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Finite-size effects in the nonphononic density of states in computer glassesEdan Lerner ^{*}*Institute for Theoretical Physics, University of Amsterdam, Science Park 904, Amsterdam, the Netherlands*

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The universal form of the density of nonphononic, quasilocalized vibrational modes of frequency ω in structural glasses, $\mathcal{D}(\omega)$, was predicted theoretically decades ago, but only recently revealed in numerical simulations. In particular, it has been recently established that, in generic computer glasses, $\mathcal{D}(\omega)$ increases from zero frequency as ω^4 , independent of spatial dimension and of microscopic details. However, it has been shown [Lerner and Bouchbinder, *Phys. Rev. E* **96**, 020104(R) (2017)] that the preparation protocol employed to create glassy samples may affect the form of their resulting $\mathcal{D}(\omega)$: glassy samples rapidly quenched from high-temperature liquid states were shown to feature $\mathcal{D}(\omega) \sim \omega^\beta$ with $\beta < 4$, presumably limiting the degree of universality of the ω^4 law. Here we show that exponents $\beta < 4$ are seen only in small glassy samples quenched from high-temperature liquid states—whose sizes are comparable to or smaller than the size of the disordered core of soft quasilocalized vibrations—while larger glassy samples made with the *same protocol* feature the universal ω^4 law. Our results demonstrate that observations of $\beta < 4$ in the nonphononic density of states stem from finite-size effects, and we thus conclude that the ω^4 law should be featured by any sufficiently large glass quenched from a melt.

DOI: [10.1103/PhysRevE.101.032120](https://doi.org/10.1103/PhysRevE.101.032120)**I. INTRODUCTION**

The form of low-frequency spectra of glassy solids remains a subject of lively debates, despite decades of theoretical [1–7], numerical [8–15], and experimental research [16–19]. Low-frequency excitations are believed to be responsible for several important glassy phenomena, ranging from yielding [20–23] and shear banding [24–27] to anomalous wave attenuation [28]. Relations between low-frequency excitations and both relaxation in supercooled liquids [29–31] and low-temperature thermodynamics of glassy solids [32–34] have also been suggested.

Numerical investigations of computer glass models have contributed significantly to our understanding of many problems in glass physics and, in particular, to revealing the structural and statistical properties of soft, quasilocalized modes (QLMs) in glassy solids. Pioneered by Schober and coworkers decades ago [8,9], computational investigations have revealed that the lowest-frequency vibrational modes in computer glasses are quasilocalized: they consist of a disordered core, decorated by an algebraically decaying field away from the core [13,35,36]. Furthermore, it was discovered that the distribution $\mathcal{D}(\omega)$ of these nonphononic modes follows a universal $\sim \omega^4$ law (with ω denoting the frequency), independent of spatial dimension [14] or microscopic details [13,15]. Remarkably, these findings are consistent with theoretical predictions put forward some time ago [1–3].

Universality of the nonphononic $\mathcal{D}(\omega) \sim \omega^4$ density of states with respect to glass preparation protocol was also reported [37–39], but with some caveats; in particular, in Ref. [37] it was shown that glassy samples created by an

infinitely fast quench from high-temperature liquid states, or by continuous quenches at very high rates, feature $\mathcal{D}(\omega) \sim \omega^\beta$ with $\beta < 4$. In the same reference it was further suggested that the observation of $\beta < 4$ could be consistent with the Soft Potential Model [1,2], which states that very unstable states—presumably similar to those obtained by instantaneous *overdamped* quenches of high-temperature liquid states—could feature $\beta < 4$.

