Searching for the Gardner Transition in Glassy Glycerol

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We search for a Gardner transition in glassy glycerol, a standard molecular glass, measuring the third harmonics cubic susceptibility $\chi_3^{(3)}$ from slightly below the usual glass transition temperature down to 10 K. According to the mean-field picture, if local motion within the glass were becoming highly correlated due to the emergence of a Gardner phase then $\chi_3^{(3)}$, which is analogous to the dynamical spin-glass susceptibility, should increase and diverge at the Gardner transition temperature T_G. We find instead that upon cooling $|\chi_3^{(3)}|$ decreases by several orders of magnitude and becomes roughly constant in the regime $100-10$ K. We rationalize our findings by assuming that the low temperature physics is described by localized excitations weakly interacting via a spin-glass dipolar pairwise interaction in a random magnetic field. Our quantitative estimations show that the spin-glass interaction is twenty to fifty times smaller than the local random field contribution, thus rationalizing the absence of the spin-glass Gardner phase. This hints at the fact that a Gardner phase may be suppressed in standard molecular glasses, but it also suggests ways to favor its existence in other amorphous solids and by changing the preparation protocol.

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At low temperatures, glasses display a set of anomalies compared to their crystalline counterparts. For instance, the specific heat and thermal conductivity violate the Debye law and the vibrational properties are different from the ones predicted by the Debye theory of phononic excitations [1,2]. These concomitant phenomena have been investigated extensively both at the theoretical and experimental level starting from the 1970s [3,4]. The central physical question underpinning this field of research is the nature of the excitations that govern the low temperature physics of amorphous solids. One of the main proposals is that those are associated with disordered independent two-level systems (TLS) [3,4]. Although the TLS theory allows us to explain many experimental results, some puzzles remain unsolved [5], and theoreticians still wonder about the possible collective nature of the low-energy excitations [5,6]. The recent solution of simple structural glass models obtained in the limit of infinite spatial dimensions [7,8] has introduced a new possibility in this research effort: amorphous solids may undergo upon compression or cooling a new kind of phase transition, called Gardner transition, that changes their nature, in particular their low temperature properties.

Let us first recall the main results of the infinite dimensional solution that are relevant for the problem we focus on. Within this approach an amorphous solid is described in terms of a metabasin of configurations in which the liquid remains trapped at the glass transition. Since within the mean-field theory (realized in the infinite dimensional limit) barriers between metabasins are divergent, amorphous solids correspond to separate ergodic components that can therefore be studied using a thermodynamical formalism [9]. The main result found in studying infinite dimensional hard and harmonic spheres [7,10] is that these systems undergo a Gardner phase transition when lowering the temperature or increasing the pressure: below the critical temperature or above a critical pressure the metabasin associated with the solid formed at the glass transition breaks down in a multitude of glassy states organized in a hierarchical fashion [11–13]. This hierarchy is of the very same nature as that found in the spin-glass state in certain mean-field spin-glass models [14]. This Gardner phase brings about soft modes [15], diverging susceptibilities, and collective excitations [16,17], and therefore is said to be marginal. Remarkably, it plays a central role in the quantitative understanding of the critical properties of three dimensional packings of spheres at jamming [18]. It is therefore also a possible candidate to explain the anomalous low temperature properties of amorphous solids.

Whether or not a Gardner transition takes place for generic model systems is a question that has been investigated in the past few years. Already at the mean-field level it has been shown that the emergence of a Gardner phase may depend on the model (interaction potential) and on the cooling procedure; proximity to jamming favors its existence [10,19] while for some interaction potentials, well annealed glasses do not undergo a Gardner transition upon cooling [20,21]. Therefore, the emergence of Gardner physics, even at the mean-field level, is not generic and may depend on the physical context, interaction potential, and preparation details. Similar results have also been found in simulations, where evidence of the Gardner transition has been found mainly in hard sphere systems [16,22–24]. On the experimental side, favorable but somewhat indirect evidence has been reported in granular glasses [25], colloidal glasses [26], and in two molecular glasses exhibiting a strong Johari-Goldstein β peak [27]. From the theoretical point of view, going beyond the realm of mean-field theory and including finite dimensional fluctuations is very challenging: the Gardner transition is alike to the spin-glass transition in a field [23,28], for which renormalization group results are not conclusive on the possibility of having a transition in three dimensions [28–31].

