Colloids in a periodic potential: Driven lattice gas in continuous space

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Motivated by recent studies of colloidal particles in optical-tweezer arrays, we study a two-dimensional model of a colloidal suspension in a periodic potential. The particles tend to stay near potential minima, approximating a lattice gas. The interparticle interaction, a sum of Yukawa terms, features short-range repulsion and attraction at somewhat larger separations, such that two particles cannot occupy the same potential well, but occupation of adjacent cells is energetically favored. Monte Carlo simulation reveals that the equilibrium system exhibits condensation, as in the Ising model or lattice gas with conserved magnetization; the transition appears to be continuous at one-half occupancy. We study the effect of biased hopping, favoring motion along one lattice direction, as might be generated by a steady flow relative to the potential array. This system is found to exhibit features of the driven lattice gas: the interface is oriented along the drive, and appears to be smooth. A weak drive facilitates ordering of the particles into high- and low-density regions, while stronger bias tends to destroy order, and leads to very large energy fluctuations. We also study ordering in a moving periodic potential. Our results suggest possible realizations of equilibrium and driven lattice gases in a colloidal suspension subject to an optical tweezer array.

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I. INTRODUCTION

Lattice gas models are an essential tool of statistical mechanics. In addition to their role as minimal models of fluids, they are important in the study of phase transitions in adlayers [1–3]. The equilibrium properties of this class of models are well understood [4]. Lattice gases are readily extended to the study of time-dependent phenomena, by defining a stochastic particle dynamics (typically via hopping) that obeys detailed balance with respect to the Hamiltonian. In driven lattice gas models, also known as *driven diffusive systems* (DDSs) [5–7], hopping is biased to favor motion along one of the principal lattice directions, with periodic boundaries along this direction. Since DDS dynamics violates detailed balance, it represents an out-of-equilibrium situation. The driven system exhibits a nonequilibrium stationary state with a nonzero current, and anisotropic ordering [8,9]. DDSs have been studied extensively as prototypes of nonequilibrium phase transitions. Despite their simplicity, these systems exhibit surprising characteristics [6], for example, the *increase* of the critical temperature with drive strength in the case of attractive interactions. A driven fluid in continuous space was also found to possess an unusual phase diagram [10]. In this case, the drive appears to oppose ordering, as reflected in a reduced critical temperature, compared with equilibrium.

Although much effort has been devoted to studying DDSs theoretically and in simulations, there are as yet no experimental realizations. Colloidal suspensions offer some promise in this regard. With the availability of optical-tweezer arrays, the dynamics of colloidal particles in an external potential has been investigated intensively [11–15]. A study of colloidal particles in a periodic potential revealed a variety of dynamic regimes as a function of the viscous friction coefficient [11]. At low friction, the motion at long times consists of jumps between adjacent potential minima, resembling that of particles in a lattice gas with nearest-neighbor hopping.

Motivated by this correspondence, we study a two-dimensional model system in continuous space, of colloid-like particles in a periodic background potential. The potential is taken so that the particles spend most of the time near a potential minimum. The interparticle potential, a sum of Yukawa terms, features short-range repulsion and attraction at somewhat larger separations, such that two particles cannot occupy the same potential well, but occupation of adjacent cells is energetically favored. We study the phase behavior in Monte Carlo simulations, beginning with the undriven (equilibrium) case, followed by an exploration of the effects of driving, and a brief examination of another nonequilibrium version, in which there is no drive, but the external potential is time dependent.

Although the present model includes, in the interests of computational efficiency, certain unrealistic features, we believe that it represents a significant step toward devising a system capable of experimental realization, and exhibiting properties characteristic of DDSs. Our results suggest that it is possible to realize lattice-gas-like systems, both equilibrium and driven, in a colloidal suspension subject to a periodic external potential.

In the following section we define the model and simulation method. In Sec. III we present simulation results, while Sec. IV contains a summary and discussion of open issues, regarding both the model system and possible realizations of DDSs in experiments on colloids.

