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Stochastic ratcheting of two-dimensional colloids: Directed current and dynamical transitions

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We present results of molecular dynamics simulations for two-dimensional repulsively interacting colloids driven by a one-dimensional asymmetric and commensurate ratchet potential, switching on and off stochastically. This drives a time-averaged directed current of colloids, exhibiting resonance with change in ratcheting frequency, where the resonance frequency itself depends nonmonotonically on density. Using scaling arguments, we obtain analytic results that show good agreement with numerical simulations. With increasing ratcheting frequency, we find nonequilibrium reentrant transitions between solid and modulated liquid phases.

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Introduction. A flashing ratchet refers to a time-averaged directed motion of Brownian particles under the influence of a spatially periodic and asymmetric potential, with the potential height varying with time, either deterministically or stochastically [1–4]. Stochastic ratcheting has been studied extensively in the context of the active dynamics of molecular motors [5–8] and the dynamics of colloidal dispersion in electrical [9–11], magnetic [12,13], or optical drive [14,15]; as a mechanism of particle segregation [16–18] or transport of cold atoms in optical lattice [19]; and in the motion of flux quanta [20,21]. While a large body of work has concentrated on the ratcheting of individual particles, fewer studies have focused on the effects of interaction [22–26]. Recent studies of two-dimensional (2D) paramagnetic particles under 1D magnetic ratchets observed a relation between the overall dynamics and local particle coordination numbers [13].

In colloidal suspensions, the ratchetlike directed motion of particles has been achieved using suitable laser potentials [14,15]. Confinement and laser trapping in colloids, on the other hand, is known to give rise to interesting mechanical properties and phase transitions [27–32]. Coupling 2D interacting colloids to a 1D time-independent spatially periodic potential with periodicity commensurate with the mean particle separation leads to the phenomena of laser-induced freezing (LIF) and reentrant melting with an increase in the potential strength. This was demonstrated in experiments using the standing wave pattern of interfering laser beams [31,32] and was understood in terms of a dislocation unbinding theory [33,34].

We consider transport of a 2D system of particles interacting via soft-core repulsion and driven by a 1D asymmetric flashing ratchet, using molecular dynamics (MD) simulations in the presence of a Langevin heat bath. The ratcheting potential breaks time-reversal symmetry and generates an averaged directed current along the direction of ratcheting [Fig. 1(a)]. We choose a periodicity of the potential commensurate with the interparticle separation. At switching frequencies much faster than the intrinsic relaxation times, the time scale required for particles to relax over a single valley of the external ratchet potential, the system experiences a time-averaged effective periodic potential, which in the limit of weak asymmetry is expected to lead to a situation similar to that of LIF. However, at intermediate switching frequencies the system is driven

