Nature of the glass transition in 2D colloidal suspensions of short rods

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Abstract

The glass transition, a many-body system undergoes an apparent dynamic arrest with no appearance of long-range order, is one of the most challenging problems in condensed matter physics. Recently attention has been focused on the puzzling two-step transition observed in colloidal suspensions of ellipsoids. It was observed that micro nematic domains appearing with increasing packing density suggesting that such micro domains may be the structure origin of the two-step glass transition. Here we report an experimental study of monolayers of colloidal rods which exhibits a two-step glass transition with no appearance of pseudo-nematic domains. Instead, it was found that parallel and perpendicularly packed rods form local free energy minima in configurational space, separated by an activation barrier. This barrier increases significantly when rotational glass transition is approached, thereby the rotational motion is frozen while the translational one remains diffusive. We propose that the activation barrier for rotation is the key to understanding the two-step glass transition in general.

1. Introduction

In the Landau paradigm of phase transitions [1], e.g. when water freezes into ice, the solidification is understood as a spontaneous symmetry breaking phenomenon, the system develops a long-range order and acquires an emergent property of shear rigidity. The phase transition can be understood, sometimes quantitatively, by a local order parameter. The success of this Landau paradigm is immense, from alloys to superconductors [2]. Still there are many problems in condensed matter fall outside of the Landau paradigm of symmetry breaking and local order parameter. For example, the apparent phase transitions in liquid helium films, in superconductor films, in 2D solid melting, etc, cannot be understood using Landau’s approach. No local order parameter can be defined to distinguish the apparent phases of matter. The theory of Kosterlitz and Thouless [3] based on topological order and topological defects gave a self-consistent explanation to these problems. The introduction of topology into statistical mechanics by Kosterlitz and Thouless represented a paradigm shift from that of Landau.

The glass transition may require another new paradigm for quench disordered phases of matter [4]. Glass transition refers to the ubiquitous phenomena in many condensed matter systems that an apparent dynamic arrest occurs with no appearance of long-range order [5]. This phenomenon occurs in window glasses [6], in disordered type-II superconductors in a magnetic field [7], in disordered magnets [8],
in colloidal matter on a rough substrate [9], in multi-components colloidal glasses [10, 11], and even in seemingly simple 2D colloidal matter of monodispersed ellipsoids [12], or rods [13]. The microscopic origins of the various glass transitions remain controversial.

For a glassy system, there is no positional order to associate the glassy phase with, thus the apparent shear rigidity has been treated as a diverging viscosity [6], i.e. the system appears elastic only at finite time and length scales. However, two recent independent experiments [10, 11] suggested that 2D colloidal glasses, consisted of polydispersed microspheres, appear to be elastic at large length and long time scales, again raising the question on the physical mechanisms of the glass transition. In this context, the apparent 2D glass transitions in monodispersed anisotropic particles are particularly interesting. It is now well documented [12, 14–17] that monodispersed colloidal ellipsoids can form a stable glass phase in 2D. The transition into the glass phase seems to involve two steps [12, 16, 18]. With increasing packing density, the system first goes from an isotropic liquid to an orientational glass phase with orientational dynamics frozen, but there are still large clusters of particles can move translationally. Only at an even higher packing density does the system become translationally frozen. Given the monodispersity nature of the particles, it is highly expected that the mechanism(s) of the glass transition can be readily explored.

It has been suggested that local nematic structures may provide a mechanism for the glass transition of colloidal ellipsoids [14–16]. Specifically, for ellipsoids with large aspect ratio \( p > 2.5 \), it has been proposed that the formation of pseudo-nematic domains leads to a decoupling between translational and orientational motion [12]. For \( p < 2.5 \), however, the pseudo-nematic domains disappear, the two glass transitions merge into a one-step transition [15, 16, 19].