In this paper we show that the observations of $\mathcal{D}(\omega) \sim \omega^\beta$ with $\beta < 4$ made in Ref. [37] are, in fact, the result of too-small glassy samples employed. The results presented here for $\mathcal{D}(\omega)$ were obtained using the same computer glass model as employed in that reference. We prepared large ensembles of glassy samples by instantaneous quenches from high-temperature liquid states, and by gradually varying the system size N , we show in what follows that the exponent β that characterizes the distribution of soft, nonphononic QLMs varies from $\beta < 4$ in small systems to $\beta = 4$ in larger systems. We further show that the characteristic length ξ_g of soft QLMs—recently shown to be well represented by the spatial form of linear responses to localized force dipoles [36]—is comparable to the system sizes in which $\beta < 4$ is observed, suggesting that finite-size effects in $\mathcal{D}(\omega)$ depend on the ratio ξ_g/L , with L denoting the linear system size. We expect our results to serve as constraints on the formulation of future theories about the statistical mechanics of mesoscale elasticity in glassy solids.

II. MODELS, METHODS, AND UNITS

The model employed here is described in detail in, e.g., Ref. [38]; it is a binary mixture of “large” and “small” particles interacting via a radially symmetric, purely repulsive $\propto r^{-10}$ pairwise potential, with r denoting the distance

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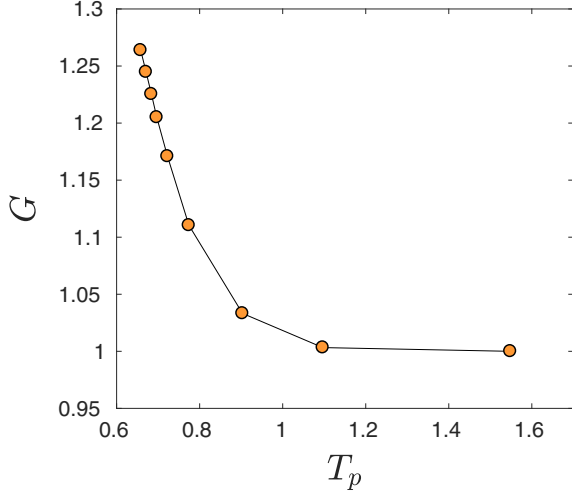


FIG. 1. Sample-to-sample mean athermal shear modulus G , plotted against the equilibrium parent temperature T_p from which our glassy samples were instantaneously quenched. In this work we express temperatures in terms of the onset temperature of the high- T_p plateau of G , and elastic moduli in terms of the high- T_p plateau of G itself.

between pairs of particles. In what follows, we refer to the *parent temperature* T_p of an ensemble of glassy samples as the equilibrium temperature from which they were instantaneously quenched; see, e.g., Refs. [36,38]. Parent temperatures T_p are expressed in terms of the onset parent temperature T_{onset} above which the sample-to-sample mean of the athermal shear modulus G [38] of underlying inherent states plateaus, as shown in Fig. 1. We express lengths in terms of $a_0 \equiv (V/N)^{1/3}$ where $V = L^3$ is the volume. The mass m is the same for all particles and is chosen as our microscopic units of mass. Elastic moduli are expressed in terms of the high-parent-temperature plateau G_∞ of the sample-to-sample mean athermal shear modulus (as seen in Fig. 1), frequencies in terms of c_∞/a_0 where $c_\infty \equiv \sqrt{G_\infty/\rho}$ represents the speed of sound of glasses quenched from $T_p > T_{\text{onset}}$, and $\rho = mN/V$ is the mass density.

Glassy samples were made by first equilibrating independent liquid states at a very high temperature of

$T = 2.5 > T_{\text{onset}}$, and then instantaneously quenching those states to zero temperature using a standard conjugate gradient algorithm. We prepared 320 000, 80 000, 20 000, 5000, and 1250 glassy samples of $N = 2048$, 8192, 32 768, 131 072, and 524 288 particles, respectively. Ensemble sizes were chosen such that the quality of the statistics is independent of system size.