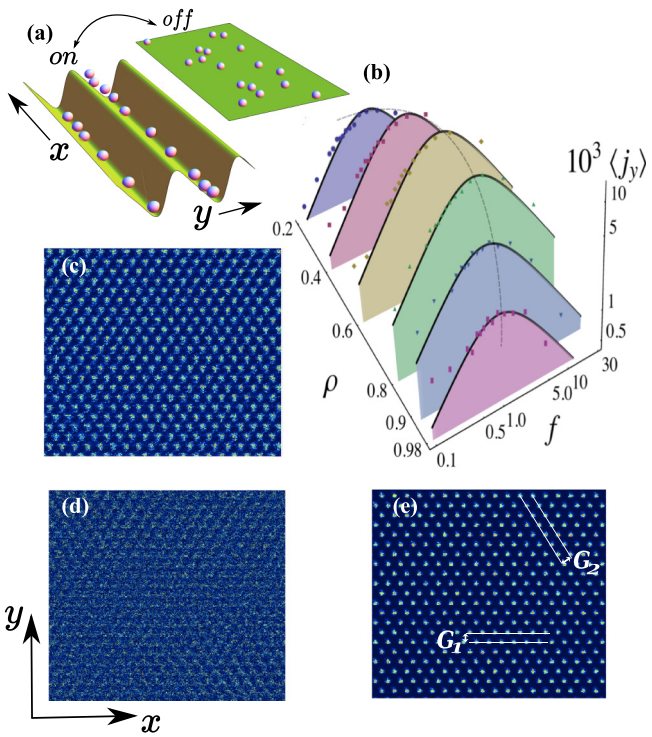

FIG. 1. (Color online) (a) Schematic of 2D colloids in 1D asymmetric ratchet potential periodic in the y direction and constant in the x direction. The arrow between the (green) corrugated and flat surfaces denotes switching of the external potential between the on and off states with rate f. (b) Time-averaged directed current along the y direction $\langle j_y\rangle$ as a function of frequency f and density ρ. The dashed line indicates variation of the maximal current with density. Superimposed positions of 10^3 uncorrelated configurations, from center-of-mass coordinates, are shown at a density of $\rho = 1.0$ with ratcheting frequencies (c) $f = 0.11$, (d) $f = 1.67$, and (e) $f = 10$. The color code denotes the local density of points from red (light) (high) to blue (dark) (low). The reciprocal lattice vectors $\mathbf{G}_{1,2}$ and the corresponding lattice planes are indicated in (e).

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out of equilibrium and carries an averaged directed current. We present the transport properties and the relation between structure and transport in this system.

In the 2D ratchet system that we study, the averaged directed current shows nonmonotonic variation with density and ratcheting frequency, with the maximal current achieved at their intermediate values. The behavior differs significantly in the detailed functional dependence from the 1D ratchet. Using scaling arguments, we derive expressions for the directed current that fully capture the simulation results. Our study of 2D ratchet reveals two fascinating properties that are unlike the 1D ratchet: (i) With increasing ratcheting frequency we find reentrant nonequilibrium phase transitions between solid and modulated liquid phases, as the averaged directed current shows nonmonotonic variation, and (ii) crossover from ballistic to diffusive transport with density, captured by a nonmonotonic density dependence of resonance frequency. Our predictions are amenable to verification in experiments on, e.g., sterically stabilized colloids driven by suitably tunable optical or magnetic ratchets [13,14].

Model. As a model colloid, we consider a system of purely repulsive particles interacting via a shifted and truncated soft-core potential $\beta U(r) = (\sigma/r)^{12} - 2^{-12}$ with a cutoff distance $r_c = 2\sigma$, so that $\beta U(r) = 0$ for $r > r_c$. Here $k_B T = 1/\beta$ and σ set the energy and length scales, respectively. The asymmetric ratchet potential $U_{\rm ext}(y,t) = V_0(t)[\sin(2\pi y/\lambda) + \alpha \sin(4\pi y/\lambda)]$, where $V_0(t)$ switches between U_0 and 0 with a switching rate f, which we also refer to as frequency. We use the asymmetry parameter $\alpha = 0.2$ [see Fig. 1(a)]. In all our simulations we set $\beta U_0 = 1$. The external potential is kept commensurate with the density of the particles, such that $\lambda = a_y$, with the separation between consecutive lattice planes $a_y = \sqrt{3}a/2$ in a triangular lattice at a density $\rho = 2/\sqrt{3}a^2$. Molecular dynamics simulations are performed using the standard leapfrog algorithm [35] with a time step $\delta t = 0.001\tau$, where $\tau = \sigma\sqrt{m/k_B T}$ is the characteristic time scale. We choose the mass of the particles $m = 1$ and set the temperature $T = 1.0\epsilon/k_B$ by using a Langevin thermostat [36] with an isotropic friction $\gamma = 1/\tau$. At each time step, a trial move to perform switching of the external potential strength between 0 and U_0 is performed and accepted with probability $f\delta t$. We used $N = 4096$ particles in our simulations.

