Disentangling defects and sound modes in disordered solids

S. Wijtmans¹ and M.L. Manning¹

¹Syracuse University, Syracuse, New York 13244, USA

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We develop a new method to isolate localized defects from extended vibrational modes in disordered solids. This method augments particle interactions with an artificial potential that acts as a high-pass filter: it preserves small-scale structures while pushing extended vibrational modes to higher frequencies. The low-frequency modes that remain are “bare” defects; they are exponentially localized without the quadrupolar tails associated with elastic interactions. We identify a robust definition for the energy barrier associated with each defect, which is an important parameter in continuum models for plasticity. Surprisingly, we find that the energy barriers associated with “bare” defects are generally higher than those for defects decorated with elastic tails, suggesting that elastic interactions may help to constitutively activate particle rearrangements.

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Under applied stress, solids can flow plastically. In crystals, this flow is controlled via rearrangements at lattice defects, which are the dislocations [1], easily identified by looking at the bond orientational order. In disordered solids, such as granular materials [2–5] and bulk metallic glasses [6–8], the structural defects are not easily identifiable using structural data, though the rearrangements are still localized [8–9].

Localized defects can self-organize, changing the bulk properties of materials. For example, shear bands, which are regions that flow faster than the rest of the material, develop when defects co-localize [10–11]. In other regimes, self-organization of defects leads to avalanching, where a material deforms elastically until a stress-lowering rearrangement at one defect triggers a cascade of rearrangements at other defects [12]. Interesting memory effects [13–14] may also be generated by self-organization of defects.

Defects play an important role in several mesoscopic phenomenological theories of plasticity, such as the theory of shear transformation zones (STZ) [9–13], which given a population and energy scale for defects, predicts how the solid will fail, and the theory of soft glassy rheology (SGR) [15], which assumes a population of yielding mesoscopic regions.

Isolating and quantifying features of defects in disordered solids has proved difficult. Finding the precise set of particle displacements that allow the system to rearrange at the lowest energy cost is computationally intractable. Recent observations that the low-frequency linear vibrational modes are strongly correlated with particle rearrangements and plasticity [17–18] suggest that low-frequency localized excitations have very low energy barriers and therefore identify defects. However, this conjecture is difficult to test because individual low-frequency normal modes are quasi-localized with long-ranged quadrupolar tails due to elastic interactions.

This conjecture is supported by a recently developed algorithm that highlights particles with the largest displacements in the lowest frequency modes, and finds that they do cluster into soft spots that identify locations where rearrangements are likely to occur [19]. This method has been extended to glasses at various temperatures [20], in different geometries [21], and to identify the approximate directions of particle displacements [22]. A second method based on machine learning can also identify structural defects [23].

However, all of these algorithmic approaches do not allow calculation of energy barriers for localized defects. The soft spots algorithm contains systematic errors due hybridization of soft spots and sound modes [19], while the machine learning algorithm does not identify directions of particle displacement and does not yet provide strong physical insight [23].

In this Letter, we develop a new method that pushes phonon-like modes out of the low frequency spectrum, leaving isolated excitations behind. This indicates that quasi-localized modes are in fact hybrids between phonons and localized defects, and allows us to test the hypothesis that localized excitations have low energy barriers. Surprisingly, we find that “bare” defects have higher energy barriers than quasi-localized modes decorated with quadrupolar tails. Because the statistics of energy barriers are an important model parameter in continuum theories of plasticity, this suggests that elastic interactions can facilitate flow in a different way than previously thought.

We study 50:50 mixtures of 2500 bidisperse disks with diameter ratio 1.4, at a packing fraction of $\phi = 0.9$, significantly larger than the jamming $\phi \sim 0.84$ [24]. Their interaction potential is Hertzian: $V = \frac{1}{2}k\delta^{3/2}$, where $\delta$ is the particle overlap. Lengths are in units of the mean particle diameter, and energies are in units of $\kappa$. We analyze mechanically stable packings in a periodic box of linear size $L \sim 47$ generated by an infinite temperature quench [24].
To find vibrational modes, we use ARPACK \footnote{V. 23} to calculate eigenvalues, \( \lambda \), and eigenvectors, \( \mathbf{e}_i \), of the dynamical matrix, \( \mathbf{M} \), which describes the linear response of the packing to particle displacements \footnote{24}. At large displacements, broken contacts alter the response \footnote{27}, but there is always a well-defined linear regime \footnote{28, 29}, and in our packings at \( \phi = 0.9 \) that regime is large.

