Boson-peak vibrational modes in glasses feature hybridized phononic and quasilocalized excitations

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A hallmark of structural glasses and other disordered solids is the emergence of excess low-frequency vibrations, on top of the Debye spectrum $D_{\text{Debye}}(\omega)$ of phonons ($\omega$ denotes the vibrational frequency), which exist in any solid whose Hamiltonian is translationally invariant. These excess vibrations — a signature of which is a THz peak in the reduced density of states $D(\omega)/D_{\text{Debye}}(\omega)$, known as the boson peak — have resisted a complete theoretical understanding for decades. Here, we provide direct numerical evidence that vibrations near the boson peak consist of hybridizations of phonons with many quasilocalized excitations, the latter were recently shown to generically populate the low-frequency tail of the vibrational spectra of structural glasses quenched from a melt and of disordered crystals. Our results suggest that quasilocalized excitations exist up to and in the vicinity of the boson-peak frequency, and hence constitute fundamental building blocks of the excess vibrational modes in glasses.

I. INTRODUCTION

The behavior of many mechanical, transport and thermodynamic properties of solids can be explained based on their vibrational spectra [1]. Examples include the specific heat [1], heat transport [2], elastic moduli [3, 4], and nonlinear failure mechanisms [5, 6]. Unlike crystalline materials, for which the low-frequency spectrum is well-understood [1], there are still open questions regarding the nature of low-frequency vibrational excitations in disordered solids [7–22]. In particular, disordered solids feature excess low-frequency modes on top of the Debye phononic spectrum $D_{\text{Debye}}(\omega) \sim \omega^{d-1}$ (here $\omega$ denotes the vibrational frequency and $d$ is the dimension of space), which stem from their intrinsic structural disorder and micromechanical frustration [23].

Observing the abundance of excess low-frequency excitations in disordered solids is commonly achieved, especially experimentally, by dividing the measured vibrational density of states (VDoS) $D(\omega)$ by the expectation $D_{\text{Debye}}(\omega)$ based on Debye’s theory of low-frequency phonons. The reduced VDoS $D(\omega)/D_{\text{Debye}}(\omega)$ generically exhibits a peak — commonly referred to as the boson peak — at a characteristic frequency commonly referred to as the boson-peak frequency $\omega_{\text{BP}}$, usually in the THz range [16, 24].

Several explanations for the emergence of the boson peak have been suggested in the literature in the past decades. These include effects of the unjamming criticality [18, 25, 26], a broadening of the so-called van-Hove singularity [15], effects of elastic-moduli spatial fluctuations [17, 27], and others [11, 13, 14, 28, 29]. We highlight here in particular Refs. [8–10], where it was suggested that quasilocalized excitations (QLEs) play a crucial role, not just in the low-frequency ($\omega \to 0$) tail of the VDoS, but also in the physics of the boson peak at higher frequencies.

In recent years, computational studies have established many of the generic properties of QLEs; in particular, it has been shown that QLEs populate the low-frequency tail of structural glasses [30], independently of the details of the interparticle potential [31] or the glass formation protocol [32, 33]. In a particular range of glass-sample sizes [34], QLEs assume the form of (quasilocalized) vibrational modes. In addition, it has been demonstrated that these low-frequency excitations follow a universal nonphononic VDoS, scaling as $D_{\text{ql.f}}(\omega) \sim \omega^4$ [20] in any spatial dimension $d \geq 2$ [35]. Finally, the quasilocalized nature of QLEs has been elucidated as they were shown to feature a disordered core of linear size $\xi$ (of a few atomic distances) accompanied by long-range displacement fields that decay as $1/r^{d-1}$ [20]. A review of past and recent efforts to understand the emergent statistics and properties of QLEs can be found in [20].

