_3+\mathbf{q}_0}\\&+\frac{i}{N^2m}\frac{|\mathbf{k}_1|^2S(\mathbf{q}_0)S_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2+\mathbf{q}_0,\mathbf{n}_3)}{S(\mathbf{k}_1)S(\mathbf{n}_1)S(\mathbf{n}_3)}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{n}_1+\mathbf{n}_2+\mathbf{n}_3+\mathbf{q}_0}\\&+\frac{i}{N^2m}\frac{|\mathbf{k}_1|^2S(\mathbf{q}_0)S_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3+\mathbf{q}_0)}{S(\mathbf{k}_1)S(\mathbf{n}_1)S(\mathbf{n}_2)}\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{n}_1+\mathbf{n}_2+\mathbf{n}_3+\mathbf{q}_0}\\&+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)\\&\equiv\tilde{\mathcal{V}}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{n}_1,\mathbf{n}_2,\mathbf{n}_3,\mathbf{q}_0)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{n}_1+\mathbf{n}_2+\mathbf{n}_3+\mathbf{q}_0}\end{aligned}\tag{66}$$

It would be an interesting result that the response of the vertices for the 4-point dynamic correlation function vanish much like in the iMCT case. From a strictly mathematical point of view, we know that

the vertices of the homogeneous GMCT (both diagonal and off-diagonal) are simply linear combinations of the homogeneous MCT vertices [41, 25]. Given the striking resemblance of the homogeneous and inhomogeneous hierarchies, it would not be surprising that within a reasonable set of approximations we could also write the inhomogeneous vertex (42) as a linear combination of (29), which would in turn lead to the following result : $\tilde{\mathcal{V}}_4 = 0$. This is however a combinatorially complicated task, but we believe that a well written script in a symbolic programming language could manage this task. We leave this for future work.

Lastly we need to determine the variation $\delta/\delta U J_4^{-1}$. To do so, consider the following identity involving $J_4 \propto \langle A^{(2)}|A^{(2)}\rangle$:

$$\frac{1}{2!}\sum_{\mathbf{k},\mathbf{k}'}\langle\dot{\hat{\rho}}_{\mathbf{k}_1}\hat{\rho}_{\mathbf{k}_2}|\dot{\hat{\rho}}_{\mathbf{k}}\hat{\rho}_{\mathbf{k}'}\rangle\langle\dot{\hat{\rho}}_{\mathbf{k}}\hat{\rho}_{\mathbf{k}'}|\dot{\hat{\rho}}_{\mathbf{k}_3}\hat{\rho}_{\mathbf{k}_4}\rangle^{-1} = \delta_{\mathbf{k}_1,\mathbf{k}_3}\delta_{\mathbf{k}_2,\mathbf{k}_4}$$

To obtain the response of the inverse generalised current J_4^{-1}, one must consider the variation of the identity :

$$\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\sum_{\mathbf{k},\mathbf{k}'}\langle\dot{\hat{\rho}}_{\mathbf{k}_1}\hat{\rho}_{\mathbf{k}_2}|\dot{\hat{\rho}}_{\mathbf{k}}\hat{\rho}_{\mathbf{k}'}\rangle\langle\dot{\hat{\rho}}_{\mathbf{k}}\hat{\rho}_{\mathbf{k}'}|\dot{\hat{\rho}}_{\mathbf{k}_3}\hat{\rho}_{\mathbf{k}_4}\rangle^{-1} = 0 \tag{67}$$

where we can apply the chain rule and then re-arrange in favour of the wanted result. We note however that the right hand side of this identity is a simple combination of Kronecker deltas due to the presence of the time derivatives which breaks the exchange symmetry $\mathbf{k}_1 \leftrightarrow \mathbf{k}_2$. Unfortunately a closed form for this inverse could not be found analytically without any justified approximations. We will therefore simply write $\delta/\delta U(\mathbf{q}_0)J_4^{-1} = \tilde{J}_4^{-1}$ for the current-current correlation response and suggest determining this inverse numerically when the equations are solved. We now write down the equation of motion for the 5-point susceptibility:

$$\begin{aligned}
\hat{\mathcal{T}}_4[F_4,F_6,\mathbf{q}_0,t] = {} & \ddot{\chi}_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,\mathbf{q}_0,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0} + \nu_4\dot{\chi}_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,\mathbf{q}_0,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0} \\
& + \left(\frac{|\mathbf{k}_1|^2}{S(\mathbf{k}_1)} + \frac{|\mathbf{k}_2|^2}{S(\mathbf{k}_2)}\right)\chi_5(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4,\mathbf{q}_0,t)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0} \\
& + \frac{1}{(2!)^2(3!)^2}\sum_{\mathbf{k},\mathbf{k}'}\sum_{\mathbf{t}_1,\mathbf{t}_2}\sum_{\mathbf{t}_1',\mathbf{t}_2'}\int_0^t d\tau\,\mathcal{V}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{t}_1,\mathbf{t}_2,\mathbf{k}_1+\mathbf{k}_2-\mathbf{t}_1-\mathbf{t}_2) \\
& \qquad \times \chi_7(\mathbf{t}_1,\mathbf{t}_2,\mathbf{k}_1+\mathbf{k}_2-\mathbf{t}_1-\mathbf{t}_2;\mathbf{t}_1,\mathbf{t}_2,\mathbf{k}_1+\mathbf{k}_2-\mathbf{t}_1'-\mathbf{t}_2',\mathbf{q}_0,t-\tau) \\
& \qquad \times \mathcal{V}_4^\dagger(\mathbf{k},\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}_0-\mathbf{k};\mathbf{t}_1',\mathbf{t}_2',\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}_0-\mathbf{t}_1'-\mathbf{t}_2') \\
& \qquad \times J_4^{-1}(\mathbf{k},\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}_0-\mathbf{k};\mathbf{k}',\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}_0-\mathbf{k}') \\
& \qquad \times \dot{F}_4(\mathbf{k}',\mathbf{k}_1+\mathbf{k}_2-\mathbf{q}_0-\mathbf{k}';\mathbf{k}_3,\mathbf{k}_4,\tau)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0} \\
& + \frac{1}{(2!)^2(3!)^2}\sum_{\mathbf{k},\mathbf{k}'}\sum_{\mathbf{t}_1,\mathbf{t}_2}\sum_{\mathbf{t}_1',\mathbf{t}_2'}\int_0^t d\tau\,\mathcal{V}_4(\mathbf{k}_1,\mathbf{k}_2;\mathbf{t}_1,\mathbf{t}_2,\mathbf{k}_1+\mathbf{k}_2-\mathbf{t}_1-\mathbf{t}_2) \\
& \qquad \times F_6(\mathbf{t}_1,\mathbf{t}_2,\mathbf{k}_1+\mathbf{k}_2-\mathbf{t}_1-\mathbf{t}_2;\mathbf{t}_1',\mathbf{t}_2',\mathbf{k}_1+\mathbf{k}_2-\mathbf{t}_1'-\mathbf{t}_2',t-\tau) \\
& \qquad \times \mathcal{V}_4^\dagger(\mathbf{k},\mathbf{k}_1+\mathbf{k}_2-\mathbf{k};\mathbf{t}_1',\mathbf{t}_2',\mathbf{k}_1+\mathbf{k}_2-\mathbf{t}_1'-\mathbf{t}_2') \\
& \qquad \times J_4^{-1}(\mathbf{k},\mathbf{k}_1+\mathbf{k}_2-\mathbf{k};\mathbf{k}',\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}') \\
& \qquad \times \dot{\chi}_5(\mathbf{k}',\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}';\mathbf{k}_3,\mathbf{k}_4,\mathbf{q}_0,\tau)\delta_{\mathbf{k}_1+\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{q}_0}
\end{aligned} \tag{68}$$

where we have collected all terms which do not contain any susceptibilities in a general functional of the 4 and 6-point dynamic correlation function that we denote $\hat{\mathcal{T}}_4$. We see that just as χ_3 is dependent on χ_5, we find that χ_5 in turn depends on χ_7, the 7-point dynamical susceptibility. Equation (68) above forms the second iteration of the infinite hierarchy of equations of motion for the dynamical susceptibilities that our framework can generate.

3.2.2 Generalisation to Arbitrary Order

Akin to inhomogeneous GMCT, we may extend the hierarchy of the dynamical susceptibilities to arbitrary order. This new hierarchy inherits the properties of the GMCT one, with the exception that it is now a set of linear coupled integro-differential equations. Starting from (44) We find for the variation of the frequency term:

$$\begin{aligned}\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\Omega^2_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{q}_1,...,\mathbf{q}_n) = \Bigg[&-\frac{n!}{m}(\mathbf{k}_1\cdot\mathbf{q}_1)S(\mathbf{q}_0)\delta_{\mathbf{k}_1,\mathbf{q}_1+\mathbf{q}_0}\delta_{\mathbf{k}_2,\mathbf{q}_2}\times ...\times\delta_{\mathbf{k}_n,\mathbf{q}_n}\\ &+\frac{n!}{m}|\mathbf{k}_1|^2 S(\mathbf{q}_0)\delta_{\mathbf{k}_1,\mathbf{q}_1+\mathbf{q}_0}\delta_{\mathbf{k}_2,\mathbf{q}_2}\times ...\times\delta_{\mathbf{k}_n,\mathbf{q}_n}\Bigg]\\ &+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...+(\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\end{aligned} \tag{69}$$

Moving onto the the variation of the vertices, we find

$$\begin{aligned}&\lim_{U\to 0}\frac{\delta}{\delta U(\mathbf{q}_0)}\mathcal{V}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{p}_1,...,\mathbf{p}_{n+1})\\ &\quad=\Bigg[\frac{-i(n+1)!}{Nm}(\mathbf{k}_1\cdot(\mathbf{p}_1+\mathbf{q}_0))S(\mathbf{q}_0)\delta_{\mathbf{k}_1-\mathbf{p}_1-\mathbf{q}_0,\mathbf{p}_2}\delta_{\mathbf{k}_2,\mathbf{p}_3}\times ...\times\delta_{\mathbf{k}_n,\mathbf{p}_{n+1}}\\ &\quad-\frac{i\mathbf{k}_1\cdot(\mathbf{k}_1-\mathbf{q}_0)}{N^n m}\frac{S_{2n+1}(\mathbf{k}_1-\mathbf{q}_0,\mathbf{k}_2,...,\mathbf{k}_n;\mathbf{t}_1,...,\mathbf{t}_{n+1})}{S(\mathbf{k}_1-\mathbf{q}_0)S(\mathbf{t}_1)\times ...\times S(\mathbf{t}_{n+1})}\delta_{\mathbf{k}_1+...+\mathbf{k}_n,\mathbf{t}_1+...+\mathbf{t}_{n+1}+\mathbf{q}_0}\\ &\quad+\frac{i|\mathbf{k}_1|^2}{N^n m}\frac{S(\mathbf{q}_0)S_{2n+1}(\mathbf{k}_1-\mathbf{q}_0,\mathbf{k}_2,...,\mathbf{k}_n;\mathbf{t}_1,...,\mathbf{t}_{n+1})}{S(\mathbf{t}_1)\times ...\times S(\mathbf{t}_{n+1})}\delta_{\mathbf{k}_1+...+\mathbf{k}_n,\mathbf{t}_1+...+\mathbf{t}_{n+1}+\mathbf{q}_0}\\ &\quad-\frac{i|\mathbf{k}_1|^2}{N^n m}\frac{S_{2(n+1)}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{t}_1,...,\mathbf{t}_{n+1},\mathbf{q}_0)}{S(\mathbf{t}_1)\times ...\times S(\mathbf{t}_{n+1})}\delta_{\mathbf{k}_1+...+\mathbf{k}_n,\mathbf{t}_1+...+\mathbf{t}_{n+1}+\mathbf{q}_0}\\ &\quad+\left\{\frac{i|\mathbf{k}_1|^2}{N^n m}\frac{S_{2n+1}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{t}_1+\mathbf{q}_0,\mathbf{t}_2,...,\mathbf{t}_{n+1})}{S(\mathbf{k}_1)S(\mathbf{t}_2)\times ...\times S(\mathbf{t}_{n+1})}+(\mathbf{t}_1\leftrightarrow\mathbf{t}_2)+...+(\mathbf{t}_1\leftrightarrow\mathbf{t}_{n+1})\right\}\Bigg]\\ &\quad+(\mathbf{k}_1\leftrightarrow\mathbf{k}_2)+...+(\mathbf{k}_1\leftrightarrow\mathbf{k}_n)\\ &\quad\equiv\tilde{\mathcal{V}}_{2n}(\mathbf{k}_1,...,\mathbf{k}_n;\mathbf{p}_1,...,\mathbf{p}_{n+1},\mathbf{q}_0)\delta_{\mathbf{k}_1+...+\mathbf{k}_n,\mathbf{p}_1+...+\mathbf{p}_{n+1}+\mathbf{q}_0}\end{aligned} \tag{70}$$

To generalise the discussion around (66), showing that the response of an *arbitrary* vertex vanishes within a reasonable set of approximations would have two principal consequences. Firstly, it would drastically simplify the equations of motion for the susceptibilities by removing two integrals involving memory kernel evaluations. Secondly, vanishing vertex responses of any order would imply that the 'coupling' of the modes is insensitive to external perturbations (at least to linear order) which in turn implies a very strong robustness of the hierarchy of equations of motion.

