Disordered Crystals Reveal Soft Quasilocalized Glassy Excitations

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Structural glasses formed by quenching a melt are known to host a population of low-energy quasilocalized (nonphononic) excitations whose frequencies ω follow a universal $\sim\omega^4$ distribution as $\omega \to 0$, independently of the glass formation history, the interparticle interaction potential, or spatial dimension. Here, we show that the universal quartic law of nonphononic excitations also holds in disordered crystals featuring finite long-range order, which is absent in their glassy counterparts. We thus establish that the degree of universality of the quartic law extends beyond structural glasses quenched from a melt. We further find that disordered crystals, whose level of disorder can be continuously controlled, host many more quasilocalized excitations than expected based on their degree of mechanical disorder—quantified by the relative fluctuations of the shear modulus—as compared to structural glasses featuring a similar degree of mechanical disorder. Our results are related to glasslike anomalies experimentally observed in disordered crystals. More broadly, they constitute an important step toward tracing the essential ingredients necessary for the emergence of universal nonphononic excitations in disordered solids.

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Introduction.—It is now established that structural glasses formed by quenching a melt generically feature a population of soft, quasilocalized (nonphononic) excitations (QLEs) [1]. These excitations play key roles in determining various static, mechanical, transport, and dynamic properties of glasses, and possibly of some supercooled liquids as well [2–5]. QLEs have been envisioned, and their importance has been underlined, since the late 1980s by several workers, as described in detail in a recent review [1]. A first-principle understanding of low-energy quasilocalized excitations in structural glasses is still incomplete, despite some recent progress in identifying and solving potentially relevant mean-field models [6–9].

Intriguingly, the vibrational density of states (VDOS) $\mathcal{D}(\omega)$ of low-energy QLEs—of vibrational frequency ω—has been shown to follow a universal law in structural glasses, of the form $\mathcal{D}(\omega) \sim \omega^4$, independently of the interparticle interaction potential [10,11], spatial dimension [12], or the glass formation protocol [13–15]. Does this nonphononic quartic law remain valid in a broader class of disordered solids, beyond structural glasses formed by quenching a melt? Can it be observed in disordered solids featuring some long-range order? In this Letter, we address these questions by studying the statistical, structural, and mechanical properties of soft excitations that emerge in a class of disordered crystals. To this aim, we study a variant of a model put forward by Barrat and coworkers [16–18] in which microscopic disorder can be gradually introduced into an initially perfectly crystalline solid, until the occurrence of a global amorphization transition of the disordered crystal into a glassy state. Related procedures were carried out in other systems [19–22].

In this Letter, we employ a three-dimensional disordered crystal model in which the external pressure is fixed under variations of the microscopic disorder, controlled by a dimensionless parameter $\delta \in [0, 1]$ ($\delta = 0$ corresponds to a perfect crystal). Additional model details appear below and in [23]. After identifying the amorphization transition point, we study the vibrational spectra of disordered crystals *prior* to amorphization, i.e., in states featuring finite long-range crystalline order. We find that these disordered crystals host a population of QLEs whose frequencies obey the universal $\sim\omega^4$ nonphononic law. We thus establish that the degree of universality of the nonphononic quartic law of QLEs in disordered media extends beyond structural glasses quenched from a melt. These results also shed new light on low-temperature, glasslike thermodynamic, transport, and vibrational anomalies observed in laboratory disordered crystals [34–44].

Finally, we assess the degree of mechanical disorder of these disordered crystals, and compare their relative abundance of QLEs to that found in structural glasses featuring similar degrees of mechanical disorder. This analysis reveals, surprisingly, that these disordered crystals host many more QLEs as compared to structural glasses. We also show that the micromechanical properties of QLEs in disordered crystals, e.g., their degree of spatial localization and stability bounds, reveal striking similarities to the corresponding QLEs' properties in structural glasses.

Disordered crystals model and the amorphization transition.—We employ a binary system of N Lennard-Jones particles among which a fraction x is of one species ("small" particles) and the remaining are of another species ("large" particles). One species may be regarded as "impurity atoms" relative to the other species ("host atoms"), as is extensively used in laboratory disordered crystals. The parameter δ determines the effective interaction of the two species, such that $\delta = 0$ corresponds to a monodisperse system, while $\delta = 1$ corresponds to the largest-considered interaction contrast between the two species. Details about the pairwise interaction potential and system sizes employed are provided in [23]. Throughout this Letter, we fix $x = 1/2$, such that $\delta = 1$ corresponds to a 50:50 binary mixture extensively employed in computer glass-forming models. We have verified that the key results reported below do not qualitatively depend on the precise choice of x.

