Geometry of hopping processes and local excitations in glasses

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In amorphous materials, groups of particles can rearrange locally into a new stable configuration. Such elementary excitations are key as they determine the response to external stresses, as well as to thermal and quantum fluctuations. Yet, understanding what controls their geometry remains a challenge. Here we build a scaling description of their geometry and energy in terms of the distance to an instability, corresponding to a vanishing frequency \( \omega_c \) in their vibrational spectrum, as predicted for instance at the dynamical transition in mean field approaches of supercooled liquids. We successfully test our predictions in materials with a gapped spectrum and in regular ultrastable glasses. Our analysis explains why string-like rearrangements are observed to relax liquids only at low temperatures, as otherwise excitations display too small displacements to probe the granularity of the material.

If a liquid is cooled rapidly enough to avoid crystallization, its dynamics rapidly slows down until the glass transition where equilibration cannot be achieved: a glass is formed, and the material acts as a solid. What controls the dynamics in such supercooled liquids is a long-standing question of condensed matter [1, 2]. Yet, new observations further constrain the descriptions of this phenomenon. The ‘swap’ Monte-Carlo algorithms [3] (in which nearby poly-disperse particles can exchange positions, in addition to their usual translation move) can speed up the dynamics by 15 orders of magnitude or more, and can change the glass transition temperature \( T_g \) by up to a factor two [4]. Because swap algorithms achieve thermal equilibrium, theories of the glass transition in which thermodynamics governs kinetics [5, 6] appear ill suited to explain such a dramatic difference [7] (see [8] for an alternative view). Several theoretical works (including real-space [9], replica [10] and mode-coupling [11] approaches) predict that the dynamical transition temperature \( T_c \) below which thermal activation becomes the dominant mechanism of relaxation [1] decreases in the presence of swap, plausibly explaining the speed up of this algorithm. However, understanding the dynamics in the vicinity of \( T_c \) in finite dimension \( d \) remains a challenge. By contrast, in the infinite dimensional limit [12], mean-field treatments are exact: one finds that a gap in the vibrational spectrum of magnitude \( \omega_c \) appears for \( T < T_c \) that grows upon cooling [13], and that the relaxation time diverges at \( T = T_c \) [14]. For finite \( d \), the vibrational spectrum instead presents a pseudo-gap consisting of quasi-localized modes (QLMs) [15]. Moreover, thermally activated events or ‘hopping processes’ still occur for \( T < T_c \), leading to a finite relaxation time. The geometry of these excitations changes at lower temperature and eventually becomes string-like with particles that exchange positions [16, 17]. The cause for the presence of strings is still debated: they may correspond to elementary rearrangements, or, alternatively, be entropically favored and consist of a sequence of many successive rearrangements [18].

In a parallel development, there has been recently a considerable effort to analyze both QLMs as well as elementary excitations as a function of glass stability [19–27]. Numerically, liquids are equilibrated at a parent temperature \( T_p \) before being rapidly quenched to \( T = 0 \), thus obtaining an inherent structure where the Hessian of the energy can be analyzed, and where excitations can be triggered using a short thermal cycle. Strikingly, it is found that the density of excitations is reduced by several decades as \( T_p \) decreases [26], and that the characteristic number of particles involved rapidly decreases upon cooling. The former observation is consistent with recent experiments on vapor-deposited glasses [28, 29].

In this Letter, (i) we use mean-field and real-space arguments to express the length \( \ell_{\text{loc}} \), displacement \( \delta_{\text{loc}} \), number of particles \( N_{\text{loc}} \) and energy \( E_{\text{loc}} \) of low-energy excitations, assuming the presence of an underlying gap of magnitude \( \omega_c \) in the vibrational spectrum. We find our predictions to be accurately satisfied in gapped glasses [30, 31]. (ii) Our analysis implies scaling relations between local properties \( \ell_{\text{loc}}, \delta_{\text{loc}}, N_{\text{loc}}, E_{\text{loc}} \) that we find to be accurately satisfied in regular ultrastable (gapless) glasses. Together with mean field results predicting a growing gap \( \omega_c \) upon cooling, our results explain why excitations become smaller in stable glasses. (iii) We find that elementary excitations become string-like in ultrastable glasses, as we observed previously for glasses with a large gap \( \omega_c \) [31]. Our analysis supports that such strings are elementary excitations, that only occur away from an instability when excitations have large enough displacements to probe the granularity of the material.

