PHYSICAL REVIEW B **72**, 064204 (2005)

Nonlinear susceptibility in glassy systems: A probe for cooperative dynamical length scales

Jean-Philippe Bouchaud[1,*] and Giulio Biroli[2,†]

[1]*Service de Physique de l'État Condensé, Orme des Merisiers, CEA Saclay, 91191 Gif sur Yvette Cedex, France and Science and Finance, Capital Fund Management, 6-8 Bd Haussmann, 75009 Paris, France*

[2]*Service de Physique Théorique, Orme des Merisiers, CEA Saclay, 91191 Gif sur Yvette Cedex, France*

(Received 22 February 2005; published 11 August 2005)

We argue that for generic systems close to a critical point, an extended fluctuation-dissipation relation connects the low frequency nonlinear (cubic) susceptibility to the four-point correlation function. In glassy systems, the latter contains interesting information on the heterogeneity and cooperativity of the dynamics. Our result suggests that if the abrupt slowing down of glassy materials is indeed accompanied by the growth of a cooperative length ℓ, then the nonlinear, 3ω response to an oscillating field (at frequency ω) should substantially increase and give direct information on the temperature (or density) dependence of ℓ. The analysis of the nonlinear compressibility or the dielectric susceptibility in supercooled liquids, or the nonlinear magnetic susceptibility in spin-glasses, should give access to a cooperative length scale, that grows as the temperature is decreased or as the age of the system increases. Our theoretical analysis holds exactly within the mode-coupling theory of glasses.

DOI: 10.1103/PhysRevB.72.064204 PACS number(s): 64.70.Pf, 61.43.Fs, 64.60.Cn

I. INTRODUCTION

A yet unexplained property of fragile glasses is the extremely fast rise of their relaxation time (or viscosity) as the temperature is lowered, much faster than predicted by a simple thermal activation formula.[1] If interpreted in terms of an effective activation energy, the latter increases by a factor of 5–10 between $1.5T_g$ and the glass transition temperature T_g. The basic mechanism for this increase is not well understood, but it is reasonable to think that it is intimately related to cooperative effects[2,3] and possibly to the presence of an underlying critical point.[4–11] The dynamics becomes sluggish and the activation energy increases because larger and larger regions of the material have to move in a correlated way to allow for a substantial motion of individual particles. Long *time* scales must be somehow associated with large *length* scales. Although the idea of a cooperative length has been discussed in the context of glasses for many years,[2,12] it is only recently that proper measures of cooperativity (and of the size of the rearranging regions) were proposed theoretically[8] (see Ref. 13 for earlier insights) and measured in numerical simulations[13–16] (see also Refs. 12, 17, and 18 for related experimental work). The idea is to measure how the dynamics is correlated in space; technically, this involves a four-point correlation function which measures the spatial correlations of the temporal correlation [see Eq. (9) below for a more precise definition]. Recent extensive numerical evaluations of this four-point correlation function in Lennard-Jones systems have confirmed the existence of a growing length scale as temperature is decreased,[11,15,16] and have shown that different observables, such as the relaxation time or the diffusion constant, scale as powers of this length, emphasizing its crucial importance as far as the physics is concerned. In the framework of granular systems, diverging length scales near the jamming transition have also been reported in numerical studies of model systems.[19]

Although many different theoretical approaches to the glass transition[4–11] can potentially explain the existence of such a growing dynamical correlation length, these theories lead to rather different quantitative predictions for the behavior of the four-point correlation function (see the detailed discussion in Ref. 20). Thus, experiments measuring directly this four-point function would be extremely valuable to refine our understanding of the glass phenomenon and prune down the number of candidate models. Up to now, unfortunately, only indirect experimental indications of a cooperative length scale associated to heterogeneous dynamics have been reported.[12,17,21]

On a different front, that of spin-glasses, length scale ideas have also been expressed in recent years to account for nonequilibrium phenomena such as aging, memory, and rejuvenation effects.[22–27] Although spin-glass order is not easy to define nor to detect, the idea is that some kind of domain growth occurs, whereby spin-glass correlations establish on larger and larger length scales as the age of the system increases. The growth of this "coherence length" has been established numerically by comparing two replicas of the same system.[28–32] This trick is obviously inaccessible to experimentalists, who have nevertheless provided indirect evidence of a growing length scale, and some indications on its rate of growth with time and temperature.[25,33–35] Again, a direct measure of this length scale is lacking—finding a clear-cut experimental signal of a cooperative length in disordered, amorphous systems would certainly be a major breakthrough.[12]

The aim of this paper is to point out that in *slow* glassy systems at equilibrium, the nonlinear (cubic) response to an external field (electric, magnetic, pressure, etc.) in fact probes directly the four-point correlation function mentioned above, and therefore the cooperative length it may contain. Our main prediction, detailed below, is that the 3ω harmonic response to an ac field of frequency ω and amplitude h is given by $\chi_3(\omega,T)h^3$, where the nonlinear susceptibility χ_3 behaves at low frequency as

$$\chi_3(\omega,T) = \frac{\chi_s^2}{k_B T}\ell^{2-\bar{\eta}}\mathcal{H}(\omega\tau). \tag{1}$$

In the above relation, χ_s is the static linear susceptibility, $\mathcal{H}$ a certain complex function that depends weakly on temperature, and τ is the temperature dependent relaxation time of the system, which can be directly measured using the linear susceptibility. The cooperative length ℓ (measured in units of the microscopic length ξ obtained from the two point correlation function) is expected to grow as the temperature is reduced, and $\bar{\eta}$ an exponent related to the spatial structure of the four-point correlation function.[36] Our central result, Eq. (1), that we will motivate below, states that (a) the nonlinear susceptibility has the same frequency scaling as the linear susceptibility, which is not surprising and (b) it grows as the cooperative length increases, which should allow a direct experimental test of the relationship between length and time scales in glassy systems.

The above prediction holds for equilibrium systems; we will however see below that in the case of glasses and spin-glasses in a field, $\mathcal{H}(0)=0$. Below the glass transition temperature, on the other hand, the system by definition falls out of equilibrium. Its dynamics becomes nonstationary and exhibits aging, which means that the effective relaxation time of the system increases with the age t_w of the system.[37,38] This increase of the relaxation time is again most probably related to the growth of a coherence length in the system, $\ell_w=\ell(t_w)$. On very general grounds,[38] one expects that the generalization of the equilibrium result (1) to the aging case will read, for a large class of systems:

$$\chi_3(\omega,t_w) = \frac{\chi_s^2}{k_B T}\ell_w^{2-\bar{\eta}}\tilde{\mathcal{H}}\left(\frac{h(1/\omega)}{h(t_w)}\right), \tag{2}$$

where h is a certain increasing function, which in simple domain growth models is the typical size of the domains. From experimental results,[37] a plausible guess is that simple aging will hold in spin-glasses, i.e., $h(t_w)=t_w$. (Of course, more complicated scaling forms, with an infinite number of time domains, may hold in some cases, such as models with full replica symmetry breaking.[38]) In the above equation, $\tilde{\mathcal{H}}$ is another scaling function, which also contains possible violations of the standard fluctuation dissipation theorem and the appearance of a nontrivial, t_w dependent, effective temperature.[39] Equation (2) should in any case allow one to extract from nonlinear aging susceptibilities a nonequilibrium coherence length, in a much more direct way than previous attempts.

As for comparison with previous works, the divergence of the *static* nonlinear susceptibility at the spin-glass (in zero field) or dipolar-glass transition, displayed by Eq. (1) at $\omega=0$, is of course well documented, both theoretically[40] and experimentally.[40–42] The generalization to the dynamical nonlinear susceptibility in the critical region was also discussed[22,43,44] but not, to the best of our knowledge, its generalization to the nonequilibrium, aging regime, Eq. (2). The situation for glassforming liquids is quite different, since no static phase transition with a diverging static susceptibility has ever been identified, neither in experiments nor in simulations. Purely based on an analogy with spin-glasses, it was suggested in Ref. 45 that the nonlinear dielectric constant of molecular glasses might grow as the glass phase is approached (although this was not borne out by the experiments done at that time[45]). A similar suggestion was made in Ref. 46 concerning the nonlinear compressibility of soft sphere binary mixtures, with numerical results that are not incompatible with a substantial increase of $\chi_3(\omega\sim 0)$ as the temperature is lowered. We will show below that such a growth is indeed expected, although the theoretical situation for glass formers is much less clear than for spin-glasses—in particular, although χ_3 is growing may never diverge in glass formers. Different scenarios for the glass transition can be envisaged and lead to quite different predictions, for example on the value of $\bar{\eta}$ and on the relationship between ℓ and τ (or t_w).

