Spatial structure of quasi-localized vibrations in nearly jammed amorphous solids

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The low-temperature properties of amorphous solids are widely believed to be controlled by low-frequency quasi-localized modes. What governs their spatial structure and density is however debated. We study these questions numerically in very large systems as the jamming transition is approached and the pressure \( p \) vanishes. We find that these modes consist of an unstable core in which particles undergo the buckling motions and decrease the energy, and a stable far-field component which increases the energy and prevents the buckling of the core. The size of the core diverges as \( p^{-1/4} \) and its characteristic volume as \( p^{-1/2} \). These features are precisely those of the anomalous modes known to cause the Boson peak in the vibrational spectrum of weakly-coordinated materials. From this correspondence we deduce that the density of quasi-localized modes must go as \( g_{\text{loc}}(\omega) \sim \omega^4/p^2 \), in agreement with previous observations. Our analysis thus unravels the nature of quasi-localized modes in a class of amorphous materials.

Introduction. The low-temperature \( T \lesssim 1 \text{K} \) properties of amorphous solids are universal, and markedly different from those of crystals [1, 2]. Their specific heat increases linearly with \( T \) and their thermal conductivity increases as \( T^2 \) [1, 2]. To explain these observations, Anderson et al. [3] and Phillips [4] proposed the famous two-level systems model, that was later on extended to the soft potential model [5, 7]. This theory postulates that amorphous solids have low-frequency quasi-localized vibrational modes in addition to phonons, which can cause double-well structures in the energy landscape. The universal properties of amorphous solids can then be explained in terms of the quantum tunneling of these two-level systems and their interactions with phonons.

However, the current theory is phenomenological and does not specify the nature of these localized modes, which remains a matter of debate [8, 9]. This state of affaire led to a considerable effort to characterize quasi-localized modes numerically. Schober and Laird detected them in density of states (vDOS) [15], in consistence with the assumption of the soft potential model [5, 7]. (ii) Display a vibrational density of states (v DOS) \( g_{\text{loc}}(\omega) \) that follows a power-law \( g_{\text{loc}}(\omega) \propto \omega^4 \) [16–20] where \( \omega \) is the frequency, in agreement with previous arguments for disordered bosonic systems [9, 21]. (iii) Decay algebraically in space as long as they are not hybridized with phonon [17, 22]. This decay is rapid enough for their participation ratio to scale as \( 1/N \) as for truly localized modes, where \( N \) is the number of particles. (iv) Are suppressed if pre-stress is removed [19, 23]. (v) Play an important role in mechanical failure under load [24, 26] and in the structural relaxation near the glass transition [27, 28]. Interestingly, their characteristic frequency appears to rise rapidly with approaching that transition, in concert with a local measure of elastic stiffness [29]. Despite these recent advances, understanding what fixes the nature and density of these modes remains a challenge.

In this letter we seek to resolve these questions by studying the spatial architecture of these modes, and how it is affected by the proximity of the jamming transition. The latter is reached in finite-range interacting particles as the pressure \( p \) vanishes [30, 31]. A well-known property of the vibrational spectrum of amorphous solids, an excess modes with respect to the Debye density of states called the Boson peak [2], is singular at that point. The associated modes, called ”anomalous” in this context, have been characterized in detail [31, 42]. By considering very large systems, we can study localized soft modes even close to jamming. We find that these modes consist of an unstable core in which particles undergo the buckling motions and decrease the energy, and a stable far-field component which increases the energy and prevents the buckling of the core. We find that the size of the core diverges as \( p^{-1/4} \) and its characteristic volume as \( p^{-1/2} \). All these features are precisely those of the anomalous modes at the Boson peak frequency if pre-stress is removed (corresponding to removing all forces between interacting particles). Our analysis thus supports that localized soft modes are anoma-

Methods. We used monodisperse, three-dimensional packings of particles with mass \( m \) interacting through a finite-range, harmonic potential (see Ref. [19] for details):

\[
\phi(r) = \frac{\epsilon}{2} \left( 1 - \frac{r}{\sigma} \right)^2 H(\sigma - r),
\]

where \( \sigma \) is the particle diameter, \( \epsilon \) is the characteristic energy, and \( H(r) \) is the Heaviside step function. Length,
we calculate the contribution $\delta E$ where the displacement is large and heterogeneous, whose one are shown. We observe that these modes present a core component (reads [45]): $\lambda^k = (\omega^k)^2$, where $\omega^k$ is its eigenfrequency. Note that the eigenvectors are ortho-normalized. After removing three translational zero modes, eigenmodes are sorted in ascending order of their eigenvalues, i.e., $\omega^1 < \omega^2 < \cdots < \omega^{N-3}$. Then for a given mode, $\omega^j$ is negative up to some length scale we denote as $\xi$, and decays rapidly with distance as expected from the decay of the displacements themselves.

Results. Figure 1 shows the visualization of the lowest-frequency modes of the systems at high ($p = 0.05$) and low ($p = 0.001$) pressures. Each arrow indicates an eigenvector, only those larger than 1% of the largest one are shown. We observe that these modes present a core where the displacement is large and heterogeneous, whose size appears to increases as pressure decreases.