Disordered crystals are generated by first placing the two particle species randomly on a perfect face-centered-cubic (fcc) lattice, and setting $\delta = 0$. Then, for each value of the parameter $\delta > 0$, the potential energy is relaxed under fixed zero confining pressure to obtain a disordered crystal. This energy relaxation step spontaneously generates positional disorder and internal stresses, absent in the reference fcc lattice. Continuously varying δ gives rise to a class of disordered crystals with a varying degree of disorder. Once δ exceeds a critical value—to be determined next—the disordered crystal collapses into a glassy state via a so-called "amorphization transition" [16–18].

To quantitatively identify this transition, we study the behavior of χ, the relative sample-to-sample fluctuations of the shear modulus G, defined as $\chi \equiv \sqrt{N\overline{(G-\bar{G})^2}}/\bar{G}$, where $\bar{\bullet}$ denotes a sample-to-sample average. The quantifier χ was discussed at length in [11,24–27] and was shown to determine wave attenuation rates in the harmonic regime [25,27]. Here, we use a jackknifelike method to approximate χ; see details in [26]. We also consider the ratio $G_{\rm na}/G$ of the nonaffine $G_{\rm na} = V^{-1}((\partial^2 U/\partial\gamma\partial\boldsymbol{x})\cdot\boldsymbol{\mathcal{H}}^{-1}\cdot(\partial^2 U/\partial\boldsymbol{x}\partial\gamma))$ to total shear modulus G. Here, γ is the shear-strain parameter [11], $U(\boldsymbol{x})$ denotes the potential energy as a function of the particles spatial coordinates $\boldsymbol{x}$, and $\boldsymbol{\mathcal{H}}$ is the Hessian matrix. The nonaffine contribution to the shear modulus, $G_{\rm na}$, is intimately related to material disorder, most notably to deviations from crystalline long-range order, and hence is of interest here.

Figure 1 displays our results for χ (main panel) and $G_{\rm na}/G$ (inset) under variations of the control parameter δ. While $G_{\rm na}/G$ varies smoothly and monotonically with δ, $\chi(\delta)$ exhibits a rather sharp peak, which enables the identification of the amorphization transition. The corresponding onset threshold δ_c, which for $x = 1/2$ takes the value $\delta \approx 0.49$, is marked by the vertical dashed line. The black squares pertain to the disordered-crystal ensembles that we study next, for which $\delta < \delta_c$.

To further substantiate the amorphization transition at δ_c and to establish the existence of finite long-range order in our disordered crystals for $\delta < \delta_c$, we consider the pair correlation function $g(r)$ among the "large" particle species; results for $\delta = 0.38, 0.44, 0.49, 0.70$ are shown in Fig. 2. We find that solids with $\delta \lesssim \delta_c$ all feature finite long-range order, indicated by the presence of peaks in $g(r)$ that persist to the largest distances r considered. In particular, long-range order is clearly present in the ensembles pertaining to $\delta = 0.38$ to $\delta = 0.44$, which are

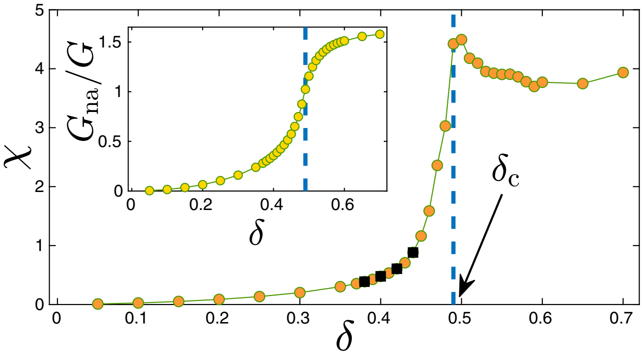

FIG. 1. Dimensionless quantifiers of mechanical disorder across the amorphization transition: the main panel shows the relative sample-to-sample fluctuations of the shear modulus χ; see definition in main text. These data were measured over about 700 independent realizations of systems of $N = 108\,000$ particles at fixed vanishing pressure. The vertical line marks an estimation of the transition point at $\delta_c \approx 0.49$. The black squares mark the disordered-crystal ensembles studied below, namely $\delta = 0.38$, 0.40, 0.42, 0.44, all of which are located *below* the amorphization transition. Inset: the ratio $G_{\rm na}/G$ vs δ; see text for discussion.

