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T-structured fluid and jamming in driven Brownian rotators

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Abstract. – The dynamics of two-dimensional suspensions of Brownian rods which are driven by an external torque are explored by computer simulation and instability theory. For increasing density the system self-organizes into a "T-structured fluid", where neighbouring rods preferentially orient perpendicular to each other until an overlap density is reached. Then a sharp jamming transition occurs towards a dynamically blocked state with parallel neighbouring rods.

Colloidal suspensions are excellent model systems for phase transitions in equilibrium and non-equilibrium [1]. In particular, active particles [2, 3] and systems driven by external fields [4] show a wealth of non-equilibrium phase transformations. Brownian rotators, as realized by rodlike colloidal particles which are driven by an external torque, are an important example with many potential applications as Brownian motors [5] and as mixers in micro- and nanofluidic devices [6, 7].

In this letter we study the self-organization of an ensemble of interacting driven Brownian rotators. In particular, we study a simple two-dimensional model of mutually repellent Brownian rods which are driven by a constant external torque using non-equilibrium computer simulations. The motivation to study such a simple model stems from the fact that the key ingredients can be realized in quite different experimental set-ups: first, suspensions of non-spherical birefringent particles can be driven by circularly polarized light, as has recently been demonstrated by Chaikin and coworkers [1, 8] and by Bishop and coworkers [9], see also ref. [10] for the basic physics. The magnitude of the torques acting on these particles is easily tunable by the applied power. These rotators can also be confined to two dimensions either by an optical tweezer or in a pendent liquid-gas interface [11]. A second realization are star-shaped rotators in a Langmuir monolayer whose rotational motion can be micromanipulated using optical tweezers [12]. Finally, rod-like dust particles [13, 14] in a quasi–two-dimensional plasma sheath can in principle be rotated by external fields as well.

For strong external torques, we find a novel self-organization behaviour of the rods. Below an overlap density, the system self-organizes into a "T-structured fluid", where neighbouring rods preferentially orient perpendicular to each other avoiding mutual overlap by a coordinated

motion. This dynamical T-structure which is gradually increasing with density is reminiscent to the static three-dimensional house-of-card structure as found in the gelling of clay particles [15]. If an overlap density is exceeded, a sharp jamming transition occurs towards a dynamically blocked state where neighbouring rods are aligned preferentially with a parallel orientation. This is a structural non-equilibrium phase transition which we call jamming [16,17] since it originates from a mutual dynamical hindrance of the rods. We may view this effect as the rotational counterpart of systems translationally driven by an external force [18,19].

In our model, we consider N two-dimensional rod-like particles moving in an area A which are characterized by their center of mass and their orientation. The rods interact via a Yukawa segment model which was shown to be a good representation of charged rod-like colloids [20]. There are $N_\mathrm{b} = 10$ equidistant beads along the rods; the bead spacing along the rods is given by the bead diameter σ, which sets the total rod length as $L = N_\mathrm{b}\sigma$. The bead diameter σ serves as a natural length scale. The repulsive interaction potential between beads of different rods is $V(r) = U_0\,\sigma\,\frac{\exp[-\kappa(r-\sigma)/\sigma]}{r}$, where r is the interbead distance, U_0 is an energy scale and κ is the dimensionless screening parameter. The number density of the rods is specified in terms of the dimensionless area packing fraction $\eta = \pi N L^2/4A$ and the system temperature is T.

The rods are moving according to Brownian dynamics [21] with rotational friction γ_R and anisotropic translational frictions $\gamma_{||}$ and $\gamma_\perp$, parallel and perpendicular to their orientations [21]. These friction coefficients were taken from ref. [22] for a rod aspect ratio of 10 and a rod diameter σ. The standard Langevin dynamics with neglected hydrodynamic interactions was adopted with random kicks of the solvent which keeps the suspension of rods at constant thermal energy $k_\mathrm{B}T$. On top of that the rods are exposed to a constant external torque D which tends to rotate all rods in the same direction driving the system into a non-equilibrium steady state [23].

We solved the Langevin equation for the rod centers and orientations in a Brownian dynamics computer simulation with a finite time step $\Delta t = 0.009\tau_\mathrm{B}$, where $\tau_\mathrm{B} = \gamma_{||}\sigma^2/U_0$ is a suitable Brownian time scale. The rods are in a quadratic box with periodic boundary conditions in x and y directions. Typically, we simulated $N = 80$ rods for 300000 time steps to get into the steady state. Statistics in the steady state were gathered by a further time average over typically 100000 time steps. In our simulations we fixed the external torque to $D/U_0 = 150$ and the screening parameter to $\kappa = 2$. We varied the rod density η and the reduced temperature $T^* = k_\mathrm{B}T/U_0$.

