Pattern switching and polarizability for colloids in optical-trap arrays

C. Reichhardt and C. J. Olson Reichhardt

Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

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We show that colloidal molecular crystal states interacting with a periodic substrate, such as an optical-trap array, and a rotating external field can undergo a rapid pattern switching in which the orientation of the crystal changes. In some cases, a martensiticlike symmetry switching occurs. It is also possible to create a polarized state where the colloids in each substrate minimum develop a director field which smoothly rotates with the external drive, similar to liquid-crystal behavior. These results open the possibility for creating different types of devices using photonic band-gap materials, and should be generalizable to a variety of other condensed matter systems with multiple particle trapping.

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There are a wealth of systems with commensurate and incommensurate states which can be modeled as interacting particles on ordered substrates, including vortices in type-II superconductors with periodic pinning assemblies [1], vortices in Bose-Einstein condensates with optical-trap arrays [2], atoms on surfaces [3], electrons on periodic landscapes [4], and cold atoms or molecules in optical traps where Wigner crystal states can occur [5]. Another system of this type that has been attracting growing interest is charge-stabilized colloidal particles in the presence of optical-trap arrays, where a remarkably wide variety of crystalline states [6-13], sliding dynamics [14,15], and nonequilibrium phenomena [16] can be realized. An attractive aspect of the colloidal system is that the dynamics of the individual particles can be observed directly, making colloids on periodic substrates an ideal system for studying different types of behaviors occurring on periodic substrates that could be realized on much smaller scales. Additionally, the ability to create and rapidly control colloidal crystal structures has a wide range of applications in photonic and phononic materials, optical switches [17], photonic band-gap materials [11], and self-assembly of nanostructures.

Crystalline states known as colloidal molecular crystals (CMCs), which have an additional orientational degree of freedom, have been shown to form in the presence of a twodimensional periodic substrate when there are an integer number of colloids per substrate minimum [6-9]. The orientational degree of freedom permits the equilibrium colloidal states to exhibit spin type ordering, including a ferromagnetic state where all the CMCs point in the same direction, an antiferromagnetic state, and herringbone states [8,9]. Here we show that when an in-plane external rotating electric field is applied with a magnitude small enough that the colloids remain confined within the individual traps, the orientational ordering, structure, and direction of the CMCs can be directly controlled. In particular, we find that ferromagnetically interacting CMCs can undergo a switching phenomenon where the orientation rapidly switches between different directions. For certain regimes, the colloids in each substrate minimum polarize and develop a director field which smoothly follows the rotating external drive, similar to a liquid-crystal state. For dimers with herringbone ordering, the rotating drive can induce symmetry changes in the CMC structure, giving rise to a martensitic switching behavior. The ability to create colloidal crystal structures that can rapidly switch among different orientations and structures could have a profound impact in creating different types of devices utilizing photonic band-gap materials such as sensors, optical switches, and filters. Similar rapid structure transitions have already been achieved by applying mechanical stress to macroscopic elastic hole structures [18,19]. Here we show that similar types of structural transitions occur for colloids subjected to an external field rather than to mechanical stress. The general physics of the colloidal system studied in this work may also be realized for other systems of interacting particles on periodic substrates, such as ions or localized electrons on ordered substrates or cold-atom Wigner crystal states [20] in the presence of periodic optical-trap arrays. If similar polarization and switching dynamics can be realized in these systems, a unique class of nanodevices could be explored.

We model an assembly of N_c colloidal particles interacting with a two-dimensional triangular optical-trap array at fillings of either two or three colloids per trap. The system has periodic boundary conditions in the x and y directions and the dynamics of the colloids are obtained by integrating the following overdamped equation of motion:

$$\eta \frac{d\mathbf{R}_i}{dt} = \sum_{j=1}^{N_c} -\nabla V(R_{ij}) + \mathbf{F}_s + \mathbf{F}_{ext}.$$
 (1)

The colloid-colloid interaction is repulsive and has the screened Coulomb form $V(R_{ii}) = (E_0/R_{ii}) \exp(-\kappa R_{ii})$ where $E_o = Z^{*2}/4\pi\epsilon\epsilon_0 a_0$, Z^* is the unit of charge, ϵ is the solvent dielectric constant, η is the damping constant, $R_{ij} = |\mathbf{R}_i - \mathbf{R}_j|$, and $\mathbf{R}_{i(j)}$ is the position of colloid i(j). The system size is measured in units of a_0 and the screening length $1/\kappa$ is fixed at $2a_0$. Force and time are measured in units of $\tau = \eta a_0^2 / E_0$ and $F_0 = E_0 / a_0$. We neglect hydrodynamic interactions since we are working in the low volume, highly charged, electrophoretic limit [21]. The substrate force comes from a triangular substrate of strength F_s with $\mathbf{F}_{s} = \sum_{k=1}^{3} F_{s} \sin(2\pi b_{k}/a_{0}) [\cos(\theta_{k})\hat{x} - \sin(\theta_{k})\hat{y}]$ where b_k $=x\cos(\theta_k)-y\sin(\theta_k)+a_0/2, \quad \theta_1=\pi/6, \quad \theta_2=\pi/2, \text{ and } \theta_3$ =5 $\pi/6$. The initial colloidal positions are found by simulated annealing [6], and we obtain the same ordered ground states observed in previous studies [6,7,9]. After annealing, a



