# Cooperative behavior and pattern formation in mixtures of driven and nondriven colloidal assemblies

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We simulate a disordered assembly of particles interacting through a repulsive Yukawa potential with a small fraction of the particles coupled to an external drive. Distortions in the arrangement of the nondriven particles produce a dynamically induced effective attraction between the driven particles, giving rise to intermittent one-dimensional stringlike structures. The velocity of a moving string increases with the number of driven particles in the string. We identify the average stable string length as a function of driving force, background particle density, and particle charge. This model represents a type of collective transport system composed of interacting particles moving through deformable disorder.

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The collective transport of interacting particles driven through disordered backgrounds has been studied extensively in systems such as moving vortex lattices in the presence of random pinning [1], driven charge density waves [2], and sliding friction [3]. A variety of nonequilibrium behaviors occur in these systems, including fractal flow patterns, avalanches, nonlinear velocity-force responses, and dynamic reordering transitions. Here, the disorder in the substrate is quenched and does not change with time, but substrates which distort and respond to the driven particles are also possible, such as in the case of a single colloidal particle driven through a disordered background of other particles which are not coupled to the external drive [4,5]. Both simulations [5] and experiments [4] show that such a system can exhibit a nonzero threshold force for motion as well as a nonlinear velocity vs applied force response. The driven particle moves in an intermittent manner and causes local rearrangements and distortions in the surrounding bath of colloids. An open question is what happens if there are multiple particles driven through a background of nondriven particles, rather than only a single driven particle? In particular, it is possible that the ability of the surrounding media to react to the motion of driven particles could induce an emergent effective driven particle-particle interaction, leading to new types of moving structures or to pattern formation that is not observed in driven systems with fixed or quenched background disorder.

Recent studies have focused on a related system in which two sets of identical repulsive particles are driven in opposite directions [6–9]. Here, a laning phenomenon occurs, and the species segregate into multiple streams flowing past one another. This model has been studied in terms of pedestrian dynamics, where the particles represent people moving in opposite directions [8], as well as in the context of charged colloids in the presence of a driving field [7]. The colloidal systems are particularly attractive for further study since binary mixtures of oppositely charged colloids which can undergo laning transitions in the presence of a driving field have been produced recently in experiment [9]. This type of system opens a wealth of new experimental possibilities.

To our knowledge, the case of a small fraction of particles driven through a responsive but undriven background has not been studied previously. By using a smaller ratio of driven particles compared to previous work [7], we identify and characterize individual strings of moving particles, which has not been done in previous studies. Here we show that when multiple particles move through a background of nondriven particles, an effective emergent attraction arises between the driven particles even though all of the pairwise interactions between the particles are repulsive. We find that intermittent moving strings form which have an average stable length that is a function of the driving force, system density, and particle charge. Strings that are longer than average are very short lived. The string velocity increases monotonically with the string length. We also study the power spectra of the velocity fluctuations, which has never been considered before in these types of systems. We show that the power spectrum can be very valuable in distinguishing between the cases of single driven particles, small fluctuating strings, and states with fully formed lanes of moving particles.

