Thermal origin of quasi-localised excitations in glasses

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Abstract

Key aspects of glasses are controlled by the presence of excitations in which a group of particles can rearrange. Surprisingly, recent observations indicate that their density is dramatically reduced and their size decreases as the temperature of the supercooled liquid is lowered. Some theories predict these excitations to cause a pseudo-gap in the spectrum of quasi-localised modes of the Hessian, \(D_L(\omega)\), while others predict a gap in \(D_L(\omega)\) that grows upon cooling. To unify these views and observations, we generate glassy configurations of controlled gap magnitude \(\omega_c\) at temperature \(T = 0\), using so-called ‘breathing’ particles, and study how such gapped states respond to thermal fluctuations. We find that (i) the gap always fills up at finite \(T\) with \(D_L(\omega) \approx A_4(T) \omega^4\) and \(A_4 \sim \exp(-E_0/T)\) at low \(T\), (ii) \(E_0\) rapidly grows with \(\omega_c\), in reasonable agreement with a simple scaling prediction \(E_0 \sim \omega_c^2\) and (iii) at larger \(\omega_c\) excitations involve fewer particles, as we rationalise, and eventually become string-like. We propose an interpretation of mean-field theories of the glass transition, in which the modes beyond the gap act as an excitation reservoir, from which a pseudo-gap distribution is populated with its magnitude rapidly decreasing at lower \(T\). We discuss how this picture unifies the rarefaction as well as the decreasing size of excitations upon cooling, together with a string-like relaxation occurring near the glass transition.

1 Introduction

A key feature of structural glasses is that groups of particles can rearrange locally between two stable states, a motion that can be triggered by quantum or thermal fluctuations, or by exerting an external stress or strain. Specifically at low temperature, quantum tunnelling between these states can lead to two-level systems (TLS) that are then the dominant source of excitations [1–3]. Furthermore, for any temperature below the glass transition, plasticity upon stress loading occurs when such a group becomes unstable, corresponding to a saddle-node bifurcation in the potential energy landscape [4, 5] leading to a rearrangement denoted shear transformation [6]. Finally, relaxation in supercooled liquids near their glass transition occurs via thermally activated events, observed to become more and more string-like upon cooling [7, 8].

Understanding how temperature or system preparation controls the density of these excitations remains a challenge. It is, however, a question of practical importance since the density of shear transformations controls for instance the glass brittleness [9–11], the rarefaction of activated events near the glass transition controls its fragility [12], and the density of TLS (recently observed to be almost absent in ultra-stable glasses [13, 14]) affects the decoherence in cubits [15]. Another tantalising question concerns the possible unification of these excitations into a common description.

These excitations should affect the low-frequency spectrum of the Hessian of the energy landscape, since groups of particles that can easily rearrange tend to have a small linear restoring force [4, 16, 17]. Indeed, in numerical glasses, quasi-localised modes are found at low-frequencies [18]. Recently there has been a considerable effort to analyse them [19–25]. In most glasses, it is found that in inherent structures\(^1\), the density of quasi-localised modes \(D_L(\omega) \approx A_4(T) \omega^4\), with \(\omega\) the frequency. Most remarkably, \(A_4(T)\) is reduced by several decades as \(T\) is reduced by 30% [25, 26] (a similar finding was obtained for the density of TLS [27]). Furthermore, quasi-localised modes also display a lower participation ratio upon cooling. A unifying explanation for these facts is currently missing\(^2\).

On the theoretical side, two distinct approaches have been proposed. On the one hand, the \(\omega^4\) power law has been rationalised by making specific assumptions on the disorder and by assuming that modes were non-interacting [29, 30], or by modelling a quench from \(T = \infty\) and including interactions [17]. On the other hand, in mean-field calculations in infinite dimensions for temperatures below the mode-coupling temperature \(T_c\) [31] the spectrum of the Hessian becomes gapped (excluding obvious long wave-length Goldstone modes that are always present). Below \(T_c\), the gap is predicted to grow as \(T\) decreases [32–34]. A gap was also predicted from real-space stability arguments in finite dimensions for continuously polydisperse particles, at very low energies and zero temperature [35]. Nevertheless, it is currently unclear if a gap truly exists in finite dimension and at finite temperature.

\(^1\)Obtained by rapidly quenching an equilibrated liquid (at initial temperature \(T\)) to zero temperature.

\(^2\)TLS were proposed to be controlled by the mosaic length scale that diverges at the Kauzmann temperature \(T_K\) in mean field approaches [28]. Yet this description predicts a growing (instead of decreasing) length scale and a mild (a factor 10 at most) decrease of density of excitations upon cooling.
In this article we seek a unifying scenario for these facts and different approaches, by studying the stability of gapped spectra with respect to thermal fluctuations. Specifically, we use ‘breathing’ particles\(^3\) [37] in order to generate athermal ultra-stable glasses of controlled gap magnitude \(\omega_c\). Then, as sketched in Fig. 1, we transiently reheat these glasses, with a standard molecular dynamics simulation, at a low temperature \(T_a\) for a duration \(t_a\), before quenching them back to zero temperature. Our central results are that (a) thermal fluctuations, even small, destroy the gap and we recover a density \(D_L(\omega) \approx A_4 \omega^4\); the prefactor \(A_4(T_a, t_a)\) depends very mildly on \(t_a\) but presents an Arrhenius dependence on temperature with \(A_4 \sim \exp(-E_a/T_a)\) (in our temperature units the Boltzmann constant \(k_B = 1\)). (b) The activation energy \(E_a\) rapidly increases with the gap magnitude \(\omega_c\). (c) We introduce a novel algorithm to decompose the rearrangements into elementary excitations, and find that they involve fewer particles for larger gap values, and eventually become string-like for our largest gap. We propose a scaling argument for their decreasing size. Overall, these results suggest to describe equilibrated liquids perturbatively as \textit{gapped states decorated by thermally activated excitations whose characteristic energy is controlled by the gap itself}, leading to a contribution with \(A_4 \sim \exp(-E_a(\omega_c(T))/T)\). We discuss the implication of this picture, sketched in Fig. 1, for the density of these various excitations as well as for the glass transition.

![Figure 1](image_url)

**Figure 1.** When a gapped glass is heated to a temperature \(T_a\) for a duration \(t_a\), as sketched in the inset, modes beyond the gap act as a reservoir of excitations that can be thermally activated. It fills up the gap, leading to a pseudo-gap \(D_L(\omega) \approx A_4 \omega^4\). This effect is exponentially diminished if \(T_a\) decreases or \(\omega_c\) increases (as predicted by the mean-field description near the glass transition).

