Emergence of Long-Range Correlations and Rigidity at the Dynamic Glass Transition

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At the so-called dynamic glass transition predicted by the mean-field replica approach equilibrium liquid's translational symmetry is spontaneously broken, albeit at the microscopic level. We show that this fact implies the emergence of Goldstone modes and long-range density correlations. We derive and evaluate a new statistical mechanical expression for the glass shear modulus.

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Introduction.—The difference between a liquid and a solid manifests itself in their (linear) response to a small volume-preserving shear deformation: a liquid flows, whereas in a solid internal forces arise which are proportional to and oppose the deformation. The solid's rigidity is an emergent [1] property: it amounts to transmitting internal forces opposing a volume-preserving deformation over macroscopic distances but it does not originate from long-range intermolecular interactions. However, it is intuitively plausible, and it was argued at a phenomenological level [2,3], that the emergence of rigidity has to be accompanied by the appearance of long-range correlations.

Interestingly, the first microscopic argument for the existence of long-range correlations in crystalline solids was given in 1966 by Wagner [4], preceding phenomenological analyses of Refs. [2,3]. However, the part of his paper dealing with solids seems to have been overlooked. More importantly, while he showed that in crystalline solids broken translational symmetry implies long-range correlations, he did not connect the latter to solids' rigidity.

The intimate relation between long-range correlations and rigidity was established somewhat indirectly. Bavaud *et al.* [5] analyzed the standard expression for the elastic moduli, which is derived by calculating free energy change due to a deformation. They proved that this expression reduces itself to the inverse of the isothermal compressibility if both intermolecular interactions and density correlation functions are short range. Thus, they showed that for systems with short-range interactions the absence of long-range density correlations implies the vanishing of the shear modulus.

The microscopic argument for the existence of long-range correlations in crystals was independently derived by Szamel and Ernst [6]. They proposed an expression for a displacement field in crystals in terms of the microscopic density and showed that the displacement field defined in this way exhibits long-range correlations, as expected on the basis of phenomenological arguments [2,3].

Here we consider an amorphous solid which appears as a consequence of the so-called dynamic glass transition [7] predicted by the mean-field replica approach [8–10]. We show that, in close analogy with crystalline solids, broken translational symmetry of this amorphous solid implies the emergence of long-range correlations and rigidity.

It should be recalled that, although macroscopically the amorphous solid's density is constant, microscopically it is randomly nonuniform. Thus, a rigid translation of the amorphous solid produces an equivalent but different state. To describe this randomly broken translational symmetry, and amorphous solids in general, we use the replica approach of Franz and Parisi [8,9]. In this approach, the appearance of the amorphous solid manifests itself in nontrivial replica off-diagonal densities. We argue that the existence of distinct, rigidly shifted states of the amorphous solid manifests itself in a family of equivalent sets of replica off-diagonal densities. Next, we propose an expression for the displacement field in terms of microscopic two-point replica off-diagonal densities. We show that the displacement field defined in this way exhibits long-range correlations. Since the displacement field is defined in terms of microscopic two-point replica off-diagonal densities, long range of the displacement field's correlations implies a slow decay of a four-point replica off-diagonal correlation function.

In addition, we derive and evaluate a new expression for the shear modulus of amorphous solids. To this end we evaluate microscopically the force needed to maintain a shear deformation and compare it to an expression obtained from the macroscopic theory of elasticity.

Family of equivalent glassy states.—Following Franz and Parisi [8,9] we consider a system of particles coupled to a frozen (referred to as ''quenched'') configuration of the same system via an attractive potential

$$\epsilon \sum_{i,j} w(|\mathbf{r}_i - \mathbf{r}_j^0|), \tag{1}$$

where $\mathbf{r}_i$ and $\mathbf{r}_j^0$ are positions of particles i and j in the system and in the quenched configuration, respectively, and $w(r)$ is a monotonic function with a minimum at $r = 0$. We assume that as the amplitude ϵ of the potential is being gradually reduced to 0, if the temperature is low enough or if the density is high enough, the system gets stuck in a metastable state characterized by nontrivial correlations with the quenched configuration. The

appearance of such a metastable state is termed the dynamic glass transition [8–10]. To facilitate averaging over a distribution of quenched configurations we replicate the system s times and at the end we take the limit $s \to 0$ [11]. At the level of the replicated system, the dynamic glass transition manifests itself in the appearance of nontrivial two-point replica off-diagonal densities $n_{\alpha\beta}(r)$, $\alpha \neq \beta$. With $w(r)$ having a minimum at $r = 0$, these functions exhibit a pronounced peak at $r = 0$, oscillate in phase with the equilibrium pair distribution function $g(r)$, and decay to n^2 at large distances (n is the average density). In the following the metastable state with all replica off-diagonal densities having the main peak at $r = 0$ will be referred to as the "classical" state.