Scaling description for local excitations. We consider a material that undergoes an elastic instability when a control parameter \( \epsilon \) vanishes, for instance \( \epsilon \sim (T_c - T) \) if the instability is driven by temperature. Infinite dimensional [13, 32] calculations as well as effective medium theory [33] then predict a vanishing gap frequency \( \omega_c \sim \sqrt{\epsilon} \) above which the spectrum of the Hessian of the energy is a semicircle. In finite dimensions, hopping processes between stable configurations will, how-
ever, occur.

To estimate their spatial extension, we consider two replicas of the system in the glass phase \((\epsilon = T_c - T > 0)\), and denote by \(Q(r)\) their overlap. \(Q(r)\) characterizes the similarity between two configurations at location \(r\), and is unity if they are identical [34]. In mean field, the free energy of this coupled system undergoes a saddle-node bifurcation as \(\epsilon \to 0\) [35] for some finite value of the overlap \(Q^*\). To describe the spatial fluctuations of \(Q(r)\), we use the following Ginzburg-Landau free energy [36, 37] [38]:

\[
F[Q] = \int d^d r [\epsilon(Q - Q^*) + \frac{1}{3}(Q - Q^*)^3 + \frac{1}{2}(\nabla Q)^2]
\]

\[
\approx \int d^d r [2\epsilon(Q - Q_{eq})^2 + \frac{1}{2}(\nabla Q)^2],
\]

(1)

where the second line is obtained by performing an expansion around the minimum of the free energy \(Q_{eq}\) satisfying \(Q_{eq} - Q^* = \sqrt{\epsilon}\). The overlap \(Q(r)\) displays thermal fluctuations, whose length scale and correlation volume can be deduced from the correlation function \(G(r) = \langle Q(r)Q(0)\rangle - \langle Q(0)\rangle^2\). For the quadratic free energy of Eq. (1), this classical computation gives:

\[
G(r) \sim \frac{1}{\sqrt{d-2}} \exp(-r/\xi) \quad \text{with} \quad \xi \sim \epsilon^{-1/4}.
\]

(2)

A similar length scale was predicted to affect the dynamics in mode-coupling theory [39, 40] and was observed to characterize the linear response near an instability [41]. Eq. (2) also leads to a characteristic volume where fluctuations are correlated. A simple integration [42] leads to \(V \sim \xi^2 \sim 1/\sqrt{\epsilon}\) independently of the dimension (a relation between volume and length already known to hold near jamming [23, 43]). In what follows we make the natural assumption that low-energy elementary excitations do occur on the characteristic volume and length scale of spontaneous fluctuations, such that their number of particles \(N_{loc} \sim V\) and their length \(\ell_{loc} \sim \xi\).

To obtain the characteristic displacement and energy scale of local excitations, we perform an expansion of a symmetric double well \(E(X) = -m_2 \omega_c^2 X^2 + \chi X^4 + o(X^4) \equiv E_2 + E_4\) (for an asymmetric double well, both the energy barrier and difference generically scale as the result we obtain [31]). Here \(X\) is the norm of the displacement field, that satisfies \(X^2 \sim N_{loc} \delta_{loc}^2\) where \(\delta_{loc}\) is the particle displacement. One readily obtains that the energy barrier between the two minima is \(E_{loc} \sim m_2^2 \omega_c^2 / \chi\) [44] and the distance between the local minima follows \(X^2 \sim m_2 \omega_c^2 / \chi\), implying that \(\delta_{loc}^2 \sim m_2 \omega_c^2 / \chi N_{loc}\).

