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Disorder and excess modes in hard-sphere colloidal systems

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Abstract – The anomalous thermodynamic properties of glasses remain incompletely understood, notably the anomalous peak in the heat capacity at low temperatures; it is believed to be due to an excess of low-frequency vibrational modes and a manifestation of the structural disorder in these systems. We study the thermodynamics and vibrational dynamics of colloidal glasses and (defected) crystals. The experimental determination of the vibrational density of states allows us to directly observe a strong enhancement of low-frequency modes. Using a novel method (ZARGAR R. *et al.*, *Phys. Rev. Lett.* **110** (2013) 258301) to determine the free energy, we also determine the entropy and the specific heat experimentally. It follows that the emergence of the excess modes and high values of the specific heat are directly related and are specific to the glass: even for solids containing a very large amount of defects, both the low-frequency density of states and the specific heat are significantly smaller than for the glass.

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The vibrational density of states (DOS) and the normal modes of solids provide a direct route to study its thermodynamics and mechanical properties [1]. In perfect crystalline solids, because of their long-range order, vibrational states are well understood as plane-wave phonon modes [2–5]. However, for more disordered systems the nature of vibrations remains elusive [6]. Structurally disordered systems such as liquids, glasses and amorphous materials exhibit a number of peculiar properties that are anomalous compared to those of the crystals [6–16]. These properties include anomalous acoustic behavior, a peak in the temperature dependence of the specific heat C_p/T^3 , and a Boson peak observed in inelastic scattering of light or neutrons [13–16]. These suggest the existence of an excess vibrational density of states over and above the predictions of the Debye model: at the maximum in C_p/T^3 , the vibrational density of states, $D(\omega)$, scaled with the DOS of a perfect crystal, goes through a maximum which is called the “Boson peak” [17–22]. The relation between the Boson peak and the disorder remains a topic that is hotly debated [12,13]; it seems clear that there is some relation between the two, but a direct link between the Boson peak and structural disorder in glassy systems has been difficult to demonstrate [17–25].

In this letter, we study the effects of structural disorder on the vibrational modes and the thermodynamics of colloidal hard spheres. This system, as we will show below, allows us to determine both the DOS and the thermodynamic properties of the glassy and crystalline states, and to provide a direct comparison between the two states of matter. We apply the covariance matrix analysis [7,26,27] to determine the density of states and the normal modes of vibrations from the particle displacements for a nearly perfect crystal, crystals with different amounts of defects, and for completely disordered systems (glasses). We find that there is a strong enhancement of low-frequency modes in the DOS for glasses however, no significant excess of modes is observed for very defected crystals, regardless of the amount of disorder (crystalline defects) present. We also experimentally determine the entropy, which is a measure of the specific heat at constant temperature, for several hard-sphere systems with different amounts of disorder. We show that while the specific heat increases gradually with increasing amount of disorder for crystals, it shows a discontinuous jump between a very defected crystal and a glass, as does the amplitude of the excess modes. These observations confirm independently that the excess of modes is an intrinsic property of glassy systems that is uniquely due to their vibrational

entropy and is not directly related to structural defects, as the fraction of particles with local sixfold symmetry in the very defected crystal is only slightly larger than that of the glassy system.

We use sterically stabilized fluorescent polymethylmethacrylate particles suspended in a refractive index and density matched mixture of *cis*-decalin and cycloheptyl bromide which are the best model system for hard spheres [28]. Our particles have a diameter $\sigma = 1.7 \mu\text{m}$ and polydispersity $\simeq 4\%$. The disordered system is obtained in a second system with $\sigma = 1.5 \mu\text{m}$ and polydispersity $\simeq 7\%$ to prevent crystallization. In both systems an organic salt, tetrabutylammonium bromide, is used to screen any possible residual charges. Colloidal hard-sphere systems at volume fractions between 0.54 and 0.62 are prepared by diluting sediments that are centrifuged to random close packing ($\phi_{rcp} \simeq 0.64$) [29]. Fast confocal microscopy allows us to determine the structure and dynamics of our particles that are subject to thermal agitation. For the DOS, we acquire sequences of 2D images on a 3D system; this allows us to follow the dynamics of the individual particles. Note that, following the vibrations of particles in a full three-dimensional system requires an extremely fast scanning microscopy which is not feasible with the current equipment even for small three-dimensional systems. To understand what is the effect of choosing a 2D slice of a 3D system, previously the density of states for a two-dimensional slice of a 3D system and for a three-dimensional system was studied by performing extensive simulations [7,8]; it was shown that the low-frequency part of the two spectra have a qualitatively similar behavior: the signature of the presence of the soft modes is present both in the full 3D system, and in its 2D cut.