In what follows we discuss the statistical properties of vibrational frequencies ω associated with vibrational modes ψ_ω , which are solutions to the eigenvalue problem

$$\mathcal{M} \cdot \psi_\omega = m\omega^2 \psi_\omega, \quad (1)$$

where $\mathcal{M} \equiv \frac{\partial^2 U}{\partial \mathbf{x} \partial \mathbf{x}}$ is the Hessian of the potential $U(\mathbf{x})$ evaluated at a local minimum, and \mathbf{x} denotes the particles' coordinates. For each member of our ensembles of glassy samples we calculated the lowest 100 vibrational frequencies using ARPACK [40]. A discussion regarding the conditions under which $D(\omega)$ can be cleanly observed, without hindrance by hybridizations with phonons, can be found in Ref. [41].

III. RESULTS

As mentioned above, in Ref. [37] it was shown that glassy samples quenched instantaneously from high parent temperatures T_p feature a density of nonphononic quasilocalized modes $\mathcal{D}(\omega) \sim \omega^\beta$ with $\beta < 4$. In Fig. 2 we show the low-frequency regime of the *total* density of states $D(\omega)$ measured in our glassy samples. Importantly, we note that below the lowest phonon frequency $2\pi c_\infty/L$, the nonphononic and total density of states are the same, i.e., $D(\omega) = \mathcal{D}(\omega)$. Consistent with Ref. [37], we find $\beta \approx 3.65$ for $N = 2048$; however, upon increasing the system size we find $\beta \approx 3.8$ for $N = 8192$, and $\beta \approx 4$ for $N \geq 32\,768$, in agreement with previous results [15,38,42].

What gives rise to this finite-size effect in the exponent β ? We assert below that the finite-size effect seen in the exponent β is the result of soft QLMs not “fitting” well in the simulation box. To establish this, we build on the recent result [36] that the length that characterizes the linear response to local force dipoles agrees well with the core size of QLMs. We thus first aim at measuring the length that characterizes the response to local force dipoles in our glassy samples.

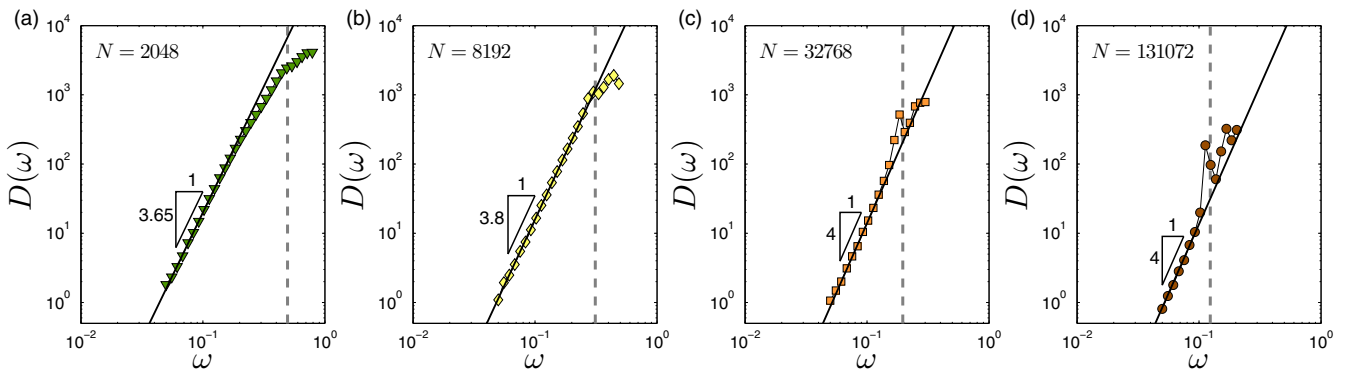


FIG. 2. The low-frequency regime of the total density of states $D(\omega)$, measured in our ensembles of glassy samples quenched from high temperatures of (a) $N = 2048$, (b) $N = 8192$, (c) $N = 32\,768$, and (d) $N = 131\,072$ particles. The vertical dashed lines mark the first phonon frequency $2\pi c_\infty/L$. These data show that $D(\omega) \sim \omega^\beta$ with $\beta < 4$ is a finite-size effect.