In the following section we will give some physical arguments that motivate our results, and muster the predictions of different theoretical models for glassformers. A more detailed and technical discussion is then presented in Sec. III. Finally our conclusions are presented in Sec. IV.

II. PHYSICAL ARGUMENTS AND RESULTS

A. Spin-glasses

1. Order parameter and nonlinear susceptibility

Let us first focus on spin-glasses in zero external magnetic field, $H=0$. These systems are thought, both theoretically and experimentally, to have a nonzero transition temperature below which the magnetization profile, $\langle s_x\rangle$, freezes into one (or more) amorphous configurations. The ordered state is characterized by a nonzero Edwards-Anderson (EA) parameter $q=[\langle s_x\rangle^2]$, where $\langle\cdots\rangle$ indicates thermal averaging and the brackets a spatial (or disorder) average. These systems display an unusual type of long-range order, which cannot be detected using either one body or two-body spin-spin correlations: because the ordering pattern is random, the average magnetization $[\langle s_x\rangle]$ remains zero and the spin-spin correlation $[\langle s_x s_y\rangle]$ short-ranged, even in the spin-glass phase. Correspondingly, the linear susceptibility, related by a fluctuation dissipation theorem (FDT) to the integral of the spin-spin correlation function, does not diverge as T_g is approached, even if some long-ranged correlations appear in the system. The way to get rid of the spurious cancellation between strongly correlated and strongly anticorrelated spins is well known: exactly as one should square $\langle s_x\rangle$ to obtain a nonzero Edwards-Anderson parameter, one should also square $\langle s_x s_y\rangle$ before averaging over disorder. The integral over space of that quantity now diverges as T_g is approached, and in fact has two interesting physical interpretations.

(1) The first one is the susceptibility of the spin-glass order parameter to small random ordering fields. Imagine one adds small random magnetic fields h_x on every site. Using linear response, one can write, for a given sample at $T>T_g$ and $H=0$:

$$\delta\langle s_x\rangle = \frac{1}{k_B T}\sum_y \langle s_x s_y\rangle_0 h_y, \tag{3}$$

where the subscript 0 means that the correlation functions are evaluated at zero external field. Squaring this relation, summing over x and averaging over the random fields gives the sensitivity of the EA order parameter to a random pinning field:

$$\chi_{\mathrm{SG}} = \frac{\partial q}{\partial \langle h^2 \rangle} = \frac{1}{N(k_B T)^2} \sum_{x,y} [\langle s_x s_y \rangle_0^2]. \tag{4}$$

Clearly, the divergence of χ_{SG} signals an incipient instability towards an ordering pattern favored by the small pinning fields, exactly as the divergence of the usual two-body susceptibility signals an instability to ferromagnetic order, triggered by a small (uniform) magnetic field.

(2) As defined above, χ_{SG} has a clear theoretical interpretation but seems hard to access experimentally. Fortunately, χ_{SG} has another interpretation in terms of a nonlinear susceptibility, which can be directly measured. The intuitive idea is that the nonlinear susceptibility is actually a measure of the (quadratic) dependence of the linear susceptibility on the external field. Using again FDT the change of the connected correlation function between two spins (and hence of the linear susceptibility) induced by the field contains the term:

$$\delta[\langle s_x \rangle \langle s_y \rangle] \simeq \sum_{z,z'} \langle s_x s_z \rangle_0 \langle s_y s_{z'} \rangle_0 \frac{h^2}{(k_B T)^2}. \tag{5}$$

Averaging over space (or over disorder), only the terms $(z=y,x,z'=x,y)$ survive, the first one giving $[\langle s_x s_y \rangle_0^2]$ as in χ_{SG}. A more precise treatment for Ising spins at zero field leads to the exact relation $\chi_3(\omega=0) = -(3\chi_{\mathrm{SG}}-2)/(k_B T)$.[3]

Therefore, the static nonlinear susceptibility of spin-glasses diverges as the spin-glass transition temperature T_g is approached, a well-known effect that allows one to measure some of the critical exponents experimentally.[40,41] The physics behind the correlation induced amplification of χ_3 is clear: the influence of the polarization of spin s_x on s_y may be either positive or negative, but it has the same sign as the reverse influence of s_y on s_x. Therefore, the quadratic effect of an external field h on the dynamical correlation between any pair of spins has a well defined sign, in turn leading to a diverging nonlinear susceptibility as the size of correlated regions increases, even if the linear susceptibility remains small.

2. Nonzero external field and $T < T_g$

The case where a nonzero external field H is present is more subtle. In mean-field, the spin-glass phase survives in a whole region of the T, H plane, below the de Almeida-Thouless (AT) line. The spin-spin correlation function $[(\langle s_x s_y \rangle - \langle s_x \rangle \langle s_y \rangle)^2]$ is long-ranged in the whole spin- glass phase but is no longer directly related to the static nonlinear susceptibility. Some exact compensation mechanism[47,48] actually cancels the divergence in the combination of four-spin correlations appearing in $\chi_3(\omega=0)$. Therefore, the nonlinear susceptibility is finite in the whole spin-glass phase (including $H=0$ in strict mean-field). There is in particular no divergence of $\chi_3(\omega=0)$ on the AT line, except at $H=0$; rather, the nonlinear susceptibility is *discontinuous* across the AT phase transition.[49]

Within the droplet theory, on the other hand, the spin-glass is detroyed by any nonzero field in finite dimensions. Both the spin-glass and nonlinear susceptibilities are therefore finite when $H \neq 0$.[22,50] For $H=0$, a compensation mechanism similar to that of mean-field glasses is also at play, but does not prevent the nonlinear susceptibility to diverge for all $T \leqslant T_g$.[22] The replica field theory of spin-glasses in finite dimensions seems also compatible with a diverging nonlinear susceptibility for $T < T_g$ at zero field.[51]

3. Dynamical nonlinear susceptibility

The above qualitative arguments for the static nonlinear susceptibility can be extended to the dynamical case as well. As will be recalled below, the dynamical FDT gives

$$\langle s_x \rangle (t_1) = \frac{1}{k_B T} \sum_y \int dt_3 \frac{d}{dt_3} \langle s_x(t_1) s_y(t_3) \rangle_0 h_y(t_3). \tag{6}$$

Therefore, the change in the connected dynamical correlations between $s_x(t_1)$ and $s_y(t_2)$, induced by a uniform, but time dependent external field, will contain a term like

$$\sum_{z,z'} \int dt_3 dt_4 \frac{d}{dt_3} \langle s_x(t_1) s_z(t_3) \rangle_0 \frac{d}{dt_4} \langle s_y(t_2) s_{z'}(t_4) \rangle_0 h(t_3) h(t_4). \tag{7}$$

Repeating the same argument developed in the static case, i.e., averaging over space (or disorder) and using FDT to relate the connected correlation function to the dynamical linear susceptibility leads to a nonlinear response function that reads

$$\chi_3(t_1; t_2, t_3, t_4) \sim \sum_y \frac{d^3}{dt_2 dt_3 dt_4} [\langle s_x(t_1) s_y(t_3) \rangle_0 \langle s_x(t_4) s_y(t_2) \rangle_0]. \tag{8}$$

Taking t_1, t_2, t_3, t_4 all within an interval of the order of the relaxation time τ of the system, we see that the correlation function entering χ_3 above defines a cooperative length scale ℓ, such that the dynamics of s_x and s_y within this time interval is dominated by common events. This in turn leads to our scaling prediction, Eq. (1), near the transition temperature. The exact result for the dynamical $\chi_3(t_1; t_2, t_3, t_4)$ needs to be worked out carefully (see Sec. III), since FDT for higher order correlations is more involved than for two point functions.[52] Although different from the above naive expression, it indeed contains four-spin correlation functions that capture the cooperativity of the dynamics. Intuitively, again, the nonlinear response is strong if on the scale of the relaxation time, two spins feedback on each other's dynamics—this cross correlation is squared and survives averaging, even if the correlation itself is of random sign.

