Collective Sliding States for Colloidal Molecular Crystals

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We study the driving of colloidal molecular crystals over periodic substrates such as those created with optical traps. The $n$-merization that occurs in the colloidal molecular crystal states produces a remarkably rich variety of distinct dynamical behaviors, including polarization effects within the pinned phase and the formation of both ordered and disordered sliding phases. Using computer simulations, we map the dynamic phase diagrams as a function of substrate strength for dimers and trimers on a triangular substrate, and correlate features on the phase diagram with transport signatures.

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A wide range of condensed matter systems can be modeled as a collection of interacting particles on a periodic substrate under equilibrium and nonequilibrium conditions, where the number of particles may be commensurate or incommensurate with the substrate. Examples of such systems include molecules and atoms on corrugated surfaces, models of sliding interfaces, vortices interacting with artificial pinning arrays in superconductors, and vortices in Bose-Einstein condensates interacting with optical trap arrays. Recently, there has been growing interest in studying colloidal particles interacting with one-dimensional or two-dimensional periodic or quasiperiodic substrates such as those generated with optical traps. Sufficiently strong traps can capture multiple colloids, and at commensurate fillings where the number of colloids is equal to an integer multiple of the number of traps, the colloids in each trap undergo an effective $n$-merization which causes them to act like rigid objects such as dimers or trimers. This produces an orientational degree of freedom and leads to the formation of ordered states which have been termed colloidal molecular crystals. Simulations and experiments have shown that these systems can undergo interestingly ordered to orientationally ordered to disordered transitions as a function of substrate strength. Additional theoretical and numerical studies demonstrated that colloidal molecular crystal systems can exhibit a rich variety of equilibrium states which have ferromagnetic, antiferromagnetic, and other types of spinlike symmetries.

Although individual colloids have been driven over periodic substrates, and colloidal assemblies have been depinned from random substrates, the nonequilibrium dynamics of colloidal molecular crystals in the presence of an additional driving force has not been studied previously. The $n$-merization that occurs in the colloidal molecular crystal system could be expected to produce dynamical sliding behaviors that are distinct from those of sliding point particles. In this work, we show that the pinning force passes through a series of peaks as a function of colloid density that are associated with the formation of commensurate colloidal molecular crystal states. At the commensurate densities, the scaling of the depinning threshold indicates that the pinning is collective, while pronounced changes occur in the depinning threshold as a function of substrate strength that correlate with changes in the symmetry of the colloidal molecular crystal states. Changes in the structure of the colloidal molecular crystals also produce features in the velocity-force curves. We specifically map the dynamical phase diagram for dimer and trimer states at filling fractions of 2 and 3 colloids per trap. In addition to providing an understanding of the general phenomenon of sliding phases on periodic substrates, our results could also be of importance for the development of externally driven dynamical assembly techniques for colloids and other particulate matter systems.

We consider a two-dimensional system with periodic boundary conditions in the $x$ and $y$ directions containing $N_c$ colloidal particles. The dynamics of a single colloid $i$ at position $R_i$ is governed by the overdamped equation of motion

$$\eta \frac{dR_i}{dt} = F_i^c + F_i^s + F_d, \quad (1)$$

where we set $\eta = 1$. The colloid-colloid interaction force is $F_i^c = -\sum_{j \neq i} V(R_{ij})$ where the potential has a Yukawa form, $V(R_{ij}) = (E_0/R_{ij}) \exp(-\kappa R_{ij})$ and where $R_{ij} = |R_i - R_j|$, $E_0 = Z^{*2}/(4\pi\epsilon\epsilon_0 a_0)$, $\epsilon$ is the solvent dielectric constant, $Z^*$ is the effective charge, and 1/$\kappa$ is the screening length. Lengths are measured in units of $a_0$, assumed to be on the order of a micron, forces are measured in units of $F_0 = E_0/a_0$, and time is measured in units of $\tau = \eta/E_0$. The substrate force $F_s$ arises from a triangular substrate with