To penalize long-range collective motion, we augment the system with an artificial square grid with lattice constant \( a \) of spring-like interactions. These spring-like interactions link the coarse-grained particle motions \( \mathbf{\tilde{u}} \), and generate an augmented potential energy \( \mathbf{\tilde{U}} \) and dynamical matrix \( \mathbf{\tilde{M}} = \mathbf{M} + \mathbf{M}^\dagger \):

\[
\mathbf{\tilde{U}} = \frac{1}{2} \mathbf{u} \cdot (\mathbf{M} + \mathbf{M}^\dagger) \cdot \mathbf{u} \\
= \frac{1}{2} (\mathbf{u} \cdot \mathbf{M} \cdot \mathbf{u} + \mathbf{K}_{kl}(\mathbf{\tilde{u}}_{k\gamma} - \mathbf{\tilde{u}}_{l\delta})^2) \\
\mathbf{\tilde{u}}_{k\gamma} = \sum_{\mathbf{i}} \mathbf{u}(x_{i\gamma})e^{-(x_{i\gamma} - k\gamma a)^2}/\sigma^2 ,
\]

where \( k, l \) are grid point indices and \( \mathbf{\tilde{u}}_{k\gamma} \) represents a Gaussian weighting of particle displacements. We choose this weighting because for \( \sigma \gtrsim a \), the sum of the Gaussian terms on each individual particle is nearly identical, with fluctuations on the order of \( 1 \times 10^{-8} \) for \( a = \sigma \). We fix \( \sigma = a \) to limit the inclusion of redundant particle displacements in coarse-grained grid points. Then \( \mathbf{M}^\dagger \) becomes:

\[
\mathbf{M}_{ij\alpha\beta}^\dagger = \mathbf{K}_{ik}(\mathbf{W}_{il}\mathbf{W}_{jl} - 2\mathbf{W}_{il}\mathbf{W}_{jk})
\]

with \( \mathbf{W}_{il} = \exp\left(-\left((x_{i\alpha} - l_\alpha a)^2 + (x_{i\beta} - l_\beta a)^2\right)/\sigma^2\right) \), and \( \mathbf{K}_{kl} = \mathbf{K} \) for neighbors on the square lattice and 0 otherwise. The motion of each grid point, defined by Eq. 3, is illustrated schematically in Fig. 1(a): average displacements in the same direction are penalized. Because \( \mathbf{\tilde{M}} \) is no longer guaranteed to be positive definite, augmented eigenvalues \( \tilde{\lambda} \) can be negative.

Fig. 1(b) shows the particle displacements in a typical low-frequency hybridized mode derived from the standard dynamical matrix \( \mathbf{M} \). A typical low-frequency eigenvector of the augmented dynamical matrix \( \mathbf{\tilde{M}} \), shown in Fig. 1(c), is localized, and the inset shows it has an exponential decay in amplitude, indicating a “bare” defect.

Fig. 1(d,e) show the sum of the magnitudes of the 30 lowest frequency modes for typical standard and augmented matrices, respectively. In Fig. 1(e), large magnitudes occur in the same localized regions as in Fig. 1(d), indicating that the augmented potential does not interfere with small-scale structure or alter the locations of the soft spots. It does suppress the background associated with extended excitations, making soft spots easier to identify.

An analysis of sums over eigenvectors, like those shown in Fig. 1(d,e) with \( K = 1 \) and \( g = 5 \), suggests fixing \( K = 1 \) is sufficient to push extended modes to higher frequencies. For larger values of \( K \), \( \mathbf{M}^\dagger \) begins to dominate \( \mathbf{M} \) and soft spot resolution decreases.

The augmented potential acts as a high-pass filter, increasing the energy of modes with lengthscales smaller than \( \sim g/2 \). This is shown in Fig. 2(a), which compares the standard \( \mathbf{U} \) and augmented \( \mathbf{\tilde{U}} \) energies associated with particle displacements along an artificially generated plane wave. To verify this same effect for vibrational modes, we compare each augmented mode to the
standard mode with which it has the largest dot product. Fig 2b) examines the difference in energies of each pair of modes as a function of frequency, averaged over 20 packings. We fix \( g = 5 \), corresponding to 9.47 particle diameters between grid points, as it generates the largest difference in energy between low and high frequency modes [30].

To confirm that modes with increased energies are indeed extended, we split eigenvectors into three groups. Modes with participation ratio < 0.1 are “localized”. Remaining modes are characterized by integrals over the Fourier spectrum, \( I_f \), and \( L_2 \) distance between their cumulative distribution functions (cdfs) and the typical boson peak cdf \( L_{BP} \) [27]: \( I_f > 0.3 - 3 * L_{BP} \) are “phonon-like”, and the remainder are “boson peak” modes.

While augmented and standard matrices have a similar total density of states at low frequencies, the fraction of those modes that are extended differs. Fig. 2c) shows the difference in the density of phonon-like and boson peak states between \( \tilde{M} \) and \( M \), while the inset compares the integrated number of localized modes, because there are not enough statistics to compare the density. This shows the number of plane-wave-like and boson peak-like states is significantly suppressed in the augmented system at low frequency, while the number of localized modes is significantly increased.

These observations demonstrate that when extended modes are pushed to higher frequencies, the remaining low-frequency normal modes are “bare” localized excitations. This confirms our intuition that quasi-localized modes are hybridizations of plane waves and the fundamental defects that accommodate flow in solids.