4. The aging regime

In the low temperature, spin-glass phase, the relaxation time τ is infinite and the age of the system t_w plays a crucial role—all time dependent correlation functions depend explicitly on t_w, which sets the time scale for the relaxation of the system,[38] and also for higher order correlation functions, such as the four-point correlation. Intuitively, exciting the system with a field of frequency $1/t_w$ will give the nonlinear response of a spin-glass equilibrated only up to a certain length scale $\ell_w = \ell(t_w)$.[53]

Interestingly, and contrarily to standard ordering transitions, spin-glasses are thought to be *critical* throughout their low temperature phase, in the sense that the space integral of the connected correlation function $[\langle s_x s_y \rangle_c^2]$ diverges for all $T < T_g$. In the special case of mean-field spin-glasses, the static nonlinear susceptibility within one phase is, as mentioned above, finite (except for $T = T_g$ and $H = 0$).[47] Except if some further cancellations operate at finite frequencies, an equation similar to Eq. (2) should hold in the aging phase of mean-field spin-glasses, but with an infinite number of time domains rather than a simple scaling variable such as ωt_w.[38] The explicit calculation of $\widetilde{\mathcal{H}}$ in the context of a spherical p-spin model would be extremely interesting; in particular one may ask whether the effective temperature appearing in the nonlinear response is the same as that appearing in the linear response.[54]

In the droplet picture,[22,50] and for the replica theory in finite dimensions,[51] the static nonlinear susceptibility diverges for all $T < T_g$ (at least for $H = 0$), and one should certainly observe a nonlinear susceptibility increasing as in Eq. (2). In the original droplet model with activated dynamics one predicts a function $\widetilde{\mathcal{H}}$ of $\ln \omega / \ln t_w$ associated with the logarithmic growth of $\ell(t_w)$. The peak value $\chi_3(t_w, \omega = 1/t_w)$ should grow as $\ell(t_w)^{2-\bar{\eta}}$. The numerical value of $2 - \bar{\eta}$ is yet unknown, but following Fisher and Huse,[22] one may expect $d - 3\theta \leq 2 - \bar{\eta} \leq d - \theta$ with $\theta \approx 0.2$ in three dimensions.

B. Structural glasses

1. Four-point density functions

Let us now discuss structural glasses. The important lesson we learn from spin-glasses is that a nontrivial amorphous type of long-range order can set in. In the case of glasses, the subtlety comes from the absence of quenched disorder; however, there has now been many papers exploring the idea of self-induced disorder which could drive a similar transition in homogeneous, frustrated systems (see, e.g., Ref. 38, and references therein). This has led, in particular, to the "Random First Order Transition" scenario,[4] where a glass transition of the same nature as the spin-glass transition in mean-field p-spin models takes place (see Ref. 7 for recent quantitative results). Whether a true transition of this type can exist in real systems with finite range interactions is still an actively debated issue; it is nevertheless extremely fruitful to explore the properties of systems for which this transition is, in some sense, nearly present. The order parameter in the would-be glass phase is the amplitude of the frozen in (random) density fluctuations $\delta\rho_x$. As for spin-glasses, the average of this quantity is zero, but $[\langle \delta\rho_x \rangle^2]$ is not, and plays the role of the Edwards-Anderson parameter. (The square brackets above now means an average over times longer than the relaxation time of the system, see Sec. II B 3.) Similarly, one expects $[\langle \delta\rho_x \delta\rho_y \rangle]$ not to show any interesting features (beyond that typical of a liquid structure factor), whereas its square may reveal long-range cooperative dynamics. The analog of the spin-glass and nonlinear susceptibilities discussed previously can be easily found in the case of glasses: the former can be seen as the susceptibility to a random external pinning field[55] that triggers the appearance of one particular type of frozen density fluctuation, whereas the latter is directly related to the nonlinear compressibility, i.e., the response to a pressure field that couples to the density. Other nonlinear responses to a field that couples to the degrees of freedom undergoing a glass transition are also relevant (for example, the dielectric response when the dipoles are strongly coupled to the translational degrees of freedom, such as in glycerol, OTP, etc.).

Let us directly focus on the dynamical susceptibility and postpone the discussion of its static limit to Sec. II B 3. Indeed, the analogy with spin-glasses has to be taken with a grain of salt (see Sec. II B 3). The dynamical four-point density function is defined as

$$G_4(r,t) = \langle \delta\rho_x(t=0)\,\delta\rho_x(t)\,\delta\rho_{x+r}(t=0)\,\delta\rho_{x+r}(t)\rangle - C^2(t), \quad C(t) \equiv \langle \delta\rho_x(t=0)\,\delta\rho_x(t)\rangle. \qquad (9)$$

The point of this paper is that this correlation function is related to the dynamical nonlinear response of the system to an external excitation that couples to the density.[56] Once again, the idea is that the change of the two point correlation between x and y induced by the external field of frequency $\omega \sim \tau^{-1}$ will be large if on that time scale, the dynamics at these two points is strongly correlated, which is true precisely if $G_4(x-y,\tau)$ is large. The extended, nonlinear FDT discussed in the next section makes this statement more precise and finally leads to our central results, Eq. (1) and Eq. (26) below. Now, recent numerical [13–16] and theoretical work[8,9,11,20,57] have focused on the above choice of four point density function. Its integral over space $\chi_4(t) = \int d^d r G_4(r,t)$ (divided by V) gives the variance of the correlation function $C(t)$ for a system of finite volume V,[58] and is therefore a good quantitative measure for dynamical heterogeneities. This quantity was unambiguously shown to display a peak at $t = \tau$, of increasing amplitude as the temperature is decreased and the glass temperature is approached,[13–16] signaling increased cooperativity in the dynamics and the growth of a length scale ℓ, which should in turn show up in the nonlinear response of the system.

2. Different scenarios for the glass transition: Qualitative predictions

We therefore expect, on very general grounds, the nonlinear response to a field that couples to degrees of freedom undergoing a collective freezing phenomenon, to increase substantially (as $\ell^{2-\bar{\eta}}$) as the glass phase is approached. However, as we discuss now, the details of this increase do depend on the specific scenario at play. Most important in that respect is the quantitative relation between the cooperative length scale ℓ and the relaxation time τ, which is often a power-law $\tau \sim \ell^z$ where z is the dynamical exponent.

One scenario for the glass state is based on the idea that some mobility defects are needed to trigger the dynamics, which slows down at low temperatures because these defects become rare.[57,59,60] Kinetically constrained models provide an interesting framework to quantify this idea. The class of so-called "East" models seems able to capture some of the phenomenology of fragile glasses and predict a temperature dependent exponent $z=z_0/T$, which may become large at low temperatures.[60,61] This is very important since the relaxation time of the system is known to increase by 15 orders of magnitude as the temperature is reduced from $1.5T_g$ to T_g. But if, say, $z=15$ the cooperative length ℓ would only increase by a very moderate factor 10.

