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LETTER

# Heterogeneous dynamics, marginal stability and soft modes in hard sphere glasses

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**Abstract.** In a recent publication we established an analogy between the free energy of a hard sphere system and the energy of an elastic network. This result enables one to study the free energy landscape of hard spheres, which was previously accessible only via density functional theory. In our formalism normal modes can easily be defined and computed. In this work we use these tools to analyze the activated transitions between meta-basins, both in the ageing regime deep in the glass phase and near the glass transition. We observe numerically that structural relaxation occurs mostly along a very small number of nearly unstable extended modes. This number decays for denser packing and is significantly lowered as the system undergoes the glass transition. This observation supports the assertion that structural relaxation and marginal modes share common properties. In particular, theoretical results show that these modes extend at least on some length scale  $l^* \sim (\phi_c - \phi)^{-1/2}$  where  $\phi_c$  corresponds to the maximum packing fraction, i.e. the jamming transition. This prediction is consistent with very recent numerical observations of dynamical length scales in sheared systems near the jamming threshold, where a similar exponent is found, and with the commonly observed growth of the rearranging regions with compression near the glass transition.

**Keywords:** Brownian motion, disordered systems (theory), dynamical heterogeneities (theory), slow relaxation and glassy dynamics

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A colossal effort has been made to characterize the spatial nature of the structural relaxation near the glass transition. Numerical simulations [1] and experiments [2, 3] have shown that the dynamics in supercooled liquids is heterogeneous and becomes more collective as the glass transition is approached. Both the string-like [4] and the compact [5] aspects of the particle displacements have been emphasized. Nevertheless, the cause of such collective motions remains debated [6, 7]. To make progress, one would like to relate these motions to other objects. A possible candidate is the excess of low frequency modes present in all glasses, the so-called boson peak [8]. Because these modes shift in general to lower frequencies as the temperature increases toward the glass transition temperature  $T_g$ , it has been proposed that they are responsible for the melting of the glass [9, 10]. This suggests the use of widely employed tools, such as the low frequency instantaneous normal modes [11] or the negative directions of saddles of the potential energy landscape [12], to analyze the collective motions causing relaxation. Nevertheless, this approach has the major drawback of being based on energy instead of free energy. As such, it cannot be applied for example to hard spheres or colloids, where structural relaxation is also known to be collective; see e.g. [3]. In this case barriers between metastable states are purely entropic. More generally, one expects entropic effects to be important for glasses where hard core repulsions and non-linearities are not negligible, which is presumably the case in general above  $T_g$  [13].

Recent developments make this analysis possible for hard sphere systems. In [14], we derived an analogy between the free energy of a hard sphere glass and the energy of a weakly connected network of logarithmic springs. This allows us to define normal modes, that can be compared with the dynamics. Furthermore, recent results [15]–[17] valid for weakly connected networks, such as elastic particles near jamming [18, 19]—where scaling laws relating packing geometry and vibrational properties were first observed—or simple models of silica [20], apply for characterizing these modes: (i) excess modes appear above some frequency  $\omega_{AM}$  which depends on the pressure  $p$  and the coordination  $z$ , whose definition will be recalled below for hard spheres. These *anomalous modes* extend at least on a length scale  $l^*$ , which depends on  $z$  and diverges near maximum packing [16, 17, 19]. The predicted dependence of  $l^*$  was checked in [21] by considering the response to a point force. (ii) Metastable states can exist only if they contain a configuration for which  $\omega_{AM} > 0$ . This leads to a non-trivial scaling relationship between  $p$  and  $z$  that must be satisfied in the glass phase. Numerically, we observed that the hard sphere glass lies close to marginal stability: the coordination is just sufficient to maintain rigidity [14]. This implies that anomalous modes are present at very low frequency.