The soft-core particles, in the absence of an external potential, freeze at a density $\rho^* \approx 1.01$ [see Fig. 1(a) in the Supplemental Material [37]]. The limit of $\alpha = 0$ and $V_0(t) = U_0$ corresponds to the equilibrium situation of laser-induced freezing [33]. At a density close to the liquid-solid transition, the system freezes into a triangular lattice solid (LIF) that remelts into a density modulated liquid with increasing U_0 [32,33]. In soft-core particles, the LIF with $\beta U_0 = 1$ occurs at $\rho = 0.95$ [34]. A similar freezing transition at this density is observed for a weakly asymmetric ratchet ($\alpha = 0.2$) of strength $\beta U_0 = 1$ in the limit of high switching frequency, much faster than the typical relaxation time, such that the colloids experience an effective periodic potential [see Fig. 2(d) in the Supplemental Material [37]]. In the other limit of extremely slow switching, the system comes to quasiequilibrium with the instantaneous strength of the external potential and one obtains a slow variation between a modulated liquid and a solid phase. The most interesting dynamics takes place at intermediate frequencies. The ratchet-driven averaged directed current shows resonance with frequency and nonmonotonic variation with density [Fig. 1(b)]. At suitable densities, the system shows a dynamical reentrant transition from a soft solid to modulated liquid to a solid with an increase in ratcheting frequency [Figs. 1(c)–1(e) and 5].

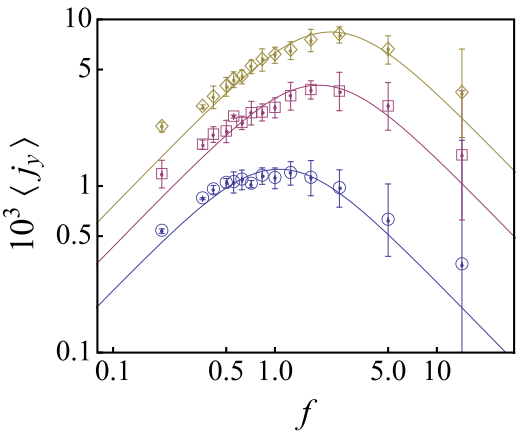

FIG. 2. (Color online) Average directed current as a function of switching rate f at particle densities $\rho = 0.1$ (○), 0.5 (□), and 1.0 (◇). The solid lines show the fit to Eq. (2).

Transport properties. The steady-state dynamics is characterized in terms of a space- and time-averaged directed current of particles flowing along the direction of ratcheting

$$\langle j_y\rangle = \frac{1}{\tau_m}\frac{1}{L_x L_y}\int^{\tau_m} dt \int^{L_x} dx \int^{L_y} dy\, j_y(x,y,t), \qquad (1)$$

where the time averaging is done over $\tau_m = nt_p$, with $t_p = 1/f$ and n denoting a large number of switchings, chosen to be 200 in all our simulations.

For small switching frequencies $f \ll \nu$, the inverse of the intrinsic relaxation time, the system is close to thermodynamic equilibrium. The directed current increases as $\langle j_y\rangle \sim f$ starting from zero at $f = 0$ in agreement with linear response [5,38,39]. The frequency dependence at a high switching rate was calculated earlier using an asymptotic expansion to give $\langle j_y\rangle \sim 1/f$ [39,40].

