THERMAL RATCHET EFFECT IN FERROFLUIDS

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Introduction. The extraction of directed motion from random fluctuations is an old and controversial problem in statistical mechanics with a long and interesting history [1]. Although excluded by the second law of thermodynamics for equilibrium systems, rectification of fluctuations *is* possible in systems driven sufficiently far away from thermal equilibrium [2, 3]. The problem has gained renewed attention under the trademarks of "thermal ratchets" and "Brownian motors" due to its possible relevance for biological transport [4, 5] and the prospects of nano-technology [6, 7, 8].

Ferrofluids are ideal systems to investigate such fluctuation driven transport phenomena, both theoretically and experimentally [9]. The main reasons are the following. First, the magnetic energy of a ferromagnetic particle in a ferrofluid is for typical magnetic fields comparable to its thermal energy at room temperature. Hence, the rotational dynamics of the ferromagnetic grains is strongly influenced by thermal fluctuations [10]. Second, appropriate time-dependent potentials can be easily designed with the help of external magnetic fields. Third, due to the viscous coupling to the carrier liquid the rectification of *microscopic* orientational fluctuations of the ferromagnetic grains manifests itself in a *macroscopic* torque per fluid volume, which can be easily detected experimentally.

In the present contribution we investigate how a suitably designed time dependent external magnetic field *without net rotating component* may rectify fluctuations of the ferrofluid particle orientation and set up a *noise-induced* rotation of the ferromagnetic grains. We will thus show how the angular momentum can be transferred from an *oscillating* magnetic field to a ferrofluid *at rest.* This is a rather indirect and subtle aspect of the interplay between the rotational Brownian motion of ferrofluid particles and their relaxational dynamics in an external magnetic field.

In the present introductory discussion we will use two approximations which simplify the analysis without compromising the effect under consideration. The first is to neglect the Neel-relaxation of the magnetization, *i.e.,* the rotation of the magnetization vector with respect to the ferromagnetic particle. This is justified for particle sizes that are not too small and amounts to assuming that the magnetic moments are firmly attached to the geometry of the particles. Any reorientation of the magnetic moment hence requires a rotation of the particle as a whole. The second approximation is to neglect interactions between the particles. We hence assume a sufficiently diluted ferrofluid and work in a *single particle* picture.

1. Basic equations. We consider the rotational motion of a spherical particle of volume V and magnetic moment $\boldsymbol{\mu}$ immersed in a liquid with a dynamic viscosity η and subjected to a horizontal, time dependent, spatially homogeneous magnetic field $\mathbf{H}$. The field is composed of a constant part H_x parallel to the x-axis and an oscillatory part $H_y(t)$ with period $2\pi/\omega$ along the y-direction

$$\mathbf{H} = (H_x, H_y(t), 0), \qquad H_y(t + 2\pi/\omega) = H_y(t). \tag{1}$$

Different choices for the time dependence of H_y are of interest [11]. Here we will focus on the case

$$H_y(t) = H_y^{(1)} \cos(\omega t) + H_y^{(2)} \sin(2\omega t + \delta) \ , \tag{2}$$

where the amplitudes $H_y^{(1,2)}$, and the phase δ are control parameters. The main features of this time dependence are a zero average over one period, the presence of a higher harmonic of the basic frequency, and a purely oscillatory character, *i.e.,* the absence of any *net rotating component.*

The orientation of the particle at time t is described by the unit vector $\mathbf{e}(t) = \boldsymbol{\mu}(t)/\mu$, where μ denotes the modulus of the magnetic moment. Its time dependence is governed by

$$\partial_t \mathbf{e} = \mathbf{\Omega} \times \mathbf{e} \ , \tag{3}$$

where $\mathbf{\Omega}(t)$ denotes an instantaneous angular velocity of the particle. Changes of $\mathbf{\Omega}$ are due to torques on the particle. Denoting by ∇ the angular part of the three dimensional Nabla operator, the magnetic torque

$$\mathbf{N}_{\text{mag}} = -\mathbf{e} \times \nabla U = \mu \, \mathbf{e} \times \mathbf{H} \tag{4}$$

derives from the potential energy

$$U(\mathbf{e}, t) = -\mu \, \mathbf{e} \cdot \mathbf{H}(t) \tag{5}$$

of a magnetic dipole in an external field [12]. Furthermore, the viscosity η of the carrier liquid gives rise to a viscous torque [13]

$$\mathbf{N}_{\text{visc}} = -6\eta V \mathbf{\Omega} \ . \tag{6}$$

Additionally, the interaction between the rotating particle and the surrounding liquid also causes thermal fluctuations which generate a stochastic torque

$$\mathbf{N}_{\text{stoch}} = \sqrt{12\eta V k_B T} \, \boldsymbol{\xi}(t) \ . \tag{7}$$

