Editors' Suggestion

Unifying Description of the Vibrational Anomalies of Amorphous Materials

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The vibrational density of states $D(\omega)$ of solids controls their thermal and transport properties. In crystals, the low-frequency modes are extended phonons distributed in frequency according to Debye's law, $D(\omega) \propto \omega^2$. In amorphous solids, phonons are damped, and at low frequency $D(\omega)$ comprises extended modes in excess over Debye's prediction, leading to the so-called boson peak in $D(\omega)/\omega^2$ at ω_{bp}, and quasilocalized ones. Here we show that boson peak and phonon attenuation in the Rayleigh scattering regime are related, as suggested by correlated fluctuating elasticity theory, and that amorphous materials can be described as homogeneous isotropic elastic media punctuated by quasilocalized modes acting as elastic heterogeneities. Our numerical results resolve the conflict between theoretical approaches attributing amorphous solids' vibrational anomalies to elastic disorder and localized defects.

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The distribution in the frequency, ω, of the vibrational modes of solids, or density of states (DOS), is a fundamental material property controlling, e.g., their specific heat and thermal conductivity [1,2]. At small frequencies, crystals' DOS is populated by phonons (plane waves) and follows Debye's law, $D(\omega) = A_D\omega^2$. The vibrational properties of amorphous materials deviate from that of crystals in several aspects. First, the reduced density of states $D(\omega)/\omega^2$ exhibits a peak at the Boson peak frequency, ω_{bp}, in the terahertz regime for molecular solids. The boson peak reveals an excess of modes over Debye's prediction. Competing theories have attributed this anomaly to elastic disorder [3–8], localized harmonic [9,10] or anharmonic vibrations [11,12], anharmonic effects [13,14], broadening of the lowest van Hove singularity of the transverse phonon branch [15,16]. Second, the low-frequency DOS of amorphous solids is the superposition [17–20] of extended modes complying to Debye's prediction, $D(\omega) = A_D\omega^2$, and of quasilocalized modes (QLMs) distributed in frequency as $D_{\mathrm{loc}}(\omega) = A_4\omega^4$. Finally, in amorphous solids, the extended low-frequency modes are not phonons. Rather, phonons attenuate while propagating with a frequency-dependent rate, $\Gamma(\omega)$. In the absence of temperature induced anharmonic effects [21], phonons' attenuation rate Γ crossovers from a Rayleigh scattering regime [22], $\Gamma \propto \omega^4$, to a disordered-broadening regime, $\Gamma \propto \omega^2$ [23–27], as the phonon frequency increases, as observed in recent studies [28–30].

Since the vibrational anomalies of amorphous solids occur in different frequency regimes, it is not clear that there should be a relation between them [29,31]. However, there are indications suggesting such a relation. For instance, numerical results indicate a correlation between A_4 and ω_{bp} [31,32]. Furthermore, fluctuating elasticity theory [3–5,33] (FET), in its extended version incorporating an elastic disorder correlation length ξ_e [34,35] (corr-FET) suggests a correlation between ω_{bp} and the attenuation rate of sound waves in the Rayleigh scattering regime, $\Gamma/\omega_{\mathrm{bp}} \propto \gamma(\omega/\omega_{\mathrm{bp}})^4$. Here, γ is a disorder parameter controlling the scaling of the fluctuations $\sigma_\mu^2(N)$ of the shear modulus on the coarse-grained size [3–5,33], $\sigma^2(N)/\mu^2 = \gamma/N$, μ being the average modulus, and $\omega_{\mathrm{bp}} = c_s/\xi_e$ with c_s the sound velocity of transverse waves. A connection between γ and A_4 has also been observed [36,37]. While sound attenuation appears to correlate with the fluctuations of the elastic moduli [17,30], the validity of corr-FET is debated. It has been suggested, for instance, that corr-FET is only qualitatively accurate [29,38] or that corr-FET prediction holds with ω_{bp} replaced by $\omega_0 = c_s/a_0$, with $a_0 = \rho^{-1/d}$ and ρ the number density [30]. Henceforth, it is still unclear if boson peak, quasilocalized modes, and sound attenuation are related.