2 Generating gapped glasses

To generate ultra-stable glasses displaying a finite gap, we follow a procedure similar to [35]. We consider ‘breathing’ particles whose individual size can vary according to an energetic cost of characteristic stiffness \(K\) (see Appendix B). The particles interact with a repulsive potential, up to a finite cutoff radius, chosen such that the potential remains continuous up to its third derivative [38] and thus allowing for a well-defined Hessian. At a given temperature, this system is known to be thermodynamically equivalent to a system of given (and continuous) polydispersity, yet using a usual molecular dynamics (MD). Including this breathing degrees of freedom leads to a giant shortening of the equilibration time, comparable to that of swap algorithms [37, 39]. In practice, we perform MD with breathing particles for a long duration \(t_p\) at a temperature \(T_p(K)\), chosen such as to minimise the energy of the states eventually obtained (see Appendix C), before quenching using a ‘FIRE’ algorithm [40] in which particles can still breathe.

The polydispersity obtained for various values of stiffness \(K\) is shown in Fig. 2(a) for \(N = 8000\) particles, in three dimensions and at fixed pressure. Next, we freeze the radius of each particle, and compute the usual Hessian of the potential energy: its eigenvectors correspond to the vibrational modes of the glass, and its eigenvalues are denoted \(\omega^2\) since they correspond directly to the frequencies of vibrational modes, as we take the particle mass is unity. Showing that these states are gapped requires considerable statistics; in fact, we collect the spectra of \(n = 4000\) independent realisations (see Appendix A for a precise statement) and average them in order to obtain the density of vibrational modes \(D(\omega)\). For such system size, one finds quasi-localised modes softer than the first plane waves [20]. \(D(\omega)\) turns out to display a gap: there are no quasi-localised modes below a finite frequency \(\omega_c\). Because \(\omega_c\) is even higher than the frequencies of the first plane waves for \(K = \{10^2, 10^3\}\), this requires us to manually remove them in order to measure the density of \textit{quasi-localised} modes \(D_L(\omega)\), as shown in Fig. 2(b). We extract \(\omega_c\) by fitting a power law \(D_L(\omega) \sim (\omega - \omega_c)^c\), and obtain the values \(\omega_c = \{1.64, 1.19, 0.85, 0.65\}\), for \(K = \{10^2, 10^3, 3 \times 10^3, 10^4\}\), indicated with markers in Fig. 2(b). Note that if we consider instead the minimal frequency observed as an estimate for \(\omega_c\), this does not affect our conclusions below (see Appendix D).

![Figure 2](image_url)

**Figure 2.** (a) Distribution of particle radii, normalised by the number density \(\rho = N/V\), for different values of stiffness \(K\). (b) Density of quasi-localised modes displaying a finite gap \(\omega_c\), in contrast to the usual pseudo-gap scaling \(D_L(\omega) \sim \omega^4\) indicated with a dashed line. The gap values \(\omega_c \approx \{1.64, 1.19, 0.85, 0.65\}\) corresponding respectively to \(K = \{10^2, 10^3, 3 \times 10^3, 10^4\}\) are indicated using ticks, following the same color code. Physically, decreasing \(K\) results to a larger gap and thus a more stable glass, and is associated to a larger polydispersity.
3 Filling up the gap via thermal activation

![Figure 3.](image)

(a) Density of soft quasi-localised modes \( D(\omega) \) after reheating for a fixed duration \( t_a = 500 \) at different temperatures \( T_a \) (following the protocol sketched in Fig. 1). For reference, the mode-coupling temperature in this system is \( T_c \approx 0.3 \). The first plane wave is indicated by a black arrow, and the gap \( \omega_c = 1.64 \) (the largest one we generate) by a black tick. We emphasise that, before reheating, we had \( D_L(\omega < \omega_c) = 0 \), so that the corresponding modes have been activated by thermal fluctuations. (b) Prefactor \( A_t \) as a function of reheating temperature \( T_a \) for different durations \( t_a \). (c) Collapse of the different curves \( A_t(T_a, t_a) \), supporting the functional form \( A_t = f(t_a^2 \exp(-E_a/T_a)) \) where the function \( f \) is linear at small arguments, supporting an Arrhenius behaviour at low temperature \( T_a \) (see Appendix D). Remarkably, this collapse indicates that for a given gap, the distribution of excitation energies is characterised by a single scale (presumably a lower cutoff, see below).

Interestingly, we find in Fig. 3(d) that this energy scale \( E_a \) very strongly increases with gap magnitude \( \omega_c \) (see below for a proposed explanation). The dynamical exponent \( \gamma(\omega_c) \) is also shown in inset, and remains smaller than 0.2 in all the range of initial gaps that we probe.

4 Modes beyond the gap act as an excitation reservoir

To rationalise these findings, we consider the path of minimal energy connecting two states associated to one excitation, and denote by \( s \) the curvilinear coordinate along it. The Taylor expansion of the energy along this path from the state 1, by definition the one of minimal energy, reads:

\[
E(s) = \frac{1}{2!} \lambda_1 s^2 + \frac{1}{3!} \kappa_1 s^3 + \frac{1}{4!} \chi_1 s^4 + O(s^5). \tag{2}
\]

In general such a path is not straight, in the 3\( N \)-dimensional configurational space, and neither does it lie fully along the associated soft quasi-localised mode of frequency \( \omega_1 \). Nevertheless, for soft modes with low barriers we expect generally the path and the mode to nearly coincide, thus we have approximately \( \omega_1^2 \approx \lambda_1 \).

Thermal activation towards state 2 will lead to a smaller frequency \( \omega_2^2 \approx \lambda_2 \). From Eq. (2) it is straightforward to obtain the expansion from state 2, and the transformation \((\lambda_2, \kappa_2, \chi_2) = g(\lambda_1, \kappa_1, \chi_1)\). The joint distribution follows \( P(\lambda_2, \kappa_2, \chi_2) = |g'(\lambda_2, \kappa_2, \chi_2)| P(\lambda_1, \kappa_1, \chi_1) \) where the Jacobian determinant \( g'(\lambda_2, \kappa_2, \chi_2) \sim \lambda_2 \) for small \( \lambda_2 \) (see Appendix E). One expects \( P(\lambda_1, \kappa_1, \chi_1) \) to be smooth, implying that for small \( \lambda_2 \) one has \( P(\lambda_2, \kappa_2, \chi_2) \sim \lambda_2 \) or equivalently \( P(\omega_2, \kappa_2, \chi_2) \sim \lambda_2 d\lambda_2/d\omega \sim \omega_2^3 \). After integrating on \( \kappa_2 \) and \( \chi_2 \) one gets \( D_L(\omega_2) \sim \omega_2^3 \). See [30] for a more general argument along the same line. Thus one expects to observe a pseudo-gap following thermally activated excitations. One effect will deplete the spectrum even further: in the case of a cubic pseudo-gap, the low-frequency spectrum is dominated by states 2 very close to a saddle node bifurcation (at the spinodal). However, once interactions among excitations are taken into account\(^4\), configurations with such a large density of states near saddle node bifurcation can be shown to be unstable and display avalanche-type events, \( i.e. \) where the relaxation of one excitation can destabilise others in turn [17]. This effect will increase the pseudo-gap exponent to

\(^4\)Such interactions are relevant even at high temperature near the glass transition [41]. Note that when quenching the system to zero temperature after a reheating, interactions with relaxing vibrational modes may also destabilise excitations which are close to their spinodal.
Values larger than three\(^5\).