However, possible relations between QLEs and vibrations at higher frequencies, especially at and in the vicinity of the boson-peak frequency, have not yet been fully elucidated. In fact, in Ref. [21], it was argued that vibrations around $\omega_{\text{BP}}$ — coined ‘stringlets’ therein — are distinct from QLEs (termed ‘four-leaf-like’ vibrations in [21], due to their quadrupolar far fields), the latter presumably only emerge at lower frequencies $\omega \ll \omega_{\text{BP}}$.

Here, we employ numerical simulations of a two-dimensional (2D) glass former (see model details in Sect. II) and a variety of recently developed tools to demonstrate that harmonic vibrations at the vicinity of the boson-peak frequency $\omega_{\text{BP}}$ consist of hybridization (mixing) of QLEs and phonons. In particular, this is achieved by utilizing the framework put forward in Ref. [36], which allows to effectively de-hybridize individual QLEs from hybridized vibrations. Our findings support the assertion of Refs. [8–10] that QLEs are the
II. GLASS-FORMING MODEL

We employed the same well-studied two-dimensional \( d = 2 \) glass forming model of Ref. [21]. This model is composed of a 50:50 binary mixture of ‘large’ and ‘small’ particles of equal mass, interacting via a purely repulsive \( \sim r^{-10} \) pairwise potential \( r \) denotes the pairwise distance between particles) that is smoothed up to two derivatives at some cutoff pairwise distance. The size ratio between the effective diameters of the ‘large’ and ‘small’ particles is 1.4, and the number density is set to \( N/V = 0.86 \) (in simulation units). Further details about the interaction potential and the units employed can be found, e.g., in Ref. [37].

We prepared 1000 independent glassy samples of \( N = 8100 \) particles, using the aforementioned glass-forming model. This ensemble size is required to ensure the statistical convergence of the VDoS. The computer glass transition temperature of this model is estimated as \( T = 0.5 \) (in simulation units). We prepared glasses by first equilibrating the system at high-temperature liquid states with some future research questions.

III. DEFINITION AND CALCULATION OF QUASILOCALIZED EXCITATIONS

Previous work on soft excitations in glasses [36, 39, 40] has demonstrated that QLEs can be isolated and dehybridized from other QLEs and phononic excitations using various cost functions, which make use of the known spatial localization properties of QLEs. The arguments of these cost functions are generalized directions \( z \) in the glass’s \( Nd \)-dimensional configuration space.

In what follows, we employ a cost function \( C(z) \) that only depends on the harmonic approximation to the potential energy of the glass [36]. It reads

\[
C(z) = \frac{z \cdot \mathcal{H} \cdot z}{\sum_{(ij)} z_{ij} \cdot z_{ij}},
\]

A. Vibrational spectrum

In Fig. 1, we present key features of the vibrational spectrum of the employed glass-forming model. The full VDoS, obtained by diagonalizing the Hessian matrix (see definition below), is shown in Fig. 1a; the dashed vertical line (also in Figs. 1b-c) marks the boson-peak frequency, estimated following [21] as \( \omega_B \approx 1.37 \). The latter is defined as the maximum of the reduced VDoS \( D(\omega)/\omega \sim D(\omega)/D_{Debye}(\omega) \), shown in Fig. 1c. Figure 1b presents a zoomed-in view of the low-frequency regime of the VDoS of Fig. 1a. The distinct peaks in Fig. 1b correspond to discrete phonon bands, broadened by disorder [34]. Vibrational modes occurring below the lowest-frequency phonon band are typically quasi-localized modes [20]; an example of one with \( \omega \approx 0.11 \) is shown in Fig. 1d. Panels (e) and (f) show examples of a long-wavelength phonon and a boson-peak mode, respectively. While the boson-peak mode (i.e. a mode whose vibrational frequency is close to \( \omega_B \)) in panel (f) is clearly complex, it features a distinct phononic background pattern. To highlight this point, we added to panel (f) a red bar, which corresponds to the wavelength \( 2\pi c_s/\omega_B \approx 21.1 \) (in simulation units) of a plane-wave phonon of frequency \( \omega_B \). The red bar matches well the spatial periodicity of the wave-like pattern seen in the mode’s structure. Similar comparisons are shown in Fig. 3e and in Appendix A.
FIG. 1. (a) The vibrational spectrum $D(\omega)$ of the 2D computer glass former employed in this work. The vertical dashed line marks the boson-peak frequency (defined as shown in panel (c)). (b) A zoomed-in view on the low-frequency regime of the vibrational spectrum shown in panel (a). The vertical green and red lines mark the frequencies of the quasilocalized vibrational mode shown in panel (d), and of the phononic vibrational mode shown in panel (e), respectively. The vertical dashed line marks the boson-peak frequency (cf. panel (a)), populated by vibrational modes of similar spatial features as the one shown in panel (f). Modes are plotted by quivering the eigenfunctions obtained from diagonalizing the Hessian matrix. The red bar in panel (f) represents the phononic wavelength $2\pi c_s/\omega_{BP} \approx 21.1$, which matches well the mode’s wave-like background patterns, and see further examples in Fig. 5 below. (c) The reduced spectrum $D(\omega)/\omega$ is plotted vs. $\omega$; following [21], we estimate the maximum to occur at $\omega_{BP} \approx 1.37$ (vertical dashed line). The faded vertical lines represent the frequencies of the QLEs used in the analysis of Sect. V, see text for additional details.