Ultimately, the term \(E_4 = \chi X^4\) stems from the quartic non-linearity in the inter-particle interaction potential, which we assume to be short ranged. We denote its characteristic magnitude \(\kappa\), being a microscopic quantity it takes a finite value as \(\omega_c \to 0\). Writing that the total quartic term is a sum of the microscopic ones leads to:

\[
E_4 \sim N_{loc} \kappa \delta_{loc}^4\]

implying that \(\chi \sim \kappa / N_{loc}\). This scaling relation is confirmed empirically for QLMs in the Supporting Information (SI). We thus obtain \(\delta_{loc}^2 \sim m_2 \omega_c^2 / \kappa\) and \(E_{loc} \sim m_2^2 \omega_c^4 N_{loc} / \kappa\). Overall we get the following scaling description (disregarding constant prefactors):

\[
N_{loc} \sim \frac{1}{\omega_c}, \quad E_{loc} \sim \omega_c^3, \quad \delta_{loc} \sim \omega_c, \quad \ell_{loc} \sim \frac{1}{\sqrt{\kappa} \omega_c}.
\]

(3)

Thus, we predict that close to an instability (e.g. \(\omega_c \sim \sqrt{\epsilon} \sim \sqrt{T_c - T}\)), hopping processes are extended with small characteristic displacement and energy scales. Away from an instability, they become small with large displacements and energy.

**Gapped glasses.** We first test Eq. (3) in three-dimensional gapped glasses, obtained with ‘breathing’ particles [9, 30]. We use the protocol and parameters of [31] reviewed in the SI. In a nutshell, we perform molecular dynamics (MD) simulations in which the radius of all \(N\) particles is an additional degree of freedom, whose stiffness \(K\) controls the particle polydispersity. A long run at finite temperature is followed by an instantaneous quench to zero temperature. We then freeze the particle radii and measure the vibrational spectrum, which presents a gap of magnitude \(\omega_c\) that strongly depends on \(K\).

Next, we study elementary excitations using thermally activated rearrangements. They are obtained by heating our samples with standard (non-breathing) MD to a temperature \(T_a\) for a duration \(t_a\), followed by an instantaneous quench using the “FIRE” algorithm. In practice, \(T_a\) and \(t_a\) are chosen so as to trigger one rearrangement in average (up to 4 rearrangements per sample in practice), which we then separate in individual ones using an algorithm developed in [31]. A displacement field \(\delta R = \{\delta R_i\}, i = 1...N\), which is a vector of dimension \(Nd\), is associated to each excitation. We focus on elementary excitations that go to higher energy states. In doing so, we eliminate events where one double well is very asymmetric and lies close to a saddle-node bifurcation (and would then present a tiny activation barrier not captured by our scaling assuming a symmetric well). These events can also be suppressed if the quench is not instantaneous, as we use below.

For each gap magnitude, we obtain of the order of 100 excitations. We consider the median of the following observables: (i) The number of particles involved in a rearrangement \(N_{loc} = NP_\ell\), where \(P_\ell\) is the participation ratio of \(\delta R\). (ii) The particle characteristic displacement \(\delta_{loc} \equiv X/\sqrt{N_{loc}}\) where \(X = ||\delta R||\). (iii) The length \(\ell_{loc}\) defined from the second moment of the position of the particles involved in the rearrangement. Namely, \(\ell_{loc} \equiv 2\sqrt{T} \sum I \equiv \sum_i m_i ||\Delta R_i||^2\), \(m_i = ||\delta R_i||^2 / \sum_j ||\delta R_j||^2\) and \(\Delta R_i = R_i - \sum_j m_j R_j\) is the relative position of particle \(i\) with respect to the center of the rearrangement. (iv) The energy difference before and after the rearrangement \(E_{loc}\). We compare this
last quantity to another estimate of the characteristic energy, obtained in [31] by fitting $A_1(T_a)$ by an Arrhenius behavior, where $A_1$ is the prefactor of the pseudo-gap $D_L(\omega) = A_4 \omega^4$ that appears after our temperature cycle.

Our results are presented in Fig. 1 (left column) (see SI for the whole distributions): the vanishing scale of particle displacement $\delta_{loc} \sim \omega_c$ is shown in (a), and $E_{loc} \sim \delta_{loc}^3$ is tested in panel (b). It is found to be slightly smaller but comparable to the previously reported quantity $E_{loc}$ [31]. $N_{loc} \sim 1/\delta_{loc}$ is tested in panel (c), and $\ell_{loc} \sim 1/\sqrt{\delta_{loc}}$ in (d). Overall, we find a very good agreement between predictions and measurements.

