

Anomalous non-linear response of glassy liquids: general arguments and a Mode-Coupling approach

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Abstract

We study theoretically the non-linear response properties of glass formers. We establish several general results which, together with the assumption of Time-Temperature Superposition, lead to a relation between the non-linear response and the derivative of the linear response with respect to temperature. Using results from Mode-Coupling Theory (MCT) and scaling arguments valid close to the glass transition, we obtain the frequency and temperature dependence of the non-linear response in the α and β -regimes. These results are extended to the case of inhomogeneous perturbing fields and confirmed by an exact analysis of the non-linear response of the spherical p-spin model.

I. INTRODUCTION AND MOTIVATIONS

There is mounting evidence that the glass transition is a collective phenomenon, possibly related to the appearance and growth of *amorphous long range order* [1, 2, 3]. Standard two-body correlations are blind to this strange type of order and declare the glass to be, structurally, a frozen liquid. The signature of such an exotic scenario must be sought for in higher order, spatio-temporal correlation functions. A natural candidate, which also appears in the context of spin-glasses, is the four-body correlation function which can be thought of as the spatial correlation of the local two body temporal correlation [4, 5]. Its integral over space defines a ‘dynamical susceptibility’ called χ_4 , which has been intensively studied in the past few years, both theoretically and numerically [6, 7, 8, 9, 10, 11]. One finds that χ_4 reaches a peak value for time scales of the order of the relaxation time of the system τ_α , and the height of this peak increases as the temperature is reduced, as a clear sign of the growth of some dynamical correlation length as the glass transition is approached. From an experimental point of view, however, four-point correlation functions are very difficult to measure directly, except in cases where one can monitor the trajectory of individual particles – for example granular systems where χ_4 can be measured directly and again shows interesting features as the system jams [12, 13, 14]. It is therefore important to investigate alternative quantities that can both be measured experimentally and probe the non standard nature of the glassy correlations.

One possibility, advocated in [15], is to study the non linear response to a small external field. In spin-glasses, it is well known that the (cubic) non-linear magnetic susceptibility $\chi_3(\omega)$ is the natural probe for the appearance of spin-glass order [16, 17, 18]. The static non-linear susceptibility actually diverges at the spin-glass transition, signaling the appearance of long range amorphous order in these systems. For glasses, it was argued in [15], on the basis of physical and heuristic arguments, that the non-linear dielectric susceptibility $\chi_3(\omega)$ should exhibit a growing peak around $\omega\tau_\alpha = 1$, while $\chi_3(\omega = 0)$ should remain trivial, in contrast with the case of spin-glasses. However, the detailed shape of $\chi_3(\omega)$ in the glassy region is beyond the grasp of these heuristic arguments. Since the corresponding experiments are currently being performed [19, 20], it is quite important to get more precise predictions on the expected shape of $\chi_3(\omega)$. This is the primary aim of the present study, where we obtain for the first time, within the context of the Mode Coupling Theory (MCT) of the

glass transition, some precise information on the non-linear susceptibility, concerning both its frequency and temperature dependence.

Another interesting quantity, extensively studied in the past few years, is the derivative of the standard two-body correlation $C(\tau)$ or susceptibility $\chi_1(\omega)$ with respect to the temperature (or the density) – a quantity called $\chi_T = T\partial C(\tau)/\partial T$ (or $\chi_\rho = \rho\partial C(\tau)/\partial\rho$) in [21]. This is clearly an easily accessible quantity, which also shows a peak at times of the order of τ_α and whose height grows as the temperature is lowered [21, 22]. This has led to direct estimates of the size of the dynamical correlation length in supercooled liquids and glasses [21, 22, 23]. The relation between χ_4 and χ_T is however highly non-trivial and has been investigated thoroughly in [10, 11]. It was realized in these papers that the existence of conserved quantities (energy, density) crucially affects the properties of χ_4 , which depends both on the thermodynamic ensemble (NVE vs. NPT for example) and on the dynamics (Brownian vs. Newtonian for example). The true glassy correlation length, on the other hand, does not depend on these choices, and therefore the direct interpretation of $\chi_4(t)$ in terms of a correlation volume is somewhat obscured. At a deeper level, the basic ingredient leading to the critical behaviour of χ_4 turns out to be entirely contained in the response function χ_T itself, as the field theoretical analysis of [10] explicitly demonstrates and the numerical results presented there fully confirm. For example, for Brownian dynamics or for Newtonian dynamics in the NVE ensemble, $\chi_4 \approx \chi_T$, whereas for Newtonian dynamics in the NVT ensemble, one rather finds $\chi_4 \approx \chi_T^2$, see [10, 11] for a detailed discussion.

A natural question is therefore the relation between the non-linear susceptibility $\chi_3(\omega)$ and the dynamical susceptibilities χ_T and χ_4 . The conclusion of this present study is that the complications brought about by conserved quantities that affect χ_4 *do not* contribute to the non-linear susceptibility which is again controlled by the critical properties of χ_T . We establish simple identities between χ_3 and χ_T which hold whenever Time-Temperature superposition holds, i.e. all dependence of the linear response of the system on external parameters (temperature, density, electric field, ...) comes through the dependence of the relaxation time on these parameters. This is a strict statement within MCT, where a true dynamical phase transition takes place.

Whenever this property holds, our central result is that $\chi_3(\omega)$ takes in the α -region the

scaling form conjectured in [15]:

$$\chi_3(\omega) \simeq \xi_\alpha^{2-\bar{\eta}} \mathcal{G}(\omega\tau_\alpha), \quad (1)$$

where ξ_α is the dynamical correlation length which also appears in χ_T , and $\bar{\eta}$ a certain critical exponent that within MCT is equal to minus 2.

For symmetry reasons, the quadratic non-linear susceptibility $\chi_2(\omega)$ is zero for unpolarized systems. This would not be the case for example for polarized systems, or when considering the response to a density perturbation. The non-linear response to a density perturbation contains a quadratic term. The arguments presented below make it clear that in that case $\chi_2(\omega)$ itself is directly related to χ_T . We therefore expect that all the results presented below apply to describe the long-wavelength ($q \rightarrow 0$) non-linear compressibility of supercooled liquids or colloids. By adapting the arguments presented below to the explicit results obtained for Inhomogenous Mode Coupling Theory [24] we also extend these results to finite wavevectors q .

The organisation of the paper is as follows. We first introduce the theoretical framework needed to deal with non-linear response to an external field and establish some general relations between different quantities that naturally appear (Section II). We then exploit the Time-Temperature superposition (TTS) properties of the correlation function of glassy system to establish the scaling form of the non-linear susceptibility (Section III). Using scaling arguments, explicit expressions can be easily derived with the context of MCT, where TTS holds asymptotically. We summarize our central results at the end of Section III, see in particular Fig. 1. In the second, more technical part of the paper (Section IV), we provide a more rigorous analysis of the Mode-Coupling equations in the presence of an external field, and analyze the solutions to second order in the field, in order to obtain in a more rigorous way the results given in Section III on the basis of a scaling analysis. We end by a conclusion with open problems, possible extensions and experimental suggestions.

II. NON-LINEAR SUSCEPTIBILITY: GENERAL FRAMEWORK

In this Section, we introduce the formalism needed to deal with non-linear response and establish a general relation between the non-linear susceptibility and a dynamical response function recently introduced in the literature, which was argued to capture the critical spatio-

temporal correlations of the dynamics in the glassy region. In order to remain close to recent and ongoing experiments on glycerol, we use below the language of dielectric susceptibility. However, as mentioned in the Introduction, our arguments and results apply to more general non-linear susceptibilities (mechanical, magnetic, etc.).

A. Linear and non linear response: small field expansion

Let us consider a dipolar molecular liquid in presence of a small external electric field oscillating at frequency ω in the z -direction. We denote it as:

$$\mathbf{E}(t) = \mathbf{z}E(t) \equiv \mathbf{z}E \cos(\omega t), \quad (2)$$

where \mathbf{z} is the unit vector in the z direction and $E(t) = E \cos(\omega t)$ is its z -component with peak field amplitude E

When the external field is sufficiently small, the polarization vector (per particle) $\mathbf{P}(t, E)$ can be expanded in powers of E . In the following we will denote $P(t, E)$ its z -component. Due to the rotational symmetry in the x - y plane the other components are identically zero. Furthermore, because of the up-down symmetry in the z direction, the polarization must be an odd function of E , i.e., $P(t, -E) = -P(t, E)$. As a consequence, the expansion of P in powers of E contains only odd terms:

$$P(t, E) = P_1(t)E + P_3(t)E^3 + O(E^5), \quad (3)$$

where $P_1(t)$ and $P_3(t)$ can be expressed as functional derivatives of the magnetization with respect to the external field:

$$\begin{aligned} P_1(t) &\equiv \int_{t_1 < t} dt_1 \left. \frac{\delta P(t)}{\delta E(t_1)} \right|_{E=0} \cos(\omega t_1) \\ P_3(t) &= \frac{1}{6} \int_{t_1, t_2, t_3 < t} dt_1 dt_2 dt_3 \left. \frac{\delta^3 P(t)}{\delta E(t_1) \delta E(t_2) \delta E(t_3)} \right|_{E=0} \cos(\omega t_1) \cos(\omega t_2) \cos(\omega t_3). \end{aligned} \quad (4)$$

It is important to remark that the linear and non-linear response kernels in the above integrals are time translation invariant (TTI), i.e. they do not change if all time variables are shifted by the same amount. This comes from the fact that they are *equilibrium* response functions. Using this result and the specific form of the external field, Eq. (2), one finds:

$$P(t, E) = E \Re(\chi_1(\omega) e^{i\omega t}) + \frac{E^3}{4} \Re(\chi_{1,2}(\omega) e^{i\omega t} + \chi_3(\omega) e^{3i\omega t}) + O(E^5). \quad (5)$$

which defines the usual frequency dependent linear susceptibility, $\chi_1(\omega)$, and the frequency dependent non-linear susceptibility, $\chi_3(\omega)$, while $\chi_{1,2}(\omega)$ is the E^2 correction to the first harmonic susceptibility $\chi_1(\omega)$.

Following the same procedure, one can expand the (z -component) polarization correlation and linear response functions in powers of the electric field. The up-down symmetry in the z direction implies that they both are even functions of E . Therefore their expansion in power of E contains only even terms:

$$\begin{aligned} C(t, t') &= C_0(t, t') + C_2(t, t')E^2 + O(E^4) \\ R(t, t') &= R_0(t, t') + R_2(t, t')E^2 + O(E^4). \end{aligned} \quad (6)$$

C_0 and R_0 are the unperturbed correlation and response functions in absence of the external field. At equilibrium, they are functions only of the time difference $\tau = t - t' \geq 0$: $C_0(t, t') = C_0(t - t')$ and $R_0(t, t') = R_0(t - t')$. Moreover, the Fluctuation-Dissipation theorem (FDT) holds for the unperturbed correlation and response functions:

$$R_0(\tau) = -\frac{1}{T} \frac{\partial C_0(\tau)}{\partial \tau}. \quad (7)$$

The second-order correlation and response functions appearing in Eq. (6) are defined as:

$$C_2(t, t') = \frac{1}{2} \int_{t_1, t_2 < t} dt_1 dt_2 \left. \frac{\delta^2 C(t, t')}{\delta E(t_1) \delta E(t_2)} \right|_{E=0} \cos(\omega t_1) \cos(\omega t_2). \quad (8)$$

$$R_2(t, t') = \frac{1}{2} \int_{t_1, t_2 < t} dt_1 dt_2 \left. \frac{\delta^2 R(t, t')}{\delta E(t_1) \delta E(t_2)} \right|_{E=0} \cos(\omega t_1) \cos(\omega t_2). \quad (9)$$

The second order correlation function, $C_2(t, t')$, was introduced in the context of spin-glasses by Huse [18] in the static limit, and more recently studied in details in [26, 27]. Neither TTI nor FDT holds for $C_2(t, t')$ and $R_2(t, t')$, which are explicit functions of both t and t' . However, the response kernels appearing inside the above integrals are TTI. Therefore, one finds that $C_2(t, t')$ and $R_2(t, t')$ are periodic function in the time variable $t + t'$ with period $2\pi/\omega$. As a consequence, they be expanded in Fourier series as follows:

$$\begin{aligned} C_2(t, t') &= \sum_{n=-\infty}^{\infty} e^{in\omega(t+t')} c_n^{(\omega)}(t - t') \\ R_2(t, t') &= \sum_{n=-\infty}^{\infty} e^{in\omega(t+t')} r_n^{(\omega)}(t - t'). \end{aligned} \quad (10)$$

Since the correlation and response function have to be real, we also have that $c_n^{(\omega)}(\tau) = c_{-n}^{(\omega)}(\tau)^*$ and $r_n^{(\omega)}(\tau) = r_{-n}^{(\omega)}(\tau)^*$.

