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## Abstract

The excess low-frequency normal modes for two widely used models of glasses are studied at zero temperature. The onset frequencies for the anomalous modes for both systems agree well with predictions of a variational argument, which is based on analyzing the vibrational energy originating from the excess contacts per particle over the minimum number needed for mechanical stability. Even though both glasses studied have a high coordination number, most of the additional contacts can be considered to be weak.

## Disciplines

Physical Sciences and Mathematics | Physics

## Comments

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## Excess Vibrational Modes and the Boson Peak in Model Glasses

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The excess low-frequency normal modes for two widely used models of glasses are studied at zero temperature. The onset frequencies for the anomalous modes for both systems agree well with predictions of a variational argument, which is based on analyzing the vibrational energy originating from the excess contacts per particle over the minimum number needed for mechanical stability. Even though both glasses studied have a high coordination number, most of the additional contacts can be considered to be weak.

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Our understanding of liquids is based on the idea that liquid structure is largely determined by strong short-ranged repulsions and that the longer-ranged attractions can be treated as a perturbation [1]. Similar considerations were used to study jamming at zero temperature as a function of density  $\phi$  [2,3]. When potentials are repulsive and have a finite range, there is a sharp jamming transition at  $\phi_c$ . This Letter ties together two systems: it connects the marginally jammed state just above  $\phi_c$  of granular packings [2,3] to the boson peak of glasses [4–15]. The latter peak is believed to be key to the mysterious, apparently universal, low-temperature properties of amorphous solids. We find that the longer-ranged part of the interaction for more realistic interatomic potentials gives only small corrections to the behavior of the marginally jammed solid. Our result can be viewed as a conceptual extension of the perturbation theory of liquids to the case of jamming.

In the marginally jammed state, the low-frequency normal modes of vibration are fundamentally different from the long-wavelength plane waves expected from elasticity theory [2,3]. The unusual nature of the modes is reflected in the density of vibrational states  $D(\omega)$  versus frequency  $\omega$  [2,3]. At the transition,  $D(\omega)$  has a plateau extending to  $\omega = 0$ . This is very different from the expected Debye scaling  $D(\omega) \propto \omega^{d-1}$  in  $d$  dimensions [16], which is normally observed in solids. At densities above  $\phi_c$ , the plateau no longer extends to  $\omega = 0$  but terminates at a frequency  $\omega^*$ . The dramatic rise in  $D(\omega)$  at  $\omega^*$  corresponds to the onset of anomalous modes. Similar excess modes are observed in boson peaks of glasses.

In this Letter, we demonstrate that the theoretical framework [17–19] used to explain the modes in marginally jammed solids at zero temperature can be extended to the anomalous modes in two more realistic models of “repulsive glasses,” by which we mean systems that undergo glass transitions due to the repulsive part of their potentials as the temperature is lowered. The zero-temperature jamming transition of spheres with finite-ranged repulsions, which we will call point  $J$  to distinguish from other jamming transitions, coincides with the density at which the

system has just enough contacts to satisfy the constraints of mechanical equilibrium [2]. This is called the isostatic condition. The average coordination number  $z$  (i.e., the average number of particles with which a given particle interacts) needed for mechanical stability is  $z_c = 2d$ , where  $d$  is the spatial dimension [20,21]. By compressing the system or increasing the range of interaction, we increase  $z$ . These extra contacts suppress anomalous modes at low frequencies [17–19]. We will show that, in systems with long-ranged interactions, there is a well-defined division between a relatively few strong repulsive interactions (stiff contacts) and the more numerous weaker interactions (including attractions), which can be treated as a correction.