$$F_s = \sum_{i=1}^{3} A \sin \left( \frac{2\pi b_i}{a_0} \right) \left[ \cos(\theta_i) x - \sin(\theta_i) y \right], \quad (2)$$

where $b_i = x \cos(\theta_i) - y \sin(\theta_i) + a_0/2$, $\theta_1 = \pi/6$, $\theta_2 = \pi/2$, and $\theta_3 = 5\pi/6$. Here $A$ is the relative substrate strength and there are $N_s$ substrate minima. The initial colloidal
FIG. 1: (a) The critical depinning force $F_c$ versus filling fraction $N_c/N_s$ for a substrate with strength $A = 2.5$. Peaks appear at the commensurate fillings of $N_c/N_s = 2$, 3 and 4. (b) $V$, the average colloidal velocity per particle, vs the external drive $F_d$ for $N_c/N_s = 1.94$ (dotted line), 2 (thick line), and 2.13 (thin line). A clear depinning threshold exists which is largest at $N_c/N_s = 2$. (c) The colloidal positions (black dots) and trajectories (black lines) for $N_c/N_s = 2.0$ in the pinned triangular crystal (PC) state at $F_d = 0.0$ and $A = 0.5$. (d) Colloid positions and trajectories for $N_c/N_s = 2.0$ in the moving random (MR) state at $A = 1.5$ and $F_d/F_c = 1.1$.

positions are obtained through simulated annealing. The applied driving force $F_d = F_d\mathbf{x}$ represents the force that would be produced by an external electric field $13$. For each drive, we measure the average colloidal velocity $V = \langle N_c^{-1} \sum_i N_i d\mathbf{r}_i/dt \cdot \mathbf{x} \rangle$. The depinning force $F_c$ is defined as the value of $F_d$ at which $V = 0.025$.

In Fig. 1(a) we plot the depinning threshold $F_c$ versus the filling factor $N_c/N_s$ over the range $1.5 < N_c/N_s < 4.5$ for a system with a substrate of strength $A = 2.5$. At the integer matching fields $N_c/N_s = 2$, 3, and 4, there are clear peaks in the depinning threshold. This is similar to the behavior observed for vortices in superconductors with periodic pinning, where peaks in the critical current (which is proportional to the depinning force) appear when the vortex density is an integer multiple of the pinning site density $14,16$. In Fig. 1(b) we illustrate the velocity force curves for $N_c/N_s = 1.94$, 2.0, and 2.13. A clear single depinning threshold appears at $N_c/N_s = 2$, while two-step depinning transitions occur at the noninteger fillings. For $N_c/N_s = 1.94$, the initial depinning occurs due to the motion of monomer defects in the dimer lattice followed by the depinning of the remaining dimers, while for $N_c/N_s = 2.13$, the trimer defects in the dimer lattice depin first and then the remaining dimers depin at a higher drive.

We measure $F_c$ versus substrate strength $A$ for the case $N_c/N_s = 2.0$ and find that there are three distinct

FIG. 2: The colloidal positions (black dots) and trajectories (black lines) for $N_c/N_s = 2.0$ and (a-d) $A = 3.25$; (e,f) $A = 1.5$. (a) The pinned herringbone (PHB) state at $F_d = 0$. (b) The PHB at finite $F_d/F_c = 0.45$ showing the onset of dimer polarization in the direction of the drive. (c) The fully polarized dimers at $F_d/F_c = 0.9$ form the pinned ferromagnetic (PF) state. (d) The moving ferromagnetic (MF) state at $F_d/F_c = 1.1$ where the motion is strictly one-dimensional. (e) The PMF state for $A = 1.5$ at $F_d/F_c = 1.5$. (f) Vortex positions only in the PMF state from panel (e) showing that every other row of dimers is aligned.