Here, we introduce and verify via extensive numerical simulations a simple picture relating amorphous solids' vibrational anomalies. First, we validate corr-FET and its proposed connection between boson peak, elastic heterogeneities and sound attenuation. Then, we show that low-frequency corr-FET predictions emerge from the mechanical model introduced by Rayleigh in his seminal work [22], a homogeneous elastic continuum of shear modulus μ_0 punctuated by defects with shear modulus $\mu_0 + \delta\mu_d$, provided that the defects have linear size $\xi_d \propto \xi_e$, constant number density n, and that $\delta\mu_d \propto \mu_0$. Finally, we demonstrate that QLMs satisfy these constraints. Our results clarify that the low-frequency vibrational properties of amorphous solids are those of an elastic continuum punctuated by quasilocalized vibrational modes. Hence, our work establishes a relation between the different vibrational anomalies of amorphous solids and resolves the contrast between theoretical models attributing the boson peak anomaly to elastic disorder and localized defects.

We resort to numerical simulations to investigate vibrational properties and attenuation rate of model amorphous materials, focusing on systems of particles interacting via an Lennard-Jones-like potential $V(r, x_c)$. Here, x_c is a parameter setting the extension of the attractive well [39], which vanishes at $x_c\sigma$. This parameter influences the relaxation dynamics [40] and the mechanical response [36,37,39,41]. We follow the model of Ref. [40]. We simulate systems with a varying number of particles N, up to $N = 8\,192\,000$, in a cubic box with periodic boundary conditions, at fixed number density $\rho = 1.07$. We generate amorphous solid configurations by minimizing, via conjugate gradient, the energy of systems in thermal equilibrium at $T = 4.0\epsilon$, above the glass transition temperature for the considered x_c values [40].

We determine the two parameters entering corr-FET predictions [3–5,33–35], ξ_e and γ, investigating the dependence of the elastic properties on the system size as well as the dependence of coarse-grained elastic properties on the coarse-graining length. We have found these two approaches [36,42–44] to give consistent results, as we recap in the Supplementary Material [45]. Importantly, we find that a single length scale [36,44] characterizes the dependence the shear modulus fluctuations on the system size, so that γ is a nondimensional measure of the correlation volume, $\gamma \propto (\xi_e/a_0)^3$. In Fig. 1(a), we observe the elastic length scale ξ_e, or equivalently γ, to decrease with the attraction range x_c, consistently with previous results [36].

The estimation of ξ_e, γ, and $c_s = \sqrt{\mu/(m\rho)}$ allows us to validate if the boson peak frequency scales as $\omega_{\rm bp} \propto c_s/\xi_e$, as predicted by corr-FET for stable glasses [46]. To check this prediction, we evaluate $D(\omega)$ via the Fourier transform of the velocity autocorrelation function of $N = 256\,000$ particle systems, and $\omega_{\rm bp}$ via the scaling collapse of Fig. 1(b). The inset shows that the boson peak frequency is proportional to $c_s/\xi_{\rm e}$, validating corr-FET.

Considering that $\gamma \propto \xi_e^3$, FET [3–5,19] and corr-FET [4,5,34,35] predictions for the attenuation rate (see [45]) can be summarized as follows:

$$\Gamma\frac{\omega_0^3}{\omega^4} \propto \begin{cases} \gamma & \text{FET} \\ \gamma^2 \propto \left(\frac{\omega_0}{\omega_{\rm bp}}\right)^6 & \text{corr-FET} \end{cases}. \quad (1)$$

We remark that these predictions concerns the harmonic $T \to 0$ limit of our interest. At low temperature, $\Gamma \propto T\omega^2$ for