We consider a two-dimensional system with periodic boundary conditions in the $x$ and $y$ directions containing $N_c$ particles interacting via a screened Coulomb or Yukawa interaction. Only a small number $N_D$ of the particles couple to an external drive. The particles move in an overdamped background, and we neglect hydrodynamic effects, which is reasonable for the low volume fraction limit considered here. We use molecular dynamics (MD) to numerically integrate the overdamped equations of motion for the particles, given for particle $i$ by $\eta d\mathbf{r}_i/dt=\mathbf{F}_i^{cc}+\mathbf{F}_D^i+\mathbf{F}_i^T$. Here the damping coefficient $\eta=1$. The colloid-colloid interaction force term $\mathbf{F}_i^{cc}=-q_iA\Sigma_{j\neq i}^{N_c}\nabla_i V(r_{ij})$ contains the Yukawa interaction potential $V(r_{ij})=(q_j/|\mathbf{r}_i-\mathbf{r}_j|)\exp(-\kappa|\mathbf{r}_i-\mathbf{r}_j|)$, where $\mathbf{r}_{i(j)}$ is the position of particle $i(j)$, $q_{i(j)}$ is the charge of colloid $i(j)$, and all colloids have the same sign of charge so that all interaction forces are repulsive. Energies are expressed in units of $A=q^2/4\pi\epsilon\epsilon_0$ and lengths in terms of $a$, the average colloid spacing. The screening length $1/\kappa$ is set to 1/2 in all our simulations. We use a time step of $\Delta t=0.01$ in dimensionless units, such that a colloid moving with an average velocity of 1.5 requires roughly 100 time steps to translate by one lattice constant. The background colloids are composed of a 50:50 mixture of two charges with $q_1/q_2=1/2$, which produces a

 

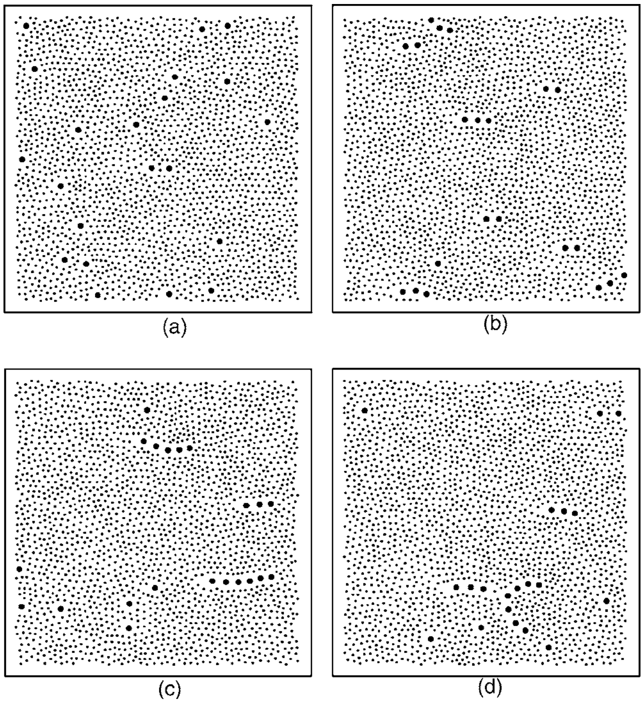


FIG. 1. Images of the system composed of a disordered array of background colloids (small circles) and particles that couple to the external drive (large circles) for $q_D/q=6.67$, $n_c=0.94$, $F_D=3.0$, and $N_D/N_c=0.01$. (a) Initial configuration; (b) the same system after $10^5$ MD steps; (c) a typical steady state snapshot after $10^6$ MD steps; (d) snapshot of a string breaking into shorter strings. The driving force is from left to right.

noncrystalline background arrangement. The average background charge $q=(q_1+q_2)/2$. For the nondriven particles, $\mathbf{F}_D^i=0$. The colloids that couple to the external field have charge $q_D$ and experience a constant drive $\mathbf{F}_D^i=F_D\hat{\mathbf{x}}$ applied after the system is equilibrated by simulated annealing. The thermal force $\mathbf{F}_i^T$ has the properties $\langle\mathbf{F}_i^T\rangle=0$ and $\langle\mathbf{F}_i^T(t)\mathbf{F}_j^T(t')\rangle=2\eta k_B T\delta_{ij}\delta(t-t')$. For a system of length $L$ the colloid density is $n_c=N_c/L$. We consider both the constant $n_c$ case, where $L$ and $N_c$ are increased simultaneously, as well as the case where $n_c$ is increased by fixing $L$ and increasing $N_c$.