IV. CONSTRUCTING SUPERPOSITIONS OF QUASILOCALIZED MODES

Given a harmonic vibrational mode $\psi$, i.e. an eigenmode of the Hessian matrix $\mathcal{H}$, we search for a normalized, linear superposition $\phi$ of QLEs of the form

$$\phi = \sum_{\ell} c_\ell \pi_\ell,$$

which has the maximal absolute value of the overlap $\phi \cdot \psi$.

To this aim, we construct another cost function $\mathcal{F}(\{c_\ell\})$ that depends on the coefficients $c_\ell$, which reads

$$\mathcal{F}(\{c_\ell\}) = \frac{(\phi \cdot \psi)^2}{(\phi \cdot \phi)(\psi \cdot \psi)}.$$

By maximizing this cost function with respect to the coefficients $c_\ell$, we obtain the superposition $\phi^\dagger$ that has the largest overlap with the vibrational mode $\psi$. In what follows, we study the properties of the QLE-constructed modes $\phi^\dagger$ and their similarity (or lack thereof) to vibrational modes at the vicinity of the boson-peak frequency $\omega_{BP}$.

V. RESULTS

Our main goal is to assess whether QLEs play the role of fundamental building blocks of vibrational modes in the vicinity of the boson-peak frequency $\omega_{BP}$. To this aim, we show in the top row of Fig. 3 three vi-
bational modes whose frequencies dwell in the vicinity of the boson-peak frequency $\omega_{BP}$. The expected phononic wavelength, assuming the plane-wave relation $2\pi c_s/\omega_{BP} \approx 21.1$, is represented by the red bar in Fig. 3c. We use these three boson-peak vibrational modes to detect 31 QLEs as described in Sect. III. The frequencies of the detected modes [42] are represented by the faded vertical lines shown in Fig. 1c; they clearly dwell at and in the vicinity of the boson-peak frequency $\omega_{BP} \approx 1.37$. The highest and lowest frequency QLEs detected are shown in Appendix B. We next use the extracted set of QLEs (“library/catalog”) to construct the fields $\phi\dagger$ following the procedure explained in Sect. IV; the constructed fields $\phi\dagger$ are shown in the bottom row of Fig. 3.

As expected, the superpositions $\phi\dagger$ are unable to generate short-wavelength phononic patterns as seen in the spatial structure of the boson-peak vibrations of Figs. 3a-c. However, the disordered, highly nonaffine cores in $\phi\dagger$ remarkably match the same patterns as seen in the harmonic vibrational modes. Since our QLE-detection scheme is not fully exhaustive, some highly nonaffine parts of the vibrational modes are absent in the reconstructed fields. Nevertheless, our results clearly indicate that vibrational modes near the boson-peak frequency consist of hybridized phonons and QLEs (see also a comparison to lower frequencies, presented in Appendix A). This is the main result of this contribution. It is important to note that the boson peak modes analyzed in Fig. 3 were extracted from a randomly selected glass sample, highlighting the generic nature of our main result.