Hence, we may then write the equation of motion for an arbitrary $(2n+1)$-th dynamical susceptibility as :

$$\begin{aligned}
&\hat{\mathcal{T}}_{2n}[F_{2n}, F_{2(n+1)}, \mathbf{q}_0, t] = \ddot{\chi}_{2n+1}(\mathbf{k}_1, ..., \mathbf{k}_n; \mathbf{k}_{n+1}, ..., \mathbf{k}_{2n}, \mathbf{q}_0, t)\delta_{\mathbf{k}_1+...+\mathbf{k}_n, \mathbf{k}_{n+1}+...+\mathbf{k}_{2n}+\mathbf{q}_0} \\
&\quad + \nu_{2n}\dot{\chi}_{2n+1}(\mathbf{k}_1, ..., \mathbf{k}_n; \mathbf{k}_{n+1}, ..., \mathbf{k}_{2n}, \mathbf{q}_0, t)\delta_{\mathbf{k}_1+...+\mathbf{k}_n, \mathbf{k}_{n+1}+...+\mathbf{k}_{2n}+\mathbf{q}_0} \\
&\quad + \left(\frac{|\mathbf{k}_1|^2}{S(\mathbf{k}_1)} + ... + \frac{|\mathbf{k}_n|^2}{S(\mathbf{k}_n)}\right)\chi_{2n+1}(\mathbf{k}_1, ..., \mathbf{k}_n; \mathbf{k}_{n+1}, ..., \mathbf{k}_{2n}, \mathbf{q}_0, t)\delta_{\mathbf{k}_1+...+\mathbf{k}_n, \mathbf{k}_{n+1}+...+\mathbf{k}_{2n}+\mathbf{q}_0} \\
&\quad + \frac{1}{(n!)^2((n+1)!)^2}\sum_{\{\mathbf{t}_j\},\{\mathbf{t}'_j\}}\sum_{\{\mathbf{k}'_j\},\{\mathbf{k}''_j\}}\int_0^t d\tau \mathcal{V}_{2n}(\mathbf{k}_1, ..., \mathbf{k}_n; \mathbf{t}_1, ..., \mathbf{t}_n, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{t}_1 - ... - \mathbf{t}_n) \\
&\qquad \times \chi_{2(n+1)+1}(\mathbf{t}_1, ..., \mathbf{t}_{n+1}; \mathbf{t}'_1, ..., \mathbf{t}'_{n+1}, \mathbf{q}_0, t-\tau) \\
&\qquad \times \mathcal{V}^\dagger_{2n}(\mathbf{k}'_1, ..., \mathbf{k}'_{n-1}, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{k}'_1 - ... - \mathbf{k}'_{n-1} - \mathbf{q}_0; \mathbf{t}'_1, ..., \mathbf{t}'_n, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{t}'_1 - ... - \mathbf{t}'_n - \mathbf{q}_0) \\
&\qquad \times J^{-1}_{2n}(\mathbf{k}'_1, ..., \mathbf{k}'_{n-1}, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{k}'_1 - ... - \mathbf{k}'_{n-1} - \mathbf{q}_0; \mathbf{k}''_1, ..., \mathbf{k}''_{n-1}, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{k}''_1 - ... - \mathbf{k}''_{n-1} - \mathbf{q}_0) \\
&\qquad \times \dot{F}_{2n}(\mathbf{k}''_1, ..., \mathbf{k}''_{n-1}, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{k}''_1 - ... - \mathbf{k}''_{n-1} - \mathbf{q}_0; \mathbf{k}_{n+1}, ..., \mathbf{k}_{2n}, \tau)\delta_{\mathbf{k}_1+...+\mathbf{k}_n, \mathbf{k}_{n+1}+...+\mathbf{k}_{2n}+\mathbf{q}_0} \\
&\quad + \frac{1}{(n!)^2((n+1)!)^2}\sum_{\{\mathbf{t}_j\},\{\mathbf{t}'_j\}}\sum_{\{\mathbf{k}'_j\},\{\mathbf{k}''_j\}}\int_0^t d\tau \mathcal{V}_{2n}(\mathbf{k}_1, ..., \mathbf{k}_n; \mathbf{t}_1, ..., \mathbf{t}_n, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{t}_1 - ... - \mathbf{t}_n) \\
&\qquad \times F_{2(n+1)}(\mathbf{t}_1, ..., \mathbf{t}_n, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{t}_1 - ... - \mathbf{t}_n; \mathbf{t}'_1, ..., \mathbf{t}'_n, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{t}'_1 - ... - \mathbf{t}'_n, t-\tau) \\
&\qquad \times \mathcal{V}^\dagger_{2n}(\mathbf{k}'_1, ..., \mathbf{k}'_{n-1}, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{k}'_1 - ... - \mathbf{k}'_{n-1}; \mathbf{t}'_1, ..., \mathbf{t}'_n, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{t}'_1 - ... - \mathbf{t}'_n) \\
&\qquad \times J^{-1}_4(\mathbf{k}'_1, ..., \mathbf{k}'_{n-1}, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{k}'_1 - ... - \mathbf{k}'_{n-1}; \mathbf{k}''_1, ..., \mathbf{k}''_{n-1}, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{k}''_1 - ... - \mathbf{k}''_{n-1}) \\
&\qquad \times \dot{\chi}_{2n+1}(\mathbf{k}''_1, ..., \mathbf{k}''_{n-1}, \mathbf{k}_1 + ... + \mathbf{k}_n - \mathbf{k}''_1 - ... - \mathbf{k}''_{n-1}; \mathbf{k}_{n+1}, ..., \mathbf{k}_{2n}, \mathbf{q}_0, \tau)\delta_{\mathbf{k}_1+...+\mathbf{k}_n, \mathbf{k}_{n+1}+...+\mathbf{k}_{2n}+\mathbf{q}_0}
\end{aligned} \tag{71}$$

where again the general functional $\hat{\mathcal{T}}_{2n}$ contains all terms generated by the linear response expansion but that do not contain any susceptibility terms.

3.2.3 General Comments on Non-Linear Susceptibilities

From the above derivation, we note that we can cast the dynamical susceptibilities into the following form :

$$\int_0^t d\tau \hat{\mathcal{D}}_{2n}\left[\{F_{2j}(\{\mathbf{k}_j\})\}, \{\chi_{2j+1}(\{\mathbf{k}_j\})\}, \mathbf{q}_0; t, \tau\right] * \chi_{2n+1}(\mathbf{k}_1, ..., \mathbf{k}_{2n}, \mathbf{q}_0, \tau) = \hat{\mathcal{T}}_{2n}[\{F_{2j}(\{\mathbf{k}_j\})\}, t] \tag{72}$$

where $j < n$ and $\hat{\mathcal{D}}_{2n}$ is a *linear non-local* differential operator. We hypothesise that $\hat{\mathcal{D}}_{2n}$ becomes *singular* at the transition temperature T_{GMCT}, akin to the (G)MCT where an eigenvalue of a critical operator vanishes [32] ; this would lead to a true divergence of the dynamical susceptibilities and justify that the structural glass transition predicted by GMCT is indeed a Landau mean-field theory. In fact, the association of the singularity of some linear operator to a growing lengthscale at the glass transition is not a new idea and has been present for some years in field-theoretical approaches to the problem [22, 16]. This extended framework is also an important test for the validity of GMCT as a valid theory of the structural glass transition.

We emphasise that (71) is a really novel result which gives us direct theoretical access to non-linear susceptibilities, and thus collective effects in supercooled liquids from a fully first-principles framework.

While it will prove to be a herculean task, we believe that this hierarchy can be numerically tackled, at least for the first few susceptibilities. We have high hopes that direct comparison with experimentally measured non-linear susceptibilities [27, 9, 24] as well as from molecular dynamics simulations in the presence of external pinning fields [44] will finally shed light on the putative idea of emerging and growing lengthscales in supercooled liquids approaching the glass transition.

4 Toy Models of the Glass Transition & Extensions to Dynamical Susceptibilities

4.1 Definitions of Toy Models

In order to get an idea of the qualitative behaviour of both GMCT and the associated dynamical susceptibilities, we study simplified versions of the microscopic equations derived in this work. These simplified models are known as *schematic* models in the glass literature. Schematic models have proven useful to study semi quantitative behaviour of different glass transition scenarios [23, 40]. These simplified models are obtained from the microscopic equations (44) and (71) by dropping all wave-vector dependence and retaining only the time-dependence in the observables. This is equivalent to restricting all-physical processes to a single arbitrary shell in momentum space. From (44) and (71), we then perform the following 'substitutions':

- The wave-number dependent many-body intermediate scattering functions

$$F_{2m}(\mathbf{k}_1, ..., \mathbf{k}_m; \mathbf{k}_{m+1}, ..., \mathbf{k}_{2m}, t) \to \phi_{2m}(t)$$

  become simple functions of time

- The wave-number dependent dynamical susceptibilities

$$\chi_{2m+1}(\mathbf{k}_1, ..., \mathbf{k}_m; \mathbf{k}_{m+1}, ..., \mathbf{k}_{2m}, \mathbf{q}_0, t) \to \chi_{2m+1}(q, t)$$

  become schematic susceptibilities. We note that we retain a single wave-number (modulus) q, to effectively account for the lengthscale over which the system is perturbed, as displayed in Fig.6.

- The frequency term and its response expansion :

$$\Omega_{2m}(\mathbf{k}_1, ..., \mathbf{k}_m; \mathbf{k}_{m+1}, ..., \mathbf{k}_{2m}) \to \mu_{2m}$$

$$\tilde{\Omega}_{2m}(\mathbf{k}_1, ..., \mathbf{k}_m; \mathbf{k}_{m+1}, ..., \mathbf{k}_{2m}, \mathbf{q}_0) \to \tilde{\mu}_{2m}$$

  where we do not retain the q dependence in $\tilde{\mu}_{2m}$ for simplicity, so as to reduce the number of parameters in the model.

- The vertices and inverse current terms are combined into a single effective coupling term that we may interpret as an inverse temperature:

$$\mathcal{V}_{2m} * \mathcal{V}^{\dagger}_{2m} * J^{-1}_{2m} \to \Lambda_{2m}$$

  and the 3 integrals that come from linear response are combined into the same coupling

$$\left\{\mathcal{W}_{2m} * \mathcal{V}^{\dagger}_{2m} * J^{-1}_{2m} + \mathcal{V}_{2m} * \mathcal{W}^{\dagger}_{2m} * J^{-1}_{2m} + \mathcal{V}_{2m} * \mathcal{V}^{\dagger}_{2m} * \tilde{J}^{-1}_{2m}\right\} \to \Lambda_{2m}$$

  This means that the 3-integrals that arise in the general functional $\hat{\mathcal{T}}$ in (72), the eigenvalue equation for the susceptibilities are effectively combined into a single integral equivalent to the integral term arising the schematic GMCT.

- We note that the coupling on the response of the memory function takes the following form : $\lambda_{2m}(q) = \Lambda_{2m}(1 - \Gamma_{2m}q^2)$. This form arises from symmetry arguments (namely rotational invariance) [18, 52]. Since it corresponds to a perturbative expansion truncated beyond linear order, we require that the product $\Gamma_{2m}q^2 < 1$.