In Fig. 1 we show four snapshots which highlight the dynamically induced attraction of the driven particles, indicated as large black dots, and the formation of the string structures for $n_c=0.94$, $q_D/q=6.67$, and $F_D=3.0$. Figure 1(a) shows the initial state in which the driven particles are well separated from each other. In Fig. 1(b), after $10^5$ MD time steps, the driven particles begin to form stringlike structures of length $N_s=2$ to 3 colloids which are aligned in the direction of the driving force (left to right in the figure). In this nonequilibrium system, despite the fact that all of the pairwise interactions are repulsive, an effective attraction emerges between the driven particles. What is difficult to convey through still images is the fact that as $N_s$ increases, the average string velocity $\langle V_x^s\rangle$ increases so that strings move considerably faster than isolated driven particles. In Fig. 1(c) after $10^6$ MD time steps, the system has reached a nonequilibrium steady state. Here the average stable length of a string $\langle N_s\rangle=5$ to 6. Strings with $N_s>6$ form occasionally but have a very short lifetime. In Fig. 1(c), strings with $N_s=6$, 5, 3, and 1 are present. All of the strings have an *intermittent* character, in that driven particles in the string detach from the string, and new driven particles join the string. Typically strings add or shed one driven particle at a time; however, long strings with $N_s=7$ or 8 tend to break up into separate strings with $N_s=3$, 4, or 5, as illustrated in Fig. 1(d).

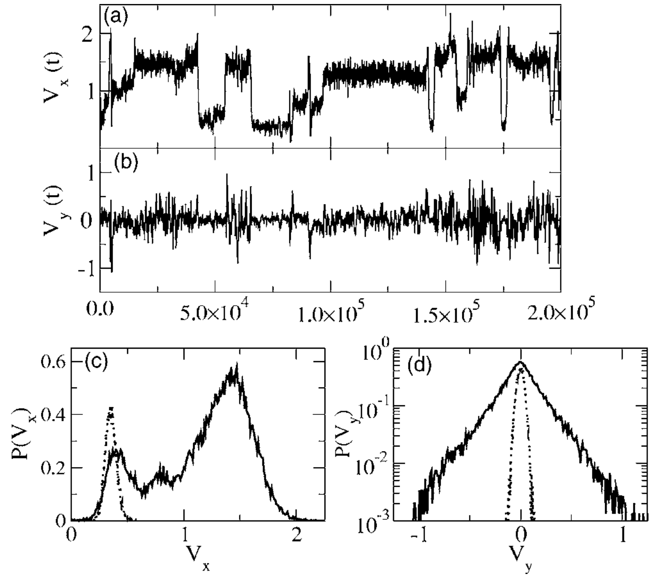


FIG. 2. (a) Time series $V_x(t)$ for a single driven colloid from the system in Fig. 1(a) with $N_D=20$. (b) The corresponding $V_y(t)$. (c) Solid line: The normalized histogram of $V_x$ for the system in (a). Dotted line: Histogram of $V_x$ for $N_D=1$. (d) Solid line: The normalized histogram of $V_y$ for the system in (b). Dotted line: Histogram of $V_y$ for $N_D=1$.

We note that if $N_D/N_c$ is increased while all other parameters are held fixed, then $\langle N_s\rangle$ remains unchanged at $\langle N_s\rangle=5$ to 6. We find the formation of stable strings over an extensive range of parameters, including all $q_D/q\geqslant1.0$ (where we have checked up to $q_D/q=40$), for $n_c>0.17$, and for arbitrarily large $F_D$. The value of $\langle N_s\rangle$ depends on the specific choice of parameters; however, the phenomenon of string formation is very robust provided that the driven particles can distort the arrangement of the background particles. In Fig. 1, $N_D/N\approx0.01$, so that the initial average distance between driven particles is many times larger than the pair-interaction length. Thus, the attractive force between driven particles is mediated by the distortion field produced in the background particles. If the fraction is increased as much as an order of magnitude up to $N_D/N_c=0.1$, the preferred length of $\langle N_s\rangle=5$ to 6 persists. We note that if $N_D/N_c$ is too high, finite size effects due to the boundary conditions become important.