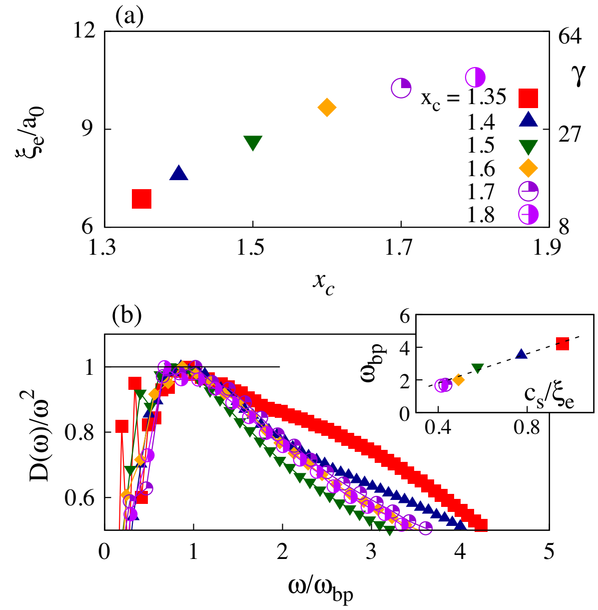

FIG. 1. (a) Dependence of elastic disorder correlation length $\xi_e \propto \gamma^{1/3}$ and disorder parameter γ on x_c, a parameter controlling the extension of the attractive well. Error bars are smaller than the symbol size. (b) Reduced $D(\omega)$, normalized by its maximum value, as a function of $\omega/\omega_{\rm bp}$. We found $\omega_{\rm bp} \simeq 4.5 c_s/\xi_e$ (inset).

$\omega \to 0$, due to anharmonic effects [24,47,48]. corr-FET can be extended to account for these anharmonic effects [49].

To validate these predictions, we evaluate Γ by exciting [50,51] a transverse acoustic wave with wave vector $\boldsymbol{\kappa}$ in which two among κ_x, κ_y, and κ_z are zero. We then evolve the system in the linear response regime to evaluate the velocity autocorrelation function, which we average over 30 phonons from independent samples for $N \leq 512\,000$, and over 15 phonons for $N \geq 512\,000$. A subsequent fit of this averaged velocity autocorrelation function to $\cos(\omega t)e^{-\Gamma t/2}$ allows extracting attenuation rate Γ and frequency ω as a function of κ.

The normalized attenuation rate $\Gamma\omega_0^3/\omega^4$ attains a constant value at low frequency, demonstrating the existence of a well-defined Rayleigh scattering regime, as illustrated in Fig. 2(a). This finding [28–30] demonstrates that, in this regime, anisotropic long-range spatial correlations in the elastic moduli [52,53] do not influence sound damping [50,54]. We test FET and corr-FET predictions, Eq. (1), by plotting $\alpha\gamma$ and $\beta\gamma^2$, with α and β constants and γ as in Fig. 1(a). Corr-FET correctly predicts the relation between sound attenuation and boson peak frequency. Furthermore, Fig. 2(a) indicates that Rayleigh's scattering regime sets in at a frequency smaller but close to $\omega_{\rm bp}$, confirming another corr-FET prediction.

Previous works did not support corr-FET. References [29,55] tested it by measuring the fluctuations of coarse-grained elastic constants defined via the so-called fully local approach [56]. We speculate this approach leads