In our MD simulations of the 2D system of soft disks, we observe the same behavior, viz., $\langle j_y\rangle \sim f$ at low frequency and $\langle j_y\rangle \sim 1/f$ at very high ratcheting frequencies (Fig. 2). The asymptotic behavior may be captured by the interpolation formula $g(\nu,f) = \nu f/(\nu^2 + f^2)$. We use a simple ansatz $\langle j_y\rangle = \kappa g(\nu,f)\rho v_0$, where κ is a dimensionless proportionality constant and ρv_0 has the dimension of current with v_0 an intrinsic velocity. As we show below, the form of v_0 and ν allows us to describe the whole density and frequency dependence of the directed current. The relation

$$\langle j_y\rangle = \kappa \frac{\nu f}{\nu^2 + f^2}\rho v_0 \qquad (2)$$

shows good agreement with simulation results (Fig. 2). The above frequency dependence is obeyed even if the ratcheting wavelength λ is incommensurate with density [see Figs. 2(a)–2(c) in the Supplemental Material [37]]. A similar frequency

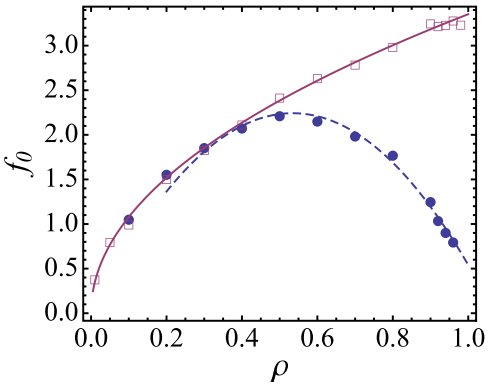

FIG. 3. (Color online) Resonance frequency f_0 as a function of the density ρ for noninteracting particles (squares) and soft-core particles (circles). The solid line shows the ballistic form $f_0 \sim \sqrt{\rho}$, while the dashed line shows the diffusive form $f_0 \sim \rho(1-\rho/\rho_c)$ with $\rho_c = 1.07$.

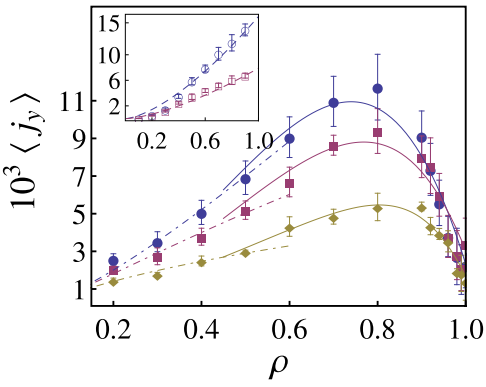

FIG. 4. (Color online) Average particle flux $\langle j_y \rangle$ as a function of density ρ for soft-core particles at ratcheting frequencies $f = 1.43$ (●), 0.71 (■), and 0.36 (◆).The dot-dashed lines are fit to Eq. (3) in the regime $\rho < 0.5$ with fitting parameter $\kappa = 0.04, 0.03, 0.02$ for the three data sets, respectively. The solid lines are fit to Eq. (4) in the regime $\rho \geqslant 0.5$ with fitting parameters $\kappa = 0.12, 0.05, 0.02$ and $\rho_c = 1.03, 1.05, 1.03$. The inset shows the same quantity for free particles at two different ratcheting frequencies $f = 0.71$ (○) and 0.36 (□). The dashed lines are fit to Eq. (3).

dependence was recently found for a stochastic pump model of the one-dimensional system of interacting particles [41]. Fitting the MD simulation data of Fig. 2 to Eq. (2), we find the resonance frequencies $f = f_0 = \nu$, which show a nonmonotonic variation with the mean density of colloids ρ (Fig. 3).

The intrinsic relaxation frequency ν, controlling the behavior of time-averaged dynamics, may arise from a ballistic or diffusive relaxation of the particles over the characteristic length scale λ. We use a ratcheting potential commensurate with the density such that $\lambda^2 \sim 1/\rho$ (for treatment using an incommensurate potential see the Supplemental Material [37]). For underdamped motion, the ballistic time scale τ_b for a particle to traverse the potential valley is obtained from the kinematic relation $\lambda \sim (U_0/\lambda)\tau_b^2$, which leads to $\tau_b \sim (\rho U_0)^{-1/2}$. On the other hand, the relaxation time in the overdamped diffusive regime is given by $\tau_D = \lambda^2/D \sim (D\rho)^{-1}$. The self-diffusion constant D decreases with density for two-dimensional repulsively interacting particles as $D = D_0(1-\rho/\rho_c)$ [42,43] [see Fig. 1(b) in the Supplemental Material [37]].