As far as the kinetics is concerned, the time scale \(t_a\) on which an excitation equilibrates depends on the energy barrier \(\Delta E\) to go from state 1 to 2. It will occur (neglecting prefactors) when \(t_a \gg t_a^* \sim \exp(\Delta E/T)\), i.e. its first-passage time. For much larger time scales, the probability of being in the excited states follows a Boltzmann factor \(\exp(-E_{12}/T)\) at small \(T\), where \(E_{12}\) is the energy difference between the two states. If all states were equilibrated, \(A_4\) would not depend on \(t_a\) (i.e. \(\gamma = 0\)). By contrast, if no states were equilibrated \(A_4\) would grow linearly in time. In that respect, our observation of the intermediate case \(\gamma \approx 0.2\) is consistent with the common picture that there is a broad distribution of barriers, so that on the time scale \(t_a\) a fraction of excitations are equilibrated, yet some barriers are still being jumped over for the first time.

For a given gap magnitude \(\omega_c\), we expect to find a lower cutoff on the distribution of barriers \(\Delta E\) (with the typical energy difference \(E_{12}\) of the associated excitations being of the same order of magnitude). Consider for instance a symmetric double well in the energy landscape and expand its energy around the maximum: \(E(s) = -\frac{1}{2} \lambda s^2 + \frac{1}{4} \lambda s^4\). It is straightforward to show that in each minimum the frequency of the soft mode scales as \(\sqrt{\lambda}\), allowing us to identify for the softest excitations \(\lambda \sim \omega_c^2\). Likewise in this example the barrier for the double well follows \(\Delta E \sim \lambda^2 \sim \omega_c^4\). This scaling holds for asymmetric double wells as well (see Appendix E). Interestingly, our measured activation energy \(E_a\) is compatible with this power-law relation, except for the smallest gap (hence less stable glass) that we study\(^6\).

Overall, this analysis supports the scenario that modes beyond the gap act as a reservoir of excitations, with a broad distribution of barriers presenting a typical cutoff \(E_a \sim \omega_c^4\) at low energies.

### 5 Rearrangements involve fewer particles and become string-like at large gaps

We introduce a novel algorithm to decompose the displacement field of a rearrangement into several elementary excitations, which is needed to study how their geometry depends on the gap magnitude. Given a displacement field (induced, in our case, by the thermal cycle), we first consider the particle with maximal displacement, and draw a sphere of radius \(\bar{R}\) around it. Beyond this sphere, all the particle displacements are set to zero (i.e. these particles are set back to their initial position in the gapped state), whereas within the sphere the displacements are preserved.

Next, we perform with that initial condition a steepest descent of the interaction energy. We find that if \(\bar{R}\) is small, all displacements go back to zero, whereas if \(\bar{R}\) is large, they do not. We consider the smallest \(\bar{R}\) of the latter case, and the displacement field obtained at the end of the corresponding gradient descent defines our first elementary excitation. Next, we subtract this obtained displacement field from the full one, and repeat the entire procedure recursively until no more excitations are found (see Appendix F for details and visual examples).

Given some individual excitation of displacement field \(\delta \vec{r}_i\), we compute from its associated participation ratio an estimate of the number of particles involved in this excitation \(NP_r \equiv \left(\sum_i ||\delta \vec{r}_i||^2\right)/\left(\sum_i ||\delta \vec{r}_i||^4\right)\). For each gap magnitude \(\omega_c\), we find about 5000 such excitations and report the mean and the median of this observable in Fig. 4(a). We find that the typical number of particles involved in one excitation decreases as \(\omega_c\) increases. We propose the following rationalisation. First, the length scale of quasi-localised modes was found to be proportional to the characteristic length scale entering the response to a local dipole [23], as proposed based on a variational arguments in [44]. Second, the length scale \(\ell_c\) entering the dipolar response was observed to decrease as the system moves away from a marginally stable phase and enters a gapped solid phase as \(\ell_c \sim 1/\sqrt{\omega_c}\) [45]. The volume of the corresponding mode was shown to go as \(\ell_c^3\) (independent of the number of dimensions) for elastic networks of springs at rest [44, 45]. Assuming that it is more generally true, and taking together these results suggest that the number of particles involved in an excitation should decrease with larger gap, as \(NP_r \sim 1/\omega_c\). As shown in the inset of Fig. 4(a), this is in reasonable agreement with our observations.

A complementary observable for the geometry is the maximum displacement \(\max_i ||\delta \vec{r}_i||\) for a given excitation, whose mean and median values for all excitations at a given \(\omega_c\) are shown in Fig. 4(b). Interestingly, this maximum displacement increases with the gap, and becomes close to the small particle diameter equal to \(d^*\).

A direct visualisation of the excitation fields reveals a (presumably related) interesting phenomenon: for our largest gap, the displacements are string-like with several particles exchanging positions, as shown in Fig. 4(c), whereas for smaller gaps they are much more compact and no permutations occur. To quantify this effect, we follow the glass transition literature [7] and measure the distinct part of the Van Hove correlation:

\[
G_d(\vec{r}, t) \equiv \frac{1}{N} \sum_{i=1}^{N} \sum_{j=1}^{N} \delta(\vec{r} - \vec{r}_j(t) + \vec{r}_i(0)),
\]

where the average is made on all the observed elementary excitations at some given \(\omega_c\). It is plotted for our stabelst system in Fig. 4(e) after radially averaging. The key observation is the presence of a very sharp peak around \(r = 0\),
which can only arise from particles replacing each other. Interestingly, if we condition our definition of the Van Hove correlation to large or small particles only, we find that the peak only persists for small particles (in red in Fig. 4(e)). Strings thus correspond to smaller particles navigating in an environment of larger ones.

Next we integrate the peak around \( r = 0 \) to quantify the number of permuting particles averaged on all elementary excitations:

\[
\langle n_p \rangle = \left\langle N \int_0^{r_c} G_d(r, t_o) 4\pi r^2 dr \right\rangle,
\]

where \( r_c \) is a cutoff that is tuned. We observe that permutations are essentially absent except for the largest considered gap, see Fig. 4(f).