We will study the onset frequency of the anomalous modes  $\omega^\dagger$  in two widely used models of glasses and the glass transition and compare the simulation results with theoretical predictions [19]. We use a mixture of 800  $A$  and 200  $B$  spheres with equal mass  $m$  interacting in three dimensions via the Lennard-Jones potential [22]:

$$V(r_{ij}) = \frac{\epsilon_{ij}}{72} \left[ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^6 \right], \quad (1)$$

where  $r_{ij}$  is the separation between particles  $i$  and  $j$  and  $\epsilon_{ij}$  and  $\sigma_{ij}$  depend on the type of particles under consideration:  $\epsilon_{AB} = 1.5\epsilon_{AA}$ ,  $\epsilon_{BB} = 0.5\epsilon_{AA}$ ,  $\sigma_{AB} = 0.8\sigma_{AA}$ , and  $\sigma_{BB} = 0.88\sigma_{AA}$ . The potential is cut off at  $r_{ij} = 2.5\sigma_{ij}$  and shifted to satisfy  $V(2.5\sigma_{ij}) = V'(2.5\sigma_{ij}) = 0$ . For these systems, the density is given in units of  $\rho = N\sigma_{AA}^3/L^3$  and  $\epsilon_{AA}$ ,  $\sigma_{AA}$  and  $m$  are set to unity. We restrict the densities to lie above  $\rho \approx 1.2$ , where the pressure is positive. When this is not the case, there can be low-frequency modes arising from rather different physics.

For comparison we also study a system with purely repulsive interactions where  $\phi_c$  exists. We simulate a mixture of 500  $A$  and 500  $B$  spheres with  $\sigma_B = 1.4\sigma_A$  and equal mass  $m$ , interacting via the repulsive Lennard-Jones potential, [1]

$$V(r_{ij}) = \begin{cases} \frac{\epsilon}{72} \left[ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - 2 \left( \frac{\sigma_{ij}}{r_{ij}} \right)^6 + 1 \right], & r_{ij} < \sigma_{ij}, \\ 0, & r_{ij} \geq \sigma_{ij}, \end{cases} \quad (2)$$

where  $\epsilon$  is the characteristic energy, and  $\sigma_{ij} = (\sigma_i + \sigma_j)/2$ . For these systems, we characterize density with the packing fraction  $\phi = \pi/6(N_A\sigma_A^3 + N_B\sigma_B^3)/L^3$ . We set  $\epsilon = 1$ ,  $m = 1$ , and  $\sigma_A = 1$ . The simulations were all carried out in a cubic box with periodic boundary conditions. We study zero-temperature ( $T = 0$ ) configurations which were obtained by quenching initially random ( $T = \infty$ ) configurations of particles to their local energy minima at  $T = 0$  using conjugate gradient energy minimization [23].

In the harmonic expansion, the energy of particle displacements  $\delta\vec{R}_i$  from their equilibrium positions is

$$\delta E = \frac{1}{2} \sum_{i,j} \left[ V''(r_{ij}) (\delta\vec{R}_{ij} \cdot \hat{r}_{ij})^2 + \frac{V'(r_{ij})}{r_{ij}} (\delta\vec{R}_{ij}^\perp)^2 \right], \quad (3)$$

where  $V'$  and  $V''$  are the first and second derivatives of the pair potential  $V(r)$  with respect to  $r$ , and  $\vec{r}_{ij} = r_{ij}\hat{r}_{ij}$  is the equilibrium separation vector between particles  $i$  and  $j$ . Here,  $\delta\vec{R}_{ij} \equiv \delta\vec{R}_i - \delta\vec{R}_j$ , and  $\delta\vec{R}_{ij}^\perp$  is the projection of  $\delta\vec{R}_{ij}$  on the plane perpendicular to  $\hat{r}_{ij}$ . We diagonalize the dynamical matrix [16] to obtain normal modes  $|n\rangle$ , and their corresponding eigenvalues  $\mathcal{E}_n$ . The normal-mode frequencies are  $\omega_n^2 = \mathcal{E}_n$ , where the index  $n$  runs from 1 to  $Nd$ . From these frequencies, we calculate the density of vibrational states,  $D(\omega)$ . It is instructive to also calculate  $D^{(0)}(\omega)$ , obtained by neglecting the second term in Eq. (4) (i.e., the stress term). This corresponds to replacing  $V(r)$  with unstretched springs, each chosen to have the same stiffness  $V''(r)$  as for the original potential.