Another well known scenario is based on the mode-coupling theory (MCT) of supercooled liquids, which predicts a dynamical singularity at a finite temperature $T_{\rm MCT}$, where the relaxation time should diverge as $\tau \sim (T-T_{\rm MCT})^{-\gamma}$,[62] with a nonuniversal exponent γ. It was recently shown that this singularity is actually accompanied by the divergence of a cooperative length ℓ, precisely defined in terms of the four-density correlation function above, Eq. (9).[9] The nature of the transition is actually equivalent to that of the mean-field p-spin glass, where both the spin-glass and the nonlinear susceptibility diverge at $T_{\rm MCT}$.[8,63] The exponent z is found to be equal to 2γ; reasonable values of z being in the range 4–6. However, the ideal MCT phase transition is "avoided" in real systems. Only the first 2–4 decades of increase of τ can be satisfactorily accounted by MCT, before some new physics come into play, that smear out the MCT transition. In the temperature region where $T<T_{\rm MCT}$, the system should, according to mean-field, be completely frozen. In finite dimensions, however, barriers to motion are finite and the dynamics is instead strongly activated. The liquid is conjectured to be a "mosaic" of local metastable glass states, that must rearrange collectively.[2,4,10] The size of these frozen clusters is the cooperative length ℓ, which increases as T decreases, but now only logarithmically with τ. Therefore, within the random first order theory (RFOT) of Ref. 4 which unifies the MCT and the mosaic scenario, one expects a crossover between a power-law increase of ℓ for $T \sim T_{\rm MCT}$ and a much more modest increase of ℓ as the temperature is reduced from $T_{\rm MCT}$ to T_g. [Below T_g, aging effects come into play and we expect that an equation similar to Eq. (2) will hold in that regime.] Finally, the "avoided critical point" scenario of Kivelson and Tarjus also predicts a cooperative length that grows weakly (logarithmically) with the relaxation time.[6]

The value of $\bar{\eta}$ in Eq. (1) above is not known either, and presumably depends both on the scenario and on the temperature regime. Negative values of $\bar{\eta} \approx -1.58$ have been reported for East models.[60] In the simplest mosaic state scenario where clusters are compact, the exponent $2-\bar{\eta}$ is equal to the dimension of space d. Rather independently of the scenario, we therefore expect a noticeable rise of the nonlinear susceptibility in supercooled liquids as the temperature is reduced: taking $\bar{\eta}=0$ and $\ell(T_g)/\ell(1.5T_g)=5$ leads to an increase of the peak value of χ_3 by a factor ≈ 25. (Note however that depending on the particular physical observable there might be other contributions coming from the temperature dependence of χ_s or, for example for the nonlinear dielectric susceptibility, from the Lorentz field effects that may play an important role in strongly dielectric systems.[64])

3. Glasses vs spin-glasses: Some caveats

The tricky aspect of the analogy between glasses and spin-glasses is that the *static* nonlinear susceptibility of glasses is in fact not expected to display any divergence. As a matter of fact, no growing correlation length has ever yet been found in any static correlation function close to the glass transition, neither in experiments nor in simulations. This is not only because the glass transition is never sharp in real glasses, but also because from a purely theoretical point of view the situation is more subtle than in spin-glasses. Within RFOT, for example, the static nonlinear susceptibility does *not* diverge. The reason is that RFOT predicts an exponentially large number of possible amorphous states and thus a nonzero configurational entropy for $T_K<T<T_{\rm MCT}$ (Refs. 7, 65, and 66) (where T_K is the "entropy crisis" temperature). Now, since equilibrium thermal averages are sums over all states, one has $\langle \delta\rho_x \delta\rho_y \rangle \equiv \Sigma_\alpha w_\alpha \langle \delta\rho_x \delta\rho_y \rangle_\alpha$, where the subscript α indicates that the average is restricted to the metastable state α, and w_α is the weight of state α. Therefore:

$$\sum_y \langle \delta\rho_x \delta\rho_y \rangle^2 \equiv \sum_{\alpha,\beta} \sum_y w_\alpha w_\beta \langle \delta\rho_x \delta\rho_y \rangle_\alpha \langle \delta\rho_x \delta\rho_y \rangle_\beta. \quad (10)$$

The divergence of the static spin-glass susceptibility is due to the diagonal terms $\alpha=\beta$. In the case of spin-glasses, the number of relevant states is effectively finite, and the above sum diverges in the spin-glass phase. Within RFOT, on the other hand, the number of relevant metastable states is so huge for $T_K<T<T_{\rm MCT}$ that the diagonal contribution, even divergent, tends to zero in the above sum, as does the off-diagonal contribution since the different frozen patterns are uncorrelated with each other. The only way to unveil any growing correlation is to focus on the terms $\alpha=\beta$, a calculation that is possible in mean-field (see previous footnote). In nonmean-field situations, this can be done in two ways: one can study (a) the static correlations but in a self-induced static pinning field (freezing all the particles outside a cavity and studying the thermodynamics inside[10]), which select a particular state α or (b) the dynamical correlations on a time scale short enough for the system to equilibrate but remain in a single state.[8] For example, the four-point correlation function defined in Eq. (9) for $t \lesssim \tau$, effectively reproduces, for τ large enough, the static sum restricted to $\alpha=\beta$,[8] since τ is the time needed to evolve from state α to any other state. More precisely, one sees that in this time regime, G_4 can be written in a form closer to the corresponding expression for spin-glasses:

$$G_4(r,t) \approx [\langle \delta\rho_x(t')\delta\rho_{x+r}(t')\rangle_\alpha \langle \delta\rho_x(t'+t)\delta\rho_{x+r}(t'+t)\rangle_\alpha]_{t'}, \quad (11)$$

where the disorder average $[\cdots]$ in the case of spin-glasses is replaced by an average over time, $[\cdots]_{t'}$, corresponding to different configurations of the self-induced disorder in glasses. Therefore, we expect that the analogy with static spin-glasses indeed makes sense, but only for $t \lesssim \tau$. A practical consequence of this observation is that the scaling function $\mathcal{H}(x)$ in Eq. (1) should tend to zero at zero frequency for glasses, at variance with the spin-glass case where it remains finite at $T=T_g^+$ and for zero field.

Even the spin-glass case turns out to be tricky since, as mentioned above, the scaling function $\mathcal{H}(x \to 0)$ in fact also goes to zero for $H \neq 0$ in the context of the full replica symmetry breaking solution.[47,48] Nevertheless, our dynamical result Eq. (26) suggests this sum rule will generically *not* hold at finite frequency whenever the four-point correlation function has a nontrivial time dependence. This statement should of course be checked explicitely for mean-field models with continuous replica symmetry breaking.[54] If indeed $\mathcal{H}(x \sim 1)$ is found to be nonzero close to the AT line, the experimental study of the dynamical nonlinear susceptibility, predicted to diverge for $\omega\tau \sim 1$, would offer a direct way to prove or disprove the existence of an AT line in real systems (see Ref. 67 for a recent discussion).

III. THEORETICAL ANALYSIS

In the rest of this paper, we give some theoretical justifications of our central result, Eq. (1). These arguments also suggest that in the nonequilibrium phase, Eq. (2) will hold. We will use the Langevin equation formalism for continuous spins,[52] but our result are expected to hold more generally (for example, if the continuous spins are replaced by interacting particles with Newtonian dynamics).

A. Linear response

We assume that the equation of motion of spin s_i is given by

$$\partial_t s_i = - \partial_{s_i} H + \eta_i(t), \tag{12}$$

where H is the Hamiltonian of the system, which we do not specify explicitly. In the case of spin glasses it contains quenched disorder and possibly one body terms ensuring an Ising-type character to the spins s_i. The coupling to an external, site dependent field $h_i(t)$, amounts to add to H the sum over spins of $h_i(t)s_i$. The Gaussian noise η_i is as usual of zero mean, white in time and decorrelated from spin to spin,

$$\langle \eta_i(t_1)\, \eta_j(t_2) \rangle = 2k_B T \delta(t_1 - t_2)\delta_{i,j}. \tag{13}$$

Since the noise is Gaussian, one can establish the following identity:

$$\langle s_i(t_1)\, \eta_j(t_2) \rangle = k_B T \left\langle \frac{\partial s_i(t_1)}{\partial h_j(t_2)} \right\rangle. \tag{14}$$

Let us first quickly re-establish the standard linear FDT. From the above equation and the equation of motion, the response of a spin to an earlier field is

$$\chi_{ij}(t_1,t_2) = \left\langle \frac{\partial s_i(t_1)}{\partial h_j(t_2)} \right\rangle = \frac{1}{k_B T} \langle s_i(t_1)[\partial_{t_2} s_j + \partial_{s_j} H(t_2)] \rangle. \tag{15}$$

The averaging above assumes the system to be in equilibrium: we average over all histories with initial conditions appearing with the equilibrium Boltzmann weight. The second term in the right-hand side is zero since, for an arbitrary observable $O(\{t_a\})$ that depends on times t_a, all posterior to t_2, one has

$$\langle O(\{t_i\}) \partial_{s_j} H(t_2) \rangle \equiv - k_B T \int \prod_a ds(t_a) P[\{s(t_a)\}|s_2] O(\{s(t_a)\}) \times ds_2 \partial_{s_2} \exp[-H(s_2)/k_B T] = 0, \tag{16}$$

where $s_2 = s(t_2)$ and the last equality holds because the last term is a total derivative. Therefore, one finds the usual FDT relation,

$$\chi_{ij}(t_1,t_2) = \frac{1}{k_B T} \frac{d}{dt_2} \langle s_i(t_1) s_j(t_2) \rangle. \tag{17}$$

Integrating this quantity over t_2 with a constant field $h_i(t_2) = h$ gives the static susceptibility χ_s, which, as is well known, is found to be the integral over space of the two-body correlation function. In the case of a static critical point where the correlation length ξ diverges, one would have $\chi_s \sim \mu^2 \xi^{2-\eta}/k_B T$, where η is the standard critical exponent of the static transition and μ the elementary magnetic moment. However, in the case of glassy systems, the two point function is not critical and one rather expects $\chi_s \sim \mu^2 \xi^d / k_B T$ where ξ remains microscopic and does not grow appreciably lowering the temperature (or increasing the density). As emphasized in Sec. II, one should in the case of amorphous systems rather focus on nonlinear effects to observe some nontrivial behavior.

