Enhancing Nanoparticle Diffusion on a Unidirectional Domain Wall Magnetic Ratchet

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Supporting Information

ABSTRACT: The performance of nanoscale magnetic devices is often limited by the presence of thermal fluctuations, whereas in micro- and nanofluidic applications the same fluctuations may be used to spread reactants or drugs. Here, we demonstrate the controlled motion and the enhancement of diffusion of magnetic nanoparticles that are manipulated and driven across a series of Bloch walls within an epitaxially grown ferrite garnet film. We use a rotating magnetic field to generate a traveling wave potential that unidirectionally transports the nanoparticles at a frequency tunable speed. Strikingly, we find an enhancement of diffusion along the propulsion direction and a frequency-dependent diffusion coefficient that can be precisely controlled by varying the system parameters. To explain the reported phenomena, we develop a theoretical approach that shows a fair agreement with the experimental data enabling an exact analytical expression for the enhanced diffusivity above the magnetically modulated periodic landscape. Our technique to control thermal fluctuations of driven magnetic nanoparticles represents a versatile and powerful way to programmably transport magnetic colloidal matter in a fluid, opening the doors to different fluidic applications based on exploiting magnetic domain wall ratchets.

KEYWORDS: Diffusion, magnetic fields, domain walls, microfluidics, ratchet effect

The ratchet effect emerged in the past as a powerful way to transport matter at the micro- and nanoscale, taking advantage of Brownian motion.1,2 The success of such concept comes from the possibility of using thermal fluctuations to obtain useful work out of a thermodynamic system, although such fluctuations produce noise, heat, and randomize the motion of nanoparticles which limits the efficiency of any device operating at a small scale.3−5 Reducing or controlling thermal fluctuations in nanoparticle systems may present different technological advantages apart from providing important fundamental insight into the dynamics and interactions of matter at such scale. In the first case, diffusion can be used as a means for mixing streams of fluids or for spreading reactants, drugs, and biological species6,7 in micro- and nanofluidic applications.8−10 On a more fundamental level, the search for strategies that enable controlling diffusion and noise has fascinated scientists for long time, since the pioneering lecture of Richard Feynman on Brownian ratchet.11

In a typical ratchet system, the random fluctuations of nanoparticles can be rectified into a directed motion by an external potential. In overdamped systems, different ratchet mechanisms can be sorted in two general classes, depending on the nature of the external potential.1 One class refers to the tilting ratchets that are typically specified by a stationary, spatially asymmetric potential landscape often accompanied by an external force. The asymmetry of the landscape is able to rectify thermal fluctuations into net motion, and the force, which determines the tilt of the total potential, can be used to...
Polarization microscope image showing a 270 nm particle trajectory transported by the rotating magnetic fields that are characterized by a time-dependent landscape, and reverse the programmable nature, that enables to remotely control, direct requires an external random evolution of the landscape. Two particular subclasses of landscapes are the translating potential landscapes usually created by using, for example, the walls or barriers in a microfluidic device. Another broad class are the pulsating ratchets that are characterized by a time-dependent landscape, in which the net particle flux arises from the periodic or random evolution of the landscape. Two particular subclasses are the flashing ratchet with the landscape switching between two states, and the traveling wave ratchet, where the landscape translates at a given speed. The latter allows dragging the particles that are trapped in the energy minima of the moving landscape.

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an epitaxial growth process and they generate strong local stray fields on the surface. In this study, we use an FGF of composition $Y_{1-x}Bi_xFe_{12}Ga_2O_{12}$ ($x = 0.5-1$) that was grown by dipping liquid phase epitaxy on a (111) oriented single crystal gadolinium gallium garnet (Gd$_3$Ga$_5$O$_{12}$) substrate. In absence of external field, the BWs in the FGF are equally spaced at a distance $\lambda/2$ with $\lambda = 2.6 \mu m$ the spatial periodicity and separate domains of opposite magnetization with saturation magnetization $M_s = 1.3 \times 10^4 A m^{-1}$. As shown in Figure 1d, under polarized light the magnetic domains are visible due to the Faraday effect. They appear as stripes of black and white colors, reflecting the direction of magnetization. An external magnetic field perpendicular to the film $H_k$ can be used to easily manipulate the size of these magnetic domains. In particular, $H_k$ increases (decreases) the width $\lambda_+ (\lambda_-)$ of the domains that have magnetization parallel (antiparallel) to the field. As shown in a previous work, for small field amplitudes, $H_k \ll H_c \approx M_s/2$, the change in these domains is small and linear with the applied field, such that $\lambda(\pm H_k) = \lambda/2 \pm \Delta(\lambda_k)$, $\Delta \propto H_k$. and the total wavelength $\lambda = \lambda_+ + \lambda_-$. remains constant. Raising the field amplitude breaks down these favorable conditions. Moreover, beyond the critical field, $H_k > H_c$, irreversible deformations of the stripe pattern occur and at very high field the FGF finally converts into a single magnetized domain. Therefore, to keep reproducible deformations of the Bloch walls, we restrict ourselves to small external fields.

