Ferrofluids as Thermal Ratchets

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Colloidal suspensions of ferromagnetic nanoparticles, so-called ferrofluids, are shown to be suitable systems to demonstrate and investigate thermal ratchet behavior: By rectifying thermal fluctuations, angular momentum is transferred to a resting ferrofluid from an oscillating magnetic field without net rotating component. Via viscous coupling the noise driven rotation of the microscopic ferromagnetic grains is transmitted to the carrier liquid to yield a macroscopic torque. For a simple setup we analyze the rotation of the ferrofluid theoretically and show that the results are compatible with the outcome of a simple demonstration experiment.

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To extract directed motion from random fluctuations is a problem at the heart of statistical mechanics with a long history [1–4]. On the one hand it is crucial for basing the second law of thermodynamics on statistical reasoning and for depriving various of Maxwell's demon type devices of their mystique. On the other hand, it is closely linked to practical issues such as the efficiency of heat engines. The problem has gained renewed attention [5–7] due to its possible relevance for biological transport [8,9] and the prospects of nanotechnology [10–12].

A particularly clear example of a so-called *ratchet* is provided by an over-damped particle in a time-dependent on-off spatially sawtooth potential subject to some random noise [13]. In the stationary (on or off) state, thermodynamic equilibrium prevails and detailed balance prohibits directed transport. Complementary, with the potential switched on and off regularly diffusion and relaxation combine to yield a nonzero average particle drift. Other means may be used to drive the system away from equilibrium. In any case a noise driven drift is to be expected generically under nonequilibrium conditions where, however, the specification of the value and even the sign of this drift can be rather subtle [7].

In the present Letter, we show that colloidal suspensions of ferromagnetic nanoparticles, so-called ferrofluids [14], are ideal systems to test theoretical predictions on fluctuation driven transport experimentally. This is due to three main reasons: First, the small size of the ferromagnetic grains (~ 10 nm) implies that their dynamics is strongly influenced by thermal fluctuations [15]. Second, spatially periodic time-dependent potentials for the orientation of the particles can be easily realized by external magnetic fields. Third, directed orientational transport manifests itself as systematic rotation of the ferromagnetic particles which can be easily detected from the resulting macroscopic torque on the carrier liquid. We show that our theoretical considerations are in agreement with a simple demonstration experiment.

Ferrofluids combine the hydrodynamic properties of Newtonian fluids with the magnetic behavior of superparamagnets [14]. Many of their fascinating properties stem from the viscous coupling between the rotation of the magnetic grains and the vorticity of the hydrodynamic flow. A direct way to set this coupling into action is with a *rotating* external magnetic field as used, e.g., to spin up a ferromagnetic drop floating under hydraulic zero-gravity [16,17]. Complementary, a rotational ferrofluid flow exposed to a static magnetic field exhibits an enhanced shear viscosity [18]. Various unusual effects such as "negative" rotational viscosity [19], magnetovortical resonance [20], and anomalously enhanced ac response due to coherent particle rotation [21] rely on the exchange of angular momentum between *rotating* particles and an *oscillating* magnetic field. In these cases the involved nonzero flow vorticity is crucial for explicitly breaking the symmetry between clockwise and counterclockwise particle rotation.

In contrast to these situations we demonstrate the transfer of angular momentum from an *oscillating* magnetic field to a ferrofluid *at rest.* A crucial ingredient of the operating mechanism is a sufficient impact of thermal noise due to random collisions between the ferromagnetic grains and the molecules of the carrier liquid.