6 Discussion

In summary, we have argued that in gapped glasses, modes beyond the gap act as an excitation reservoir for thermal activation. This effect always destroys the gap and leads to a density of quasi-localised modes \( D_L(\omega) \approx A_4(T) \omega^4 \). At low temperatures, we found that \( A_4 \sim \exp(-E_a(\omega_c)/T) \) where the typical energy scale \( E_a(\omega_c) \) is a rapidly increasing function of \( \omega_c \). A simple scaling argument gives \( E_a(\omega_c) \sim \omega_c^4 \), in good agreement with our observations, except for the smallest gap value that we explore. Finally, we observed that as the gap increases, excitations involve fewer and fewer particles and become more and more string-like. The growing length scale of the excitations as \( \omega_c \rightarrow 0 \) presumably corresponds to the previously identified growing length, characterising the elastic response of an amorphous solid near a macroscopic elastic instability [45].

Although our observations were made in ultra-stable states obtained by a specific protocol, our arguments on this reservoir effect are much more general. Assuming that this effect is at play in supercooled liquids ties together several unexplained observations, as we now discuss.

Reinterpreting mean-field descriptions of glasses: Goldstein [46] proposed early on that the glass transition takes place near some temperature \( T_c \) below which most normal modes become stable. Such an enhanced stability is consistent with the overall elastic stiffening upon cooling apparent in the bulk [47] or local [22] elastic moduli in fragile supercooled liquids. Theoretically, this view is consistent with mean-field models of the glass transition in infinite dimensions – that turn out to be closely related to Mode-Coupling Theory – [32, 33], in which the spectrum of the Hessian becomes stable and opens a gap with \( \omega_c^2 \sim (T_c - T)^{\frac{4}{3}} \) [34]. Our work suggests a natural way to extend this picture to finite dimensions as sketched in Fig. 1: the gap is decorated by excitations stemming from the reservoir of modes with \( \omega \geq \omega_c \). In this approach (i) the excitation density strongly decreases with temperature: away from \( T_c \) in the deeply supercooled regime, it should be proportional to \( \exp(-E_a(\omega_c(T))/T) \) and (ii) as \( T \) decreases, \( \omega_c \) increases and excitations are less and less extended. Point (i) is a
plausible explanation for the very rapid decay upon cooling of $A_4(T)$ [25, 26], TLS density [27] and shear transformations [10, 48] observed in ultra-stable supercooled liquids. Point (ii) is consistent with the result that TLS [27] and quasi-localised modes [25, 26] present a lower participation ratio upon cooling (such changes of geometry may lead to additional effects on their density).

**Glass transition:** The mean-field proposal that supercooled liquids present an effective gap growing upon cooling, leading to a rarefaction of thermally accessible excitations, is consistent with the observation that rearrangements become string-like with more and more particles exchanging positions upon cooling [7, 8] – since we find that excitations at large gaps are precisely like that. In our view, why elementary excitations display such a geometry at large gap is yet to be explained.

At our lowest temperatures, only one or a few strings get activated, which can only lead to a very partial relaxation of the system. Isolated strings thus belong to the class of $\beta$-relaxation in supercooled liquids, as shown in [8] for model metallic glasses (molecular and covalent liquids may certainly present other $\beta$-relaxation mechanisms governed by the local chemistry, such as dangling bonds). Yet we believe that strings, if present in sufficient density, can fully relax liquids (at least the continuously polydisperse ones receiving much attention currently). Indeed they allow for the exchange of particles with distinct radii. Such swap moves are now known to relax the system with great efficiency, so the dynamics should not be slower than the time scale to naturally operate these swaps (see [51, 52] for related discussions). If these strings are sufficient to fully relax the system, then their characteristic low-energy cutoff $E_a$ is a viable candidate to characterise the $\alpha$-relaxation time in liquids. Fragility would then stem from the rapid rise of $E_a$ with the growing gap magnitude $\omega_c$.

Note that this view, in which activation deep in the supercooled liquid phase is controlled by $T_c$, contrasts with the usual interpretation of mean-field results in which activation gets activated, which can only lead to a very partial relax-ation in supercooled liquids, as shown in [8] for model metallic glasses (molecular and covalent liquids may certainly present other $\beta$-relaxation mechanisms governed by the local chemistry, such as dangling bonds). Yet we believe that strings, if present in sufficient density, can fully relax liquids (at least the continuously polydisperse ones receiving much attention currently). Indeed they allow for the exchange of particles with distinct radii. Such swap moves are now known to relax the system with great efficiency, so the dynamics should not be slower than the time scale to naturally operate these swaps (see [51, 52] for related discussions). If these strings are sufficient to fully relax the system, then their characteristic low-energy cutoff $E_a$ is a viable candidate to characterise the $\alpha$-relaxation time in liquids. Fragility would then stem from the rapid rise of $E_a$ with the growing gap magnitude $\omega_c$.

Note that this view, in which activation deep in the supercooled liquid phase is controlled by $T_c$, contrasts with the usual interpretation of mean-field results in which activation is controlled by an entropy crisis occurring at lower temperature $T_K$ [32, 33]. The latter approach is in our opinion ruled out by the recent observation that change of kinetic rules (such as allowing for swap moves [36]) immensely affects the location of the glass transition, while leaving intact thermodynamic properties [51]. Changes of kinetic rules, however, affect the location of $T_c$ [37], thus our proposed description of activation is consistent with the observations of swap algorithms.

**Lifshitz tails:** Beyond thermal activation of excitations,

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A Nomenclature

A.1 Symbols

- $n$ number of independent realisations in the ensemble
- $N$ number of particles in one realisation, labelled with indices $i = 1, \ldots, N$
- $\vec{r}_i$ particle position; vector in three-dimensional Cartesian coordinates
- $\delta \vec{r}_i$ particle displacement vector
- $R_i$ particle radius
- $k_B$ Boltzmann’s constant
- $m$ particle mass (the same for all particles)
- $L$ linear size of the box which has the same length in three directions
- $V$ volume of the box $\equiv L^3$
- $\rho$ number density $\equiv N/(V)$
- $V_p$ volume occupied by particles $\equiv \sum_{i=1}^{N} \frac{4}{3} \pi R_i^3$
- $\phi$ packing fraction $\equiv V_p/V$
- $K$ ‘stiffness’ with which the particle radius is drawn to its initial value during preparation
- $\omega$ vibrational frequency
- $T_p$ temperature during preparation using ‘breathing’ dynamics
- $t_p$ duration at which the temperature is fixed to $T_p$ during preparation
- $T_a$ temperature during thermal activation using normal molecular dynamics (MD)
- $t_a$ duration at which the temperature is fixed to $T_a$ during thermal activation
- $T_c$ mode-coupling temperature
- $\bar{n}_c$ average number of rearrangements per realisation

A.2 Units

- Lengths ($\vec{r}$, $R$, and $L$) are shown in the unit of $d_0$: the diameter of an initially small particle.
- Energies ($E_a$) are expressed in the unit of $\varepsilon$: the prefactor of the pair interaction potential.
- Temperature ($T_p$, $T_a$, and $T_c$) is in the unit of $\varepsilon/k_B$ where $k_B$ is Boltzmann’s constant.
- Time ($t_p$, $t_a$, and $\omega^{-1}$) is shown in the unit of $t_0$ where $t_0 \equiv \sqrt{m d_0^2 / \varepsilon}$.