In order to characterize mutual orientations of neighbouring rods we introduce the order parameter

$$m = \frac{\int_0^a g(r)\cdot\cos^2(\theta(r))}{\int_0^a g(r)}\,, \tag{1}$$

where $\theta(r)$ is the relative angle between two rods with the distance r, $a = (\frac{N}{A})^{-\frac{1}{2}}$ is the average distance of the rods and $g(r)$ is the pair correlation function between the centres of the rods. Hence, if $m = 0$, neighbouring rods are perpendicular oriented while they are parallel when $m = 1$. If there is no correlation of neighbouring correlations, $m = 0.5$.

In fig. 1, the order parameter is shown *vs.* rod density for different temperatures. Obviously, m equals 0.5 for the non-interacting system at small densities. For zero temperature, there is a clear built-up of a *T-structure*, as signalled by a value for the order parameter which is close to zero. At a certain threshold density of $\eta_\mathrm{c} \approx 0.95$, the orientation of neighbouring rods drastically changes to be parallel. This is accompagnied by a sharp jump in the order parameter m from zero to a finite non-vanishing value which is clearly seen in fig. 1. Within our numerical accuracy this is a sharp discontinuous jump pointing to a true non-equilibrium phase transition. An increase in the temperature makes the jump Δm in the order parameter

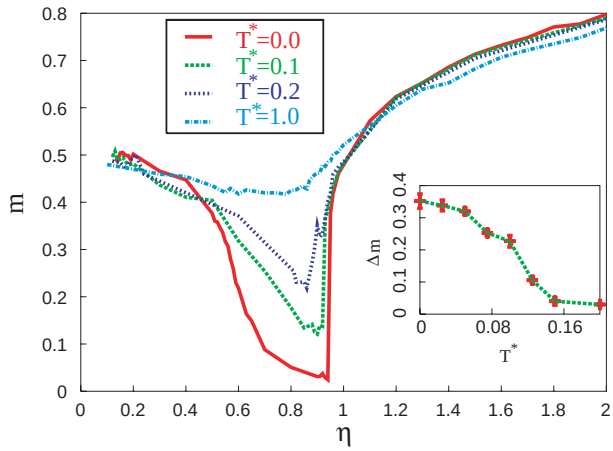

Fig. 1 – Order parameter m of neighbouring orientations *vs.* the area packing fraction η of the rods for different reduced temperatures T^*. The inset shows the maximal variation Δm of the order parameter within a small packing fraction interval of 0.01 as a function of T^*.

smaller (see the inset of fig. 1). For temperatures larger than roughly 0.15, we can only detect a continuous crossover within the statistical accuracy of our data. We have checked that the transition occurs at the same threshold density for a larger system with $N = 400$ rods indicating that there are no finite-size effects. We have further examined hysteresis effects by slowly increasing and decreasing the density but we could not find any indication of hysteresis [24].

In order to resolve dynamical anomalies at the transition, we have calculated the mean angular velocity $\langle\omega\rangle$ of the rods in the steady state. For vanishing rod densities $\eta = 0$, clearly $\langle\omega\rangle = D/\gamma_{\mathrm{R}}$, therefore it is convenient to plot a reduced mean angular velocity $\langle\omega\rangle^* = \langle\omega\rangle\gamma_{\mathrm{R}}/D$. In fig. 2, this quantity $\langle\omega\rangle^*$ is shown *vs.* the rod density η for different temperatures. Interestingly, $\langle\omega\rangle^*$ is almost independent of density. The self-organization into the T-structure clearly avoids friction by mutual collisions and allows for a fast collective rotation of the rods.

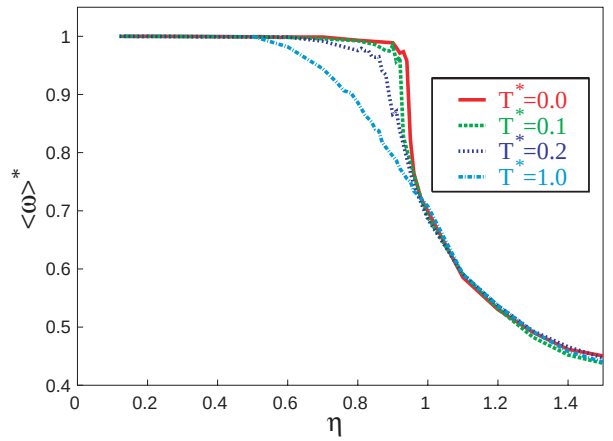

Fig. 2 – Mean angular velocity $\langle\omega\rangle^*$ normalized to its zero-density limit *vs.* area packing fraction η of the rods for different reduced temperatures T^*.