In order to illustrate the intermittent behavior and the velocity increase as the number of colloids in a string increases, in Fig. 2(a) we plot a typical time series of the velocity in the direction of drive $V_x$ for one of the driven colloids from the system in Fig. 1(a). Each point of the velocity time series is obtained by averaging the instantaneous velocity over 50 time steps. Here $V_x(t)$ shows a series of well defined increasing or decreasing jumps, with a roughly constant velocity between the jumps. The lower value that $V_x$ takes is around 0.39 corresponding to the average velocity of a driven particle that is not in a string. There are several places where $V_x=1.45$ which corresponds to the average velocity of a string with $N_s=5$. Additionally, there are some plateaus near a value of $V_x=0.7$ which corresponds to $N_s=3$. Figure 2(a) also shows that for short times, $V_x>1.5$, which corresponds to strings with $N_s>6$. This figure shows that the driven colloid is exiting and joining strings of different lengths.

Figure 2(a) is only a portion of a much larger time series which covers $10^7$ MD steps. In Fig. 2(c) we plot the histogram of this entire time series along with the histogram of $V_x$ (dotted line) for a case where there is only one driven particle in the system so that strings do not form. The $N_D=1$ curve shows a single peak in $P(V_x)$ near $V_x=0.35$, which falls near but slightly below the first peak in $P(V_x)$ for the multiple driven particle system. Thus, the lowest value of $V_x$ in Fig. 2(a) corresponds to time periods when the driven colloid is moving individually and is not part of a string. Additionally, there is a small peak in $P(V_x)$ for the $N_D=20$ system near $V_x=0.8$, which corresponds to the formation of strings with $N_s=2$. There is a large peak in $P(V_x)$ around $V_x=1.45$ corresponding to $N_s=5$ and 6. For $V_x>1.5$ there is a rapid decrease in $P(V_x)$. This result shows that there is a preferred string length of $\langle N_s\rangle \approx 5$ for the $N_D=20$ system. The average velocity of a driven particle in the $N_D=20$ case is much larger than the average velocity in the $N_D=1$ case, even through the fraction of driven particles is only $N_D/N_c=0.01$.

Driven particles traveling in strings show a more pronounced transverse wandering motion than individual driven particles. In Fig. 2(b), we plot the time series for the velocity transverse to the driving force, $V_y$, for the same particle as in Fig. 2(a). Here $\langle V_y\rangle=0$, and there are numerous spikelike events in the positive and negative directions. In Fig. 2(d) we plot the histogram of the entire time series of $V_y$ (solid line) along with the same histogram for a system containing only a single driven particle (dashed line). The $N_D=1$ curve shows a well defined Gaussian distribution of $P(V_y)$ centered at zero. For $N_D=20$, the spread in $V_y$ is much larger and the fluctuations are non-Gaussian, as indicated by the presence of large tails in the distribution. There are an excess of events at larger values of $|V_y|$ which result from the fact that the particles in a chain move in a correlated fashion. The non-Gaussian statistics also indicate that the strings have transverse superdiffusive behavior.

We next examine the average velocity of the strings $\langle V_x^s\rangle$ vs $N_s$, which we plot in Fig. 3(a) for systems with the same parameters as in Fig. 1 at $F_D=3.0$, 4.0, and 5.0. The bottom curve, which corresponds directly to the system in Figs. 1 and 2 shows that for $N_s=1.0$, $\langle V_x^s\rangle=0.35$, while for $N_s=5$ and 6, $\langle V_x^s\rangle \approx 1.4$. The string velocity increases rapidly with $N_s$ for $N_s<5$ and then increases much more slowly with $N_s$. The longer strings become considerably more winding and experience additional frictional drag, whereas the shorter strings move rigidly. Due to the meandering of the longer strings, there is a tendency for the driven particles at the back of the string to be ripped away from the string very quickly. For