In Figs. 1(a) and 1(b), we show two curves,  $D(\omega)$  (heavy curves) and  $D^{(0)}(\omega)$  (light curves), corresponding to the stressed and unstressed cases, at each density. Figure 1(a) shows results for the repulsive Lennard-Jones systems. Close to the unjamming transition, these systems are approximately equivalent to the repulsive harmonic systems studied previously [2,3]. Just above the transition,  $D(\omega)$  has a plateau down to  $\omega = 0$ . Upon compression the onset of these anomalous modes shifts up to  $\omega^*$ , below which  $D(\omega)$  decreases to zero. Figure 1(b) shows the results for the system with Lennard-Jones interactions. Because there are attractive interactions, the jamming transition lies inside the liquid-vapor spinodal [2,24] and is inaccessible. Thus, the plateau in the density of states never extends to  $\omega = 0$ .

It is important to note that  $D^{(0)}(\omega)$  falls much more sharply than  $D(\omega)$  as  $\omega$  decreases below  $\omega^*$ , but that  $D(\omega)$  and  $D^{(0)}(\omega)$  are nearly indistinguishable above  $\omega^*$ . Here,  $\omega^*$  corresponds to the onset of the anomalous modes in unstressed systems. At high densities, the sharp peaks in  $D^{(0)}(\omega)$  below  $\omega^*$  arise from linear combinations of nearly degenerate long-wavelength plane waves. (The lowest

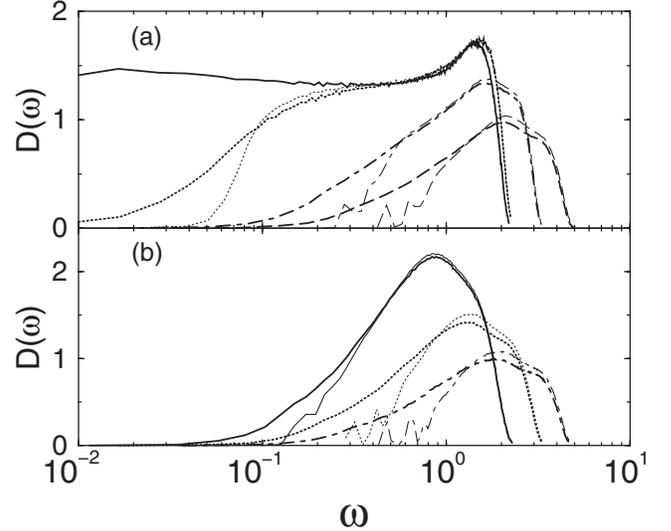


FIG. 1. Density of vibrational states of 3D glasses of  $N = 1000$  particles, averaged over 100 configurations. We plot  $D(\omega)$  for stressed systems (heavy curves) and  $D^{(0)}(\omega)$  for unstressed systems (light curves) interacting via (a) the repulsive Lennard-Jones potential at  $\phi - \phi_c = 10^{-6}$  (solid line),  $10^{-2}$  (dotted line), 0.1 (dot-dashed line), and 0.2 (dashed line); and (b) the Lennard-Jones potential at  $\rho = 1.2$  (solid line), 1.4 (dotted line), and 1.6 (dashed line).

peak has plane waves with wave vector,  $k_1 = 2\pi/L$ , where  $L$  is the box size, while the second peak has  $k_2 = 2\sqrt{2}\pi/L$  as one would expect for the two lowest frequency modes in an elastic solid.) Thus, the onset of anomalous modes lies above these peaks. In the infinite system-size limit, the peaks should smooth out to yield the normal scaling:  $D(\omega) \sim \omega^{d-1}$  in  $d$  dimensions.