Here, $\boldsymbol{\xi}(t)$ is a vector of independent δ-correlated Gaussian noise sources of zero meaning, the noise intensity is related to the temperature T and the dissipation by the fluctuation-dissipation relation, and k_B stands for the Boltzmann's constant. Denoting the moment of inertia of the particle by I, the equation of motion for $\mathbf{\Omega}$ acquires the form

$$I \partial_t \mathbf{\Omega} + 6\eta V \mathbf{\Omega} = \mu \, \mathbf{e} \times \mathbf{H} + \sqrt{12\eta V k_B T} \, \boldsymbol{\xi}(t) \ . \tag{8}$$

Eqs.(3) and (8) form a closed set of equations for the description of the rotational motion of the particle. As is well known [10], for experimentally relevant parameter values the first term on the l.h.s. of eq. (8) is five to seven orders of magnitude smaller than the second one. We may hence safely neglect inertial effects and may work in the overdamped limit

$$\mathbf{\Omega} = \frac{\mu}{6\eta V} \, \mathbf{e} \times \mathbf{H} + \sqrt{2D} \, \boldsymbol{\xi}(t) \ , \tag{9}$$

where we have introduced $D = k_B T / 6\eta V$. Using this result in (3) yields a closed equation for the time evolution of $\mathbf{e}$

$$\partial_t \mathbf{e} = \frac{\mu}{6\eta V} \, (\mathbf{e} \times \mathbf{H}) \times \mathbf{e} + \sqrt{2D} \, \boldsymbol{\xi}(t) \times \mathbf{e} \ . \tag{10}$$

In order to introduce dimensionless units, we measure time in units of the inverse of the external driving frequency, $t \mapsto t/\omega$, use $6\eta V\omega/\mu$ as a unit for the magnetic field strength, $\mathbf{H} \mapsto (6\eta V\omega/\mu)\ \mathbf{H}$, and rescale the noise intensity according to $D \mapsto \omega\ D$. The evolution equation for the orientation $\mathbf{e}$ then reads

$$\partial_t \mathbf{e} = (\mathbf{e} \times \mathbf{H}) \times \mathbf{e} + \sqrt{2D}\ \boldsymbol{\xi}(t) \times \mathbf{e}\ . \tag{11}$$

Introducing the Brownian relaxation time

$$\tau_B = \frac{3\eta V}{k_B T}\ , \tag{12}$$

we note that the rescaled, dimensionless noise intensity D, occurring in (11), just gives the ratio between the relevant deterministic and stochastic time scales in the system:

$$D = \frac{k_B T}{6\eta V\omega} = \frac{1}{2\tau_B\ \omega}\ . \tag{13}$$

To proceed, we parametrize the orientation $\mathbf{e}$ of the magnetic particle by two angles θ and φ according to

$$\mathbf{e} = (\sin\theta\cos\varphi, \sin\theta\sin\varphi, \cos\theta)\ . \tag{14}$$

From (11) we then find the following Langevin equations for the time evolution of these angles

$$\partial_t \theta = -\partial_\theta U + D\cot\theta + \sqrt{2D}\ \xi_\theta(t) \tag{15}$$

$$\partial_t \varphi = -\frac{1}{\sin^2\theta}\partial_\varphi U + \frac{\sqrt{2D}}{\sin\theta}\ \xi_\varphi(t)\ . \tag{16}$$

In the dimensionless units adopted, the noise intensity D is given by (13) and potential (5) takes the form

$$U(\theta, \varphi, t) = -\sin\theta\ (H_x \cos\varphi + H_y(t)\sin\varphi)\ . \tag{17}$$

The thermal fluctuations $\xi_\theta(t)$ and $\xi_\varphi(t)$ are given by two independent, δ-correlated Gaussian noise sources of zero meaning.