II. MODEL AND SIMULATIONS

We study a two-dimensional system of N particles interacting via a pairwise potential $u(r)$, and subject to a periodic external potential $V(x,y)$. The latter is of the form used to model an optical-tweezer array [15],

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$$V(x,y) = \frac{V_0}{1 + e^{-g(x,y)}}, \tag{1}$$

where $g(x,y)=A[\cos(2\pi x)+\cos(2\pi y)-2B]$. For $V_0>0$ the potential minima fall at the sites of the integer square lattice $(n+1/2,m+1/2)$; the parameters A and B together control the well depth and the curvature near the potential minimum. The interparticle potential is given by the sum of a short-range repulsive term and a slightly longer-range attractive term, both of Yukawa form [16]:

$$u(r) = V_1 \frac{e^{-\kappa_1 r}}{r} - V_2 \frac{e^{-\kappa_2 r}}{r} - V_c \tag{2}$$

for $r<r_c$, and $u=0$ for $r>r_c$. Here $\kappa_{1,2}$ are characteristic lengths and V_c is a constant taken so that $u(r_c)=0$. The parameter values used in this study ($\kappa_1=3.30$, $\kappa_2=2.21$, $V_1=200$, $V_2=90$, $V_0=40$, $A=5$, $B=-0.5$, and $r_c=2.5$), lead to a strong short-range repulsion that effectively prohibits two particles from occupying the same potential well at the temperatures of interest. The minimum of the interparticle potential falls at $r=1$, favoring occupation of neighboring cells. (For $r=1$, $\sqrt{2}$ and 2, one has $u=-2.347,-1.35$, and -0.283, respectively.) The ground-state energy per particle is -8.396, about 4.2 times that of the nearest-neighbor square lattice gas with unit interaction. A crude estimate of the critical temperature is then 4.2 times that of the nearest-neighbor lattice gas ($T_c \simeq 0.5673$), that is, $T_c \simeq 2.4$.

We perform Metropolis Monte Carlo (MC) simulations of the system defined above. The MC time step is defined as one attempted move per particle. In each move, a randomly chosen particle is subject to a random displacement $\Delta\mathbf{r}=(\Delta x,\Delta y)$, with components distributed uniformly on the interval $[-1,1]$.

As in studies of DDSs, the drive takes the form of a force $\mathbf{f}$, such that the work done on a particle when it suffers a displacement $\Delta\mathbf{r}$ is $\mathbf{f}\cdot\Delta\mathbf{r}$. As noted, the system is periodic in the direction of the drive (in practice, the x direction), so that $\mathbf{f}$ cannot be written as the gradient of a potential. The acceptance probability for a particle displacement $\Delta\mathbf{r}$ is

$$P(\Delta\mathbf{r}) = \min\{1,\exp[-\beta(\Delta E - \mathbf{f}\cdot\Delta\mathbf{r})]\}, \tag{3}$$

where ΔE is the change in energy and β represents inverse temperature, in units such that $k_B=1$.

The quantities of principal interest are the average energy $\langle E\rangle$, the order parameter $\mathbf{m}=(m_x,m_y)$, and the corresponding fluctuations. To define the order parameter we introduce lattice gas variables: $\sigma_{ij}=1$ if the cell centered at $(i+1/2,j+1/2)$ is occupied, and zero otherwise. Then an appropriate order parameter for a lattice gas with conserved particle density can be defined, for a system of $L\times L$ sites, via [7]

$$m_x = \frac{1}{L^3}\sum_{j=1}^{L}\left(\sum_{i=1}^{L}(1-2\sigma_{ij})\right)^2, \tag{4}$$

with m_y given by exchanging indices i and j in the sums. In a disordered phase $\langle m_x\rangle=\langle m_y\rangle=0$. In a half-occupied system, a maximally ordered, isotropic configuration (a square of side $L/\sqrt{2}$) has $m_x=m_y=1/2$. In a system with periodic boundaries, however, the surface energy is smaller in a strip configuration. If the particles occupy all sites the region $j_0 \le j \le j_0+L/2$, then $m_x=1$ and $m_y=0$; the values are exchanged for a strip oriented along the y direction. $m=\sqrt{m_x^2+m_y^2}$ characterizes overall ordering. In equilibrium, the two orientations are equally likely, but for low temperatures and reasonably large systems, the typical time for switching between them is much larger than practical simulation times, so it is interesting to characterize the degree of order by the anisotropy parameter $\Delta m \equiv m_> - m_<$, where $m_> \equiv \max\{\langle m_x\rangle,\langle m_y\rangle\}$, and $m_<$ denotes the lesser of the two components. Experience with DDSs shows that in driven systems $m_>$ is always along the drive direction.