Regular ultrastable glasses. In regular glasses, the density of QLMs does not display a gap [19–21], thus $\omega_c$ cannot be readily measured. Indeed at any finite temperature a gap must necessarily fill up in finite dimensions [31], making the characteristic frequency $\omega_c$ hard to extract. Yet, the local scaling relationships that follow from Eq. (3) can be tested. Configurations equilibrated by swap at some different parent temperature $T_P$ are taken from [27] using the specific liquid model of [45]. We instantaneously quench these configurations from $T_P$ to $T_P/3$, followed by a small cooling rate $\dot{T}$. This procedure is used so as to minimize the number of modes that are close to an instability. Thus it limits the number of excitations that go to a lower energy state upon temperature cycling, and allows us to have a better statistics on the excitations increasing energy that we consider here.

The excitations of these inherent structures are then probed by cycling temperature as above, at three different low $T_a$ for a short time $t_a$, chosen so as to obtain a collection of a few hundreds excitations in all systems for a given parent temperature. $N_{loc}$, $E_{loc}$, $\ell_{loc}$ and $\delta_{loc}$ are then extracted. As shown in Fig. 1, right column, we again find a very good agreement with our predictions. It supports our central claim: the geometrical description of localized excitations that follows from Eq. (3) holds in regular glasses.

Our second claim is that the increased stability upon cooling predicted by mean-field methods (corresponding to a growing characteristic frequency $\omega_c(T_P)$ upon cooling), together with our scaling relations Eq. (3), imply that in regular glasses local excitations must then become less extended and involve fewer particles—precisely as has been observed [20–22, 24–27], and confirmed in Fig. 1(g,h). We further predict that the characteristic energy of excitations should rapidly increase upon cooling and that the displacement decreases, as confirmed in Fig. 1(e,f).

Note that a (crude) estimate of $\omega_c(T_P)$ can be obtained by comparing the displacement magnitude $\delta_{loc}$ of the lowest-energy excitations in our samples, to those of gapped samples. It corresponds to comparing Figs. 1(a) and (e), and leads to a rapidly growing characteristic frequency upon cooling as shown in SI.

String-like hopping processes. Differences between QLMs and excitations have recently been emphasized in [46]. Here we propose the following explanation for this decoupling: for very stable glasses, excitations move particles by a distance of order of the particle size. At that point, the granularity of the materials matters: particles exchange position and excitations become string-like. This granularity needs not affect the linear response, causing the observed decoupling between excitations and QLMs.

This view is consistent with our previous observations that excitations become string-like in glasses with a large gap $\omega_c$ [31]. We now show that the same phenomenon
occurs for stable regular glasses, thus providing a simple explanation for the existence of strings (which differs from that of [47], in which strings are not elementary excitations but consist of many of them, unlike what our data support). Direct visualization of excitations signals particles taking the positions of their neighbors at low $T_p$, as exemplified in Fig. 2. As discussed in the SI, this effect is generally quantified by extracting the number of particles $n_p(r_c)$ replacing the position of others, i.e. the number of pairs for which $||R_i + \delta R_i - R_j|| < r_c d_0$, where $d_0$ corresponds to the peak of the pair distribution function and $r_c$ is some cut-off (details in SI). Fig. 2(d) shows the probability that an excitation has at least one such exchange, Fig. 2(e) shows $(n_p)$ averaged on all excitations considered. Both quantities strongly increase upon cooling. Here, the temperature cycle was tuned such that the number of excitations per particle considered is independent on $T_p$, and approximately $1.5 \times 10^{-5}$.

We find that strings only appear distinctly for the lowest temperatures probed, corresponding to $\delta_{\text{loc}} \approx 0.5$. In the gapped glasses they occurred only for the largest gap we probed [31], also corresponding to $\delta_{\text{loc}} \approx 0.5$ following Fig. 1(a). For less stable glasses, according to Fig. 1(a,e) the particle displacements are much smaller than the inter-particle distance, and particles thus cannot exchange position.