In this work, we study how low frequency modes take part in the structural relaxation, both during the ageing dynamics deep in the glass phase, and in the vicinity of the glass transition where the system is at equilibrium. We show that when relaxation occurs between metastable states, the system yields in the direction of the softest modes: most of the amplitude of the observed displacements can be decomposed on a small fraction of the modes, of the order of few per cent. This observation supports that the collective aspect of the relaxation does not stem from the non-linear coupling of localized relaxation events, but rather from the extended character of the softest degrees of freedom. This suggests that the typical size of the events relaxing the structure increases as the extension of the anomalous modes  $l^* \sim (\phi_c - \phi)^{-1/2} \sim p^{1/2}$ , which diverges deep in the glass phase.

We start by recalling some results of [14]. In a metastable state of a hard sphere system, one can define a contact network [22]: two particles  $i, j$  are said to be in contact if they collide during some interval of time  $t_1$ , where  $t_1$  is chosen to be much larger than  $\tau_c$ , the collision time, and smaller than the structural relaxation time  $\tau$  where metastability is lost. The contact force  $f_{ij}$  is defined as the time average rate of momentum exchange in collisions between  $i$  and  $j$ . We define the average contact force  $\langle f \rangle \equiv \langle f_{ij} \rangle_{ij}$ , where  $\langle \rangle_{ij}$  denotes an average over all contacts. The coordination number  $z$  of this network is defined as the average number of contacts of the particles in the system. An approximation of the Gibbs free energy  $\mathcal{G}$  can then be expressed as a sum over all the contacts  $\langle ij \rangle$ :

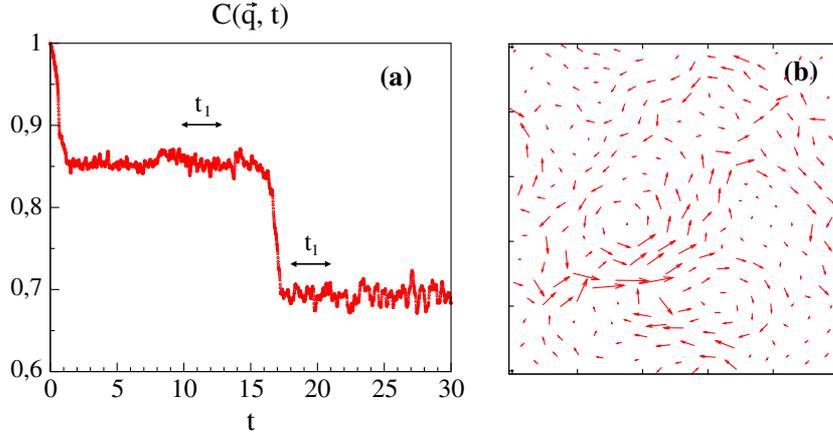
$$\mathcal{G} = -kT \sum_{\langle ij \rangle} \ln(\langle h_{ij} \rangle_t) \quad (1)$$

where  $h_{ij} = r_{ij} - r_i - r_j$  is the gap between particles  $i$  and  $j$ ,  $r_{ij}$  is the distance between them,  $r_i$  denotes the radius of particle  $i$ , and  $\langle \rangle_t$  is a time average. Equation (1) has two main limitations: (i) it is only exact near the maximum packing fraction  $\phi_c$  where the pressure diverges and (ii) to perform the time average one requires a strong separation of time scales between  $\tau_c$  and  $\tau$ . Thus equation (1) is a better approximation deep in the glass phase. Nevertheless the corrections to equation (1) are found to be rather small empirically [14, 17], and we shall use equation (1) to study the vicinity of the glass transition ( $\phi \approx \phi_0$ , where  $\phi_0$  is the packing fraction above which equilibrium cannot be reached with accessible numerical time scales) as well.

Equation (1) can be expanded around any equilibrium position<sup>3</sup>. For a contact  $ij$ , one finds for the force  $V'_{ij} = -kT/\langle h_{ij} \rangle_t$  and for the stiffness  $V''_{ij} = kT/\langle h_{ij} \rangle_t^2$ . This enables one to compute the dynamical matrix  $\mathcal{M}$  [23] which relates a small applied force to the linear displacement of the average particle positions. Normal modes can then be computed, whose angular frequencies are the square roots of the eigenvalues of  $\mathcal{M}$ . In what follows we locate quiet periods of the dynamics where  $\mathcal{M}$  can be estimated. Then, we use the normal modes to analyze the subsequent structural relaxation.