A.3 Parameters & Key quantities

| $K$       | $1\times 10^{-2}$ | $1\times 10^{-3}$ | $3\times 10^{-3}$ | $1\times 10^{-4}$ |
|-----------|-------------------|-------------------|-------------------|-------------------|
| $\omega_c$| 1.65              | 1.19              | 0.85              | 0.65              |
| $n$       | $4\times 10^{-3}$ | $4\times 10^{-3}$ | $2\times 10^{-3}$ | $3\times 10^{-3}$ |
| $N$       | $8\times 10^{-3}$ | $8\times 10^{-3}$ | $8\times 10^{-3}$ | $8\times 10^{-3}$ |
| $\phi$    | 0.62              | 0.74              | 0.77              | 0.79              |
| $\langle V \rangle$ | $3.722 \pm 0.3$ | $7.303 \pm 0.7$ | $8.607 \pm 0.9$ | $9.356 \pm 1$ |
| $\rho$    | 2.15              | 1.10              | 0.93              | 0.86              |
| $T_p$     | 0.2               | 0.39              | 0.44              | 0.49              |
| $t_p$     | $1\times 10^{-4}$ | $1\times 10^{-4}$ | $1\times 10^{-4}$ | $1\times 10^{-4}$ |
| $T_c$     | 0.304             | 0.484             | 0.525             | 0.572             |
| $T_a$     | $\{0.15, 0.20, 0.23, 0.25, 0.27, 0.30, 0.38, 0.50, 0.60, 0.80\}$ | $\{0.07, 0.10, 0.20, 0.30, 0.39, 0.50, 0.60, 0.80\}$ | $\{0.03, 0.05, 0.1\}$ | $\{0.03, 0.05, 0.10, 0.20, 0.30, 0.49, 0.60, 0.80\}$ |
| $t_a$     | $\{5e2, 2e3, 1e4\}$ | $\{1e2, 5e2, 1e4\}$ | $\{5e2\}$ | $\{1e2, 5e2, 2e3\}$ |
| $E_a$     | $0.66 \pm 0.08$   | $0.15 \pm 0.03$   | $0.042 \pm 0.016$ | $0.03 \pm 0.007$  |
| $\gamma$ | 0.2               | 0.05              | -                 | 0.003             |
| $\bar{n}_c$ | 1.4              | 1.6               | 1.9               | 3.5               |
Note that the notation $2c3 \equiv 2 \times 10^3$; that $\langle V \rangle$ is the ensemble average volume (and the uncertainty its standard deviation); and that $n_c$ has been obtained with $t_a = 500$ at the lowest $T_a$ we probed for each $K$.

## B Molecular dynamics

### B.1 Sample preparation: ‘breathing’ dynamics

We study a three-dimensional periodic particle system of $N = 8000$ particles, that is characterised by the grand potential

$$ U = \sum_{i<j} \varphi (r_{ij}, R_i, R_j) + \sum_i \mu (R_i, R_i^{(0)}) $$

(5)

where $\varphi$ is a purely repulsive inverse power-law potential, defined

$$ \varphi (r_{ij}, R_i, R_j) = \begin{cases} 
\varepsilon \left[ \frac{R_{ij}}{r_{ij}} \right]^{10} + \sum_{p=0}^{3} c_{2p} \left( \frac{r_{ij}}{R_{ij}} \right)^{2p}, & r_{ij} \leq r_c \\
0, & r_{ij} > r_c 
\end{cases} $$

(6)

with $r_c$ is the cutoff distance, $R_{ij} \equiv R_i + R_j$ (two times the average particle radius), and $r_{ij} \equiv ||\vec{r}_i - \vec{r}_j||$ (the Euclidean norm of the distance vector separating particles $i$ and $j$). $c_{2p}$ is a constant that makes $\varphi$ continuous up to the third derivative at $r_c$. Furthermore,

$$ \mu (R_i, R_i^{(0)}) = \frac{K}{2} \left( 1 - \frac{R_i^{(0)}}{R_i} \right)^2 \left( R_i^{(0)} \right)^2 $$

(7)

is a chemical potential that allows a particle to change its size from its initial value $R_i^{(0)}$ at an energetic cost that scales with a modulus $K$. For $K = \infty$ it is impossible for a particle to change its radius, while it becomes easier as $K \to 0$. The initial particle radii are bi-disperse, in a 50:50 mixture. In particular, one, randomly selected, half of the particles has $R_i^{(0)} = 0.5d_0$ and the other half has $R_i^{(0)} = 0.7d_0$ (where $d_0$ sets the unit of length of our system).

Sample preparation proceeds by instantaneously heating the initial random configuration to a temperature $T_p$, and keeping it at this temperature for a certain time $t_p$ under the constraint of a fixed pressure $p = 20.0$ (in units of $\varepsilon/d_0^3$). We then instantaneously quench the system to zero temperature by minimising the grand potential. See algorithmic details below.

### B.2 Activation by temperature: normal dynamics

We proceed by fixing the particle size, which corresponds to a potential energy

$$ U = \sum_{i<j} \varphi (r_{ij}, R_i, R_j) $$

(8)

(see Eq. (6) for the definition of $\varphi$). We then gently heat the system configuration to a certain “activation temperature” $T_a$ (at a heating rate $T_a/(10t_0)$), and keep the sample at $T_a$ for a total duration $t_a$. Thereafter we instantaneously quench the sample to zero temperature. Algorithmic details are listed below.

### B.3 Molecular dynamics algorithm

We run molecular dynamics, whereby the particle dynamics are given by Newton’s equation of motion with the gradient of the potential energy on a particle as driving force. Time is discretised using the standard velocity Verlet algorithm.

The temperature and pressure are controlled using a Berendsen thermostat [54], where the temperature is defined as the total kinetic energy $\sum_i m||\dot{\vec{r}}_i||^2/2$ (where $\dot{\bullet}$ refers to the time derivative). Note that during preparation the kinetic energy is $\sum_i (m||\dot{\vec{r}}_i||^2 + \dot{\vec{r}}_i^2)/2$. We use the ‘FIRE’ algorithm [40] to quench the systems.