To address the universality of the proposed nematic domain model [14–16], here we report an experimental study of the glass transition in 2D suspensions of colloidal rods. Like ellipsoids, colloidal rods undergo a two-step glass transition even for small aspect ratio, \( p = 1.5 \). Unlike ellipsoids, there is no evidence of pseudo-nematic domain formation at any packing density. We suggest that these new experimental observations, together with previous observations in which pseudo-nematic domains were observed [12, 16], call for a new theoretical paradigm in the understanding of colloidal glass transition, at least for 2D. We propose a local kinetics model in which the local angular distribution between colloidal rods in the glassy regimes is a manifestation of the kinetics of local rod–rod interactions. We argue that the local bond angle distribution in the glass phase represents the Boltzmann distribution of the rod–rod interactions at the moment when the system falls out of equilibrium. With this assumption, we extracted the effective free energy landscape of the rod–rod interactions. We found that the parallel and perpendicular local orientational configurations of the rods are local free energy minima, separated by an activation barrier. The large activation barriers between the local configurations hinder rotation of the rods, resulting in a decoupling between translational and orientational motion, and consequently a two-step glass transition. We believe that the activation barrier between well-defined local configurations holds the key for a universal understanding of the two-step glass transition.

2. Experimental

2.1. Colloidal samples and video microscopy.

Our colloidal rods were fabricated from the monodispersed polystyrene (PS) microspheres (Duke Standards 4204A, Themo Fisher) using a heating–stretching–cooling procedure [20]. The starting microspheres have diameters of 4.000 \( \mu m \pm 0.043 \mu m \), with a polydispersity of 1%. We prepared three batches of rods with aspect ratio \( p_1 = L/D = 5.1 \mu m/3.4 \mu m = 1.5 \), \( p_2 = 6.1 \mu m/2.7 \mu m = 2.3 \) and \( p_3 = 6.8 \mu m/2.5 \mu m = 2.7 \), where \( L \) and \( D \) are the length and diameter of the rods, respectively. The rods were characterized using a scanning electron microscope (SEM, Hitachi SU8010). The rods, when lied down on a flat surface, have cross-sectional areas \( (A) \) of 13.78 \( \mu m^2 \pm 0.84 \mu m^2 \), measured from the SEM images of \( \sim 200 \) individual rods, see figure S1 of the supplementary information (https://stacks.iop.org/NJP/22/103066/mmedia). The cleaned rods were suspended in an aqueous solution of 3 mM sodium dodecyl sulfate (SDS). The zeta potential of the rods was measured to be \( \sim 40 \) mV using a dynamic light scattering instrument (DLS, Zetasizer Nano ZS, Malvern Instruments LTD). The corresponding Debye screening length was estimated to be \( \sim 10 \) nm. However, no aggregations of the rods in a dilute suspension for a long time were observed, which demonstrates that the phase behavior of the rods is not dominated by the van der Waals interactions and the rods can be considered as hard particles [21, 22].

The suspension of the rods was confined between two glass coverslips (Themo Fisher) to produce a monolayer of the particles (figure 1(a)). The areal fraction \( (\phi) \) is defined as the ratio of the total cross-sectional areas of the rods in the field of view divided by the area of the field of view. The particle number density was calculated as \( \rho = N/S \), where \( N \) is the number of the particles in the field of view, and \( S \)
is the total area of the field of view of the camera. We then obtained the areal fraction using $\phi = Ap$. In our particle counting algorithm, only the particles with their centers-of-mass located inside the field of view are counted. This method typically gives an accurate account of the number of particles in the field of view. However, we find that even after equilibrating for a long time (hours), the sample region can have as many as $\pm 10$ particles variations when we change the field of view with a field view of $\sim 2000$ particles, giving an error bar of $\Delta \phi \sim 0.002–0.005$ in the areal density measurement, as shown in figure 1(c).

The thermal motion of the rods was recorded by an optical microscope (Nikon Ti) with a CCD camera (Basler) at 5 frames per second for low $\phi$ and 1 frame per second for high $\phi$. During measurements at each $\phi$, no drift was observed. The center positions and orientations of individual rods were obtained using the open source program ImageJ. The angular resolution was 1°, and the spatial resolution was 60 nm. More experimental details are in the supplementary information.