FIG. 3: (a) $V$ vs $F_d$ for $N_c/N_s = 2.0$ and $A = 3.25$ showing the continuous PF-MF depinning transition. Inset: $V$ vs $F_d - F_c$ in the same system indicating a power-law scaling with $\alpha = 0.5$. (b) $V$ vs $F_d$ for $N_c/N_s = 2.0$ and $A = 1.5$. The depinning occurs from a PHB state to a moving random (MR) state, and at higher drives the colloids organize into a partially-ordered moving ferromagnetic (PMF) state. Inset: $dV/dF_d$ vs $F_d$ for the same system. The sharp jump indicates that the depinning transition is discontinuous. The fluctuating region corresponds to the MR state and the onset of a regime with small fluctuations at $F_d > 1$ corresponds to the formation of the PMF state.

pinned states and four moving states. In Fig. 2 we plot the colloidal configurations at different values of $F_d$ for $A = 3.25$. At $F_d = 0$, the ground state is the pinned herringbone (PHB) structure shown in Fig. 2(a). For increasing $F_d$, the dimers become increasingly polarized in the direction of drive, as illustrated in Fig. 2(b) for $F_d/F_c = 0.45$. At sufficiently large $F_d$, the dimers are
completely aligned into the pinned ferromagnetic (PF) state shown in Fig. 2(c) for \( F_d / F_c = 0.9 \). The system depins directly into the moving ferromagnetic (MF) state, where the colloids move in one-dimensional channels as seen in Fig. 2(d). This same sequence of states also appears for higher values of \( A \). The PF-MF transition is continuous, as shown in Fig. 3(a) where we plot \( V \) versus \( F_d \) for \( A = 3.25 \). The inset of Fig. 3(a) indicates that the velocity follows a power law scaling, \( V = (F_d - F_c)^\alpha \) with \( \alpha = 1/2 \), with a turnover at higher \( F_d \) to a linear form. This exponent is consistent with elastic depinning, in which the colloids keep the same neighbors while moving. We observe the same velocity scaling for \( A \geq 2.5 \).

For \( 0.7 < A < 2.5 \), the PF state does not form. Instead, the PHB depins discontinuously into a plastically flowing state where the colloids can exchange neighbors and undergo a transverse diffusive behavior shown in Fig. 1(d). We call this the moving random (MR) state. In Fig. 2(b) we plot the velocity force curve at \( A = 1.5 \) which has a discontinuous depinning transition, as indicated in the inset of Fig. 2(b) where we show \( dV/dF_d \) versus \( F_d \). There is a clear sharp jump in \( V \) at the PHB-MR depinning transition, followed by a regime of fluctuating \( dV/dF_d \) which corresponds to the MR state. For \( F_d > 1.0 \), the fluctuations are diminished when the system forms a partially-ordered moving ferromagnetic (PMF) state, illustrated in Fig. 2(e) and (f). Here the moving colloids form a stripelike structure in which every other row of dimers is aligned with the drive. Unlike the MR state shown in Fig. 1(d), in the PMF state there is no transverse diffusion. For \( A < 0.7 \) and \( F_d = 0 \), the weak substrate causes the elastic energy cost of the PHB state to be too high, and instead the colloids form a pinned triangular crystal (PC) state illustrated in Fig. 1(c). The PC state is only weakly pinned and the depinning occurs as a continuous elastic transition to a moving triangular crystal (MC) state.