We consider a ferrofluid in a horizontal, spatially homogeneous magnetic field $\mathbf{H}$ composed of a constant part H_x parallel to the x axis and an oscillatory part $H_y f(t)$ along the y direction. The precise form of the time modulation $f(t)$ will be fixed below. As is common for diluted ferrofluids [14,15] we model the ferromagnetic particles as noninteracting spherical equal sized dipoles of volume V and magnetic moment $\mathbf{m}$ [22]. The external magnetic field gives rise to a potential $U = -\mathbf{m} \cdot \mathbf{H}$ for the direction of the magnetic moment. This direction in turn is assumed to be tightly coaligned with the orientation of the magnetic particle. A reorientation of the magnetic moment hence requires a rotation of the particle against the viscosity η of the carrier liquid characterized by the Brownian relaxation time $\tau_B = 3\eta V/k_B T$ with T denoting the temperature of the liquid. We use the dimensionless Langevin parameters $\alpha_{x,y} = mH_{x,y}/k_B T$ to measure the magnetic field strengths and scale time by twice the Brownian relaxation time. As time dependence in the ac part of the field we choose

$$f(t) = \cos(\omega t) + a \sin(2\omega t + \beta). \tag{1}$$

The parameters α_x, α_y, a, ω, and β are easily controlled in experiments.

Parametrizing the orientation of a magnetic dipole by two angles, $\mathbf{e} = \mathbf{m}/m = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ the potential U generated by the above specified magnetic field takes the form

$$U(\theta, \phi, t) = -\sin\theta[\alpha_x \cos\phi + \alpha_y f(t) \sin\phi]. \tag{2}$$

Neglecting thermal fluctuations, the particle orientation is governed by an overdamped relaxation dynamics in the force field $-\nabla U$. Accordingly, the orientation $\mathbf{e}(t)$ tends to align with the magnetic field. Since the field is restricted to the x-y plane, $\theta(t) \to \pi/2$ for $t \to \infty$. Within the plane, the angle between the field and the x axis is always less than $\pi/2$, implying the same for $\phi(t)$ when $t \to \infty$; see Fig. 1, i.e., no average rotation or torque can arise.

Things change in the presence of thermal fluctuations causing stochastic transitions between the deterministic solutions. As elucidated qualitatively with Fig. 1 the dynamical asymmetry induced by $\alpha_x \neq 0$ and $a \neq 0$ gives rise to slightly different probabilities for a 2π-phase slip in the "forward-ϕ direction" and "backward-ϕ direction," respectively. This in turn results in an average rotation of the particle, a manifestation of the ratchet effect [5,6,23] for the orientational motion of the ferromagnetic grains. Similar to situations with additive driving [24–27] the spatial symmetry of our potential $U(\theta, \phi, t)$ requires a sufficiently complex time modulation function $f(t)$ for noise induced transport to occur.

A more quantitative analysis of the effect can be made on the basis of the Fokker-Planck equation for the probability distribution $P(\theta, \phi, t)$ of the particle orientation. It is given by (see, e.g., [28])

$$\partial_t P = \nabla(P \nabla U) + \nabla^2 P, \tag{3}$$

where ∇ denotes the angular part of the nabla operator in spherical coordinates.

We have solved this equation numerically by expanding $P(\theta, \phi, t)$ in spherical harmonics. From the solution we can determine the average orientation $\langle \mathbf{e} \rangle = \int d\phi d\theta \sin\theta\, \mathbf{e}\, P(\theta, \phi, t)$ and the average torque $\mathbf{N}_p = m\langle \mathbf{e} \rangle \times \mathbf{H}$ acting upon an individual particle. For symmetry reasons only the z component of the torque is