By applying the above 'simplifications', we formally obtain two infinite hierarchies coupled integro-differential equations. By allowing for generalisation of the functional form of the integral kernels, we write:

$$\frac{\partial^2 \phi_{2n}}{\partial t^2} + \nu_{2n}\frac{\partial \phi_{2n}}{\partial t} + \mu_{2n}\phi_{2n} + \Lambda_{2n}\int_0^t d\tau M_{2n}^{(\phi)}(t-\tau)\frac{\partial \phi_{2n}}{\partial t}\bigg|_{t=\tau} = 0 \tag{73}$$

subject to initial conditions $\phi_{2n}(t=0) = 1$ and $\dot{\phi}_{2n}(t=0) = 0$. While the dynamical susceptibilities read :

$$\begin{aligned}&\left[\frac{\partial^2}{\partial t^2} + \nu_{2n}\frac{\partial}{\partial t} + \mu_{2n}\right]\chi_{2n+1}(q;t) + \tilde{\mu}_{2n}\phi_{2n}(t)\\ &+ \int_0^t d\tau \left\{\Lambda_{2n}M_{2n}^{(\phi)}(t-\tau)\frac{\partial \chi_{2n+1}(q;t)}{\partial t}\bigg|_{t=\tau} + \lambda_{2n}(q)M_{2n}^{(\chi)}(q;t-\tau)\frac{\partial \phi_{2n}}{\partial t}\bigg|_{t=\tau} + \Lambda_{2n}M_{2n}^{(\phi)}(t-\tau)\frac{\partial \phi_{2n}}{\partial t}\bigg|_{t=\tau}\right\} = 0\end{aligned} \tag{74}$$

subject to initial conditions $\chi_{2n+1}(q, t = 0) = 1$ and $\dot{\chi}_{2n+1}(q, t = 0) = 0$. We note that the generalisation of the integral kernels formally reads $M_{2n}^{(\phi)} \equiv \mathcal{M}[\{\phi_{2(j)}\}]$ some functional of correlators and $M_{2n}^{(\chi)} = \mathcal{M}'[\{\chi_{2(j+1)}\}, \{\phi_{j'}\}]$ some other functional of both the correlators and their suscepibilities which is in fact directly obtained from $\mathcal{M}[\{\phi_{2(j)}\}]$. The first hierarchy considered has $M_{2n}^{(\phi)}(t) = \phi_{2(n+1)}(t)$, and is referred to as schematic generalised mode coupling theory where each variable ϕ_{2n} can be heuristically associated with the many-body dynamic structure factors F_{2n} from earlier sections. Various parametrisation have been thoroughly studied through the years [48, 21, 33, 35] and analytical results exist for the infinite limit $n \to \infty$ in the case of the Mayer-Miyazaki-Reichmann (MMR) parametrisation [40]. For the second hierarchy considered, we find that $M_{2n}^{(\chi)}(q;t) \equiv \chi_{2(n+1)+1}(q;t)$ by simple generalisation from 71. Numerical treatments of the hierarchies requires truncation at an appropriate order N_c. Three common closure types can be identified from the literature:

- *Exponential closure* : where $M_{2N_c}^{(\phi)}(t) = 0$ is enforced by hand. Then, the corresponding $\phi_{2N_c}(t)$ decouples from the rest of the hierarchy and exhibits (oscillatory) decay behaviour. We formally denote this closure type EXP $-N_c$

- *Mean-field closures of type I* : where $M_{2N_c}^{(\phi)}(t)$ is self-consistently approximated with a product of different ϕ's :

  $$M_{2N_c}^{(\phi)}(t) = \phi_{2i_1}(t)\phi_{2i_2}(t)...\phi_{2i_p}(t) = \prod_{j=1}^{p}\phi_{2i_j}(t)$$

  possibly under the constraint that $\sum_{j=1}^{p} i_j = N_c$ (in the case of schematic GMCT). We formally denote this closure type MF $-2N_c[..., i_j^n, ...]$.

- *Mean-field closures of type II* : a slightly more general case of Mean-field closures of type I which allows for multi-linear products of lower order ϕ's

$$M^{(\phi)}_{2N_c}(t) = \alpha\phi_{2i_1}(t)\phi_{2i_2}(t)...\phi_{2i_p}(t) + \beta\phi_{2i'_1}(t)\phi_{2i'_2}(t)...\phi_{2i'_p}(t) + ...$$

Mean-field closures of type II can be found in the spin glass literature [7, 21], or can be relevant for systems where different modes of observables couple in asymmetric ways. They provide a somewhat consistent way of introducing additional parameters which unfold the parameter space into new dimensions, leading to richer bifurcation structures.

We emphasise that by construction the choice of self-consistent closure immediately determines that of the susceptibility hierarchy. It is a trivial exercise to see that exponential closures give $M^{(\chi)}_{2N_c}(q;t) = 0$. Mean-field closures of type I are slightly more complex. By the chain-rule, we find that

$$M^{(\chi)}_{2N_c}(t) = \chi_{2i_1+1}(q;t)\phi_{2i_2}(t)...\phi_{2i_p}(t) + \phi_{2i_1}(t)\chi_{2i_2+1}(q;t)...\phi_{2i_p}(t) + ... + \phi_{2i_1}(t)\phi_{2i_2}(t)...\chi_{2i_p+1}(q;t)$$

and analogously for the Mean-field closures of type II. We would like to emphasise the flexibility of these two coupled hierarchies of equations of motion for the schematic structure factors $\phi_{2n}(t)$ and their associated susceptibilities $\chi_{2n+1}(q;t)$. In addition to the freedom in the choice of self-consistent closure relation, the level dependent coefficients $\mu_{2n}, \tilde{\mu}_{2n}, \Gamma_{2n}$ as well as the coupling parameters Λ_{2n} can in principle be any desired real-number.

4.2 Ergodicity Breaking in Schematic Models

We know from prior studies [40] that closure of exponential type systematically lead to avoided glass transition singularities. This in turn implies (and we have checked this rigorously) that the associated schematic dynamical susceptibilities do not display critical-like behaviour, and are thus of no true interest in the context of this work. In the case of mean-field closures of type I & II however, the schematic generalised mode coupling theory possesses an ergodicity breaking transition that is analogous to the one encountered in the microscopic equations of motion. We can therefore hope to see diverging non-linear susceptibilities near this transition. The non-ergodic state (glass state) is defined as $\phi_2(t \to \infty) > 0$, while the ergodic state (liquid) is identified as $\phi_2(t \to \infty) = 0$. Denoting $\phi_2(t \to \infty) \equiv \phi_2^\infty$, this quantity serves as an order parameter akin to $F_2(t)$ in the microscopic theory. We recall that in the glass regime, the long-time limit of $F_2(t)$ serves as an order parameter for the transition, and that in the case of these schematic models we can analyse the true asymptotic limits.

In these schematic models, the breadth of possible parametrisations can lead to very different types of glass transition singuarities [33, 35, 37, 21]. In order to study these, we consider the long-time limit of the schematic GMCT hierarchy, which can be investigated in Laplace frequency space (using the well known property $\lim_{t\to\infty} f(t) = \lim_{z\to 0} z\mathcal{L}\{f\}(z)$), where we find that

$$\frac{\mu_{2m}\phi^\infty_{2m}}{1-\phi^\infty_{2m}} = \Lambda_{2m} M^{(\phi)\infty}_{2m} \tag{75}$$

determines the long-time behaviour of the order parameters. The glass singularity manifests itself as the point where ϕ^∞_{2m}, (also referred to as a *form factor*) is no longer zero for a given point in parameter space. We note that there are strong constraints on the solutions that we allow. For instance all physical solutions must satisfy : $\phi^\infty_{2m} \in [0, 1]$. We follow the terminology of bifurcations introduced by

Arnol'd [34]. Solving for the form factors generally amounts to solving a set of polynomial equations in these. The singularities are characterised by the degeneracy of these roots, and we denote A_l the bifurcation point associated with an l-th order degeneracy. The simplest glass transition scenario is then the A_2 transition, where non-ergodic and ergodic form factor collide. This is also known as a fold bifurcation in the literature. More generally, a singularity of type A_l consists of the coalescence of two singularities of type A_{l-1} in parameter space. Singularities of type A_3 and A_4 are known as cusps and swallowtails in the bifurcation literature. These higher order bifurcation scenarios however require a certain minimal number of variable parameters to be observed.

4.3 Numerical Results

4.3.1 Hierarchy Parametrisations

For the rest of this work, we turn our attention to two special cases of mean-field closures of type I: (i) $M^{(\phi)}_{2N_c}(t) = \phi_2(t)^{N_c+1}$ that we abbreviate with MF $-2N_c[1^{N_c+1}]$ and (ii) $M^{(\phi)}_{2N_c}(t) = \phi_2(t)\phi_{2N_c}(t)$ which we abbreviate MF $-2N_c[1^1, N_c^1]$. For any closure level N_c, the hierarchy of coupled relaxators with MF closures exhibit glass transition singularities of type A_2. We identify two important parametrisations for the hierarchies [23]:

(A) $\{\mu_{2n} = n \text{ and } \Lambda_{2n} = \Lambda n^{1-\nu}, \text{ with } \nu > 0\}$

(B) $\{\mu_{2n} = n \text{ and } \Lambda_{2n} = \Lambda(n + c), \text{ with } c \geq 0\}$

which will be referred to as model A and model B respectively from now on. For simplicity, in all cases considered we will assume that $\tilde{\mu}_{2n} = \mu_{2n}$. Model A is interesting because it allows us to probe for 3 different regimes. In the case where the exponent $\nu < 1$, we have a coupling strength Λ_{2n} which increases as the total hierarchy level N_c increases. Physically speaking, this case increases the influence of many-body correlators on the solution of the lowest order one $\phi_2(t)$. Vice-versa when $\nu > 1$, where we attribute less 'weight' to the higher-order correlators as the hierarchy height N_c increases. The special case $\nu = 1$ recovers the MMR model [23] previously mentioned. In the case of model B, we consider a linearly increasing coupling throughout throughout the hierarchy.

4.3.2 Numerical Methods

We define the principal characteristic timescale $\tau_\alpha^{(2n)}$ of the $2n$-th correlator as $\phi_{2n}(\tau_\alpha^{(2n)}) = 1\text{e-}1$, which we interpret as a de-correlation time and corresponds to the principal α-relaxation regime. Hence, whenever $\phi_{2n}(t) \leq \phi_{2n}(\tau_\alpha^{(2n)})$ we have a liquid configuration. Similarly, the secondary β relaxation time $\tau_\beta^{(2n)}$ is defined as $\phi_{2m}(\tau_\beta^{(2n)}) = \phi_{2n}^c$, where ϕ_{2n}^c is the long-time limit of the the $2n$-th correlation function at the critical point. In glasses, the β-regime is often associated with the onset of caging mechanisms where particles are effectively kinetically trapped by their neighbours.

The time-dependent solutions to the hierarchies are obtained using the algorithm described in [31], which has been suitably modified for the dynamical susceptibilities hierarchy. In brief, the two hierarchies are solved in parallel using a modified Euler's integration method with a logarithmically increasing time-step and solving for fixed-point equations until convergence at each time-step. We point out that the logarithmically increasing time-step size employed in this work can introduce very subtle errors near critical points. In our case this is especially true for growing hierarchy closure

level N. While it is very easy to solve for say $N = 10000$ on both sides of the transition, systematic studies of scaling laws near criticality are in practice very difficult to do. The results from direct integration techniques presented in the rest of this work are henceforth restricted lower values of $N < 20$.

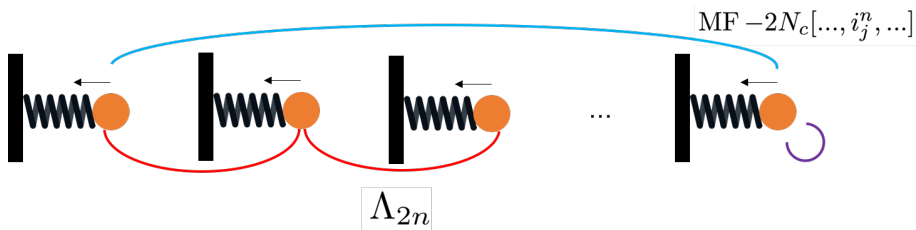
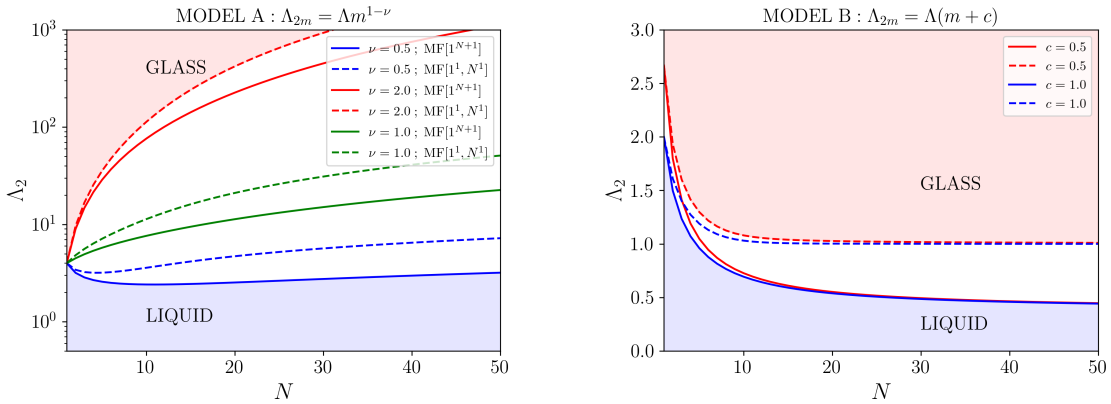


Figure 9: *Schematic illustration of the GMCT hierarchy of coupled relaxators.*

4.3.3 Phase Diagrams

We plot below phase diagrams of Model-A and Model-B for various parameter values of ν and c. We remark that the location of the transition point is strongly dependent on the choice of parametrisation as we increase the hierarchy height N. This is especially true for model-A where as N is increased, we systematically observe that Λ_2^c grows continuously and is expected to diverge as $N \to \infty$. Given that we interpret Λ_2 as an inverse temperature, we find that the structure of the hierarchy systematically accelerates the dynamics. This has also been demonstrated in the fully microscopic theory [49, 26], where increasing the hierarchy heights leads to lower T_g. We also note that for Model A, there is a notable difference in the phase-curve for a given self-consistent closure relation : mixed closures such as MF $-2N_c[1^1, N_c^1]$ systematically have higher transition points, which means that they display faster dynamics. This behaviour is intuitively expected: below the transition, we anticipate higher-order correlations to decay (much) faster than the two-body correlation function. Using a self-consistent closure involving higher-order correlation function will make the highest level of the hierarchy decay faster than if it only involved low-order ones, amd thus the whole spectrum of the dynamics will be accelerated.