B. Relation between non-linear susceptibility and second-order response

In the following we aim at establishing a relation between the second order response function defined above and the non-linear susceptibility. By definition, the electric polarization is given by the convolution of the response function with the external field:

$$P(t) = \int_{-\infty}^t dt' R(t, t') E(t'). \quad (11)$$

Therefore, using Eqs. (3) and (6), we simply get that:

$$EP_1(t) = \int_{-\infty}^t dt' R_0(t - t') E(t'), \quad (12)$$

and

$$EP_3(t) = \int_{-\infty}^t dt' R_2(t, t') E(t'). \quad (13)$$

Thus, the component of order E^3 of the polarization (related to χ_3) turns out to be just the convolution of the field with the function $R_2(t, t')$ defined in Eq. (8). As a consequence, using the above expression, together with the Fourier expansion Eq. (10) up to order E^3 , we find, for an oscillating field at frequency ω :

$$P_3(t) = \frac{1}{2} \left\{ \left[e^{i\omega t} \left(\tilde{r}_0^{(\omega)}(\omega) + \tilde{r}_1^{(\omega)}(0) \right) + \text{c.c.} \right] + \left[e^{3i\omega t} \tilde{r}_1^{(\omega)}(2\omega) + \text{c.c.} \right] \right\}, \quad (14)$$

where we denoted $\tilde{r}_0^{(\omega)}(\omega')$ and $\tilde{r}_1^{(\omega)}(\omega')$ the semi-Fourier transform (with respect to τ) at frequency ω' of the coefficients appearing in the Fourier expansion, $r_0^{(\omega)}(\tau)$ and $r_1^{(\omega)}(\tau)$. The previous equation allows us to establish a general relation between $\chi_3(\omega)$ and the Fourier transform of $r_{n=1}^{(\omega)}(\tau)$:

$$\chi_3(\omega) = 4 \tilde{r}_1^{(\omega)}(2\omega) = 4 \int_0^{\infty} d\tau e^{-2i\omega\tau} r_1^{(\omega)}(\tau). \quad (15)$$

This relation will be very useful. Using scaling arguments we will now obtain the critical behavior of $r_1^{(\omega)}(\tau)$ within MCT. The relation above will then allow us to obtain straightforwardly the scaling behaviour of $\chi_3(\omega)$.

C. Analysis of the low frequency limit

In the following we focus on the evolution of the correlation and response function for time differences $\tau = t - t'$ much smaller than the period of oscillation of the external field,

in other words in the low-frequency limit $\omega\tau \ll 1$. By definition all degrees of freedom relevant for this time-sector of the response or correlation relax on timescales much smaller than ω^{-1} . As a consequence the correlation/response functions are expected to be given by their equilibrium expression in the presence of a quasi-constant external field $E \cos(\omega t)$. Therefore, in this regime:

$$\begin{aligned} C(t, t') &= C_{eq}(t - t', E \cos(\omega t)) \\ R(t, t') &= R_{eq}(t - t', E \cos(\omega t)). \end{aligned} \tag{16}$$

Since we are interested in the small E behavior, we can expand the above expression up to second order in E . For the response function, for instance, this yields:

$$R_{eq}(\tau, E \cos(\omega t)) \approx R_0(\tau) + \frac{E^2 \cos^2(\omega t)}{2} \left. \frac{\partial^2 R_{eq}(\tau, E)}{\partial E^2} \right|_{E=0}, \tag{17}$$

where $R_0(\tau)$ is the unperturbed equilibrium response function, and the derivative is computed with respect to a constant external field. Comparing the last equation with Eq. (10) in the stationary regime ($t, t' \rightarrow \infty$ with $\tau = t - t'$ finite) we find a very simple expression for the $n = 1$ component of the expansion $r_1^{(\omega)}(\tau)$ in the regime $\omega\tau \ll 1$:

$$r_1^{(\omega)}(\tau) = \frac{1}{8} \left. \frac{\partial^2 R_{eq}(\tau, E)}{\partial E^2} \right|_{E=0} \quad (\omega\tau \ll 1) \tag{18}$$

An analogous relation holds for the correlation function:

$$c_1^{(\omega)}(\tau) = \frac{1}{8} \left. \frac{\partial^2 C_{eq}(\tau, E)}{\partial E^2} \right|_{E=0} \quad (\omega\tau \ll 1) \tag{19}$$

These general results provide important insights to understand the behaviour of the non-linear susceptibility. First of all, since the correlation and response functions appearing in Eq. (16) are defined in equilibrium in presence of a constant field, they must obey FDT. Therefore one can establish a sort of generalized Fluctuation-Dissipation relation between the second order correlation and response functions, which reads:

$$r_1^{(\omega)}(\tau) = -\frac{1}{T} \frac{\partial c_1^{(\omega)}(\tau)}{\partial \tau}, \quad (\omega\tau \ll 1) \tag{20}$$

which is however only valid in the low frequency domain $\omega\tau \ll 1$.

III. CRITICAL BEHAVIOUR OF THE NON-LINEAR SUSCEPTIBILITY: SCALING ARGUMENTS

In this section we analyze the behaviour of the non-linear susceptibility using general physical and scaling arguments, later confirmed by our exact analysis of the Mode Coupling (p-spin) equations.

A. Time-temperature superposition principle

This results in the previous Section allow us to establish an interesting relation between the second order correlation and response functions and the *dynamical response* $\chi_T(\tau) \equiv T\partial C_{eq}(\tau)/\partial T$ that was recently introduced and extensively studied in [10, 11, 21, 22, 23, 24], in particular in relation with the behaviour of the four-point dynamical correlation function. The key idea is that in the glassy dynamics regime, the equilibrium correlation function $C_{eq}(\tau)$ satisfies to a good approximation the time-temperature superposition (TTS) principle. This means that the normalized correlation function for different temperatures, densities, external fields, etc., can be written as a function of $\tau/\tau_\alpha(T, \rho, E)$, and the whole T, ρ, E dependence is captured by the structural relaxation time $\tau_\alpha(T, \rho, E)$. This becomes actually an exact statement within the α -regime of MCT, when the system approaches the dynamical critical point.

Now, since $\tau_\alpha(T, \rho, E)$ is expected to be an even function of E because of the up-down symmetry, it should rather be written as $\tau_\alpha(T, \rho, \Theta)$, with $\Theta = E^2$. Then TTS immediately leads to:

$$\frac{\partial C_{eq}(\tau)}{\partial \Theta} = \frac{\partial \tau_\alpha / \partial \Theta}{\partial \tau_\alpha / \partial T} \frac{\partial C_{eq}(\tau)}{\partial T}. \quad (21)$$

Now, around $E = 0$ one has $\partial^2 C_{eq} / \partial E^2 = 2\partial C_{eq} / \partial \Theta$; using the above results one finds the very interesting relation (valid for $\omega\tau \ll 1$):

$$c_1^{(\omega)}(\tau) = \frac{\kappa}{4} \frac{\partial C_{eq}(\tau)}{\partial T} = \frac{\kappa}{4T} \chi_T(\tau),$$

where $\kappa = \frac{\partial \tau_\alpha}{\partial \Theta} / \frac{\partial \tau_\alpha}{\partial T}$. Analogously one finds for the response function (using the FDT relation (20)):

$$r_1^{(\omega)}(\tau) = -\frac{\kappa}{4T^2} \frac{\partial \chi_T(\tau)}{\partial \tau}. \quad (22)$$

Within MCT one can understand these results by noticing that the dynamical critical temperature is expected to show a quadratic dependence on the external field (for small fields) of the form:

$$T_{MCT}(E) \approx T_{MCT}(E = 0) + \kappa E^2. \quad (23)$$

Close to the critical point, a small field changes slightly the critical temperature. Since the only thing that matters for the critical behaviour is the distance from the critical point, one finds that applying a small field is equivalent to a small change in temperature (up to the constant κ). Note that this implies that the relations found above carry over, within MCT, to the β regime as well.

More generally, the amplitude of the correlation function also depends on temperature and electric field, and this dependence brings extra contributions that affect the above equalities. However, in glassy systems, it is the relaxation time τ_α which is most sensitive to external parameters, and these correction terms can be discarded. This is not the case in spin-glasses for example, where the correlation amplitude itself depends critically on temperature.

Provided the Fourier transform of Eq. (15) is dominated by the region $\omega\tau \ll 1$, one finds, using again FDT, that the non-linear susceptibility $\chi_3(\omega)$ is given by:

$$\chi_3(\omega) \approx \kappa \frac{\partial \chi_1(2\omega)}{\partial T}, \quad (24)$$

a relation expected to hold at low enough frequencies, at least when the deviations from TTS are weak.

B. Physical consequences within MCT

The results above establish a clear connection between the dynamical response χ_T and the non-linear susceptibility χ_3 . In the following, we will exploit the consequences of this connection within the MCT framework, using scaling arguments which will be fully confirmed by the detailed analysis of Section IV below.

The behaviour of $\chi_T(\tau)$ has been studied in great detail within MCT [11, 24], where two critical relaxation regimes occur close to the MCT transition: the β -regime, with relaxation time $\tau_\beta \sim \tau_0 \epsilon^{-1/2a}$, and the α -regime, with relaxation time $\tau_\alpha \sim \tau_\beta \epsilon^{-1/2b} \gg \tau_\beta$. [$\epsilon = T - T_c$ is the distance from the Mode Coupling critical temperature T_c , τ_0 is a microscopic time scale, and a, b are known MCT exponents.] The critical properties of $\chi_T(\tau)$ have been

derived in terms of scaling functions both in the α and β -regimes. Using these results, the relations established above and the FDT of Eq. (20), one can obtain the scaling behaviour of $c_1^{(\omega)}(\tau)$ and $r_1^{(\omega)}(\tau)$ close to the MCT transition. Let us analyse in turn the β -regime and the α -regime.

1. The β -regime

Let us first analyze the regime $\omega\tau \ll 1$ where the above quasi-stationary results are valid. When $\tau_0 \ll \tau \sim \tau_\beta \ll \tau_\alpha$, one finds [11, 24]:

$$\begin{aligned} c_1^{(\omega)}(\tau) &\approx \frac{1}{\sqrt{\epsilon}} c_\beta \left(\frac{\tau}{\tau_\beta} \right) \\ r_1^{(\omega)}(\tau) &\approx \frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} r_\beta \left(\frac{\tau}{\tau_\beta} \right), \end{aligned} \quad (25)$$

where the scaling function $c_\beta(x)$ behaves asymptotically as x^a for $x \ll 1$ and as x^b for $x \gg 1$, whereas $r_\beta(x)$ behaves as x^{a-1} for $x \ll 1$ and as x^{b-1} for $x \gg 1$.

For very large frequencies, such that $\omega\tau \gg 1$, one expects very small second order correlation and response functions. The reason is that, despite the fact that the system is close to the critical point, the field oscillates so fast that the system has no time to respond and so the change due to the field is very small. Close to the critical point, one expects a scaling behavior in the β -regime generalizing the one above:

$$c_1^{(\omega)}(\tau) \approx \frac{1}{\sqrt{\epsilon}} \hat{f}_\beta \left(\frac{\tau}{\tau_\beta}, \omega\tau \right) \quad (26)$$

$$r_1^{(\omega)}(\tau) \approx \frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} \hat{g}_\beta \left(\frac{\tau}{\tau_\beta}, \omega\tau \right). \quad (27)$$

The most general assumption compatible with previous results is a factorized form for \hat{f}_β and \hat{g}_β in both regimes $\tau \gg \tau_\beta$ and $\tau \ll \tau_\beta$. In the late β -regime, one has (with L for ‘late’):

$$c_1^{(\omega)}(\tau) \simeq \frac{1}{\sqrt{\epsilon}} \left(\frac{\tau}{\tau_\beta} \right)^b f_\beta^L(\omega\tau) \quad (28)$$

$$r_1^{(\omega)}(\tau) \simeq \frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} \left(\frac{\tau}{\tau_\beta} \right)^{b-1} g_\beta^L(\omega\tau). \quad (29)$$

where both functions f_β^L, g_β^L tend to a constant when their argument $\omega\tau$ is small, and tend to zero when $\omega\tau$ is large. In the early β -regime, a similar result holds, with a priori different

scaling functions f_β^E, g_β^E (E for ‘early’):

$$c_1^{(\omega)}(\tau) \simeq \frac{1}{\sqrt{\epsilon}} \left(\frac{\tau}{\tau_\beta} \right)^a f_\beta^E(\omega\tau) \quad (30)$$

$$r_1^{(\omega)}(\tau) \simeq \frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} \left(\frac{\tau}{\tau_\beta} \right)^{a-1} g_\beta^E(\omega\tau). \quad (31)$$

These new functions f_β^E, g_β^E again tend to a constant when their argument $\omega\tau$ is small, and tend to zero when $\omega\tau$ is large.

In this regime, the explicit dependence with τ and ω occurs only through the rescaled time and frequency τ/τ_β and $\omega\tau_\beta$. In the rest of the text, we will frequently use the variables $\hat{\tau} = \tau/\tau_\beta$, $\hat{\omega} = \omega\tau_\beta$ and $x = \omega\tau$.

2. The α -regime

The behaviour of $c_1^{(\omega)}, r_1^{(\omega)}$ in the α -regime follows similar scaling laws. When $\omega\tau \ll 1$, the results of [11, 24] allow one to obtain:

$$c_1^{(\omega)}(\tau) \approx \frac{1}{\epsilon} c_\alpha \left(\frac{\tau}{\tau_\alpha} \right) \quad (32)$$

$$r_1^{(\omega)}(\tau) \approx \frac{1}{\epsilon} \frac{1}{\tau_\alpha} r_\alpha \left(\frac{\tau}{\tau_\alpha} \right),$$

The matching between the late β -regime and the α -regime determine the asymptotic behaviour of the scaling functions defined above. One finds that $c_\alpha(x \ll 1)$ behaves as x^b and $r_\alpha(x \ll 1)$ as x^{b-1} , whereas both functions tend exponentially fast to zero for $x \gg 1$.