To characterize the motions of particles in these modes, we calculate the contribution $\delta E^k$ of particle $i$ to the energy of the mode $k$, which must satisfy $\lambda^k = 2 \sum_i \delta E^k_i$. It reads [45]:

$$
\delta E^k_i = \frac{1}{4} \sum_{j \neq i} \left( \frac{u_{ij}^j}{r_{ij}} \right)^2 - \frac{f_{ij}}{r_{ij}} \left( \frac{u_{ij}^j}{r_{ij}} \right)^2 ,
$$

where $\partial i$ labels the set of particles interacting with particle $i$, $u_{ij}^j = (\hat{e}^k_i - \hat{e}^k_j) \cdot \hat{r}_{ij}$ is the relative displacement between $i$ and $j$ parallel to the bond $ij$ of direction $\hat{r}_{ij}$, $u_{ij}^j = \sqrt{|u_{ij}^j|^2 - (\hat{e}^k_i - \hat{e}^k_j) \cdot \hat{r}_{ij})^2}$ is the perpendicular component of that relative displacement, and $f_{ij} = -d\phi(r_{ij})/dr$ is the contact force. Note that $f_{ij}$ is always positive in the present system, and packings are called unstressed when setting $f_{ij} = 0$ [34].

Next we calculate the energy $\langle \delta E^k_i \rangle_k$ of the $i$-th particle with largest displacement averaged on all the quasi-localized modes we obtain at a given pressure. In Fig. 2 we plot $\langle \delta E^k_i \rangle_k$ vs. the averaged norms $\langle |e^k_i| \rangle_k$. We find that the larger the norm, the lower the energy. In particular, particles in the core (particles with large norm) even have a negative energy. This result implies that the perpendicular motion $u_{ij}^j$ is very dominant there, since it is the only negative contribution to the energy following Eq. 2, and $f_{ij}/r_{ij} \ll 1$ near jamming. Such a large perpendicular motion is a characteristic feature of anomalous modes [34, 41] and non-affine displacements under global deformations near jamming [46], and of the transition between double well potentials [47].

To study the spatial distribution of $\delta E^k$, we define the radial energy distribution function:

$$
\delta E(r) = \left\langle \frac{\sum_i \delta E^k_i \delta(r - r_i)}{\sum_i \delta(r - r_i)} \right\rangle_k ,
$$

where $r_i$ is the distance of the particle $i$ from that with the lowest energy. This function measures the average energy of particles at distance $r$ from the center of the localized mode. Fig. 3(a) shows $\delta E(r)$ for different $p$. For the moment, we focus on data that are far away from jamming, corresponding to $p = 0.05$ (black line). We observe that $\delta E(r)$ is negative up to some length scale we denote as $\xi$, here $\xi \approx 1.5$. For $r \gtrsim \xi$, $\delta E(r)$ is a positive quantity and decays rapidly with distance as expected from the decay of the displacements themselves. $\xi$ thus characterizes
the size of the unstable core of the localized modes, which is stabilized by its far field components corresponding to \( r > \xi_1 \).

We then calculate the integrated radial energy distribution function defined as:

\[
\Lambda(r) = \left\langle \sum_{r_1 \leq r} \delta E^k_i \right\rangle_k.
\]  

(4)

\( \Lambda(r) \) corresponds to the average energy the localized modes would have if the system was cut at a distance \( r \) from the center of the mode. Obviously, \( \lim_{r \to \infty} \Lambda(r) = \left\langle \lambda^k \right\rangle_k / 2 \). There is a direct link between \( \Lambda(r) \) and \( \delta E(r) \):

\[
\Lambda(r) \approx \int dr' \rho G(r') \delta E(r')
\]  

(5)

where \( \rho \) is the number density, and \( G(r) \) is the radial distribution function \([48]\). \( \Lambda(r) \) are shown for different pressures in Fig. 3(b). Again, we focus on \( p = 0.05 \) for the moment (black line). From Eq. 5, it is clear that the negativity of \( \delta E(r) \) at small distances results in the negativity of \( \Lambda(r) \) at small \( r \), which must display a minimum at a distance \( \xi_1 \) defined above. For \( r > \xi_1 \), \( \Lambda(r) \) gradually increases and becomes positive at a distance we denote \( \xi_3 \). Here \( \xi_3 \approx 15 \), which is ten-fold larger than the core size \( \xi_1 \). In practical terms, this result implies that even far from jamming, cutting the system around a localized mode at rather large distances \( r < 15 \) (and imposing external forces at the particles at the boundary to maintain force balance) would not lead to a stable system: the localized mode would still be unstable, and rearrangements would necessarily occur. The emerging physical picture for quasi-localized modes is that of a core which is passed due to Eq. (5). Lastly, \( \xi_3 \) has the length where \( \Lambda(r) \) reaches a minimum, which must satisfy \( \xi_1 \approx \xi_2 \) due to Eq. 5. Lastly, \( \xi_3 \) is smallest \( r \) for which \( \Lambda(r) \) becomes positive. \( \xi_1, \xi_2 \) and \( \xi_3 \) are indicated
in Figs. 3(a) and 3(b) by arrows for $p = 0.05$. The pressure dependences of $\xi_1$, $\xi_2$, $\xi_3$ is shown in Fig. 4(a), which supports the following power-law dependence:

$$\xi_1, \xi_2, \xi_3 \propto p^{-1/4}. \quad (6)$$

Therefore, the quasi-localized modes become more extended as $p \to 0$, and their characteristic length scale diverges at jamming.