(a) (b)

Figure 10: (a) *Phase Diagrams for schematic GMCT Model A with exponents* $\nu = 1/2, 1, 2$ *and mean-field closures discussed in the text.* (b) *Phase Diagrams for schematic GMCT Model B with parameter* $c = 1/2, 1$ *and mean-field closures discussed in the text*

The two models A & B of the hierarchies above display a bifurcation at some critical Λ_2^c, beyond which the $2n$-th point correlators admit finite values at long-times. In effect, have a single bifurcation parameter: Λ in both model A and model B, and for all parametrisations of similar types, we have an A_2 singularity. By virtue of the strong topological classification of bifurcation types we expect very similar qualitative behaviour from both models. We have verified this claim numerically for a very wide range of points in parameter space. The following discussion on the results therefore focuses on Model A, but the conclusions also extend to Model B.

4.3.4 Scaling Laws at Criticality

We study scaling relations for the principal relaxation time $\tau_\alpha^{(2n)}$ as we approach the A_2 singularity. We begin by looking at the scaling of the principal relaxation time $\tau_\alpha^{(2n)} \propto \epsilon^{-\gamma_n}$. We systematically verify that the analytical results presented in [40] and find that $\gamma_{2n} = 1.769$ is actually independent of the level of the hierarchy, as displayed for $\tau_\alpha^{(2)}, \tau_\alpha^{(4)}$ in Fig.11 below. We emphasise that this result is actually independent of the parametrisation Λ_{2n} of the system, provided that we remain at an A_2 point.

The β regime previously defined also offers interesting scaling laws. We define a relative trajectory for the $2n$-th correlator : $\Delta\phi_{2n}(t) = |\phi_{2n}(t) - \phi_{2n}^c|$ where by definition we expect $\Delta\phi_{2n}(\tau_\beta) = 0$. Arbitrarily close to the transition point, we numerically observe power-law regimes for correlators of any level and make the ansatz : $\Delta\phi_{2n}(t) \sim t^{-a_n}$ for $t < \tau_\beta$ and $\Delta\phi_{2n}(t) \sim t^{b_n}$ for $t > \tau_\beta$ for a_n, b_n level dependent exponents. We also find that the exponents extracted all satisfy the established general relations [23, 40]:

$$\gamma_n = \frac{1}{2a_n} + \frac{1}{2b_n}$$
$$1 = \frac{\Gamma(1+b_n)^2}{\Gamma(1+2b_n)} \times \frac{\Gamma(1-2a_n)}{\Gamma(1-a_n)^2} \tag{76}$$

which hold valid arbitrarily close to the critical A_2 point. This relation is one of the great successes of the Mode Coupling Theory. A possible numerical solution to (76) give $\gamma = 1.769$, $a = 0.395$ and $b = 1$. Numerical verification for different cases are shown in Fig.12. We numerically find that a_n, b_n and γ_n are independent of n for the model considered. Within reasonable numerical accuracy we are also able to confirm that the fitted exponents agree with the solution proposed above. The independence of the scaling exponents from the hierarchy level show that the inclusion of higher order correlation functions in the framework does not damage the successful parts of MCT. We note that as we study higher-order correlation functions, the time-window for the existence of these scaling laws shortens. This is expected as the criticality of higher-order correlation functions is thought to emerge only asymptotically close to the bifurcation point.

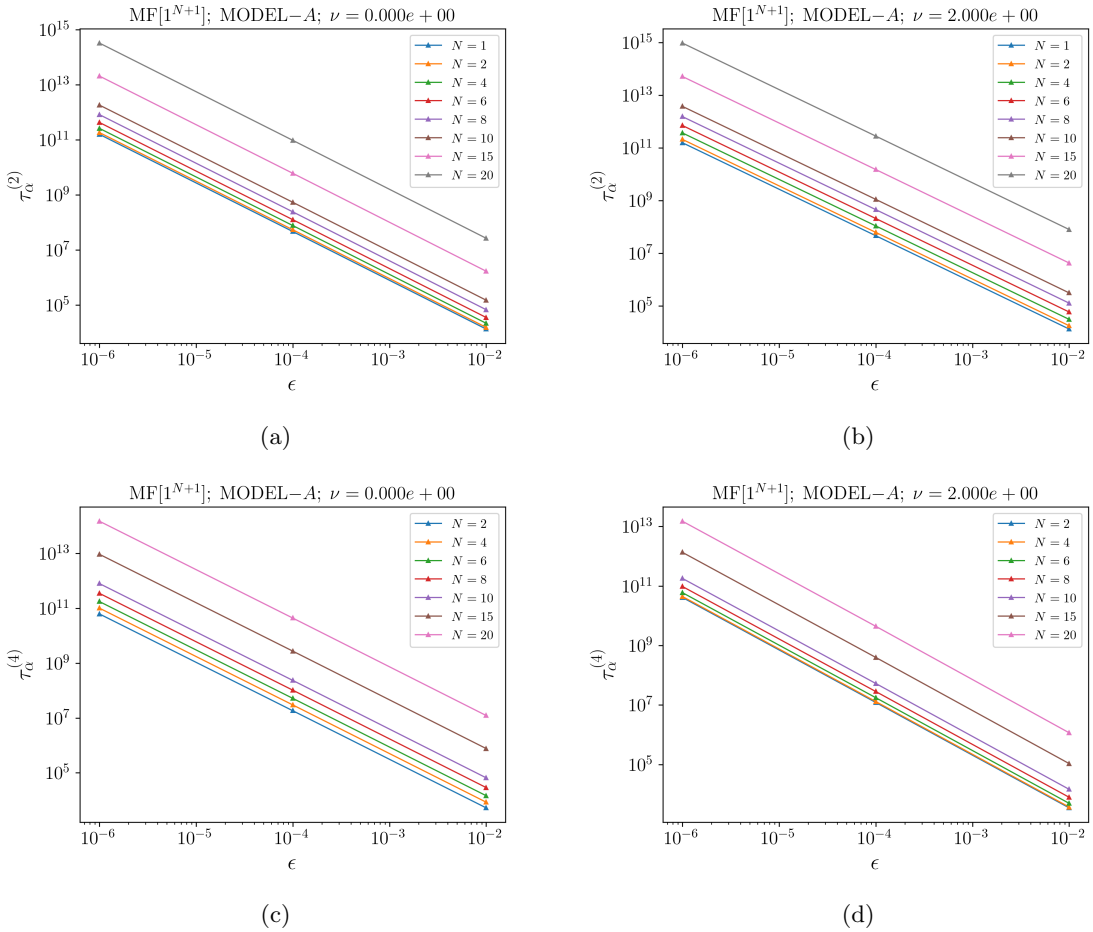


Figure 11: (a)-(b) *Scaling relation for the principal relaxation time* $\tau_\alpha^{(2)}$ *associated with correlation function* $\phi_2(t)$ *for Model A with parameters* $\nu = 0, 2$. (c)-(d) *Scaling relation for the principal relaxation time* $\tau_\alpha^{(4)}$ *associated with correlation function* $\phi_4(t)$ *for Model A with parameters* $\nu = 0, 2$.

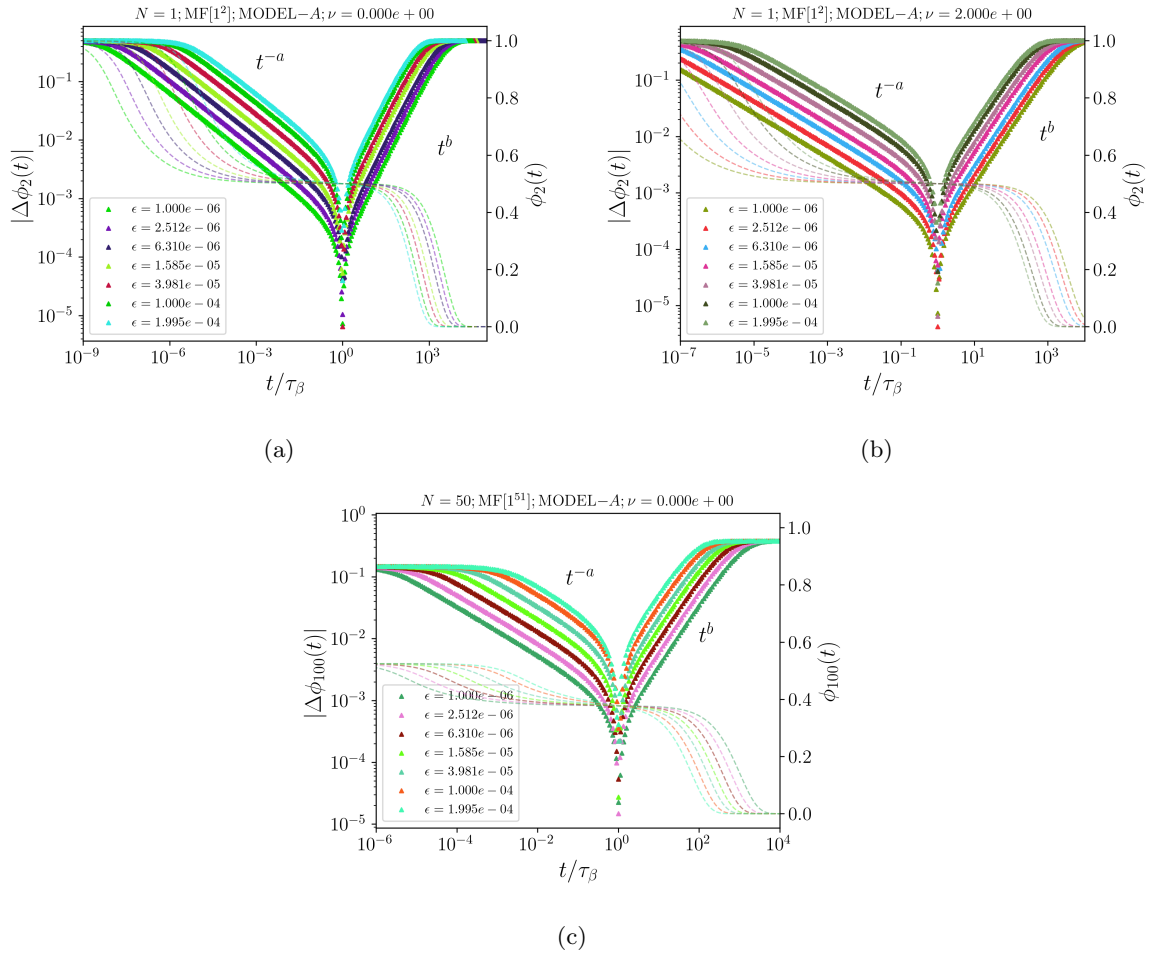


Figure 12: (a)-(b)-(c) *Relative trajectories with respect to the glass-plateau value* $|\Delta\phi_{2m}(t)| = |\phi_{2m}(t) - \phi_{2m}^{\infty}|$ *for two parametrisations of Model-A with* $\nu = 0, 2$.

4.3.5 Schematic Nonlinear Susceptibilities at Criticality

Now that the behaviour of the dynamic correlation functions is better understood near the critical point, we move on to the study of their associated dynamical susceptibilities. We pay particular attention to their global behaviour as well as near the important timescales (α, β regimes) at the onset of criticality.