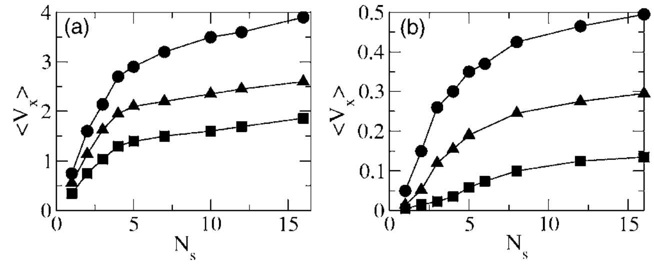


FIG. 3. $\langle V_x^s\rangle$ vs $N_s$. (a) The same system as in Fig. 1(a) with $q_D/q=6.67$ for $F_D=3.0$ (squares), 4.0 (triangles), and 5.0 (circles). (b) The same system as in (a) for $q_D/q=1.33$ and $F_D=0.5$ (squares), 0.75 (triangles), and 1.0 (circles).

curves with higher $F_D$, there is still a monotonic increase in the average velocity with $N_s$. In order to show that this phenomenon is robust over a wide range of parameters, in Fig. 3(b) we plot $\langle V_x^s\rangle$ vs $N_s$ for a system with $q_D/q=1.33$ for $F_D=0.5$, 0.75, and 1.0. For these parameters we again observe the string formation and an increase in $\langle V_x^s\rangle$ with $N_s$. The best functional fit we find is $\langle V_x^s\rangle \propto \ln(N_s)$; however, other forms can also be fit, such as two linear fits for the system with $q_D=1.33$.

In Fig. 4(a) we show the most probable number of particles $\langle N_s\rangle$ in a string for the same system in Fig. 1 for varied $F_D$. Here $\langle N_s\rangle$ goes through a maximum as a function of $F_D$. As $F_D$ decreases below $F_D<0.2$, the distortion created by the driven particles in the surrounding background particles decreases, and therefore there is less attraction between the driven particles. At high $F_D$, the velocity fluctuations along the string become important, which tends to limit $\langle N_s\rangle$. In the inset of Fig. 4(c), the corresponding $\langle V_x^s\rangle$ vs $F_D$ shows a sharp increase in $\langle V_x^s\rangle$ at $F_D=2.0$ when the longest strings form. This is followed by a saturation of $\langle V_x^s\rangle$ just above $F_D=2.0$, caused when the decreasing $\langle N_s\rangle$ counterbalances the increasing $F_D$.

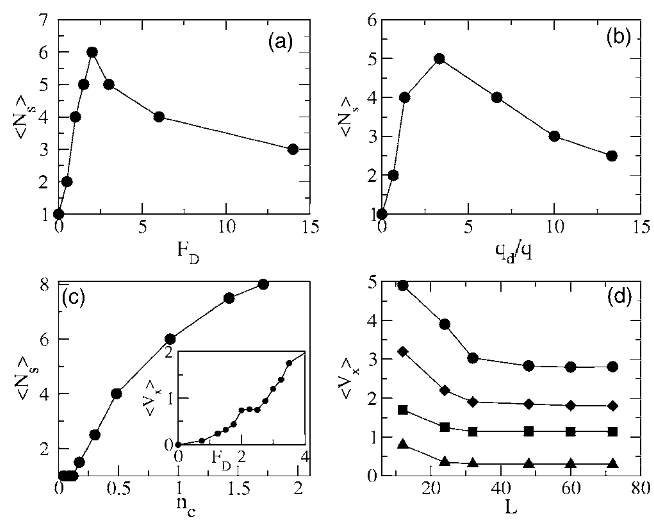


FIG. 4. (a) $\langle N_s\rangle$ vs $F_D$ for the system in Fig 1. (b) $\langle N_s\rangle$ vs $q_D$ for the same system as in (a) for $F_D=1.0$. (c) $\langle N_s\rangle$ vs $n_c$ for $q_D=6.67$ and $F_D=2.0$. Inset: $\langle V_x^s\rangle$ vs $F_D$ for the system in (a). (d) $\langle V_x^s\rangle$ vs system size $L$ for the system in Fig. 1 for $F_D=1.5$ (triangles), 3.0 (squares), 4.5 (diamonds), and 6.0 (circles).