For completeness we report that $r_c = 1.48 d_0$, $\varepsilon = 1$, $m = 1$, $d_0 = 1$, and $\Delta t = 0.005$. Furthermore $c_0 = -1.110637662511798$, $c_2 = 1.2676152372297065$, $c_4 = -0.4960406072849212$, $c_6 = 0.0660511826415732$; see Supplemental Material of [38].

## C Sample preparation

We choose $T_p$ and $t_p$ to empirically generate a configuration in the lowest possible energetic state in terms of the mean interaction energy $\langle u \rangle = \langle U \rangle/N$ (averaged on an ensemble of $n = 10$ samples). In particular, we set $t_p = 10^4$ (the highest
value we can practically reach, with each sample taking eight CPU hours to prepare). We manually optimise $T_p$ as reported in Fig. 5. Note that we verify that the $T_p$ at which we find the optimum, is robust in terms of preparation duration $t_p$, by comparing our results to those for $t_p = 500$ (dashed line in Fig. 5). Furthermore, the reader is reminded that although the particle size distribution depends on temperature while still at $T_p$, the final particle size distribution at zero temperature is independent of $T_p$. Note that our ‘breathing’ dynamics (at small $K$) are quite efficient to prepare samples in a low potential energy state. We verify this by preparing an ensemble (again $n = 10$, but with $N = 2000$ particles) with normal dynamics and a slow quench rate. We plot the potential energy $\langle u \rangle$ at different temperatures in Fig. 6. In all cases $\langle u \rangle$ at $T = 0$ is higher than that for the sample prepared using ‘breathing’ dynamics, which was prepared at a fraction of the computational costs (sample preparation is a factor of 2000 faster using ‘breathing’ dynamics).

Figure 5. Mean interaction potential energy $\langle u \rangle$ after sample preparation with ‘breathing’ dynamics for varying parent temperature $T_p$ and two different waiting times $t_p$. The different panels correspond to different $K$ as indicated. The selected temperature $T_p$ for which the potential energy is lowest for the largest practically reachable $t_p = 10^4$ is indicated using vertical lines (see Appendix A for numeric values).

D Measurement of quasi-localised modes

D.1 Spectrum of the Hessian

We extract the Hessian (or stiffness matrix) – the second derivative of interaction energy – as follows

$$H_{ij} \equiv \frac{\partial^2 U}{\partial \vec{r}_i \partial \vec{r}_j} = -\frac{d^2 \varphi(r_{ij})}{dr_{ij}^2} \frac{\vec{r}_{ij}}{r_{ij}^3} - \frac{d\varphi(r_{ij})}{dr_{ij}} \frac{1}{r_{ij}} \left( I - \frac{\vec{r}_{ij} \vec{r}_{ij}}{r_{ij}^2} \right)$$

for $i \neq j$. The diagonal

$$H_{ii} = -\sum_{i \neq j} H_{ij}$$

due to translation symmetry.

Note that $H_{ij}$ is a second-order tensor, and that $I = \delta_{\alpha\beta} \vec{e}_\alpha \vec{e}_\beta$ is a second order unit tensor. We then diagonalise the Hessian, leading to $N$ eigenvalues $\lambda$ and corresponding eigenmodes $\Phi_i$. Because all particles have a mass $m = 1$ the corresponding $N$ eigenfrequencies are

$$\omega \equiv \sqrt{\lambda}$$

We finally represent the spectrum of the Hessian as

$$D(\omega) = \frac{1}{3N - 3} \sum_{k=1}^{3N-3} \delta(\omega - \omega_k)$$
Figure 6. Mean interaction potential energy as obtained by sample preparation using ‘breathing’ dynamics (at $T = 0$ for $K = 10^2$, $t_p = 10^4$, and $T_p = 0.2$, in black) and using normal dynamics at different cooling rates (cooling from $T = 0.4$) as indicated in the legend. Note that in both cases the ensemble comprises $n = 10$ samples, but that normal dynamics are run using smaller than usual samples comprising $N = 2000$ particles ($N = 8000$ is used throughout). We verify the representativeness of these smaller samples using $N = 8000$ for $T = 10^{-3}$, shown using a dashed red line (that indeed coincides with the solid red line for $N = 2000$). The required CPU time to run the entire simulation with $N = 2000$ particles is indicated. Note that for our ‘breathing’ dynamics the time has been divided by four to correct for the difference in system size.

D.2 Density of quasi-localised modes

The density of quasi-localised modes, $D_L(\omega)$, follows from the spectrum of the Hessian in Eq. (12) by filtering plane waves that have a frequency $\omega_e < \omega_c$ (where $\omega_c$ is defined below). We identify these plane waves by their signature in participation ratio

$$P_r = \left( \frac{\sum_i ||\Phi_i||^2}{N \sum_i ||\Phi_i||^4} \right)$$

Plane waves thereby have $P_r \approx 2/3$, while quasi-localised modes have $P_r \ll 1$.

In practice, most of our samples have no plane waves below $\omega_c$, rendering filtering obsolete. In fact, we only apply filtering after sample preparation for $K = \{10^2, 10^3\}$. Since we empirically observe the plane waves to be well separated from the quasi-localised modes in terms of frequency, we remove them by removing the first $3 + 12$ eigenmodes of each realisation for $K = 10^3$ and $3 + 12 + 24$ eigenmodes of each realisation for $K = 10^2$, corresponding the 3 translational modes and the first (two) bands of plane waves. Note that $D_L(\omega)$ is not renormalised after filtering of plane waves.

We emphasise that in all other measurements $D_L(\omega) = D(\omega)$ at low frequency.

D.3 Protocol to measure $\omega_c$

We measure the gap frequency $\omega_c$ – the frequency of the first quasi-localised mode. To measure $\omega_c$, we assert that the density of soft quasi-localised modes follows

$$D_L(\omega) \sim (\omega - \omega_c)^\zeta$$

at low frequency $\omega$. We then move $\omega_c$ until the power law is most obvious at low $\omega$, as shown in Fig. 7. We then visually extract the power $\zeta$ and check that it and the extracted $\omega_c$ are consistent with extreme-value statistics. In particular, we expect

$$\omega'_\text{min} - \omega_c \sim (n')^{-1/(1+\zeta)}$$

where $\omega'_\text{min}$ is the frequency of the softest quasi-localised mode in an ensemble of $n'$ realisations chosen as a random subset of our ensemble of $n$ realisations. We consider $\omega'_\text{min}$ the average of lowest three realisations (of $n'$ realisations). Indeed, our extracted $\omega_c$ is consistent with this scaling, as shown in the insets of Fig. 7. In addition, we check that $\omega_c$ is robust to a change of system size (shown in blue in Fig. 7).