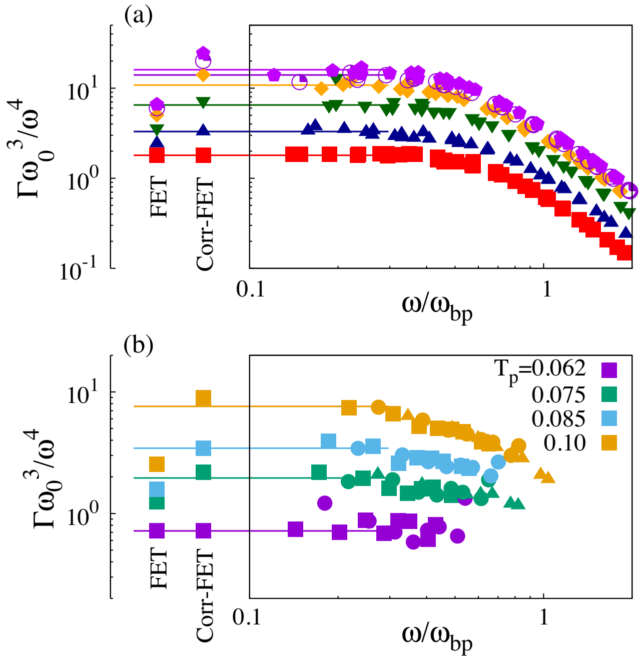

FIG. 2. (a) The frequency dependence of the scaled attenuation rate is consistent with corr-FET as concern the limiting low-frequency value, $\propto \gamma^2$. We combine data for $N = 32$ k, 64 k, 256 k, 512 k, 2048 k, and 8192 k. Symbols are as in Fig. 1. (b) Analogous results are obtained investigating the scaled sound attenuation rate of amorphous solid configurations prepared minimizing the energy of ultrastable liquids in equilibrium at temperature T_p, below the mode coupling one. Data are from Refs. [29,55], to which we refer for further details. Symbols identify the system size: 192 k (squares), 96 k (circles), 48 k (triangles).

to unreliable results as it fails to recover self-averaging [42]. Reference [30] supported the validity of FET, rather than of corr-FET, studying sound attenuation and elastic properties as a function of a parameter artificially affecting the prestress contribution to the dynamical matrix of a given system. We suspect this approach breaks the relation $\gamma \propto \xi_e^3$, leading to changes in γ at constant ξ_e, but the matter deserves further investigation.

To further support our findings, we consider that corr-FET prediction of Eq. (1) can be tested without the direct measurement of the disorder parameter, but rather inferring it from measurements of the boson peak frequency, as $\gamma \propto (\omega_0/\omega_{bp})^3$. We exploit this result to validate corr-FET against numerical data for the boson peak frequency [29] and sound attenuation [55] of ultrastable glasses. The result of this investigation further support the validity of corr-FET, as we illustrate in Fig. 2(b). The validation of corr-FET is our first main result.

We now turn our attention to the connection between corr-FET and QLMs. Fluctuating elasticity theory has been introduced considering that, in an amorphous material, "it is difficult to distinguish between 'host' and 'defect'" [10]. However, the analysis of the vibrational properties of amorphous materials revealed the existence of QLMs, extended soft mechanical regions that act as structural defects controlling the mechanical response under shear [57] and, possibly, the relaxation dynamics of supercooled liquids [58]. Hence, there could be a relation between FET and QLMs.

We establish this relation within Rayleigh's elastic model [22], an elastic continuum with shear modulus μ_0 punctuated by n defects per unit volume, each defect being a region of linear size ξ_d with shear modulus $\mu_0 + \delta\mu_d$. Within this model, FET disorder parameter [45] results in

$$\gamma \propto (n a_0^3)\left(\frac{\xi_d}{a_0}\right)^3 \frac{\delta\mu_d^2}{\mu_0^2}, \tag{2}$$

and the boson peak frequency is $\omega_{\rm bp} \propto c_s/\xi_d$, so that Rayleigh's seminal result for the attenuation rate [22] of low-frequency phonons, $\Gamma \propto (\delta\mu_d/\mu_0)^2 \xi_d^6 \omega^4$, can be expressed as $\Gamma(\omega)(\xi_d/c_s) \propto \gamma(\omega\xi_d/c_s)^4$.

Corr-FET and the defect model are consistent in their predictions for the boson peak frequency if

$$\xi_d \propto \xi_e. \tag{3}$$

If this relation holds, then the models are consistent in their predictions for the attenuation rate if Eq. (2) is satisfied, or equivalently, given Eq. (3), if $n\delta\mu^2/\mu_0^2 = \text{const}$. This occurs, e.g., if

$$n = \text{const} \tag{4}$$

$$\delta\mu_d \propto \mu_0. \tag{5}$$

We now show that QLMs satisfy Eqs. (3)–(5).

