Normal modes analysis of the microscopic dynamics in hard discs

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We estimate numerically the normal modes of the free energy in a glass of hard discs. We observe that, near the glass transition or after a rapid quench deep in the glass phase, the density of states (i) is characteristic of a marginally stable structure, in particular it displays a frequency scale $\omega^* \sim p^{1/2}$, where $p$ is the pressure and (ii) gives a faithful representation of the short-time dynamics. This brings further evidences that the boson peak near the glass transition corresponds to the relaxation of marginal modes of a weakly-coordinated structure, and implies that the mean square displacement in the glass phase is anomalously large and goes as $\langle \delta R^2 \rangle \sim p^{-3/2}$, a prediction that we check numerically.

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There is still no accepted description of why the dynamics of liquids rapidly slows down as the temperature is lowered $^1$. Goldstein $^2$ proposed that the glass transition is related to a change in the topology of the free energy landscape. In this view, below some temperature meta-stable states appear, the dynamics becomes activated and rapidly slows down. A similar scenario occurs in mean field glass models $^3, 4, 5$, which share formal similarity with the Mode Coupling Theory of liquids $^6$. The presence of an elastic instability, corresponding to the onset of activation, is also predicted in systems were particles are deposited randomly (corresponding to an infinite temperature), as the density is changed $^7$. Nevertheless, the real-space interpretation of those views is not clear. Empirically, the presence of an elastic instability near the glass transition is suggested by Raman and neutron spectra, which display a peak, the “Boson Peak”, typically shifting toward zero-frequency as the glass is heated toward the glass transition $^8$. This observation is also supported by numerical simulations $^9$. The nature of the modes forming the peak and how they depend on the structure of the glass are long-standing questions $^10$, on which recent progress was made in a-thermal assemblies of particles $^11, 12$. Elucidating these questions at finite temperature, in particular in the vicinity of the glass transition, is necessary to make progress in our microscopic understanding of the glass transition. Indeed these modes, which characterize the short-term dynamics, presumably affect how glasses flow on much longer time-scales, as supported by the observations that the short term dynamics (i) correlate with the fragility of the glass $^13$ and (ii) is a good local predictor of the propensity (the tendency of a region to flow) $^14$. To study these modes, various numerical technics can be used, such as the instantaneous normal modes analysis $^15$ or the analysis of the vibrational spectrum of inherent structures $^16$. Nevertheless, these approaches consider the energy rather than the free energy landscape. Such analysis are not adapted for colloidal systems or shaken grains $^17, 18$ or when non-linearities of the interacting potential are important, which is presumably often the case near the glass transition $^19$.

In this Letter, we study the microscopic dynamics of hard discs in the vicinity of the glass transition and deep in glass phase, via an analysis of the free energy landscape. To achieve it, we use a recent technic developed to infer the normal modes of the free energy $^20$. We show that, (i) after a rapid quench in the glass phase or near the glass transition, the density of normal modes is characteristic of a marginally rigid solid, in agreement with a former analysis of the microscopic structure of the glass $^20$, (ii) throughout the spectrum, the computed frequency of a mode gives the correct estimate for the characteristic time and amplitude of its fluctuations. This demonstrates that our mode analysis performs well in the vicinity of the glass transition. (iii) These results justify the presence of a boson peak and an anomalously large and slow microscopic relaxation. In particular, the mean square displacement must vary as $\langle \delta R^2 \rangle \sim p^{-3/2}$, in good agreement with our simulations. This signifies that, deep in the glass phase, the amplitude of the particles trajectories become infinitely larger than the instantaneous size of the cages.

As the viscosity of a super-cooled liquid increases, the dynamics become intermittent $^21, 22, 23$: there are long quiet periods where particles are rattling around their average positions, interrupted by rapid rearrangements toward new configurations. In a simulation, this intermittent dynamics can be followed via the self density correlation function $C(\vec{q}, t, t_0) = \langle \delta q \cdot \delta R_j(t + t_0) - \delta R_j(t) \rangle$, where the average is made on all particles, but not on time, and where $\delta R_j(t)$ indicates the position of particle $j$ at time $t$. This observable displays long plateaus, interrupted by sudden jumps leading to new plateaus $^20, 21$. We shall refer to these plateaus as meta-stable states. In the reference $^21$, the transition between states was studied. Here we focus on their inner dynamics.