![Figure 2](image_url)

Figure 2. Top: examples of displacement fields (2D projection, zoomed $\times 1.7$, in units of $d_0$) for (a,b) lowest and (c) highest $T_p$. Bottom: (d) fraction of string-like motions $p_s$ and (e) number of permuting particles averaged on elementary excitations $(n_p)$ as a function $T_p$. Note that (d,e) involve integration up to a cut-off distance $r_c$, see SI for its definition and robustness.

**Conclusion** We have developed a scaling description for the architecture of local excitations in glasses, expressed in terms of the distance to an elastic instability where their characteristic length diverges. In gapped glassed obtained with breathing particles, this distance is embodied in magnitude of the gap $\omega_c$. Yet this description holds quantitatively in regular glasses as well, where a gap cannot be identified—and in fact cannot exist at finite temperature in finite dimension [31].

Using the mean-field result that the gap $\omega_c$ grows upon cooling together with our arguments explains why excitations become less extended upon cooling, and leads to two other confirmed predictions. First, excitations have larger displacements in stable glasses. It plausibly explains why strings only appear in that case, since near an instability the characteristic particle displacement of excitations is much smaller than the interparticle distance, and particles cannot exchange. It also supports that such hopping processes are elementary local excitations, strengthening the connection between these phenomena. Second, we predict a rapidly growing low energy scale for local excitations, corresponding to two decades in the temperature range probed as apparent in Fig. 1(f). The density of two-level systems should be diminished by this growing energy, as observed numerically [26, 46], since it implies a larger tunneling barrier that will eventually become hard to overcome by quantum fluctuations on experimental time scales [31].

Note that our mean-field arguments appear to yield valid exponents in three-dimensional simulations. This situation is reminiscent of the jamming literature [12, 33], and suggests that structural disorder in glasses induces limited heterogeneities in their elastic properties. It would be interesting to design a Ginzburg criterion, in the spirit of [48], to estimate beyond which length scale finite dimensional effects could be detectable.

The differences between QLMs and excitations would also be interesting to study further. In the SI, we show that the length scale $\ell_c$ below which continuum elasticity breaks down when a force dipole is exerted [41] and known to characterize the core of QLMs [23, 27] indeed decouples from the excitations length scale $\ell_{\text{loc}}$ in stable glasses (gapped or regular).

Looking forward, the connection between hopping processes and elementary excitations may give a new handle to study relaxation in supercooled liquids. $E_{\text{loc}}$ is a lower cut-off on the distribution of the excitations’ energy: quasi-localized modes with frequency $\omega > \omega_c$ are present in the material that will cause excitations with higher energy and larger displacements. This effect is apparent in Fig. 1(f): higher energy excitations are indeed triggered by larger ‘activation’ temperatures $T_a$. Likewise, in a supercooled liquid, at short times we expect the lowest-energy excitations to relax, corresponding to the so-called $\beta$-relaxation. At longer times, higher energy excitations with larger displacements (and thus more likely to be string-like) must relax too. There is indeed numerical evidence that such elementary building blocks add-up to ultimately form compact rearrangements [49–51] corresponding to the $\alpha$-relaxation. In that view, the decrease of the spatial extension $\ell_{\text{loc}}$ upon cooling does not contradict the common observation that dynamical heterogeneities increase in extension upon cooling. Indeed we expect that at low temperatures, local excitations...
such as strings accumulate in regions bigger than their size where the material is elastically soft (e.g. surrounding locally favored structures, that can be hard to detect by simple correlation functions [52] but may be captured by measurements as performed by [53], employing point-to-set correlations). Such a relationship between softness and relaxation is in fact well known [54].