### 2.2. Four-point dynamic susceptibility.

Here we carry out the standard procedure of characterizing a colloidal glass transition using the empirical method of four-point dynamic susceptibility [23–25]: $\chi_4(\Delta t) = N\langle Q_2(\Delta t, w_c) \rangle^2 > < Q_2(\Delta t, w_c) >^2$

where $N$ is the number of the particles, $w_c = r_c$ or $\theta_c$, $\Delta t$, $r_c$, $\theta_c$ are the probed time, distance, and angular scales, $Q_2(\Delta t, r_c) = \frac{1}{N} \sum_{i=1}^{N} \exp\left(-\frac{2\Delta t r_c^2}{2w_c^2}\right)$ for the translational motion, and $Q_2(\Delta t, \theta_c) = \frac{1}{N} \sum_{i=1}^{N} \exp\left(-\frac{2\Delta t \theta_c^2}{2\gamma^2}\right)$ for the rotational motion. Here, to clearly demonstrate the heterogeneous dynamics in different phases, we choose the values of $r_c$ and $\theta_c$ to maximize $\chi_4$ following a previously reported practice [25]. The other choices of the length scale do not change the results. For translational motion, $r_c = 0.2 \, \mu m, 0.44 \, \mu m$ and 0.13 $\mu m$ for $\phi = 0.42, 0.81$ and 0.83, respectively. For orientational motion, $\theta_c = 3^\circ, 6^\circ$ and $2^\circ$ for $\phi = 0.42, 0.81$ and 0.83, respectively.

### 3. Results and discussion

We applied two common approaches to quantify the glass transition, the mode-coupling theory (MCT) [26–28], and the Vogel–Tammann–Fulcher (VTF) model [29]. It should be noted that the MCT formulated by Görtze is a microscopic theory [26–28], whereas the VTF scaling is an empirical approach [24, 29]. Here, we present the MCT scaling, instead of the VTF scaling, so that we can compare the results with those for ellipsoidal glass [12, 15], where the MCT scaling is found to give better description of heterogeneous glassy dynamics. A typical result using the VTF is included in the supplementary information (figure S2).

First, the self-intermediate scattering function $F_i(q, t) \equiv \frac{1}{N} \langle \sum_{j=1}^{N} e^{i q \cdot [r_i(t) - r_i(0)]} \rangle$ and orientation correlation function $L_\theta(t) \equiv \frac{1}{N} \langle \sum_{j=1}^{N} \cos n[\theta_j(t) - \theta_j(0)] \rangle$ were calculated, where $r_i(t)$ and $\theta_j(t)$ are the position and orientation of rod $j$ at time $t$, $N$ is the total number of particles, $q$ is the scattering vector, $n$ is a
positive integer, and $<>$ denotes a time average. The relaxation time $\tau$ is defined as the time at which $F_r(q,t)$ and $L_0(t)$ have decayed to $1/e$ [15, 30]. Note that $F_r(q,t)$ and $L_0(t)$ decay faster for larger value of $q$ and $n$, respectively (figure 1(b)), and different choices of $q$ and $n$ can yield the same glass transition point [12, 15]. More results of $F_r(q,t)$ and $L_0(t)$ are shown in figure S2.

The glass transition point $\phi_g$ is determined from the MCT scaling, following the previous work on the glass transition of colloidal ellipsoids [12, 15, 16]. According to the MCT, the relaxation time $\tau$ diverges algebraically at $\phi_g$: $\tau(\phi) \sim (\phi_g - \phi)^{-\gamma}$, where $\gamma = \alpha + \beta$ [26]. Here $\alpha$ and $\beta$ are exponents in the decay law $F_r(q,t) = f_q + h_q t^{-\alpha}$ and the von Schweidler law $F_r(q,t) = f_q - h_q t^{-\beta}$ for the initial $\beta$ relaxation and the crossover time to the $\alpha$ relaxation, respectively [31–33]. $f_q$ and $h_q$ are the plateau height and amplitude. We obtained $b$ from fits to $F_r(q,t)$ and $L_0(t)$, and $a$ from the relation between $a$ and $b$ [26, 27]. This approach is consistent with the common practice in characterizing the glass transition of ellipsoids [15]. The result shows that the $\tau^{-1/\gamma}$ is linear in $\phi$ for different choices of $q$ and $n$, and expectedly, all the scalings show there are two glass transitions at $\phi_g = 0.78 \pm 0.01$ for rotational motion and $\phi_g^T = 0.82 \pm 0.01$ for translational motion for rods with $p = 1.5$. Thus the orientational glass transition occurs at a lower density than the translational glass transition (figure 1(c)). Moreover, this two-step glass transition is also observed for rods with $p = 2.3$ and 2.7 (see SI, figure S4). The difference between $\phi_g^T$ and $\phi_g^T$ increases with increasing $p$, which is consistent with that in ellipsoidal systems [12, 16].