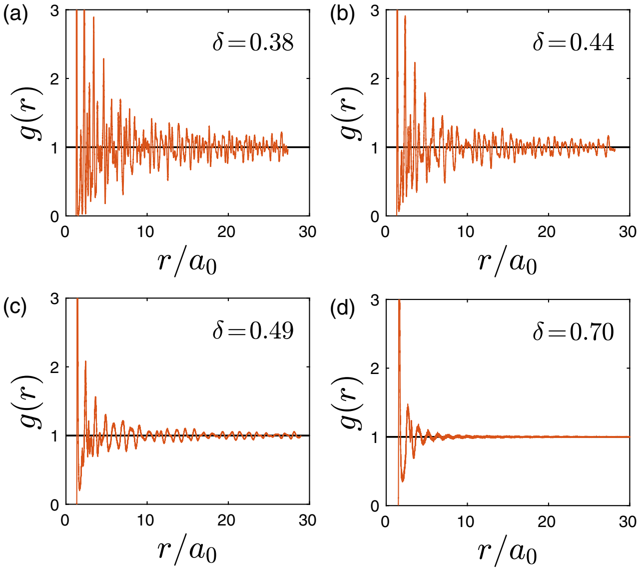

FIG. 2. (a)–(d) Pair correlation functions $g(r)$ between the "large" particle species (see Ref. [23] for details), measured for ensembles of disordered solids of $N = 108\,000$ particles created with different δ values, as indicated in the legends. r is normalized by the interparticle distance $a_0 \equiv (V/N)^{1/3}$, where V is the solid's volume. We indeed find that all the way up to the amorphization transition at $\delta_c \approx 0.49$, long-range order persists.

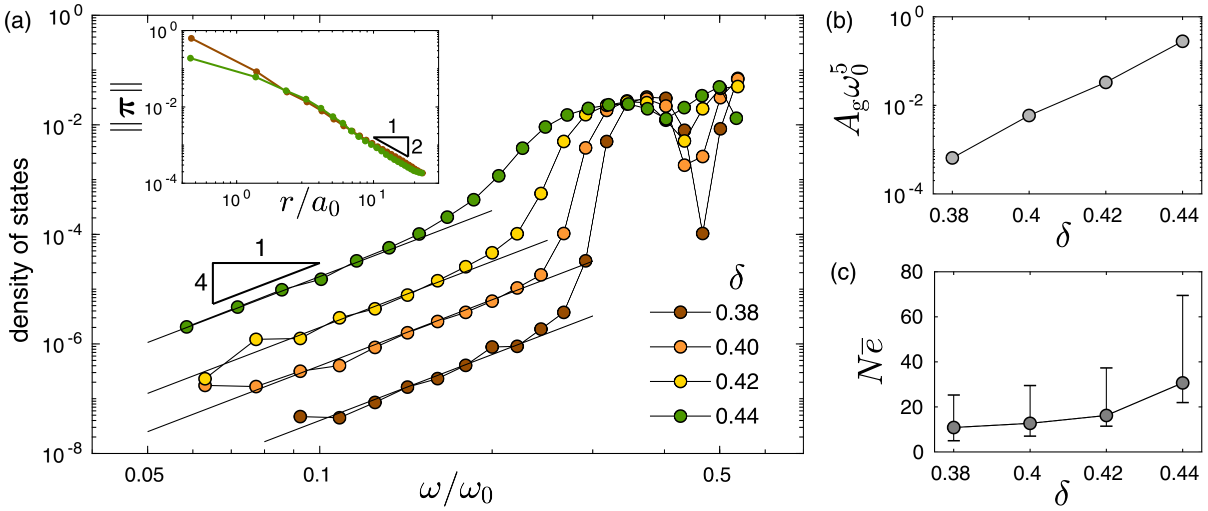

FIG. 3. (a) The vibrational density of states of disordered crystals, for different values of the parameter δ (as indicated in the legend), plotted against rescaled frequency ω/ω_0, where $\omega_0 \equiv c_s/a_0$ with c_s being the shear wave speed. The continuous lines correspond to fits to the $\sim\omega^4$ law, from which the prefactors A_g are extracted. Inset: the spatial decay of (two randomly selected) soft quasilocalized excitations $\boldsymbol{\pi}$ (obtained by the nonlinear framework presented in [23]) follows the expected continuumlike $\sim r^{-2}$ scaling in three dimensions; the colors match the δ values in the legend. (b) Dimensionless prefactors $A_g\omega_0^5$ vs δ on a log-lin scale. (c) The products $N\bar{e}$—representing the mean size of the disordered cores of QLEs (e is the participation ratio, defined in the text)—plotted vs δ. The vertical bars cover the 2nd and third quartiles.