In the underdamped case, the velocity scale is set by $v_0^b = \lambda/\tau_b = U_0^{1/2}$. Using this and $\nu = 1/\tau_b$ in this regime, one finds

$$\langle j_y \rangle \simeq \kappa \frac{f U_0}{\rho U_0 + f^2} \rho^{3/2}. \tag{3}$$

The resonance frequency is then $f_0 = (\rho U_0)^{1/2}$. On the other hand, the velocity scale in the overdamped regime may be obtained using the time scale for free diffusion $1/\rho D_0$ over the mean interparticle separation λ, $v_0^D = D_0 \rho^{1/2}$. Thus, using $\nu = 1/\tau_D$, the averaged directed current becomes

$$\langle j_y \rangle \simeq \kappa \frac{f D_0^2}{D_0^2 \rho^2 (1-\rho/\rho_c)^2 + f^2} \rho^{5/2}(1-\rho/\rho_c). \tag{4}$$

The corresponding resonance frequency is $f_0 = D_0\rho(1-\rho/\rho_c)$.

Our simulations show that the resonance frequency, and therefore the intrinsic relaxation frequency, follows ballistic behavior $f_0 \sim \sqrt{\rho}$ at low densities (and for noninteracting particles) and diffusive behavior $f_0 \sim \rho(1-\rho/\rho_c)$ at high densities (Fig. 3). The dynamical behavior changes from ballistic to diffusive with an increase in density. This may be understood in terms of what happens to a directed current in the presence of direction randomizing scattering events. At low densities, the time- and space-averaged motion of a test particle with a small number of scattering events remains ballistic on average. However, at large densities the mean free path is reduced and consequently a large number of scattering events randomizes the direction of motion leading to a predominantly diffusive dynamics.

In Fig. 4 we show the density dependence of the directed current at various switching frequencies. The plots show nonmonotonic variation, the low-density limit of which is fully captured by Eq. (3) and the high-density limit by Eq. (4). Near the density ρ_c, the system gets into a *jammed* state where the directed current vanishes as $\langle j_y \rangle \sim \rho^{5/2}(1-\rho/\rho_c)$. Note that the overall density dependence that we find in the 2D ratchet is quite unlike the $\langle j \rangle \sim \rho(1-\rho)$ behavior of the directed current found in the repulsively interacting 1D ratchet [26]. The collective dynamics of the 2D ratchet can be further characterized in terms of the density and ratcheting frequency dependence of longitudinal and transverse diffusivities $D_{x,y}(\rho, f)$ (see Figs. 3 and 4 in the Supplemental Material [37]).

Dynamical transitions. The reduction of directed current at high densities and subsequent jamming is associated with freezing of the system into a triangular lattice solid. Our MD simulations show that similar structural transitions are also associated with a change in current as a function of ratcheting frequency [Fig. 1(b)], a fully dynamical effect. In Figs. 1(c)–1(e) we plot the superimposed positions of 10^3 uncorrelated configurations from the center-of-mass frame for a system at a mean density $\rho = 1.0$ and ratcheting frequencies $f = 0.11, 1.67, 10$. This suggests a frequency-dependent reentrant transition from