Above the FGF, we demonstrate the controlled transport of three types of paramagnetic polystyrene nanoparticles with diameters $d = 540, 360$, and $270 \text{nm}$, characterized by an $\sim 40\%$ wt iron oxide content (Microparticles GmbH). When placed above the FGF, the nanoparticles show simple diffusive dynamics and are able to easily pass the BWs due to the presence of a $1 \mu m$ thick polymer coating that prevents sticking to the FGF surface, see Materials and Methods. We apply an external magnetic field rotating in the $(x,z)$ plane with frequency $f$ and amplitudes $H_x = H_{c0}(1+\beta)$ and $H_z = H_{c0}$, such that $H = H_{0}(1-\beta)$, such that $H_{c0}$ is used to control the $\beta$ field, the amplitude $H_{0} = \sqrt{(H_z^2 + H_x^2)}$, and an ellipticity parameter $\beta = (H_z^2 - H_x^2)/H_{c0}$. The value of $\beta = -1/3$ is chosen to minimize dipolar interactions between nearest particles, as they can promote the undesired chaining at high density. Although for self-consistency, we account for this small ellipticity in a full numerical model, a more tractable semi-analytical model for the special case of $\beta = 0$ is shown to work quantitatively well.

The applied field modulates the stray magnetic field generated by the FGF, leading to a spatially periodic magnetic energy landscape $U_{0}(x,t) = -U_0 \cos(k(x - v_0 t))$ that translates at a constant speed, $v_0 = \delta f$, Figure 1a, see Materials and Methods for further details and definition of $U_0$ and $k$. Its evolution at the elevation corresponding to the $270 \text{nm}$ particles is shown in Figure 1b. One may intuitively expect that the particles would follow the energy minima whose locations are given by the blue regions and travel the distance of one wavelength during one time period. However, the nature of motion and the velocity of mean drift across the BWs depend on the external frequency, as becomes evident for the simple case of no thermal fluctuations, see eq 2. The variation of $\langle \psi \rangle$ with the frequency $f$ is also shown in Figure 1c, where the experimental data (open symbols) are plotted along with the theoretical predictions (dashed and solid lines) for a $270 \text{nm}$ particle.

As it follows from our model (see eq 2 in Materials and Methods), and as confirmed by the experimental data, there exist two different dynamic states separated by the critical frequency $f_c$. At low frequencies, here $f < f_c \approx 13 \text{Hz}$, the nanoparticles indeed translate consistently with the landscape with the maximum possible speed, $x(t) = \dot{x}(t) = v_0 f$ see the range of perfectly linear increase of $\langle \psi \rangle$ with $f$ in Figure 1c. We also note that at any $0 < f < f_c$ in this “locked” regime, the particle lags behind the minimum of the potential energy landscape $U_{0\text{w}}$ whose relative position is determined by minimizing the potential given by eq 2. For $f > f_c$ the particle is unable to move together with the landscape. It starts to slip and cannot remain localized within the same minimum. In this “sliding” regime, every time period the particle covers a distance smaller than $\lambda$, resulting in a reduction of the mean speed. At high frequencies $f$, the speed of mean drift decays as $\langle \psi \rangle \approx \sqrt{\frac{v_f}{f}}$. Although the increase of $H_k$ leads to a proportional shift of the critical frequency, $f_c \propto H_0$ (see Materials and Methods) and widens the range of frequencies allowed for the synchronous regime, we keep this parameter fixed in the experiment. This is done to avoid undesirable deformations of the Bloch walls and instability of the magnetic pattern that could arise at high field amplitudes. Note also that at low and high frequencies, thus far away from $f_0$, the predictions of the model with thermal fluctuations and the experimental data are highly consistent with the deterministic (zero temperature) results given by eq 2. However, the deterministic model overshadows the experimental data close to $f_c$. Therefore, we conclude that thermal fluctuations start to significantly affect the particle speed close to the critical frequency, blurring the sharp transition from the locked to the sliding dynamics and effectively decreasing the value of $f_c$ as compared to the deterministic model.