Equation (4) is suggested by previous studies [36]. Here, we validate it investigating QLMs' density of states to estimate their number density, n. We determine $D_{\rm loc}(\omega)$ via the direct diagonalization of the Hessian of a small systems, $N = 4000$, to lift the minimum phonon frequency [59], $\propto c_s/N^{1/3}$, and make the low-frequency spectrum predominantly populated by localized modes. We average our results over at least 10^4 independent realizations. Figure 3(a) shows that data for different potentials collapse when $D_{\rm loc}$ is nondimensionalized resorting to the boson peak frequency. Assuming that $\omega_{\rm bp}$ is the maximum QLMs' frequency, this result indicates that

$$D_{\rm loc}(\omega) = A_4\omega^4 = \frac{5n}{\omega_{\rm bp}}\left(\frac{\omega}{\omega_{\rm bp}}\right)^4, \qquad \omega < \omega_{\rm bp}, \tag{6}$$

where $n = \int_0^{\omega_{\rm bp}} D(\omega)d\omega \simeq 0.005$ is the *constant* density of vibrational modes. This result validates Eq. (4). Interestingly, Ref. [31] reported $A_4^{-1/5}/\omega_{\rm bp} = (5n)^{-1/5} \simeq 2.1$ (see their Fig. 6), in quantitative agreement with our

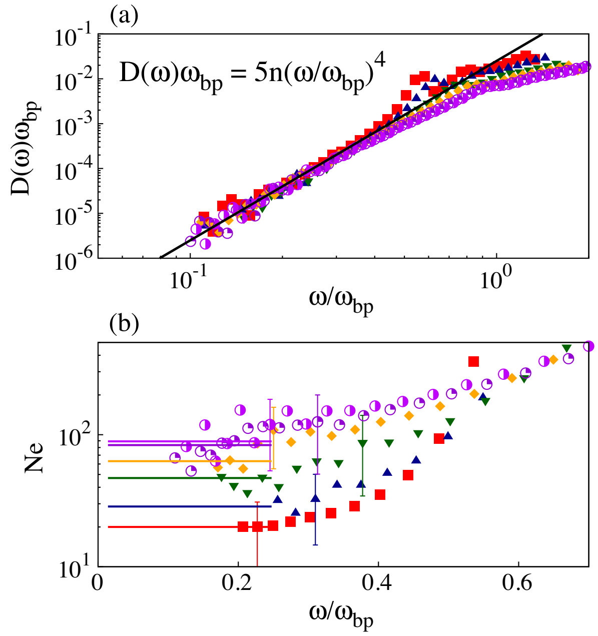

FIG. 3. (a) Scaling of the low-frequency density of states. (b) QLMs volume estimated by the low-frequency limit of Ne with e the participation ratio and N the system size. Representative error bars are shown. The full lines correspond to $\xi_e^3 \propto \gamma$. For both panels $N = 4000$ and data are averaged over at least 10^4 realizations. Symbols indicate different x_c values as in Fig. 1.

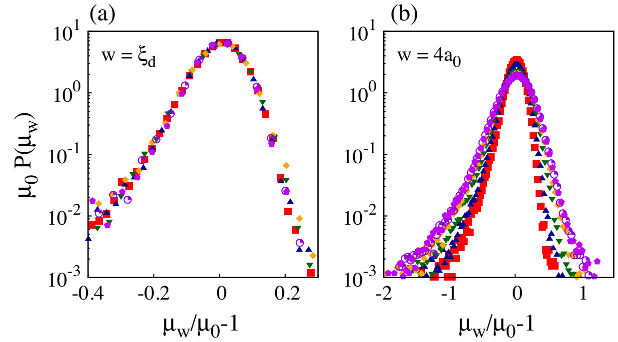

FIG. 4. Probability distribution of the local shear modulus $\mu_w/\mu_0 - 1$ coarse grained over a length scale w, μ_0 being the average modulus. In (a) the coarse graining length scale equals the defect size, $w = \xi_d = \xi_e$, whose x_c dependence is in Fig. 1(a). In panel (b), $w = 4a_0$. For each x_c, results are obtaining averaging over 50 independent $N = 256\,000$ particles configurations. Symbols are as in Fig. 1.

result, investigating a different system. This suggests n might be a universal constant. We leave to the future the investigation of this intriguing question.