This physically-motivated algorithm for the first time allows us to calculate the distribution of energy barriers associated with bare defects. We displace particles along each eigenvector and use the LBFGS line search algorithm [32] to minimize the potential energy at every step. At the first step where the minimized state is different from the initial state, we say that there has been a particle rearrangement and define an energy barrier \( \Delta U \) as the difference between the initial energy and the maximum energy attained [33].

The definition of a particle rearrangement or new state can be subtle: not all contact changes signal saddle points in the potential energy landscape [29] and the energy landscape is fractal [34]. In order to define a new state, we examine six independent criteria: i) any change in the contact network (CR+), ii) non-rattler [35] contact changes (CR−), iii) requiring more than two particles to change contacts (C2), iv) energy differences between the original and final basins of greater than \( 10^{-8} \) (E-8), v) a displacement of a single particle more than two large particle diameters in a direction perpendicular to the mode (D), and vi) requiring that more than 2 contact changing particles must be neighbors, thereby rearranging as a unit, (C2N).

For each standard mode and each of the first five definitions, we measure the ratio between the energy barrier calculated using that definition and C2N \((\Delta U/\Delta U_{C2N})\). Fig. 3a) is a plot of the distributions of these energy barrier ratios. This plot shows that metrics that have been used for energy barriers in the past [33], such as contact changes that include or exclude rattlers (CR+,CR−), generate energy barriers that are significantly lower than the other criteria and different from each other. This suggests that these criteria generate a lot of “false pos-
FIG. 3: Energy barrier ratios. (a) Ratio of energy barriers \( \Delta U/\Delta U^{C2N} \) calculated using different definitions for what constitutes a particle rearrangement, as described in the main text. Box and whiskers contains 50% and 92% of the data points, respectively, blue bars denote the median, and outliers are circles. (b) The first column compares the standard mode with the lowest C2N energy barrier \( e_{\text{min}} \) to the most locally similar augmented mode \( \tilde{e}_{\text{min}} \), while the second and third columns compare \( e_{\text{min}} \) to weighted versions of itself, as described in the main text, with statistics over 100 realizations. (inset) Localization lengths the eigenvector \( \tilde{e}_{\text{min}} \) shown for 100 realizations, calculated as shown in the inset to Fig. 1(c). Black vertical line indicates the independently chosen \( \ell^* \).

The first column of Fig. 3(b) shows the distribution of \( \Delta U(e_{\text{min}})/\Delta U(\tilde{e}_{\text{min}}) \) over 100 realizations. Surprisingly, the median value is significantly greater than unity, indicating that the “best” localized augmented mode typically has a higher energy barrier than its quasi-localized standard counterpart; only in 14% of realizations is does the augmented mode have a lower energy barrier.

To understand whether this increased energy is generated by the long-range quadrupolar tails, or subtle differences in the particle displacements inside the defect region, we examine the energy barriers of artificially localized versions of \( e_{\text{min}} \). Specifically, we compare \( e_{\text{min}} \) to a weighted version of itself; for \( e_{\text{exp}}^{\tilde{e}_{\text{min}}} \), the magnitudes of particle displacements are weighted by an exponential function with a decay length \( \ell^* \) centered at the largest particle displacement, while for \( e_{\text{out}}^{\tilde{e}_{\text{min}}} \) they are weighted by unity inside a circle of radius \( \ell^* \) and zero otherwise.

As seen in the second and third columns of Fig. 3(b), these artificially localized modes generate energy barriers that are higher than the standard modes, and comparable to or higher than the augmented modes, despite having identical structure in the defect region. This indicates that removing the long-range elastic tails from a quasi-localized mode increases the energy barrier and suggests that the quadrupolar tails decrease the energy barrier for a bare defect.

Using a simple physically-motivated augmented potential, we were able to push extended low-frequency vibrational modes to higher frequencies and isolate localized “bare” defects. Although we expected that localized bare defects would have lower energy barriers than their quasi-localized counterparts, we found the opposite. Calculations of energy barriers on artificially localized modes suggest that the long-range elastic tails in each eigenmode actually lower energy barriers, providing an explanation for this counterintuitive result.

These results should immediately improve continuum models for plasticity in amorphous solids. Our method allows the direct calculation of energy barriers associated with defects in disordered solids, an important parameter in both STZ and SGR models. While these were previously fitting parameters, one can now extract them from a simulation and test specific model predictions. In addition, it is fairly straightforward to extend this algorithm to a simulated packing undergoing simple shear (with Lees-Edwards periodic boundary conditions), and a future direction is to study the creation, annihilation and activation of defects in sheared simulations and compare to continuum model assumptions.

Our observation that defects with long-range elastic tails have lower energy barriers than bare defects suggests that the lowest energy barriers could be systematically lower at higher pressures, as non-affine contributions become weaker and well-defined elastic tails become stronger. Furthermore, defects that are well-connected to the rigid backbone may be more likely to...
fail than those that are poorly connected, generating an additional explanation for observed spatial patterns of avalanches [12, 41, 43]. It is well understood that pairs of defects have elastic interactions that lower effective energy barriers, but our work suggests that elasticity might constitutively lower energy barriers for a single defect in isolation. It would be interesting to study plasticity in a model that incorporates this effect.

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