All in all, whether standard molecular glasses display a Gardner phase, or at least some signature of it, remains an open question. The aim of this Letter is to address this issue by combining experiments and theory. At variance with previous experiments [25–27], we measure the low temperature behavior of the third harmonic susceptibility of glassy glycerol, which is a *direct* smoking gun of the transition and is expected to diverge in correspondence of the Gardner point (see below). We do not find any hint of such behavior, therefore excluding the possibility of a transition, at least down to 10 K. In parallel, from a theoretical point of view, we rationalize our findings using a phenomenological approach: at variance with previous theoretical approaches that investigated numerically the Gardner transition in finite dimension [20,21], we build up a phenomenological model of the transition itself and we show that within the assumptions considered in this framework, we cannot expect a Gardner transition in standard fragile molecular glasses, at least in typical experimental conditions.

We start by presenting the results of the experiments on third harmonic cubic susceptibility $\chi_3^{(3)}$ in glassy glycerol from 180 K $\simeq T_g - 8$ K, T_g being the usual glass transition temperature, down to 10 K. At low temperature local excitations have a dipolar moment, $\chi_3^{(3)}$ at fixed angular frequency ω is expected to probe spin-glass order [32] and therefore to diverge upon cooling if there is a Gardner transition [33]. Indeed $\chi_3^{(3)}$ is the dielectric equivalent of the dynamical spin-glass susceptibility. More precisely, dynamical critical theory leads to [34–36]

$$\chi_3^{(3)}(\omega) = \left(\frac{T_G}{|T - T_G|}\right)^{\nu(2d_f - d)} g\left[\frac{\omega}{\omega_0}\left(\frac{T_G}{|T - T_G|}\right)^{z\nu}\right], \qquad (1)$$

where ω_0 is the microscopic frequency, ν and z are the critical exponents related to the correlation length and to the relaxation time, respectively, g is a scaling function, and d_f is the fractal dimension of correlated regions (d is the spatial dimension). Using dynamical scaling, one finds that approaching T_G the third harmonics cubic susceptibility $\chi_3^{(3)}$ should increase when probed at a fixed frequency and it should reach a maximal value of $\chi_3^{(3)}(\omega) \sim (\omega_0/\omega)^{(2d_f - d)/z}$ at $T = T_G$.

Henceforth, we shall report $\delta\chi_3^{(3)}(T) \equiv \chi_3^{(3)}(T) - \chi_3^{(3)}(30\text{ K})$. The reason for this substraction is that at low temperatures the value of $|\chi_3^{(3)}|$ is typically 10^4 times smaller than around the glass transition temperature, i.e., it is so small that the residual spurious third harmonics $V_{\text{source}}^{(3)}$ of the voltage source competes with the third harmonics signal of the glycerol sample. Using the fact that the spurious third harmonics does not depend on T, we can cancel it out by subtracting the value at the reference temperature $T = 30$ K. In the Supplemental Material [37] we present more details and tests that show the efficiency of our experimental procedure.

In Fig. 1 we show the behavior of $\delta\chi_3^{(3)}$ for a frequency 9.878 Hz as a function of T—note that $|\chi_3^{(3)}(30\text{ K})| = (1.0 \pm 0.5) \times 10^{-19}\ \text{m}^2/\text{V}^2$. Our results show a decrease from 180 to 100 K. Close to the glass transition temperature T_g, $|\chi_3^{(3)}|$ probes correlated particle motion [35,38].

The decrease below T_g is explained as a progressive depletion of mobile regions inside the glass matrix, and does not provide any hint of a Gardner transition. Figure 2 focuses on temperatures below 100 K. In this regime $|\chi_3^{(3)}|$ is essentially constant. A computation of its value based on the assumption of *independent* local excitations is presented in the Supplemental Material [37] and leads to

