Rigidity of thermalized soft repulsive spheres around the jamming point

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We study the effect of thermalization on the rigidity of a randomly packed soft repulsive sphere system around the jamming point by analyzing the shear-modulus using the cloned liquid theory with the 1 step replica symmetry breaking ansatz and molecular dynamics simulations. Contrarily to the usual harmonic picture for solids, we found that the thermalized jamming system is anomalously softer than at zero temperature such that the shear-modulus becomes as small as the pressure down to vanishingly low temperatures.

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Randomly packed repulsive foams, colloids, emulsions and granular particles exhibit anomalous solid states. At high densities liquid like spontaneous structural re-arrangements of the positions of the particles become almost impossible much as deeply supercooled molecular glass forming liquids. However the contacts between the particles exhibit surprisingly rich dynamic characters around the jamming point located deep in the glassy phase as manifested in static properties [1–10], linear and non-linear rheology [11–13] and relaxations [14–15].

An important common ingredient in this class of systems is that the particles are interacting with each other through repulsive contact potentials with some definite cut-off at particle scales. Quite interestingly recent works [16–18] suggest that the solid states of such systems are only marginally stable against unharmonic, plastic deformations such that opening of a weak contact can trigger an avalanche like deformation. This is contrarily to the usual view that harmonic descriptions work well in solids because defects would require finite creation energies so that they become suppressed at low enough temperatures.

On the other hand, physically we would still expect that these glassy systems, even if they are not strictly harmonic, retain some sort of macroscopic solidity at least at low enough temperatures. A natural quantity to quantify the solidity is the shear-modulus or the rigidity \( \mu \), which is finite only in solids. Indeed a set of experiments on an emulsion system [1, 3] done at the room temperature which amounts to a rescaled temperature that these glassy systems, even if they are not strictly harmonic, retain some sort of macroscopic solidity at least at low enough temperatures. A natural quantity to quantify the solidity is the shear-modulus or the rigidity \( \mu \), which is finite only in solids. Indeed a set of experiments on an emulsion system [1, 3] done at the room temperature which amounts to a rescaled temperature that these glassy systems, even if they are not strictly harmonic, retain some sort of macroscopic solidity at least at low enough temperatures.

The purpose of this paper is to study a model system which mimic the emulsion system [19] and examine the rigidity at low but finite temperatures by theoretical and numerical approaches. For the theoretical approach we employ the cloned liquid method [20, 22] which is a 1st principle method based on the liquid theory and the replica method aimed to analyze static properties of glasses and jamming systems within the scenario of the random first order transition (RFOT) theory (see [23] for a review). To compute the rigidity in this framework we follow the scheme of [24, 25] and extend it to the jamming system. We also performed simulations of the shear-stress relaxation to obtain the thermalized rigidity. Our main results is that the thermalized rigidity behaves just as the pressure much as in the experiment but in contradiction to the harmonic picture. We argue that our result suggests some gap-less spectrum of plastic excitations between different inherent structures (energy minima) within a meta-basin (metastable state) [26].

**Model** We study a system of \( N \) particles \( (i = 1, 2, \ldots, N) \) of diameter \( a = 1 \) in the 3-dimensional space with volume \( V \), which interact with each other through a soft repulsive contact potential given below. The crucial parameter is the volume fraction \( \phi = (\pi/6)a^3\rho \) which is related to the number density \( \rho = N/V \). We denote the positions of the particles as \( \mathbf{r}_i \) and represents a pair of particles \( i \) and \( j \) as \( (ij) \). The potential energy is given by 

\[
U = \sum_{ij} \phi_r(\mathbf{r}_{ij}) \quad \text{where} \quad \phi_r = |\mathbf{r}_i - \mathbf{r}_j|.
\]

We consider a soft repulsive contact potential of the form, 

\[
\phi(r) = \epsilon(1 - r/a)^2\theta(1 - r/a), \quad \text{where} \quad \theta(r) \text{ is the step function.}
\]

This is considered as an approximate model potential for emulsions [10, 18]. We denote a rescaled temperature \( k_BT/\epsilon \), where \( k_B \) is the Boltzmann’s constant, simply as \( T \) below.