When $\omega\tau$ is not small, the scaling behaviour in the α -regime reads:

$$c_1^{(\omega)}(\tau) \approx \frac{1}{\epsilon} \bar{f}_\alpha \left(\frac{\tau}{\tau_\alpha}, \omega\tau \right) \quad (33)$$

$$r_1^{(\omega)}(\tau) \approx \frac{1}{\epsilon} \frac{1}{\tau_\alpha} \bar{g}_\alpha \left(\frac{\tau}{\tau_\alpha}, \omega\tau \right). \quad (34)$$

In the early α -regime, that is when $\tau \ll \tau_\alpha$, one finds:

$$c_1^{(\omega)}(\tau) \simeq \frac{1}{\epsilon} \left(\frac{\tau}{\tau_\alpha} \right)^b f_\alpha^E(\omega\tau) \quad (35)$$

$$r_1^{(\omega)}(\tau) \simeq \frac{1}{\epsilon} \frac{1}{\tau_\alpha} \left(\frac{\tau}{\tau_\alpha} \right)^{b-1} g_\alpha^E(\omega\tau). \quad (36)$$

Furthermore, by requiring the matching between the two regimes of large τ/τ_β and small τ/τ_α , one finds that the scaling functions $f_\alpha^E(x)$ and $g_\alpha^E(x)$ are the same as $f_\beta^E(x)$ and $g_\beta^E(x)$.

As in the β -regime, the explicit dependence with τ and ω occurs only through the rescaled time and frequency τ/τ_α and $\omega\tau_\alpha$. In the rest of the text, we will frequently use the variables $\bar{\tau} = \tau/\tau_\alpha$, $\bar{\omega} = \omega\tau_\alpha$ (and also $x = \omega\tau$).

C. Scaling behavior of $\chi_3(\omega)$

In the previous section we have determined the scaling forms governing the time and temperature dependence of $r_1^{(\omega)}(\tau)$ and $c_1^{(\omega)}(\tau)$. Using these results we can now easily analyze the critical behaviour of the non-linear susceptibility, by computing its Fourier transform at frequency 2ω , according to Eq. (15). We again focus in turn on the β -regime and then on the α -regime, before commenting on the zero and infinite frequency limits.

1. The β -regime

Let us first consider probing frequencies of the order of the inverse of the β -relaxation time. We set $\hat{\omega} = \omega\tau_\beta$ and assume that only the β -regime of $r_1^{(\omega)}(\tau)$ contributes in this regime. This assumption will be fully justified by the exact analysis of the schematic Mode Coupling equations of Sec. IV. Indeed one can show that $r_1^{(\omega)}(\tau \sim \tau_\alpha)$ is small due to the fact that the scaling function $g_\alpha(x)$, introduced in Eq. (35), vanishes as $1/x^{1+b}$ at large x . Therefore, for probing frequencies of the order of the inverse of the β -relaxation time, the contribution to the non-linear susceptibility coming from the time integral α -regime is negligible in Eq. (15).

One then finds that:

$$\chi_3(\omega) \simeq \frac{1}{\sqrt{\epsilon}} \mathcal{F}(\hat{\omega}) \quad (37)$$

where the function $\mathcal{F}(x)$ is defined as:

$$\mathcal{F}(\hat{\omega}) = \frac{4}{\hat{\omega}} \int_0^\infty du e^{-2iu} \hat{g}_\beta\left(\frac{u}{\hat{\omega}}, u\right)$$

The asymptotic behaviour of the scaling function \mathcal{F} can easily be obtained from the results of the previous section. One finds:

$$\mathcal{F}(\hat{\omega}) \simeq 4\hat{\omega}^{-b} \int_0^\infty du u^{b-1} e^{-2iu} g_\beta^L(u) \quad \omega\tau_\beta \ll 1 \quad (38)$$

$$\simeq 4\hat{\omega}^{-a} \int_0^\infty du u^{a-1} e^{-2iu} g_\beta^E(u) \quad \omega\tau_\beta \gg 1. \quad (39)$$

Using the asymptotic properties of $g_\beta^L(u), g_\beta^E(u)$ and the fact that a, b are between zero and one insures the convergence of the integrals appearing in the above equation, at both small and large u . This confirms that the scaling behaviour of $\chi_3(\omega)$ in this region is indeed dominated by the β -regime of $r_1^{(\omega)}(\tau)$. Note that in the high frequency region the ϵ dependence of $\chi_3(\omega)$ drops out, as it should in order to match the non critical τ_0^{-1} frequency regime.

2. The α -regime

We now consider the α -regime, where we set $\bar{\omega} = \omega\tau_\alpha$, and again assume that only the scaling form of $r_1^{(\omega)}(\tau)$ in this same regime, Eq. (33), contributes significantly to Eq. (15). Indeed, the contribution due to the time integral in the β -regime is at least a factor $\sqrt{\epsilon}$ smaller than the one coming from the α -regime, and yields a subleading contribution to the critical behavior of $\chi_3(\omega)$. We then find that the non-linear susceptibility scales as:

$$\chi_3(\omega) \simeq \frac{1}{\epsilon} \mathcal{G}(\bar{\omega}) \quad (40)$$

where the function $\mathcal{G}(x)$ is defined as:

$$\mathcal{G}(\bar{\omega}) = \frac{4}{\bar{\omega}} \int_0^\infty du e^{-2iu} \bar{g}_\alpha^E\left(\frac{u}{\bar{\omega}}, u\right)$$

Clearly, because of the matching of \bar{g}_α for small first arguments with \hat{g}_β at large first arguments, we find that the scaling of the early α -regime ($\omega\tau_\alpha \gg 1$) of the non-linear susceptibility matches with that of the late β -regime ($\omega\tau_\beta \ll 1$), with:

$$\chi_3(\omega) \propto \epsilon^{(b-a)/2a} \omega^{-b}. \quad (41)$$

3. Low frequency limit

In the low frequency limit, one finds that $\chi_3(\omega)$ decreases from its peak value ϵ^{-1} reached for $\omega \sim 1/2\tau_\alpha$ to a non critical, finite value given by Eq. (24). As discussed in [15], contrary to the case of spin-glasses, the non-linear susceptibility is critical only for small but non zero values of the frequencies. Zero frequency corresponds to a static equilibrium response (or correlation, via FDT). In glasses, these are not expected to have any critical behavior.

In particular, in the low frequency limit, one can expand Eq. (15) up to second order in ω . Using Eq. (17) we have:

$$\begin{aligned}\chi_3(\omega\tau \ll 1) &\approx \kappa \int_0^\infty d\tau (1 - 2i\omega\tau - 2\omega^2\tau^2) \frac{\partial R_{eq}(\tau)}{\partial T} \\ &\approx \kappa \frac{d}{dT} \left(\chi_1(0) - 2i\omega A_1 \frac{\tau_\alpha}{T} - 4\omega^2 A_2 \frac{\tau_\alpha^2}{T} \right),\end{aligned}\quad (42)$$

where the zero frequency limit of the linear susceptibility, $\chi_1(0)$, equals the static polarization fluctuations (along the z-axis) divided by temperature, $N\langle P^2 \rangle/T$. A_1 and A_2 are two temperature-independent constants defined as $A_1 = \int_0^\infty d(\tau/\tau_\alpha) C_{eq}(\tau/\tau_\alpha)$ and $A_2 = \int_0^\infty d(\tau/\tau_\alpha) (\tau/\tau_\alpha) C_{eq}(\tau/\tau_\alpha)$ (here we have used again the time-temperature superposition principle). The last equations allows us to determine the low frequency behavior of the real and imaginary part of the non-linear susceptibility:

$$\begin{aligned}\Re(\chi_3(\omega\tau \ll 1)) &\approx \kappa \frac{d\chi_1(0)}{dT} + B_1\omega^2 + O(\omega^4) \\ \Im(\chi_3(\omega\tau \ll 1)) &\approx B_2\omega + O(\omega^3),\end{aligned}$$

with $B_1 = -4\kappa A_2\omega^2 d(\tau_\alpha^2/T)/dT > 0$ and $B_2 = -2\kappa A_1\omega d(\tau_\alpha/T)/dT > 0$.

4. Large frequency limit

At very large frequencies (very small timescales) the non-linear susceptibility is vanishing because the system has not enough time to respond to the oscillating field. One could argue that the analysis of Eq. (4) at very large frequencies yields:

$$P_3(t) \sim \frac{\delta^3 P}{\delta E^3(0)} \Big|_{E=0} \left[\int_0^t dt_1 (e^{i\omega t_1} + e^{-i\omega t_1}) \right]^3. \quad (43)$$

As a result, at very large frequency the non-linear susceptibility behaves as:

$$\chi_3(\omega \rightarrow \infty) \sim \frac{1}{(i\omega)^3} \frac{\delta^3 P}{\delta E^3(0)} \Big|_{E=0}. \quad (44)$$

This analysis is oversimplified and assumes analytic properties of the function $\frac{\delta^3 P}{\delta E(t_1)\delta E(t_2)\delta E(t_3)}$ that are not granted and may depend strongly on the microscopic dynamics. For instance, in the case of Ising spins with a Monte Carlo heath bath dynamics one can easily verify that the previous arguments do not apply and the large frequency behavior is proportional to $1/(i\omega)$. The conclusion is that the high frequency behavior depends on the underlying

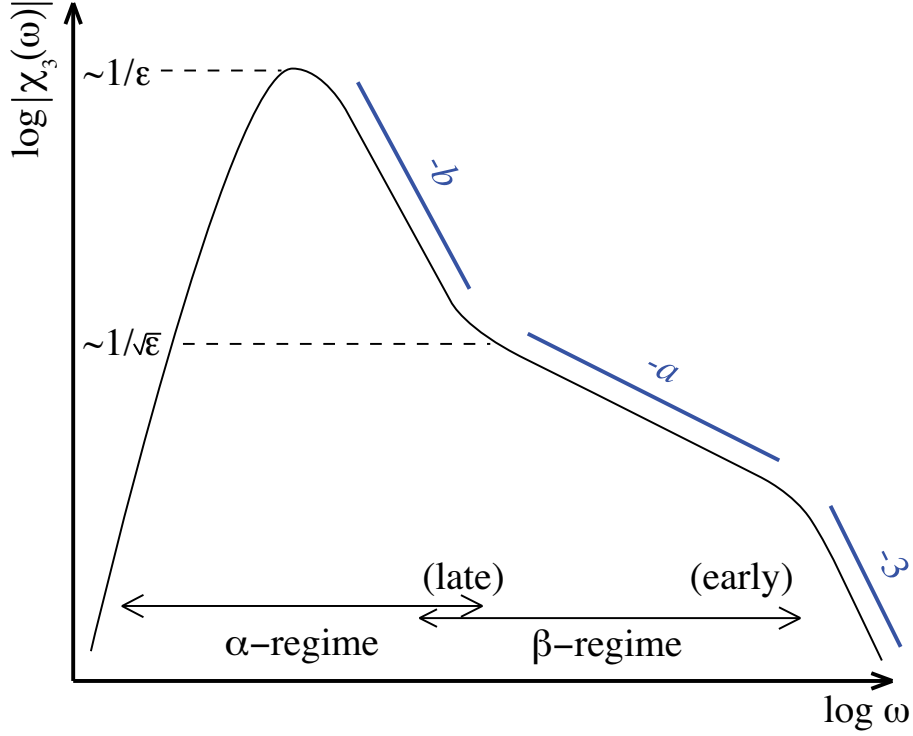


FIG. 1: Sketch of $\log |\chi_3(\omega)|$ as a function of $\log \omega$, showing five different frequency regimes: $\omega\tau_\alpha \ll 1$, $\omega\tau_\alpha \sim 1$, $\tau_\beta/\tau_\alpha \ll \omega\tau_\beta \ll 1$, $\omega\tau_\beta \gg 1$, $\omega\tau_0 \sim 1$. Note that the low frequency limit is non zero but much smaller than the peak value for T close to T_c .

microscopic dynamics and, likely, on the type of the non-linear response considered. In the case of non-linear dielectric susceptibility the underlying microscopic dynamics should be provided by Langevin equations for dipoles in a non-polar solvent (this is an approximation since at extremely high frequency inertia effects will play a role). To work out the high frequency behavior one can neglect interactions with other dipoles and the coupling to structural relaxation. Thus, the analysis of the non-linear response of a single dipole in a non-polar solvent worked out in [25] should apply. The outcome is the $1/(i\omega)^3$ behavior discussed above.

D. Summary of the main results

We summarize the main results of this paper on the frequency dependent non linear susceptibility $\chi_3(\omega)$ in Fig. 1, where the logarithm of the absolute value of $\chi_3(\omega)$ is sketched as a function of the logarithm of the frequency of the probing field. Five different frequency regimes are identified: $\chi_3(\omega)$ exhibits a peak around frequencies of the order of a half of the inverse of the structural relaxation time of the system τ_α , whose height grows as $(T - T_c)^{-1}$ as the critical temperature is approached. For higher frequencies, $\chi_3(\omega)$ decays as power laws, with an exponent equal to $-b$, then to $-a$, and finally to -3 . Note that the absolute value of both the real and the imaginary parts of χ_3 are expected to show a similar behavior.