We show in Fig.13 the solutions to the dynamical susceptibilities $\chi_3(q;t), \chi_5(q;t)$ and $\chi_{21}(q;t)$ for various value of q, relative distances $\epsilon = |\Lambda_2 - \Lambda_2^c|/\Lambda_2^c$ to the critical point Λ_2^c and hierarchy levels $N_c = 1, 2, 10$ respectively. This means that we do not show the effect from the inclusion of higher-order correlation functions to lower-order susceptibilities. Firstly, we observe that the plotted non-linear susceptibilities shown display critical-behaviour as we approach criticality. We checked consistently and we are in position to say that this extends to all dynamical susceptibilities, of any order asymptotically close to the critical point for the two models considered. Additionally, we remark a behaviour that is very similar to that of the non-Gaussian parameter $\alpha_2(t)$ and the 4-point function $\chi_4(t)$ measured from

simulations shown in Fig.5a-5b ; as the glass transition is approached, we see that the susceptibilities grow and are shifted in time. We also consistently checked and are able to say that the location of the peaks of the susceptibilties χ^*_{2n+1} coincides with the principal structural time $\tau_\alpha^{(2n)}$ of the association $2n$-point correlation function. Our schematic model also displays a non-trivial dependence on the perturbative wave-number q. We systematically find for all susceptibilities that the peak is highest in the limit $q \to 0$ and smallest for $q \to 1$ (we recall the value of $\Gamma_{2n} q^2 < 1$ is bounded). This indicates that our model captures a dependence on the lengthscale over which the system is perturbed, with highest responses for larger perturbation lengthscales. This observation suggests that we are really probing for collective dynamical responses. We hypothesise that such a perturbation lengthscale also exists in both the microscopic solutions to the hierarchies derived in the previous sections as well as in real structural glass forming materials. This would provide concrete evidence that the nonlinear susceptibilities $\chi_{2n+1}(t)$ are indeed appropriate probes for macroscopic collective dynamical responses.

We recall that in the schematic GMCT, increasing the level N_c of the hierarchy systematically accelerates the dynamics, but at relative distances to the critical points the scaling laws are rigorously preserved. In Fig. 14-15 we verify that the time-evolution of the susceptibilities has unchanged scaling. We find that at fixed ϵ, $q = 0$ and increasing N_c, the curves are simply shifted in time to the right. This holds true for susceptibilities of any order. In the case of large perturbation scales ($q \ll 1$), we notice a two step growth of all dynamical susceptibilities around $\tau_\beta^{(2n)}$ that governed by scaling laws $\chi_{2n+1}(q = 0; t) \sim t^{a_n}$ for $t < \tau_\beta^{(2n)}$ and $\chi_{2n+1}(q = 0; t) \sim t^{b_n}$ for $t > \tau_\beta^{(2n)}$. The exponents a_n, b_n are actually the same as the ones discussed in the previous sections for the behaviour of the correlation functions $\phi_{2n}(t)$ in the β-regime. We have verified this numerically, and we see in Fig. 14-15 that increasing the hierarchy level N_c does not affect the scaling of this growth. While all the high-point non-linear susceptibilities behave critically close to the transition, they are 'polluted' by an increasing number of non-linearities and the window to observe these scalings around $\tau_\beta^{(2n)}$ are shortened, much like for the dynamical correlation functions studied in the previous section. This type of two-step growth behaviour has sometimes been associated to a change in regime from dilute to dense collectively responding structures in the glassy regime [18], but more rigorous studies in this regime are necessary to shed light on this.

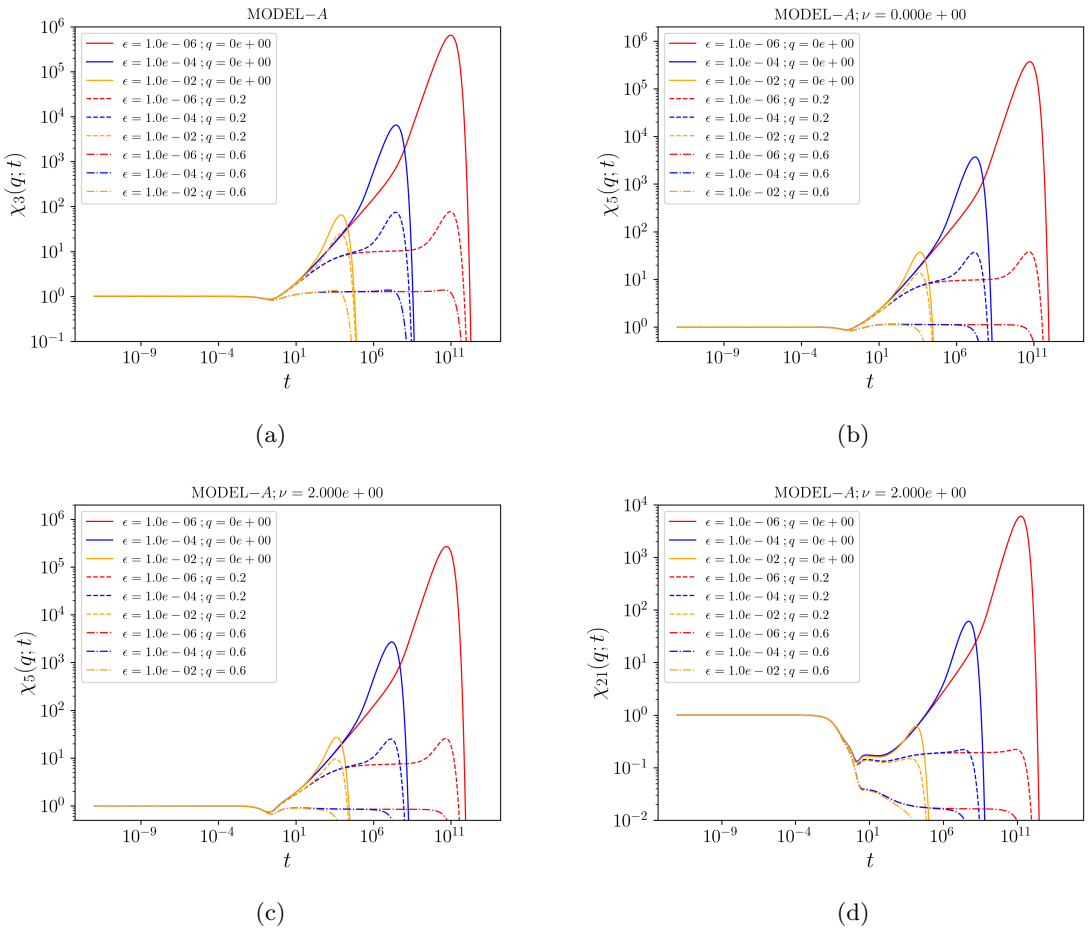
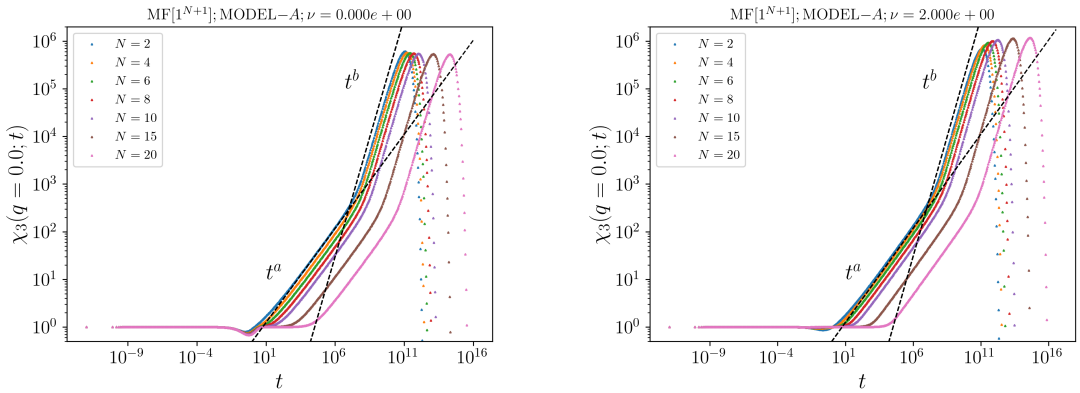
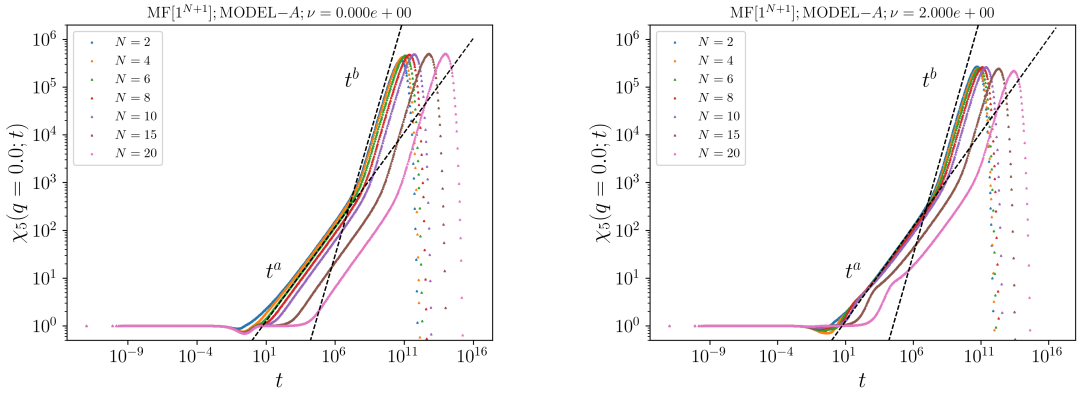


Figure 13: (a) *q-dependence of the 3-point susceptibility for Model A* $\chi_3(q;t)$ *for various relative distances to the critical point* ϵ *at hierarchy level* $N_c = 1$. (b)-(c) *q-dependence of the 5-point susceptibility for Model A* $\chi_5(q;t)$ *for various relative distances to the critical point* ϵ *and parameters* $\nu = 0, 2$ *at hierarchy level* $N_c = 2$.. (d) *q-dependence of the 21-point susceptibility for Model A* $\chi_{21}(q;t)$ *for various relative distances to the critical point* ϵ *and parameters* $\nu = 2$ *at hierarchy level* $N_c = 10$.



(a) (b)

Figure 14: (a)-(b) *behaviour of the three-point susceptibility* χ_3 *at* $q = 0$ *and* $\epsilon =$ 1e-6 *and parameter* $\nu = 0, 2$ *for increasing hierarchy level* N.



(a) (b)

Figure 15: (a)-(b) *Behaviour of the five-point susceptibility* χ_5 *at* $q = 0$ *and* $\epsilon =$ 1e-6 *and parameter* $\nu = 0, 2$ *for increasing hierarchy level* N. *Exponents* $a = 0.395, b = 1$ *are the schematic (G)MCT exponents.*

We now look of $\chi^*_{2n+1}(q)$ the peak of the non-linear susceptibilities as we approach the bifurcation point. At short wavelengths, we find that the peaks of the first few levels stabilises at a constant value of order $\mathcal{O}(10^3 - 10^4)$ for $\epsilon =$ 1e-4, while for larger wavelengths we observe a continuous decay of $\chi^*_{2n+1}(q)$ for $q > 10^{-2}$. As displayed in Fig.16, for large wave-numbers the continuous decay follows a power-law behaviour whose exponent is quantitatively dependent on both the system parametrisation and hierarchy level. We remark however that $\chi^*_{2n+1}(q \to 1) \propto q^{-\Delta_n}$ with $\Delta_n \in [3.7, 3.9]$ fitted numerically for $n = 1, 2$. By taking into account this tail-like behaviour of $\chi^*_{2n+1}(q)$ at long wavelengths, we consider a generalised scaling law also found in [64]

$$\chi^*_{2n+1}(q) \propto \frac{\xi_n^4}{\alpha_n + \beta_n(q\xi_n)^2 + \theta_n(q\xi_n)^4} \tag{77}$$

to which the data is fitted. We note that ξ_n is the 'lengthscale' (should we restore units) associated to the $(2n+1)$-th susceptibility χ_{2n+1}. We anticipate this lengthscale to diverge as well $\xi_m \propto \epsilon^{-\nu_n}$ along with $\chi^*_{2n+1}(q \to 0) \propto \epsilon^{-\psi_n}$. Analysis of $\chi^*_{2n+1}(q \to 0)$ with respect to ϵ for $n = 1$ lead to the conclusion that $\chi^*_3(q \to 0) \times \epsilon \sim \mathcal{O}(1)$. Looking at the scaling of χ_3 with ϵ in detail and find that the associated exponent is $\psi_1 = 1$. Similar analysis performed on the higher-order susceptibilities lead to the conclusion that $\psi_n = 1$ for all levels n tested, independent of MF-closure type as well. Akin to the observations made for the scaling laws of ϕ_{2n}'s, this scaling relation is robust to different parametrisation of models A and B. At low wave-numbers, the quadratic and quartic terms in the denominator of (77) can be neglected, and so we find that $\chi^*_{2n+1}(q) \propto \xi_n^4 \sim \epsilon^{-\psi_n} \Rightarrow \xi_n \propto \epsilon^{-1/4}$, independent of the closure level N_c. Interestingly, these exponents are identical to the ones determined for for fully microscopic calculations of the three-point dynamic susceptibility [64, 52].