B. Nonlinear response: The static limit

As a consequence we want to extend the above calculation to the response at time $t_1 > t_2$ to three field "kicks" at times $t_2 > t_3 > t_4$. This is given by

$$\chi_{3,ijkl}(t_1,t_2,t_3,t_4) = \left\langle \frac{\partial^3 s_i(t_1)}{\partial h_j(t_2) \partial h_k(t_3) \partial h_l(t_4)} \right\rangle = (k_B T)^{-3} \langle s_i(t_1)\, \eta_j(t_2)\, \eta_k(t_3)\, \eta_l(t_4) \rangle. \tag{18}$$

Using three times the Langevin equation of motion, and once the above trick to get rid of the final $\partial_{s_l} H(t_4)$, we find the following general relation, involving four terms:

$$\begin{aligned}(k_B T)^3 \chi_{3,ijkl}(t_1,t_2,t_3,t_4) = {} & \frac{d^3}{dt_2 dt_3 dt_4} \langle s_i(t_1) s_j(t_2) s_k(t_3) s_l(t_4) \rangle \\ & + \frac{d^2}{dt_3 dt_4} \langle s_i(t_1) \partial_{s_j} H(t_2) s_k(t_3) s_l(t_4) \rangle \\ & + \frac{d^2}{dt_2 dt_4} \langle s_i(t_1) s_j(t_2) \partial_{s_k} H(t_3) s_l(t_4) \rangle \\ & + \frac{d}{dt_4} \langle s_i(t_1) \partial_{s_j} H(t_2) \partial_{s_k} H(t_3) s_l(t_4) \rangle.\end{aligned} \tag{19}$$

Let us first analyze the static limit of this expression. From the above result, one can show in full generality that the static nonlinear susceptibility $\chi_{3s} = \chi_3(\omega = 0, T)$, obtained by integrating over all $t_2 > t_3 > t_4$ with a constant field $h_i(t) = h$ on all sites, is given by

$$(k_BT)^3\chi_{3s} = \frac{1}{N}\sum_{ijkl}\langle s_i(t_1)s_j(t_1)s_k(t_1)s_l(t_1)\rangle_c, \tag{20}$$

where the subscript c means that one takes the connected part of the correlation and N the total number of sites. This result is exact and can be obtained directly using equilibrium statistical mechanics. In the present context, only the first term in expression Eq. (19) for $\chi_{3,ijkl}(t_1,t_2,t_3,t_4)$ contributes to $\omega=0$. As discussed in Sec. II, the long range order setting in spin-glasses is unveiled not by the two-body correlation that oscillates in sign and averages to zero, but by the square of this two-body correlation. Therefore, the leading dominant term in the above sum corresponds to the square of the two-body correlation obtained pairing i, j, k, l, say, i with j and k with l within the two-body correlation length ξ (which typically remains small at all temperatures):

$$(k_BT)^3\chi_{3s} \approx -\frac{3}{N}\xi^{2d}\sum_{ik} G_{ik} \quad G_{ik} = \langle s_i(t_1)s_k(t_1)\rangle_c^2. \tag{21}$$

If one now assumes that G_{ik} scales as in usual critical phenomena[22,47]

$$G_{ik} = \frac{1}{|r_i - r_k|^{d-2+\bar{\eta}}}\mathcal{G}\left(\frac{|r_i - r_k|}{\ell}\right), \tag{22}$$

then the sum over i, k behaves as $N\ell^{2-\bar{\eta}}$, finally leading to a static nonlinear susceptibility given by

$$\chi_{3s} \approx \frac{C\mu^4\xi^{2d}}{(k_BT)^3}\ell^{2-\bar{\eta}} \sim \frac{\chi_s^2}{k_BT}\ell^{2-\bar{\eta}}, \tag{23}$$

where C is a constant, and ℓ is counted in units of the static correlation length ξ. This is the zero frequency result given in Eq. (1).

C. Nonlinear response: Dynamics

1. Some general arguments

The extension to nonzero frequency of the above result can proceed in different ways. Our result Eq. (1) can be simply seen as a standard dynamical scaling assumption close to a critical point, as is indeed correct for spin-glasses.[43,44] This result is expected to hold whenever a critical point is responsible for the simultaneous increase of the relaxation time and the cooperative length. This is true of the mode-coupling theory of glasses,[8,9] and also of other scenarios discussed in the introduction and in Sec. II, which rely on the existence of an underlying critical point.[4–7,10,11] From a more technical point of view we want to justify that the behavior of the nonlinear cubic response is the same as of the first term on the right-hand side of Eq. (19), whereas the three other terms are either negligible or of the same order of magnitude (on frequencies of the order of τ^{-1}), but not more divergent.

A simple case that can be treated in some generality is when the fluctuation of the norm of the spins can be neglected, for example for Ising spins that can be recovered from the Langevin equation in the limit of infinitely sharp double well potential that is zero if $s^2=1$ and infinite otherwise. After several integration by parts and using $s^2=1$, one can show that the three last terms of Eq. (19) do not contribute to the nonlinear ac susceptibility at low frequencies (much smaller than the microscopic, high frequency scale of the model). One is therefore left with the first term of Eq. (19), that contains three derivatives with respect to time. If one assumes that the four-body correlation $\langle s_i(t_1)s_i(t_2)s_k(t_3)s_k(t_4)\rangle$ is, for $|i-k|\sim\ell$, only a function of $(t_1-t_2)/\tau$, $(t_2-t_3)/\tau$ and $(t_3-t_4)/\tau$, the integration over t_2, t_3, t_4 with an oscillating field at frequency ω and over space directly leads to Eq. (1), i.e., a nonlinear susceptibility that scales as a certain function $\mathcal{H}$ of $\omega\tau$. This result is only justified in the low frequency domain; for high frequency, contributions from the short-time β-regime will obviously come into play. Note that very generally, we expect $\mathcal{H}$ to be nontrivial, although it does vanish at zero frequency whenever the static susceptibility is finite, as is the case for glasses and spin-glasses in an external field (see the discussion in Secs. II A 2 and II B 3).

More generally, one can argue both physically and diagrammatically that the three last terms of Eq. (19) give contributions which are at most of the same order of magnitude as the first one. From a physical point of view, these terms contain less time derivatives that the first, but also contain the local "force" acting on the configuration, $\partial_s H(t)$. Since we are interested in the low frequency response of the system, we can decompose the dynamics of the spins into a fast part s^f and a slow part s^*, that corresponds to the dynamics on a time of order τ. It is clear that the force acting on the slow modes can only lead to a slow dynamics of these modes, i.e., $|\partial_{s^*}H| \lesssim \tau^{-1}$. Therefore, for frequencies $\sim\tau^{-1}$, one has, for example,

$$\langle s_i(t_1)\partial_{s_j}H(t_2)s_k(t_3)s_l(t_4)\rangle \lesssim \tau^{-1}F\left(\frac{t_1-t_2}{\tau},\frac{t_2-t_3}{\tau},\frac{t_3-t_4}{\tau}\right), \tag{24}$$

(where F is a certain function), which after integration leads again to a result of the form (1).