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Supporting information – Geometry of hopping processes and local excitations in glasses

Parameters

We list all parameters beyond those listed in [27, 31, 45]. For gapped glasses we use ensembles comprising $n = 10^3$ samples at $N = 8000$ particles in three dimensions for four different gap frequencies $\omega_c$ ($\omega_c \equiv \omega_{-} \omega_D$, see below for the definition of $\omega_D$), prepared by [31]. For regular glasses we use ensembles comprising $n = 10^4$ configurations at $N = 2000$ particles in three dimensions for six different parent temperatures $T_p$, prepared by [27]. The relevant parameters are listed in Table I, where, in addition to the parameters described in the text:

- $d_0$, the typical inter-particle distance, is defined as the peak in the particle-particle correlation function.
- $n_{\text{all}}$ is the total number of excitations triggered using temperature cycling; $n_{\text{pos}}$ is the number of excitations going to higher energy minima (‘positive’ excitations).
- $\omega_D = [18\pi^2 \rho / (2c_t^{-3} + c_i^{-3})]^{1/3}$ is the Debye frequency, with the particle number density $\rho \equiv N/V$ and $V$ the volume; $c_t = \sqrt{G/(m \rho)}$ and $c_i = \sqrt{(B + 4G/3)/(m \rho)}$ are the transverse and longitudinal velocity, related to the shear modulus $G$ and bulk modulus $B$; $m$ is the particle mass (taken equal for all particles).
- $u_0$ is the summation of pair interaction energy divided by $N$.

Note that $\omega_D$, $G$, $B$, and $u_0$ are obtained as average values of sample-to-sample fluctuating quantities.

The units in Table I are as follows. Length ($d_0$) is in units of $D_0$, the initial diameter of small particles in gapped glasses, and the diameter of smallest particles in regular glasses (particles sizes are inverse power law distributed, and $D_0$ is the smallest diameter i.e. the lower bound of the diameter distribution). Energy ($u_0$) is in units of $\epsilon$, the prefactor of the inter-particle interaction potential. Temperature ($T_g$ and $T_p$) is in units of $\epsilon/k_B$, where we set Boltzmann’s constant $k_B$ to 1. Time ($t_{\text{a}}$, $\omega_c^{-1}$, $\omega_D^{-1}$) is in units of $\sqrt{m D_0^3/\epsilon}$, where $m$ is the particle mass (equal for all particles). Bulk modulus $B$ and shear modulus $G$ are in units of $\epsilon / D_0^3$.

| $\omega_c$ | $\omega_D$ | $u_0$ | $d_0$ | $T_g$ | $t_{\text{a}}$ | $n_{\text{all}}$ | $n_{\text{pos}}$ | separation | $G$ | $B$ |
|----------|-------------|-------|-------|-------|-----------|-----------|-----------|------------|-----|-----|
| 1.64     | 17.794      | 2.0643| 0.738 | 0.15  | 500       | 655       | 442       | yes        | 21.488 | 78.591 |
| 1.19     | 18.686      | 4.4520| 0.918 | 0.07  | 500       | 915       | 175       | yes        | 18.570 | 73.843 |
| 0.85     | 18.698      | 5.3497| 0.963 | 0.03  | 500       | 1245      | 117       | yes        | 17.581 | 72.993 |
| 0.65     | 18.565      | 5.8704| 0.988 | 0.01  | 500       | 1823      | 95        | yes        | 16.840 | 72.599 |

| $T_p$    | $\omega_D$ | $u_0$ | $d_0$ | $T_g$ | $t_{\text{a}}$ | $n_{\text{all}}$ | $n_{\text{pos}}$ | separation | $G$ | $B$ |
|----------|-------------|-------|-------|-------|-----------|-----------|-----------|------------|-----|-----|
| 0.30     | 18.134      | 4.8245| 1.305 | 0.4   | 100       | 115,196,406| 95,169,360| no         | 14.267 | 44.032 |
| 0.35     | 17.718      | 4.9115| 1.305 | 0.1   | 100       | 197,328,432| 135,235,324| no         | 13.592 | 44.542 |
| 0.40     | 17.298      | 4.9870| 1.305 | 0.01  | 100       | 167,246,406| 80,127,260| no         | 12.930 | 44.982 |
| 0.45     | 16.841      | 5.0553| 1.305 | 0.005 | 100       | 283,417,532| 96,179,264| no         | 12.233 | 45.374 |
| 0.50     | 16.361      | 5.1147| 1.305 | 0.003 | 100       | 411,560,794| 121,181,294| no         | 11.523 | 45.726 |
| 0.55     | 15.928      | 5.1668| 1.305 | 0.001 | 100       | 280,450,798| 69,107,264| no         | 10.907 | 46.028 |