While our domain wall magnetic ratchet enables full control over the mean speed of the particles moving across BWs, the instant position of the particles is affected by unavoidable thermal fluctuations, as evidenced by the trajectory in Figure 1d. The role of such fluctuations is typically quantified by measuring the mean squared displacement (MSD), which can be calculated from the positions of the nanoparticles. Because we are interested in the general effect of how the diffusive properties of a nanoparticle are influenced by the ratchet mechanism, we focus on investigating these quantities along the propulsion direction, namely the $x$-axis. Because of the nonvanishing mean drift, we define the MSD as the variance of the corresponding particle position, $\sigma^2_{x}(t) = \langle \delta x^2(t) \rangle \sim t^\alpha$, where $\delta x(t) = X(t) - \langle X(t) \rangle$. For our statistical analysis, we averaged over more than $50$ independent experimentally measured trajectories, for each of which we subtracted its mean drift, $\langle X(t) \rangle$. The exponent of the power law $\alpha$ can be used to distinguish the diffusive $\alpha = 1$ from anomalous (subdiffusive $0 < \alpha < 1$ or superdiffusive $\alpha > 1$) dynamics. Our case corresponds to normal diffusion with an effective diffusion coefficient across the BWs evaluated as

$$
D_{\text{eff}} = \lim_{t \to \infty} \frac{\sigma^2_{x}(t)}{2t} \tag{2}
$$
In Figure 2, we show that effectively, the magnetic ratchet provides a strong enhancement of the diffusion coefficient at an optimal frequency. At low frequencies, the effective drift of the nanoparticles with $d = 270$ nm corresponds to a $D_{\text{eff}}$ much lower than the diffusivity in absence of the external field, $D_0 = 1.0 \text{ µm}^2 \text{s}^{-1}$. At high frequencies, the effective diffusion becomes close to $D_0$. At intermediate frequencies, the effective diffusivity increases with a clear enhancement relative to the free diffusion, $D_{\text{eff}} \approx 5D_0$ at a frequency of $f = 14.5$ Hz, see also Figures 3b,c. In particular, as shown in the inset of Figure 2, after subtracting the mean drift we calculate the effective diffusion coefficient using eq 2 and performing a linear fit only for the region of data where the mean square displacement displays a power law behavior with $\alpha = 1$. This regime is always found in the long-time limit. Overall, the dependence of $D_{\text{eff}}$ on $f$ follows a well-defined trend, characterized by an initial sharp raise above a value of $f \approx 8$ Hz, and an exponential-like decay above a peak at 14.5 Hz, which is close to the critical frequency value measured for this size of nanoparticle, $f_c = 13.4$ Hz.