To examine if the system is in an orientational glass state between $\phi_g^T$ and $\phi_g^T$, we measured the dynamical heterogeneity for rods with $p = 1.5$, at $\phi = 0.42, 0.81$, and 0.83. The dynamical heterogeneity was quantified by the four-point dynamic susceptibility, $\chi_4$ [24–25]. $\chi_4$ is small at $\phi = 0.42$ and 0.83 (figure 2(a)) signifying a relatively homogeneous dynamics, characteristic of a colloidal liquid and fully glassy phase, respectively. The homogeneous dynamics is also confirmed by snapshots of the fast-moving particles [34]. At $\phi = 0.81$, the $\chi_4$ for the translational motion displays a large peak, while for the orientational motion $\chi_4$ remains nearly flat without a distinct peak. This suggests that the heterogeneous dynamics is strong only in the translational motion, but not the orientational motion. This is the characteristic of an orientational glass state [12].

It was suggested that ellipsoids with the large aspect ratios ($p > 2.5$) can form pseudo-nematic domains, which in turn lead to the two-step glass transition [12]. In the rod systems here, for $p = 1.5$, no pseudo-nematic domains were observed for any $\phi$. For rods with larger anisotropy $p = 2.3$ and 2.7, micro-domain structures with local nematic order can indeed be observed (see figure S3). This observation suggests that the structure of the nematic domains is one of the local configurations that drives the two-step glass transition, but not the key mechanism causing the two-step glass transition.

Next, we propose a new microscopic approach for analyzing the structural information of the glass transition. To gain insights into the nature of the glass transition in this rod system, we analyze the distribution of the relative angles between the rods. This distribution may reveal the local structures formed by the nearest neighboring particles. It should be pointed out that our approach was inspired by Chong and Götze [35] in which they first proposed to look beyond the static structure factor $S(q)$ (which is related to the radial distribution function $g(r)$ shown in figure 3(b) by a Fourier transform). In figure 3(a), the spatial distributions of the local orientational configuration for rods with $p = 1.5$ are shown in a graphical representation. The positions and orientations for all nearest neighboring particles are normalized with respect to the reference rod sketched in the center of the figure, and the nearest neighboring particles are plotted at the points. The color of the point denotes the relative orientation of the nearest neighboring particle, $\Delta \theta$. A clear observation is that with increasing packing density, the rods are oriented relative to each other in either parallel or perpendicular local configurations (see blue and red color regions). Such increased local configurations are confirmed by the radial distribution function $g(r)$. With increasing $\phi$, the first peak in $g(r)$ splits into three peaks. These three peaks represent two parallel local configurations (the first peak and the third peak) and a perpendicular local configuration (the second peak) (figure 3(b)). The peaks become more pronounced with increasing $\phi$, indicating the enhancements of the parallel and perpendicular local configurations when approaching the glass transition.