In Fig. 4(b) we show that $\langle N_s \rangle$ for a system with a fixed $F_D=1.0$ also goes through a maximum as a function of $q_D/q$. For small $q_D/q$, the driven particles produce very little distortion in the surrounding medium so there is little attraction between them. For large $q_D/q$, the repulsion between the driven particles becomes more prominent and the average velocity drops, which again reduces the distortion field. In Fig. 4(c) we plot $\langle N_s \rangle$ vs the particle density $n_c$ for a system with $q_D=6.67$ and $F_D=2.0$. At low $n_c$, all the particles are far apart and the distortion in the bath particles is reduced or absent. As $n_c$ increases, the particle-particle interactions are enhanced and the string can grow longer. Presumably for very high $n_c$, $\langle N_s \rangle$ will decrease again as the average velocity will have to be reduced. We are not able to access particle densities in this range.

In Fig. 4(d) we plot $\langle V_x^s \rangle$ vs the system size $L$ for $F_D=1.5$ (lower curve), 3, 4.5, and 6.0 (upper curve). In each case, $\langle V_x^s \rangle$ initially decreases with $L$ but reaches a saturation value by $L \approx 40$. The change in $\langle V_x^s \rangle$ at small $L$ occurs because, in small systems, the periodic boundary conditions allow driven colloids to interact strongly with their own distortion trails. For larger $F_D$, larger system sizes must be used to avoid this effect. In addition to the increase in $\langle V_x^s \rangle$ in the small systems, $\langle N_s \rangle$ also increases for the smallest systems. This emphasizes the fact that sufficiently large systems must be considered in order to avoid finite size effects. We have used $L=48$ throughout this work, which is large enough to avoid finite size effects for the quantities we have measured.

The attraction between the driven particles is mediated by the distortion in the surrounding bath of colloids. The concept of nonequilibrium depletion forces, which are highly anisotropic and have both an attractive and a repulsive portion, was introduced recently for a case where two large colloidal particles have an additional bath of smaller particles flowing past them [10]. The surrounding particle density is higher in front of the large particle and lower in back, which causes an anisotropic attraction force between the large colloids that aligns them with the direction of flow. Although this model was only studied for two particles, it seems to capture several of the features that we find in our simulations, including the alignment of the strings in the direction of drive. This suggests that nonequilibrium depletion force models could be applied for multiple large particles. The longer strings move faster since the front particle has additional particles pushing on it from the back. Since the chains are aligned, the cross section with the background particles remains the same as for a single driven particle; however, the string is coupled $N_s$ times more strongly to the external field. As longer strings become increasingly meandering, the cross section with the background particles increases and limits the speed that long strings can attain. The role of the effective cross section of the chain in determining the chain speed is also supported by work on a related system of chains of ions moving through a dense jellium target. In Ref. [11], the stopping power per ion for an ion chain moving parallel to its length initially decreases with chain length for short chains before saturating for longer chains. The stopping power, which would correspond to an effective viscosity contribution in our context, is reduced for the chain of ions compared to the value it would have for an equivalent number of isolated ions due to dynamic interferences between the moving ions.