studied next. We focus on this intermediate δ range because it is below the amorphization transition, yet the disordered crystalline states in this range feature disorder levels (cf. Fig. 1) that make the identification of glassy properties quantitatively and computationally feasible.

Universal nonphononic VDOS.—To study QLEs in disordered crystals, it is important to select sufficiently small systems such that a phonon-free frequency window opens below the first shear-wave frequency, as explained at length in [1]. Furthermore, since QLEs that emerge in structural glasses are characterized by a core size ξ_g—usually on the order of a few interparticle distances [1,14]—it is also important that the linear system size is chosen to be sufficiently larger than ξ_g, in order for QLEs to fit comfortably in the simulation box [1,45]. To accommodate these two requirements, we opt for studying emergent QLEs in disordered crystals of $N = 1372$ particles. We prepared 667 900, 297 600, 150 000, and 109 600 independent realizations of disordered crystals with $\delta = 0.38$, 0.40, 0.42 and 0.44 (all below δ_c), following the protocol described above and in [23]. For each disordered crystal, we calculated the first 30 nontrivial vibrational modes $\boldsymbol{\psi}_\omega$ as defined in [23].

The resulting VDOS are shown in Fig. 3(a), which presents a key result of this Letter. We find that disordered crystals featuring finite long-range order host a population of quasilocalized modes whose frequencies obey the universal $\sim\omega^4$ law. We next turn to discussing the nonuniversal prefactor A_g in the full expression of the VDOS, $\mathcal{D}(\omega) = A_g\omega^4$. The prefactor A_g, of dimension $[\text{time}]^5$, serves as an indicator of the abundance of soft QLEs [13,46]. As such, it was the focus of several recent studies [11,14,26,47,48]; notably, A_g strongly correlates with the tensile fracture toughness of glassy solids [47]. The prefactors A_g of our disordered crystals—obtained here by fits to a $\sim\omega^4$ power law at low frequencies (straight lines)—are reported in Fig. 3(b). We find an approximate exponential dependence of A_g on the parameter δ, reminiscent of the Boltzmann-like dependence of A_g on the *equilibrium* parent temperature of structural glasses quenched from deeply supercooled liquids [14,49]. A similar exponential decay of A_g was also observed recently in a mean-field spin-glass model [6].

QLE properties.—We next study some of the structural properties of low-frequency QLEs that emerge in our disordered crystals. To this aim, we invoke the nonlinear framework of [28,29], which provides a robust representation of soft QLEs, denoted as $\boldsymbol{\pi}$ (see also Ref. [23]). We first focus on the spatial decay of QLEs, which is presented in the inset of Fig. 3(a) for two randomly selected nonlinear modes. It is observed that similarly to QLEs in structural glasses [1], QLEs in our disordered crystals feature a disordered core of a few interparticle distances, followed a power law $\sim r^{-2}$ tail (i.e., $\sim r^{-\mathrm{đ}+1}$ in three dimensions, $\mathrm{đ} = 3$).

The degree of spatial localization of QLEs is further discussed next, by considering the participation ratio e, defined as $e(\boldsymbol{\pi}) \equiv N^{-1}(\sum_i \boldsymbol{\pi}_i \cdot \boldsymbol{\pi}_i)^2/\sum_i(\boldsymbol{\pi}_i \cdot \boldsymbol{\pi}_i)^2$. Here, $\boldsymbol{\pi}_i$ denotes the đ-dimensional vector of Cartesian components pertaining to the ith particle of a QLE $\boldsymbol{\pi}$. The participation ratio $e(\boldsymbol{\pi})$ quantifies the degree of spatial localization of $\boldsymbol{\pi}$: $\boldsymbol{\pi}$'s that are localized on a core of N_c particles follow $Ne \approx N_c$, whereas spatially extended $\boldsymbol{\pi}$ follow $Ne \sim N$ (or $e \sim 1$). In Fig. 3(c), we plot the factored means $N\bar{e}$ vs δ; here, $\bar{e}$ was calculated over 1000 QLEs (for each value of δ) in systems of $N = 16384$. We find that moving away from the amorphization transition, giving rise to less disordered states, leads to a stronger localization of QLEs. This enhanced localization is reminiscent of the stronger localization of QLEs observed in structural glasses with increasing glass stability, as shown in, e.g., [14,15]. In addition, we find that QLEs in our disordered crystals satisfy similar stability bounds as in structural glasses [23].