As shown in $^21$, in meta-stable states the Gibbs free
energy of a hard sphere system can be approximated as:
\[ \mathcal{G} = -k_B T \sum_{ij} \ln(h_{ij}), \]
where the sum is made on all pairs of particles \( ij \) in contact, i.e. colliding with each other in a specific meta-stable state. \( T \) is the temperature, \( \langle h_{ij} \rangle \equiv \langle \|\vec{R}_i - \vec{R}_j\| \rangle - (\sigma_i + \sigma_j)/2 \) is the average gap between particles measured in a given meta-stable state, and \( \sigma_i \) is the diameter of particle \( i \). Eq. (11) draws an analogy between the free energy of hard spheres and the energy of a system of logarithmic springs. Eq. (11) can be expanded for small displacements around the average particles position \( \delta \vec{R}_i = \vec{R}_i - \langle \vec{R}_i \rangle \). This enables to define the dynamical matrix \( \mathcal{M} \) [24], which describes how the average particles positions respond to a small external force. The eigenvectors \( \delta \vec{R}^n \) of \( \mathcal{M} \) are the vibrational modes [22]. For particles of unit mass, we define as usual their frequency \( \omega \) as the square root of the corresponding eigenvalue of \( \mathcal{M} \). In what follows \( \omega \) is used to label the normal modes. We shall see below that this label corresponds, within a good approximation, to the physical oscillation frequency of the motion.

In practice, we use an event-driven code to simulate a two-dimensional bidisperse hard discs system [20, 21] and compute \( \mathcal{M} \). Half of the particles have a unit diameter, while the others a diameter of 1.4 and their mass is \( m \equiv 1 \). We choose \( k_B T \) as our unit of energy. We can equilibrate this system up to \( \phi < \phi_0 \approx 0.79 \). To generate larger packing fractions and enter in the aging regime, we proceed as follows: we decompress jammed configurations of [20] of packing fraction \( \phi_c \approx 0.83 \) by decreasing the particle diameters by some fraction, and assign a random velocity to every particle. As the dynamics run, we compute the self-density correlation function and determine its plateaus or meta-stable states. For each of them, we consider a time interval \([t, t + t_1]\) corresponding to a single state, where \( t_1 \) is chosen to be much larger than the average collisional time of a contact \( \tau_c \). In what follows \( t_1 \) corresponds typically to 100 \( \tau_c \) but our results are robust to other choices as long as \( t_1 \gg \tau_c \). The network of contacts and the average particle positions \( \langle \vec{R}_i \rangle \) are computed during this interval. This enables to compute \( \mathcal{G} \) and \( \mathcal{M} \) numerically.

To compute the normal modes and the density of states, we diagonalize \( \mathcal{M} \). Results are shown in Fig. 1. Curves are labeled by the average contact force \( \langle f \rangle \equiv \langle 1/(h_{ij}) \rangle_{ij} \), where the average is made on all contacts. \( \langle f \rangle \) diverges near \( \phi_c \) and scales as the pressure. The densest system we can equilibrate corresponds to \( \langle f \rangle \approx 18 \). For larger \( \langle f \rangle \) the system is a glass. To compare various packing fractions, the frequency axis is rescaled by the square root of the typical interaction stiffness \( k \), of order \( k^{1/2} \sim \langle f \rangle \) in our units. We observe that: (i) for all \( \langle f \rangle \), \( D(\omega) \) increases rapidly from zero-frequency to reach a maximum at some frequency \( \omega^* \), before decaying again. In our finite size system the modes constituting the low-frequency part of the spectrum are not plane-wave like [24]. This can be inferred from the inset of Fig. 1, where \( D(\omega) \) is normalized by its Debye value (\( \sim \omega \) in two dimensions): no plateau can be detected at low frequency, where a peak appears at some frequency \( \omega_{bp} \) significantly smaller than \( \omega^* \). (ii) \( \omega^* \) decreases toward zero in the rescaled units as \( \langle f \rangle \) diverges. Fig. 2 shows the dependence of \( \omega^* \) with \( \langle f \rangle \). We observe that the scaling
\[ \omega^* \sim \langle f \rangle^{1/2} \] holds well from the glass transition toward our densest packing. Scaling behaviors in the vibrational spectrum, together with the fact that the lowest-frequency vibrations observed are not plane wave like, were first observed in elastic non-thermal particles [20]. This was later explained as follows: in a weakly-coordinated network of springs at rest, i.e. with a small coordination number \( z \) close to a critical value \( z = 2d \) (\( d \) is the spatial dimension), extended modes, quite different from plane waves, appear already in the low-frequency part of the spectrum. The frequency scale of these anomalous modes is \( \omega^* \sim k^{1/2}(z - z_c) \) [11]. In a system of repulsive particles with non-zero forces, these modes are shifted toward a lower frequency [27]. To avoid an elastic instability, the system must display enough contacts. In the marginally stable case where there are just enough contacts to be rigid, the spectrum displays anomalous modes up to zero frequency, but present a characteristic frequency at \( \omega^* \sim \langle f \rangle^{1/2} \), such that \( D(\omega^*) \) is of order of its typical value \( k^{-1/2} \). This estimate of \( D(\omega^*) \) agrees with Fig. 1, where \( D(\omega^*) \) appears to remains of the same order in rescaled frequency. Thus both the scaling of \( \omega^* \) and the observation that anomalous modes are present at frequency \( \omega_{bp} \ll \omega^* \) support that the glass phase lies close to marginal stability, as was already suggested by a direct measure of the coordination in the glass phase [20].