D.4 Protocol to fit $A_4$

$A_4$ is extracted from $D_L(\omega)$ by fitting

$$D_L(\omega) = A_4 \omega^4$$

For $K = 10^3$, $\omega_c = 1.30 \pm 0.02$, and for $K = 10^2$, $\omega_c = \{1.26 \pm 0.03, 1.73 \pm 0.05\}$ (where the uncertainty refers to the standard deviation).
Figure 7. Fit of $\omega_c$ by asserting the power law scaling in Eq. (14) and check by extreme value statistics as in Eq. (15) (insets), for all considered $K$.

(i.e. Eq. (1)) for frequencies below the first plane wave (for $K = 10^2$) and for frequencies below $\omega_c$ (for $K = 10^3$, $3 \times 10^3$, and $10^4$). Note that consequently $D_L(\omega) = D(\omega)$ in the relevant frequency range for all these measurements. The mean and the error of $A_4$ follow as the mean and standard deviation of $\{\ln D(\omega_i) - 4 \ln \omega_i\}$ where $\omega_i$ corresponds to the position of the bins of $D(\omega)$.

We verify that the value of $A_4$ that we fit is robust to a mild decrease of system size (using $n = 2000$ realisations of $N = 4000$ particles, compared to an ensemble of $n = 4000$ realisations of $N = 8000$ particles). We find that both the density of soft quasi-localised modes and the extracted $A_4$ are robust to the change of system size, as reported in Fig. 8.

Figure 8. (a) $D(\omega)$ for different system sizes $N$ and different activation temperatures $T_a$ (as indicated in the legend). (b) $A_4$ fitted on (a) as a function of $1/T_a$. Both plots are for $K = 10^2$ and $t_a = 500$.

D.5 Protocol to fit $E_a$ and $\gamma$

Our protocol to fit $E_a$ and extract $\gamma$ consists of two steps. 1) We first collapse the curves of $A_4(T_a)$ for different $t_a$. Thereto we shift the horizontal axis of e.g. Fig. 3(b) in accordance with assuming a functional dependence

$$A_4 = A_4 \left( t_a e^{-E_a/T_a} \right)$$

(17)
until the curves for different \( t_a \) collapse to a single curve (e.g. Fig. 3(c)), by optimising the ratio \( \gamma/E_a \). 2) On the master curve we next fit \( E_a \) of low \( T_a \). Since we know the ratio \( \gamma/E_a \), the fitted value of \( E_a \) gives us direct access to \( E_a \). Specifically, we fit \( \ln(A_4) \) vs \( 1/T_a - \gamma/E_a \ln t_a \) using linear regression to get \( E_a \) and its error at low \( T_a \) (the lowest 5 data points in Fig. 3(c)).

D.6 Results for different \( K \)

In Fig. 9 we show the collapse of different waiting times \( t_a \) and the fit of \( E_a \) at low \( T_a \) for all ensembles that are not shown in the main text (notably Fig. 3). Note that for \( K = 3 \times 10^3 \) we extract \( E_a \) by directly fitting for low \( T_a \) for a single \( t_a \).

\[ A_4 \sim \exp(-E_a/T_a) \]

Figure 9. Fitting of \( E_a \) and \( \gamma \) for all \( K \) not shown in the main text. The fitted values are reported in Appendix A.

D.7 Robustness of \( E_a \)

In Fig. 10 we verify that the consistency with \( E_a \sim \omega_c^4 \) is robust to a different measure of the softest quasi-localised mode after sample preparation. In particular, we compare with \( \omega_{\min} \).

\[ E_a \sim \omega_c^4 \]

Figure 10. \( E_a \) as a function of \( \omega_c \) (open markers) or as a function of \( \omega_{\min} \) (solid markers).

E The Jacobian of the transformation from \((\lambda_1, \kappa_1, \chi_1)\) to \((\lambda_2, \kappa_2, \chi_2)\)

\[ E(s) = \frac{1}{2!} \lambda_1 s^2 + \frac{1}{3!} \kappa_1 s^3 + \frac{1}{4!} \chi_1 s^4 + \text{constant} \]

Figure 11. Double-well potential.

The potential around the state 1 is given by:

\[ E(s) = \frac{1}{2!} \lambda_1 s^2 + \frac{1}{3!} \kappa_1 s^3 + \frac{1}{4!} \chi_1 s^4 + \text{constant} \]
with the joint distribution \( P(\lambda_1, \kappa_1, \chi_1) \). For the new minimum, state 2, the potential reads:

\[
E(s) = \frac{1}{2!} \lambda_2 (s + s_0)^2 + \frac{1}{3!} \kappa_2 (s + s_0)^3 + \frac{1}{4!} \chi_2 (s + s_0)^4 + \text{constant}
\]  

(19)

where \( s_0 \) is the shift along \( s \), see Fig. 11; and the corresponding joint distribution is \( P(\lambda_2, \kappa_2, \chi_2) \).

The relation between two sets of coefficients \( \lambda_1, \kappa_1, \chi_1 \) and \( \lambda_2, \kappa_2, \chi_2 \)

\[
\begin{cases}
\chi_1 = \chi_2 \\
\kappa_1 = \kappa_2 + \chi_2 s_0 \\
\lambda_1 = \lambda_2 + \kappa_2 s_0 + \frac{1}{2} \chi_2 s_0^2
\end{cases}
\]  

(20)

where \( s_0 \) as a function of \( (\lambda_2, \kappa_2, \chi_2) \) follows from the fact that the linear term vanishes in Eqs. (18) and (19). In particular,

\[
6\lambda_2 + 3\kappa_2 s_0 + \chi_2 s_0^2 = 0
\]  

(21)

The joint distribution \( P(\lambda_2, \kappa_2, \chi_2) \) is given by

\[
P(\lambda_2, \kappa_2, \chi_2) = \left| \det \left( \frac{d\lambda_1 d\kappa_1 d\chi_1}{d\lambda_2 d\kappa_2 d\chi_2} \right) \right| P(\lambda_1, \kappa_1, \chi_1)
\]  

(22)

where

\[
\left| \det \left( \frac{d\lambda_1 d\kappa_1 d\chi_1}{d\lambda_2 d\kappa_2 d\chi_2} \right) \right| = \left| \det \left( \frac{\partial\lambda_1}{\partial\lambda_2} \frac{\partial\lambda_1}{\partial\kappa_2} \frac{\partial\lambda_1}{\partial\chi_2} \frac{\partial\kappa_1}{\partial\lambda_2} \frac{\partial\kappa_1}{\partial\kappa_2} \frac{\partial\kappa_1}{\partial\chi_2} \frac{\partial\chi_1}{\partial\lambda_2} \frac{\partial\chi_1}{\partial\kappa_2} \frac{\partial\chi_1}{\partial\chi_2} \right) \right| = \left| 1 + \kappa_2 \frac{\partial s_0}{\partial\lambda_2} + \chi_2 \frac{\partial s_0}{\partial\kappa_2} \right|
\]  