One may wonder about the role of conserved variables like energy or density on the above results. As recalled in the introduction, we know that these conserved variables can dramatically change the scaling behaviour of χ_4 for example, which also diverges as $(T - T_c)^{-1}$ within a p-spin framework with Langevin dynamics, but diverges as $(T - T_c)^{-2}$ when the contribution of conserved variables is taken into account [10, 11]. From a diagrammatic point of view, this is due to the presence of ‘squared ladder’ diagrams which gives the dominant contribution to χ_4 . One can check that due to the causality of the response functions, these diagrams in fact are absent when one computes the non-linear susceptibility and the above results are expected to hold for a *bona fide* MCT theory of liquids. Beyond MCT, we expect that in the α -regime, $\chi_3(\omega)$ will take the following scaling form [15]:

$$\chi_3(\omega) \simeq \xi^{2-\bar{\eta}} \mathcal{G}(\omega\tau_\alpha), \quad (45)$$

where ξ is the dynamical correlation length which also appears in χ_T . One could in fact generalize the Inhomogeneous MCT calculation of [24] to account for a space and time dependent source term, that would describe the non-linear response to an oscillating field with wave-vector q and frequency ω . The corresponding behaviour of $\chi_3(\omega, q)$ can be guessed by adapting the above scaling arguments to the explicit results of [24] on the wavevector-dependent dynamical response. This leads to the following predictions for $\chi_3(\omega, q)$. In the β regime one finds:

$$\chi_3(\omega, q) = \xi^2 H_\beta(\omega\tau_\beta, q\xi) \quad \xi = \epsilon^{-1/4}, \quad \tau_\beta = \epsilon^{-\frac{1}{2a}}$$

where the scaling function $H_\beta(x, y)$ is equal to $\mathcal{F}(x)$ for $y = 0$, i.e. for a uniform electric field one finds back Eq. (37). For large y one expects a power law behavior such as $\frac{h_\beta(x)}{y^2}$ (where

$h_\beta(x)$ is a certain scaling function). As discussed in [24] this is needed to cancel out the diverging prefactor ξ^2 and match the critical behavior to the non-critical one taking place for $q \propto O(1)$. The asymptotic behavior with respect to x is identically to the one already described for homogeneous fields. Very small x correspond to the matching between α and β regimes. Since in the α regime χ_3 is expected to diverge as ξ^4 , the matching imposes the behavior at small x : $\frac{h_\beta^L(y)}{x^b}$ (where $h_\beta^L(y)$ is another scaling function). For large x values, the field varies so rapidly that the system has not enough time to adjust and to respond to the field. Again, in order to cancel the diverging prefactor and match the non-critical behavior one expects a large x behavior such as $\frac{h_\beta^E(y)}{x^a}$ (where $h_\beta^E(y)$ is a third scaling function). It would be interesting to specify in more details the shape of the scaling functions $h_\beta, h_\beta^L, h_\beta^E$.

In the α regime one expects:

$$\chi_3(\omega, q) = \xi^4 H_\alpha(\omega\tau_\alpha, q\xi) \quad \xi = \epsilon^{-1/4}, \quad \tau_\alpha = \epsilon^{-\frac{1}{2a} - \frac{1}{2b}}$$

where the scaling function $H_\alpha(x, y)$ is equal to $\mathcal{G}(x)$ for $y = 0$, i.e. for a uniform electric field one finds back eq. (40). The same kind of arguments used above suggests for large y a power law behavior: $H_\alpha(x, y) \simeq \frac{h_\alpha(x)}{y^4}$ (where $h_\alpha(x)$ is a scaling function). For very small x the scaling function vanishes in order to match the $x = 0$ value corresponds to the non-critical (non diverging) static non-linear susceptibility. For large x values in order to match the β regime one expects a behaviour such as $\frac{h_\alpha^E(y)}{x^b}$ (where $h_\alpha^E(x)$ is another scaling function).

IV. THE p -SPIN IN FULL GLORY

In this section we study a specific microscopic model which allows one to check the assumptions discussed in the previous section and to derive analytically, in a simplified setting, the critical behaviour of the non-linear susceptibility. The model we consider is the spherical p -spin model, a mean field model with quenched disorder and p -body interactions, which belongs to the universality class of the discontinuous spin glasses. This class of analytically solvable models is able to reproduce many features of glass-forming liquids, and provides a mean-field paradigm of the glass transition [1]. In particular, the spherical p -spin glass shows a dynamical transition as the temperature is lowered below T_c , described by the schematic Mode Coupling Theory for supercooled liquids. At T_c the structural relaxation time of the system diverges as a power law, due to the emergence of an exponentially large

number of metastable states (which, in mean field, are separated by infinite barriers). As in MCT, the dynamical transition is accompanied by the divergence of dynamical correlations [5]. Since the model is completely connected one can probe these correlations only through the dynamical susceptibility, $\chi_4(t)$, which indeed diverges in a critical way at the transition [5, 28] (for the full MCT one can also obtain real space dynamic correlations, see [24]). In the following we will compute the non-linear susceptibility for the p -spin model and show that it allows one to probe the same divergent dynamical correlations captured by χ_4 . A previous work [29] has already considered the effect of an oscillating field on the dynamics of this model but it focused on non-equilibrium dynamics and not on dynamical correlations.

A. The model and the Langevin dynamics

The Hamiltonian of the model is:

$$\mathcal{H} = - \sum_{i_1 < \dots < i_p} J_{i_1 \dots i_p} S_{i_1} \cdots S_{i_p}, \quad (46)$$

where $J_{i_1 \dots i_p}$ are Gaussian quenched random couplings with zero mean and variance $\overline{J_{i_1 \dots i_p}^2} = p! J_p^2 / 2N^{p-1}$. We consider the spherical version of the model, where the N spins are continuous real variables with a global constraint: $\sum_i S_i^2 = N$.

We assume that the dynamics of the model is described by the Langevin equation:

$$\frac{dS_i}{dt} = - \frac{\partial \mathcal{H}}{\partial S_i} + \eta_i(t), \quad (47)$$

where $\eta_i(t)$ is a Gaussian white noise, with zero mean, $\langle \eta_i(t) \rangle = 0$ and variance $\langle \eta_i(t) \eta_j(t') \rangle = 2T \delta_{ij} \delta(t - t')$. We shall use the Martin-Siggia-Rose formalism to derive the dynamical effective action [32]. The probability of a given dynamical trajectory of the spins is thus equal to:

$$P(\{S_i\}) = \int \mathcal{D}\eta \mathcal{W}(\eta) \prod_i \delta \left(\frac{dS_i}{dt} + \frac{\partial \mathcal{H}}{\partial S_i} - \eta_i(t) \right),$$

where $\mathcal{W}(\eta) \propto \exp\{-1/2 \sum_i \int dt dt' \eta_i(t) 2T \delta(t - t') \eta_i(t')\}$. Using the integral representation of the delta functions, introducing the new variables $\hat{S}_i(t)$, and integrating over the noise, the partition function of the dynamical process reads [33]:

$$\begin{aligned} \mathcal{Z} &= \int \mathcal{D}S P(\{S_i\}) = \int \mathcal{D}S \mathcal{D}\hat{S} \\ &\times \exp \left\{ - \sum_i \int dt i \hat{S}_i(t) \left[\dot{S}_i(t) - T i \hat{S}_i(t) + \frac{\partial \mathcal{H}}{\partial S_i} \right] \right\}. \end{aligned} \quad (48)$$

All the dynamical observables can be derived from the generating functional \mathcal{Z} by introducing two (time-dependent) fields $h_i(t)$ and $\hat{h}_i(t)$ conjugated to S_i and \hat{S}_i and taking the appropriate derivatives of \mathcal{Z} with respect to these fields.

In order to study the critical behaviour of the non-linear susceptibility, $\chi_3(\omega)$, we introduce an external oscillating uniform “magnetic” field, coupled to all the spins of the system, of the form $h(t) = h \cos(\omega t)$. The partition function, Eq. (48), can be written in terms of an effective action as $\mathcal{Z} = \int \mathcal{D}S \mathcal{D}\hat{S} \exp\{\mathcal{L}(S, \hat{S})\}$, where

$$\mathcal{L}(S, \hat{S}) = - \sum_i \int dt \left\{ i\hat{S}_i(t) \left[\dot{S}_i(t) - T i\hat{S}_i(t) + Z(t)S_i(t) - h(t) \right] + \mathcal{L}_J^i(S, \hat{S}) \right\}. \quad (49)$$

$\sum_i \mathcal{L}_J^i(S, \hat{S})$ is the part of the effective action which depends on the quenched disorder (which we evaluate below), $Z(t)$ is a Lagrange multiplier which enforces the spherical constraint, and $h(t)$ is the external magnetic field.

By using the fact that $\sum_{i_1 < \dots < i_p} = \frac{1}{p!} \sum_{i_1, \dots, i_p}$, the derivative of the Hamiltonian, Eq. (46), with respect to the spin S_i reads:

$$\mathcal{L}_J^i(s, \hat{S}) = \frac{1}{(p-1)!} \sum_{i_2, \dots, i_p} J_{i i_2 \dots i_p} i\hat{S}_i(t) S_{i_2}(t) \cdots S_{i_p}(t).$$

We can now perform the integral over the quenched disorder which yields:

$$\begin{aligned} \overline{\exp\left\{\sum_i \mathcal{L}_J(S, \hat{S})\right\}} &\propto \exp\left\{\frac{NpJ_p^2}{4} \int dt_1 dt_2 \right. \\ &\times \left(\mathbf{S}(t_1) \cdot \mathbf{S}(t_2)\right)^{p-2} \left[\left(i\hat{\mathbf{S}}(t_1) \cdot i\hat{\mathbf{S}}(t_2)\right) \left(\mathbf{S}(t_1) \cdot \mathbf{S}(t_2)\right) \right. \\ &\left. \left. + (p-1) \left(i\hat{\mathbf{S}}(t_1) \cdot \mathbf{S}(t_2)\right) \left(\mathbf{S}(t_1) \cdot i\hat{\mathbf{S}}(t_2)\right) \right] \right\}, \end{aligned}$$

where we have introduced the notation:

$$\mathbf{S}(t_1) \cdot \mathbf{S}(t_2) = \frac{1}{N} \sum_{i=1}^N S_i(t_1) S_i(t_2).$$

B. Equations of motion of the magnetization, the correlation function, the response function, and the function $Z(t)$

In the thermodynamic limit ($N \rightarrow \infty$) we can take the saddle point value of the effective action and determine the equations of motion of all the dynamical observables. For instance,

from the equality $\langle \partial \mathcal{L} / \partial i \hat{S}_i \rangle = 0$, we obtain the equation of motion of the magnetization, $m(t) = (1/N) \sum_i \langle S_i(t) \rangle$:

$$\begin{aligned} \frac{\partial \mathcal{L}}{\partial i \hat{S}_i(t)} &= -\dot{S}_i(t) - Z(t)S_i(t) + h(t) + 2Ti\hat{S}_i(t) \\ &+ \frac{pJ_p^2}{4} \int dt_1 \left[2i\hat{S}_i(t_1) (\mathbf{S}(t) \cdot \mathbf{S}(t_1))^{p-1} + 2(p-1)S_i(t_1) (\mathbf{S}(t) \cdot i\hat{\mathbf{S}}(t_1)) (\mathbf{S}(t) \cdot \mathbf{S}(t_1))^{p-2} \right], \end{aligned} \quad (50)$$

Using the fact that $\langle \hat{S}_i \rangle = 0$, and the definition of the correlation and response functions:

$$\begin{aligned} C(t, t') &= \frac{1}{N} \sum_{i=1}^N \langle S_i(t) S_i(t') \rangle \\ R(t, t') &= \frac{1}{N} \sum_{i=1}^N \left. \frac{\partial \langle S_i(t) \rangle}{\partial h(t')} \right|_{h_i=0} = \langle S_i(t) i\hat{S}_i(t') \rangle, \end{aligned} \quad (51)$$

we get:

$$\dot{m} = -Z(t)m(t) + h(t) \frac{p(p-1)J_p^2}{2} \int_{-\infty}^t dt_1 m(t_1) R(t, t_1) [C(t, t_1)]^{p-2}. \quad (52)$$

Here, we have replaced the fluctuating two points functions by their average (correlation and response). This is exact in the large N limit. We have also used the fact that $R(t_1, t_2) = 0$ for $t_2 > t_1$ due to causality. Finally, notice that we have applied the field at a very distant time in the past, equal to minus infinity for all practical purposes. In this way we are sure that the system has reached a steady state on times $t \propto O(1)$ and above T_c .