Our entire crystal is polycrystalline, allowing to perform the measurements on perfect and defected crystals on the same sample but at different regions that are characterized by a different defect density. The 2D slices are taken in a field of view $105 \mu\text{m} \times 105 \mu\text{m}$ and at a distance of $25\text{--}30 \mu\text{m}$ away from the coverslip, deep enough to avoid the effects of the boundary. We note that hard spheres nucleate with their hexagonal plane, *i.e.* the $(1,1,1)$ -plane of the fcc crystal, perpendicular to the external boundary and then fill the cell maintaining the same orientation. The cover slide thus dictates the orientation of the crystal but, at sufficiently long distances, has no effect on its thermodynamic properties. In other words, the orientation of crystalline planes is due to its growth history, but at the measurement location (at $25\text{--}30 \mu\text{m}$ from the cover plate), the crystal behaves like a bulk crystal, *i.e.* its vibrational properties are unaffected by the presence of the wall. For amorphous phases however, the wall effects propagate at most over the dynamical correlation length, which is less than a few times of the particle diameter [30].

Acquiring about 5500 images with a rate of 25 frames per second, we follow positions of about 2600 particles in the 2D confocal plane. For determining the free energy,

we perform full 3D scans of the particle positions, that allows us to determine the free volumes of a given static configuration [31]. Figure 1 shows a 2D configuration of the particles for a nearly perfect crystal, crystals with three different amounts of disorder and for a supercooled liquid, all at the same volume fraction $\phi = 0.56$; crystals are more and more defected going from (a) to (d). The particles are colored according to their hexagonal order; to quantify the order in our systems, we calculate the bond orientational order parameter, $\Psi_6 = \frac{1}{N_{nn}} \sum_k \exp(6i\theta_{jk})$ in which N_{nn} is the number of nearest neighbors and θ_{jk} indicates the direction of particle j with respect to its nearest neighbors k [32]. Figure 1(f) demonstrates the probability density distribution of Ψ_6 . We find that for the nearly perfect crystal and the two more imperfect crystals, corresponding, respectively, to fig. 1(a)–(c), the distribution shows a high peak at $\Psi_6 = 1$: all the particles are sixfold coordinated. For the glassy sample, the histogram is broad with $\Psi_6 < 1$ (fig. 1(f)). The very defected crystal however, represents both features: a small peak at $\Psi_6 = 1$ and a rather broad distribution for $\Psi_6 < 1$ (fig. 1(d) and (f)).

Following the motion of particles in real time, we obtain all particle positions $x = x(t)$, $y = y(t)$ as functions of time using standard particle tracking software [33]. Denoting $u_i(t)$ the components of the particle displacements from the average positions along the confocal plane $u_i(t) = \{(x_i(t) - \langle x_i \rangle), (y_i(t) - \langle y_i \rangle)\}$, we obtain the displacement correlation matrix (of dimension twice the number of particles) as

$$D_{ab} = \langle u_{\mu i} u_{\nu j} \rangle, \quad \mu, \nu = x, y, \quad (1)$$

where $a, b = 1, 2, \dots, 2N$ matrix index on the left runs both over the particle indices and the Cartesian components of displacements. The averaging $\langle \dots \rangle$ has been done over the period of measurement, which is about 220 seconds.