We consider a bidisperse two-dimensional hard sphere system. Half of the particles have a diameter  $\sigma_1 = 1$ , the other a diameter 1.4; their mass is  $m = 1$ , and energies are expressed in units of  $kT$ . To study the ageing dynamics, configurations are generated in the glass phase ( $\phi_0 \approx 0.79 \leq \phi \leq \phi_c \approx 0.84$ ) as in [14]. An event-driven code is used to simulate the dynamics. We observe long quiet periods, or metastable states, interrupted by sudden rearrangements, or ‘earthquakes’. Such earthquakes correspond to collective motions of a large number of particles, and have been observed in various other ageing systems, such as colloidal paste and laponite [24], and in Lennard-Jones simulations [25, 26]. Even for our largest numerical box of  $N = 1024$  particles, deep in the glass phase these events generally span the entire system. They appear as drops in the self-scattering function  $C(\vec{q}, t) \equiv \langle \exp[i\vec{q} \cdot (\vec{R}_i(t) - \vec{R}_i(0))] \rangle_i$ , where  $\langle \rangle_i$  is an average over all particles and  $\vec{R}_i(t)$  is the position of particle  $i$  at time  $t$ . An example of earthquake is shown in figure 1.

<sup>3</sup> In a metastable state the average contact forces must be balanced on each particle; otherwise particles would accelerate. Within the approximation of equation (1), which estimates forces within a few per cent accuracy throughout the glass phase [14], this implies that the average configuration in a metastable must be at a minimum of  $\mathcal{G}$ , since forces are balanced only for such configurations.



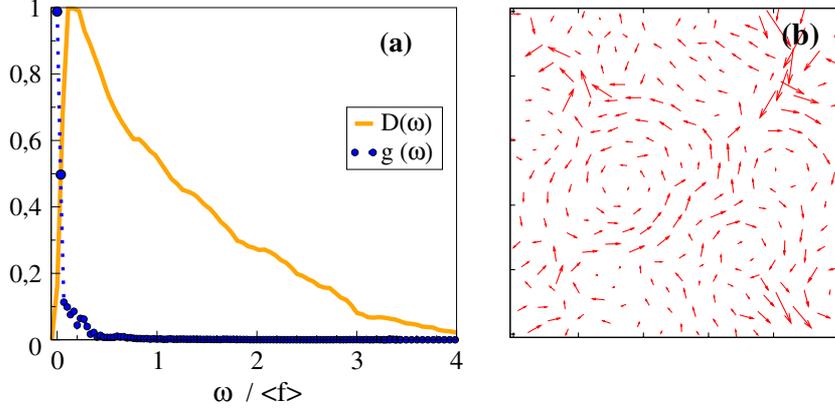
**Figure 1.** Left: self-density correlation function  $C(\vec{q}, t)$  versus time (expressed in units of  $\sigma\sqrt{m/kT}$ ) for  $q = 2\pi/\sigma_1$  in a system of  $N = 256$  particles, at packing fraction  $\phi = 0.837$ . Metastable states appear as plateaus of  $C(\vec{q}, t)$ , whereas the drops of  $C(\vec{q}, t)$  are the aforementioned earthquakes. Time averages are made during the time segments  $t_1$ . Right: displacement field of the corresponding earthquake. Arrows connect the average particle positions before and after the earthquake, they are amplified four times here for visibility. For similar data for a 3D LJ case see [25].