The above described construction can also be performed with a potential that pins the system in a position shifted with respect to the quenched configuration,

$$\epsilon \sum_{i,j} w(|\mathbf{r}_i - \mathbf{r}_j^0 - \mathbf{a}|). \tag{2}$$

The metastable states obtained with potentials (1) and (2) are identical up to a rigid shift by vector $\mathbf{a}$ of the system relative to the quenched configuration. At the level of the replicated system this translation corresponds to the following transformation of two replica densities,

$$n_{\alpha 0}(r) \to n_{\alpha 0}(|\mathbf{r} - \mathbf{a}|), \qquad n_{0\alpha}(r) \to n_{0\alpha}(|\mathbf{r} + \mathbf{a}|) \tag{3}$$

for $\alpha > 0$ and keeping all other two replica densities (both diagonal and off-diagonal) unchanged. In the following we consider only densities $n_{\alpha 0}$, $\alpha > 0$, since densities $n_{0\alpha}$ can be obtained from the former ones.

This observation implies that instead of one glassy state we have a continuous family of states that can be labeled by a vector $\mathbf{a}$ denoting translation with respect to the classical state. This is analogous to what is found in other systems with broken continuous symmetry [2].

We note that the rigid shift described above breaks replica symmetry. However, since replica nonsymmetric states obtained by rigid translations arise due to broken translational symmetry, they have the same free energy as the classical state (which is replica symmetric [8,9]). Thus, the present case is fundamentally different from replica symmetry breaking in mean-field spin glasses [12]. In the latter case, replica symmetry breaking originates from the existence of a multitude of different glassy states unrelated by translations. Importantly, the replica nonsymmetric state minimizes the free energy.

Microscopic definition of displacement field.—According to the conventional definition, a displacement field in crystalline solids is defined in terms of departures of particles' instantaneous positions from their lattice sites. Notably, this definition implicitly assumes that each particle can be assigned to a specific lattice site. Thus, it is not applicable for, e.g., crystals with vacancies or interstitials. This fact was one of the motivations for a microscopic definition of the displacement field [6,13].

Since amorphous solids do not have any underlying crystalline lattice, it is impossible to use the conventional definition. An operational definition that is closest to the one used for crystals uses a displacement field defined in terms of departures of the instantaneous positions with respect to the average positions. While a procedure like this can be, at least in principle, implemented in a computer simulation [14] or an experiment [15], it is not clear how to formulate it theoretically. The definition we propose is similar in spirit to this procedure in that it relates microscopic and average densities of the amorphous solid.

We propose the following definition of (the Fourier transform of) the displacement field in terms of microscopic replica off-diagonal density,

$$\mathbf{u}(\mathbf{k}) = -\frac{1}{\mathcal{N} s} \int d\mathbf{r}_1 e^{-i\mathbf{k}\cdot\mathbf{r}_1} \int d\mathbf{r}_{21} \sum_{\alpha>0} \frac{\partial n_{\alpha 0}(\mathbf{r}_1, \mathbf{r}_2)}{\partial \mathbf{r}_1} \times \sum_{i,j} \delta(\mathbf{r}_1 - \mathbf{r}_i^\alpha)\delta(\mathbf{r}_2 - \mathbf{r}_j^0). \tag{4}$$

Here $n_{\alpha 0}(\mathbf{r}_1, \mathbf{r}_2)$ is the classical two-point replica off-diagonal density and $\sum_{i,j}\delta(\mathbf{r}_1 - \mathbf{r}_i^\alpha)\delta(\mathbf{r}_2 - \mathbf{r}_j^0)$ is the microscopic two-point replica off-diagonal density with $\mathbf{r}_i^\alpha$ and $\mathbf{r}_j^0$ being positions of particles i and j in replica α and 0, respectively. Finally, $\mathcal{N}$ is the normalization factor, $\mathcal{N} = (3s)^{-1} \int d\mathbf{r}_{21} \sum_{\alpha>0} [\partial_{\mathbf{r}_1} n_{\alpha 0}(\mathbf{r}_1, \mathbf{r}_2)]^2$. Note that definition (4) is symmetric with respect to replica indices α, $\alpha > 0$. This reflects the fact that the system is first deformed and only subsequently replicated. Thus, deformations in all $\alpha > 0$ replicas are the same.