FIG. 1. (Color online) Colloid positions (balls) and trajectories (lines) in a portion of the sample over several ac drive periods for a system with three colloids per substrate minimum. (a) In the locked (L) phase at $F_s=2.5$ and $A_{ac}=0.5$, the trimers remain fixed in the same orientation. (b) In the switching (S) phase at $F_s=4.0$ and $A_{ac}=1.45$, the trimers switch between six different orientations in a single drive period. (c) In the continuously polarized (CP) phase at $F_s=6.5$ and $A_{ac}=2.5$, the trimers rotate continuously with the ac drive. (d) In the partially depinned phase at $F_s=2.5$ and $A_{ac}=0.65$, one of the possible dynamical modes is shown where one third of the colloids depin and switch in a large triangular pattern while the other colloids remain pinned and switch in a smaller star pattern [22].

circular ac drive of amplitude A_{ac} is applied with $\mathbf{F}_{ext} = A_{ac} \sin(2\pi\nu t)\hat{\mathbf{x}} + A_{ac} \cos(2\pi\nu t)\hat{\mathbf{y}}$, where $\nu = 5 \times 10^{-6}$ inverse simulation time steps.

We first study the case of three colloids per potential minimum. The trimers that form in each trap are all aligned in the same direction via an effective ferromagnetic coupling with neighboring trimers [6-8]. Under a rotating external drive, five distinct dynamical responses appear. In Fig. 1(a) we plot the trimer trajectories over several ac drive periods for a system with $F_s=2.5$ and $A_{ac}=0.5$ in the locked (L) phase, where the trimers undergo a small elliptical motion but remain aligned in a fixed orientation [22]. The trajectories at F_s =4.0 and A_{ac} =1.45, shown in Fig. 1(b), are much more complicated and the distorted circular orbits indicate that the trimers change their orientation periodically, leading us to term this the switching (S) phase. Here the triangle formed by the colloids in each trimer is no longer equilateral; instead, one colloid moves further away from the other two, elongating the trimer. The resulting polarized trimer has a director, and in the lowest-energy state this director would remain aligned with the rotating drive; however, the outer colloid of the trimer experiences a corrugated potential generated by the underlying substrate and is unable to rotate smoothly. Instead, the trimer first undergoes a slow continuous displacement and begins to compress, followed by an abrupt switching from one symmetry direction of the triangular substrate to another. Within a single ac period there are six switching events, as shown in Fig. 2(b) where we present



FIG. 2. The instantaneous x component V_x of the colloid velocity. (a) The trimer CP phase in Fig. 1(c) showing a smooth sinusoidal curve. (b) The trimer S phase from Fig. 1(b) showing a sinusoidal curve with six spikes per period that correspond to the switching events. (c) The dimer CP regime for the system in Fig. 4(f) where the dimers continuously rotate. (d) The dimer S phase for the system in Fig. 4(b).

the instantaneous x component V_x of the velocity for the system in Fig. 1(b). The smooth sinusoidal motion is superimposed with six sharp spikes per period. In Fig. 1(c) we show that at F_s =6.5 and A_{ac} =2.5 the trimers are more strongly polarized and the director continuously follows the ac drive, as indicated by the two sets of circular trajectories. In Fig. 2(a) the corresponding V_x has only a smooth sinusoidal motion. The ability of the director of the polarized trimer to couple with and follow an external field has many similarities to liquid crystal systems and opens the possibility of realizing colloidal versions of liquid crystal devices.

Phases with multiple switching dynamics can also appear. For example, Fig. 1(d) illustrates a partially depinned state at $F_s=2.5$ and $A_{ac}=0.65$ in the trimer system where one third of



FIG. 3. (Color online) Phase diagrams as a function of ac amplitude A_{ac} versus substrate strength F_s . Filled circles: locked (L) phase; open triangles: switching (S) phase; hatched squares: continuoulsy polarized (CP) phase; plus signs: partially depinned phase; diamonds: depinned phase. (a) Trimer state. The L phase is illustrated in Fig. 1(a), the S phase in Fig. 1(b), the CP phase in Fig. 1(c), and the partially depinned phase in Fig. 1(d). (b) Dimer state. The L HB phase is illustrated in Fig. 4(a), the S HB phase in Fig. 4(b), and the CP phase in Fig. 4(f). Along the boundary of the S and CP phases we find the martensitic switching regime which is a combination of S and CP.