In what follows the average particle position in a metastable state  $l$  is denoted as  $|R^l\rangle \equiv \{\langle \vec{R}_i \rangle_t\}, i = 1, \dots, N$ . In practice the time averaging  $\langle \rangle_t$  is over a long time  $t_1$  corresponding to few hundred collisions per particle (we use  $t_1 = 10^5, 5 \times 10^4$  numerical time steps for respectively  $N = 1024$  and  $256$  particles). The earthquake displacement field  $|\delta R^e\rangle$ , between two metastable states  $l$  and  $m$ , is then defined as  $|\delta R^e\rangle \equiv |R^m\rangle - |R^l\rangle$ ; see figure 1(b). During earthquakes, we find that the average particle displacement is typically 10%–20% of the particle diameter, and tends to decrease with the pressure.

To analyze these displacement fields, we compute the average of the particle positions and the contact network in the metastable state prior to the earthquake<sup>4</sup>. This enables us to define  $\mathcal{M}$  and the normal modes  $|\delta R^\alpha\rangle$ , where the label  $\alpha = 1, \dots, 2N$  ranks the modes in order of increasing frequencies  $\omega^\alpha$ . An example of the density of states  $D(\omega)$  is shown in figure 2(a), and the lowest frequency mode in figure 2(b). We indeed observe extended anomalous modes at very low frequencies, in agreement with the marginal stability inferred from the microscopic structure of the glass [14]. Note that we occasionally observe a few unstable modes even deep in the glass phase, implying the presence of saddles (and multiple configurations of free energy minima) or ‘shoulders’ in the metastable states we are considering. In the present work we do not focus on this aspect, and treat unstable modes the same as the rest.

We then project the earthquake displacement  $|\delta R^e\rangle$  on the modes and compute  $c_\alpha = \langle \delta R^e | \delta R^\alpha \rangle / \langle \delta R^e | \delta R^e \rangle$ , where  $\langle \delta R^e | \delta R^\alpha \rangle \equiv \sum_i \delta \vec{R}_i^e \cdot \delta \vec{R}_i^\alpha$ . The  $c_\alpha$  satisfy  $\sum_\alpha c_\alpha^2 = 1$  since the normal modes form a unitary basis. To study how the contribution of the modes

<sup>4</sup> Very close to  $\phi_c$  (for  $\langle f \rangle > 5 \times 10^3$ ), ‘rattlers’ [18] are present, which are systematically removed from our analysis [14].



**Figure 2.** Left, straight curve:  $D(\omega)$  computed in the metastable state prior to the earthquake of figure 1 versus  $\omega/\langle f \rangle$ , the angular frequency rescaled with respect to the average contact force  $\langle f \rangle$ . Dotted curve:  $g(\omega)$  versus  $\omega/\langle f \rangle$ . Right: lowest frequency normal mode.

depends on frequency, we define  $g(\omega) = \langle c_\alpha^2 \rangle_\omega$ , where the average is over all  $\alpha$  such that  $\omega^\alpha \in [\omega, \omega + d\omega]$ . Figure 2(a) shows  $g(\omega)$  for the earthquake shown in figure 1. The average contribution of the modes decreases very rapidly with increasing frequency, and most of the displacement projects onto the excess modes present near zero frequency. This implies that the free energy barrier crossed by the system is located in the direction of the softest degrees of freedom.

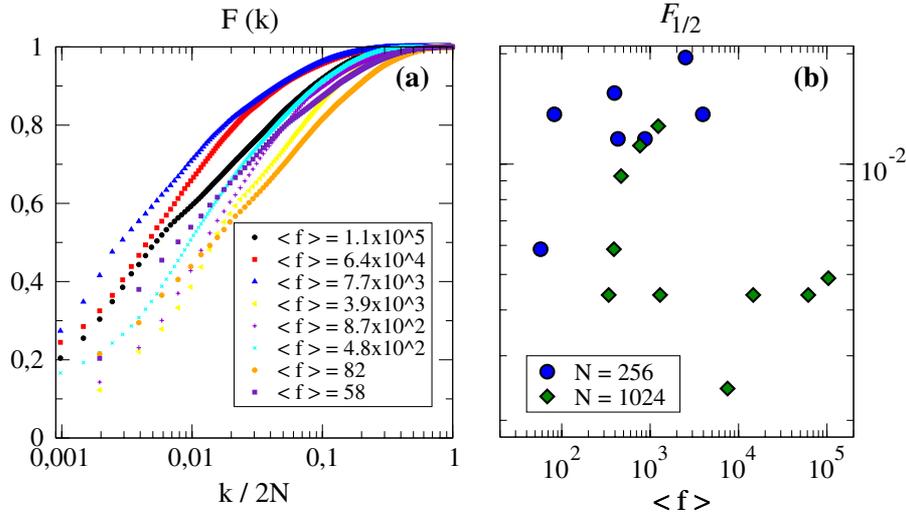
To make this observation systematic, we introduce the label  $i$  to rank the  $c$  values in decreasing order:  $c_1 > c_2 \cdots > c_{2N}$ . Then we define