Mechanical-disorder quantifiers.—We turn now to examining how dimensionless quantifiers of mechanical disorder relate to each other in disordered crystals, and how those compare to the corresponding relations in structural glasses. For example, a scaling relation between A_g and the strength of spatial fluctuations of the shear modulus quantified by χ has been demonstrated in computer glasses [26,47,48]. The generality of such relations between different measures of mechanical disorder remains largely unexplored across various classes of disordered solids. To shed light on this important issue, we employ, in addition to our disordered crystals dataset, a dataset of a simple glass-forming model (see Ref. [23] for details) that can be supercooled down to extremely low temperatures using the swap Monte Carlo algorithm [30], spanning a broad range of mechanical disorder.

In Fig. 4(a), we plot $G_{\rm na}/G$ vs χ for both disordered crystals and structural glasses. Interestingly, while the data are somewhat noisy, both datasets appear to follow similar trends. This is an interesting commonality, which emerges for the two classes of disordered solids that are generated by very different preparation protocols. It may suggest that the two quantifiers in both classes are sensitive to mechanical disorder in a similar fashion. The situation is dramatically different, however, when comparing the relation between the prefactor A_g—made dimensionless by multiplying it by ω_0^5—and χ, considered in both disordered crystals and structural glasses, as done in Fig. 4(b). Here, we find that these two classes of disordered solids exhibit very different $A_g\omega_0^5$–χ relations. Most notably, for similar values of the dimensionless prefactor $A_g\omega_0^5$, which encodes information about both the number and characteristic frequency of QLEs [14,46], χ of disordered crystals is significantly smaller. Put differently, we observe that disordered crystals appear to feature many more QLEs than expected from the $A_g\omega_0^5$–χ relation of structural glasses. Finally, we note that in a recent work it was found that χ of a broad variety of (computer) structural glasses appears to be bounded from below by $\chi_0 \approx 0.85$ [50], marked by the vertical, dash-dotted line in Fig. 4(b). Interestingly, in our disordered crystals, for the range of δ studied here, χ breaks this apparent bound by a significant factor.

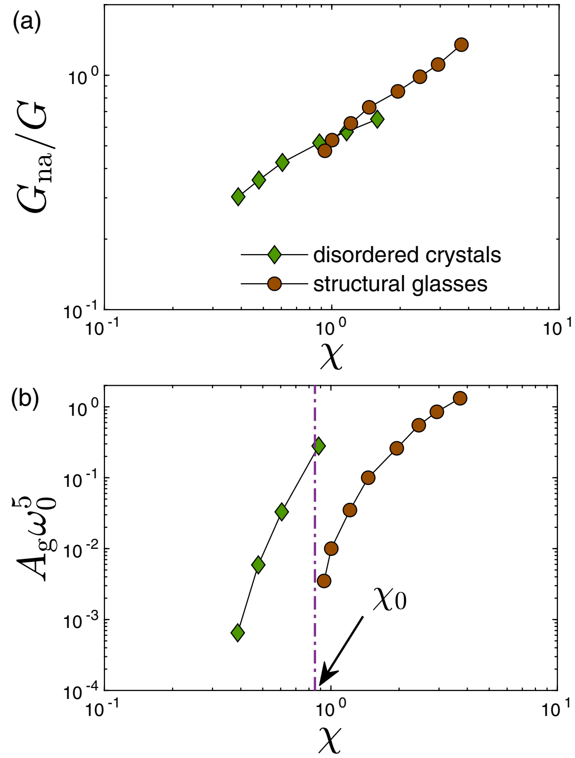

FIG. 4. Relations between quantifiers of mechanical disorder. (a) $G_{\rm na}/G$ vs χ for disordered crystals and structural glasses, as indicated by the legend. The two datasets appear to approximately form a single function; see text for further discussion. (b) The dimensionless nonphononic VDOS prefactor $A_g\omega_0^5$ vs χ for disordered crystals and structural glasses [the same symbols as in panel (a)]. In sharp contrast to the results of panel (a), we find that the two classes of disordered solids follow very different curves. In particular, the disordered crystals appear to have anomalously low χ values relative to the number of QLEs they host.