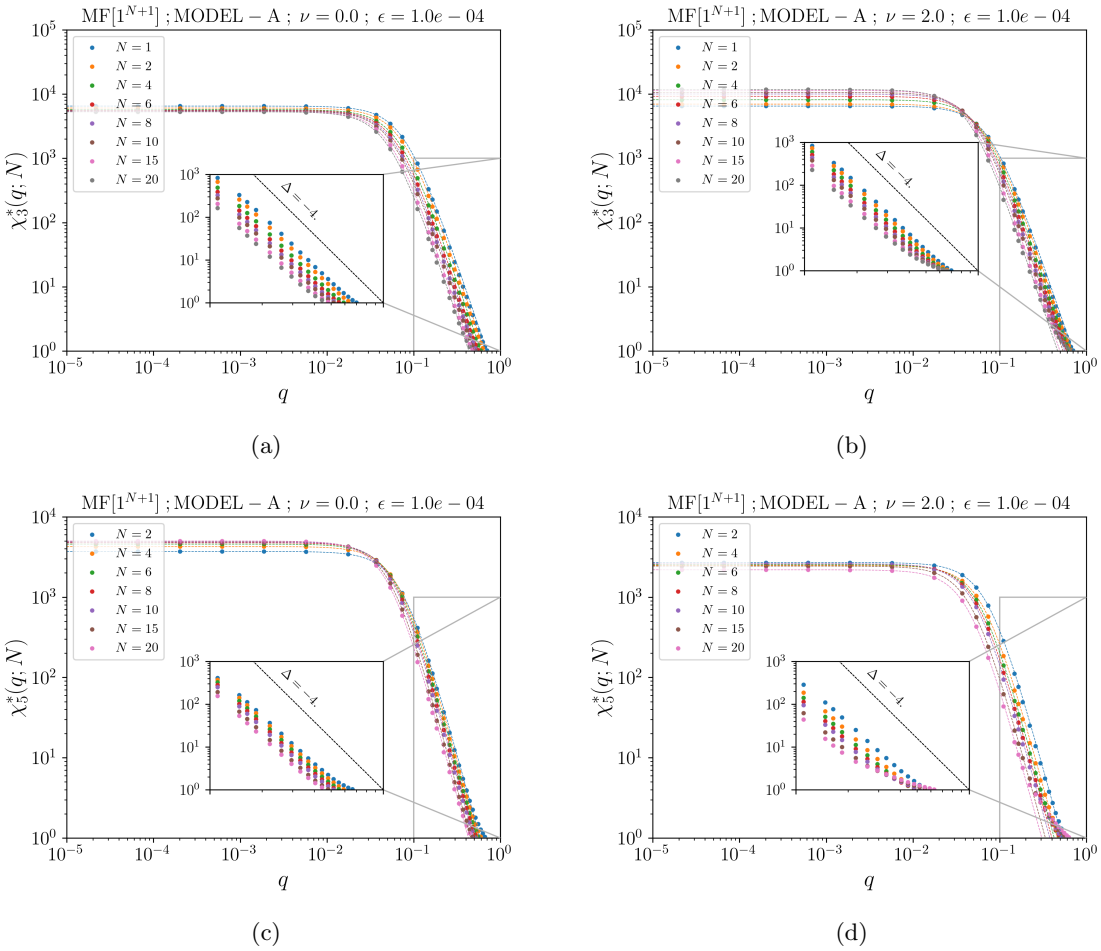


Figure 16: *Scaling of* χ_3^* ((a)-(b)) *and* χ_5^* ((c)-(d)) *for Model A with* $\nu = 0., 2.$ *and various closure levels ranging from 1 to 20. Results fitted with* (77). *Inset to guide the eye at the short-wavelength power-law decay with exponents* Δ_m.

5 Conclusion

In this work we extended the recently derived generalised mode-coupling theory of glass-forming liquids to inhomogeneous environments in the presence of a spatially varying external field. We find that the form of the inhomogeneous and homogeneous hierarchies of order parameters are very similar, the main differences coming in the form of non-translationally invariant quantities. Taking the zero-field limit of this new theory provides a way to consistently obtain off-diagonal Generalised Mode Coupling Theory, which is believed to be an important improvement on diagonal Generalised Mode Coupling Theory. Indeed, the theory developed in [23] employs a diagonal approximation, which restricts some momentum integrations to specific shells in $\mathbf{k}$-space. Regarding inhomogeneous Generalised Mode Coupling Theory as a Landau theory for phase transitions [5], our formalism enables us to systematically take variations of the many-body intermediate scattering functions F_{2m} with respect to the pinning field to obtain equations of motion for the many-body dynamical susceptibilities $\chi_{2m+1}(\mathbf{q})$ where $\mathbf{q}$ is the wave-length of the induced perturbation. much like in spin glasses, these non-linear susceptibilities are excellent candidates to probe for collective effects in the structural glass transition.

From the hierarchical structure of iGMCT, we find that the dynamical susceptibilities form of hierarchy of coupled *linear integro-differential* equations which take as initial conditions full solutions to the homogeneous GMCT. This is the main drawback of solving for the microscopic equations as solving for GMCT is already a herculean task [49, 50]. In the spirit of early GMCT studies [23, 40], we explore numerically a mathematically similar but much simpler hierarchy of coupled integro-differential equations where all wave-vector dependence has been dropped. In this simpler model, we find that that dynamical susceptibilities of any order are critical-like near the A_2 glass transition singularity, and we are able to extract diverging length scales with critical exponents $\nu = 1/4$ for susceptibilities of any order in the limit of $\mathbf{q} \to 0$. Furthermore, we report on a multi-step responses in time to an external perturbation hinting at very complicated dynamical processes.

Future endeavors will have the intention to build on this work for more complicated (polydisperse) glass-formers to investigate growing length-scales in different glass transition scenarios. Accounting for polydispersity will involve a generalisation of the dynamic structure factors and non-linear dynamical susceptibilities to tensor quantities : $F_{2n}^{\alpha_1,\ldots,\alpha_n}$ and $\chi_{2n+1}^{\alpha_1,\ldots,\alpha_n\gamma}$ with the presence of multiple infinitesimal generating terms accounting for each species: $U^{\gamma}(\mathbf{q}_0)$.

Lastly, we point out that carefully designed experiments are capable of measuring these high-point dynamical susceptibilities [12, 2]. It would be interesting to compare these with microscopic results from our susceptibility hierarchy. The novel framework developed in this work promises to build an important bridge between dynamical and thermodynamical theories of the structural glass transition.

A Convolution Approximations

We show the general framework required to derive generalised convolution approximations for high-point static structure factors within the formalism of Density Functional Theory (DFT). These quantities are useful since many-body static structure factors serve as initial conditions for (Generalised) Mode Coupling Theories. We first briefly summarise the results from [8]. We consider a one-component fluid in an external field. We denote Ξ the grand-canonical partition function (GPF), $z(\mathbf{r}) = \Lambda^{-3}\exp[\beta\psi(\mathbf{r})]$ the local activity, with $\psi(\mathbf{r}) = \mu - \varphi(\mathbf{r})$ which are respectively the chemical potential and the external potential. As usual the microscopic density is defined as $\rho(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_j)$. We denote $\mathcal{F}[\rho(\mathbf{r})]$ the free-energy of the system, which is a functional of the density. We now introduce a set of important distribution functions.

1. The n-particle density, denoted $\rho^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n)$ which is generated by derivatives of the GPF

$$\rho^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n) \equiv \frac{\left(\prod_{j=1}^n z(\mathbf{r}_j)\right)}{\Xi} \frac{\delta^n \Xi}{\delta z(\mathbf{r}_1)...\delta z(\mathbf{r}_n)} \tag{78}$$

2. The n-particle distribution function $g^{(n)}$

$$g^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n) = \frac{\rho^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n)}{\prod_{j=1}^n \rho^{(1)}(\mathbf{r}_j)} \tag{79}$$

3. The correlation function $h^{(n)}$

$$h^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n) = \frac{\left(\prod_{j=1}^n z(\mathbf{r}_j)\right)}{\left(\prod_{j=1}^n \rho^{(1)}(\mathbf{r}_j)\right)} \frac{\delta^n \log(\Xi)}{\delta z(\mathbf{r}_1)...\delta z(\mathbf{r}_n)} \tag{80}$$

4. The density correlation function $H^{(n)}$

$$\begin{aligned} H^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n) &= \left\langle \left[\rho(\mathbf{r}_1) - \rho^{(1)}(\mathbf{r}_1)\right]...\left[\rho(\mathbf{r}_n) - \rho^{(1)}(\mathbf{r}_n)\right] \right\rangle \\ &= \frac{\delta^n \log(\Xi)}{\delta \log(z(\mathbf{r}_1))...\delta \log(z(\mathbf{r}_n))} \end{aligned} \tag{81}$$

   Following [39], we note that $H^{(n)}$ is also generated by the Grand potential Ω, for $n \geq 2$:

$$H^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n) = -\frac{\delta\beta\Omega}{\delta\beta\psi(\mathbf{r}_1)...\delta\beta\psi(\mathbf{r}_n)} \tag{82}$$

   and also that this many-body distribution functions is directly proportional to the many-body structure factors that Generalised Mode Coupling Theories take as input : $S_n(\mathbf{k}_1, ..., \mathbf{k}_{n-1}) = \rho^{-1}\tilde{H}^{(n)}(\mathbf{k}_1, ..., \mathbf{k}_{n-1})$.

5. The inverse of $H^{(n)}$, denoted $K^{(n)}$

$$\begin{aligned} K^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n) &= -\frac{\delta^n \beta\mathcal{F}}{\delta\rho^{(1)}(\mathbf{r}_1)...\delta\rho^{(1)}(\mathbf{r}_n)} \\ &= \frac{(-1)^n (n-2)!}{[\rho^{(1)}(\mathbf{r}_1)]^{n-1}} \delta(\mathbf{r}_1 - \mathbf{r}_2)...\delta(\mathbf{r}_1 - \mathbf{r}_n) - c^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n) \end{aligned} \tag{83}$$

and its version in Fourier-space, assuming translational invariance :

$$\tilde{K}^{(n)}(\mathbf{k}_2, ..., \mathbf{k}_n) = \frac{(-1)^n (n-2)!}{\rho_b^{n-1}} - \tilde{c}^{(n)}(\mathbf{k}_2, ..., \mathbf{k}_n) \tag{84}$$

6. The direct correlation function $c^{(n)}$, generated by the excess free energy $\mathcal{F}_{\text{exc}}$

$$c^{(n)}(\mathbf{r}_1, ..., \mathbf{r}_n) = \frac{\delta c^{(n-1)}(\mathbf{r}_1, ..., \mathbf{r}_{n-1})}{\delta \rho^{(1)}(\mathbf{r}_n)} = -\frac{\delta \beta \mathcal{F}_{\text{exc}}}{\delta \rho^{(1)}(\mathbf{r}_1) ... \delta \rho^{(1)}(\mathbf{r}_n)} \tag{85}$$

We note that the following functional identity immediately follows

$$\int d\mathbf{r}_3 K^{(2)}(\mathbf{r}_1, \mathbf{r}_3) H^{(2)}(\mathbf{r}_3, \mathbf{r}_2) = \delta(\mathbf{r}_1 - \mathbf{r}_2) \tag{86}$$