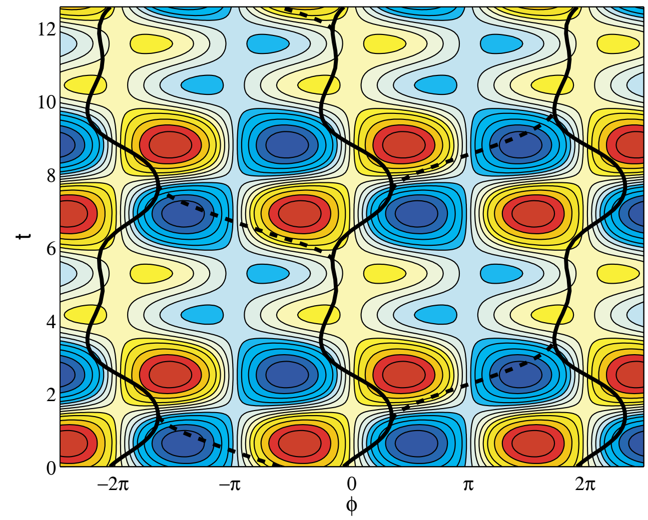

FIG. 1 (color). Space-time plot of the potential $U(\theta = \pi/2, \phi, t)$, from Eq. (2) for $\alpha_x = 0.3$, $\alpha_y = 1$, $a = 1$, $\omega = 1$, and $\beta = 0$. Blue and red regions correspond to small and large values of U, respectively. In the long-time limit, the deterministic dynamics approaches $\theta(t) = \pi/2$ and a periodic $\phi(t)$, represented by either of the full black lines. In the presence of thermal noise, transitions between these deterministic solutions become possible schematically indicated by the dashed lines. The spatial asymmetry and temporal anharmonicity of the potential results in slightly different rates for noise induced increments and decrements of ϕ, respectively. As a result a noise driven rotation of the particles arises.

different from zero. Because of the viscous coupling between the ferromagnetic grains and the carrier liquid the individual torques combine to yield a macroscopic torque per fluid volume

$$\mathbf{N} = n\mathbf{N}_p = \mu_0 M_s \langle \mathbf{e} \rangle \times \mathbf{H} = \mu_0 \mathbf{M} \times \mathbf{H}. \tag{4}$$

Here n denotes the number density of the particles, $\mathbf{M} = M_s\langle \mathbf{e} \rangle$ the magnetization of the ferrofluid, $M_s = mn/\mu_0$ its saturation value, and μ_0 the permeability of free space. A nonzero torque after averaging (4) over one period of the external ac driving is the macroscopic signature of a directed orientational transport.

An explicit expression for the torque can be obtained from an approximate solution of the Fokker-Planck equation (3) by adopting the effective field method [29]. It gives rise to a relaxation equation [30] for the average orientation $\langle \mathbf{e} \rangle$ which for a fluid at rest reduces to

$$\partial_t \langle e_i \rangle = -\sum_j \left[\left(1 - \frac{\langle e \rangle}{\alpha_{\text{eff}}}\right)\delta_{ij} + \left(3\frac{\langle e \rangle}{\alpha_{\text{eff}}} - 1\right)\frac{\langle e_i \rangle}{\langle e \rangle}\frac{\langle e_j \rangle}{\langle e \rangle} \right] \times (\alpha_{\text{eff}} - \alpha)_j, \tag{5}$$

where $\langle e \rangle$ denotes $|\langle \mathbf{e} \rangle|$ and the so-called effective field $\alpha_{\text{eff}}(\langle e \rangle)$ is given by the inverse of the Langevin function $\langle e \rangle = \mathcal{L}(\alpha) = \coth\alpha - 1/\alpha$. The numerical integration of Eq. (5) yields rather accurate approximations for the orientation $\langle e_i \rangle(t)$, which deviate from the numerical solution of the Fokker-Planck equation (3) by less than a few percent. Nevertheless, the final values for the time-averaged magnetic torque (4) differ by a factor between 2 and 3. This is due to the fact that the time average $\overline{N}_z$ is much smaller than the typical values of the time-dependent component N_z of the torque.