Diagonalizing D_{ab} we obtain the eigenvalues, λ_a , and the corresponding $2N$ normal modes of the system. Results are presented in terms of the mode frequencies which are related to the eigenvalues as

$$\omega_a = \sqrt{1/\lambda_a}. \quad (2)$$

We note that the covariance matrix analysis is extensively used to study the vibrational density of states for different systems [4,7–9,27,34]. This method allows us to gain information on the nature of low-frequency modes, but completely ignores dissipative effects and anharmonicity that are present in the system. On the other hand, for the dynamical matrix method while both dissipative effects and anharmonicity are taken into account, all spatial information is lost, which is not the case for our method. Connecting the two methods is not possible here: the eigenvalues of the covariance matrix λ_a with the dimension of length squared, are related to those of the dynamical matrix ω_a with the dimension of inverse time,

Fig. 1: (Color online) Configuration of the particles in a 2D plane. The average position of the particles are shown for (a) a nearly perfect crystal, ((b), (c)) two more imperfect crystals, (d) for a crystal with a large amount of disorder, a “very defected crystal” and (e) for a supercooled liquid, all at the same volume fraction $\phi = 0.56$. The amount of disorder increases from (a) to (e). Particles are colored according to the hexagonal order. A Voronoi tessellation is shown for each configuration. (f) The probability density distribution of the bond orientational parameters for a nearly perfect crystal, crystals with defect, and for liquids.

as $\lambda_a = (k_B T)/(m\omega_a^2)$ with $k_B T$ the thermal energy, only for systems with non-dissipative dynamics at equilibrium which is not valid for our dense hard spheres. Therefore, the combined effects of damping, anharmonicity and hydrodynamic interactions can alter the frequency scale of the covariance matrix analysis from the real frequency of the damped anharmonic system. Thus, ω_a are the temporal frequencies the system would have if it were harmonic and undamped.

The resulting density of states, $D(\omega)$, is shown in fig. 2; the DOS is plotted *vs.* the frequency for a nearly perfect crystal, crystals with three different amounts of disorder and for a supercooled liquid all at the same volume fraction $\phi = 0.56$. Since “hard” modes are expected to have eigenvalues proportional to the pressure, we scale out this effect by plotting the density of states in terms of the scaled frequency ω/p [7,8,26,35]; to do so, we use the Hall [36] equation of state for the crystal and the Liu [37] equation of state for the supercooled liquid and glassy phases. Scaling the frequency with the pressure, allows us to compare the DOS for systems with different amounts of disorder and different volume fractions, since trivial effects due to the density change are already scaled out. The density of states is normalized such that $\int_0^\infty D(\omega) d\omega = 1$. It has been established [28] that for high frequencies the experimental noise becomes important due to the lack of accuracy in determining individual particle positions. Dashed lines show the limits below which the frequencies should remain unaffected by noise (fig. 2).

Fig. 2: (Color online) The density of states for a nearly perfect crystal, crystals with three different amounts of disorder, and for a completely disordered system all at the same volume fraction $\phi = 0.56$. Dashed and dotted lines show the upper limit of the frequency until which the data is not affected by noise for, respectively, crystals and liquid. The DOS of the nearly perfect crystal is subtracted from all the densities of states (inset).

Figure 2 shows that for a given low frequency, the observed density of states for defected crystals is larger than the DOS for a perfect crystal; the difference increases with increasing disorder. For the supercooled liquid however, the difference is much larger than any of the crystal samples (fig. 2). We also find that the DOS shows a shift towards lower frequencies with increasing disorder. The

most striking observation is however the large difference between all crystalline samples and the supercooled liquid, which shows a large peak at low frequencies. We note that, we compare the DOS for (defected) crystals and completely disordered system at the same volume fraction $\phi = 0.56$, for which the pressure is $p_{Hall} = 12.21\rho k_B T$ for the crystals and $p_{Liu} = 22.97\rho k_B T$ for the supercooled liquid; ρ is the number density. Scaling the frequency with p changes the height of the supercooled DOS with respect to the crystal DOS by a factor of $22.97/12.21 = 1.88$. However, this factor is considerably smaller than the observed difference between the crystal and the disordered system (fig. 2). We note also that the shape of the DOS spectrum for the (defected) crystals and the disordered system are qualitatively similar around the peaks, but as we show later, what is important is the large difference in the intensity.