$$F(k) \equiv \sum_{i=1}^k c_i^2. \quad (2)$$

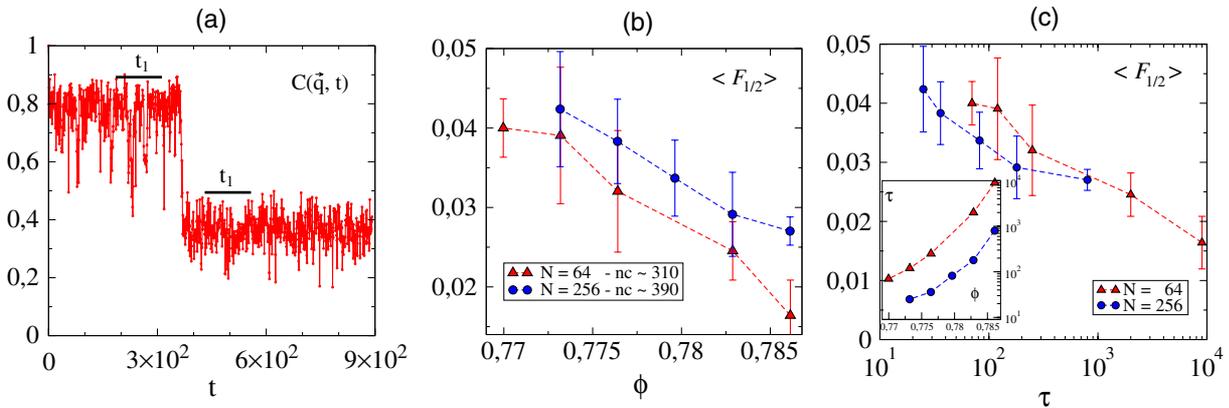
$F(k)$  indicates which fraction of the total displacement is contained in the  $k$  most contributing modes. If  $F(k) = 1 \forall k$  then only one mode contributes. If  $F(k) = k/2N$  all modes contribute equally. We then define  $k_{1/2}$  as the minimum number of modes contributing to 50% of the displacements, i.e. the smallest  $k$  for which  $F(k) > 1/2$ . Figure 3 shows  $F(k)$  and  $F_{1/2} \equiv k_{1/2}/(2N)$  for the 17 cracks studied. Figure 3(b) shows that  $0.2\% < F_{1/2} < 2\%$  for all the events studied throughout the glass phase. We thus systematically observe that the extended earthquakes correspond to the relaxation of a small number of degrees of freedom.

We now extend this analysis to an equilibrated supercooled liquid. We equilibrate at  $0.77 \leq \phi \leq 0.786$ . As previously observed e.g. in [5], the dynamics is heterogeneous in time, and sudden rearrangements still occur on time scales of the order of  $\tau$ , the time scale of the  $\alpha$  relaxation<sup>5</sup>. We use the procedure previously described to determine the average configuration of metastable states, and to define the displacement relaxing the structure; see figure 4(a). We start by studying five packing fractions in a system of  $N = 64$  particles. For this size, rearrangements generally span the entire system. A similar observation was

<sup>5</sup> We define  $\tau$  as the time for which  $C(\vec{q}, \tau) = 0.3$ .