For sufficiently weak magnetic field we may use $\mathcal{L}(\alpha) \simeq \alpha/3$ which makes Eq. (5) linear in $\langle \mathbf{e} \rangle$. It is then easily solved for a time dependence of the field α as specified by Eq. (1). The resulting time-averaged torque (4), however, vanishes identically. Exploring the nonlinear regime perturbatively by using $\mathcal{L}(\alpha) \simeq \alpha/3 - \alpha^3/45$ we find to leading order for the time-averaged z component of the torque

$$\overline{N}_z = \mu_0 \frac{M_s^2}{3\chi} \alpha_y^3 \alpha_x \frac{a}{30} \frac{\omega^2(\omega \cos\beta + 2\sin\beta)}{(1+\omega^2)(4+\omega^2)^2}, \quad (6)$$

where χ denotes the magnetic susceptibility of the ferrofluid. This expression provides a useful approximate formula for the nontrivial dependencies of the torque on the parameters of the problem. In particular, it shows that both the static magnetic field component α_x and the anharmonic time dependence (1) of the oscillatory component are essential for directed rotational transport to occur. The effective field method may be a useful tool for the analysis of other ratchet systems as well [31].

To verify our theoretical findings we have designed a simple demonstration experiment as sketched in Fig. 2. The setup is a torsion balance similar to those used in string galvanometers. A hollow plastic sphere with inner diameter 16 mm was filled with a ferrofluid APG 933 (Ferro-Tec), with the following specifications: density $\rho = 1100\ \mathrm{kg/m^3}$, susceptibility $\chi = 1.09$, saturation magnetization $M_s = 18$ kA/m, dynamic viscosity $\eta = 0.1$ Pas. The container was suspended on a Kevlar fiber with a length of 20 cm and 10 μm diameter. The oscillatory magnetic field was generated by a pair of Helmholtz coils of 100 windings each via a computer generated signal of the form specified in Eq. (1). The rms-field strength in the center of the coil amounted to about $H_y \simeq 2.1$ kA/m. The static field component $H_x \simeq 9$ kA/m was generated by a commercial electromagnet (Bruker). The applied frequency was $\nu = 200$ Hz. In the choice of the frequency and the ratio between the amplitudes of the static and oscillating field we were guided by our numerical solutions of the Fokker-Planck equation (3). The dimensionless parameters describing the setup are $\alpha_x = 1.72$, $\alpha_y = 0.4$, $a = 1$. The dimensionless frequency ω depends on the Brownian relaxation time τ_B which was used as a fit parameter.

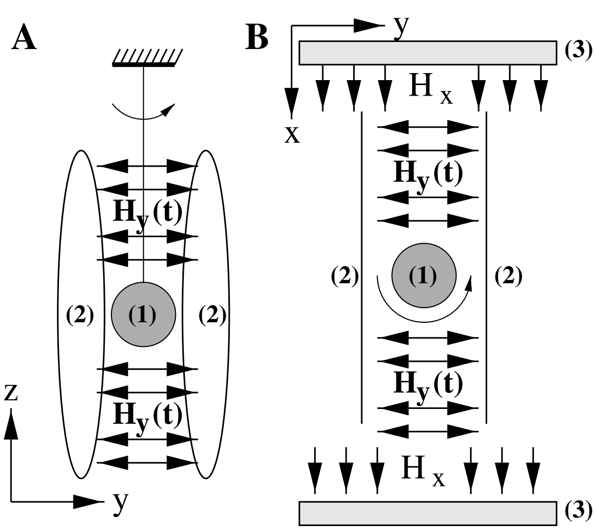

FIG. 2. Sketch of the experimental setup. (A side view, B top view). A hollow plastic sphere (1) is filled with ferrofluid and suspended on a thin Kevlar fiber. The time dependent magnetic field in the y direction is generated with a pair of Helmholtz coils (2), the static field in x direction stems from a commercial electromagnet (3). Noise assisted transfer of angular momentum from the magnetic field to the ferrofluid manifests itself in a rotation of the sphere.

The main qualitative result of the experiment is the unambiguous demonstration of the proposed thermal ratchet effect: After switching on the fields the ferrofluid sphere immediately starts to rotate. Switching off either the static field α_x or the modulation amplitude a the torque disappears. A reversal of the sign of either α_x or a causes a reversal of the sample's rotation sense.