2. Diagrammatic analysis

One can understand the above result from a different point of view using diagrams for a general Langevin equation, which leads to a dynamical field theory with the spin field s and the response field $\hat{s}$.[68] The nonlinear cubic response χ_3 at time t_1 to three instantaneous fields at times t_2,t_3,t_4 can be written in full generality as (see Fig. 1)

$$\chi_3(t_1;t_2,t_3,t_4) = \int dt_1'dt_2'dt_3'dt_4'\Gamma_{\hat{s},s,s,s}(t_1';t_2',t_3',t_4')\chi(t_1-t_1')\chi(t_2' - t_2)\chi(t_3'-t_3)\chi(t_4-t_4'), \tag{25}$$

where $\Gamma_{\hat{s},s,s,s}$ is the amputated vertex with legs $\hat{s}$, s, s, s (for simplicity we skip here the space indices). Note that the vertex $\Gamma_{s,s,s,s}$ is zero by causality because it contains for sure a closed loop of response functions. The other vertices do not appear because the correlation function $\langle\hat{s}\hat{s}\rangle$ vanishes by causality.

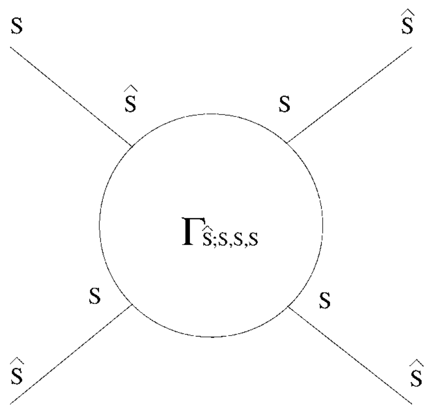

FIG. 1. General diagrammatic representation of the nonlinear cubic response.

Now let us consider the diagrams contributing to the connected four body correlation function. There is a first contribution G_4^a obtained by plugging three two body correlation functions into $\Gamma_{\hat{s},s,s,s}$, see Fig. 2. It is straightforward to check, using FDT, that this first series of diagrams, G_4^a, is directly related to the nonlinear cubic response. If G_4^a was the only contribution, then one would find an extended FDT where only the first term on the right-hand side of Eq. (19) contributes.

There is another contribution, G_4^b, that corresponds to constructing ladders with the irreducible vertices $\Gamma^{\text{irr}}_{s,s;\hat{s},\hat{s}}$, $\Gamma^{\text{irr}}_{s,s;s,\hat{s}}$, $\Gamma^{\text{irr}}_{\hat{s},\hat{s};s,\hat{s}}$, $\Gamma^{\text{irr}}_{\hat{s},\hat{s};\hat{s},\hat{s}}$, $\Gamma^{\text{irr}}_{\hat{s},\hat{s};s,s}$ on the left of $\Gamma_{\hat{s},s,s,s}$ and the irreducible vertices $\Gamma^{\text{irr}}_{s,s;\hat{s},\hat{s}}$, $\Gamma^{\text{irr}}_{s,\hat{s};\hat{s},\hat{s}}$, $\Gamma^{\text{irr}}_{\hat{s},\hat{s};\hat{s},\hat{s}}$, $\Gamma^{\text{irr}}_{\hat{s},\hat{s};s,\hat{s}}$, $\Gamma^{\text{irr}}_{\hat{s},s;s,\hat{s}}$, $\Gamma^{\text{irr}}_{s,s;s,\hat{s}}$, on the right of $\Gamma_{\hat{s},s,s,s}$ (see Fig. 3). [We recall that the irreducible vertex $\Gamma^{\text{irr}}_{1,2;3,4}$ is the sum of all Feynman diagrams contributing to $\Gamma_{1,2;3,4}$ (the amputated vertex) that has the property that cutting two internal lines does not separate the diagram into two disconnected parts, such that one part contains the lines 1,2 and the other one the lines 3,4.] Finally, the last contribution, G_4^c, is formed by plugging together the same irreducible diagrams used in G_4^b but without making use of $\Gamma_{\hat{s},s,s,s}$.

In the case of the critical equilibrium dynamics of spin-glasses, Eq. (1) is already known[43,44] and could have been guessed *a priori* from the general scaling properties of second order phase transitions with a single diverging length (and time) scale. As a consequence in this case G_4^b, G_4^c are of the same order or less divergent than G_4^a. The case of structural glasses is *a priori* more tricky, since there is no consensus on the effective critical microscopic model that would describe them. However, if we take as an established fact (at least numerically) that the four body correlation is governed by a length scale that increases as the glass is approached, then this effect has to be contained in (at least) one of the three contributions G_4^a, G_4^b, G_4^c. Now, the nonlinear response certainly contains the contribution related to G_4^a; therefore both χ_3 and G_4 grow (or even diverge) similarly unless another family of (more) diverging diagrams (the ones contributing to G_4^b, G_4^c) can be constructed. We believe that this is a rather unlikely scenario and instead expect that in general $\Gamma_{\hat{s},s,s,s}$ contains the leading divergence. As a consequence G_4^a and G_4^b, and therefore χ_3 and G_4, are of the same order of magnitude whereas G_4^c is subdominant. In this case Eq. (1) holds. Strictly speaking, these arguments only prove that if the nonlinear cubic dynamical response diverges, a similar (or stronger) divergence is expected for the four body correlation function, but not vice versa. Therefore it would be important to check our prediction for specific models of the glass transition, in the spirit of Ref. 20. Although the above general arguments are clearly incomplete, we want to emphasize here they are indeed correct within the mode-coupling theory of the glass transition. This can be seen using the ideas of Ref. 10 which establish the validity of our central result, Eq. (1).

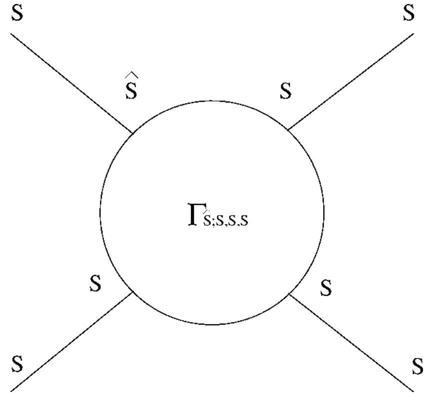

FIG. 2. Diagrammatic representation of G_4^a.

Finally, let us remark that the extension to the nonequilibrium case can be tackled in a similar way. In particular, since the four-body correlation function diverges with t_w (Ref. 69) in spin-glasses, and the classification in terms of G_4^a, G_4^b, G_4^c carries over to the nonequilibrium case, the above discussion can be generalized to the nonequilibrium case as well.

In summary, we have shown in this section that for glassy systems close to a critical point, where the relaxation time and cooperative length diverge, an extended approximate FDT relates the nonlinear susceptibility to the four-point correlation function in the low frequency domain,

$$(k_BT)^3\chi_{3,ijkl}(t_1,t_2,t_3,t_4) \sim \frac{d^3}{dt_2dt_3dt_4}\langle s_i(t_1)s_j(t_2)s_k(t_3)s_l(t_4)\rangle, \tag{26}$$

where ~ means that right and left-hand side have the same critical behavior. The additional terms missing in the above equation are either of the same order of magnitude, or negligible.