We refer to this equation as OZ-2 from now on. Substituting the results summarised above gives :

$$\int d\mathbf{r}_3 \left(\frac{1}{\rho^{(1)}(\mathbf{r}_1)} \delta(\mathbf{r}_1 - \mathbf{r}_3) - c^{(2)}(\mathbf{r}_1, \mathbf{r}_3) \right) \frac{\delta^2 \log(\Xi)}{\delta \log(z(\mathbf{r}_3)) \delta \log(z(\mathbf{r}_2))} = \delta(\mathbf{r}_1 - \mathbf{r}_2)$$

$$\Rightarrow \int d\mathbf{r}_3 \left(\frac{1}{\rho^{(1)}(\mathbf{r}_1)} \delta(\mathbf{r}_1 - \mathbf{r}_3) - c^{(2)}(\mathbf{r}_1, \mathbf{r}_3) \right) \left(\rho^{(1)}(\mathbf{r}_2) \rho^{(1)}(\mathbf{r}_3) h^{(2)}(\mathbf{r}_2, \mathbf{r}_3) + \rho^{(1)}(\mathbf{r}_2) \delta(\mathbf{r}_2 - \mathbf{r}_3) \right) = \delta(\mathbf{r}_1 - \mathbf{r}_2)$$

$$\Rightarrow h^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = c^{(2)}(\mathbf{r}_1, \mathbf{r}_2) + \int d\mathbf{r}_3 c^{(2)}(\mathbf{r}_1, \mathbf{r}_3) \rho^{(1)}(\mathbf{r}_3) h^{(2)}(\mathbf{r}_2, \mathbf{r}_3)$$

which is the Ornstein Zernike [39] relation ! We seek to generalise to higher order correlation functions in order to derive convolution approximations in momentum space. To do so we consider successive functional derivatives of the functional identity above. For instance:

$$\begin{aligned}
\frac{\delta}{\delta \rho^{(1)}(\mathbf{r}_4)} \int d\mathbf{r}_3 K^{(2)}(\mathbf{r}_1, \mathbf{r}_3) H^{(2)}(\mathbf{r}_3, \mathbf{r}_2) &= \int d\mathbf{r}_3 \left\{ K^{(3)}(\mathbf{r}_4, \mathbf{r}_1, \mathbf{r}_3) H^{(2)}(\mathbf{r}_3, \mathbf{r}_2) + K^{(2)}(\mathbf{r}_1, \mathbf{r}_3) \frac{\delta H^{(2)}(\mathbf{r}_3, \mathbf{r}_2)}{\delta \rho^{(1)}(\mathbf{r}_4)} \right\} \\
&= \int d\mathbf{r}_3 \left\{ K^{(3)}(\mathbf{r}_4, \mathbf{r}_1, \mathbf{r}_3) H^{(2)}(\mathbf{r}_3, \mathbf{r}_2) + \int d\mathbf{r}_5 K^{(2)}(\mathbf{r}_1, \mathbf{r}_3) \frac{\delta \log(z(\mathbf{r}_5))}{\delta \rho^{(1)}(\mathbf{r}_4)} \frac{\delta H^{(2)}(\mathbf{r}_3, \mathbf{r}_2)}{\delta \log(z(\mathbf{r}_5))} \right\} \\
&= \int d\mathbf{r}_3 \left\{ K^{(3)}(\mathbf{r}_4, \mathbf{r}_1, \mathbf{r}_3) H^{(2)}(\mathbf{r}_3, \mathbf{r}_2) + \int d\mathbf{r}_5 K^{(2)}(\mathbf{r}_1, \mathbf{r}_3) H^{(3)}(\mathbf{r}_3, \mathbf{r}_2, \mathbf{r}_5) \frac{\delta \log(z(\mathbf{r}_5))}{\delta \rho^{(1)}(\mathbf{r}_4)} \right\} \\
&= \int d\mathbf{r}_3 \left\{ K^{(3)}(\mathbf{r}_4, \mathbf{r}_1, \mathbf{r}_3) H^{(2)}(\mathbf{r}_3, \mathbf{r}_2) + \int d\mathbf{r}_5 K^{(2)}(\mathbf{r}_1, \mathbf{r}_3) H^{(3)}(\mathbf{r}_3, \mathbf{r}_2, \mathbf{r}_5) K^{(2)}(\mathbf{r}_5, \mathbf{r}_4) \right\}
\end{aligned}$$

and hence, OZ-3 reads:

$$\int d\mathbf{r} \left\{ K^{(3)}(\mathbf{r}_4, \mathbf{r}_1, \mathbf{r}) H^{(2)}(\mathbf{r}, \mathbf{r}_2) + \int d\mathbf{r}' K^{(2)}(\mathbf{r}_1, \mathbf{r}) H^{(3)}(\mathbf{r}, \mathbf{r}_2, \mathbf{r}') K^{(2)}(\mathbf{r}', \mathbf{r}_4) \right\} = 0 \tag{87}$$

which, up to relabelling this result is in agreement with [8]. Assuming translational invariance, OZ-3 (87) above reads in Fourier space after rearranging in favour of $\tilde{H}^{(3)}$:

$$\tilde{H}^{(3)}(\mathbf{k}_1, \mathbf{k}_2) = \frac{-\tilde{K}^{(3)}(\mathbf{k}_1, \mathbf{k}_2) \tilde{H}^{(2)}(\mathbf{k}_2)}{\tilde{K}^{(2)}(\mathbf{k}_1) \tilde{K}^{(2)}(\mathbf{k}_1 + \mathbf{k}_2)}$$

From which we easily obtain:

$$S_3(\mathbf{k}_1, \mathbf{k}_2) \approx S(\mathbf{k}_1)S(\mathbf{k}_2)S(\mathbf{k}_1 + \mathbf{k}_2) \tag{88}$$

by neglecting higher-order direct correlation functions $\tilde{c}^{(n)}$ for $n \geq 3$. To obtain the higher-order convolution approximations, the same method applies.

S_4 **convolution** : We consider a variation of OZ-3 with respect to the density field, which gives OZ-4, then expressed in Fourier space

$$\begin{aligned}\tilde{H}^{(4)}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3) = &- \frac{1}{\tilde{K}^{(2)}(\mathbf{k}_1)\tilde{K}^{(2)}(\mathbf{k}_2)\tilde{K}^{(2)}(\mathbf{k}_3)}\Big[\tilde{K}^{(4)}(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3)\tilde{H}^{(2)}(\mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3) \\ &+\tilde{K}^{(3)}(\mathbf{k}_1, \mathbf{k}_2)\tilde{H}^{(3)}(\mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_3)\tilde{K}^{(2)}(\mathbf{k}_3) + \tilde{K}^{(3)}(\mathbf{k}_1, \mathbf{k}_3)\tilde{H}^{(3)}(\mathbf{k}_1 + \mathbf{k}_3, \mathbf{k}_2)\tilde{K}^{(2)}(\mathbf{k}_2) \\ &+\tilde{K}^{(2)}(\mathbf{k}_1)\tilde{H}^{(3)}(\mathbf{k}_2 + \mathbf{k}_3, \mathbf{k}_1)\tilde{K}^{(3)}(\mathbf{k}_2, \mathbf{k}_3)\Big]\end{aligned}$$

which eventually gives

$$S_4(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4) = S(\mathbf{k}_1)S(\mathbf{k}_2)S(\mathbf{k}_3)S(\mathbf{k}_4)\Big(S(\mathbf{k}_1 + \mathbf{k}_2) + S(\mathbf{k}_1 + \mathbf{k}_3) + S(\mathbf{k}_2 + \mathbf{k}_3) - 2\Big) \tag{89}$$

We summarise below the results for the next two orders.

S_5 **convolution** :

$$\begin{aligned}S_5(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4, \mathbf{k}_5) =& S(\mathbf{k}_1)S(\mathbf{k}_2)S(\mathbf{k}_3)S(\mathbf{k}_5)\Big\{6S(\mathbf{k}_4) - 2\Big(\frac{S_3(\mathbf{k}_4, \mathbf{k}_1 + \mathbf{k}_2 + \mathbf{k}_3)}{S(\mathbf{k}_5)} + \frac{S_3(\mathbf{k}_1, \mathbf{k}_4)}{S(\mathbf{k}_1)} \\ &+ \frac{S_3(\mathbf{k}_2, \mathbf{k}_4)}{S(\mathbf{k}_2)} + \frac{S_3(\mathbf{k}_3, \mathbf{k}_4)}{S(\mathbf{k}_3)}\Big) - S_3(\mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_4) - S_3(\mathbf{k}_1 + \mathbf{k}_3, \mathbf{k}_4) - S_3(\mathbf{k}_2 + \mathbf{k}_3, \mathbf{k}_4) \\ &+ \frac{1}{S(\mathbf{k}_5)}\Big(\frac{S_4(\mathbf{k}_1 + \mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4)}{S(\mathbf{k}_3)} + \frac{S_4(\mathbf{k}_1 + \mathbf{k}_3, \mathbf{k}_2, \mathbf{k}_4)}{S(\mathbf{k}_2)} + \frac{S_4(\mathbf{k}_2 + \mathbf{k}_3, \mathbf{k}_1, \mathbf{k}_4)}{S(\mathbf{k}_1)}\Big) \\ &+ \frac{S_4(\mathbf{k}_2, \mathbf{k}_3, \mathbf{k}_4)}{S(\mathbf{k}_2)S(\mathbf{k}_3)} + \frac{S_4(\mathbf{k}_1, \mathbf{k}_2, \mathbf{k}_4)}{S(\mathbf{k}_1)S(\mathbf{k}_2)} + \frac{S_4(\mathbf{k}_1, \mathbf{k}_3, \mathbf{k}_4)}{S(\mathbf{k}_1)S(\mathbf{k}_3)}\Big\}\end{aligned}$$

S_6 **convolution :**

$$
\begin{aligned}
&S_6(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,\mathbf{k}_5)\\
&= S(\mathbf{k}_2)S(\mathbf{k}_3)S(\mathbf{k}_4)S(\mathbf{k}_5)S(\mathbf{k}_6)\Bigg\{-24S(\mathbf{k}_1)+6\Bigg(\frac{S_3(\mathbf{k}_1,\mathbf{k}_5)}{S(\mathbf{k}_5)}+\frac{S_3(\mathbf{k}_1,\mathbf{k}_4)}{S(\mathbf{k}_4)}+\frac{S_3(\mathbf{k}_1,\mathbf{k}_3)}{S(\mathbf{k}_3)}\\
&+\frac{S_3(\mathbf{k}_1,\mathbf{k}_2)}{S(\mathbf{k}_2)}+\frac{S_3(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4+\mathbf{k}_5)}{S(\mathbf{k}_6)}\Bigg)+2\Bigg(S_3(\mathbf{k}_1,\mathbf{k}_4+\mathbf{k}_5)+S_3(\mathbf{k}_1,\mathbf{k}_3+\mathbf{k}_5)\\
&+S_3(\mathbf{k}_1,\mathbf{k}_3+\mathbf{k}_4)+S_3(\mathbf{k}_1,\mathbf{k}_3+\mathbf{k}_4+\mathbf{k}_5)+S_3(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_5)+S_3(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_4)\\
&+S_3(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_4+\mathbf{k}_5)+S_3(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_3)+S_3(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_5)+S_3(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4)\Bigg)\\
&-2\Bigg(\frac{S_4(\mathbf{k}_1,\mathbf{k}_4,\mathbf{k}_5)}{S(\mathbf{k}_4)S(\mathbf{k}_5)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_5)}{S(\mathbf{k}_3)S(\mathbf{k}_5)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_4)}{S(\mathbf{k}_3)S(\mathbf{k}_4)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_5)}{S(\mathbf{k}_2)S(\mathbf{k}_5)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_4)}{S(\mathbf{k}_4)S(\mathbf{k}_2)}\\
&+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3)}{S(\mathbf{k}_2)S(\mathbf{k}_3)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4+\mathbf{k}_5)}{S(\mathbf{k}_2)S(\mathbf{k}_3+\mathbf{k}_4+\mathbf{k}_5)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_2+\mathbf{k}_4+\mathbf{k}_5)}{S(\mathbf{k}_3)S(\mathbf{k}_2+\mathbf{k}_4+\mathbf{k}_5)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_4,\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_5)}{S(\mathbf{k}_4)S(\mathbf{k}_6)}\\
&+\frac{S_4(\mathbf{k}_1,\mathbf{k}_5,\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4)}{S(\mathbf{k}_5)S(\mathbf{k}_6)}\Bigg)-\Bigg(\frac{S_4(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_4+\mathbf{k}_5)}{S(\mathbf{k}_3)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_4,\mathbf{k}_3+\mathbf{k}_5)}{S(\mathbf{k}_4)}\\
&+\frac{S_4(\mathbf{k}_1,\mathbf{k}_5,\mathbf{k}_3+\mathbf{k}_4)}{S(\mathbf{k}_5)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_4+\mathbf{k}_5)}{S(\mathbf{k}_2)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_4,\mathbf{k}_2+\mathbf{k}_5)}{S(\mathbf{k}_4)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_5,\mathbf{k}_2+\mathbf{k}_4)}{S(\mathbf{k}_5)}\\
&+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_5)}{S(\mathbf{k}_2)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_2+\mathbf{k}_5)}{S(\mathbf{k}_3)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3+\mathbf{k}_4)}{S(\mathbf{k}_2)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_3+\mathbf{k}_4,\mathbf{k}_3+\mathbf{k}_5)}{S(\mathbf{k}_6)}\\
&+\frac{S_4(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_2+\mathbf{k}_4)}{S(\mathbf{k}_3)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_3+\mathbf{k}_5,\mathbf{k}_2+\mathbf{k}_4)}{S(\mathbf{k}_5)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_3,\mathbf{k}_5)}{S(\mathbf{k}_5)}+\frac{S_4(\mathbf{k}_1,\mathbf{k}_4,\mathbf{k}_2+\mathbf{k}_3)}{S(\mathbf{k}_4)}\\
&+\frac{S_4(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_3,\mathbf{k}_4+\mathbf{k}_5)}{S(\mathbf{k}_6)}\Bigg)+\frac{S_5(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_4,\mathbf{k}_5)}{S(\mathbf{k}_2)S(\mathbf{k}_4)S(\mathbf{k}_5)}+\frac{S_5(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_5)}{S(\mathbf{k}_2)S(\mathbf{k}_3)S(\mathbf{k}_5)}+\frac{S_5(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4)}{S(\mathbf{k}_2)S(\mathbf{k}_3)S(\mathbf{k}_4)}\\
&+\frac{S_5(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4+\mathbf{k}_5)}{S(\mathbf{k}_2)S(\mathbf{k}_3)S(\mathbf{k}_6)}+\frac{S_5(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_4,\mathbf{k}_3+\mathbf{k}_5)}{S(\mathbf{k}_2)S(\mathbf{k}_4)S(\mathbf{k}_6)}+\frac{S_5(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_4,\mathbf{k}_2+\mathbf{k}_5)}{S(\mathbf{k}_3)S(\mathbf{k}_4)S(\mathbf{k}_6)}\\
&+\frac{S_5(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_5,\mathbf{k}_3+\mathbf{k}_4)}{S(\mathbf{k}_6)S(\mathbf{k}_2)S(\mathbf{k}_5)}+\frac{S_5(\mathbf{k}_1,\mathbf{k}_2+\mathbf{k}_3,\mathbf{k}_4,\mathbf{k}_5)}{S(\mathbf{k}_4)S(\mathbf{k}_5)S(\mathbf{k}_6)}+\frac{S_5(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_5,\mathbf{k}_2+\mathbf{k}_4)}{S(\mathbf{k}_6)S(\mathbf{k}_3)S(\mathbf{k}_5)}+\frac{S_5(\mathbf{k}_1,\mathbf{k}_3,\mathbf{k}_4,\mathbf{k}_5)}{S(\mathbf{k}_3)S(\mathbf{k}_4)S(\mathbf{k}_5)}\Bigg\}
\end{aligned}
$$