We now recapitulate the theoretical ideas that address the properties of high-coordination systems [19]. In general, extra contacts increase the frequency of the lowest-frequency anomalous modes in two ways: (i) they can increase the energy cost of a mode by adding extra nodes so that some bonds are unstretched during an oscillation, and (ii) they can leave the number of nodes fixed but instead increase the average restoring force (and therefore the energy) for the normal-mode displacement.

A variational argument calculates an upper bound for the energy of a normal mode by minimizing the energy with respect to these two contributions. The first term in Eq. (3) indicates that a good trial function would have nodes, i.e., small values of  $(\delta\vec{R}_{ij} \cdot \hat{r}_{ij})^2$ , where  $V''$  is large. Thus the  $z_1 N/2$  contacts with the highest values of  $V''$  introduce nodes in the trial function. The remaining  $(z - z_1)N/2$  contacts increase the energy of the trial mode by increasing the restoring force for displacements according to the first term in Eq. (3) [19]. We rewrite Eq. (3) as

$$\delta E^{(0)} = \delta E_1 + \delta E_2, \quad \delta E = \delta E^{(0)} + \delta E_3, \quad (4)$$

where  $\delta E_1$ ,  $\delta E_2$ , and  $\delta E_3$  represent the energy costs asso-

ciated, respectively, with the  $z_1 N/2$  stiffest contacts, the remaining  $(z - z_1)N/2$  weak contacts, and the contribution of the stress term [the second term in Eq. (3); see Ref. [18]]. Note that  $\delta E^{(0)}$  is the energy cost of a mode for the unstressed system.

We now construct approximate expressions for these contributions. Reference [17] argues that  $\delta E_1 \approx A_1 k_1 (z_1 - z_c)^2$ , where  $k_1 = \langle V'' \rangle_1$  is the average over the  $z_1 N/2$  contacts with the highest values of  $V''$ . This is consistent with earlier simulation results for harmonic springs for  $z_1 - z_c \leq 3$  [18]. While  $k_1$  varies strongly with density and potential, we expect  $A_1$  to depend only weakly on these quantities [25]. To obtain  $A_1$ , we compare to simulations of unstressed systems with springs of equal stiffness. The precise value of  $A_1$  depends on which point in the density of states we choose to represent the onset of anomalous modes. In the following analysis, we use the value  $\omega^\dagger$  where  $D^{(0)}(\omega)$  reaches 0.25 of its maximum height, which fixes  $A_1 = 0.018$ . We would have obtained a somewhat different constant if we had compared with data at a different point in the rise, but the difference would be small because  $D^{(0)}(\omega)$  rises abruptly.

To estimate  $\delta E_2$ , we assume that for the normalized trial mode, the displacements of  $i$  and  $j$  are uncorrelated with each other if  $i$  and  $j$  are connected by weak contacts. Then  $\langle (\delta \vec{R}_{ij} \cdot \hat{r}_{ij})^2 \rangle \approx 2/Nd$  for an  $N$ -particle system in  $d$  dimensions [19]. Thus  $\delta E_2 \approx \frac{1}{Nd} \sum_{ij}^w V''(r_{ij})$ , where the sum  $\sum^w$  runs only over pairs of particles  $i$  and  $j$  connected by the  $(z - z_1)N/2$  weakest contacts. We have shown numerically that this approximation is reasonable for a system with particles connected by harmonic springs at their equilibrium lengths with two very different stiffnesses. For such a system, the stiff springs contribute only to  $\delta E_1$ , the weak springs contribute only to  $\delta E_2$ . Our expressions for  $\delta E_2$  and  $\delta E_1$  were thus verified cleanly.