Importantly, definition (4) is only applicable to infinitesimally small deformations of the classical state. A similar restriction applies to the definition of the displacement field in crystals proposed in Refs. [6,13].

Finally, the difference between (4) and the definition proposed in Refs. [6,13] originates from the fact that in an amorphous solid (at the level of replicated theory) the displacement can only be revealed through the change of the 0α sector of replica off-diagonal densities. In contrast, in crystalline solids the displacement can be "read off" from the change of the density.

Long-range density correlations.—First, we adapt the calculation presented in Ref. [6] and prove that correlations of displacement field (4) exhibit a small wave vector divergence. Then we show that long-range displacement field correlations imply slow decay of a component of a four-point replica off-diagonal correlation function.

We start with Bogoliubov's inequality [2,16] $\langle |A|^2 \rangle \langle |B|^2 \rangle \geq |\langle AB \rangle|^2$ with $A = V^{-1/2} \hat{\mathbf{n}} \cdot \mathbf{u}^*(\mathbf{k})$ and $B = V^{-1/2} s^{-1} \sum_{\alpha>0} \hat{\mathbf{n}} \cdot i\mathcal{L}_\alpha \mathbf{g}_\alpha(\mathbf{k})$, where $\hat{\mathbf{n}}$ is an arbitrary unit vector, $\mathbf{g}_\alpha$ is the Fourier transform of the microscopic momentum density in replica α, $\mathbf{g}_\alpha(\mathbf{k}) = \sum_i m \mathbf{v}_i^\alpha e^{-i\mathbf{k}\cdot\mathbf{r}_i^\alpha}$, with m denoting the particle's mass and $\mathbf{v}_i^\alpha$ being the

velocity of particle i in replica α, and $\mathcal{L}_\alpha$ is the Liouville operator [2] in replica α.

The cross term can be evaluated using the self-adjoint property of $\mathcal{L}_\alpha$. In the $\mathbf{k} \to \mathbf{0}$ limit we obtain $\langle AB\rangle = -k_B T/s$. $\langle |B|^2\rangle$ can be expressed in terms of the correlation function of the stress tensor. The stress tensor in replica α is defined through the continuity equation for the momentum in replica α, $i\mathcal{L}_\alpha \mathbf{g}_\alpha(\mathbf{k}) = -i\mathbf{k}\cdot\overleftrightarrow{\sigma}_\alpha(\mathbf{k};t)$.

Substituting $\langle AB\rangle$ and $\langle |B|^2\rangle$ into Bogoliubov's inequality, and taking the $s \to 0$ limit we obtain

$$\lim_{s\to 0}\frac{s}{V}\langle|\hat{\mathbf{n}}\cdot\mathbf{u}(\mathbf{k})|^2\rangle \geq \frac{1}{k^2}\frac{(k_B T)^2}{\lim_{\mathbf{k}\to\mathbf{0}}\frac{1}{V}\langle\delta\Sigma(\mathbf{k})\rangle}, \tag{5}$$

where

$$\delta\Sigma(\mathbf{k}) = |\hat{\mathbf{k}}\cdot\overleftrightarrow{\sigma}_1(\mathbf{k})\cdot\hat{\mathbf{n}}|^2 - |\hat{\mathbf{k}}\cdot\overleftrightarrow{\sigma}_1(\mathbf{k})\cdot\hat{\mathbf{n}}||\hat{\mathbf{k}}\cdot\overleftrightarrow{\sigma}_2(\mathbf{k})\cdot\hat{\mathbf{n}}| \tag{6}$$

and $\hat{\mathbf{k}} = \mathbf{k}/k$.

Bound (5) and (6) for the small wave vector behavior of the displacement field correlations is compatible with the result of a phenomenological analysis which predicts a k^{-2} divergence [2]. This agreement, and the closeness of the steps outlined above and the general scheme presented in Chap. 7 of Ref. [2] provide an additional justification for definition (4) and for recognizing the displacement field as a "hydrodynamic Goldstone mode."