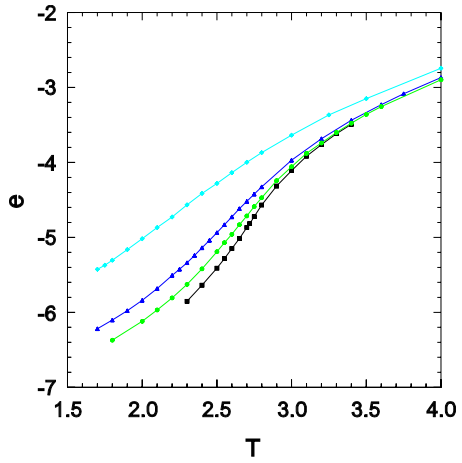

FIG. 1. (Color online) Energy per particle versus temperature for $L=10$, 20, 30, and 50 (upper to lower). Error bars are smaller than the symbols.

III. SIMULATION RESULTS

A. Equilibrium properties

We study half-occupied systems, $N=L^2/2$, in periodic cells of linear size $L=10$, 20, 30, and 50. Our results represent averages over the stationary regime of ten or more independent realizations, each of 10^6 or more time steps. Figure 1 shows that the mean energy e per particle exhibits the qualitative behavior typical of lattice gases or fluids with a hard core and short-range attraction. Evidence of a phase transition is seen in the progressive growth, with system size, of the peak in the specific heat per particle, $c=\mathrm{var}(E)/NT^2$, shown in Fig. 2. The specific heat maxima fall at temperatures $T_m=2.10(2)$, 2.50(2), 2.63(1), and 2.66(1) for sizes $L=10$, 20, 30, and 50, respectively. (Figures in parentheses denote statistical uncertainties.) A quadratic fit of T_m versus $1/L$ yields the estimate $T_c=2.74(4)$ for the critical temperature in the infinite-size limit. We also note that the maximum specific heat values, $c_m(L)$, grow approximately linearly with $\ln L$, compatible with a critical point in the Ising model universality class.

Figure 3 shows the order parameter m for $L=30$ and 50. The results are consistent with a transition near $T=2.8$ or so. Of note is the much smaller finite-size effect in Δm as com-

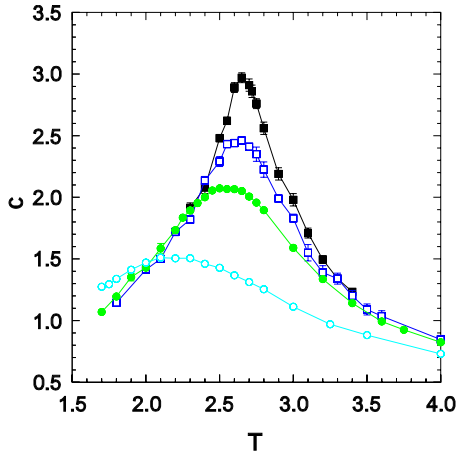

FIG. 2. (Color online) Specific heat per particle versus temperature for $L=10$, 20, 30, and 50, in ascending order.

pared with m in the high-temperature regime, as the fluctuations in the two components of **m** are nearly equal. The inset shows that $\chi \equiv (L^2/T)\mathrm{var}(m)$ exhibits a well-defined maximum in the vicinity of the transition.

Based on the present results, we can conclude that the half-filled system exhibits an (apparently continuous) order-disorder transition near a temperature of about 2.75. Further details on the nature of the transition (which from symmetry considerations should belong to the Ising model universality class), must await more extensive studies using larger systems.