By multiplying Eq. (50) by $S_i(t')$, using that $\langle S_i(t') \frac{\partial \mathcal{L}}{\partial i \hat{S}_i(t')} \rangle = 0$ and averaging over the sites, we get the equation of motion for the correlation function:

$$\begin{aligned} \frac{\partial C(t, t')}{\partial t} &= -Z(t)C(t, t') + h(t)m(t') + 2TR(t', t) \\ &+ \sum_p \frac{pJ_p^2}{2} \left\{ \int_{-\infty}^{t'} dt_1 R(t', t_1) [C(t, t_1)]^{p-1} + (p-1) \int_{-\infty}^t dt_1 R(t, t_1) C(t_1, t') [C(t, t_1)]^{p-2} \right\}. \end{aligned} \quad (53)$$

Similarly, multiplying Eq. (50) by $\hat{S}_i(t')$, using that $\langle i\hat{S}_i(t') \frac{\partial \mathcal{L}}{\partial i \hat{S}_i(t')} \rangle = \delta(t' - t)$ and averaging over the sites, one gets the equation for the evolution of the response function:

$$\begin{aligned} \frac{\partial R(t, t')}{\partial t} &= -Z(t)R(t, t') + \delta(t - t') \\ &+ \frac{p(p-1)J_p^2}{2} \int_{t'}^t dt_1 R(t_1, t') R(t, t_1) [C(t, t_1)]^{p-2} \end{aligned} \quad (54)$$

Notice that we used the fact that $\langle i\hat{S}_i(t) i\hat{S}_i(t') \rangle = 0$. We can now also obtain the equation of motion of the Lagrange multiplier, $Z(t)$, by enforcing the constraint $C(t, t) = 1$, i.e.,

$[\partial_t C(t, t') + \partial_{t'} C(t, t')]_{t=t'} = 0$, which yields:

$$Z(t) = h(t)m(t) + T + \frac{p^2 J_p^2}{2} \int_{-\infty}^t dt_1 R(t, t_1) [C(t, t_1)]^{p-1}. \quad (55)$$

When $h(t) = 0$, one sees from this equation that $Z(t)$ tends to a time independent constant Z_0 in the stationary regime, $T > T_c$, $t \rightarrow \infty$.

C. Small field expansion

In this section we discuss the small field expansion of the magnetization, the correlation and the response function, and derive the equation of motion of the second-order correlation and response functions introduced in the previous section.

Because of the up-down symmetry in the direction of the field, the effective action $\mathcal{L}(S, \hat{S})$, Eq. (49), is invariant under the transformation $h \rightarrow -h$ and $S \rightarrow -S$, $\hat{S} \rightarrow -\hat{S}$. This yields $m(t, h) = -m(t, -h)$, i.e., the magnetization is an odd function of the magnetic field, and thus only the odd derivatives of m with respect to h are non-zero. Conversely, the correlation and response functions are even functions of the external field (as well as the function $Z(t)$), and their expansion in powers of h contains only even terms. Therefore, up to the third order in h , one can write:

$$\begin{aligned} m(t) &= m_1(t)h + m_3(t)h^3 + O(h^5) \\ C(t, t') &= C_0(t, t') + C_2(t, t')h^2 + O(h^4) \\ R(t, t') &= R_0(t, t') + R_2(t, t')h^2 + O(h^4) \\ Z(t) &= Z_0 + Z_2(t)h^2 + O(h^4) \end{aligned} \quad (56)$$

where C_0 and R_0 are the correlation and response functions in absence of the oscillating magnetic field. In the stationary regime ($t, t', \tau = t - t' \propto O(1)$), TTI and FDT hold for the unperturbed correlation and response function, which satisfy the following equation for $\tau > 0$:

$$\frac{dC_0(\tau)}{d\tau} + TC_0(\tau) + \frac{p}{2T} \int_0^\tau du [C_0(\tau - u)]^{p-1} \frac{dC_0(u)}{du} = 0. \quad (57)$$

This is basically the schematic Mode Coupling equation for the density-density correlation functions in supercooled liquids, which leads to the well known shape of $C_0(\tau)$ characterized by two power-law relaxation regimes (β -relaxation and α -relaxation) and an intermediate plateau.

Plugging the previous equation, together with Eqs. (56), into Eqs. (52), (53), (54) and (55), we easily obtain a system of coupled integro-differential equations for $m_1(t)$, $m_3(t)$, $C_2(t, t')$, $R_2(t, t')$ and $Z_2(t)$. In particular, up to $O(h)$, from Eq. (52) we get:

$$\frac{d}{dt} m_1(t) = -Z_0 m_1(t) + \cos(\omega t) + \frac{p(p-1)J_p^2}{2} \int_0^t dt_1 m_1(t_1) R_0(t, t_1) [C_0(t, t_1)]^{p-2}. \quad (58)$$

As expected, the Green's function of the equation above satisfies exactly the same equation as $R_0(t, t')$. As a result we have that [see Eq. (12)]:

$$m_1(t) = \int_0^t dt' R_0(t, t') \cos(\omega t') \quad (59)$$

Furthermore, from Eqs. (51b), (56c) and (59) we have that [see Eq. (13)]:

$$m_3(t) = \int_0^t dt' R_2(t, t') \cos(\omega t'). \quad (60)$$

As a matter of fact, the equation of motion of the magnetization is superfluous. Indeed, by taking the derivative of Eq. (52) with respect to $h(t')$ we get exactly the equation for the dynamical evolution of the response function. Therefore, in the following we will only focus on the three equations (53)-(55).

Let us define the vector:

$$\mathbf{V}(t, t_1) = [C_2(t, t_1), R_2(t, t_1), Z_2(t)].$$

Eqs. (53)-(55) can be rewritten as:

$$\mathcal{O} \cdot \mathbf{V} = \mathbf{S}, \quad (61)$$

where \mathcal{O} is an integro-differential operator and \mathbf{S} is a source term which reads:

$$\begin{aligned} \mathbf{S}(t, t') &= \{ \cos(\omega t) m_1(t'), 0, \cos(\omega t) m_1(t) \} \\ &= \left\{ \frac{1}{4} \left[R_0(\omega) \left(e^{i\omega(t+t')} + 1 \right) + \text{c.c.} \right], 0, \frac{1}{4} \left(R_0(\omega) \left(e^{2i\omega t} + 1 \right) + \text{c.c.} \right) \right\}. \end{aligned} \quad (62)$$

We now write down the explicit expressions of the nine elements of the linear operator \mathcal{O} , omitting (t, t') in the l.h.s.. From the equation of the dynamical evolution of the correlation function, Eq. (53), we get:

$$\begin{aligned} \mathcal{O}_{11} \cdot C_2 &= \left(\frac{\partial}{\partial t} + Z_0 \right) C_2(t, t') - \frac{p(p-1)J_p^2}{2} \left[\int dt_1 \left(R_0(t', t_1) [C_0(t, t_1)]^{p-2} \right. \right. \\ &\quad \left. \left. + (p-2) R_0(t, t_1) C_0(t_1, t') [C_0(t, t_1)]^{p-3} \right) C_2(t, t_1) - \int dt_1 R_0(t, t_1) [C_0(t, t_1)]^{p-2} C_2(t_1, t') \right]. \end{aligned} \quad (63)$$

Analogously, we can determine the other elements:

$$\begin{aligned} \mathcal{O}_{12} \cdot R_2 &= -2T R_2(t', t) \\ &- \frac{pJ_p^2}{2} \left[(p-1) \int dt_1 C_0(t_1, t') [C_0(t, t_1)]^{p-2} R_2(t, t_1) + \int dt_1 [C_0(t, t_1)]^{p-1} R_2(t', t_1) \right] \end{aligned} \quad (64)$$

and

$$\mathcal{O}_{13} \cdot Z_2 = C_0(t, t') Z_2(t). \quad (65)$$

In the same way, we can analyze the equation for the response function, Eq. (54), which yields:

$$\mathcal{O}_{21} \cdot C_2 = -\frac{p(p-1)(p-2)J_p^2}{2} \int dt_1 R_0(t_1, t') R_0(t, t_1) [C_0(t, t_1)]^{p-3} C_2(t, t_1), \quad (66)$$

$$\mathcal{O}_{22} \cdot R_2 = \left(\frac{\partial}{\partial t} + Z_0 \right) R_2(t, t') \quad (67)$$

$$\begin{aligned} &- \frac{p(p-1)J_p^2}{2} \left\{ \int dt_1 R_0(t_1, t') [C_0(t, t_1)]^{p-2} R_2(t, t_1) \right. \\ &\quad \left. + \int dt_1 R_0(t, t_1) [C_0(t, t_1)]^{p-2} R_2(t_1, t') \right\}, \\ \mathcal{O}_{23} \cdot Z_2 &= R_0(t, t') Z_2(t). \end{aligned} \quad (68)$$

Finally, considering the equation for $Z(t)$, Eq. (55), one gets:

$$\mathcal{O}_{31} \cdot C_2 = -\frac{p^2(p-1)J_p^2}{2} \int dt_1 R_0(t, t_1) [C_0(t, t_1)]^{p-2} C_2(t, t_1), \quad (69)$$

$$\mathcal{O}_{32} \cdot R_2 = -\frac{p^2 J_p^2}{2} \int dt_1 [C_0(t, t_1)]^{p-1} R_2(t, t_1), \quad (70)$$

$$\mathcal{O}_{33} \cdot Z_2 = Z_2(t). \quad (71)$$

The above expressions can be greatly simplified by noting that in presence of an oscillating field of frequency ω , $R_2(t, t')$ and $C_2(t, t')$ are invariant under the shift of both times t and t' by any multiple of π/ω . Similarly, the function $Z_2(t)$ turns out to be a periodic function of t of frequency 2ω . Therefore, as discussed in the previous section, the correlation function, the response function and the function $Z_2(t)$ can be expanded in Fourier series [see Eq. (10)]. Notice that the source term have exactly the same periodicity as R_2 , C_2 and Z_2 , but contains only the 0-th and the first harmonics (i.e., $n = 0, 1, -1$). This implies that:

$$\begin{aligned} R_2(t, t') &= r_0^{(\omega)}(\tau) + e^{i\omega(t+t')} r_1^{(\omega)}(\tau) + e^{-i\omega(t+t')} r_{-1}^{(\omega)}(\tau) \\ C_2(t, t') &= c_0^{(\omega)}(\tau) + e^{i\omega(t+t')} c_1^{(\omega)}(\tau) + e^{-i\omega(t+t')} c_{-1}^{(\omega)}(\tau) \\ Z_2(t) &= z_0^{(\omega)} + e^{2i\omega t} z_1^{(\omega)} + e^{-2i\omega t} z_{-1}^{(\omega)}, \end{aligned} \quad (72)$$

where $\tau \equiv t - t'$. Here we recall that, plugging Eq. (72) into Eq. (60) we get that the non-linear susceptibility is given by the Fourier transform of $r_1^{(\omega)}(\tau)$ at frequency 2ω [Eq. (15)].

Using these properties, the integrals of Eqs. (63-71) can be simplified. Notice that, as expected (due to the fact that the correlation and the response function have to be real), $r_{-1}^{(\omega)}$ turns out to be equal to $r_1^{(\omega)*}$, and similar identities hold for $c_{-1}^{(\omega)}$ and $z_{-1}^{(\omega)}$. Therefore, we are left with three coupled integro-differential equations for $r_1^{(\omega)}$, $c_1^{(\omega)}$ and $z_1^{(\omega)}$. Up to order \hbar^2 , Eq. (55) reads:

$$z_1^{(\omega)} - \frac{p^2 J_p^2}{2} \int_0^\infty d\sigma e^{-i\omega\sigma} [C_0(\sigma)]^{p-2} [C_0(\sigma) r_1^{(\omega)}(\sigma) + (p-1)R_0(\sigma) c_1^{(\omega)}(\sigma)] = R_0(\omega) \quad (73)$$

Analogously, for the first equation, Eq. (53), we have:

$$\begin{aligned} R_0(\omega) = & \left(Z_0 + i\omega + \frac{d}{d\tau} \right) c_1^{(\omega)}(\tau) + e^{i\omega\tau} C_0(\tau) z_1^{(\omega)} \quad (74) \\ & - \frac{pJ_p^2}{2} \left\{ \int_{-\infty}^{t'} d\sigma \left[e^{-i\omega(\sigma+\tau)} C_0(\sigma+\tau) r_1^{(\omega)}(\sigma) + (p-1) e^{-i\omega\sigma} R_0(\sigma) c_1^{(\omega)}(\sigma+\tau) \right] [C_0(\sigma+\tau)]^{p-2} \right. \\ & + (p-1) \int_{-\infty}^t d\sigma \left[e^{i\omega(\tau-\sigma)} C_0(\tau-\sigma) (C_0(\sigma) r_1^{(\omega)}(\sigma) + (p-2) R_0(\sigma) c_1^{(\omega)}(\sigma)) \right. \\ & \left. \left. + e^{-i\omega\sigma} R_0(\sigma) C_0(\sigma) c_1^{(\omega)}(\tau-\sigma) \right] [C_0(\sigma)]^{p-3} \right\}. \end{aligned}$$

Finally, Eq. (54) becomes the central equation that we will need to solve to get $r_1^{(\omega)}(\tau)$:

$$\begin{aligned} 0 = & \left(Z_0 + i\omega + \frac{d}{d\tau} \right) r_1^{(\omega)}(\tau) + e^{i\omega\tau} R_0(\tau) z_1^{(\omega)} \\ & - \frac{p(p-1)J_p^2}{2} \int_0^\tau d\sigma [C_0(\sigma)]^{p-3} \left\{ e^{-i\omega\sigma} R_0(\sigma) C_0(\sigma) r_1^{(\omega)}(\tau-\sigma) \right. \\ & \left. + e^{i\omega(\tau-\sigma)} R_0(\tau-\sigma) [C_0(\sigma) r_1^{(\omega)}(\sigma) + (p-2) R_0(\sigma) c_1^{(\omega)}(\sigma)] \right\} \quad (75) \end{aligned}$$

D. $\omega \rightarrow 0$ limit

Let us first consider the $\omega \rightarrow 0$ limit (i.e. ω much smaller than the inverse of all characteristic times), where the system is subjected to an adiabatically varying external magnetic field, h . In this case, at stationarity, both TTI and FDT hold for the full correlation and response functions. As a result, one has that $C(t, t') = C(t - t')$ and $R(t, t') = R(t - t')$; and $-(1/T) \partial[C(\tau)]/\partial\tau = R(\tau)$. Since FDT holds for the unperturbed response and correlation

functions, $C_0(\tau)$ and $R_0(\tau)$, we have that:

$$-\frac{1}{T} \frac{\partial [c_1^{(\omega=0)}(\tau)]}{\partial \tau} = r_1^{(\omega=0)}(\tau) \quad (76)$$

at all times. Using this property, Eqs. (73)-(75) reduce to a single equation for $r_1^{(\omega=0)}(\tau)$. (Note that $z_1^{(\omega=0)} = \partial Z / \partial h^2 = 1/T$). It is straightforward to show that this equation is exactly given by the derivative with respect to h_0^2 of the equation for the full correlation function in presence of a constant external field [30]. Indeed, by definition we have that $c_1^{(\omega=0)}(\tau) = \partial C(\tau) / \partial h^2$, and $r_1^{(\omega=0)}(\tau) = \partial R(\tau) / \partial h^2$.