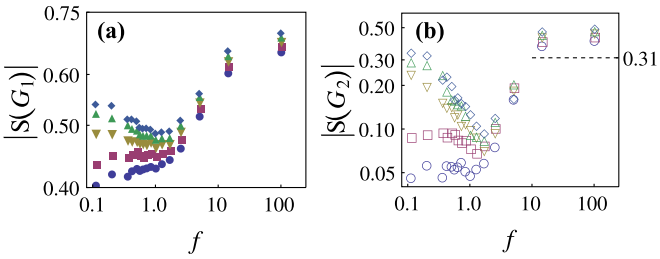

FIG. 5. (Color online) Amplitude of the steady-state structure factor for the reciprocal lattice vectors (a) $\mathbf{G}_1$ and (b) $\mathbf{G}_2$ as a function of ratcheting frequency f for densities $\rho = 0.98$ (●, ○), 0.99 (■, □), 1.00 (▼, ▽), 1.01 (▲, △), and 1.02 (◆, ◇).

a triangular lattice solid ($f = 0.11$) to a density modulated liquid ($f = 1.67$) to again a triangular lattice solid ($f = 10$) order. Note from Fig. 2 that the modulated liquid at $\rho = 1.0$ and $f = 1.67$ corresponds to the resonance frequency in directed current.

The interplay of structure and dynamics is further quantified with the help of the time-averaged steady-state structure factor $S(\mathbf{G}) = \langle \frac{1}{N^2} \sum_{i,j} \exp[-\mathbf{G} \cdot (\mathbf{r}_i - \mathbf{r}_j)] \rangle$ with reciprocal lattice vectors $\mathbf{G}_1 = (0, \pm 2\pi/a_y)$ and $\mathbf{G}_2 = (\pm 2\pi/a, \pm 2\pi/\sqrt{3}a)$ [see Fig. 1(e)]. In Fig. 5 we show the frequency dependence of $|S(\mathbf{G}_{1,2})|$ at various densities. The presence of a ratcheting potential keeps $|S(\mathbf{G}_1)| > |S(\mathbf{G}_2)|$ corresponding to stronger density modulation in the y direction. The nonmonotonic variation of $|S(\mathbf{G}_1)|$ with frequency quantifies a reduction followed by an increase in this density modulation. At very high frequencies, the solid order parameter $|S(\mathbf{G}_2)| > 0.31$ for densities $\rho \gtrsim 0.96$, signifying freezing into a triangular lattice structure [see Figs. 1(a) and 2(d) in the Supplemental Material [37]], reminiscent of the LIF transition [34]. The solid order parameter $|S(\mathbf{G}_2)|$ at densities $\rho \geqslant 1$ shows significant nonmonotonic variation with frequency, pointing to a dynamical reentrant transition from a solid to a modulated liquid to a solid phase. Thus the ratcheting frequency provides a means to structural control during transport and may be utilized in experiments.

Summary and outlook. Our study of a 2D system of soft-core particles under 1D ratchet drive has shown an interesting relation between transport properties and structural phases. Using scaling arguments, we obtained the density and ratcheting frequency dependence of the averaged directed current $\langle j_y \rangle$, which fully captured the simulation results. The resonance frequency of $\langle j_y \rangle$ showed a curious crossover from ballistic to diffusive behavior with increasing density, related to a reduction of the mean free path. Within a range of densities, we found a dynamical reentrant transition from the solid to the modulated liquid to the solid phase with increasing ratcheting frequency. The fact that the ratcheting frequency provides control over both the emergent directed current and structural phases may have useful applications.

Our predictions may be verified in experiments on repulsively interacting colloids confined within glass plates, e.g., using magnetic ratcheting [13] or optical ratcheting [14] in a suitably modified 2D laser trapping setup [32]. For example, polysterene beads have a density of 1.05 g/cm^3, i.e., a bead of diameter of $\sigma \approx 5$ μm has mass $m \approx 6.9 \times 10^{-14}$ Kg. Given $k_B T = 4.2 \times 10^{-21}$ Nm at room temperature, the unit of time $\tau = \sigma\sqrt{m/k_B T} \approx 0.02$ s. Thus the dimensionless frequency range of $f = 0.1$ to 100 studied here corresponds to a range of 5 Hz to 5 KHz and the resonance at $f_0 \approx 1$ means a frequency of 50 Hz.

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