To close this section we show a representative configuration (Fig. 4) for $L=50$ at temperature $T=2$. Here the system has separated into distinct high- and low-density phases. The latter is quite dilute, while the former clearly reflects the periodic potential, and possesses a low density of vacancies

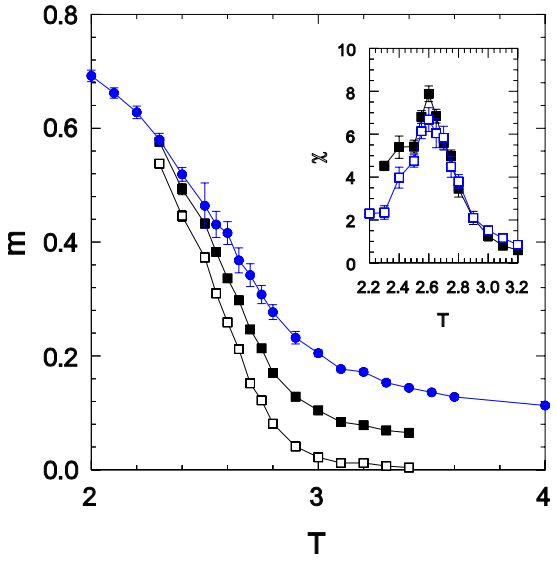

FIG. 3. (Color online) Order parameter m (filled symbols: upper, $L=30$; lower, $L=50$), and anisotropy Δm (open symbols, $L=50$) versus temperature. Inset: scaled variance of order parameter, $L=30$ (open symbols) and $L=50$ (filled symbols).

FIG. 4. Typical configuration, $L=50$, $T=2$.

or vacancy pairs, as is typical of a lattice gas well below the critical temperature. The interface between the high- and low-density regions meanders considerably, and is quite rough. (A quantitative discussion of the interface roughness is given below.)

B. Driven system

We determined the stationary properties for various drive strengths f, for system sizes $L=20$ and 30, following the procedure described above. The mean energy is plotted versus temperature in Fig. 5 for various drive intensities. For a relatively weak drive ($f=1$), the energy is only slightly greater than in equilibrium; at larger drives we observe a substantial increase in energy.

In Fig. 6 we compare the scaled energy variance c in equilibrium and under drive $f=1$. Although c does not represent the specific heat for $f>0$, it is nevertheless reasonable to suppose that a singularity in c (or a sharp peak, in a finite system) marks a phase transition. It is therefore interesting to note that the $f=1$ data exhibit a sharper peak (and at a

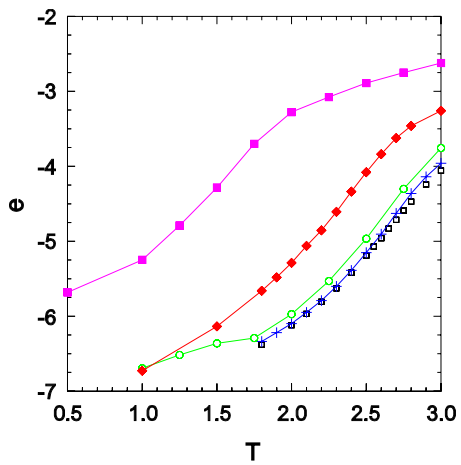

FIG. 5. (Color online) Mean energy per particle versus temperature for $L=30$ and (right to left) drive strength $f=0$, 1, 2, 4, and 7.

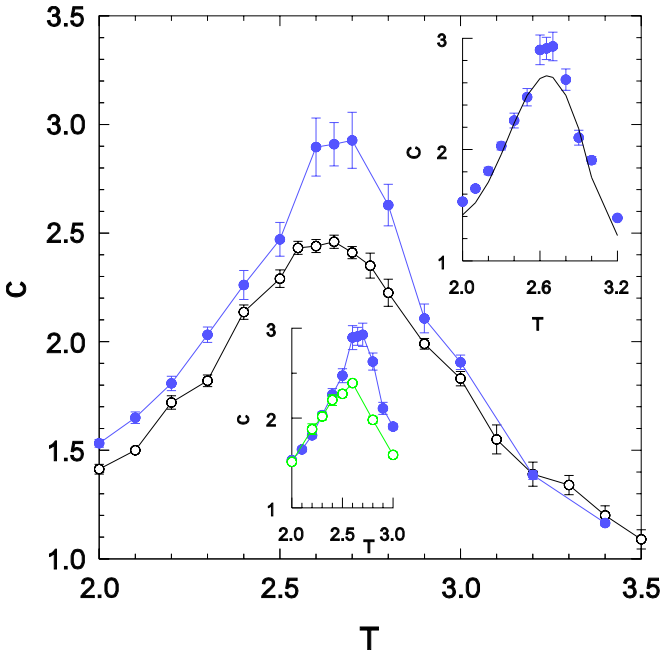

FIG. 6. (Color online) Scaled energy variance c versus temperature T for drive $f=1$ (upper curve) and in equilibrium (lower). Upper inset: comparison ($f=1$, $L=30$) of the scaled variance (points) and de/dT (smooth curve). Lower inset: comparison ($f=1$) of the scaled variance for system sizes $L=20$ (open symbols) and $L=30$ (filled symbols).