**Cloned liquid approach** In the cloned liquid approach we consider a system of \( m \) replicas, each of which is a system of \( N \) particles. In order to study the glassy phase we employ the 1 step replica symmetry breaking (RSB) ansatz which is known to capture the phenomenology of glasses [23]. The 1-RSB ansatz amounts to consider a sort of ‘molecular’ liquid [20]. The coordinates of the particles \( \mathbf{r}_i^a \ (a = 1, \ldots, m; \ i = 1, 2, \ldots, N) \) is decomposed as, 

\[
\mathbf{r}_i^a = \mathbf{R}_i + \mathbf{u}_i^a \quad \text{with} \quad \mathbf{R}_i = \frac{1}{m} \sum_{a=1}^{m} \mathbf{r}_i^a
\]

where \( \mathbf{u}_i^a \) stands for fluctuation of the particle belonging to the \( a \)-th replica with respect to the center of mass \( \mathbf{R}_i \) of a ‘molecule’. Physically the size of the molecule which we denote as \( A \) represents the size of cage in the metastable states. The fluctuations within the molecules (cages) are
assumed to obey the Gaussian statistics with the mean 0 and variance \( \langle (u_i^a)^2 (u_i^b)^2 \rangle_{\text{cage}} = A (\delta_{ab} - \frac{1}{m}) \delta_{ij} \). Here \( \mu \) (and \( \nu \)) represents a component of 3-dimensional vectors \( \mu = x, y, z \). These parameters \( A \) and \( m \) are optimized to minimize the free-energy \( F_m / m \).

In order to study static response to shear \( (x_{ij} \rightarrow x_{ij} + \gamma z_{ij}, y_{ij} \rightarrow y_{ij}, z_{ij} \rightarrow z_{ij}) \) of the replicated system, the free-energy \( F_m \) of the whole system would be expanded in power series of the shear-strain \( \gamma_a (a = 1, 2, \ldots, m) \) as \( F_m = F_m (\{ \gamma \}) / V + \sum_{a=1}^{m} \sigma_a \gamma_a + \left( \frac{1}{2} \right) \sum_{a,b=1}^{m} \mu_a \mu_b \gamma_a \gamma_b + \ldots \). where \( \sigma_a \) is the shear-stress of \( a \)-th replica, i.e. \( \sigma_a = (1 / V) \sum_{i,j} \langle (r_{ij}^a) \rangle \) with \( \langle r_{ij} \rangle = \hat{x}_i \hat{r} \hat{r} \hat{r} \). Here we introduced short-hand notations like \( x \equiv x / r \) with \( r = |r| \). The shear-modulus or rigidity (matrix) \( \mu_{ab} \) can be expressed by a fluctuation formula,

\[
\beta \mu_{ab} = \frac{1}{V} \left[ \sum_{\{kl\}} \langle \beta \sigma (r_{kl}^b) \rangle \langle \beta \sigma_b (r_{kl}^b) \rangle - \sum_{\langle mn \rangle \not\in \{kl\}} \langle \beta \sigma (r_{kl}^b) \sigma (r_{mn}^b) \rangle - \langle \beta \sigma (r_{kl}^b) \rangle \langle \beta \sigma (r_{mn}^b) \rangle \rangle \right]
\]

where \( \beta = 1 / T \) and \( \langle \ldots \rangle \) represents a thermal average, which is the local shear-stress associate with a pair of particles. The expression Eq. (1) is derived by simplifying the one given in [24] assuming rotational symmetry, which is valid in the present system, and generalizing the formula to the replicated system. It is more useful than other equivalent expressions involving the 2nd derivative of the potential which can be problematic for the contact potential systems.

Within the 1-RSB ansatz we expect that the rigidity matrix \( \mu_{ab} \) takes the generic form \( \mu_{ab} = \hat{\mu} (\delta_{ab} - \frac{1}{m}) \). Here \( \hat{\mu} = -m \mu_{a \neq b} \) can be regarded as the rigidity of metastable states which is our main concern in the present paper.

The thermodynamic properties of the present system were studied in detail by the cloned liquid approach at the level of 1-RSB in [22]. They are the same as those of the hard-sphere systems [21] in the low temperature limit at \( \varphi < \varphi_{\text{GCP}} \). Here \( \varphi_{\text{GCP}} \) is the so called glass close packing (GCP) density which is an ideal jamming density where the reduced pressure \( \beta P / \rho \) of the equilibrium hard-sphere glass state diverges.

We can assume that the cage size \( A \) is very small deep in the glassy phase. First we note that if \( A = 0 \), \( \mu_{ab} = 0 \) since the cloned liquid as a whole is just a liquid. Next we consider contribution from the fluctuations inside the cages. Focusing on the 2nd term on the r.h.s. of Eq. (1) we notice that it can be separated into three-point terms \( \sum_{i<j<k} \) and four-point terms \( \sum_{i<j<k<l} \). The 1st non-vanishing contribution to \( \hat{\mu} \) at order \( O(A) \) comes from the three-point terms,

\[
\beta \hat{\mu} = -A \beta \mu \left[ \int d^d r_0 d^d r_1 d^d r_2 \left( \nabla^2 \cdot \nabla_0 \right) \beta \sigma (r_{01}) \beta \sigma (r_{12}) g_3 (r_{01}, r_{12}, r_{12}; T^*) \right] + \ldots \quad (a \neq b)
\]