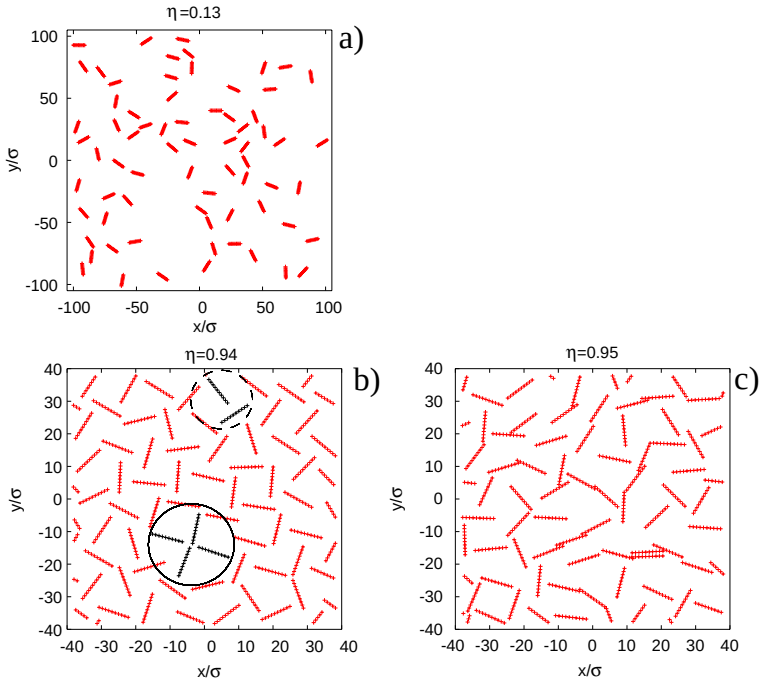

Fig. 3 – Simulation snapshots in the steady state at $T^* = 0$ for three different densities. All beads are shown as a cross. The size of the periodic simulation box is given in the xy plane in units of the bead distance σ. a) Low-density snapshot at $\eta = 0.13$ revealing almost freely rotating rods. b) Density below the jamming transition, $\eta = 0.94$. The T-structure is clearly visible, *e.g.* marked by the broken circle. A four-rod collision event is marked in the solid circle. c) Density above the jamming transition, $\eta = 0.95$. Jamming shows up as preferred parallel orientation of neighbouring rods.

The sharp transition from a T-structure to parallel rods is dynamically accompanied by a drastic change in $\langle\omega\rangle$ which gives clear significance for a dynamical *jamming*. Therefore, we call the phase transition "jamming transition". Again, consistent with the order parameter analysis of fig. 1, the sharp transition becomes a smooth crossover when $T^* > 0.15$.

More detailed information about the change of rod configurations near the jamming transition is contained in typical simulation snapshots of the steady state in fig. 3 at zero temperature. First, as a reference situation, we show a low-density snapshot at $\eta = 0.13$ in fig. 3a, where the dynamics of neighbouring rods are almost uncorrelated, hence the rods are almost free rotators. Snapshots slightly below, $\eta = 0.94$, and above, $\eta = 0.95$, the jamming transition are presented in figs. 3b and c. A typical T-structure below jamming is indicated by a broken sphere in fig. 3b. On the other hand, above the density threshold, jammed neighbouring rods with parallel orientations are clearly visible in fig. 3c. Finally, simulation data for the location of the threshold density η_c associated with the loss of the T-structure and the dynamical jamming are presented in fig. 4 in the temperature-density plane. Note that we have restricted the temperature to values where the transition is accompanied by a finite jump in the order parameter. More details of an end-point where this jump vanishes have to be explored in future studies.

Let us now present a theory for the location of the jamming transition in the temperature-density plane which is based on a simple instability analysis of the T-structured fluid. The

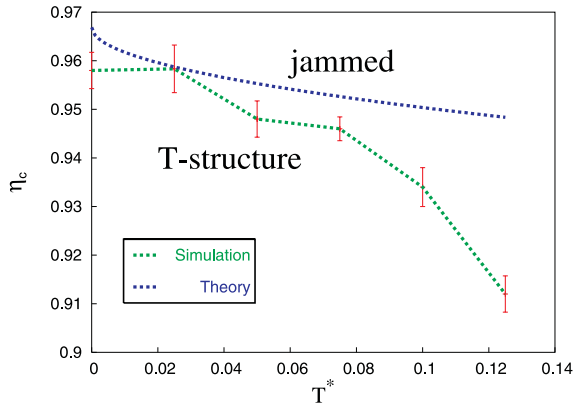

Fig. 4 – Phase diagram for the threshold density η_c in the temperature-density plane separating the jammed from the T-structured region. Both simulation results (with their statistical uncertainties) and data from the instability analysis are compared.