slightly higher temperature) than in equilibrium. This suggests that a weak drive facilitates ordering. For $f=1$, the scaled variance (upper inset) follows the same trends as, but is generally greater than, the specific heat, de/dT. (In this case we obtain de/dT via numerical differentiation of a polynomial fit to the energy data.) This suggests that the drive causes fluctuations beyond those generated by thermal mechanisms. (That is, the drive realizes work on the system but is not included in the energy balance.) The dependence of var(e) on system size (lower inset) is qualitatively similar to that found in equilibrium; we estimate $T_c=2.85(5)$ for $f=1$. Analysis of the order parameter (Fig. 7) confirms that a weak drive ($f=1$, 2) enhances ordering, whereas a stronger one inhibits it. These data suggest a transition temperature of $T_c \approx 2$, for $f=7$.

The effect of increasing drive at fixed temperature is shown in Figs. 8 (energy and its variance) and 9 (order parameter). The energy increases slowly with f for a weak drive, rapidly for intermediate drive strength, and then exhibits a steady, more gradual growth for $f>8$ or so. The energy variance shows a marked peak (near $f=4$ for $T=2.5$, and $f=6$ for $T=2.0$), which appears to be associated with destruction of an ordered arrangement. The amplitude of this peak is much larger than in equilibrium. For larger drives, var(e) increases steadily with f, as the drive forces particles out of the periodic potential minima. Figure 9 again shows that a weak drive enhances ordering. The order parameter reaches a maximum near $f=1-2$, and then decays rapidly when the drive is increased further, and begins to dominate interparticle attraction.

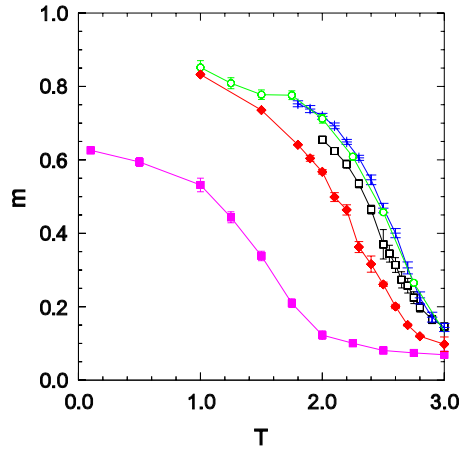

FIG. 7. (Color online) Order parameter versus temperature for $L=30$ and drive $f=0$ (open squares), 1 (+), 2 (open circles), 4 (diamonds), and 7 (filled squares).

The stationary current density j, defined as the mean displacement $\langle \Delta x \rangle$ per site and unit time, is plotted as a function of drive in Fig. 10. For the range of parameters studied here, j is an increasing function of both f and T. In the disordered phase (upper set of points in Fig. 10), the current grows linearly with f for small f, and then shows signs of saturating at larger values of the drive. At lower temperatures and weak drives, such that the system is ordered, the current is severely reduced, but it takes values comparable to those at higher temperature once f is large enough to disorder the system. The latter event is signaled by a sharp peak in the variance of the energy (Fig. 8); the singularity (if any) in the current is much weaker. Our results suggest that below T_c, the particles are organized into dense stripes oriented along the drive, as in DDSs; this is confirmed in the configurations shown in Fig. 11. Comparison of three configurations for $T=2$, with