Note, furthermore, that: (i) In preparing regular glasses, a protocol is adopted where we instantaneously quench to $T_p/3$, and then slowly quench rate at a rate $T = 10^{-3}$ so that the fraction of ‘positive’ excitation is not low (see Table I). We checked that if the glasses are instead prepared by instantaneous quench (like we do for the ‘breathing’ particles), not more than 5% of excitations are ‘positive’ excitations at the highest $T_p$ we consider, which is inefficient to obtain good statistics. (ii) Since less than 10% of samples rearrange in regular glasses, we assume that each rearrangement is an elementary excitation, and we do not apply our separation algorithm [31]. (iii) For the ‘breathing particles’ the pressure is fixed to a constant value. This is why $d_0$ is different at different $\omega_c$, and we adopt the notation $V \equiv \langle V_s \rangle$, with $V_s$ the volume of the individual samples. In regular glasses, instead, the volume is fixed to a constant value.
Quartic term distribution for quasi-localized modes

Here we show that the participation ratio $P_n$ of quasi-localized modes is proportional to the inverse of the coefficient $\chi$ of quartic term along quasi-localized modes, both in gapped glasses and in regular glasses. In particular, we have explicitly $NP_n \equiv 1/\sum_i |\Psi_i|^4$ where $i$ is the eigenmode component on the $i$th particle. $\chi \equiv (\partial^4 U)_{ijkl} \Psi_i \Psi_j \Psi_k \Psi_l$ where the $(\partial^4 U)_{ijkl}$ is the fourth order (spatial) derivative of the total interaction potential energy, it is a rank four tensor of size $(Nd)^4$. For details see [31, 56]. The scatter plots in Fig. 3, show that at low $NP_n$ the scaling is consistent with $NP_n \sim 1/\chi$. In gapped glasses, 25 samples at $N = 32000$ are used to calculate $NP_n$ and $\chi$, and these samples are also used in section .

![Figure 3. $NP_n$ vs $1/\chi$ in gapped glasses (left) and in regular glasses (right).](image)

Probability distribution functions for hopping processes

Fig. 4 shows the distributions of the bare quantities $NP_r$, of the energy difference $E_{12}$, of the length $\sqrt{I}$, and $||\delta R||/\sqrt{NP_r}$ of thermally-activated rearrangements, i.e., ‘hopping processes’. The medians are shown using a vertical line, defining $N_{loc} \equiv NP_r$, $E_{loc} \equiv E_{12}/u_0$, $\ell_{loc} \equiv 2\sqrt{I}/d_0$, and $\delta_{loc} \equiv \text{median}(||\delta R||)/(N_{loc}d_0)$, these values are reported in Fig. 1. Data are taken with the conditioning on excitations going to higher energy state: $E_{12} > 0$ both in gapped glasses (first row) and regular glasses (second row). Distributions are peaked around a maximum, except for the energy distribution whose distribution is maximum in zero (corresponding to symmetric double wells).

![Figure 4. The first row shows the corresponding distributions for gapped glasses. The second row shows the distributions for regular ultrastable glasses at highest number of excitations $N_p$ we have at each $T_p$. The solid vertical lines indicate the median values $N_{loc}$, $E_{loc}u_0$, $\ell_{loc}d_0/2$, and $\delta_{loc}d_0$ that are discussed in main text.](image)
Characterization of string-like hopping processes