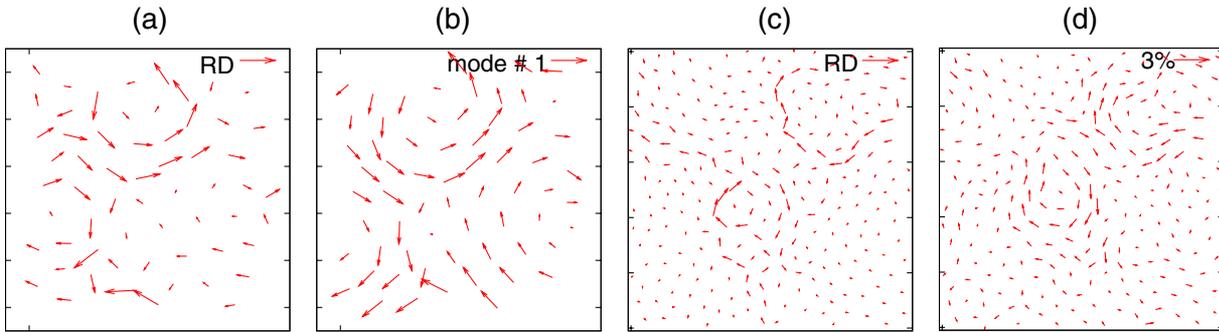


**Figure 3.** Left: examples of  $F(k)$  versus  $k/2N$  for systems with different average contact force  $\langle f \rangle$ .  $\langle f \rangle$  is measured before the earthquake. It is proportional to the pressure near  $\phi_c$ , and is of the order of 20 near the glass transition  $\phi_0$ . Right:  $F_{1/2}$  versus  $\langle f \rangle$  for  $N = 256$  (circles) and  $N = 1024$  (diamonds) particles.



**Figure 4.** (a)  $C(\vec{q}, t)$  for an equilibrated system of  $N = 64$  particles at  $\phi = 0.786$ . The segment  $t_1$  corresponds to  $10^4$  numerical time steps, which corresponds roughly to 300 collisions on average per particle. (b)  $\langle F_{1/2} \rangle$  versus  $\phi$  for  $t_1 = 5 \times 10^4$  (circles) for  $N = 256$  and  $t_1 = 10^4$  (triangles) for a system with  $N = 64$ . The average number of collisions per particle is written in the legend and denoted by  $nc$ . (c)  $\langle F_{1/2} \rangle$  versus  $\tau$  for two different system sizes. Inset: relaxation time  $\tau$  versus  $\phi$  for  $N = 64$  (triangles) and  $N = 256$  (circles).

made for a three-dimensional Lennard-Jones system of 125 particles [5]. For each packing fraction,  $F(k)$  and  $F_{1/2}$  are computed for seven relaxation events. Results for  $\langle F_{1/2} \rangle$  are presented in figure 4(b) as a function of packing fraction. We find that  $\langle F_{1/2} \rangle \leq 4\%$  for all  $\phi$  studied, implying that also in this region of the supercooled liquid phase a small fraction of the low frequency modes contribute to the relaxation events. Interestingly, this fraction



**Figure 5.** (a) Displacement field corresponding to the relaxation event of figure 4(a). Arrows were multiplied by 1.2. (b) Lowest frequency normal mode, which has the highest projection in this particular case. In this example one mode contributes most of the displacement. (c) Relaxation event in a system of  $N = 256$  particles at  $\phi = 0.786$ . (d) Projection of the displacement (c) on the 3% of the modes that contribute the most to it.

decays significantly as the relaxation time grows, suggesting a rarefaction of the number of directions along which the system can yield near the glass transition. This is supported by other numerical approaches [27, 28]. For the largest  $\phi \approx 0.786$  we studied,  $\langle F_{1/2} \rangle \leq 2\%$ , which implies that the collective event relaxing the system corresponds mainly to one or two modes. Figures 5(a) and (b) compare one event and the mode that contributes most to it, which turns out to be the softest mode in this particular example.