IV. CONCLUSION

In conclusion, we have shown in this paper that if the abrupt slowing down of glassy materials is accompanied by the growth of a cooperative length ℓ, then the nonlinear, 3ω response to an oscillating field (at frequency ω) should substantially increase and give precious information on the temperature (or density) dependence of ℓ. The theoretical moti-

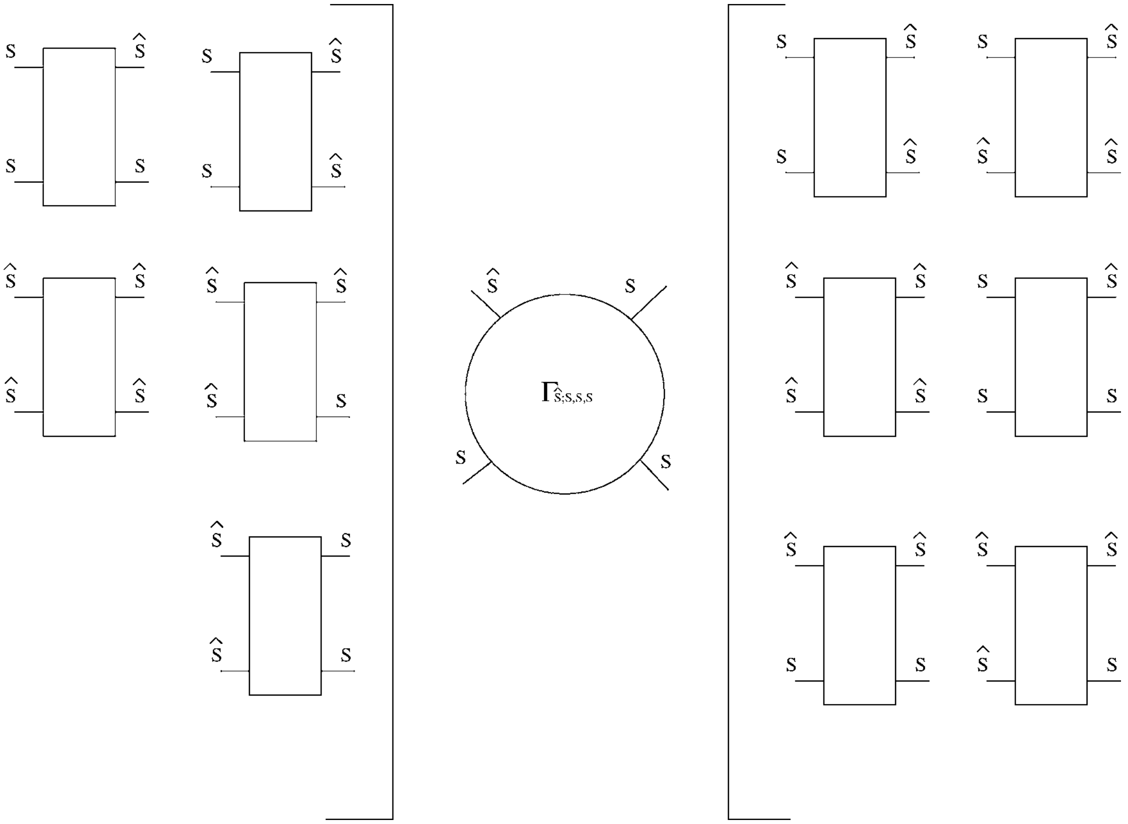

FIG. 3. Diagrammatic representation of G_4^b.

vation is that the nonlinear susceptibility is approximatively related, for glassy systems close to a critical point, to the four-point correlation function that captures dynamical cooperativity. This relation is certainly correct within the context of the mode-coupling theory of glasses, but should hold in other cases as well.

In supercooled liquids, the analysis of the nonlinear compressibility (sound wave harmonics) should allow one to probe directly the existence of a growing cooperative length. This should also be true of the nonlinear dielectric susceptibility, at least in systems where the dipoles are strongly coupled to the glassy degrees of freedom. Although early experiments seemed to show no interesting effects,[45] we believe that more systematic studies should be performed,[64] especially now that numerical simulations have unambiguously shown the growth of a cooperative length in the four-point function.[11,13–16] These experiments should also allow one to bridge the gap between the length-scales observed on simulation time scales and the length-scales observed experimentally on much larger time-scales close to the glass transition temperature.[12] The study of nonlinear specific heat effects, although more complex, may be interesting too.[70,71] From a more general point of view any nonlinear dynamical response (for example, nonlinear rheology in soft glassy materials) should be worth studying if the corresponding linear response can be used as a probe of slow dynamics.

In spin-glasses, nonlinear ac magnetic susceptibility measurements in nonzero field could shed light on the existence of a de Almeida-Thouless line. In the aging phase, such measurements should allow one to test in more details the length scale ideas put forward in Refs. 22–27. Compared to the case of glasses, the experimental situation is particularly encouraging since the nonlinear susceptibility is already known to diverge at the spin-glass transition. There should be a clear trace of this divergence in the aging phase, except if some unlikely cancellation occurs at nonzero frequency (such a cancellation indeed operates in the static limit for spin-glasses in mean-field, but not in finite dimensions). The effect of temperature cycling on the nonlinear susceptibility should then give direct indications of the mechanisms of rejuvenation and memory.[25,26] We therefore hope that the ideas expressed in this paper will help shed light on the issue of dynamical heterogeneity and cooperativity in disordered, amorphous systems.

ACKNOWLEDGMENTS

We thank L. Berthier, A. Billoire, A. Bray, C. De Dominicis, V. Dupuis, M. Mézard, M. Moore, E. Vincent, and M. Wyart, for discussions. We are grateful to F. Ladieu and D. L'Hote for important comments, in particular on Lorentz field effects, and for the interest and enthusiasm with which they started experiments along the lines of this study. We also thank P. Young for a crucial remark on the case of a spin-glass in a field, and L. Borland for carefully reading the manuscript. G. B. is partially supported by the European Community's Human Potential Programme Contract No. HPRN-CT-2002-00307 (DYGLAGEMEM).

[*] Electronic address: bouchau@spec.saclay.cea.fr

[†] Electronic address: biroli@cea.fr

[1] See, e.g., P. G. Debenedetti and F. H. Stillinger, Nature (London) **410**, 259 (2001).

[2] G. Adam and J. H. Gibbs, J. Chem. Phys. **43**, 139 (1965).

[3] P. G. Debenedetti, *Metastable Liquids, Concepts and Principles* (Princeton University Press, Princeton, 1996).

[4] For a review of recent models, see F. Ritort and P. Sollich, Adv. Phys. **52**, 219 (2003).

[5] T. R. Kirkpatrick and P. G. Wolynes, Phys. Rev. B **36**, 8552 (1987); T. R. Kirkpatrick, D. Thirumalai, and P. G. Wolynes, Phys. Rev. A **40**, 1045 (1989).

[6] J. P. Sethna, J. D. Shore, and M. Huang, Phys. Rev. B **44**, 4943 (1991); **47**, 14661 (1993).

[7] G. Tarjus and D. Kivelson, J. Chem. Phys. **103**, 3071 (1995); D. Kivelson and G. Tarjus, Philos. Mag. B **77**, 245 (1998).

[8] M. Mézard and G. Parisi, J. Chem. Phys. **111**, 1076 (1999); J. Phys.: Condens. Matter **12**, 6655 (2000); M. Mézard, *First Steps in Glass Theory*, in *More is Different*, edited by Ong and Bhatt (Princeton University Press, Princeton, 2002).

[9] S. Franz and G. Parisi, J. Phys.: Condens. Matter **12**, 6335 (2000); C. Donati, S. Franz, G. Parisi, and S. C. Glotzer, J. Non-Cryst. Solids **307**, 215 (2002).

[10] G. Biroli and J. P. Bouchaud, Europhys. Lett. **67**, 21 (2004).

[11] J. P. Bouchaud and G. Biroli, J. Chem. Phys. **121**, 7347 (2004).

[12] S. Whitelam, L. Berthier, and J. P. Garrahan, Phys. Rev. Lett. **92**, 185705 (2004).

[13] M. M. Hurley and P. Harrowell, Phys. Rev. E **52**, 1694 (1995), and references therein.

[14] For an enlightening review, see M. D. Ediger, Annu. Rev. Phys. Chem. **51**, 99 (2000).

[15] R. Yamamoto and A. Onuki, Phys. Rev. Lett. **81**, 4915 (1998) and references therein.

[16] C. Bennemann, C. Donati, J. Baschnagel, and S. C. Glotzer, Nature (London) **399**, 246 (1999); S. C. Glotzer, V. V. Novikov, and T. B. Schroeder, J. Chem. Phys. **112**, 509 (2000); N. Lacevic, F. W. Starr, T. B. Schroeder, and S. C. Glotzer, *ibid.* **119**, 7372 (2003), and references therein.

[17] L. Berthier, Phys. Rev. E **69**, 020201(R) (2004).

[18] E. R. Weeks, J. C. Crocker, A. C. Levitt, A. Schofield, and D. A. Weitz, Science **287**, 627 (2000).