We verify Eq. (3), which is supported by previous investigations [20,37], evaluating QLM size via the mode participation ratio $e = (1/N)[\sum_{i=1}^{N}(\vec{u}_i \cdot \vec{u}_i)^2]^{-1}$, where $\vec{u}_i$ is the displacement vector of particle i in the considered mode. The participation ratio is $\mathcal{O}(1)$ for extended modes, and $\mathcal{O}(1/N)$ for localized ones. Hence, Ne estimates the number of particles involved in the mode, and we expect $\xi_d^3 \propto \gamma \propto \lim_{\omega\to 0} Ne(\omega)$ if Eq. (3) holds. While our Ne data are noisy, despite our significant statistics, they are indeed compatible with this expected scenario as we demonstrate in Fig. 3(b), where full lines correspond to $a\gamma(x_c)$, with a constant.

Since QLMs correspond to soft mechanical regions, we assume that their typical shear modulus is encoded in the left tail of the distribution of the shear modulus coarse grained at the QLMs' size ξ_d, $P(\mu_{\xi_d})$, which is known to be anomalous [30,42]. Hence, if Eq. (5) holds, the left tails of distributions corresponding to different x_c should collapse, when the distributions are plotted versus $\mu_{\xi_d}/\mu_0 - 1$, with μ_0 the average shear modulus. To check this prediction, we associate to each particle a shear modulus, taking into account the nonaffine contribution, and coarse grained it at different length scales [45]. Figure 4(a) shows that the left tails of the distribution of the shear modulus coarse grained at $w = \xi_d$ collapse—indeed, the whole distribution does it. We remark that this collapse is not trivial as it occurs at a coarse-graining length scale at which the distributions are far from being Gaussian. Indeed, the collapse does not occur at smaller coarse-graining length scale, as we illustrate in Fig. 4(b) where we fix, as an example, $w = 4a_0$.

Overall, these results show that QLMs satisfy Eqs. (3)–(5) and demonstrate that corr-FET predictions are recovered within a defect picture, if defects are identified with the QLMs. Hence, elastic disorder and defect based interpretations of the anomalous vibrational properties of amorphous materials are intimately related rather than contrasting. This result is our second major finding.

To rationalize this result, we consider that within Rayleigh model sound attenuation is strongly dependent on the defect size, $\Gamma \propto \xi_d^6$, and the deviation of the elastic properties of the defects from the average, $\Gamma \propto \delta\mu_d^2$. We thus understand that QLMs dominate sound attenuation as they stand out as the largest and softest elastic heterogeneities [58].

In the defect picture, the density of states of amorphous materials is approximated by

$$D(\omega) = n\frac{\omega^4}{\omega_{\rm bp}^5}\theta(\omega_{\rm bp} - \omega) + A_D\frac{\omega^2}{\omega_D^3}\theta(\omega_D - \omega) \quad (7)$$

with $\theta(x)$ the Heaviside step function, n weakly system dependent if not constant, and A_D fixed by the normalization constraint. The characteristic QLMs size determines the boson peak, which is therefore not related to the first van Hove singularity of transverse waves [60].

The defect picture does not rely on the introduction of defects of a specific size but rather on the existence of a characteristic size, ξ_d. It is then of interest to consider the distribution of the defect sizes, $P(\xi)$. This distribution can be obtained from the distribution in frequency of the modes, given the QLMs "dispersion relation" $\xi = \xi(\omega)$, one might infer from Fig. 3, as $D_{\rm loc}(\omega)d\omega = P(\xi)d\xi$.

The relation between boson peak, sound attenuation, and QLMs we have established calls for reconsidering previous works relating the elastic properties of amorphous materials to those of disorder mass-spring networks, e.g., see for a review [61]. While elastic disorder induces a boson peak, our results suggest that only elastic networks with a disorder engineered to reproduce the observed connection between boson peak, localized modes and sound attenuation are relevant models for amorphous materials.

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