The observed enhancement of diffusion can be explained by formulating a theoretical model that explicitly takes into account the interaction of the nanoparticle with the magnetic landscape generated by the FGF surface and the thermal noise. Details of the derivation are given in Materials and Methods. In the reference frame moving with the magnetic potential, the behavior of the nanoparticle is equivalent to the motion in an effective tilted periodic potential $V(u)$ as given by eq 5. For such a potential, the effective diffusion coefficient, defined via eq 2, can be expressed as eq 7, which admits an accurate numeric evaluation. We used eq 7 in Figure 3a–c to fit the experimental data for the three types of nanoparticles and find that our theoretical approach captures very well the observed diffusion enhancement for all cases. The explicit form of the effective potential, eq 5, with the tilt $\alpha_f$ and the amplitude of the landscape $\sigma_f$ admits an intuitively clear interpretation of the observed frequency-dependent diffusive regimes, as also illustrated in Figure 3d–f. Indeed, at subcritical frequencies, $f \leq f_c$, the potential barrier the particle needs to overcome to escape from a minimum drops with the frequency as $\Delta V/k_B T \approx \Delta \nu_{m} (1-f/f_{c})^{1/2}$, where $\Delta \nu_{m} = \lambda^2 f_{c}/(4\pi D_0)$ is double the amplitude of the landscape relative to thermal energy, cf. eq 5. At low frequencies ($f \ll f_{c}$), the tilt is negligible and the particle is strongly trapped in a minimum of the magnetic landscape, as shown in Figure 3d. Since in this case the potential barrier is maximum, $\Delta V/k_B T \approx \Delta \nu_{m}$ and is too high for the particle to escape, the effective diffusivity is nearly vanishing, $D_{\text{eff}} \ll D_0$. At intermediate frequencies, as $f$ tends to $f_c$ but remains smaller than $f_c$, the potential barrier decreases with $f$ and disappears at $f = f_c$. In this frequency range, the barriers become progressively more accessible for the Brownian particle, explaining the observed significant enhancement of the effective diffusivity at frequencies $f \approx f_c$, see Figure 3e. Beyond $f = f_c$, there exist no minima in the potential, and at high frequencies $f \gg f_c$, the particle does not feel the landscape, as shown in Figure 3f. As a result, the diffusive motion occurs effectively at a constant force $\alpha_f$ and therefore corresponds to free diffusion, when ideally $D_{\text{eff}} = D_0$.

Further, since the reduced model was developed for the zero field ellipticity, $\beta = 0$, we have performed Brownian dynamic simulations of the full system with $\beta = -1/3$ to confirm the negligible effect of $\beta$ on the basic physics. The simulation (blue lines) agree very well with the model and the experimental data. The only difference is that the system with $\beta = -1/3$ is characterized by slightly higher values of the critical frequency compared to the case $\beta = 0$, which is in agreement with our earlier observation that the value of $f_c$ at any $\beta \neq 0$ is generally smaller than that for $\beta = 0$ at otherwise identical conditions. Taking together, our findings show that each type of nanoparticle investigated exhibit an enhanced diffusive behavior with the highest diffusion coefficient measured for the intermediate size, $d = 360$ nm. This outcome can be understood by considering the balance between two opposite effects. First, reducing the nanoparticle diameter $d$ increases thermal fluctuations and its free diffusion coefficient $D_0 = k_B T/\zeta$, where $\zeta$ is the friction coefficient of a spherical particle immersed in water. Note that if in the bulk $\zeta = 3\eta d/\rho$, with the dynamic viscosity $\eta = 10^{-3} \text{Pa} \cdot \text{s}$, the presence of the FGF surface leads to effectively larger values of $\zeta$. Second, for our system smaller particles come closer to the FGF surface, and thus they are strongly attracted by its stray field, which results in an effective suppression of their thermal fluctuations. For example, for 270 nm particles an effective enhancement of the friction coefficient due to the presence of the FGF surface is estimated to be 25–30%, as follows from the reduction of the diffusion coefficient (cf. the inset of Figure 2) relative to the bulk diffusivity.

While we have analyzed the transport of single particles, our magnetic ratchet enables also the collective motion for an ensemble of nanoparticles. We demonstrate in Figure 4a, where a dense suspension of 270 nm particles are trapped and transported across the FGF surface, see also corresponding Video S2 in the Supporting Information. In the absence of applied field (ratchet off) the colloidal suspension shows simple diffusive dynamics as illustrated by the Gaussian distribution of the displacements perpendicular (along the x-axis) and parallel (along the y-axis) to the magnetic stripes, Figure 4b,c. Here we used $P(\delta x) = (2\pi\sigma_x^{-2})^{-1/2} \exp(-\delta x^2/2\sigma_x^2)$ with the variance $\sigma_x^2(t) = \langle \delta x^2(t) \rangle$, $\delta x(t) = X(t) - \langle X(t) \rangle$, and $X(t) = x(t) - x(0)$ defined as earlier and considered at a sufficiently large time. The distribution $P(\delta y)$ is defined in a similar way, for which $\langle Y \rangle \equiv 0$, implying no mean drift and free
diffusion in the y-direction. We note that when the ratchet is off, \( H_0 = 0 \), the magnetic landscape \( U_m \propto H_0 \) becomes effectively flat and does not affect the dynamics of the particle. In this situation, the mean drift disappears, \( \langle v \rangle = 0 \), and therefore \( \langle X \rangle = \langle v \rangle t = 0 \). As a result, when the ratchet is off, both distributions (green lines in Figure 4b,c) are identical with \( \sigma_x^2 = \sigma_y^2 \) and thus featuring an isotropic free diffusion dynamics.