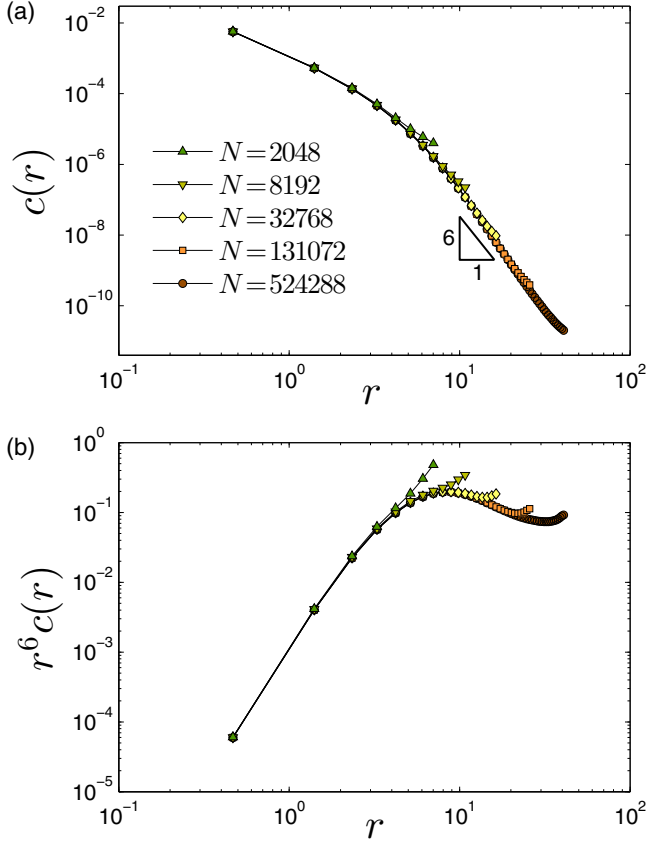


FIG. 3. (a) Decay functions $c(r)$ calculated on the responses to local force dipoles, as defined in the text, plotted against the distance r from the applied force dipole, for different system sizes as reported in the legend. We expect to see a crossover to the continuum r^{-6} scaling at $r \gtrsim \xi_g$, with ξ_g representing the QLMs' core size. (b) Scaling the decay functions by r^6 shows that the smallest systems employed here are too small to exhibit the crossover to the expected continuum scaling.

To this aim, we carry out the following procedure, also employed in Ref. [36]; we randomly select pairs ij of weakly interacting particles and calculate the response $\mathbf{z}^{(ij)}$ as

$$\mathbf{z}^{(ij)} = \mathcal{M}^{-1} \cdot \mathbf{d}^{(ij)}, \quad (2)$$

where the (dimensionless) dipole force $\mathbf{d}^{(ij)}$ is defined as

$$\mathbf{d}^{(ij)} \equiv \frac{\partial r_{ij}}{\partial \mathbf{x}}, \quad (3)$$

and r_{ij} is the pairwise distance between particles i and j . We normalized the fields $\mathbf{z}^{(ij)}$, namely, $\hat{\mathbf{z}}^{(ij)} = \mathbf{z}^{(ij)} / |\mathbf{z}^{(ij)}|$, and, for each other pair of particles $lq \neq ij$, we calculate the projection $\hat{\mathbf{z}}^{(ij)} \cdot \mathbf{d}^{(lq)}$. Finally, we define the decay function $c(r)$ as

$$c(r) \equiv \left\langle \text{median}_{r_{ij,lq} \approx r} [(\hat{\mathbf{z}}^{(ij)} \cdot \mathbf{d}^{(lq)})^2] \right\rangle_{ij}, \quad (4)$$

where $r_{ij,lq}$ stands for the distance between the average position of the pair ij and the average position of the pair lq , and the angular brackets stand for an average over different pairs ij to which a force dipole is applied, and over different glassy samples. We employ the median over pairs lq that are at a distance $\approx r$ from the pair ij —rather than the mean—in order

to suppress large fluctuations that can occur in the projections [43]. We further note that the choice to apply force dipoles only to weakly interacting pairs is motivated in Ref. [36], where it was shown that this strategy produced responses that are generally softer, and thus better representatives of soft QLMs.