FIG. 4. (Color online) Colloid positions (balls) and trajectories (lines) in a portion of the sample over several ac drive periods for a system with two colloids per substrate minimum. (a) The locked herringbone state at F_s =3.0 and A_{ac} =1.0. (b) The switching regime at F_s =3.0 and A_{ac} =1.25. (c) A snapshot of the colloid positions in the switching regime at the initiation of a switch; (d) in the middle of the switch; (e) upon completion of the switch. (f) The continuously polarized phase at F_s =3.0 and A_{ac} =2.0.

the colloids depin and move in large triangular orbits while the other colloids remain pinned and form smaller asymmetric star orbits. The velocities V_x (not shown) in this regime have spikes similar to those in Fig. 2(b). For $A_{ac} > F_s$, the system enters a different regime where the colloids continuously depin and undergo sliding dynamics, as described in Ref. [15].

Figure 3(a) shows the phase diagram of the different regimes for A_{ac} vs F_s . For weak F_s , the S phase appears only in a small window. The continuously polarized (CP) phase occurs for $F_s > 5.5$, and the width of this phase increases with increasing F_s since the trimers are increasingly compressed by the stronger trapping potential. Near the center of the trap, the potential appears parabolic, so the substrate symmetry that determines the orientation of the trimers become less important and the discrete switching of the S phase cannot occur for the compressed trimers. A partially depinned phase such as that shown in Fig. 1(d), in which only a portion of the colloids are depinned, occurs in a narrow region below the sliding or depinned phase.

We next consider the case of two colloids, which form dimers, per substrate minimum. The ground state has a herringbone (HB) structure, as in Fig. 4(e); there are three pos-

sible orientations. In the L phase, illustrated at F_s =3.0 and $A_{ac} = 1.0$ in Fig. 4(a), the colloids undergo small rotations but the orientation of the HB state remains fixed. The S phase is more complicated for the dimer HB phase than for the trimers, and the complex orbits of the dimers are illustrated in Fig. 4(b) for F_s =3.0 and A_{ac} =1.25. Each switch occurs in three stages. First, there is an abrupt buckling of every other HB row such that the dimers in the buckled row are alternately oriented perpendicular and parallel to the HB axis. This is illustrated in Fig. 4(c); the HB orientation prior to the switch was horizontal as in Fig. 4(a). After the sudden buckling, the dimers in the buckled rows begin to tilt gradually, and at the same time the dimers in the unbuckled rows also begin to tilt, as shown in Fig. 4(d). Finally, there is a second sudden switch when the dimers all snap into the orientation seen in Fig. 4(e). This same pattern of switching occurs for all three HB orientations. The sharp switches appear as spikes in V_x in Fig. 2(d).

For higher A_{ac} , the dimers lose the HB orientation and instead form the fully polarized or ferromagnetic (FM) state illustrated in Fig. 4(f) for F_s =3.0 and A_{ac} =2.0. In this CP regime, the dimers rotate continuously with the external drive, as indicated by the sinusoidal velocity response in Fig. 2(c). Between the S and CP regimes, the system exhibits a combination of the S and CP phases where the switching HB configuration can change into the FM configuration and rotate continuously for a period of time before jumping back to the HB switching state again. This behavior can be viewed as martensitic switching between two different crystalline symmetries.

In Fig. 3(b), the phase diagram for the dimer system as a function of F_s and A_{ac} indicates that the S and CP phases appear only for $F_s > 2.25$. The CP phase grows rapidly in extent with increasing F_s due to the reduction of the dimerdimer interactions as the dimers are compressed by the substrate, which destroys the HB ordering. A CMC with nonferromagnetic-type ordering, such as the dimer HB phase, arises only due to an effective multipole interaction between *n*-mers [9]. As F_s increases and the *n*-mers compress, the pole moment is reduced. We expect that similar types of dynamical behaviors will arise for colloids on square substrates where similar spin type orderings are expected to occur [6,9].

In summary, we have shown that colloidal molecular crystals on periodic substrates exhibit a remarkable pattern of switching and polarization effects in the presence of a circular ac external drive. We specifically examine a triangular substrate with three and two colloids per trap which form ferromagnetic trimer or herringbone dimer ground states. The trimers show a switching regime in which the partially polarized ferromagnetically ordered trimers rapidly change their global orientation. The dimers exhibit structural switching into different states as well as a martensitic switching between a herringbone and a ferromagnetically ordered state. For strong substrates, both the dimers and the trimers enter a strongly or fully polarized state where the *n*-mers each develop a director that couples to the external drive. This results in a continuous rotation of the *n*-mers with the external drive which has similarities to liquid-crystal dynamics. The ability to create colloidal molecular crystalline structures that can undergo rapid pattern switching or that have liquidcrystal behaviors has exciting implications for creating a different class of photonic or phononic drives, switches, displays, and dynamical materials. Additionally, dynamical behaviors observed for the colloidal system may be scaled down to systems such as Wigner crystals on periodic substrates or crystalline cold atoms or molecules on optical-trap

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arrays. Realizing switching dynamics in these systems also has the potential to provide a basis for the development of new types of nanodevices.

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