Another characterisation of these modes is their participation ratio $P^k = \frac{1}{N} \left[ \sum_i \left( e_i^k \cdot e_i^k \right)^2 \right]^{-1}$. The quantity $NP^k$ is an estimate of the number of particles involved in the mode $k$ \[1\]-\[13]. We define the average volume of the localized modes as $V = \langle NP^k \rangle_k$, whose dependence on $p$ is shown in Fig. 4(b). Once again we find a singular behavior near jamming, consistent with:

$$V \propto p^{-1/2}. \quad (7)$$

Discussion. The scaling results Eqs. (6) and (7) support that the quasi-localized modes are the anomalous modes responsible for the Boson peak in these systems, whose properties we now recall. Near jamming, the density of vibrational modes exhibits a flat spectrum $g(\omega) \sim \omega^0$ at frequencies $\omega > \omega_*$ \[31, 32\], where $\omega_* \propto p^{1/2}$. Anomalous modes at $\omega_*$ are spatially extended, but can be characterized by finite correlation length which diverges at the jamming transition as $l_c \propto p^{-1/4}$ \[12, 35, 38\]. A length scale that also characterizes the response to a local perturbation \[50, 52\]. These results can be derived via effective medium calculations \[37, 39\]. Anomalous modes at $\omega_*$ and the response to a local perturbation are also tied together by a recent variational argument \[42\] showing that the later can be used as building blocks to reconstruct the former. These building blocks can be localized on a length scale $l_\rho$ and on a characteristic volume $V \propto p^{-1/2}$ \[42\] without affecting significantly their frequency scale. ($V \propto p^{-1/2}$ differs from $l_\rho^d$ where $d$ is the spatial dimension due to the algebraic decay of the mode magnitude in space). The architecture we discovered for quasi-localized modes is thus fully consistent with that of the building blocks of anomalous modes.

This correspondence can be used to explain the existence of the quasi-localized modes and to predict how their density depend on the distance to jamming. In the absence of pre-stress (obtained by dropping the second term in Eq. (2)), there are no anomalous modes at frequencies $\omega < \omega_*$, a frequency beyond which they suddenly appear and their density becomes large \[34\]. Due to this large density, it is plausible that these modes hybridize and are thus extended. In the stressed system, the energy of anomalous modes decreases approximately by $-p$ due to the second term in Eq. (2) \[34\]. As a result, anomalous modes populate the entire frequency range $0 < \omega < \omega_*$, an effect coined marginal stability. This effect lifts the degeneracy of the anomalous modes, which then become quasi-localized on the characteristic length scale of the building blocks that constitute them. This view is consistent with the finding that quasi-localized modes are mostly apparent in the stressed system, and disappear when pre-stress is removed \[16, 23\]. The integrated density of anomalous modes in this frequency range is of order $\omega_*$. This scaling, implied by the flat density of anomalous modes, simply states that there is one anomalous modes in this frequency range every volume $V$. Let us assume that a finite fraction of these modes become quasi-localized. From general arguments \[21\], we know that their density must follow $g_{\text{loc}}(\omega) \sim c(p)\omega^4$. Requiring that:

$$\int_0^{\omega_*} g_{\text{loc}}(\omega) d\omega \sim \omega_* \sim \frac{1}{V} \quad (8)$$

fixes $c(p) \sim V^{-1}\omega_*^{-5} \propto p^{-2}$, as indeed found numerically \[19\]. Our approach thus rationalises why the density of quasi-localized modes explodes near jamming.

Overall, our work supports that quasi-localized modes correspond to the anomalous modes known to control the boson peak in finite-range interacting systems. Although this correspondence is most stringently tested near jamming where both objects display singular properties, we expect it to hold true away from jamming as well. If so, our conclusion should hold in Lennard-Jones glasses, where the boson peak can also be interpreted in terms of the distance to a jamming transition (that cannot vanish however due to long-range interactions) \[53\], but also in chalcogenide glasses and silica where jamming corresponds to the point where the covalent network becomes rigid \[39, 54\].

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We first estimated the width of the band of the lowest frequency phonon by the same method as Ref. [22]. We then fitted the peak of the vDOS of the lowest frequency phonon to the Gaussian function and obtained the mean frequency $\omega_1$ and the standard deviation $\Delta \omega_1$ of the band. We finally estimated the width of the band to be $3\Delta \omega_1$, and we picked up only the vibrational modes that satisfy $\omega_k^b \in [\omega_1 - 3\Delta \omega_1, \omega_1 + 3\Delta \omega_1]$ in our analysis.