*Size effect:* We now consider a system of  $N = 256$  particles. An example of relaxation is shown in figure 5(c). We observe for this system size that a larger fraction of the particles stay motionless. It is also interesting to compare for the same system size the ageing dynamics deeper in the glass phase; e.g. figures 1(b) with 5(c): as was previously observed for LJ systems [25], the collective rearrangements at equilibrium involve fewer particles than earthquakes, but move them more. Nevertheless in the equilibrated case as well, we shall see now that the observed displacement projects on a very small fraction of the normal modes. To study this question we perform the analysis introduced above, and compute  $\langle F_{1/2} \rangle$  by averaging on twelve events for the five packing fractions considered. Results are shown in figure 4(b): they are qualitatively similar to those for the system with  $N = 64$ , but the values of  $\langle F_{1/2} \rangle$  are larger by approximately 0.5%. Nevertheless, it is well known that the glass transition occurs at lower packing fraction for smaller systems [29]; see the inset of figure 4(c). When plotted versus relaxation time, see figure 4(c), the difference between the values of  $\langle F_{1/2} \rangle$  in the two systems is reduced and less systematic, and  $\langle F_{1/2} \rangle$  becomes in fact smaller for  $N = 256$ . Thus, even when embedded in a system containing quiet regions, relaxation occurs along the softest modes.

For kinetically constrained models [30], that have been proposed as paradigms for glassy dynamics, heterogeneous dynamics can arise from simple local microscopic rules. Our result that collective rearrangements correspond mostly to a few (and therefore necessarily reasonably extended) modes supports an alternative view: elementary relaxation events are already extended objects, as are the soft degrees of freedom of the system. We now argue that these modes are the anomalous modes [15, 16] described in the introduction. Our justification for this lies in the microscopic structure of the glass: as

evoked above, imposing the marginal stability of these modes leads to a power-law relation between coordination and pressure that is indeed observed in the glass phase [14]. If other soft objects (e.g. local configurations particularly soft due to disorder) were driving the dynamics, it is unlikely that the glass would freeze in this specific region of the phase space<sup>6</sup>.

More needs to be known on the statistical properties of the anomalous modes, for example on the curly aspect of their displacement field or on their apparent capacity to form intense flow lines or strings. This problem turns out to be equivalent to the statistics of force chains in amorphous solids [31], a much studied problem in the granular literature, but which still lacks a definite answer. Nevertheless, we know that the anomalous modes are characterized by some length scale  $l^* \sim p^{1/2}$  above which the softest modes must extend [15]. The observation that the softest modes dominate the relaxation supports the assertion that regions of size at least  $l^* \sim p^{1/2} \sim (\phi_c - \phi)^{-1/2}$  must rearrange. Very recent numerical work on sheared systems near their jamming threshold [32] tend to support our views, as in this case rearrangements are characterized by a diverging length scale with an exponent  $0.6 \pm 0.1$ , consistently with the idea that relaxation occurs along anomalous modes. This is also consistent with the growing dynamical length scale observed near the glass transition. Nevertheless in this case the length scales at play, typically about 5 or 10 particles, are too small for comparing different theories [33]. More stringent tests could be performed near maximum packing, where a diverging length scale is expected. This could be tested experimentally, e.g. by considering the intermittent ageing dynamics of colloids at large osmotic pressures.

Our work supports a unified description of the structural relaxation and the packing geometry, where the dynamics corresponds to the collapse of anomalous modes, and where the microscopic structure is fixed by their marginal stability. Note that the theoretical framework used here applies identically in three dimensions, where we expect our results to be valid as well. This scenario may also hold for other glasses, for example in the Lennard-Jones case where anomalous modes are also present [34]. Nevertheless in this case, as for any long-range interaction potentials,  $l^*$  is bounded above and does not diverge in the glass phase.

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<sup>6</sup> This region may correspond to the critical surface of mode coupling theory, whose definition also depends on coordination; see [25, 26] and references therein.

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