In Fig. 4 we summarize the different states in a dynamic phase diagram for \( N_c/N_s = 2.0 \). We plot \( dF_c/dA \) versus \( A \) in the upper inset of Fig. 4 to show how changes in \( F_c \) correlate with different phases of the system. The PHB-PC transition is marked by a discontinuous jump down in \( F_c \) when the PC phase forms, as indicated by the sharp peak in \( dF_c/dA \). A smaller feature in \( dF_c/dA \) occurs at \( A = 2.5 \) when the depinning changes from the discontinuous PHB-PMF transition to the continuous PF-MF transition. The PF phase forms for \( A > 2.5 \) and the PHB-PF transition shifts to lower \( F_d \) with increasing \( A \). Since the PHB state forms due to the effective quadrupole interaction between the dimers \[ 16 \], as \( A \) increases, the colloids forming each dimer are pulled closer together, reducing the quadrupole moment and facilitating the formation of the PF and MF states. The MR state appears in a narrow window between the two elastic depinning transitions, PHB-MC and PHB-PMF. In the MR regime, there is a competition between the quadrupole moment, which prevents the dimers from aligning into the PF state, and the tendency of the external drive to align the dimers. For \( 0.7 < A \leq 1.25 \), the MR state orders into a MC state for increasing \( F_d \) rather than forming the PMF state. For small \( A \), \( F_c \) exhibits a scaling \( F_c \propto A^2 \), consistent with collective pinning in two dimensions \[ 18 \]. A similar scaling occurs for \( A > 0.7 \) as well. In contrast, for a single colloid moving over the periodic substrate, we find the linear behavior \( F_c \propto A \), as shown in the lower inset of Fig. 4. The collective pinning behavior that occurs in the PHB state for large \( A \) likely arises because the objects that are depinning are dimers rather than single particles.

For \( N_c/N_s = 3.0 \), we find a similar set of pinned and moving phases as shown in the dynamic phase diagram for \( F_d \) vs \( A \) in Fig. 5(a). At this filling, there is only a single pinned phase, the pinned trimer crystal (PTC) illustrated in Figs. 5(b) and (c) for \( A = 0.5 \) and \( A = 2.5 \), and so there is no discontinuous transition in \( F_c \) vs \( A \) such as that seen at the PHB-PC transition for \( N_c/N_s = 2.0 \). In Fig. 5(a), the PTC-MC depinning transition for \( A < 1.0 \) is elastic, while for \( A > 1.0 \) the PTC-MR depinning transition is followed at higher drives by the organization of the colloids into either the moving crystal (MC) state or into the moving stripe (MS) state illustrated in Fig. 5(d). In the MS state, the colloids are strictly confined to move along one-dimensional rows with no transverse diffusion. The oriented trimer structure found in the PC is lost in the MS state; however, some orientational ordering of
FIG. 5: (a) The dynamic phase diagram for $F_d$ vs $A$ with $N_c/N_s = 3.0$. Open circles: depinning threshold. PTC: pinned trimer crystal; MC: moving triangular crystal; MR: moving random; MS: moving stripe. (b) Colloid positions in PTC at $A = 0.5$ and $F_d = 0$. (c) Colloid positions in PTC at $A = 2.5$ and $F_d = 0$. The size of each trimer is reduced compared to panel (b). (d) The MS state at $A = 2.5$ and $F_d = 1.5$.

The trimer persists in the MS state, producing a zig-zag structure. The MS state reaches its maximum extent at the transition between the MC and MS phases at $A = 1.6$. The scaling of $F_c$ with $A$ is consistent with collective depinning for low $A$, while for $A > 3.0$, there is a rollover to a more linear regime, indicative of single particle depinning. In the high-$A$ regime, the depinning occurs via the hopping of individual colloids from well to well, followed at higher drives by more general motion of all the colloids.

We expect that a similar type of phase diagram will occur for the higher order molecular crystals. Additional phases are likely to occur at incommensurate fillings. Thermal fluctuations could produce interesting results and new effects since it has been shown that the disordering transition depends on the substrate strength [14, 15, 16].

In summary, we find that a remarkably rich variety of dynamical sliding states can be realized for colloidal molecular crystals under an external drive with varied substrate strength. For dimer colloidal molecular crystals, these states include moving ordered, partially ordered, and moving random phases. The external drive can induce transitions within the pinned phase itself such as a polarization from a pinned herringbone to a pinned ferromagnetic state. The onset of the different phases can be identified through features in the transport response and depinning threshold. We map the dynamical phase diagrams for dimer and trimer states and find that similar features exist for both fillings. Our results should be useful for the general understanding of sliding states for complex particles on periodic substrates and may also have potential applications for creating dynamically induced self-assembled structures.

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