Upon application of the rotating field, \( H_0 > 0 \), the particles are immediately localized along the BWs, forming parallel chains and being consecutively transported across the magnetic platform. The ratchet transport features a similar dispersion along the perpendicular direction as in the free case, as shown by the blue line in Figure 4b. Along the transport directions we observe a stronger confinement with a narrower distribution of displacement \( P(\delta x) \), where the mean drift is subtracted by putting \( \langle X(i) \rangle = \langle v \rangle t \). Video S2 in the Supporting Information shows the versatility of our magnetic ratchet approach, as now the nanoparticles can be easily trapped or released and transported to the right or left by just inverting the chirality of the rotating field, \( H_x \rightarrow -H_x \) which keeps \( H_0 \) and \( \beta \) unchanged. While these features have been previously reported for microscale systems, our experimental realization proves its potential for further miniaturization of the transported elements and opens the doors toward applications in magnetic drug delivery systems using the higher surface to volume ratio of nanoparticles.

To conclude, we have reported the controlled transport and diffusion enhancement of nanoparticles in a magnetic ratchet generated at the surface of a ferrite garnet film. In contrast to previous experimental works on microscopic particles confined in a rotating optical ring, above patterned plasmonic or lithographic landscapes, our nanoparticles are trapped and controlled on an extended surface in the absence of any topographic relief that can perturb the transport via steric interactions. The advantage of using the garnet film as a source of magnetic background potential is that the domain wall motion has negligible intermittent behavior and hysteresis. This makes the cyclic displacements and the device performance fully reversible. We have also reported in the past the giant diffusion in a magnetic ratchet of microscopic colloids. However, the significant advantage of the present implementation is that the particle fluctuations can also be controlled along the direction of motion and remain completely

Figure 3. (a–c) Effective diffusion coefficient \( D_{\text{eff}} \) versus frequency \( f \) for three types of particles with diameters 270 nm (a), 360 nm (b), and 540 nm (c). All panels show the experimental data (open circles), predictions of the reduced theoretical model with \( \beta = 0 \) according to eq 7 (red line) and estimates of the effective diffusion coefficient from numerical simulations of the full model with \( \beta = -1/3 \), eq 3 (blue lines). The experimental parameters used are \( H_0 = 1200 \text{ A m}^{-1} \) and \( \beta = -1/3 \) that reflect the values used for the simulations. For the three types of particles we also indicate the critical frequencies \( f_c \) used for the model (red) and simulation (blue). (d–f) Magnetic potential \( V(u) \) in units of thermal energy \( k_B T \) in the moving reference frame (eq 5) evaluated for a 270 nm particle and at a frequency of \( f = 1 \text{ Hz} \) (d), \( f = 11 \text{ Hz} \) (e), and \( f = 15 \text{ Hz} \) (f). Here the critical frequency is \( f_c = 13 \text{ Hz} \).
independent of the deformation of the BWs in the lateral directions. This crucial difference has important implications for the design of channel-free nanofluidic devices where the colloidal motion can be confined to a straight line. Further, our work goes beyond a previous one, centered on trapping fluctuating nanoparticles along the BWs in an FGF. Here, we not only demonstrate the possibility to transport nanoscale objects at a well-defined speed but also that our magnetic ratchet system can be used to tune the diffusive properties of the particles, increasing their effective diffusion constant by almost an order of magnitude with respect to the previous case.

Another future avenue of this work is to investigate the dynamics of dense suspensions of interacting nanoparticles, and how collective effects alter the average particle flow. Increasing their density, however, requires a visualization technique different from fluorescent labeling, to avoid artifacts during the tracking mechanism. While with relatively larger particles, the average speed of a colloidal monolayer was found to decrease with respect to the individual case, the presence of stronger thermal fluctuations for smaller particles may cause different nontrivial effects on the overall system dynamics. Finally, our controlled enhanced diffusion in a transported ratchet may be used as a pilot system for fundamental studies related to transport, diffusion, and their complex relationship at the nanoscale.