We estimate  $\delta E_3$  as follows. For uncorrelated displacements between particles  $i$  and  $j$ ,  $\langle (\delta \vec{R}_{ij}^\perp)^2 \rangle \approx 2(d-1)/dN$ , leading to  $\delta E_3 = A_3 \mathcal{S}$ , where  $\mathcal{S} \equiv 1/N \sum_{ij} [V'(r_{ij})/r_{ij}]$ , and  $A_3 \approx (d-1)/d$  is of order unity.

Note that  $\delta E_3$  does not depend on  $z_1$ , so it does not affect the energy minimization with respect to  $z_1$  or  $z_1$  itself. In order to compute  $z_1$ , we thus compute the total energy cost in absence of stress:

$$\delta E^{(0)} = 0.018 k_1 (z_1 - z_c)^2 + \frac{1}{Nd} \sum_{ij}^w V''(r_{ij}), \quad (5)$$

where  $k_1 = \langle V'' \rangle_1$  is the average over the  $z_1 N/2$  contacts with the highest values of  $V''$ , and  $\sum_{ij}^w$  sums over the pairs of particles connected by the  $(z - z_1)N/2$  contacts with the smallest values of  $V''$ . By minimizing  $\delta E^{(0)}$  with respect to  $z_1$ , we obtain an upper bound on the energy and therefore the frequency,  $\omega^* = \sqrt{\delta E_{\min}^{(0)}}$ , of the lowest-frequency anomalous modes in the unstressed systems.

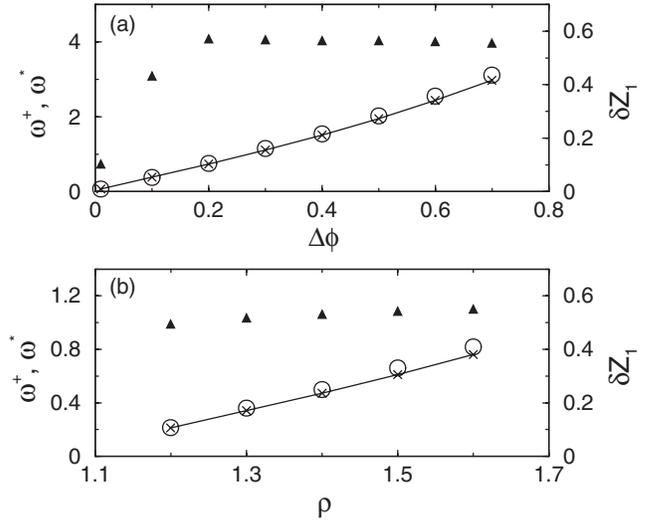


FIG. 2. Left axis: The characteristic frequency  $\omega^\dagger$  (open symbols) calculated from numerical simulations and the corresponding theoretical predictions  $\omega^*$  (crosses). Lines are to guide the eye and connect the theoretical points. Right axis: the fractional deviation from isostaticity  $\delta Z_1 \equiv (z_1 - z_c)/z_c$  (closed symbols), as functions of (a) distance from the unjamming transition,  $\Delta\phi = \phi - \phi_c$ , for repulsive Lennard-Jones mixtures, and (b) density  $\rho$  for Lennard-Jones mixtures.

In Fig. 2, we compare this theoretical prediction for  $\omega^*$  with the onset of anomalous modes,  $\omega^\dagger$ , from simulations in the unstressed system [determined by where  $D^{(0)}(\omega)$  reaches 0.25 of its maximum value]. The agreement is excellent everywhere, with no adjustable parameters. We have verified that the agreement is equally good (though with a different coefficient  $A_1$ ) if we define  $\omega^\dagger$  by a different point in the rise of  $D^{(0)}(\omega)$ , as long as we are consistent in our definition of  $\omega^\dagger$  for all potentials and densities.