We check that \( D(\omega) \) reflects well the dynamics. We project the dynamics on the normal modes and define

FIG. 1: Densities of states \( D'(\omega') \equiv \langle f \rangle D(\omega) \) vs. rescaled frequency \( \omega' = \omega/\langle f \rangle \) for different values of \( \langle f \rangle \) in a system of \( N = 256 \) particles. We used \( t_1 = 5 \times 10^4 \). Inset: \( D(\omega')/\omega' \) vs. \( \omega' \) for \( \langle f \rangle = 18 \).
apparent in the inset of Fig. (1) as the Boson Peak, which
ful distribution of relaxation time scales of the micro-
discontinuous. The non-linearities do not appear to dominate the
mode dynamics in a dense disordered assembly of col-
trum, the non-linearities do not appear to dominate the

where $\langle R(t + t_w) | \delta R^w \rangle \cdot \langle \delta R(t_w) | \delta R^w \rangle \rangle_{t_w}$,

where $| \delta R(t + t_w) \rangle$ is the displacement field from the aver-
age configuration, $\langle \delta R^w | \delta R^w \rangle \equiv \sum_i \delta R^w_i \cdot \delta R^w_i$, and where the average is made on all time segments $[t_w, t_w + t]$ in-
cluded in a meta-stable state. If the modes were longitudi-
dinal plane-waves, $C_\omega(t)$ would describe the relaxation of
density fluctuations. Here this is not at all the case. W e
damped oscillations, whose amplitude and period follow:

$C_\omega(t) = ( \langle R(t + t_w) | \delta R^w \rangle \cdot \langle \delta R(t_w) | \delta R^w \rangle \rangle_{t_w}$, \hspace{1cm} (3)

We observe that the modes display weakly damped oscillations, whose amplitude and period follow:

$\langle A^2(\omega) \rangle \sim \frac{1}{\omega^2}$, \hspace{1cm} (4)

$\tau(\omega) \sim \frac{1}{\omega}$. \hspace{1cm} (5)

These results hold true even for the low-frequency part of the spectrum, although more scattering is found there
29. Thus, even for the lowest-frequency part of the spec-
trum, the non-linearities do not appear to dominate the
mode dynamics in a dense disordered assembly of col-
liding hard spheres, although the interacting potential is discontinuous.

This observation implies that $D(\omega)$ gives a rather faith-
ful distribution of relaxation time scales of the micro-
scopic dynamics. This allows us to identify the peak

appears as a similar hump in Raman or neutron spectra
in molecular liquids 8,10. Near the glass transition, this
peak appears at a frequency significantly smaller than
$\omega^*$. The scaling behavior of $D(\omega)$ supports that these
modes correspond the anomalous modes characterizing
weakly-coordinated structures, whose geometry is stud-
ied in 27. It would be interesting to go further and test if
the early-$\beta$ relaxation 6, which characterizes the relax-
ation on time scales longer than the boson peak, is due to
the weakly-damped relaxation of anomalous modes even softer than the peak, or to other non-linear processes not
captured by the present analysis. Unfortunately this relax-
ation is difficult to study numerically using Newtonian
dynamics, and our present analysis should be performed
in three-dimensions, where numerical and experimental
data exist, to test this possibility.