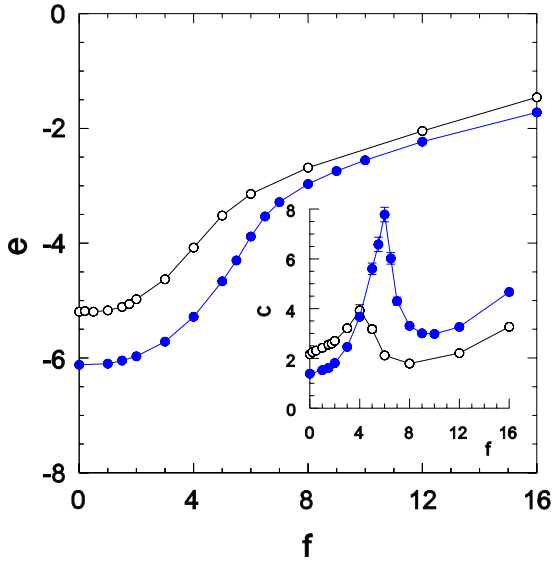

FIG. 8. (Color online) Energy per particle versus drive f for temperature $T=2.5$ (open symbols) and 2.0 (filled symbols), $L=30$. Inset: c versus f for the same parameters.

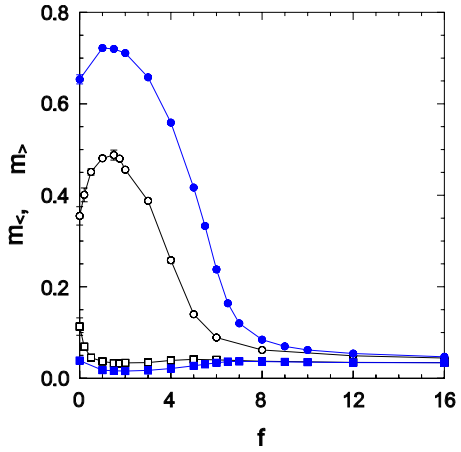

FIG. 9. (Color online) Order parameter components $m_>$ (upper curves) and $m_<$ (lower curves) versus drive f for $T=2.5$ (open symbols) and 2.0 (filled symbols), $L=30$.

drive $f=1$ and 5 (Fig. 11, upper panels) and with zero drive (Fig. 4) shows that the interface is smoothest in the weakly driven system. (Under a stronger drive the interface appears to be more diffuse.)

To quantify the effect of the drive on the interface, we calculate the interface width. Following the procedure used in Ref. [19], we first coarse-grain the configuration, to eliminate vacancies from the high-density strip and isolated occupied sites from the low-density region. This permits unambiguous definition of interfaces between the two phases. Then each column j (or row, should the interfaces be oriented vertically) is characterized by a height h_j, given by the distance between the interfaces. The interface width w is then defined via $w^2=\mathrm{var}(h_j)$, evaluated by averaging over columns and over many configurations during the evolution. Figure 12 shows that in the absence of a drive, w^2 increases linearly with system size L, as expected for the rough interfaces characteristic of a two-dimensional lattice gas. In the presence of a modest drive ($f=1,2$) the interface width is sharply reduced; the data are consistent with w^2 attaining a finite limit as $L\to\infty$ (i.e., an asymptotically *smooth* interface), though the present data are insufficient to fix the large-L behavior definitively.

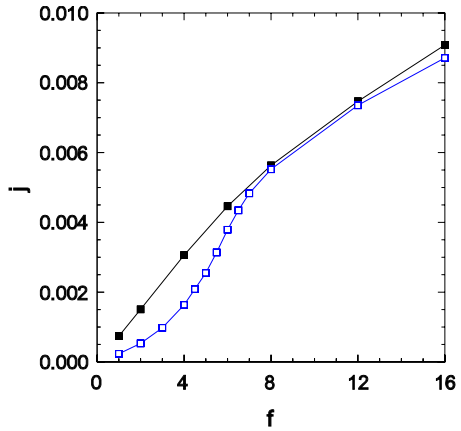

FIG. 10. (Color online) Current density j versus drive f for $T=3.5$ (filled symbols) and 2.0 (open symbols), $L=30$.