The observable of interest in the present investigation is the time and ensemble averaged torque (4) exerted by the magnetic field upon the magnetic particle in the long time limit, *i.e.,* after the initial transient has died out. Since the magnetic field (1) is constrained to the x-y plane, only the z-component of this magnetic torque can be different from zero. Suppressing the subscript $_{\text{mag}}$ from now on, we denote the averaged z-component of the magnetic torque by $\overline{\langle N_z \rangle}$, where $\langle \ldots \rangle$ stands for the ensemble average over the different realizations of the noise terms in (15), (16) and the overbar represents the time average over one period of the magnetic field. Using (1) and (4), we get

$$\overline{\langle N_z \rangle} = \frac{1}{2\pi}\int_0^{2\pi} \mathrm{d}t\ \langle \sin\theta(t)\,(-\,H_x\ \sin\varphi(t) + H_y(t)\ \cos\varphi(t)) \rangle\ . \tag{18}$$

Exploiting (17) and (16), one readily finds the equivalent expressions

$$\overline{\langle N_z \rangle} = \frac{1}{2\pi}\int_0^{2\pi} \mathrm{d}t\ \langle \partial_\varphi U(\theta(t), \varphi(t), t) \rangle = \frac{1}{2\pi}\int_0^{2\pi} \mathrm{d}t\ \langle \partial_t\varphi(t)\ \sin^2\theta(t) \rangle \tag{19}$$

For reasons of ergodicity, the ensemble average in (18) is equivalent to a time average of a single realization over an infinite time interval. Then the extra time average over one period of the external driving drops out and we are left with

$$\overline{\langle N_z \rangle} = \lim_{(t_f - t_i)\to\infty} \frac{1}{t_f - t_i} \int_{t_i}^{t_f} \mathrm{d}t\, \partial_t \varphi(t)\, \sin^2\theta(t) \tag{20}$$

and similarly for the equivalent expressions in (18), (19).

2. Deterministic dynamics. In the absence of thermal fluctuations, the particle orientation is governed by the overdamped deterministic relaxation dynamics given by (15), (16) with $D = 0$. Hence, at any given moment, the orientation $\mathbf{e}(t)$ tends to align with the instantaneous magnetic field $\mathbf{H}(t)$, which lies in the x-y-plane. A careful analysis [14] of the various possible initial conditions reveals that in all cases $\theta(t)$ converges eventually to $\pi/2$ for $t \to \infty$. This in turn implies that $\varphi(t)$ approaches a *periodic* long time behaviour. An explicit example is displayed in Fig. 1.

From (20) we hence infer

$$\overline{\langle N_z \rangle} = \lim_{(t_f - t_i)\to\infty} \frac{\varphi(t_f) - \varphi(t_i)}{t_f - t_i} = 0 \tag{21}$$

and, therefore, in the absence of thermal fluctuations no particle rotation will occur and no average torque can arise. The effect under discussion is hence indeed