In figure 3(a), we notice that, immediately adjacent to the reference particle, the angular distribution of its nearest neighbors is highly anisotropic. This anisotropy increases sharply with increasing packing density as the system approaches the first transition, when the orientational degrees of freedom freeze out. The obvious question is whether this effect, as shown in the striking graphical representation in figure 3(a), reveals any hint of the microscopic mechanism of the two-step glass transition as shown in figure 1. Here we propose a kinetic hypothesis that this locally anisotropic angular distribution represents the spatial distribution just before the system falls out of equilibrium, i.e. becomes stuck. Since the detailed balance should be preserved locally for the fluctuations between the rods prior to being stuck, thus one can use Boltzmann statistics to extract the effective free energy landscape of the rod–rod interactions from their relative angle distribution.
Figure 2. Dynamical heterogeneity confirms the existence of an orientational glass state. The four-point dynamic susceptibility, $\chi_4$, for translational motion and orientational motion for the rods with aspect ratio, $p = 1.5$. Insets are bright-field micrographs of fast-moving particles for translational motion (red squares) and orientational motion (green dots) at, from top to bottom, $\phi = 0.42, 0.81$ and $0.83$. The lack of orientational cooperative motion and the appearance of strong translational cooperative motion at $\phi = 0.81$ indicate that the orientational motion is frozen but the translational motion remains, confirming the existence of the orientational glass phase. The fast-moving particles are defined as the particles with the 10% largest displacements over the lag time $\Delta t$ that maximizes $\chi_4$. Scale bar: 20 μm.

It is worth noting that using local fluctuating configurations in a 2D colloidal lattice to extract effective potential was successfully attempted previously [36, 37]. After binning the data in figure 3(a), for the nearest neighbor range $r/D \leq 1.8D$, we obtained the probability distribution of local orientational configuration, $P(\Delta \theta)$. The distribution shows a clear increase of the probability of the parallel and perpendicular configurations and an obvious decrease of the intermediate configurations as $\phi$ increases (figure 4(a)). The question here is whether there is a simple physical mechanism for this anisotropic probability distribution. Here we argue that this angular dependence of the probability distribution may be the relic of the kinetic arrest at the glass transition. Namely, just before the system falls out of equilibrium, the rods locally can rearrange due to thermal agitation and thus one may use the Boltzmann distribution, assuming $P(\Delta \theta) \propto e^{-V(\Delta \theta)/k_BT}$, where $V(\Delta \theta)$ is the effective potential energy (Gibbs free energy) of the local configuration $\Delta \theta$, $k_B$ is the Boltzmann constant and $T$ is the temperature. Using this kinetic model, we estimate $V(\Delta \theta)$ from $k_BT \ln P$ (figure 4(b)). Indeed the low energy configurations belong to the parallel or perpendicular local structure of the rods, and the angled configurations are less energy favorable. We suggest that this effective potential energy in the local nearest neighbor configuration space is the key to the two-step glass transition. Once a larger probed neighbor range is chosen, the energy difference between
Figure 3. Rods form local orientational structures approaching the rotational glass transition. (a) The spatial distribution of the orientational configuration of nearest neighboring rods ($\Delta \theta$) at $\phi = 0.42$ (top panel) and 0.73 (bottom panel) for the rod monolayer with $p = 1.5$. Blue and red colors represent the parallel and perpendicular configurations, respectively. More orientational structures have formed when approaching rotational glass transition point. Arrows in bottom panel indicate that particles can keep its orientation while move along the axis of the neighboring particles, i.e. rotation is frozen while the translation is possible. (b) The radial distribution function $g(r)$. For clarity, the curve has been shifted vertically for different $\phi$.

favorable and unfavorable configurations decreases (figure S5). This is reasonable since the system is more isotropic at larger distance. It is expected that such preferred configurations disappear at large distances.

One immediate reward of our local configuration analyses is that now one can quantitatively address the structural origin associated with the two-step glass transition, by presenting the effective activation barrier $\Delta V (= V_{\text{max}} - V_{\text{min}})$ as a function of packing density (figure 5). Interestingly, the barrier shows a plateau as the system enters into the orientational glass regime. Further increasing density $\phi$ the barrier rises again when the system enters the second glass transition where the translational motion freezes. The plateau in the activation barrier is particularly striking. It suggests that the barrier to rotation in the orientational glass state is weakly dependent on the density. Motivated by this puzzling effect, we repeated measurements on colloidal rods of two additional aspect ratios, $p = 2.3$ and 2.7, the results are plotted together with $p = 1.5$ in figure 5. It is found that the plateau effect is present for all three values of aspect ratios. Intuitively, one expects the barrier between the parallel and perpendicular configurations to be larger for particles of larger aspect ratios. This seems to be true at low packing density, in the fully liquid phase, e.g. see figure 5 at $\phi = 0.65$. The fact that the plateau of the activation barrier is smaller for particles of large aspect ratios can be explained if the height of the plateau represents a threshold or critical barrier that forces the system out of equilibrium. Since it takes longer time for longer rods to rotate relative to each other, thus the system will be stuck at a lower barrier height. This decreased activation barrier suggests the freezing transition is a kinetic effect, which is further demonstrated by the change of the effective potential energy at different $\phi$. For example, the effective potential energy changes by $\sim 2k_B T$ between $\phi = 0.43$ and 0.78 for the $p = 1.5$ system. Thus one expects that the dynamics changes by about a factor of $\sim e^2 = 7.4$. However, the corresponding dynamics slows down by a factor of $\sim 40$ (figure S2(b)). Thus, it is clear that this structural barrier does not play the decisive role to the slowing relaxation dynamics. Moreover, if the dynamics would indeed be related to a simple barrier, it is expected an exponential time dependence of the correlation function, which is not the case (figure S2(b)). These thereby suggest the kinetic nature of the glass transition here.