In what follows we rather focus on the amplitude of
the short-time relaxation, in particular the particle mean-

the fluctuations of a crystal: the modes is assumed. Eq.(8) must be compared with stability of the structure and the harmonic behavior of systems of different modes is independent, one gets:

$$\langle \delta \hat{R}^2 \rangle \sim 1/N \sum_i \langle \delta \hat{R}^2 \rangle_i$$

We then average on all particles and define $$\langle \delta \hat{R}^2 \rangle = 1/N \sum_i \langle \delta \hat{R}^2 \rangle_i$$ where $$N$$ is the system size. Using the modes normalization $$\langle \delta \hat{R}(\omega)^2 \rangle_i = 1/N$$ and applying Eqs.(5) lead to:

$$\langle \delta \hat{R}^2 \rangle \sim \int \frac{D(\omega)}{\omega^2} d\omega \geq \int \frac{D(\omega)}{\omega^2} d\omega$$

Since $$D(\omega)$$ reaches the order of its typical value $$1/\sqrt{\xi} \sim 1/\langle f \rangle$$ for $$\omega \geq \omega^*$$, the last integral is dominated by the lowest bound and one gets:

$$\langle \delta \hat{R}^2 \rangle \geq \frac{D(\omega^*)}{\omega^*} \sim \langle f \rangle^{-3/2}$$

It is easy to show from Eq.7 that the scaling [8] becomes an equality if $$D(\omega)$$ does not increase more rapidly than linearly between 0 and $$\omega^*$$. The bound [8] holds in any dimension $$d \geq 2$$, as long as the marginal stability of the structure and the harmonic behavior of the modes is assumed. Eq.[8] must be compared with the fluctuations of a crystal: $$\langle \delta \hat{R}^2 \rangle \sim h^2 \sim \langle f \rangle^{-2}$$ (with log $$N$$ corrections in two dimensions). In other words, near maximum packing, the amplitude of particles motions becomes infinitely smaller in the crystal, where it is simply of the order of the inter-particle gap $$h$$. In the hard sphere glass, particles diffuse much more than the size of their instantaneous cage $$h$$, and the size of the region they visit in a meta-stable state is at least of order $$h^{3/4}$$.

To check this prediction, we consider various metastable states, and measure the mean square displacement $$\langle \delta \hat{R}^2 \rangle$$ by averaging on some long time $$t_1$$, see Fig.4. Our numerical result agrees well with [8]. As for Eq.(2), the scaling prediction captures the behavior also near the glass transition, where the system can be equilibrated.

We have used and tested a free energy-based approach that allows us to perform a mode analysis at non zero-temperature, in particular in the vicinity of the glass transition. We have observed that, near the glass transition or after a rapid quench, the spectrum of the free energy is characteristic of a marginally rigid solid, as confirmed further by the scaling of the amplitude of the mean square displacement in the glass phase. The observation that on the time scales we can probe the system remains near the onset of rigidity supports the proposition, expressed originally by Goldstein in terms of free energy landscape, that the dynamics dramatically slows down once meta-stable states appear. Our analysis provides a spatial interpretation of this landscape picture, where the elastic instability is governed at a microscopic level by coordination and contact forces, and where the soft modes at play are collective rearrangements, whose characteristic length scale is limited near the glass transition and diverges near the maximum packing of hard spheres [11]. Finally, the observation that the microscopic dynamics, and activation events [21], are dominated by soft modes is consistent with the observation that the short term dynamics is a good indicator of propensity [14]. It suggests that correlations between coordination, contact force, and propensity may exist; and that soft modes may be the elementary objects to consider to describe microscopically activation.

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[28] For examples of lowest-frequency modes, see [21].
[29] Sometimes one or a few unstable modes are observed at very low frequency, indicating a shoulder or a saddle in the free energy landscape. In this case the values of \langle A^2 \rangle and \tau are found to be of the order of those of the lowest-frequency stable modes.
[30] In two dimensions, this analysis is only possible in a finite size system, since plane waves lead to a log N divergence of the mean square displacement.