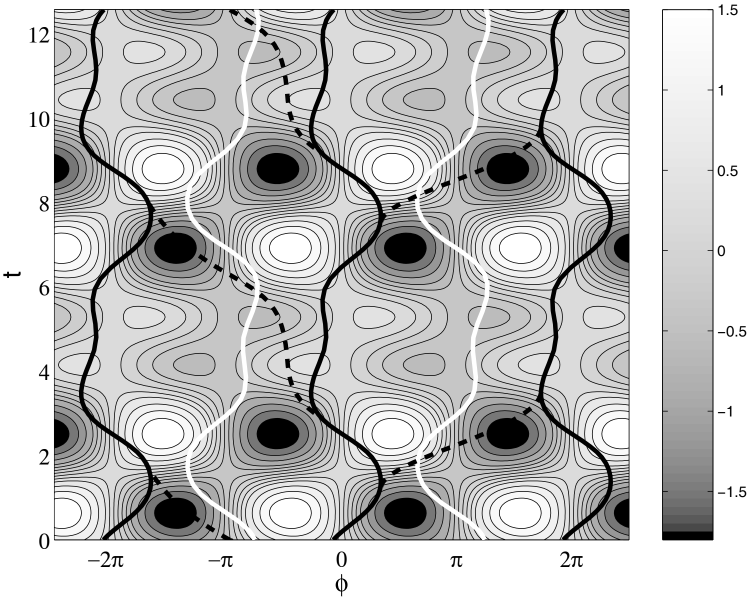

Fig. 1. Space-time plot of the potential $U(\theta = \pi/2, \varphi, t)$, eq.(17), for the time dependence (2) with $H_x = 0.3$, $H_y^{(1)} = H_y^{(2)} = 1$, and $\delta = 0$. Dark and bright regions correspond to small and large values of U, respectively. In the long-time limit, the deterministic dynamics (15), (16) with $D = 0$ approaches $\theta(t) = \pi/2$ and a periodical $\varphi(t)$, oscillating back and forth, as represented by either of the full black lines. In the presence of a small amount of noise, occasional transitions across the unstable deterministic orbits shown as full white lines become possible which are schematically indicated by the dashed lines. The spatial asymmetry and temporal anharmonicity of the potential conspire to yield slightly different rates for noise induced increments and decrements of φ, respectively. As a result, a noise driven rotation of the particles arises.

intimately connected with the presence of thermal fluctuations. Only their interplay with the specifically designed external magnetic field gives rise to the rotation of the ferromagnetic grains in the ferrofluid.

3. Stochastic dynamics. It is easy to understand intuitively why the deterministic situation described above changes fundamentally in the presence of fluctuations. As shown qualitatively in Fig. 1, for small noise intensities, the time dependence of $\varphi(t)$ and $\theta(t)$ will most of the time closely follow the deterministic trajectories. But additionally there is now the possibility for rare, fluctuation induced *transitions* between different deterministic solutions, as qualitatively indicated by the dotted lines in Fig. 1. From the detailed form of the stable and unstable orbits it is plausible that the rates for forward (increasing φ by 2π) and backward transitions (decreasing φ by 2π) will in general be different from each other. Hence, on average a net rotation of the particle will occur, implying with (21) that $\overline{\langle N_z \rangle} \neq 0$. This is a manifestation of the so-called ratchet effect [3], in which an unbiased potential and undirected Brownian fluctuations cooperate to produce directed transport.

The possibility for a noise-induced rotation may be inferred without explicitly solving the equations of motion from a general investigation of the space-time symmetries of the system. A first observation is that at any given instance of time t, potential (17) is "symmetric" in φ, i.e., there exists a $\Delta\varphi(t)$ such that $U(\theta, -\varphi, t) = U(\theta, \varphi + \Delta\varphi(t), t)$. Contrary to several other ratchet devices, our system is, therefore, not characterized by a spatially asymmetric potential but exhibits a *dynamical symmetry breaking* that gives rise to a preferential direction of rotation in the generic case. As discussed in detail in [11], the crucial ingredients for an operation of the ratchet mechanism in the present system is the constant component of the field in the x-direction and the second harmonic in the time dependence of the oscillating field component. In fact, one can show that the induced torque must be an even function of the constant field component in the x-direction and hence $H_x = 0$ implies $\overline{\langle N_z \rangle} = 0$. Moreover, if one can find a Δt such that $H_y(t) = -H_y(t + \Delta t)$, again no average torque can arise. Hence, with a simple sinusiodal time dependence for the field $H_y(t)$ no ratchet effect will occur, which may be the reason why in the experiments reported in [15] no *rotation* of a liquid ferrofluid drop in crossed constant and oscillating magnetic fields was observed.

A more quantitative analysis of the stochastic dynamics builds on the numerical solution of the Fokker–Planck equation equivalent to (15), (16). Expanding the time dependent probability density $P(\theta, \varphi, t)$ in spherical harmonics, accurate results for $\overline{\langle N_z \rangle}$ can be obtained [11]. Among other things it is interesting to study the dependence of the transferred angular momentum on the particle size. Fig. 2 shows how the torque changes with a relative particle size for a special set of parameters and two qualitatively different ways of modifying the size of the particles. It is important to note that in both cases the torque changes the sign when changing the size. For this choice of parameters, we have, hence, a somewhat unusual situation that in a polydisperse ferrofluid the larger ferromagnetic particles rotate clockwise, whereas the smaller ones rotate counter-clockwise. In translational ratchets similar effects of current inversion may be used for size separation. Such an application is less obvious for the rotational ratchet considered here. It should also be noted that the noise induced torques and rotation frequencies are rather small.