where we introduced a short-hand notation \( T^* = T / m \) and \( \beta^* = 1 / T^* \). In the 2nd equation we approximated the three-point correlation function \( g_3 (r_1, r_2) \) by the Kirkwood approximation in order to make an analytical progressions, \( g_3 (r_1, r_2, r_{12}) \simeq g (r_1) g (r_2) g (r_{12}) \) where \( g (r) \) is the radial distribution function. We switched to the polar coordinates with radial variables \( r_1, r_2 \) and solid angles \( \varOmega_1, \varOmega_2 \). We also introduced the cavity function \( g_3 (r; T) \equiv e^{-\beta \varphi (r)} g (r; T) \) and a function \( \Delta (r; T) \equiv d^d r e^{-\beta \varphi (r)} \) which becomes a delta function in the \( T \rightarrow 0 \) limit enabling analytical computations.

We also have to take into account the renormalization of the interaction between ‘molecules’ due to fluctuations inside the molecules [20,22]. It amounts to replace the original potential by a renormalized one \( m \varphi (r) \rightarrow \varphi_{\text{eff}} (r) \) in the above computation. For the present system we find \( e^{-\beta \varphi_{\text{eff}} (r)} \simeq \theta (r - a) + \sqrt{\frac{\pi}{4 A}} m \delta (r - a) + \ldots \) around the jamming point \( \varphi = \varphi_{\text{GCP}} \) using the results of [22] and assuming that \( A / m \) is small. Finally we obtain,

\[
\hat{\mu} = \frac{T A}{m m \pi a^3} \varphi (a; T^*, \varphi)^3 \left[ \alpha_1 - \alpha_2 \sqrt{\frac{A}{m}} + \ldots \right]
\]

with \( \alpha_1 = (113/120) \pi^2 \) and \( \alpha_2 = (376709 / 22050) \pi^2 \).

The above expression Eq. (4) implies that the scaling property of the rigidity \( \hat{\mu} \) around the jamming point \( \varphi = \varphi_{\text{GCP}} \) is dominated by that of the parameter \( m \). The latter is known to behave as \( m (T, \varphi) = \sqrt{T} \bar{m} (\varphi - \varphi_{\text{GCP}} / \sqrt{T} \), with \( \bar{m} (x) \simeq c_1 |x| \) for \( x < 0 \) and \( \bar{m} (x) \simeq c_1 / x \) for \( x > 0 \). Now we find the rigidity behaves around \( \varphi = \varphi_{\text{GCP}} \) as,

\[
\lim_{T \rightarrow 0} \hat{\mu} = c_+ (\varphi - \varphi_{\text{GCP}}) \quad \varphi > \varphi_{\text{GCP}} \quad (5)
\]

and

\[
\lim_{T \rightarrow 0} \beta \hat{\mu} = \frac{c_-}{\varphi_{\text{GCP}} - \varphi} \quad \varphi < \varphi_{\text{GCP}}
\]
which can be regarded as an entropic rigidity. Here the numerical prefactor are $c_\alpha = c_{1 \alpha}$ and $c_+ = c_{1 +}$ where $c = (6 \phi_{\text{GCP}}/\pi) \alpha_{\text{GCP}} y(1; 0, \phi_{\text{GCP}})^2 (a_1 - 2 \phi_{\text{GCP}})$ with $\alpha_{\text{GCP}}$ being the optimized value of $A/m$ at $\varphi = \varphi_{\text{GCP}}$. Using the values of $\alpha_{\text{GCP}}, \varphi_{\text{GCP}}, y(\varphi_{\text{GCP}})$, $c$ and $c'$ given in [22], we find $c_\alpha \simeq 0.1239496$ and $c_+ \simeq 0.694315$.

A remarkable feature is that the behaviour of the rigidity $\mu$ found above is exactly the same as that of the pressure $P$ [2, 21, 22]. Quite interestingly this is consistent with the result of the experiment on the emulsion [9, 21, 22]. Quite interestingly this is consistent with the result of the the experiment on the emulsion [9, 21, 22] and the MD simulations at finite temperatures [28].

In order to solve the paradox, we next examine $\alpha$ using the values of $\alpha_{\text{GCP}}, \varphi_{\text{GCP}}, y(\varphi_{\text{GCP}})$, $c$ and $c'$ given in [22], we find $c_\alpha \simeq 0.1239496$ and $c_+ \simeq 0.694315$.

MD simulations of shear-stress relaxation A natural protocol to probe the rigidity of glassy states is to observe the plateau modulus which appears in the shear-stress relaxation processes. To study the relaxation deep in the glassy regime systematically, we employ the standard protocol of aging experiments [21, 22]: (1) thermalize the system at a high temperature $T_1$, (2) quench the temperature down to the working temperature $T$ and let the system relax for a waiting time $t_w$ (3) put a small shear-strain $\gamma$ on the system (4) measure the shear-stress $\sigma(\tau)$ as a function of the time $\tau$ elapsed afterwards.