VI. EXCESS VIBRATIONAL MODES

While our QLE-detection scheme may not be entirely exhaustive, we can assess how the number of excess modes as gleaned from the bare VDoS of our computer glasses compares with the number of QLEs we have detected. To this aim, we plot in Fig. 4a the cumulative VDoS $F(\omega) \equiv \int_0^{\omega} D(\omega')d\omega'$ of our computer glass ensemble. Next, we invoke Debye’s theory of phonons [1] (which is a continuum approximation) and superimpose in Fig. 4a the cumulative phononic density of states $F_D(\omega)$ up to frequency $\omega$ in a system of linear size $L$,

$$F_D(\omega) = \int_{2\pi c_s/L}^{\omega} \frac{\omega' - (2\pi c_s/L)^2}{2\pi c_s/L} d\omega' = \frac{\omega^2}{\omega_D} - \omega_D^{-1} \omega - \frac{(2\pi c_s/L)^2}{2\pi c_s/L}.$$  (5)

The latter is plotted as the dashed line in Fig. 4a (recall that our glasses are two-dimensional), and note that the lower integration limit in Eq. (5) is the minimal phonon frequency in a system of linear size $L$. At low frequencies, the two curves overlap, and before $\omega_{BP}$ (marked by the highlighted vertical dashed lines in Fig. 4) the curves depart from each other, indicating the emergence of excess vibrational modes on top of phonons.

In Fig. 4b, we show (dashed black line) the difference $F(\omega) - F_D(\omega)$ [43], multiplied by $2N$, which quantifies the number of excess modes up to frequency $\omega$ in a single glass sample. We superimpose on it (thin red line) the number of QLEs, up to frequency $\omega$, obtained using the QLE-detection method applied as in Fig. 3 above. The two curves reveal good quantitative agreement. In particular, note that our QLE-library contains modes with frequencies up to about $\omega \approx 1.8$ (see the highest frequency QLE of our library in Fig. 6b below). On average, the expected number of excess modes up to $\omega = 1.8$ is $\approx 28$, according to the data of Fig. 4b, i.e. in good agreement with the actual number of QLEs (31) in our library. This agreement lends quantitative support to our computational approach and conclusions.
FIG. 3. Comparison between vibrational boson-peak modes and the corresponding constructed superpositions of quasilocalized modes. Panels (a)-(c) show vibrational modes $\psi$, whose frequencies $\omega \approx \omega_{BP}$. The red bar shows the expected phononic wavelength, obtained from the plane-wave relation $2\pi c_s/\omega_{BP} \approx 21.1$ (in simulational units) at the boson peak frequency; it matches well the characteristic scale of the wave-like background patterns seen in these boson-peak modes, and see additional examples in Fig. 5 below. Panels (d)-(f) show the modes $\phi$ constructed following the scheme spelled out in Sect. IV — solely from the 31 QLEs detected as explained in Sect. III. Our scheme cannot reproduce the short wavelength phononic background, but captures well the nonaffine parts of the vibrational modes. The values of the overlaps $\psi \cdot \phi$, between the vibrational modes $\psi$ and the constructed fields $\phi$, are 0.49, 0.42 and 0.41 for the pairs of fields in the upper, middle and bottom rows, respectively.
VII. SUMMARY AND OUTLOOK

Using recently developed tools to single out QLEs in model glass formers [36, 39, 40], we provided strong visual evidence that vibrational modes at the vicinity of the boson-peak frequency \( \omega_{\text{BP}} \) in a generic 2D glass former consist of hybridized QLEs and phonons. Our results suggest that the ‘excess modes’ universally seen as a boson-peak in glasses’ reduced spectra (and in disordered crystals [22]) are intimately related to the emergence of QLEs in those systems. In addition, we establish that quasilocalized excitations are plentiful (i.e. are not rare) — and coupled among themselves and with phonons — at and in the vicinity of the boson-peak frequency, consistent with the suggestions of [8–10], but at odds with recent claims [21].