4. Phenomenological approach. It has been pointed out that the described effect may be discussed also on the macroscopic level using standard

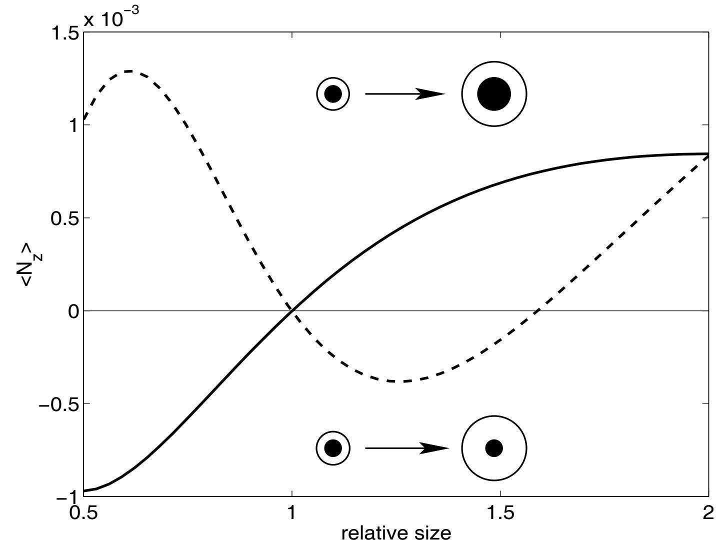

Fig. 2. Dependence of $\overline{\langle N_z \rangle}$ on the relative particle size for $H_x = 0.3$, $H_y^{(1)} = H_y^{(2)} = 1$ and $D = 0.2$, and $\delta = 1.73$. The full line corresponds to a proportional change of the magnetic and hydrodynamic radii (top sketch), the dashed line is for a change of the hydrodynamic radius only (bottom sketch).

phenomenological laws for the magnetic relaxation in ferrofluids [16]. In order to elucidate the relation between microscopic and phenomenological theory in a systematic manner, the Fokker-Planck equation may be solved approximately by expanding its solution in powers of H/D, i.e., in the ratio between magnetic and thermal energy. A standard perturbation theory reveals that the first non-zero result for the time averaged torque $\overline{\langle N_z \rangle}$ is obtained in the fourth order in H/D [11]. The explicit analytical result in this order reads

$$\overline{\langle N_z \rangle}_p = \frac{H_x\,(H_y^{(1)})^2\,H_y^{(2)}}{40}\;\frac{9(1+29D^2)\cos\delta + 2D(1+99D^2)\sin\delta}{(1+D^2)(1+4D^2)(1+9D^2)(1+36D^2)}\;. \tag{22}$$

In accordance with our symmetry considerations, this expression shows that the constant field in the x-direction (i.e., $H_x \neq 0$) as well as the second harmonic in the time dependence of the oscillating field in thr y-direction (i.e., $H_y^{(2)} \neq 0$) are crucial for the rotation of the particles to occur. It is of course impossible to perform the limit $D \to 0$ in this expression since it is correct only to the fourth order in H/D. We hence find that a purely phenomenological description of the effect is indeed possible, as is the case for many other system exhibiting the ratchet effect [3] as well. However, taken on its own, it is less instructive since it hides the microscopic origin of the noise induced particle rotation and masks the indispenibility of thermal fluctuations.

From expression (22) we infer that the sign of $\overline{\langle N_z \rangle}$ is uniquely determined by the values of the parameters and that a macroscopic rotation may occur for arbitrarily small values of the magnetic field. Both features distinguish our effect from the instability of the rest state of a ferrofluid volume in a harmonically oscillating field as investigated recently in a setup similar to ours [17]. In this case a self-sustained rotation of the ferrofluid sample occurs via a Hopf-bifurcation at a critical amplitude of the magnetic field, for which both directions of rotation are equally probable. It would be interesting to investigate the interplay between both effects experimentally and to verify that the inclusion of the higher harmonic in the time dependence as well as of the constant field in the x-direction induces an imperfect Hopf-bifurcation with a preferred rotation direction.

5. Summary. Orientational fluctuations of ferrofluid particles may be rectified by suitably designed time dependent magnetic fields. As a result, angular momentum may be transferred in a systematic way from an oscillatory magnetic field without net rotating component to a ferrofluid at rest. The effect is a realization of the so-called ratchet mechanism and demonstrates that ferrofluids are convenient systems to investigate various properties of Brownian motors, both theoretically and experimentally.

The discussed noise induced rotation is yet another example for the interesting consequences of the subtle interplay between magnetic and rotational degrees of freedom in ferrofluids. Moreover, it shows how investigations of ferrofluids may shed light upon the potentially important operating principles of nano-science.

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