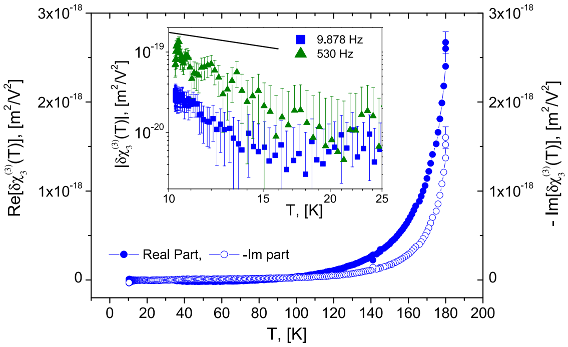

FIG. 1. Temperature dependence of $\delta\chi_3^{(3)}(T) \equiv \chi_3^{(3)}(T) - \chi_3^{(3)}(30\text{ K})$ where $\chi_3^{(3)}$ is the third harmonics cubic susceptibility of glassy glycerol, here measured at an electrical frequency $f = 9.878$ Hz. The left axis is for the real part data, while the right axis is for the opposite of the imaginary part data. Inset: temperature evolution of $|\delta\chi_3^{(3)}(T, f)|$ for $T \leq 25$ K, where $f = 9.878$ Hz (squares) or $f = 530$ Hz (triangles). The solid line is an example of the $1/T^2$ dependence expected for non interacting TLSs.

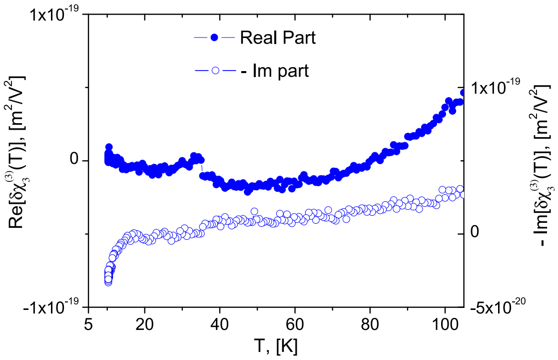

FIG. 2. Close-up view of the low T part of Fig. 1.

a value $0.9 \times 10^{-19}\ \mathrm{m}^2/\mathrm{V}^2$ which agrees well with the one found by experiments [39]. Note that in the regime [10 K; 16 K], one sees a very small increase of $|\delta\chi_3^{(3)}(T,f)|$ upon cooling. This phenomenon, which is hardly above our experimental uncertainty, see the error bars given in the inset of Fig. 1, was systematically found in the several experiments that we carried out either by varying the value of the electric field E or the angular frequency ω. It can be explained using TLS theory, which predicts a behavior $|\delta\chi_3^{(3)}(T,f)| \propto 1/T^2$; see the solid line in the inset of Fig. 1 and the Supplemental Material [37] for more detail. All in all, our experimental results do not show any evidence of a Gardner transition from T_g down to 10 K, and they are quantitatively compatible with a scenario based on independent local excitations. Because we are limited to $T \geq 10$ K, we cannot strictly exclude that some Gardner transition might happen at a critical temperature below 8 K.

In order to rationalize these findings we use a real space approach. Our main assumption is that thermal fluctuations in glasses are due to localized excitations corresponding to partial local atomic motion within the frozen glass matrix [42]. This description naturally connects to the one put forward in the past for the low temperature properties of molecular glasses, which is based on two-level systems (TLS) [3,5] as well as to many theories of rheology of amorphous solids, which are based on localized soft spots of particles that are prone to rearrangement [43,44]. Besides, recent simulations exhibit localized excitations in models of molecular glasses [21]. The interaction between excitations is mediated by the electric and the elastic fields. Since the local conformations corresponding to the excitations are random the resulting couplings are random. From this real space perspective, the Gardner phase would be a spin-glass phase arising from the interaction of local excitations.

In order to study the Gardner phase, we model the localized excitations as N degrees of freedom located in random positions in space. Their density is $\rho = N/V$, where V is the total volume of the system. Each one of them will be denoted $\sigma_{\mathbf{x}}$, where $\mathbf{x}$ is the corresponding position. Each localized excitation can be in $m_{\mathbf{x}}$ ($\mathbf{x}$-dependent) states, which correspond to the possible conformations of the localized excitation, i.e., of the local atomic positions belonging to the excitation. For simplicity, in the following we take $m_{\mathbf{x}} = 2$ for any $\mathbf{x}$ as done for TLS, and use a notation where $\sigma_{\mathbf{x}} \in \{-1, 1\}$ correspond, respectively, to the low and high energy state of the local excitations. Our arguments and conclusions carry over straightforwardly for $m_{\mathbf{x}} > 2$. The corresponding Hamiltonian reads

$$H = -U_0 \sum_{i \neq j} \frac{1}{|\mathbf{x}_i - \mathbf{x}_j|^3} u_{ij} \sigma_{\mathbf{x}_i} \sigma_{\mathbf{x}_j} - \frac{1}{2} \sum_{i=1}^{N} \epsilon_i \sigma_{\mathbf{x}_i}. \quad (2)$$