Measuring the density of states for a nearly perfect crystal, we identify a peak and a shoulder. However, ideally, there are two peaks in the vibrational spectrum for a perfect crystal that are vestiges of van Hove singularities. In our experiments the van Hove singularities may be rounded by several factors, *e.g.* size polydispersity of particles, uncertainties in finding particle positions and the limitation due to the finite number of frames; on the other hand, at low frequencies the density of states is hardly affected by these [27]. We investigate the effects of the finite number of statistics on the vibrational density of modes for a nearly perfect crystal. We find that the low-frequency part of the DOS spectrum remains unchanged with increasing the number of statistics by almost one order of magnitude (see [38] for a more detailed discussion).

To investigate the effects of disorder more consistently, we subtract the DOS of the nearly perfect crystal from all the densities of states (fig. 2 inset). The very existence of the peak in the density of states for supercooled liquid with respect to that of a perfect crystal implies a strong excess of low-frequency modes. We find that the excess of low-frequency modes or equivalently, the height of the peak is very small for defected crystals compared to completely disordered systems (fig. 2 inset), implying that the excess of modes is an intrinsic property of glasses and does not arise due to structural disorder in crystals.

Plotting $D(\omega)/\omega$ and $D(\omega)/\omega^2$ is a usual way to identify the excess modes for respectively a 2D and 3D system. However, it was shown previously that the density of states for a 2D slice through a 3D system, that we measure, differs from that for a 2D system [4]: for a 2D slice of a 3D system, it is shown that the Debye behavior is $D(\omega) \sim \omega^3$. To measure the excess of modes with respect to the corresponding crystal, we subtract the DOS of the nearly perfect crystal from all the densities of states (see [39] for a similar analysis); in our system we can be assured that this is (nearly) defect free, as we directly visualize all the defects.

To check whether the DOS for a very defected crystal represents more crystalline features or is more liquidlike,

Fig. 3: (Color online) Density of states for a nearly perfect crystal, two more imperfect crystals and several crystals with a large amount of disorder.

we measure the density of states for several crystals with a large amount of defect, *i.e.* with the wide distribution of the Ψ_6 , lies between crystal and liquid, (*e.g.* fig. 1(d) and (f)); we find that for all very defected crystals, the observed DOS stays much closer to that of the perfect crystal and far below the DOS for the liquids (see fig. 3), implying that there is a discontinuous jump between the vibrational density of states for a very defected crystal and that of a supercooled liquid or a glass. This happens in spite of the observation that the fraction of particles with local sixfold symmetry in the very defected crystals is only slightly larger than in a glassy system.

We note that for non-overdamped systems the density of states obtained from different regions of the same sample do not differ significantly from each other, since the DOS reports the vibrational properties of the whole sample. However, since our colloidal system is an overdamped system, the vibrational state inferred from each image is basically independent from each other.

We proceed by investigating the disorder dependence of the excess of modes in more detail (fig. 4). For each system we quantify the disorder exploiting two quantities: 1) the bond order parameter and 2) the number of nearest neighbors; a particle is considered as a defect if the former is less than 0.95 or the latter differs from six, the expected value for a 2D crystalline lattice. Disorder is then defined as the fraction of defected particles relative to the total number of particles, *i.e.* it is zero for a perfect crystal and one for a completely disordered system. However, for our experimental system, the disorder is 0.14 for the nearly perfect crystal, 0.35, 0.55 for the two more imperfect crystals, 0.87 for the very defected crystal and 0.99 for the completely disordered system. We find that, although the amounts of disorder in a supercooled liquid is only slightly larger than that in very defected crystals, the amplitude of the excess modes for the former is significantly larger, while it is negligible for the latter (fig. 4). This unambiguously shows, for the first time, that the excess modes is not directly related to the disorder, but rather to the nature of the glassy state.