We thus propose a universal mechanism for the two-step glass transition of anisotropic particles: the large activation energy barriers between the local configurations promote the hindrance of the local rotation of anisotropic particles. This results in a decoupling between translational and orientational motion, consequently leading to a two-step glass transition. We should point out that the decoupling of the orientational and translational degrees of the freedoms occurs in the equilibrium isotropic-nematic-solid transition of two-dimensional hard ellipses studied by computer simulation [38]. It is possible that an
Figure 4. Increase of concentration of the rods results in free energy barriers in orientational configurations. (a) The probability of $\Delta \theta$ at different $\phi$ for rod monolayers with $p = 1.5$. (b) Potential energy $V(\Delta \theta)$ of the orientational configuration at different $\phi$. $V(\Delta \theta)$ was estimated from $-\ln P(\Delta \theta)$. The relative arrangements of rods at three relative angles are shown.

Figure 5. Activation barrier $\Delta V$ increases sharply when approaching the orientational glass transition point. The vertical lines indicate the orientational glass transition points for the rods with different aspect ratios, $p$. The transition points, are 0.74 for $p = 2.7$, 0.75 for $p = 2.3$, and 0.78 for $p = 1.5$. The horizontal lines indicate the heights of barrier plateaus.

underlying connection between the equilibrium and the non-equilibrium phase transitions exists for the anisotropic particles. This issue deserves more future studies.

Moreover, it is interesting to compare our observations here with that in the two-stage melting of a 2D crystal [3, 39, 40]. From the solid side, the first transition is edge dislocation unbinding, free edge dislocations allow parts of the lattice to slide past each other, making the effective shear modulus vanish at long length and time scales, rendering the system a liquid [3]. However, the orientational order remains, hence a hexatic phase [39]. Only at a higher temperature when disclinations unbind, one part of the system can rotate freely relative to another, resulting in the isotropic liquid [39]. Here the situation is strikingly similar: starting from the frozen glass state, with lowering packing density, when the rods can slide past each other, it first gives rise to an orientationally frozen glass but a translationally flowing liquid [12]. Only when density is further lowered, when the rods have free space to rotate, an isotropic liquid forms.
4. Conclusions

We found that a two-step glass transition in a 2D colloidal suspension of rods during which there is no pseudo-nematic domain formation. Instead we observed that there exist locally favorable configurations due to shape anisotropy of the rods. We propose a kinetic hypothesis in which it is assumed that the distribution of the relative angles between the rods in the glassy state represents the local equilibrium configurations of the rods before they become stuck, one can invoke Boltzmann distribution to extract a local free energy. The existence of two preferred configurations implies a free energy barrier between them. This barrier becomes more pronounced with increasing particle concentration as expected. We conclude that this energy barrier, which promotes the arrest of the particle rotation, is the mechanism of a two-step glass transition in colloidal suspensions of rods. And we believe the energy barrier is the key to understand the two-step glass transition in other systems of anisotropic particles beyond rods and ellipsoids. We hope that future theoretical studies will develop the local equilibrium model we proposed here to account for the details of packing density dependence of the effective barrier through the two-step glass transition.

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