Since the dynamical transition temperature of the spherical p -spin model in the T - h plane is (at small fields) of the form $T_c = T_c^{(h=0)} + \kappa h^2$ [30], taking the derivative with respect to h^2 is equivalent (up to a constant) to taking the derivative with respect to temperature [see Eq. (23)]:

$$c_1^{(\omega=0)}(\tau) \propto \frac{\partial C(\tau)}{\partial T} = \frac{1}{T} \chi_T(\tau). \quad (77)$$

As a result $c_1^{(\omega=0)}(\tau)$ behaves exactly as the non-linear susceptibility $\chi_T(\tau)$, whose critical behavior is well known [10, 11, 21, 22, 24], and whose critical properties close to the dynamical transition have been reviewed in the previous section and are summarized in FIG. 3. Thanks to FDT, the critical behaviour of $r_1^{(\omega=0)}(\tau)$ can be obtained by taking the time derivative of $\chi_T(\tau)$ with respect to temperature.

E. β -regime

We now focus on the behaviour of the second order correlation and response function in the case where the frequency of the external oscillating field is finite. We start by analyzing the integrals in the β -regime close to the dynamical transition ($\tau \sim \tau_\beta$; $\tau_\beta \sim \epsilon^{-1/2a}$). In this regime the equilibrium response and correlation functions satisfy the following scaling laws, up to subleading terms:

$$\begin{aligned} C_0(\tau) &\simeq q + \sqrt{\epsilon} c_{0\beta} \left(\frac{\tau}{\tau_\beta} \right), \\ R_0(\tau) &\simeq \frac{\sqrt{\epsilon}}{\tau_\beta} r_{0\beta} \left(\frac{\tau}{\tau_\beta} \right). \end{aligned} \quad (78)$$

We first consider the case of small enough frequencies, such that $\omega\tau \ll 1$ (e.g., for frequencies of the order of the inverse α -relaxation time). In this regime, as discussed in the previous

section, one can establish relations between $c_1^{(\omega)}$, $r_1^{(\omega)}$ and χ_T , that lead to:

$$\begin{aligned} c_1^{(\omega)}(\tau) &= \frac{1}{\sqrt{\epsilon}} c_\beta \left(\frac{\tau}{\tau_\beta} \right), \\ r_1^{(\omega)}(\tau) &= \frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} r_\beta \left(\frac{\tau}{\tau_\beta} \right), \end{aligned} \quad (79)$$

with scaling functions $c_\beta(x)$, $r_\beta(x)$ that we have already discussed above.

For larger values of the frequency, close to the critical point one expects that the most general scaling behavior for the response and correlation functions is given by Eq. (26), in terms of scaling functions of two variables τ/τ_β and $\omega\tau$. However, as discussed in Sec. III B 1, in the early and late β -regime the τ/τ_β and the $\omega\tau$ dependences are expected to factorize, and one has [see Eqs. (28-31)]:

$$c_1^{(\omega)}(\tau) \simeq \frac{1}{\sqrt{\epsilon}} c_\beta(\hat{\tau}) f_\beta^{E(L)}(\hat{\omega}\hat{\tau}) \quad (80)$$

$$r_1^{(\omega)}(\tau) \simeq \frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} r_\beta(\hat{\tau}) g^{E(L)}(\hat{\omega}\hat{\tau}). \quad (81)$$

where $f_\beta^{E(L)}$ is either f_β^E or f_β^L , depending whether we study the early or late β -regime. The function $f_\beta^{E(L)}(x)$ is expected to be of order 1 for $x \ll 1$, and to drop to zero for $x \gg 1$.

The cut-off function $g^{E(L)}(\hat{\omega}\hat{\tau})$ has asymptotic behavior similar to $f_\beta^{E(L)}(\hat{\omega}\hat{\tau})$. In particular, since for $\hat{\omega}\hat{\tau} \ll 1$ FDT must hold, we have that $g^{E(L)}(x) \approx f_\beta^{E(L)}(x)$ when $x \ll 1$.

In the following we shall check that indeed these scaling forms are correct. In order to do that we need to analyze in detail Eqs. (74) and (75). Since this is rather technical and requires cutting integral in different pieces in order to analyze their scaling behaviour, we leave the details for Appendix 1 and skip directly to the final results. Let us start with the analysis of eq. (75). The contribution of all the dominant terms (computed in Appendix 1 and proportional to $r_1^{(\omega)}(\tau)$) can be simplified into:

$$\left[Z_0 - p \frac{1 - q^{p-1}}{2T} - p(p-1) q^{p-2} \frac{1 - q}{2T} \right] r_1^{(\omega)}(\tau). \quad (82)$$

Using the fact that $Z_0 = T + p/2T$, and the equation for the plateau at the dynamical transition $q = (p-2)/(p-1)$ [30], we find that the quantity in the square bracket is exactly zero at T_c . (We recall that $T_c = [p(p-2)^{p-2}/2(p-1)^{p-1}]^{1/2}$). More precisely, at temperature $T = T_c + \epsilon$, the first subleading terms are of the order $1/\tau_\beta$, see Eqs. (95) and (97). Since this term is of order $\sqrt{\epsilon}/\tau_\beta$ can be dropped. Putting all the terms of order $1/\tau_\beta$ together we

get the following equation for the scaling functions in the β -regime:

$$\begin{aligned}
& p(p-1)q^{p-2} \left\{ \int_{\hat{\tau}/2}^{\hat{\tau}} ds \cos(\hat{\omega}s) r_{0\beta}(s) \hat{g}_\beta(\hat{\tau}-s, x-\hat{\omega}s) \right. \\
& \quad \left. + \int_0^{\hat{\tau}/2} ds \cos(\hat{\omega}s) r_{0\beta}(s) [\hat{g}_\beta(\hat{\tau}-s, x-\hat{\omega}s) - \hat{g}_\beta(\hat{\tau}, x)] \right\} \\
& + \frac{p(p-1)(p-2)}{2} q^{p-3} \frac{1-q}{T} \left[r_{0\beta}(\hat{\tau}) \hat{f}_\beta(\hat{\tau}, x) + c_{0\beta}(\hat{\tau}) \hat{g}_\beta(\hat{\tau}, x) \right] = 0. \tag{83}
\end{aligned}$$

In a similar way, from the equation for the second order correlation function, Eq. (74) we can derive another equation for the scaling function \hat{f}_β in the β -regime. Using the fact that $c_1^{(\omega)}(\tau) = c_1^{(\omega)}(-\tau)$, it is more convenient (when deriving the equations) to write down the scaling equation for $[c_1^{(\omega)}(\tau) + c_1^{(\omega)}(-\tau)]/2$.

We find that the leading order is $O(1)$. Neglecting all the subleading contributions, this equation reads:

$$\begin{aligned}
r_0(\omega) = & -\frac{p(p-1)(p-2)}{2} q^{p-3} \frac{1-q}{T} \hat{f}_\beta(\hat{\tau}, x) c_{0\beta}(\hat{\tau}) \\
& -p(p-1)q^{p-2} \left\{ \int_0^{\hat{\tau}/2} ds \cos(\hat{\omega}s) r_{0\beta}(s) \left[\hat{f}_\beta(\hat{\tau}-s, x-\hat{\omega}s) + \hat{f}_\beta(\hat{\tau}+s, x+\hat{\omega}s) \right] \right. \\
& \left. + \int_{\hat{\tau}/2}^{\infty} ds \cos(\hat{\omega}s) r_{0\beta}(s) \left[\hat{f}_\beta(\hat{\tau}-s, x-\hat{\omega}s) + \hat{f}_\beta(\hat{\tau}+s, x+\hat{\omega}s) \right] \right\}. \tag{84}
\end{aligned}$$

We now analyse Eq. (83) in the different regimes. In order to get simplified scaling equations, we use the hypothesis we have made above and check the consistency of the assumptions. First let us consider the $x = \hat{\omega}\hat{\tau} \rightarrow 0$ behaviour for $\hat{\tau}$ both in the late and early β regime. In all these cases one can show that plugging our scaling ansatz into the equations one finds $f_\beta^{E(L)}(0) = 1$ and $g_\beta^{E(L)}(0) = 1$ and that $c_\beta(\tau)$ and $r_\beta(\tau)$ verifies the same equations as $\partial C_{eq}(\tau)/\partial h^2$ and $\partial R_{eq}(\tau)/\partial h^2$ in the β regime. This is indeed very much expected, actually unavoidable, from our analysis of the $\omega = 0$ limit and therefore we do not reproduce the details.

Let us focus now on the large x behaviour. The analysis of the large x behaviour is very similar for all the scaling functions. Thus, we detail it only for one of them, $f_\beta^L(x)$. In order to obtain the equation verified by this function we consider the limit $\hat{\tau} \gg 1$, keeping x finite, of eq. (84). In this limit we have that $c_1^{(\omega)}(\hat{\tau}) \approx \hat{\tau}^b/\sqrt{\epsilon} f_\beta^L(x)$, $c_{0\beta}(\hat{\tau}) \approx -B\hat{\tau}^b$ and

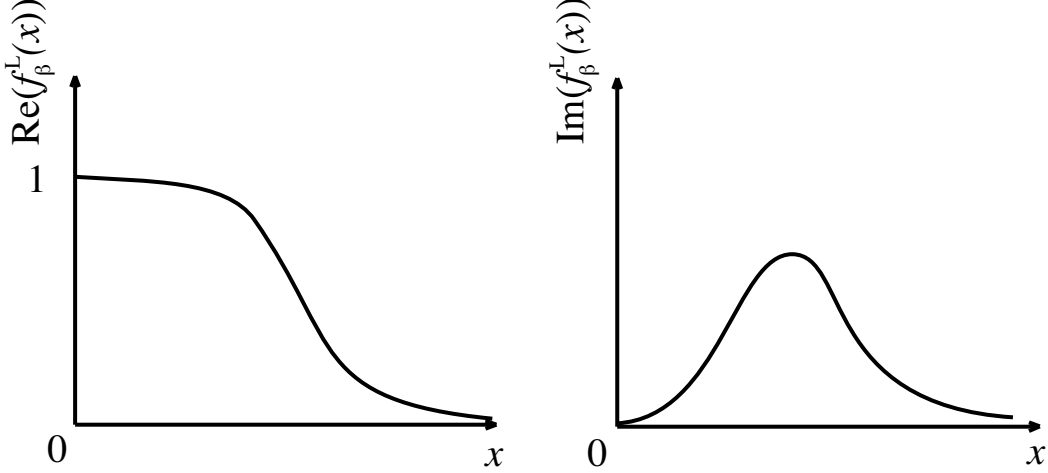


FIG. 2: Sketch of the real and imaginary part of the function f_β^L as a function of $x = \omega\tau$.