We note that the equations for $S_{5,6}$ above can be further simplified by plugging in the appropriate convolution approximations of lower order.

B Numerical Methods for Schematic Models & Parametrisation Details

We outline the numerical algorithm used to solve the schematic GMCT and schematic dynamical susceptibilities. The essence of the numerical technique lies in the treatment of the convolution integral, where we use:

$$\begin{aligned}
\int_0^t duM(t-u)\dot{F}(u) &= \int_0^{t_2} duM(t-u)\dot{F}(u) + \int_{t_2}^t duM(t-u)\dot{F}(u) \\
&= M(t-t_2)F(t_2) - M(t)F(0) - \int_0^{t_2} du\dot{M}(t-u)F(u) + \int_{t_2}^t duM(t-u)\dot{F}(u) \\
&= M(t-t_2)F(t_2) - M(t)F(0) - \sum_{j=1}^{N}\int_{t_{j-1}}^{t_j} du\dot{M}(t-u)F(u) - \sum_{j=1}^{N'}\int_{t_{j-1}}^{t_j} duM(u)\dot{F}(t-u) \\
&\approx M(t_{i-i_2})F(t_{i_2}) - M(t_i)F(0) - \sum_{j=1}^{N}(M(t_{i-j}) - M(t_{i-j+1}))\mathcal{I}[F(t_j)] \\
&\quad - \sum_{j=1}^{N'}(F(t_{i-j}) - F(t_{i-j+1}))\mathcal{I}[M(t_j)]
\end{aligned} \tag{90}$$

where the sum limits satisfy $N\Delta t = t_2$ and $N'\Delta t = t - t_2$. WLOG we will assume $\Delta t = 1$. We have also used

$$\int_{t_{j-1}}^{t_j} du\dot{\alpha}(u)\beta(u) \approx \frac{1}{2}(\alpha(t_j) - \alpha(t_{j-1}))(\beta(t_j) + \beta(t_{j-1})) \tag{91}$$

We can re-write the hierarchies (73), (74) in the form $A\phi_{2n}(t_i) + BM_{2n}^{(\phi)}(t_i) + C = 0$ and $A_X\chi_{2n+1}(t_i) + B_X M_{2n}^{(\chi)}(t_i) + C_X = 0$. The exact coefficients are

$$A = \left(\mu_n + \frac{3\nu_n}{2\delta t} + \frac{2}{\delta t^2}\right) + \Lambda_n\mathcal{I}[M_n^{(\phi)}(t_1)] \tag{92}$$

$$B = -\Lambda_n\phi_n(0) + \Lambda_n\mathcal{I}[\phi_n(t_1)] \tag{93}$$

$$\begin{aligned}
C &= -\frac{2\nu_n}{\delta t}\phi_n(t_{i-1}) + \frac{\nu_n}{2\delta t}\phi_n(t_{i-2}) - \frac{5}{\delta t^2}\phi_n(t_{i-1}) + \frac{4}{\delta t^2}\phi_n(t_{i-2}) - \frac{1}{\delta t^2}\phi_n(t_{i-3}) \\
&+ \Lambda_n\left(M_n^{\phi}(t_{i-i_2})\phi_n(t_{i_2}) - M_n^{(\phi)}(t_{i-1})\mathcal{I}[\phi_n(t_1)] - \phi_n(t_{i-1})\mathcal{I}[M_n^{(\phi)}(t_1)]\right) \\
&- \Lambda_n\sum_{j=2}^{i_2}\left(M_n^{(\phi)}(t_{i-j}) - M_n^{(\phi)}(t_{i-j+1})\right)\mathcal{I}[\phi_n(t_j)] \\
&- \Lambda_n\sum_{j=2}^{i-i_2}\left(\phi(t_{i-j}) - \phi(t_{i-j+1})\right)\mathcal{I}[M_n^{(\phi)}(t_j)]
\end{aligned} \tag{94}$$

for the hierarchy of the dynamical correlation functions and are

$$A_X = \left(\mu_n + \frac{3\nu_n}{2\delta t} + \frac{2}{\delta t^2}\right) + \Lambda_n \mathcal{I}[M_n^{(\phi)}(t_1)] \tag{95}$$

$$B_X = -\lambda_n(q)\phi_n(0) + \lambda_n(q)\mathcal{I}[\phi_n(t_1)] \tag{96}$$

$$\begin{aligned}
C_X = &-\frac{2\nu_n}{\delta t}\chi_{2n+1}(t_{i-1}) - \frac{5}{\delta t^2}\chi_{2n+1}(t_{i-1}) + \frac{\nu_n}{2\delta t}\chi_{2n+1}(t_{i-2}) + \frac{4}{\delta t^2}\chi_{2n+1}(t_{i-i_2}) - \frac{1}{\delta t^2}\chi_{2n+1}(t_{i-3}) + \bar{\mu}_n\phi_n(t_i) \\
&+ \Lambda_n\left(M_n^{(\phi)}(t_{i-i_2})\phi(t_{i_2}) - M_n^{(\phi)}(t_i)\phi_n(0) - (M_n^{(\phi)}(t_{i-1}) - M_n^{(\phi)}(t_i))\mathcal{I}[\phi_n(t_1)] - (\phi_n(t_{i-1}) - \phi_n(t_i))\mathcal{I}[M_n^{(\phi)}(t_1)]\right) \\
&+ \Lambda_n\Bigg(M_n^{(\phi)}(t_{i-i_2})\chi_{2n+1}(t_{i_2}) - M_n^{(\phi)}(t_{i-1})\mathcal{I}[\chi_{2n+1}(t_1)] - \chi_{2n+1}(t_{i-1})\mathcal{I}[M_n^{(\phi)}(t_1)] \\
&\qquad - M_n^{(\phi)}(t_i)\chi_{2n+1}(0) + M_n^{(\phi)}(t_i)\mathcal{I}[\chi_{2n+1}(t_1)]\Bigg) \\
&+ \lambda_n(q)\left(M_n^{(\chi)}(t_{i-i_2})\phi_n(t_{i_2}) - M_n^{(\chi)}(t_{i-1})\mathcal{I}[\phi_n(t_1)] - \phi_n(t_{i-1})\mathcal{I}[M_n^{(\chi)}(t_1)] + \phi_n(t_i)\mathcal{I}[M_n^{(\chi)}(t_1)]\right) \\
&- \Lambda_n\sum_{j=2}^{i_2}\left(M_n^{(\phi)}(t_{i-j}) - M_n^{(\phi)}(t_{i-j+1})\right)\mathcal{I}[\chi_{2n+1}(t_j)] \\
&- \Lambda_n\sum_{j=2}^{i-i_2}\left(\chi_{2n+1}(t_{i-j}) - \chi_{2n+1}(t_{i-j+1})\right)\mathcal{I}[M_n^{(\phi)}(t_j)] \\
&- \Lambda_n\sum_{j=2}^{i_2}\left(M_n^{(\phi)}(t_{i-j}) - M_n^{(\phi)}(t_{i-j+1})\right)\mathcal{I}[\phi_n(t_j)] \\
&- \Lambda_n\sum_{j=2}^{i-i_2}\left(\phi(t_{i-j}) - \phi(t_{i-j+1})\right)\mathcal{I}[M_n^{(\phi)}(t_j)] \\
&- \lambda_n(q)\sum_{j=2}^{i_2}\left(M_n^{(\chi)}(t_{i-j}) - M_n^{(\chi)}(t_{i-j+1})\right)\mathcal{I}[\phi_n(t_j)] \\
&- \lambda_n(q)\sum_{j=2}^{i-i_2}\left(\phi_n(t_{i-j}) - \phi_n(t_{i-j+1})\right)\mathcal{I}[M_n^{(\chi)}(t_j)]
\end{aligned} \tag{97}$$

for the dynamical susceptibilities. The algorithm requires as input the first 2N points of the solution for $\chi_{2n+1}(q;t)$. We do so by considering a Taylor development.

$$\begin{aligned}
\phi_n(t) &= \phi_n(0) + \dot{\phi}_n(0)t + \frac{1}{2}\ddot{\phi}_n(0)t^2 + \mathcal{O}(t^3) \\
\chi_{2n+1}(t) &= \chi_{2n+1}(0) + \dot{\chi}_{2n+1}(0)t + \frac{1}{2}\ddot{\chi}_{2n+1}(0)t^2 + \mathcal{O}(t^3)
\end{aligned} \tag{98}$$

OVERDAMPED scheme : then we have as initial conditions $\phi_n(0) = \chi_{2n+1}(0) = 1$. From the e.o.m. we determine that

$$
\begin{aligned}
\phi_n(t) &= 1 - \frac{\mu_n}{\nu_n} t + \frac{\mu_n}{\nu_n^2} \left[\mu_n + M_m^{(\phi)}(0) \right] t^2 \\
\chi_{2n+1}(t) &= 1 - \frac{\mu_n + \bar{\mu}_n}{\nu_n} t + \frac{\mu_n}{\nu_n^2} \left[\mu_n + \bar{\mu}_n + \mu_n \bar{\mu}_n + M_n^{(\chi)}(0) \right] t^2 + \frac{\mu_n + \bar{\mu}_n}{\nu_n^2} M_n^{(\phi)}(0) t^2
\end{aligned}
\tag{99}
$$

UNDERDAMPED scheme : then we have as initial conditions $\phi_n(0) = \chi_{2n+1}(0) = 1$ and $\dot{\phi}_n(0) = \dot{\chi}_{2n+1}(0) = 0$. This leads to the following initialisation terms :

$$
\begin{aligned}
\phi_n(t) &= 1 - \frac{mu_n}{2} t^2 \\
\chi_{2n+1}(t) &= 1 - \frac{\mu_n + \bar{\mu}_n}{2} t^2
\end{aligned}
\tag{100}
$$

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