**Materials and Methods.** **Experimental System and Methods.** We give further details on the sample preparation and experimental setup. In order to decrease the strong magnetic attraction, we coat the FGF with a $h = 1 \, \mu m$ thick layer of a photosensitive (AZ-1512 Microchem, Newton, MA), that is, a light curable polymer matrix, using spin coating at 3000 rpm for 30 s (Spinco Ws-650Sz, Laurell). Before the experiments, each type of particle is diluted in highly deionized water and deposited above the FGF, where they sediment due to the magnetic attraction to the BWs.

External magnetic fields were applied via custom-made Helmholtz coils connected to two independent power amplifiers (AMP-1800, Akiyama), which are controlled by a wave generator (TG1244, TTi). Particle positions and dynamics are recorded using an upright optical microscope (Eclipse Ni, Nikon) equipped with a $100 \times 1.3$ NA oil immersion objective and a CCD camera (Basler Scout scA640-74 fC, Basler) working at 75 frames per second. The resulting field of view is $65 \times 48 \, \mu m^2$.

Video microscopy and particle tracking routines are used to extract the particle positions $\{x_i(t), y_i(t)\}$ with $i = 1, \ldots, N$, from which the mean speed $\langle v \rangle$ is obtained performing both time and ensemble averages. To calculate the mean square displacement and diffusion coefficient, we use $N$ trajectories with length $l_{\text{threshold}} = 200$ frames, corresponding to a measurement time of $\Delta t = 200/75 = 2.6$ s.

**Theoretical Model.** The motion of paramagnetic colloidal particles above the FGF can be well interpreted within a simple model. In an external magnetic field $\mathbf{H}$, a spherically magnetically polarizable particle of volume $V$ behaves as an induced magnetic dipole with the moment $\mathbf{m} = \frac{V}{2} \mathbf{H}$ and the energy of interaction $U_m = -\mu_m \mathbf{m} \cdot \mathbf{H}/2$, where $\chi$ is the susceptibility and $\mu_0 = 4\pi \times 10^{-7} \, \text{H m}^{-1}$ is the magnetic permeability of free space. The total magnetic field above the FGF is given by the superposition $\mathbf{H}^{\text{eff}} = \mathbf{H}^{\text{sub}} + \mathbf{H}^{\text{mod}}$ of the external modulation, $\mathbf{H}^{\text{mod}} = H_0(\sqrt{1 + \beta \cos \omega t}, 0, -\sqrt{1 - \beta \sin \omega t})$, and the stray field of the garnet film $\mathbf{H}^{\text{sub}} \approx (4M_s/\pi) \exp(-kz)(\cos kx, 0, -\sin kx)$, justified under the condition $H_{0} \ll M_s$, where $M_s$ is the saturation magnetization, $\omega = 2\pi f$ is the angular frequency, $k = 2\pi/\lambda$ is the wavenumber, $H_0$ is the amplitude, and $\beta$ is the ellipticity of the modulation.

**Figure 4.** (a) Sequence of snapshots showing the collective transport of 270 nm particles. At time $t = 7.2$ s the magnetic ratchet is switched on and all the nanoparticles are transported to the right (arrow at bottom) at an average speed $\langle v \rangle = 2.6 \, \mu m \, s^{-1}$ (field parameters $f = 1 \, Hz$, $H_0 = 1200 \, A \, m^{-1}$, $\beta = -1/3$). The green (middle figure) and blue (bottom figure) lines are trajectories of a single particle. Scale bar is $5 \, \mu m$ for all images, see also Video S2 in the Supporting Information. (b,c) Probability distributions of the particle position perpendicular $P(\delta x)$ (b) and parallel $P(\delta y)$ (c) to the BWs. The scattered points are experimental data while the solid lines are Gaussian fits with $\sigma_x^2$ and $\sigma_y^2$ for the variance in the corresponding directions. In (b), the distribution was calculated by subtracting the drift as described in the text.
Being interested in the transport properties across the magnetic stripes and evaluating the magnetic force exerted on the particle, \( F(x,t) = -\partial_t U_m(x,t) \), with \( U_m = -\mu_0 V f (H^0 + H_{sub})^2/2 \), in the overdamped limit we obtain an equation of motion,

\[
\dot{x}(t) = -\zeta \lambda f \left( \sqrt{1 + \beta} \cos \omega t \cos k x \right) + \zeta(t)
\]