A bi-disperse system of the ratio of radii 1.4 is employed. The relaxation is simulated by solving the underdamped Langevin dynamics using a velocity Verlet algorithm with an integration step $h = 0.01$. The unit of time $t_{\text{micro}} = 1$ is the relaxation time of the non-interacting system. The shear-strain $\gamma$ is applied by an affine transformation and the Lee-Edwards boundary conditions.

In Fig. 1 a), b), we show a representative set of data of the stress relaxation. A notable feature is that there are two kinds of plateaus: the higher one at shorter $\tau$ and lower one at longer $\tau$. The relaxation between the former to the latter strongly depend on the waiting time $t_w$.

We interpret the two plateaus as the following. We regard the higher plateau as the rigidity $\mu_{\text{harmonic}}$ of inherent structures (IS) or energy minima, which is essentially the same as rigidity at zero temperature [2, 3]. For sufficiently short time scales $\tau$, the dynamics should be harmonic so that $\mu_{\text{harmonic}}$ only takes into account harmonic thermal fluctuations around ISs. Indeed we observe that $\gamma$; $T = 10^{-5}$.

![Fig. 1: Relaxation of the shear-stress in the system with $\varphi = 0.67$ ( $\varphi_j \sim 0.648$) at $T = 10^{-5}$. The initial temperature is $T_i = 10^{-2}$ where the system is in the liquid state. The number of particles used is $N = 800$. Comparing with some data of $N = 2400$ we found no appreciable finite size effects within our time window. The strength of the shear-strain is $\gamma = 2.5 \times 10^{-3}$. Comparing with some data of $\gamma = 2.5 \times 10^{-4}$ we confirmed that the linear response holds. The average over initial configurations is taken over 4096 samples. Panel a): data are obtained at $T = 10^{-5}$ after waiting times $t_w = 3 \times 10^{-2}, 10^3, 3 \times 10^3, 5 \times 10^3, 10^4, 3 \times 10^4, 10^5$ from the bottom to the upper curves. The top curve (indicated as $\mu_{\text{harmonic}}$) is the data obtained at $T = 0$ after aging for $t_w = 10^5$ at $T = 10^{-5}$. The data of the pressure $P (T = 10^{-5}, t_w = 10^5)$ is also shown for a comparison. Panel b): data at different temperatures $T = 10^{-5}, 10^{-6}, 10^{-7}$ with $t_w = 3 \times 10^3$ are shown for a comparison.

The initial part of the dynamics depend little on the temperature. The plateau value $\mu_{\text{harmonic}} \simeq 0.047$ agrees well with the value obtained directly at zero temperature [3].

On the other hand the other plateau is much lower than $\mu_{\text{harmonic}}$ and lies at the level of the pressure $P$. This would be interpreted as the rigidity of metabasins (MB) which is a union of ISs [20]. The strong waiting time dependence implies that the system is searching for ISs with lower free-energy within a MB. Then we may interpret the rigidity Eq. [6] and Eq. [5] found at the level of 1-RSB as the rigidity of the MBs rather than ISs.

Discussions The transitions between the ISs inside a MB necessarily involve plastic processes. Indeed we can see avalanche like processes as shown in Fig. 2 at the time scale corresponding to the shear-stress relaxation between the two plateaus. We speculate that they are the floppy modes which are released by opening contacts [17]. Presumably they are extended over the so called isostatic length scale [3]. It is natural to expect that the distribution of the excitation energies of the floppy modes is gap-less so that there is a fraction of active floppy modes
even at vanishingly low temperatures. The floppy modes do not change the distances between the particles in contact but can rotate the contacts. This scenario naturally explains why there is a significant shear-stress relaxation while the pressure remains almost constant in time as shown in Fig. 2. From the theoretical point of view, it would be very interesting to try to understand these features in terms of the $1 + \infty$ RSB scenario \cite{30}.

To conclude, we found that the rigidity of the repulsive jamming system survives at finite temperatures but in a peculiar way: the shear-modulus becomes substantially smaller than at zero temperature even at vanishingly low temperatures, contrary to usual crystals and glasses.

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\[ \text{FIG. 2: Snapshots of a plastic event during stress-relaxation. Here we used a 2-dimensional version of the model at } \phi = 0.85 \text{ (} \phi J \sim 0.84\text{). The system is initially perturbed by } \gamma = 0.05 \text{ and let to relax at } T = 10^{-6} \text{. The configuration of particles are represented by the circles and that of the contact forces } f_{ij} = -d\phi(r_{ij})/dr_{ij} \text{ are represented by bonds whose thickness is proportional to } f_{ij} \text{. The panels a) and b) show the snapshots before/after a plastic event (whose duration is about } 10^4 \text{). In panel c) the configuration of the particles before/after the event are shown by lighter/darker colors.} \]

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