Our results show that even though a typical repulsive amorphous solid may have a high coordination number and therefore appear to be far from the unjamming transition, most of the contacts are weak. The number of stiff contacts is only slightly in excess of the minimum needed for mechanical stability. This is shown in Fig. 2 (solid triangles) for both potentials at all densities studied. Even for the Lennard-Jones system, where the power-law tail of the interaction leads to a divergent total coordination, we find that  $(z_1 - z_c)/z_c < 0.6$  at all densities. Thus  $(z_1 - z_c)/z_c$  is a small parameter.

While the results of Fig. 2 are for unstressed systems, real glasses have nonzero stresses. To estimate the onset of anomalous modes for systems with stress, we must calculate the total energy cost of a mode,  $\delta E = \delta E^{(0)} + \delta E_3$ . Figure 3 shows that for our systems,  $\delta E^{(0)} \approx -\delta E_3$ , where  $\delta E^{(0)}$  is evaluated at its minimum with respect to  $z_1$  for both potentials at different densities. Thus, there is a near cancellation of two large terms leading to a small value of  $\delta E$  (with a large uncertainty) and therefore a very low

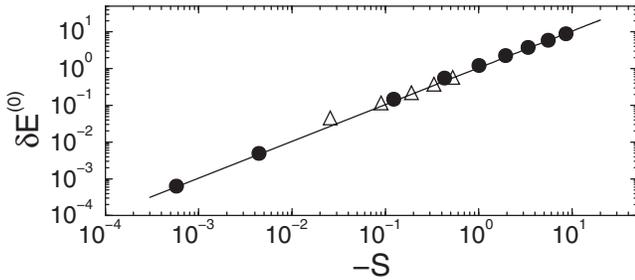


FIG. 3. The energy cost of the lowest-frequency anomalous mode for an unstressed system,  $\delta E^{(0)}$ , as a function of  $S \equiv 1/N \sum_{ij} V'(r_{ij})/r_{ij}$  for repulsive Lennard-Jones mixtures (solid circles) and Lennard-Jones mixtures (open triangles). The contribution of the stress term to the energy cost of a mode is  $\delta E_3 = A_3 S$ , where  $A_3$  is of order unity. The straight line fit corresponds to  $\delta E^{(0)} = -1.6S$ .

onset frequency for the anomalous modes in the stressed systems. This is consistent with the finding that the expected scaling of  $D(\omega) \sim \omega^2$  for plane-wave normal modes is eclipsed by an approximately linear frequency dependence at small  $\omega$  for both potentials. The near cancellation may be a result of the history of how the system was prepared [18].

Our results provide a plausible explanation for the origin of the excess vibrational modes of the boson peak in repulsive glasses. For two commonly studied models, we have shown that the boson peak derives from the same low-frequency anomalous modes that arise at point  $J$  for systems with finite-ranged repulsions. Several theories have been advanced previously for the boson peak [4–11]. Fractal systems [4] as well as disordered ones [7–9] can exhibit excess vibrational modes, but glasses are typically not fractal and low-coordination crystals can also display excess modes [15]. Approaches by Phillips [26], Thorpe [27], and Alexander [21] are also based on the idea that a minimum average coordination number is needed to prevent zero-frequency modes. However, those theories are limited to nonrigid covalent networks or systems interacting with attractive potentials, respectively. By contrast, Ref. [19] argues that the soft mode analysis can not only be applied to repulsive glasses, as we have done here, but also to low-coordination crystals and network glasses such as silica and crystalalite.

For our model repulsive glasses, we have shown that  $(z_1 - z_c)/z_c$  remains small even when  $z/z_c$  is arbitrarily large. It is for this reason that the marginal-coordination approach can be applied to these systems with high coordination. As long as  $(z_1 - z_c)/z_c$  is small, the behavior of the glass is governed by the physics of point  $J$  and isotaticity. Even though the unjamming transition itself may be inaccessible—as it is in the Lennard-Jones system studied here—the effect of the longer-ranged part of the potential can be treated as a correction. This theory can

therefore be viewed as a conceptual generalization of the perturbation theory of liquids to the case of jamming.

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