with \( F(x,t) = -\zeta \lambda f \left( \sqrt{1 + \beta} \cos \omega t \cos k x \right) \) and a random variable \( \zeta(t) \) taking account of thermal fluctuations. Here, \( f_c = 8 \mu_0 \chi VM/\mu_0 \exp(-kz)/\chi^2 \), and \( \zeta(t) \) obeys the properties \( \langle \zeta(t) \rangle = 0 \), \( \langle \zeta(t) \zeta(t') \rangle = 2d_0 \delta(t - t') \), where \( d_0 \) is the free diffusion coefficient, \( k_0 T \) is the thermal energy, and \( \zeta \) is the friction coefficient. By using \( f_c \) and \( d_0 \) as fitting parameters, we numerically integrate eq 3 and evaluate the velocity of mean drift \( \langle \nu \rangle \) and effective diffusion \( D_{eff} \) which capture well the experimental data.

To gain further insights, we approximate the general model for an arbitrary \( \beta \), eq 3, by a more tractable special case \( \beta = 0 \), which corresponds to a traveling wave potential, \( U_m(x,t) = U_m(x - \nu_0 t) \). In the reference frame comoving with the speed \( \nu_0 = \lambda f_c \) in terms of a new variable \( u(t) = x(t) + \nu_0 t \), we obtain \( u(t) = \lambda f_c \sin ku + \zeta(t) \). The deterministic velocity of mean drift is known to be \( \langle \nu \rangle_{t=0} = \begin{cases} \lambda f_c, & f \leq f_c, \\ \lambda f_c - \sqrt{\nu_0^2 f_c^2 - f_c^2}, & f > f_c \end{cases} \) \( \text{(4)} \)

Here, \( f_c \) plays the role of the critical frequency that separates the low frequency domain with the maximum possible speed of mean drift, \( \langle \nu \rangle = \nu_0 \propto f \) \( (f < f_c) \) from the high frequency domain where its efficiency progressively drops down, \( \langle \nu \rangle \propto f - \nu_0 \) \( (f > f_c) \). For nanoparticles, thermal fluctuations are, however, inevitable, and we consider their thermal motion in the associated potential

\[
\frac{V(u)}{k_0 T} = -\frac{\lambda f_c}{D_0} - \frac{\lambda f_c}{D_0 k} \cos ku
\]

which admits evaluation of the velocity of mean drift and effective diffusion \( \text{eq 5} \)

\[
\langle \nu \rangle = \lambda f_c - \frac{D_0}{k} \frac{1 - \exp \left( -\frac{\lambda f_c}{D_0 k} \right)}{\langle L_u(u) \rangle_n}
\]

\( \text{eq 6} \)

\[
D_{eff} = D_0 \frac{\langle L_u(u) \rangle_n}{\langle L_u(u) \rangle_n}
\]

\( \text{eq 7} \)

Here, \( L_u(u) = \langle \exp \left( \pm (V(u) - V(u) + \nu')/k_0 T) \right) \rangle \) are evaluated for potential \( \text{eq 5} \) and \( \langle \nu \rangle = \lambda^{-1} \int_0^\infty \rangle_t \) denotes the average over one wavelength of the landscape.

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.8b04248.

VideoS1 shows the transport of dilute 270 nm paramagnetic colloidal particles deposited above an FGF. The particles are translated at an average speed of \( \nu = 5.2 \mu m s^{-1} \) by an external rotating magnetic field with frequency \( f = 2 \) Hz, amplitude \( H_0 = 1200 \text{ A/m} \) (ellipticity \( \beta = -1/3 \)). The video corresponds to Figure 1d of the manuscript (AVI).

VideoS2 illustrates the collective transport of 270 nm paramagnetic colloids deposited above an FGF. Initially the particles perform simple Brownian movement across the FGF surface and, after few seconds, the rotating magnetic field is applied, and the particles are transported towards right at an average speed of \( \nu = 2.6 \mu m s^{-1} \). The rotating field has frequency \( f = 1 \) Hz, total amplitude \( H_0 = 1200 \text{ A/m} \) and ellipticity \( \beta = -1/3 \). The video corresponds to Video 4a of the manuscript (AVI).

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**Notes**

The authors declare no competing financial interest.

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