Finally, since the displacement field is defined in terms of microscopic two-point replica off-diagonal densities, the left-hand-side (LHS) of Eq. (5) can be rewritten in terms of four-point replica off-diagonal densities,

$$\begin{aligned}\text{LHS} = \frac{1}{\mathcal{N}V}\int d\mathbf{r}_1\cdots d\mathbf{r}_4\,\hat{\mathbf{n}}\cdot\frac{\partial n_{10}(\mathbf{r}_1,\mathbf{r}_2)}{\partial\mathbf{r}_1}\hat{\mathbf{n}} \\ \cdot\frac{\partial n_{10}(\mathbf{r}_3,\mathbf{r}_4)}{\partial\mathbf{r}_3}[n_{1010}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4) \\ - n_{1020}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)]e^{-i\mathbf{k}\cdot\mathbf{r}_{13}}.\end{aligned} \tag{7}$$

The combination of Eqs. (5) and (7) proves the slow decay of a component of a four-point replica off-diagonal correlation function of the amorphous solid. An analogous calculation for a crystal proves the slow decay of correlations of high Fourier components of the (nontranslationally invariant) two-particle density [4,6,13].

Shear modulus.—The derivation of a new formula for the amorphous solid's shear modulus proceeds in two steps. First, we consider the change of replica off-diagonal densities upon an infinitesimally small, long-wavelength deformation. Next, we calculate microscopically the force needed to maintain such a deformation. Comparison of this force with the corresponding macroscopic expression allows us to identify the shear modulus.

According to Eq. (3), under an infinitesimally small uniform translation, replica off-diagonal densities $n_{\alpha 0}$, $\alpha > 0$, change in the following way: $n_{\alpha 0}(r) \to n_{\alpha 0}(r) - \mathbf{a}\cdot\partial_{\mathbf{r}} n_{\alpha 0}(r)$. To generalize this formula to an infinitesimally small nonuniform translation we imagine a pinning potential which deforms the system by imposing an infinitesimal shift that depends on the position of the particle. This suggests the following generalization,

$$n_{\alpha 0}(r_{12}) \to n_{\alpha 0}(\mathbf{r}_1,\mathbf{r}_2) = n_{\alpha 0}(r_{12}) - \mathbf{a}(\mathbf{r}_1)\cdot\partial_{\mathbf{r}_1} n_{\alpha 0}(r_{12}), \tag{8}$$

where $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$ and $\mathbf{a}(\mathbf{r}_1)$ is an infinitesimal, long-wavelength deformation imposed on the system. We emphasize that (8) is only assumed for a long-wavelength volume-preserving deformation. Specifically, we assume a transverse deformation such that $\partial_{\mathbf{r}}\cdot\mathbf{a}(\mathbf{r}) = 0$.

Equation (8) is inspired by the assumption made by Triezenberg and Zwanzig [17], and by Lovett *et al.* [18] in their analyses of surface tension. A similar assumption was made by Szamel and Ernst [6] in their derivation of shear modulus of crystalline solids. Finally, assumption (8) is consistent with our microscopic definition of the displacement field: if all densities $n_{\alpha 0}$, $\alpha > 0$, are changed as indicated in (8) then the Fourier transform of the average displacement field is equal to $\langle\mathbf{u}(\mathbf{k})\rangle = \int d\mathbf{r} e^{-i\mathbf{k}\cdot\mathbf{r}}\mathbf{a}(\mathbf{r})$.

Next, we calculate the force exerted on one replica of the system by the pinning potential chosen in such a way that it imposes the replica off-diagonal densities (8).

First, we identify the pinning potential needed to maintain densities (8). Since we are only interested in infinitesimally small deformations and thus infinitesimally small changes of replica off-diagonal densities, we resort to a (static) linear-response type relation. Thus, we express the pinning potential using a functional derivative of the inter-replica potential with respect to the replica off-diagonal density,

$$\sum_{\beta>0}\int d\mathbf{r}_3 d\mathbf{r}_4\left(\frac{\delta V_{\alpha 0}(\mathbf{r}_1,\mathbf{r}_2)}{\delta n_{\beta 0}(\mathbf{r}_3,\mathbf{r}_4)}\right)_n[-\mathbf{a}(\mathbf{r}_3)\cdot\partial_{\mathbf{r}_3} n_{\beta 0}(r_{34})]. \tag{9}$$

Note that since we have assumed a volume-preserving deformation, the functional derivative in expression (9) should be taken at constant density.