(23)

From Eq. (21) we find that

\[
\frac{\partial s_0}{\partial\lambda_2} = -\frac{6}{3\kappa_2 + 2\chi_2 s_0}
\]  

(24)

\[
\frac{\partial s_0}{\partial\kappa_2} = -\frac{3s_0}{3\kappa_2 + 2\chi_2 s_0}
\]  

(25)

And thus:

\[
\left| \det \left( \frac{d\lambda_1 d\kappa_1 d\chi_1}{d\lambda_2 d\kappa_2 d\chi_2} \right) \right| = \left| -\frac{3\kappa_2 - \chi_2 s_0}{3\kappa_2 + 2\chi_2 s_0} \right| = \left| \frac{6\lambda_2}{3\kappa_2 s_0 + 2\chi_2 s_0^2} \right|
\]  

(26)

If the excited state of the double-well potential is close to the spinodal case, \( \lambda_2 \) is small (as it is in Fig. 11). In particular, when \( \lambda_2 \approx 0 \), it follows that \( s_0 \approx -3\kappa_2/\chi_2 \). Inserting this in Eq. (26) gives:

\[
\left| \det \left( \frac{d\lambda_1 d\kappa_1 d\chi_1}{d\lambda_2 d\kappa_2 d\chi_2} \right) \right| \approx \frac{2\lambda_2 \chi_2}{3\kappa_2^2} \sim \lambda_2
\]  

(27)

We have thus found that the joint distribution

\[
P(\lambda_2, \kappa_2, \chi_2) \sim \lambda_2 P(\lambda_1, \kappa_1, \chi_1)
\]  

(28)

Hence, the marginal distribution \( P(\lambda_2) \sim \lambda_2 \) after integrating out \( \kappa_2 \) and \( \chi_2 \):

\[
D(\omega_2) = P(\lambda_2) \frac{d\lambda_2}{d\omega_2} \sim \omega_2^3
\]  

(29)

**Gap in energy barrier distribution**

For a given \( \lambda_1 \) we define \( c(\kappa_1, \chi_1) = \lambda_1 \chi_1/\kappa_1^2 \), which smoothly varies in a narrow range from 1/3 (for a symmetric double well) to 3/8 (for a spinodal). Then we can express the energy barrier as

\[
\Delta E = \lambda_1^2 \left( 3 - \sqrt{9 - 24c} \right)^2 \frac{\left( -3 + 12c + \sqrt{9 - 24c} \right)}{192 \chi_1 c^2}
\]  

(30)

The function \( c \) is slowly varying and not singular so that

\[
\Delta E \sim \lambda_1^2 \sim \omega_2^3
\]  

(31)

Similarly, the energy difference

\[
E_{12} = \lambda_1^2 \left( 3 + \sqrt{9 - 24c} \right)^2 \frac{\left( -3 + 12c - \sqrt{9 - 24c} \right)}{192 \chi_1 c^2} \sim \lambda_1^2 \sim \omega_2^3
\]  

(32)

(except in the case of a symmetric double well).
Geometry of rearrangements

F.1 Protocol to separate rearrangements

The displacement field between the states before and after reheating may contain more than one elementary excitation. We extract them one-by-one from this displacement field, by assuming them linearly independent. This corresponds to the following algorithm:

1. Find the particle with the largest displacement.
2. Place a small sphere centred at this particle with a radius \( \tilde{R}^{(i)} = (V/N)^{1/3} \) (with \( i \) the increment number, starting at \( i = 0 \)).
3. Set all displacements outside the sphere equal to zero. The particle displacements inside the sphere are not changed.
4. Minimise the energy \( U \) (every particle is free to move).
5. Increase the radius of sphere: \( \tilde{R}^{(i+1)} = \tilde{R}^{(i)} + \Delta \tilde{R} \), and reset the displacements as in step 3 (the particle displacements outside the sphere are set to zero and those inside the sphere equal to the original particle displacements).
6. Repeat steps 4 and 5, until the localised mode is identified. In particular, stop when \( |U^{(i+1)} - U^{(i)}| < 10^{-6} \) and, to avoid stopping too early, the norm of the displacement field is larger than \( 10^{-2} \). Note that \( U^{(i)} \) refers to the potential energy after energy minimisation, in step 4, for increment \( i \).

The local rearrangement is then the displacement field after the last energy minimisation. We then subtract it from the original displacement and continue to extract the next elementary excitation, by repeating this algorithm. We continue to do so until we have extracted all elementary excitations. In particular, we stop \( \tilde{R}^{(i)}/L > \sqrt{3}/2 \) (with \( L \) the linear size of the simulation box).

F.2 Results

Five representative samples (for two different \( K \)) showing our separation protocol are shown in Fig. 12, whereby the displacement field of each elementary excitation is plotted using a different colour. On average, we measure \( \{1.4, 1.6, 1.9, 3.5\} \) elementary excitations for \( K = \{10^2, 10^3, 3 \times 10^3, 10^4\} \) at the lowest thermal activation \( T_a = \{0.15, 0.07, 0.03, 0.03\} \) with \( t_a = 500 \).

![Figure 12](image)

Figure 12. Individual local rearrangements projected on the \( xy \) plane, shown using different colours, in five randomly chosen examples (from those samples that show more than one local rearrangement) for: (top) \( K = 10^2, T_a = 0.15, \) and \( t_a = 500 \), (bottom) \( K = 10^4, T_a = 0.03, \) and \( t_a = 500 \).

We, furthermore, include the distribution of the participation ratio of the elementary excitations in Fig. 13(a), whereby the different colours correspond to the different data points in Fig. 4(a). We observe that the elementary excitations become more localised for larger gaps. Likewise, we include the distribution of the maximum displacement of each elementary excitation in Fig. 13(b) (the different colours correspond to the different data points in Fig. 4(b)). In this case we observe
that the maximum displacement increases for our largest \( \omega_c \) (in black) as the result of string-like motion. This is supported by the distinct part of the Van Hove correlation in Fig. 13(c) that displays a sharp peak around \( r = 0 \) only for our largest \( \omega_c \) (in black). For this configuration, we plot the distribution of the number of permuting particles, \( \#n_p \), inside the ‘string’ in Fig. 13(d).

**Figure 13.** Probability distribution of (a) the participation ratio \( NP_r \) and (b) the maximal particle displacement \( \max_i \{||\delta \vec{r}_i||\} \) at different \( \omega_c \). (c) The distinct part of the Van Hove correlation normalised by the number density \( \langle G_d \rangle / \rho \) at different \( \omega_c \). (d) Histogram of the number of particles that permute per realisation, \( \#n_p \) (for largest \( \omega_c \)) at two different cutoff distances \( r_c / d_0^* = \{0.025, 0.05\} \).