Our results are shown in Fig. 3; in Fig. 3(a) we show the raw decay functions $c(r)$ measured as explained above, where the different symbols represent the different system sizes considered. Since $c(r)$ scales as the strain field squared, we expect that at large distances r from the applied force dipole, $c(r)$ should scale as r^{-6} (in three dimensions). In order to better visualize the characteristic decay length, we plot in Fig. 3(b) the product $r^6 c(r)$. This representation of the decay functions reveals that the crossover to the expected continuum behavior is *not seen at all* for $N = 2048$, and only initial signs of this crossover are seen for $N = 8192$. We note that the uprise seen at the very last points of each signal is a result of the periodic boundary conditions employed. Finally, the shape of $c(r)$ depends on several details, in particular the distance to the unjamming transition [10,44,45] as discussed and shown in Refs. [43,46].

The signals for $N > 8192$ indicate that the crossover length scale to the expected continuum behavior—estimated as the maximum of the bump in $r^6 c(r)$ —is about eight particle diameters long, indicating that the effective diameter of QLMs is ≈ 16 particle diameters long [twice the effective radius as extracted from $c(r)$]. Clearly, for $N \lesssim 16^3$ QLMs cannot comfortably fit in the glassy solid. We propose that this explains why finite-size effects in $\mathcal{D}(\omega)$ are observed in these smaller system sizes.

IV. SUMMARY, DISCUSSION, AND OUTLOOK

In this work we have shown that generic computer glasses quenched quickly from high-parent-temperature liquid states feature the universal exponent $\beta = 4$ in their nonphononic density of states $\mathcal{D}(\omega) \sim \omega^\beta$ —if large-enough glassy samples are considered—and that smaller exponents $\beta < 4$ result from a finite-size effect. We further showed that $\beta < 4$ is seen when the system size is comparable to the core size of QLMs (represented by the response to local force dipoles [36]). Our findings correct the misinterpretation of numerical results presented and discussed in Ref. [37], where it was incorrectly concluded that exponents $\beta < 4$ are not a finite-size effect, but rather have to do with the physical process of glass formation by rapid quenches from high-temperature liquid states. Our results also cast doubts on the claims made in Ref. [47], where the change in β induced by deeper supercooling was attributed to the suppression of phononic (plane-wave) modes.

Our findings strongly suggest that the universality of the ω^4 law extends to glasses formed from *any* parent temperature, conditioned that those glasses are sufficiently larger than the core size of QLMs. Importantly, in Ref. [36] it was shown that the core size of QLMs decreases with decreasing parent temperature, explaining why the ω^4 law of the nonphononic density of states can be measured in smaller glassy samples quenched from low parent temperatures, or quenched at slow rates into the glassy phase.

Further support for the physical picture proposed in this work was put forward in Ref. [35], where it was shown that the far, algebraically decaying field of QLMs stabilizes their energetically unstable core. It is therefore reasonable to expect that if QLMs are deprived from their far fields—as expected to occur in small glassy samples—those QLMs will be less stable and have lower frequencies, possibly leading to $\beta < 4$ in their nonphononic density of states.

Finally, we propose that the finite-size effect reported here, in which a system constrains itself by virtue of its small size, giving rise to reduced stability manifested by the appearance of soft modes, might be an instance of a more general phenomenon. For example, in Ref. [48] it was reported that small packings of frictionless hard spheres possess relatively fewer small gaps between nearby particles that are *not* in contact, compared to the abundance of small gaps in larger packings. Since smaller gaps are known to stabilize hard-sphere

packings [48], that phenomenon bears similarities to the reduced stability that small computer glasses possess, as reported here.

In light of the findings reported here, it seems important to explore which additional finite-size effects on QLMs appear in computer glasses, and how those effects influence our insights and conclusions regarding the properties of macroscopic structural glasses.

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