We have decomposed the interaction between the local excitations in an amplitude, which decreases as the cube of the distance between excitations, and in a random adimensional coupling u_{ij}, which depends on the local stress tensors and electric dipoles corresponding to the different states of the local excitations, see, e.g., [3–5] for a similar modeling for TLS. U_0 is the energy scale of the interaction (measured in temperature times unit of volume). The fact that local excitations can be in states with different local energies is encoded in the random positive ϵ_is. The model is effectively a spin glass since the couplings u_{ij} are characterized by an even distribution. This follows from the fact that u_{ij} is bilinear in the dipolar electric moments and the strain tensors associated with the interacting local excitations [3–5]. Since their distribution in space is statistically symmetric under rotation, in particular under a change of sign, the probability of u_{ij} and $-u_{ij}$ are identical. Note that there are correlations between couplings u_{ij} associated with the same excitations, i.e., u_{ij}, u_{ik} are correlated random variables. The local positive energies ϵ_i are assumed to be independent random variables with a density distribution $(1/\Delta_{\mathrm{typ}}) f(\epsilon/\Delta_{\mathrm{typ}})$, where Δ_{typ} is the typical value of ϵ_i for a localized excitation. We expect, although it is not a crucial ingredient for what follows, that Δ_{typ} is of the order of the typical effective barrier for β relaxation below the glass transition temperature, i.e., thousands of kelvins.

Our aim here is not to construct the precise phase diagram of this model, for which a precise characterization of the probability distribution of the u_{ij}'s and ϵ_i's would be required, but instead we want to investigate the possible existence of the Gardner phase based on order of magnitude estimations. In order to do that, one of the key ingredients is the amount of local excitations per unit volume, which can be estimated from TLS physics, since those correspond to a very low-energy flank of the distribution. Results on TLS tell us that $f(0) > 0$ and that the density of thermally active localized excitations at, say, 10 K is around $1/(7\ \mathrm{nm})^3$ [3,4]. Since those are expected to be characterized by $\epsilon \ll \Delta_{\mathrm{typ}}$, we obtain that

$$\int_0^{10\ \mathrm{K}} d\epsilon \frac{1}{\Delta_{\mathrm{typ}}} f\left(\frac{\epsilon}{\Delta_{\mathrm{typ}}}\right) \simeq \frac{10\ \mathrm{K}}{\Delta_{\mathrm{typ}}} f(0) \simeq \frac{1}{(7\ \mathrm{nm})^3},$$

which sets the scale of $f(x)$ (energies are expressed in units of temperature). Note that assuming a Δ_{typ} of the order of a few thousands of kelvin (see above) one gets approximatively an excitation per nanometer cube, which seems reasonable for typical excitations. In order to connect with the notation used for TLS, we recall that in that case $\epsilon = \sqrt{\delta^2 + \Delta_0^2}$ where the potential disorder energy δ and the coupling tunnel energy Δ_0 are distributed with a density $\rho(\delta, \Delta_0) = \bar{p}/\Delta_0$ [3,4] where $\bar{p}$ is a constant. This yields—see Supplemental Material [37] and [3,4]—$f(0)/\Delta_{\mathrm{typ}} \simeq \bar{p}\ \ln(\Delta_0^{\max}/\Delta_0^{\min})$, where $\ln(\Delta_0^{\max}/\Delta_0^{\min}) \simeq 20$. An important hypothesis for our arguments is that the function $f(x)$ is regular. This amounts to assuming that $f(x)/f(0)$ starts from one at $x = 0$, varies for x of the order of one, and eventually goes to zero for larger x's. As far as order of magnitude estimates are concerned, we can use the simple form $f(x) = f(0)\theta(x - 1)$, with $\theta(x)$ the Heaviside function.

The main issue we wish to address is whether the interaction between local excitations is large enough to lead to a spin-glass phase. In order to work this out, one has to compare the value of the interaction to the local energy difference ϵ_i; if the latter is too strong then a local excitation is subjected to a very strong bias toward the low-energy state $\sigma_i = -1$ and its physical behavior is insensitive to the other ones, i.e., no long-range order can be present. Note that even though the interaction is a power law, it is short ranged as far as spin-glass order is concerned [45], i.e., the effective field due to the interactions with the other local excitations is dominated by the closest excitations [46].