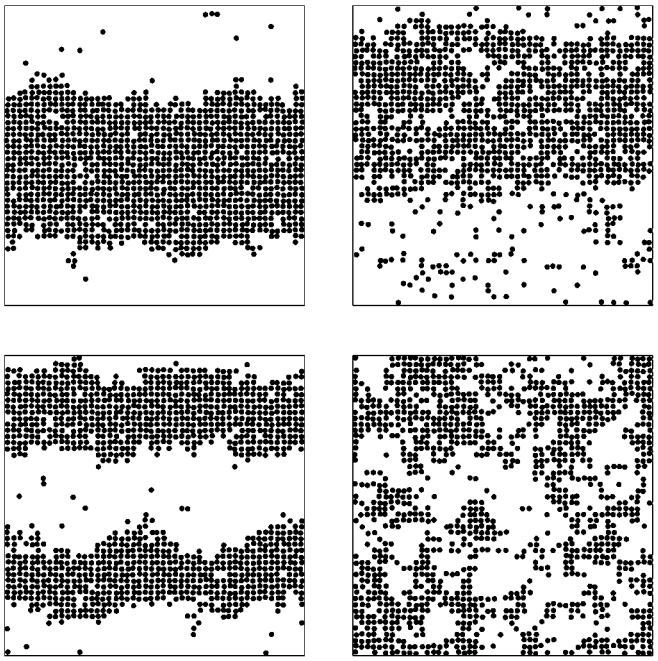

FIG. 11. Typical configurations in the driven system, $L=50$. Upper left: $T=2$, $f=1$; upper right: $T=2$, $f=5$; lower left: two strip configuration, $T=2$, $f=1$; lower right: $T=3$, $f=1$. The drive is directed to the right.

The inset of Fig. 12 shows that, unlike in the driven lattice gas, a strong drive causes the interface to roughen, as particles are pulled from the potential minima, and ordering is reduced. This is also evident in the configuration for $T=2$ and $f=5$ shown in Fig. 11. This relatively strong drive induces many vacancies in the high-density phase, and causes the interfacial region to become diffuse.

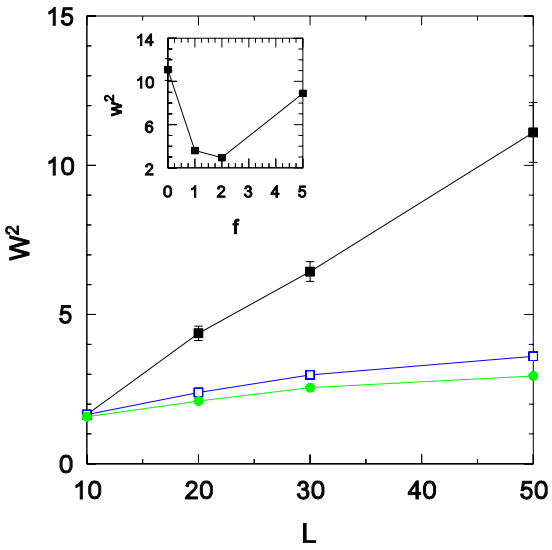

FIG. 12. (Color online) Squared interface width w^2 versus L for (top to bottom) drive $f=0$, 1, and 2, $T=2$. Inset: w^2 versus f for $L=50$, $T=2$.

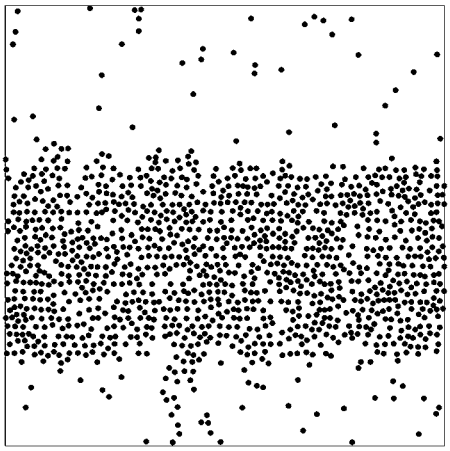

FIG. 13. Typical configuration under moving background potential, $L=50$, $v=0.1$, $T=1.5$.

The lower left panel of Fig. 11 shows a configuration well below the transition temperature, in which two separate stripes have emerged. Such configurations form readily if a drive (along x) is applied to a system that has phase separated (under zero drive) with the interfaces oriented along the y direction. As in the driven lattice gas, this appears to be a very long-lived metastable state at low temperature [7], as would be expected in the absence of significant fluctuations in the interface positions. Figure 11 also shows a typical configuration in the driven system ($f=1$), above the critical temperature ($T=3$); in this case no particle strips are evident, and there is no obvious signature of the drive in the particle configuration. (On general grounds one does expect to observe anisotropic, possibly long-range correlations in a driven system above its critical point [17]. We defer analysis of correlation functions to future work.)