$r_{0\beta}(\hat{\tau}) \approx (Bb/T)\hat{\tau}^{b-1}$. In this regime, the leading terms are of order $\hat{\tau}^{2b}$:

$$\begin{aligned}
0 = & \left\{ \frac{(p-2)(1-q)}{2} f_\beta^L(x) - bq f_\beta^L(x) \int_0^{1/2} du \cos(ux) u^{b+1} \right. \\
& - bq \int_{1/2}^1 du \cos(ux) u^{b-1} [(1+u)^b f_\beta^L(x(1+u)) + (1-u)^b f_\beta^L(x(1-u))] \\
& \left. - bq \int_1^\infty du \cos(ux) u^{b-1} [(1+u)^b f_\beta^L(x(1+u)) + (u-1)^b f_\beta^L(x(u-1))] \right\} \hat{\tau}^{2b}. \quad (85)
\end{aligned}$$

This implies that the term into the curl brackets must vanish for all x . This defines an eigenvector equation for $f_\beta^L(x)$. For this eigenvector equation to be obeyed, $f_\beta^L(x)$ must go to zero for large x as $\cos(x+\varphi)/x^{1+b}$ (φ is a phase obtained from Eq. (85)), coming from the neighborhood of $u=1$ in the above integrals. The other functions f_β^E , g_β^L , g_β^E behave exactly in the same way.

Finally, it is important to notice that the functions $f_\beta^L(x)$, $g_\beta^L(x)$, $f_\beta^E(x)$, $g_\beta^E(x)$ are in principle complex functions. As before, we only discuss $f_\beta^L(x)$, since other functions behave similarly. In the limit $\omega \rightarrow 0$, since $c_1^{(\omega)}(\tau)$ is nothing but the derivative with respect to a constant field of the correlation function, $c_1^{(\omega)}(\tau)$, $r_1^{(\omega)}(\tau)$ and, hence, $f_\beta^L(0)$ must be real. As a consequence, we expect that while the real part of $f_\beta^L(x)$ is of $O(1)$ at small x and decreases to zero as $x \gg 1$, the imaginary part of $f_\beta^L(x)$ is equal to zero for $x=0$, then it reaches a maximum around $x \sim 1$, and it decreases to zero at large x . The behavior of the real and the imaginary part of the function $f_\beta^L(x)$ are schematically sketched in FIG. 2.

F. α -regime

We now consider the α -regime, where the unperturbed correlation and response functions can be expressed as scaling functions of the variable τ/τ_α . The α relaxation time τ_α diverges as $\epsilon^{-1/2a-1/2b}$ as the dynamical transition is approached. We consider frequencies ω of order of the inverse of τ_α and set: $\omega = \bar{\omega}/\tau_\alpha$, $\bar{\tau} = \tau/\tau_\alpha$. In this regime one has:

$$\begin{aligned} C_0(\tau) &\simeq q + c_{0\alpha}(\bar{\tau}), \\ R_0(\tau) &\simeq \frac{1}{\tau_\alpha} r_{0\alpha}(\bar{\tau}). \end{aligned} \tag{86}$$

In the following we will focus on the early α -regime, where $c_{0\alpha}(\bar{\tau}) = c_{0\beta}(\bar{\tau}) \approx -B\bar{\tau}^b$ and $r_{0\alpha} = r_{0\beta} \approx (Bb/T)\bar{\tau}^{b-1}$. As discussed in Sec. III B 2, close to the critical point the most general scaling form for the second order correlation and response functions in the α -regime corresponds to the one of Eq. (33). However, in the early α -regime the scaling laws for $c_1^{(\omega)}$ and $r_1^{(\omega)}$ can be factorized in the following way [see Eq. (35)]:

$$\begin{aligned} c_1^{(\omega)}(\tau) &\simeq \frac{1}{\epsilon} c_\alpha(\bar{\tau}) f_\alpha^E(\omega\tau), \\ r_0^{(\omega)}(\tau) &\simeq \frac{1}{\epsilon} \frac{1}{\tau_\alpha} r_\alpha(\bar{\tau}) g_\alpha^E(\omega\tau). \end{aligned} \tag{87}$$

As before we shall verify that this scaling ansatz is consistent with the dynamical equations and obtain the asymptotic behavior of f_α^E and g_α^E . This turns out to be particularly easy since plugging the scaling Ansatz into the full equations one finds that f_α^E and g_α^E verify the very same equations than f_β^L and g_β^L . This is expected from the physical point of view since early α and late β behavior should match. As a consequence, their asymptotic behavior is the same one discussed in the previous section.

In Fig. 3 we show a sketch of the behavior of $|c_1^{(\omega)}(\tau)|$ that summarizes all our previous findings.

One can now easily derive explicitly the critical behaviour of the non-linear susceptibility, $\chi_{3\omega}(\omega)$ close to the dynamical transition, as done in Sec. III.

V. CONCLUSION

In this work, we have studied in detail the non-linear response of supercooled liquids. Although we are able to provide precise statements within a Mode-Coupling approach, some

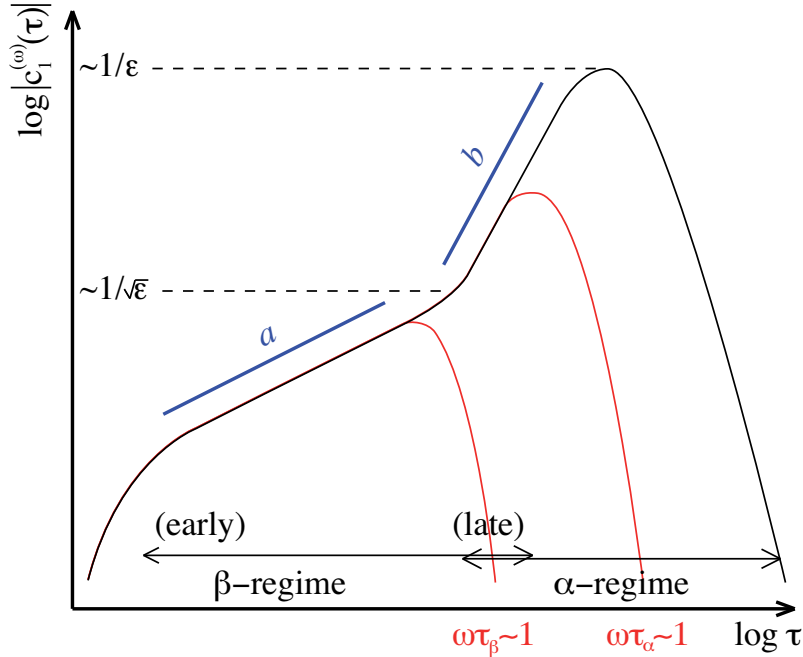


FIG. 3: Sketch of $\log|c_1^{(\omega)}(\tau)|$ as a function of $\log \tau$. In the $\omega \rightarrow 0$ limit $c_1^{(\omega)}(\tau)$, behaves as $\chi_T(\tau)$, i.e., it scales as $1/\sqrt{\epsilon}(\tau/\tau_\beta)^a$ in the early β -regime and as $1/\sqrt{\epsilon}(\tau/\tau_\beta)^b$ in the late β -regime (or, equivalently, as $1/\epsilon(\tau/\tau_\alpha)^b$ in the early α -regime). In the α -regime $|c_1^{(\omega)}(\tau)|$ reaches a maximum of order $1/\epsilon$. At finite frequency ω , $|c_1^{(\omega)}(\tau)|$ drops to values of $O(1)$ as $\tau \gtrsim 1/\omega$ (red curves).

of our results are in fact more general and only require Time-Temperature Superposition to hold. An important theoretical result is the relation (24) between the non-linear response $\chi_3(\omega)$ and the temperature derivative of the usual linear susceptibility, $d\chi_1(2\omega)/dT$, valid at small frequencies. This emphasizes again that the non trivial collective properties encoded in three- and four-point correlations and susceptibilities are revealed (at zero wave-vector) by the temperature (or density) derivative of standard two-body correlations and response. For larger frequencies, the main results of this paper are summarized in Fig. 1, where the MCT predictions are sketched. Five different frequency regimes are identified: $\chi_3(\omega)$ exhibits a peak around frequencies of the order of half the inverse of the structural relaxation time of the system τ_α . The height of the peak grows as $(T - T_c)^{-1}$ as the critical temperature is

approached. For higher frequencies, $\chi_3(\omega)$ decays as power laws, with an exponent equal to $-b$ in the late β regime, to $-a$ in the early β regime, and finally to -3 at high frequencies.

Our results should be directly applicable to the non-linear dielectric constant of molecular glasses in the weakly supercooled regime where MCT is expected to be relevant, and for describing the non-linear compressibility of hard-sphere colloids close to the glass transition, where MCT does a fair job at describing their relaxation properties. However, it is well known that MCT fails for deeply supercooled liquids, when activated events start playing a major role in the relaxation. The detailed shape of $\chi_3(\omega)$ would require a full theoretical description of the dynamics in this regime, which is unavailable to date. Still, the general low frequency relation between χ_3 and $d\chi_1/dT$, supplemented with the property of Time-Temperature superposition, allows one to give a firmer basis to the scaling relation conjectured in [15], namely that:

$$\chi_3(\omega) \approx \chi_3^* \mathcal{G}(\omega\tau_\alpha), \quad (88)$$

where \mathcal{G} is a scaling function, and $\chi_3^* \propto d\ln\tau_\alpha/d\ln T$ is the peak value of the temperature derivative of $\chi_1(\omega)$, as measured in [21, 22, 23]. Following [10, 11, 21], we expect χ_3^* to increase as a power of the dynamical correlation length $\xi(T)$. The detailed shape of \mathcal{G} would obviously be worth knowing in order to compare with upcoming experimental results. As a guide, we give the result obtained assuming a Havriliak-Nagami form for the susceptibility and the validity of the relation between χ_3 and $d\chi_1/dT$ at all frequencies, which has no justification apart from suggesting possible fitting functions. One finds:

$$\mathcal{G}_{HN}(u) = \frac{(iu)^b}{(1 + (iu)^b)^{1+c}}, \quad (89)$$

where b, c are fitting exponents.

Among open problems worth investing is the extension of the present theory to the aging regime of glasses and spin-glasses. From an experimental point of view, a detailed study of the role of the electric field on the glass properties of dipolar liquids (such as glycerol) would be very interesting. For example, the evolution of the glass transition temperature as a function of the field E would allow one to measure the proportionality coefficient κ appearing in Eq. (24). In spin-glasses, a detailed measurement of $\chi_3(\omega)$ would allow to shed light on the existence of spin-glass transition at non zero field, as argued in [15]. The behaviour of $\chi_3(\omega, t_w)$ in the aging phase would furthermore be a very useful probe of the

aging process in spin-glasses, in particular during rejuvenation cycles. Numerical simulations of $\chi_3(\omega, t_w)$, using the zero-field techniques developed in [26, 27], would be worth pursuing.

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Appendix 1: Analysis of Eq. (75)

For simplicity, and because in this appendix ω is just an external parameter, we omit the second argument of \hat{f}_β and \hat{g}_β , writing for instance $\hat{g}_\beta(\tau/\tau_\beta)$ for $\hat{g}_\beta(\tau/\tau_\beta, \omega\tau)$.

We start by analyzing the first integral appearing in Eq. (75). Since $R_0(\tau)$ is singular at small times, it is convenient to rewrite it in the following way:

$$\begin{aligned} & (p-1) \int_0^\tau d\sigma e^{-i\omega\sigma} [C_0(\sigma)]^{p-2} R_0(\sigma) \left[r_1^{(\omega)}(\tau - \sigma) \right. \\ & \quad \left. - r_1^{(\omega)}(\tau) \theta\left(\frac{\tau}{2} - \sigma\right) \right] \\ & + (p-1) r_1^{(\omega)}(\tau) \int_0^{\tau/2} d\sigma e^{-i\omega\sigma} [C_0(\sigma)]^{p-2} R_0(\sigma). \end{aligned}$$

Using FDT one can easily evaluate the second integral which, up to subleading terms equals:

$$\begin{aligned} & (p-1) r_1^{(\omega)}(\tau) \int_0^{\tau/2} d\sigma e^{-i\omega\sigma} [C_0(\sigma)]^{p-2} R_0(\sigma) \\ & = \frac{1 - [C_0(\tau/2)]^{p-1}}{T} r_1^{(\omega)}(\tau) \\ & = \left[\frac{1 - q^{p-1}}{T} + \frac{q^{p-1} - [C_0(\tau/2)]^{p-1}}{T} \right] r_1^{(\omega)}(\tau). \end{aligned} \tag{90}$$

In order to evaluate the first integral, it is useful to split the integration domain into four time intervals: $(0, \delta\tau_\beta)$, $(\delta\tau_\beta, \tau/2)$, $(\tau/2, \tau - \eta\tau_\beta)$, $(\tau - \eta\tau_\beta, \tau)$, where δ and η will be sent to

zero at the end of the calculation. The contribution due to the first interval reads:

$$\begin{aligned}
& (p-1) \int_0^{\delta\tau_\beta} d\sigma e^{-i\omega\sigma} [C_0(\sigma)]^{p-2} R_0(\sigma) \\
& \quad \times \left[r_1^{(\omega)}(\tau - \sigma) - r_1^{(\omega)}(\tau) \right] \\
& \simeq \frac{1}{T} \int_0^{\delta\tau_\beta} d\sigma \frac{d}{d\sigma} [C_0(\sigma)]^{p-1} \left[\frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} r_\beta^E \left(\frac{\sigma}{\tau_\beta} \right) \frac{\sigma}{\tau_\beta} \right] \\
& \propto \epsilon^{1/a-1/2} \delta,
\end{aligned}$$

which turns out to be a subleading contribution in the $\delta \rightarrow 0$ limit. We now evaluate the integral in the interval $(\tau - \eta\tau_\beta, \tau)$:

$$\begin{aligned}
& (p-1) \int_{\tau-\eta\tau_\beta}^{\tau} d\sigma e^{-i\omega\sigma} [C_0(\sigma)]^{p-2} R_0(\sigma) r_1^{(\omega)}(\tau - \sigma) \\
& = (p-1) q^{p-2} e^{-i\omega\tau} R_0(\tau) \int_0^{\eta\tau_\beta} d\sigma r_1^{(\omega)}(\sigma) \\
& = -\frac{(p-1)}{T} q^{p-2} e^{-i\omega\tau} R_0(\tau) c_1^{(\omega)}(\eta\tau_\beta) \propto \frac{\eta^a}{\sqrt{\epsilon}},
\end{aligned}$$

which vanishes in the limit $\eta \rightarrow 0$. The last equality has been derived using the generalized FDT, Eq. (20), which holds for small enough times (such that $\omega\tau \ll 1$).