Then, we calculate the force per unit volume exerted by the above pinning potential on the replica α. The Fourier transform of this force, after integrating by parts and using the transverse character of the deformation, can be written in the following way:

$$\begin{aligned}\mathbf{F}_\alpha(\mathbf{k}) = -\frac{1}{V}\int d\mathbf{r}_1\cdots d\mathbf{r}_4 e^{-i\mathbf{k}\cdot\mathbf{r}_{13}}[\partial_{\mathbf{r}_1} n_{\alpha 0}(r_{12})] \\ \times\sum_\beta\left(\frac{\delta V_{\alpha 0}(\mathbf{r}_1,\mathbf{r}_2)}{\delta n_{\beta 0}(\mathbf{r}_3,\mathbf{r}_4)}\right)_n[\partial_{\mathbf{r}_3} n_{\beta 0}(r_{34})]\cdot\mathbf{a}(\mathbf{k}).\end{aligned} \tag{10}$$

Finally, we take the long-wavelength limit and expand the right-hand side of Eq. (10) in a power series in $\mathbf{k}$. The zeroth order term vanishes because no force is needed in order to accomplish a rigid shift of the amorphous solid. The first order term vanishes by symmetry. The second order term, in the $s \to 0$ limit and for a shear deformation

involving y-dependent translation along the x axis, can be written in the following form:

$$F_x(k_y) = \mu k_y k_y a_x(k_y), \tag{11}$$

where μ is given by

$$\mu = \frac{k_B T}{2V} \int d\mathbf{r}_1 \cdots d\mathbf{r}_4 y_{13}^2 [\partial_{x_1} n_{10}(r_{12})][\partial_{x_3} n_{10}(r_{34})] \times \left[\left(\frac{\delta[\beta V_{10}(\mathbf{r}_1, \mathbf{r}_2)]}{\delta n_{10}(\mathbf{r}_3, \mathbf{r}_4)} \right)_n - \left(\frac{\delta[\beta V_{10}(\mathbf{r}_1, \mathbf{r}_2)]}{\delta n_{20}(\mathbf{r}_3, \mathbf{r}_4)} \right)_n \right]. \tag{12}$$

Macroscopically, for an isotropic solid the force needed to maintain a long-wavelength shear deformation is given by a formula identical to Eq. (11), where μ is the shear modulus. This fact allows us to identify μ given by Eq. (12) as the shear modulus of the amorphous solid.

To evaluate μ one has to use a concrete (albeit necessarily approximate) implementation of the replica approach in order to calculate replica off-diagonal densities and functional derivatives in Eq. (12). To obtain $n_{\alpha 0}(r)$ we use the recently proposed version [19] which is consistent with mode-coupling theory [20]. To calculate the derivatives we use an additional approximation [21],

$$\delta c_{\alpha 0}(\mathbf{r}_1, \mathbf{r}_2) = -\beta V_{\alpha 0}(\mathbf{r}_1, \mathbf{r}_2)[n_{\alpha 0}(\mathbf{r}_1, \mathbf{r}_2)/n^2]. \tag{13}$$

Here $\delta c_{\alpha 0}$ is the change of the replica off-diagonal direct correlation function due to a weak replica off-diagonal potential $V_{\alpha 0}$. Approximation (13) combined with replica Ornstein-Zernicke equations allow us to get explicit expressions for functional derivatives that enter Eq. (12).

In Fig. 1 we show the shear modulus for the hard sphere glass calculated using as an equilibrium input the Percus-Yevick structure factor.

Discussion.—We identified Goldstone modes and long-range correlations appearing in the amorphous solid due to the spontaneously broken translational symmetry. We derived a new expression for the shear modulus of the amorphous solid [24]. This expression is complementary to the standard one. In particular, our expression can be used to evaluate the shear modulus of the hard sphere glass whereas the standard formula is not applicable for hard sphere systems. In contrast to the result obtained in a recent replica approach study [22] (which was based on the standard formula) we found a discontinuous change of the shear modulus at the dynamic glass transition [23].

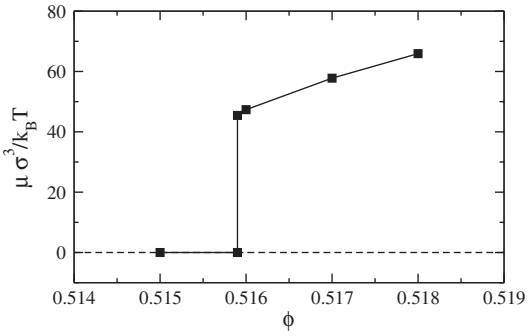

FIG. 1. Dimensionless shear modulus, $\mu\sigma^3/k_B T$, for the hard sphere glass as a function of the volume fraction, $\phi = n\pi\sigma^3/6$. The shear modulus changes discontinuously at the dynamic transition which occurs at $\phi_c = 0.5159$.

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Note added.—Recently, we became aware of a related approach [24] which used the language of density functional theory of a disordered glassy state. This approach is, however, much more difficult to implement to obtain explicit results.

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