Discussion and outlook.—We studied the emergence of glasslike properties in disordered crystals, most notably the emergence of low-energy, quasilocalized excitations (QLEs), whose existence has been recently established in structural glasses quenched from a melt [1,10,12,13]. By continuously varying the degree of mechanical disorder in a disordered crystals model that features an amorphization transition, we found that *below* the transition—where the disordered crystals feature finite long-range order—QLEs emerge and their frequencies follow the universal $\sim\omega^4$ nonphononic VDOS, as in many classes of structural glasses [1,10,12,13] and other models [6,7,9,51–54]. As such, our results extend the degree of universality of the ω^4 law to include disordered crystals.

We showed that the nonuniversal prefactor A_g of the universal $\sim\omega^4$ VDOS, which is related to the number of QLEs [14], varies substantially with a control parameter of the model, similarly to an apparently related variation observed in structural glasses [14,15]. We further found that the micromechanical properties of QLEs in disordered crystals, e.g., their spatial localization properties, resemble those observed in structural glasses, and that they satisfy a similar stability bound. Finally, considering interrelations between different quantifiers of mechanical disorder in both disordered crystals and structural glasses, striking similarities and differences emerged. We discovered that the relation between a measure of elastic nonaffinity, G_{na}/G, and a measure of shear modulus fluctuations, χ, appears to form a similar trend for both disordered crystals and structural glasses. On the other hand, not only disordered crystals feature χ values lower than the lowest value ever obtained in structural glasses [50], but their $A_g(\chi)$ relation also implies that they host many more QLEs than the corresponding structural glasses of the same χ.

The extended validity of the universal glassy $\sim\omega^4$ VDOS that includes also disordered crystals, as established here, appears to be intimately related to known glasslike anomalies experimentally observed in laboratory disordered crystals at cryogenic temperatures [34–39,41–44]. Most notably, our findings regarding the low-frequency $\sim\omega^4$ VDOS appear to be related to the existence of a $\sim T^5$ contribution to the low T specific heat in disordered crystals [39,41–44], in addition to a $\sim T$ contribution associated with two-level systems [55–57] and Debye's $\sim T^3$ contribution of phonons [58]. In particular, as the prefactor of the $\sim T^5$ contribution is predicted to be determined by A_g, theoretical models such as the one studied here could guide experiments and explain the to-be-observed variation of the low T specific heat in disordered crystals as a function of the degree of disorder, controlled by the species concentration x. Our findings may also be related to low-temperature anomalies in the heat conductivity [38] and vibrational anomalies, such as the emergence of a Boson peak [43], documented in disordered crystals. Overall, our findings support the experimentally inferred suggestion that the absence of long-range order is neither sufficient nor necessary for the existence of the low-energy excitations [37,38].

To more quantitatively explore the relations between the model studied here and laboratory disordered crystals, future work should realistically model the interactions between different species (e.g., between impurity and host atoms, captured here by the parameter δ) and vary the species concentration x (fixed here at $x = 1/2$, but see preliminary data for varying x in [23]), which is the key control parameter in laboratory binary disordered crystals. In particular, as the prefactor of the $\sim T^5$ contribution to the specific heat would be proportional to the prefactor A_g of the $\sim\omega^4$ VDOS, such future studies would need to resolve $A_g(\delta, x)$ [the results for $A_g(\delta, x = 1/2)$, for δ defined in [23], are shown in Fig. 3(b)]. Likewise, extensions to multicomponent disordered crystals can be explored.

Our results also raise basic questions about disordered solids in a broader context. One class of such questions is concerned with the identification of the essential ingredients necessary for the emergence of universal nonphononic excitations in disordered solids. In this context, it would be interesting to explore the relations between our findings and the model of [53], which *a priori* assumes the existence of soft quasilocalized excitations, and that of [59]. Another interesting question is concerned with the origin of the similar trends featured by the relation between G_{na}/G and χ in both disordered crystals and structural glasses, observed in Fig. 4(a), which is yet another topic for future investigation.

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