[19] G. Marty and O. Dauchot, Phys. Rev. Lett. **94**, 015701 (2005).

[20] L. Silbert, A. Liu, and S. Nagel, cond-mat/0501616 (unpublished), and references therein; see also M. Wyart, S. Nagel, and T. Witten, cond-mat/0409687 (unpublished).

[21] C. Toninelli, M. Wyart, L. Berthier, G. Biroli, and J.-P. Bouchaud, Phys. Rev. E **71**, 041505 (2005).

[22] H. Silescu, J. Non-Cryst. Solids **243**, 81 (1999).

[23] D. S. Fisher and D. A. Huse, Phys. Rev. B **38**, 373 (1988).

[24] A. J. Bray and M. A. Moore, Phys. Rev. Lett. **58**, 57 (1987).

[25] G. J. M. Koper and H. J. Hilhorst, J. Phys. (France) **49**, 429 (1988).

[26] J. P. Bouchaud, in *Soft and Fragile Matter*, edited by M. E. Cates and M. Evans Edts (IOP, New York, 2000); J. P. Bouchaud, V. Dupuis, J. Hammann, and E. Vincent, Phys. Rev. B **65**, 024439 (2002), see also Ref. 35.

[27] P. E. Jonsson, R. Mathieu, P. Nordblad, H. Yoshino, H. A. Katori, and H. Ito, Phys. Rev. B **70**, 174402 (2004).

[28] L. Berthier and P. C. W. Holdsworth, Europhys. Lett. **58**, 39 (2002).

[29] H. Rieger, *Ann. Rev. of Comp. Phys. II*, edited by D. Stauffer (World Scientific, Singapore, 1995).

[30] T. Komori, H. Yoshino, and H. Takayama, J. Phys. Soc. Jpn. **68**, 3387 (1999); **69**, 1192 (2000); **69**, 228 (2000).

[31] E. Marinari, G. Parisi, J. Ruiz-Lorenzo, and F. Ritort, Phys. Rev. Lett. **76**, 843 (1996).

[32] L. Berthier and J.-P. Bouchaud, Phys. Rev. B **66**, 054404 (2002).

[33] L. Berthier and A. P. Young, Phys. Rev. B **69**, 184423 (2004); **71**, 214429 (2005).

[34] Y. G. Joh, R. Orbach, G. G. Wood, J. Hammann, and E. Vincent, Phys. Rev. Lett. **82**, 438 (1999).

[35] P. E. Jonsson, H. Yoshino, P. Nordblad, H. Aruga Katori, and A. Ito, Phys. Rev. Lett. **88**, 257204 (2002).

[36] F. Bert, V. Dupuis, E. Vincent, J. Hammann, and J. P. Bouchaud, Phys. Rev. Lett. **92**, 167203 (2004).

[37] E. Vincent, J. Hammann, M. Ocio, J.-P. Bouchaud, and L. F. Cugliandolo, in *Proceeding of the Sitges Conference on Glassy Systems*, edited by E. Rubi (Springer, Berlin, 1996) (eprint cond-mat/9607224).

[38] J.-P. Bouchaud, L. Cugliandolo, J. Kurchan, and M. Mézard, in *Spin-Glasses and Random Fields*, edited by A. P. Young (World Scientific, Singapore, 1998), and references therein; see also Ref. 39.

[39] See R. Monasson, Phys. Rev. Lett. **75**, 2847 (1995) for the introduction of the concept of random pinning field in the context of structural glasses.

[40] Y. Singh, J. P. Stoessel, and P. G. Wolynes, Phys. Rev. Lett. **54**, 1059 (1985).

[41] S. Franz, M. Cardenas, and G. Parisi, J. Phys. A **31**, L163 (1998).

[42] L. Cugliandolo, in *Slow Relaxations and Nonequilibrium Dynamics in Condensed Matter*, Les Houches, Session LXXVII, edited by J. L. Barrat, M. Feigelman, J. Kurchan, and J. Dalibard (Springer-EDP Sciences, New York, 2003).

[43] See e.g., K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986).

[44] E. Vincent and J. Hammann, J. Phys. C **20**, 2659 (1987).

[45] M. Maglione, U. T. Höchli, and J. Joffrin, Phys. Rev. Lett. **57**, 436 (1986).

[46] A. Zippelius, Phys. Rev. B **29**, 2717 (1994).

[47] G. Parisi and P. Ranieri, J. Phys. A **30**, L415 (1997).

[48] L. Wu, Phys. Rev. B **43**, 9906 (1991).

[49] H. M. Carruzzo and C. C. Yu, Phys. Rev. E **66**, 021204 (2002).

[50] C. De Dominicis, I. Giardina, E. Marinari, O. C. Martin, and F. Zuliani, cond-mat/0408088 (unpublished).

[51] A. Crisanti, T. Rizzo, and T. Temesvari, cond-mat/0302538 (unpublished).

[52] C. de Dominicis has informed us that the results of Ref. 47 have been revised: the claim that the four point correlation function that appears in nonlinear susceptibility is massive does not hold in finite dimensions, although sufficiently large length scales must be probed to see the difference with mean-field.

[53] A. J. Bray and M. A. Moore, J. Phys. C **17**, L613 (1984).

[54] We are grateful to A. P. Young for a detailed explanation of this point.

[55] G. Semerjian, L. Cugliandolo, and A. Montanari, J. Stat. Phys. **115**, 493 (2004).

[56] C. Chamon, M. P. Kennett, H. E. Castillo, and L. F. Cugliandolo, Phys. Rev. Lett. **89**, 217201 (2002); H. E. Castillo, C. Chamon, L. F. Cugliandolo, J. L. Iguain, and M. P. Kennett, Phys. Rev. B **68**, 134442 (2003).

[57] A. Andreanov, G. Biroli, and J. P. Bouchaud (unpublished).

[58] L. Berthier, Phys. Rev. Lett. **91**, 055701 (2003).

[59] J. P. Garrahan and D. Chandler, Phys. Rev. Lett. **89**, 035704 (2002); J. P. Garrahan and D. Chandler, Proc. Natl. Acad. Sci. U.S.A. **100**, 9710 (2003).

[60] L. Berthier and J. P. Garrahan, Phys. Rev. E **68**, 041201 (2003); J. Phys. Chem. B **109**, 3578 (2005).

[61] P. Sollich and M. R. Evans, Phys. Rev. Lett. **83**, 3238 (1999).

[62] W. Götze and L. Sjögren, Rep. Prog. Phys. **55**, 241 (1992); W. Götze, Condens. Matter Phys. **1**, 873 (1998); W. Kob, in *Les Houches, Session LXXVII* (Springer-EDP Sciences, Berlin, 2003).

[63] A. Crisanti and H.-J. Sommers, Z. Phys. B: Condens. Matter **87**, 341 (1992).

[64] F. Ladieu and D. L'Hote (unpublished).

[65] A. P. Young and Helmut G. Katzgraber, Phys. Rev. Lett. **93**, 207203 (2004), and references therein.

[66] See, e.g., J. Cardy, *Scaling and Renormalization in Statistical Physics* (Cambridge University Press, Cambridge, 1996).

[67] N. O. Birge and S. R. Nagel, Phys. Rev. Lett. **54**, 2674 (1985).

[68] C. Schick, M. Merzlyakov, and A. Hensel, J. Chem. Phys. **111**, 2695 (1999).

[69] Note that the ratio of nonlinear effects to linear effects is, in order of magnitude, given by $\chi_s h^2/k_B T$, i.e., the ratio of the energy of the field to the thermal energy.

[70] Within a field theoretical perspective (Ref. 69) the growing correlation length ℓ_w is due to soft-modes, related to the reparametrization invariance of the dynamical action.

[71] For practical reasons one has to introduce an overlap function in the definition of G_4 or focus on slightly different observables (Refs. 15 and 16).

[72] To derive this result, one should compute χ_3 within one of the states that dominates the partition function at temperature $T < T_{\mathrm{MCT}}$. It is easy to check, using the results of Ref. 72, that the Gaussian fluctuations having a vanishing mass at T_{MCT} give the same contribution to both the spin-glass and the nonlinear susceptibility. Hence, both diverge as $1/\sqrt{T_{\mathrm{MCT}}-T}$.