Our findings reinforce the statement of Ref. [44] — based on experimental evidence — that vibrations constituting the boson peak in amorphous solids have a local character. In addition, the construction of boson-peak vibrational modes by hybridized QLEs (and phonons) somewhat echoes the ideas of Ref. [26], where it was suggested that excess vibrational modes can be constructed from (quasilocalized) responses to local force dipoles.

Our results, which indicate that QLEs play important roles in the physics of the boson peak in glasses, may also imply that the boson-peak frequency \( \omega_{\text{BP}} \) is related to the characteristic frequency \( \omega_g \) of QLEs [20, 32, 37]. The latter can be identified from the QLEs VDoS, whose dimensional form reads \( D_{\text{QLE}}(\omega) \sim \omega^4/\omega_g^5 \), as explained in [20, 32]. Indeed, a close relation between \( \omega_g \) and \( \omega_{\text{BP}} \) has been suggested and indirectly supported in [32]. The findings of the present work support such an intimate relation, which should be further explored in future work.

In addition, future work should establish the generality of the conclusions arrived at in this work, across different glass-forming models, across different glass-formation protocols, and also in three dimensions, and make them more quantitative. In particular, it would be interesting to examine whether the removal of internal stresses in model disordered solids — as done e.g. in Refs. [18, 45, 46] —, or the introduction of strong attractive forces — as done e.g. in Ref. [38] — have a measurable effect on the nature of vibrational modes at the boson-peak frequency.

Finally, we remark that methods to exhaustively compute the entire population of QLEs from a single computer glass are still under development; once such tools become available, future research should quantify the physics of QLE-QLE and QLE-phonon hybridizations. Such studies will inform us about how vibrational modes are expected to behave in glasses approaching the thermodynamic \( N \rightarrow \infty \) limit. In this context, we note that the computational results presented in this contribution has very recently led to some closely related theoretical and experimental (reanalysis of available data) progress [43].

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Appendix A: Lower-frequency vibrations

In Fig. 5, we show for comparison the results of the same analysis performed in Fig. 3 of the main text, this time applied to a vibrational mode with frequency \( \approx \omega_{\text{BP}}/4 \) (top row), and to a vibrational mode with frequency \( \approx \omega_{\text{BP}}/2 \) (bottom row). It is observed that the “imperfections” seen in the wave patterns of vibrational modes at these lower frequency in fact originate from hybridization with quasilocalized excitations.

Appendix B: Detected QLEs

In Fig. 6, we show the highest and lowest frequency QLEs detected using the method explained in Sect. III. The QLEs shown here have \( \omega = 0.69 \) (panel (a)) and \( \omega = 1.78 \) (panel (b)). Both modes show the same generic structural features of a disordered core decorated by quadrupolar algebraic far-field decays.
FIG. 5. Comparison between vibrational modes $\psi$ with (panel (a)) $\omega \approx \omega_{\text{BP}}/2$ and $\omega \approx \omega_{\text{BP}}/4$ (panel (c)), and the corresponding constructed superpositions $\phi_i$ (panels (b) and (d), respectively) of quasilocalized excitations extracted as explained in Sect. IV of the main text. The superpositions show that the “imperfections” seen in the wave patterns of these lower frequency vibrational modes in fact originate from hybridization with quasilocalized excitations. The red bars represent the phononic wavelength $2\pi c_s/\omega$.

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FIG. 6. Examples of detected QLEs. The frequencies of these modes are $\omega = 0.69$ (panel (a)) and $\omega = 1.78$ (panel (b)). In both cases, the detected QLEs reveal the generic structure of a disordered core decorated by quadrupolar, algebraically decaying far-fields.

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