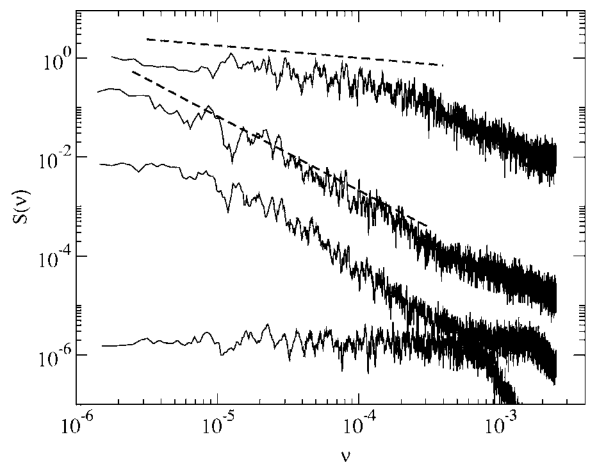


FIG. 5. The power spectra $S(\nu)$ obtained from the velocity fluctuations in the $x$-direction $V_x(t)$ for a system with the same parameters as in Fig. 2. $\nu$ is given in units of inverse molecular dynamics steps. Bottom curve: $S(\nu)$ for a single driven particle. The remaining curves show $S(\nu)$ for $V_x(t)$ for multiple driven particles from Fig. 2(a). Second lowest curve: $N_D/N_c=0.01$. Second highest curve: $N_D/N_c=0.1$. Top curve: $N_D/N_C=0.4$. The lower dashed line is a power law fit with $\alpha=1.5$ and the upper dashed line is a power law fit with $\alpha=0.25$. The curves have been offset in the $y$ direction for presentation purposes.

The power spectrum of the velocity fluctuations in the direction of drive has not been examined in previous work on these types of systems. This is defined as follows:

$$S(\nu) = \left| \int V_x(t) e^{-2\pi i \nu t} dt \right|^2. \quad (1)$$

If the velocity fluctuations are highly intermittent, we would expect to observe a $1/f^\alpha$ noise signal. In Fig. 5 we plot the power spectrum for the system in Fig. 2 for a single particle (lowest curve), and for multiple driven particles at $N_D/N_c=0.01$, $N_D/N_s=0.1$, and $N_D/N_s=0.4$ (top curve). The curves have been shifted from each other in the $y$ direction for clarity. The lower dashed line is a $1/f^\alpha$ fit for $\alpha=1.5$ and the upper dashed line is the same for $\alpha=0.25$. Here the single driven particle shows a white noise spectrum which reflects the lack of correlations in the velocity fluctuations. Additionally, the histogram of velocities for a single driven particle from Fig. 2 shows a Gaussian distribution in this case. If $F_D$ is much lower and close to the single driven particle depinning threshold, the velocities become non-Gaussian and a $1/f$ noise signal is observed [12]. In the case where the intermittent strings form, at $N_D/N_c=0.01$ and 0.1, the velocity histograms are non-Gaussian as in Fig. 2 and the velocity fluctuations are highly intermittent. In these cases, the power spectrum has a $1/f^\alpha$ form with $\alpha > 1.0$. When the density of driven particles becomes large, stable lanes of driven particles form and the moving particles fluctuate rapidly. The velocity histograms again take a Gaussian form and the power spectrum becomes white with $\alpha \approx 0$. The noise spectrum of the velocity fluctuations is thus another measure which can be used to identify the different flow regimes.

In conclusion, we have investigated a system of repulsively interacting Yukawa particles which form a disordered background for a small fraction of Yukawa particles that couple to an external drive. In this system, despite the fact that all of the pairwise interactions are repulsive, there is an emergent effective attraction between the driven particles which results in the formation of one-dimensional intermittent stringlike structures. The velocity of a string increases with the number of particles in the string. The attraction between the driven particles is mediated by the distortion of the surrounding particles and is likely a result of the recently proposed nonequilibrium depletion forces. Longer strings move faster than shorter strings due to the additional pushing force from the back particles in the string. For a single driven particle at high drives the power spectrum of the velocity fluctuations has a white noise characteristic, while in the intermittent string regime, the power spectrum is of the form $1/f^{\alpha}$ with $\alpha > 1.0$. In the regime where close to half of the particles are driven, the noise spectrum becomes white again. This system represents a class of collective transport where multiple interacting colloids are driven through a deformable disordered background, which is distinct from the case where the background disorder is quenched.

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