A more quantitative comparison between theory and experiment was difficult for the following reason: Our setup was designed to be as sensitive as possible with respect to the appearance or not of a rotation of the drop. The actual rotation speeds of the sphere then became usually so large that the field had to be switched off after a short time in order not to destroy the experiment. We hence resorted to the following semiquantitative procedure to determine the torque: Starting with the sphere at rest and then letting act the magnetic field for a fixed amount of time (a few seconds), the resulting final rotation speed of the sphere was used as a measure for the torque. In this way we have experimentally recorded the torque (in arbitrary units) as a function of the phase angle β in (1). The results are shown in Fig. 3 together with a fit to our approximate theoretical result given in Eq. (6).

Theory and experiment are compatible if the Brownian relaxation time is chosen as $\tau_B \simeq 1.8$ ms. Using $\tau_B = 3\eta V/k_B T$ this corresponds to a particle diameter of about 35 nm, which exceeds the typical size by about a factor of 3. However, in real ferrofluids size, shape, and magnetic moment of the ferromagnetic grains vary quite significantly (see, e.g., [14], p. 41), a situation only poorly described by our model involving a single relaxation time. Moreover, since the transferred angular momentum increases with the particle diameter [31] the above value for τ_B is likely to characterize the largest grains in the population rather than the average.

In conclusion, we have shown that by rectifying rotational Brownian motion angular momentum can be transferred from an oscillating magnetic field to a ferrofluid at

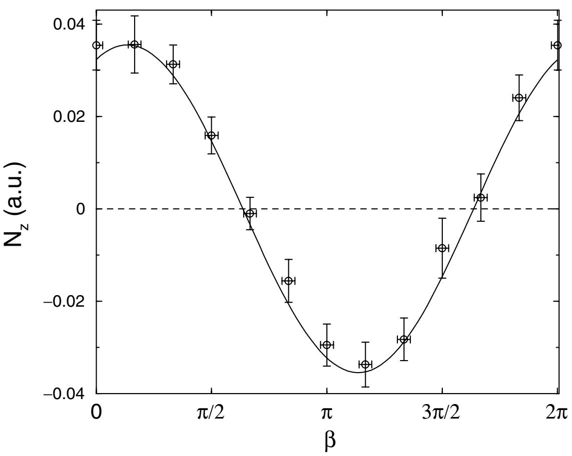

FIG. 3. Magnetic torque transferred to the ferrofluid as a function of the phase angle β for $\alpha_x = 1.72$, $\alpha_y = 0.4$, $a = 1$. The symbols are experimental results obtained with the setup shown in Fig. 2. The solid line is a fit to the approximate analytical expression (6), $N_z = A(\omega \cos\beta + 2\sin\beta)$ with the amplitude A and the frequency ω as fit parameters. The value $\omega \simeq 4.41$ obtained translates into a fit for the Brownian relaxation time of $\tau_B \simeq 1.8$ ms.

rest. A unique feature ferrofluids offer in comparison with other experimental realizations of ratchets is the combined action of many individual nanoscale ratchets to yield a *macroscopic* thermal noise induced transport effect. More precise experiments are needed to verify quantitatively our theoretical predictions about the dependence of the torque on the magnetic field strength and the frequency of the external driving.

Many other investigations using ferrofluids as thermal ratchets are conceivable. A particularly interesting line is linked with the occurrence of inversion points β_i of the rotation; see Fig. 3. When keeping β fixed to such an inversion point and varying instead another parameter of the system the rotational speed of the magnetic grains will generically exhibit a change of sign as a function of this parameter as well. Since $\alpha_{x,y}$ or the dimensionless ω (due to its dependence on τ_B) depend on the size of the magnetic grains in a polydisperse ferrofluid under the same experimental conditions the larger and smaller particles may rotate in opposite directions.

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