To characterize string-like motion in regular glasses, we use the distinct part of the Van Hove correlation [16]:

\[
G_d(R, t_a) = \frac{1}{N} \left\langle \sum_{i=1}^{N} \sum_{j \neq i} \delta(R - R_j(t_a) + R_i(0)) \right\rangle
\]  (4)

where \( R_j(t_a) \) is the position of particle \( j \) at an instance in time \( t_a \). We already extensively report this quantity in [31] for gapped glasses, and report it here for regular glasses for different \( T_p \) in Fig. 5(left), whereby we radially average and define \( r = ||R|| \). The peak at \( r = 0 \) corresponds to ‘strings’: the fraction of particles that replace another particle (that ‘permute’). The result can be interpreted as the number of permuting particles:

\[
\langle n_p \rangle \equiv \left\langle N \int_{0}^{r_c} G_d(r, t_a) 4\pi r^2 dr \right\rangle
\]  (5)

where \( r_c \) is a cutoff that is tuned, based on the inset of Fig. 5(left), and show its influence to the results reported in the main text in the right panels of Fig. 5. Note that we consider the highest \( T_a \) from Table 1 for each \( T_p \), which corresponds to a fraction of particles that rearrange \( e_p \equiv n_{pos} / (nN) \) around \( 1.5 \times 10^{-5} \). As a reminder: the excitations are conditioned to going to a higher energy state.

![Figure 5](image)

**Figure 5.** Left: Van Hove correlation \( G_d(r, t_a) \) (radially averaged) at three lowest \( T_p \), in which the fraction of particles that rearrange \( e_p \) is around \( 1.5 \times 10^{-5} \). At large \( r \), \( G_d \) is equal to the particle number density \( \rho \equiv N/V \). The peak around \( r = 0 \) corresponds to particles that replace to another (see inset for a log-log plot of this peak). Right panels: Fig. 2(d,e) in the main text at \( r_c/d_0 = 0.05 \), complemented here with data at \( r_c/d_0 = 0.1 \).

**Crude estimation of \( \omega_c \) in regular glasses**

In this section, we estimate \( \omega_c \) in regular glasses. We suppose in gapped glasses and regular glasses \( \omega_c \) varies with \( \delta_{loc} \) in the same way. Since we know both \( \omega_c \) and \( \delta_{loc} \) in regular glasses, we fit the data by \( \ln(\omega_c) = c_1 + c_2 \ln(\delta_{loc}) \) to extract \( c_1 \) and \( c_2 \). We employ them to get an estimate of \( \omega_c \) at the lowest energy excitations (lowest \( \delta_{loc} \) at each \( T_p \)). The results, in Fig. 6, \( \omega_c \) increases with decreasing \( T_p \), as we expect.

![Figure 6](image)

**Figure 6.** The estimated \( \omega_c \) vs \( T_p \), in regular glasses.
Measurement of $\ell_c$

To extract the typical global length $\ell_c$, we perturb the glasses with a local dipole force and look at the correlation function $c(r)$ which is defined in as [27, 41]. $\ell_c$ is defined as the length where rescaling $c(r)$ collapses the data, see Fig. 7. Note that for this global measurement we use a bigger system (25 samples at $N = 32000$; whereby we checked that these $\ell_c$ collapse the rescaled $c(r)$ at $N = 8000$ as well, except for a small difference at our smallest gap) at each $\tilde{\omega}_c$ in gapped glasses, and 100 samples at $N = 2000$ at each $T_p$ in regular glasses to extract $\ell_c$.

![Image](image1)

Figure 7. Correlation function $c(r)$ of the response to a dipole force response (left) and rescaling to extract $\ell_c$ (right), for gapped glasses (top) and regular glasses (bottom). Note that we find for gapped glasses $A_1 = \{1.1, 6.2, 10, 15\}$, and for regular glasses $A_1 = \{1, 1.2, 1.4, 1.6, 1.9, 2.3\}$.

In Fig. 8, we show that $\ell_c$ decouples from the excitation length scale $\ell_{\text{loc}}$ in stable (i.e. gapped and regular) glasses, at large $\delta_{\text{loc}}$.

![Image](image2)

Figure 8. Fig. 1(d,h) from the main text with superimposed $\ell_c$ (open markers). $\ell_c$ in regular glasses is shown as a function of $\delta_{\text{loc}}$ for the smallest $T_a$ for each parent temperature $T_p$ (for that reason we only show $\ell_{\text{loc}}$ for those $T_a$). Notice that, like $\ell_{\text{loc}}$, $\ell_c$ is reported in units of $d_0$. 