Fig. 4: (Color online) Amplitude of the excess modes (Y-axis at the left) is plotted *vs.* the scaled amount of disorder for a nearly perfect crystal, crystals with three different amounts of disorder and for a supercooled liquid all at the same volume fraction $\phi = 0.56$ (squares). The entropy per particle, which is indeed a measure of the specific heat for a system of hard spheres at constant temperature, in units of k_B (Y-axis at right) is plotted *vs.* the scaled amount of disorder for a nearly perfect crystal, crystals with four different amounts of disorder and for two supercooled liquids all at the same volume fraction $\phi = 0.56$ (circles). The amount of disorder is scaled with respect to that in the nearly perfect crystal. Dotted line shows the difference between the entropy of a crystal and a liquid calculated in [1]. Dashed curve is plotted as a guide for eye.

A unique feature of hard-sphere colloids is that due to the absence of interactions, the free energy can be obtained directly from the configuration of the particles [31,40]. To study the effects of disorder on the thermodynamics, we then measure the entropy for several 3D subsystems with different amounts of defects [31]; each subsystem that we measure contains around 2500 particles. We calculate the free energy F , from which the entropy follows directly as $-F/T$. We determine the free volume for each individual particle; once the free volume is known, the free energy can be obtained directly from the cell model [31]. Note that, for a system of hard spheres at constant temperature T , any changes in the entropy indicates a change in the specific heat according to: $S(T) = \int C_p(T) \frac{dT}{T}$. Results are shown in fig. 4 where the entropy is plotted *vs.* the disorder. Interestingly, we find that, similarly to the amplitude of the excess modes, the specific heat for crystals increases gradually with increasing disorder and shows a significant jump between a very defected crystal and a supercooled liquid (fig. 4). We observe again that while the difference between the amount of disorder for the very defected crystal and the supercooled liquid is very small, the difference between their measured entropy is very large, implying that the thermodynamic first-order phase transition between crystal and liquid is not much affected by the disorder, *i.e.* the transition between crystal and liquid remains as a first order, even if the crystal is strongly disordered. This observation confirms also that the excess of modes is a property of liquid-like phase that is uniquely due to its vibrational entropy and is not related

Fig. 5: (Color online) Spatial distributions of the normal modes at $\omega/p = 0.37$ for (a) a nearly perfect crystal, (b) a more imperfect crystal, (c) a very defected crystal and (d) a supercooled liquid all at the same volume fraction $\phi = 0.56$.

to structural disorder in crystals, in agreement with a common assertion that the excess of modes are a dynamic phenomenon.

We now explore the effects of disorder on the normal modes of the crystalline systems. Figure 5(a)–(d) shows the spatial distribution of the normal modes at a low frequency for, respectively, a nearly perfect crystal, a more imperfect crystal, a very defected crystal, and a supercooled liquid corresponding, respectively, to figs. 1(a), (b), (c) and (d). We find that the lowest frequency modes in the crystals exhibit mostly plane-wave-like features that extend over very large length scales. This becomes less and less evident with increasing the amount of disorder (fig. 5(a)–(c)). For the completely disordered system, the very lowest frequency modes show spatially correlated motions only over a few particle diameters (fig. 5(d)). See [38] for further discussions.

For a perfect crystal the spatial distribution of the normal modes would show plane waves that extend over the size of the system. The spatial distribution of the normal modes for our nearly perfect crystal shows wave-like features rather than very extended plane waves. The observation of wave-like features for experimental (defected) crystal is in fact quite common in these systems [3,4]. We note that, one may also look at these patterns as being non-affine patterns as discussed in [41].

In summary, we present the first experimental evidence that the excess of modes is an intrinsic property of glasses and does not arise due to structural disorder in crystals. This conclusion takes advantage of the opportunities offered by hard spheres, in which the crystal and glass

phases can be compared at the same volume fraction, and for which a simultaneous measure of disorder and free energy is possible with experiments. We show that this happens in spite of the observation that the fraction of particles with local sixfold symmetry in the very defected crystals is only slightly larger than in a glassy system. We show that the vibrational and thermodynamical properties, *i.e.* the DOS and the entropy, for a defected crystal are significantly different from those for a supercooled liquid or a glass, implying the different nature of the glass compared to a defected crystal.

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