Not all local excitations can be considered active. In fact, for a given temperature T and a given observation time τ, some of them are frozen out and cannot change state, or they are just too slow to give rise to collective behavior and cannot participate to the putative spin-glass state. As a consequence, to be relevant, a local excitation must have an ϵ_i less than a certain energy $\bar{\epsilon}(T, \tau) \leq \Delta_{\mathrm{typ}}$ which depends on T and τ. On general grounds one expects this energy scale to be less than or equal to Δ_{typ}, and to decrease with T and increase with τ [48]. The precise expression of $\bar{\epsilon}(T, \tau)$ is not needed for our arguments.

Using the simplified form of $f(x)$ we therefore find that the density of active local excitations is

$$\mathcal{N}[\bar{\epsilon}(T, \tau)] = f(0) \frac{\bar{\epsilon}(T, \tau)}{\Delta_{\mathrm{typ}}}.$$

From (2) the strength of the interaction between the local excitations is $\mathcal{I} = U_0/\ell^3$, where $\ell(T)$ is the typical distance between them. Hence, the interaction strength is proportional to the density of localized excitation, which by the previous equation is proportional to the typical strength $\bar{\epsilon}(T, \tau)$ of the random fields. These relations therefore allow us to establish a direct comparison between $\mathcal{I}$ and $\bar{\epsilon}(T, \tau)$:

$$\mathcal{I}[\bar{\epsilon}(T, \tau)] = \frac{U_0}{\ell^3} \simeq U_0 \mathcal{N}[\bar{\epsilon}(T, \tau)] = k\bar{\epsilon}(T, \tau), \qquad (3)$$

where $k = U_0 f(0)/\Delta_{\mathrm{typ}} = U_0 \bar{p}\ \ln(\Delta_0^{\max}/\Delta_0^{\min})$. For molecular glass former prepared under normal quenched condition k is of the order 0.002–0.02 [49,50]. In the Supplemental Material [37] we work out this value for glycerol, and show that even considering the additional modes showing up in the Boson peak region, k may reach 0.04 at most. This implies that the strength of the interaction $\mathcal{I}[\bar{\epsilon}(T, \tau)]$ is generically much smaller than the typical local energy $\bar{\epsilon}(T, \tau)$ [51]. Therefore, we expect that the Gardner spin-glass phase should be suppressed as we explain now. Indeed, theoretical studies have shown that random fields hamper the existence of long-range order: within the droplet model an infinitesimal random field is enough to destroy the spin-glass phase [52]; whereas within mean-field theory a finite field strength, comparable to the coupling strength, is needed [53]. Simulations and experiments have confirmed the negative role of the field: for three dimensional short-range spin glasses [54,55], if a transition takes place, it does so for field strengths much lower than the coupling strength. For three dimensional dipolar spin-glasses, a model similar to the one studied in this Letter, even without a field the existence of long-range spin-glass order is not established [56], thus making the fate of the spin-glass phase in a field even more uncertain in this case. All that leads us to the conclusion that in the present case, where the interaction strength between local excitations is typically twenty to fifty times smaller than the value of the local random field, the emergence of the spin-glass phase, and hence of the Gardner phase, is unlikely.

The natural question that comes out from the conclusions above is why molecular glasses are so different from colloidal and granular ones for which instead strong signatures of Gardner physics have been found [16,22–27]. Our results point toward two possible reasons. On the one hand, colloids and granular systems are prepared in such a way that the resulting solids are much less annealed, since the timescale for microscopic motion is much larger (10^{-6} s for colloids and a fraction of seconds for granular media). This leads to a much higher density of soft localized excitations, and in consequence to an increase of the interactions term over the random field one, thus favoring the existence of the Gardner phase. On the other hand, the proximity to the jamming transition that takes place for both systems also transforms the nature of their excitations. Indeed, at jamming, on top of localized excitations there are also delocalized ones [57,58], which could favor the Gardner transition. How the mechanisms outlined above conspire together to lead to Gardner physics in three dimensional colloidal and granular systems is not clear. Simulations and experiments can help clarify this issue. Direct analysis of the nature of excitations, as the ones performed numerically in [59], are instrumental. Another possibility is studying systems where the two mechanisms above are separated, e.g., ellipsoids or hard spheres under SWAP dynamics [60,61]. To find a Gardner transition in molecular glasses, it would be interesting to find protocols to prepare very poorly annealed systems. Another possibility is to study network glasses, such as amorphous silica (SiO_2), whose structure is close to be marginally connected [62,63] and may then display Gardner physics.

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