stability of the T-structure is mostly governed by the situation where four rods are close to contact. This situation is sketched in fig. 5. For hard needles of length L, the maximal packing density of a T-structure can be achieved if all rotators are placed on a simple square lattice with lattice constant $L/\sqrt{2}$ which corresponds to a maximal packing fraction $\eta_0 = \pi/2 = 1.57\ldots$. We approximate the short-range fluid structure by this idealized solid and assume an averaged width δ above which the needle end points are smeared, see again fig. 5. Upon the action of the external torque D, it takes a passage time $\tau = 2\delta\gamma_R/LD$ to rotate one rod across the confining cone shown as a hatched area in fig. 4. During this time the rods feel collisions with the neighbouring rods by an averaged force $\bar{F}$ which can be estimated as follows. We assume that the leading part of the collisions comes from the outermost segment interactions $V(r)$ alone. We define an interaction range ζ needed to compete with the external torque D via $\frac{L}{2}|\mathrm{d}V(\zeta)/\mathrm{d}r| = D$. Typically, this range is smaller than the width δ. Assuming a homogeneous distribution of the rod ends in their hatched area of width δ, we get $\bar{F} = (2D/L)4(\zeta/\delta)^2$, where $(2D/L)$ is the typical force for interacting rods within a distance ζ, and $4(\zeta/\delta)^2$ is the conditioned probability to find *two* rod ends with a distance smaller than ζ. During the passage time τ there will be a mean displacement δ' of the end points by a combination

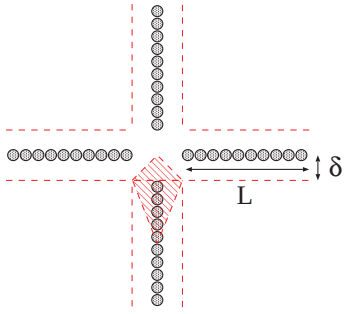

Fig. 5 – Schematic plot of four colliding rods in the T-structured phase as marked by a solid circle in fig. 3b). The hatched area is one key quantity in the instability analysis.

of rotation and translation parallel and perpendicular to the rod orientation and thermal diffusion: $\delta' = \bar{F}\Gamma\tau + \delta_T$ with $\Gamma = 1/\gamma_{||} + 1/\gamma_\perp + L^2/4\gamma_\mathrm{R}$ and the thermal displacement δ_T given by $\delta_T = \sqrt{k_\mathrm{B}T\Gamma\tau}$. If δ' is larger than δ, the situation is unstable, the stability limit is hence reached for $\delta' = \delta$ which has the numerical solution δ_c. For zero temperature, the analytical solution is $\delta = \delta_\mathrm{c} = 4\zeta\sqrt{\Gamma\gamma_\mathrm{R}}/L$. Finally, this threshold value in δ translates into the instability packing fraction

$$\eta_\mathrm{c} = \pi/2(1 + 2\delta_\mathrm{c}/L)^2. \tag{2}$$

For finite temperature, the instability theory predicts a square-root singularity in temperature $\eta_\mathrm{c}(T) = \eta_\mathrm{c}(T = 0) + \mathrm{O}(\sqrt{T})$. For the parameters used in the simulation we obtain $\eta_\mathrm{c} = 1.0$ for $T = 0$. The temperature dependence is shown in fig. 4. As expected, the instability theory is an upper bound as compared to the simulation data. But on the relative fine density scale shown, it is fair to say that the theory provides a quantitative description of the location of the jamming phase transition.

In conclusion, we have shown that two-dimensional driven rotators self-organize into a T-structured fluid upon an external torque. The characteristics of this T-structured fluid is that orientations of neighbouring rods are perpendicular. If the rod density exceeds a critical one, there is a jamming transition towards a dynamically blocked state where orientations of neighbouring rods are parallel. These theoretical predictions which are based on simulations and an instability analysis can be verified in experiments. Among the most promising candidates are colloidal rods confined with an optical tweezer close to a plate, a set-up which has been used recently [11]. When circularly polarized light is applied, it should be possible to realize our model and confirm the T-structure directly in real space. Since hydrodynamic interactions are neglected in our model, it is appropriate only for highly charged rod-like suspensions or for rods adjacent to a substrate. However, we think that the general mechanism of the T-structure formation will be stable when hydrodynamic interactions will be added [25]. Finally, the T-structure and the jamming transition can be exploited to tailor the efficiency of fluid mixing in micro- and nanofluidic devices by using an ensemble of driven colloidal rotators. For example, the diffusion of tracer particles is expected to be greatly enhanced in the T-structured phase.

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