We are now left with the computation of the integral in the two central intervals. The integral in the interval $(\tau/2, \tau - \eta\tau_\beta)$ is given by:

$$\begin{aligned}
& (p-1) \int_{\tau/2}^{\tau-\eta\tau_\beta} d\sigma e^{-i\omega\sigma} [C_0(\sigma)]^{p-2} R_0(\sigma) r_1^{(\omega)}(\tau - \sigma) \\
& = (p-1) q^{p-2} \int_{\tau/2}^{\tau-\eta\tau_\beta} d\sigma e^{-i\omega\sigma} \frac{\sqrt{\epsilon}}{\tau_\beta} r_{0\beta} \left(\frac{\sigma}{\tau_\beta} \right) \\
& \quad \times \frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} \hat{g}_\beta \left(\frac{\tau - \sigma}{\tau_\beta} \right) \\
& = \frac{(p-1)q^{p-2}}{\tau_\beta} \int_{\hat{\tau}/2}^{\hat{\tau}} ds e^{-i\hat{\omega}s} r_{0\beta}(s) \hat{g}_\beta(\hat{\tau} - s).
\end{aligned} \tag{91}$$

Here we have taken the limit $\eta \rightarrow 0$ directly. Analogously, we can evaluate the integral in the last interval $(\delta\tau_\beta, \tau/2)$:

$$\begin{aligned}
& (p-1) \int_{\delta\tau_\beta}^{\tau/2} d\sigma e^{-i\omega\sigma} [C_0(\sigma)]^{p-2} R_0(\sigma) \\
& \quad \times \left[r_1^{(\omega)}(\tau - \sigma) - r_1^{(\omega)}(\tau) \right] \\
& = \frac{(p-1)q^{p-2}}{\tau_\beta} \int_0^{\hat{\tau}/2} ds e^{-i\hat{\omega}s} r_{0\beta}(s) \\
& \quad \times [\hat{g}_\beta(\hat{\tau} - s) - \hat{g}_\beta(\hat{\tau})].
\end{aligned} \tag{92}$$

Now we consider the other two integrals of Eq. (75). In order to do that it is convenient to change the integration variable $\sigma \rightarrow \tau - \sigma$ and to rewrite them as (up to subleading terms):

$$\begin{aligned}
& (p-1) \int_0^\tau d\sigma e^{i\omega\sigma} R_0(\sigma) \\
& \times \left[[C_0(\tau - \sigma)]^{p-2} r_1^{(\omega)}(\tau - \sigma) \right. \\
& \quad + (p-2) [C_0(\tau - \sigma)]^{p-3} R_0(\tau - \sigma) c_1^{(\omega)}(\tau - \sigma) \\
& \quad - [C_0(\tau)]^{p-2} r_1^{(\omega)}(\tau) \theta\left(\frac{\tau}{2} - \sigma\right) \\
& \quad \left. - (p-2) [C_0(\tau)]^{p-3} R_0(\tau) c_1^{(\omega)}(\tau) \theta\left(\frac{\tau}{2} - \sigma\right) \right] \\
& + (p-1) [C_0(\tau)]^{p-2} \frac{1-q}{T} r_1^{(\omega)}(\tau) \\
& + (p-1)(p-2) [C_0(\tau)]^{p-3} \frac{1-q}{T} R_0(\tau) c_1^{(\omega)}(\tau).
\end{aligned}$$

Again, one can show that the contribution at short time can be neglected in the $\delta \rightarrow 0$ limit. The integrals in the interval $(0, \delta\tau_\beta)$ yield:

$$\begin{aligned}
& (p-1) \int_0^{\delta\tau_\beta} d\sigma e^{i\omega\sigma} R_0(\sigma) \\
& \quad \times \left[[C_0(\tau - \sigma)]^{p-2} r_1^{(\omega)}(\tau - \sigma) - q^{p-2} r_1^{(\omega)}(\tau) \right] \\
& = \frac{(p-1)q^{p-2}}{\sqrt{\epsilon}\tau_\beta T} \left(\frac{\tau}{\tau_\beta}\right) \int_0^{\delta\tau_\beta} d\sigma \frac{\sigma}{\tau_\beta} \frac{d}{d\sigma} C_0(\sigma) \\
& \propto \epsilon^{1/a-1/2} \delta.
\end{aligned}$$

and,

$$\begin{aligned}
& (p-1)(p-2) \int_0^{\delta\tau_\beta} d\sigma e^{i\omega\sigma} R_0(\sigma) \tag{93} \\
& \quad \times \left[[C_0(\tau - \sigma)]^{p-3} R_0(\tau - \sigma) c_1^{(\omega)}(\tau - \sigma) \right. \\
& \quad \quad \left. - q^{p-3} R_0(\tau) c_1^{(\omega)}(\tau) \right] \\
& \approx \frac{(p-1)(p-2)}{T} R_0(\tau) \frac{d}{d\tau} \left(R_0(\tau) c_1^{(\omega)}(\tau) C_0(\tau)^{p-3} \right) \\
& \quad \times \int_0^{\delta\tau_\beta} d\sigma \sigma \frac{d}{d\sigma} C_0(\sigma) \propto \epsilon^{1/a} \delta.
\end{aligned}$$

Similarly, in the interval $(\tau - \eta\tau_\beta, \tau)$, using the generalized FDR relation, one has:

$$\begin{aligned}
& (p-1) \int_{\tau-\eta\tau_\beta}^{\tau} d\sigma e^{i\omega\sigma} R_0(\sigma) \left[[C_0(\tau-\sigma)]^{p-2} r_1^{(\omega)}(\tau-\sigma) \right. \\
& \quad \left. + (p-2) [C_0(\tau-\sigma)]^{p-3} R_0(\tau-\sigma) c_1^{(\omega)}(\tau-\sigma) \right] \\
&= -\frac{(p-1)}{T} e^{i\omega\tau} R_0(\tau) \\
& \quad \times \int_0^{\eta\tau_\beta} d\sigma \frac{d}{d\sigma} \left([C_0(\sigma)]^{p-1} c_1^{(\omega)}(\sigma) \right) \\
&= -\frac{(p-1)}{T} e^{i\omega\tau} R_0(\tau) c_1^{(\omega)}(\eta\tau_\beta) \propto \frac{\eta^a}{\sqrt{\epsilon}}
\end{aligned}$$

We have now to evaluate the two integrals in the two central intervals. We first consider the following term:

$$\begin{aligned}
& (p-1) \int_{\tau/2}^{\tau-\eta\tau_\beta} d\sigma e^{i\omega\sigma} R_0(\sigma) [C_0(\tau-\sigma)]^{p-2} r_1^{(\omega)}(\tau-\sigma) \\
&= (p-1) q^{p-2} \int_{\tau/2}^{\tau-\eta\tau_\beta} d\sigma e^{i\omega\sigma} \\
& \quad \times \left[\frac{\sqrt{\epsilon}}{\tau_\beta} \hat{g}_\beta \left(\frac{\sigma}{\tau_\beta} \right) \frac{1}{\sqrt{\epsilon}} \frac{1}{\tau_\beta} \hat{g}_\beta \left(\frac{\tau}{\tau_\beta} - \frac{\sigma}{\tau_\beta} \right) \right] \\
&= \frac{(p-1)q^{p-2}}{\tau_\beta} \int_{\hat{\tau}/2}^{\hat{\tau}} ds e^{i\hat{\omega}s} r_{0\beta}(s) \hat{g}_\beta(\hat{\tau}-s),
\end{aligned} \tag{94}$$

which, together with Eq. (91) gives a contribution of the form:

$$\frac{2(p-1)q^{p-2}}{\tau_\beta} \int_{\hat{\tau}/2}^{\hat{\tau}} ds \cos(\hat{\omega}s) r_{0\beta}(s) \hat{g}_\beta(\hat{\tau}-s) \tag{95}$$

We then consider the same integral in the interval $(\delta\tau_\beta, \tau/2)$, which yields:

$$\begin{aligned}
& (p-1) \int_{\delta\tau_\beta}^{\tau/2} d\sigma e^{i\omega\sigma} R_0(\sigma) \\
& \quad \times \left[[C_0(\tau-\sigma)]^{p-2} r_1^{(\omega)}(\tau-\sigma) - q^{p-2} r_1^{(\omega)}(\tau) \right] \\
& \simeq \frac{(p-1)q^{p-2}}{\tau_\beta} \int_0^{\hat{\tau}/2} ds e^{i\hat{\omega}s} r_{0\beta}(s) \\
& \quad \times [\hat{g}_\beta(\hat{\tau}-s) - \hat{g}_\beta(\hat{\tau})],
\end{aligned} \tag{96}$$

which, together with Eqs. (92) gives

$$\begin{aligned}
& \frac{2(p-1)q^{p-2}}{\tau_\beta} \int_0^{\hat{\tau}/2} ds \cos(\hat{\omega}s) r_{0\beta}(s) \\
& \quad \times [\hat{g}_\beta(\hat{\tau}-s) - \hat{g}_\beta(\hat{\tau})],
\end{aligned} \tag{97}$$

Finally, we analyze the two integrals which contains $c_1^{(\omega)}(\tau - \sigma)$. The first one reads:

$$\begin{aligned}
& (p-1)(p-2) \int_{\tau/2}^{\tau-\eta\tau\beta} d\sigma e^{i\omega\sigma} R_0(\sigma) [C_0(\tau - \sigma)]^{p-3} \\
& \quad \times R_0(\tau - \sigma) c_1^{(\omega)}(\tau - \sigma) \\
& = (p-1)(p-2) q^{p-3} \int_{\tau/2}^{\tau-\eta\tau\beta} d\sigma e^{i\omega\sigma} \frac{\sqrt{\epsilon}}{\tau_\beta} r_{0\beta} \left(\frac{\sigma}{\tau_\beta} \right) \\
& \quad \times \frac{\sqrt{\epsilon}}{\tau_\beta} r_{0\beta} \left(\frac{\tau}{\tau_\beta} - \frac{\sigma}{\tau_\beta} \right) \frac{1}{\sqrt{\epsilon}} \hat{f}_\beta \left(\frac{\tau}{\tau_\beta} - \frac{\sigma}{\tau_\beta} \right) \\
& = \frac{\sqrt{\epsilon}(p-1)(p-2)q^{p-3}}{\tau_\beta} \int_{\hat{\tau}/2}^{1-\eta} ds e^{i\hat{\omega}s} \\
& \quad \times r_{0\beta}(s) r_{0\beta}(\hat{\tau} - s) \hat{f}_\beta(\hat{\tau} - s) \propto \frac{\sqrt{\epsilon} \log \eta}{\tau_\beta}.
\end{aligned}$$

This term is $\sqrt{\epsilon}$ smaller than the contributions given by Eqs. (95) and (97), and can thus be neglected. The second term is given by:

$$\begin{aligned}
& (p-1)(p-2) \int_{\delta\tau_\beta}^{\tau/2} d\sigma e^{i\omega\sigma} R_0(\sigma) \\
& \quad \left[[C_0(\tau - \sigma)]^{p-3} R_0(\tau - \sigma) c_1^{(\omega)}(\tau - \sigma) \right. \\
& \quad \quad \left. - q^{p-3} R_0(\tau) c_1^{(\omega)}(\tau) \right] \\
& = \frac{\sqrt{\epsilon}(p-1)(p-2)q^{p-3}}{\tau_\beta} \int_{\delta}^{\hat{\tau}/2} ds e^{i\hat{\omega}s} r_{0\beta}(s) \\
& \quad \times \left[r_{0\beta}(\hat{\tau} - s) \hat{f}_\beta(\hat{\tau} - s) - r_{0\beta}(\hat{\tau}) \hat{f}_\beta(\hat{\tau}) \right],
\end{aligned}$$

which again leads to a subleading contribution.

In order to get the scaling equation for $r_1^{(\omega)}(\tau)$ from Eq. (75), one should also evaluate $z_1^{(\omega)}$. However, we notice that $z_1^{(\omega)}$ must be of order one in the whole frequency domain, as it is just the quantity which enforce the spherical constraint. For instance, in the $\omega \rightarrow 0$ limit it is proportional to the energy per spin and it is equal to $1/T$. As a consequence, we can skip the explicit evaluation of $z_1^{(\omega)}$, keeping in mind that it should result in a contribution of $O(1)$, which is, thus, negligible in Eq. (83).

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