C. Moving background potential

We performed some preliminary studies of another method for perturbing the system out of equilibrium. In this case there is no drive, but the external potential is time dependent, given by $V(x-vt,y)$, with the function $V(x,y)$ as in Eq. (1). [Since the potential is periodic, the first argument of V is effectively $x-vt(\mathrm{mod}\ 1)$.] The particles are dragged along by the moving potential array, as in *optical peristalsis* [18].

Our results in this case are based principally on studies using $L=30$; the background potential amplitude V_0 is reduced from 40 to 10, and the trial particle displacement Δx is uniform on $[-0.5,0.5]$ (similarly for Δy). Otherwise the parameters are as in the studies reported above. For velocities $v\leq 0.02$, there is anisotropic, lattice-gas-like ordering for $T<T_c$. (The critical temperature is about 2.5 for $v=0.01$.) For larger velocities, $v=0.05$ or 0.1, the critical temperature is reduced considerably ($T_c\simeq 1.8$ for $v=0.1$). The interface between phases is again oriented along the x direction, but for $v\geq 0.05$ the particle configuration (Fig. 13) shows little evidence of periodic structure. While there is some tendency for particles to form rows of constant y (as expected, if particles follow the moving potential wells), the positions along the drive direction seem quite irregular.

IV. CONCLUSIONS

We study a two-dimensional model of colloidal particles subject to a periodic external potential and an interaction that favors ordering in a lattice-gas-like arrangement. Stationary properties are determined via Monte Carlo simulation. In equilibrium, the specific heat and the order parameter exhibit behaviors typical of a second-order phase transition. When the hopping dynamics is biased by a weak drive, we observe anisotropic phase separation: the interface is oriented along the drive, and is much smoother than in equilibrium, as found in DDSs [19]. Similarly to attractive DDSs, in which ordering is facilitated by the drive, we find that a weak drive favors ordering. A strong drive, by contrast, tends to destroy order, and provokes very large energy fluctuations. (The strong drive pulls particles off the minimum-potential positions, which is not possible on the lattice.) Preliminary analysis indicates that a colloidal system under a moving background potential may offer another realization of anisotropic phase separation.

A principal characteristic of driven systems is ordering into stripes or lanes oriented along the drive. This tendency has been noted in the present system, and in DDSs in general, as well as in model systems of (1) particles interacting via short-range attraction and long-range (Coulombic) repulsion, subject to an external drive and a quenched disordered background [20]; and (2) a binary colloid mixture with particles driven in opposite directions, in both two and three dimensions [21]. It therefore seems likely that lane formation is a generic feature of ordered phases in driven systems.

Our results suggest that a suspension of colloidal particles with short-range repulsive forces and slightly longer-range attraction, in the presence of a periodic external potential (with a lattice constant matched to the interparticle potential minimum), provides a good candidate for realization of both equilibrium and driven lattice gases. Although the physics of such a continuous space system is richer than the corresponding lattice model, there is good reason to expect that similar scaling properties will emerge, in a suitable range of parameter values. While the equilibrium model could in principle be realized using a planar optical-tweezer array to provide the external potential, the driven system must, by nature, have periodic boundary conditions along the drive direction. This suggests a cylindrical geometry, with the driving force provided by Stokes drag on the particles in steady Couette flow. (This scheme, without the optical-tweezer array, could also be used to realize the type of system studied in Ref. [10].) Alternatively, one could impose relative rotational motion between the colloidal suspension and the tweezer array. We defer analysis of the feasibility of such a setup to future work.

Many additional aspects of the system remain to be explored theoretically and in simulations. While the present study uses (because of its greater efficiency) Monte Carlo simulation to map out equilibrium properties and nonequilibrium steady states, the driven system should be investigated

via Langevin dynamics. Aside from affording a better account of nonequilibrium stationary and time-dependent properties, this method would allow for inclusion of hydrodynamic interactions between the particles and with container walls. Preliminary studies of the model system studied here using the Langevin equation in fact yield qualitatively similar results to those reported above [22]. It would also be interesting to remove the periodic boundaries in the direction perpendicular to the drive, and the restriction to two dimensions. More detailed characterization of the phase transitions exhibited by this system, both in and out of equilibrium, using large-scale simulations, are planned for future work.

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