

Role of interactions in ferrofluid thermal ratchets

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(Received 21 September 2006; published 27 March 2007)

Orientational fluctuations of colloidal particles with magnetic moments may be rectified with the help of external magnetic fields with suitably chosen time dependence. As a result a noise-driven rotation of particles occurs giving rise to a macroscopic torque per volume of the carrier liquid. We investigate the influence of mutual interactions between the particles on this ratchet effect by studying a model system with mean-field interactions. The stochastic dynamics may be described by a nonlinear Fokker-Planck equation for the collective orientation of the particles which we solve approximately by using the effective field method. We determine an interval for the ratio between coupling strength and noise intensity for which a self-sustained rectification of fluctuations becomes possible. The ratchet effect then operates under conditions for which it was impossible in the absence of interactions.

DOI: [10.1103/PhysRevE.75.031118](https://doi.org/10.1103/PhysRevE.75.031118)

PACS number(s): 05.40.-a, 82.70.-y, 75.50.Mm, 05.60.-k

I. INTRODUCTION

Thermal ratchets or Brownian motors are devices that are able to rectify thermal fluctuations and thereby extract directed motion from irregular microscopic chaos. Banished from equilibrium by the second law of thermodynamics they may nevertheless be found in various nonequilibrium situations (for a review see [1]). Having been used for a long time as thought models to highlight some subtle points in the foundations of statistical mechanics [2], they recently became objects of more practical interest due to their possible relevance for biological transport mechanisms [3,4] and potential applications in nanotechnology [5–7].

Among the many devices that have been proposed as Brownian motors [6], those based on ferrofluids [8,9] are of particular interest since they allow a rather direct observation of the ratchet effect on the macroscopic level. Ferrofluids are suspensions of ferromagnetic nanoparticles in carrier liquids such as water or oil combining the hydrodynamic behavior of Newtonian fluids with the magnetic properties of superparamagnets [10]. The orientational Brownian motion of the ferromagnetic grains can be rectified with the help of a suitably chosen time-dependent magnetic field. Due to the viscous coupling between the rotation of the magnetic grains and the local vorticity of the hydrodynamic flow the angular momentum produced by the ratchet is transferred to the carrier liquid and may be detected as hydrodynamic torque per fluid volume [8].

Although the interactions between the ferromagnetic particles mediated by the carrier liquid are of crucial importance for the macroscopic manifestation of the ratchet effect it was neglected in the theoretical modeling done so far [8,9]. In the present paper we want to remedy this drawback and elucidate the influence of interactions on the ratchet effect in ferrofluids. This will be done using a simplified mean-field model which allows to analytically discuss some of the

qualitative changes that occur. Although the mean-field approximation is a rather crude description of the intricate direct (dipole-dipole) and indirect (hydrodynamic) interactions between the ferrofluid particles it is a reasonable first step in the analysis and will provide interesting insight into the possible influence of interactions on the ratchet mechanism.

General aspects of the interplay between the ratchet effect and interactions were already discussed in [11] and [1]. In particular, it was shown that under certain conditions the interactions may give rise to a spontaneous breakdown of symmetries that formerly inhibited the ratchet effect. The relevance of interactions in biological applications was investigated in [4].

Here we will find by studying a concrete, experimentally accessible example of a ratchet that depending on the ratio between coupling strength and noise intensity qualitatively different regimes are possible. In some of these regimes a self-sustained ratchet effect may occur under circumstances for which no such effect would show up in the absence of interactions. The discussion will be centered around a nonlinear Fokker-Planck equation for the stochastic dynamics of the collective orientation of the ferrofluid particles [12] which is approximately solved by adapting the well-known effective-field method from the theory of ferrofluids [13] to the analytical investigation of the ratchet effect in these systems.

The paper is organized as follows. In Sec. II we introduce the model and the basic equations and derive the nonlinear Fokker-Planck equation for the collective orientation of the ferromagnetic particles. Section III contains some numerical solutions of this Fokker-Planck equation providing first insight into the different regimes that are possible in the system. In Sec. IV we introduce the effective-field method and show how it may be adapted to the present situation. Using this method we will characterize in Sec. V the different regimes found in Sec. III, determine their stability, and discuss the different manifestations of the ratchet effect. Finally, Sec. VI contains our conclusions.

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II. THE MODEL

We consider the overdamped dynamics of N identical spherical particles with magnetic moments \mathbf{m}_i , $i=1, \dots, N$, dispersed in a Newtonian liquid with viscosity η . The particles are under the influence of a time-dependent external magnetic field with constant x component and time-periodic y component

$$\mathbf{H} = (H_x, H_y(t), 0), \quad H_y\left(t + \frac{2\pi}{\Omega}\right) = H_y(t). \quad (1)$$

A suitable example for the time dependence of the y component is [9]

$$H_y(t) = \alpha \cos(\Omega t) + \beta \cos(2\Omega t + \delta), \quad (2)$$

with the scalar parameters α, β , and δ .

The change of orientation $\mathbf{u}_i = \mathbf{m}_i/m_i$ of particle i is given by $\partial_t \mathbf{u}_i = \boldsymbol{\omega}_i \times \mathbf{u}_i$, where $\boldsymbol{\omega}_i$ denotes the angular velocity the particle.

The particle orientations tend to align with the magnetic field. Since the latter is confined to the x - y plane the average orientations will have zero z components and the main effects will show up in the time dependence of their x and y components. To keep the analysis simple we will therefore only consider the dynamics in the x - y plane and put $u_{i,z} \equiv 0$ for all i . It is then convenient to parametrize the orientations in the form

$$\mathbf{u}_i = \begin{pmatrix} \cos \phi_i \\ \sin \phi_i \\ 0 \end{pmatrix}. \quad (3)$$

In the overdamped limit the angular velocities $\boldsymbol{\omega}_i$ are determined by the balance of torques

$$0 = \mathbf{N}_{\text{magn},i} + \mathbf{N}_{\text{visc},i} + \mathbf{N}_{\text{stoch},i} + \mathbf{N}_{\text{int},i}. \quad (4)$$

The different contributions in this equation denote the magnetic torque due to the interaction with the external magnetic field

$$\mathbf{N}_{\text{magn},i} = m \mathbf{u}_i \times \mathbf{H} = m [H_x \sin \phi_i - H_y(t) \cos \phi_i] \mathbf{e}_z, \quad (5)$$

the viscous and stochastic torques

$$\mathbf{N}_{\text{visc},i} = -6\eta V \boldsymbol{\omega}_i = -6\eta V \partial_t \phi_i \mathbf{e}_z, \quad (6)$$

and

$$\mathbf{N}_{\text{stoch},i} = \sqrt{12\eta V k_B T} \xi_i(t) \mathbf{e}_z, \quad (7)$$

respectively, describing the interaction with the carrier fluid, and the torque arising from the interaction between the particles

$$\mathbf{N}_{\text{int},i} = \frac{K}{N} \sum_{j=1}^N \mathbf{u}_j \times \mathbf{u}_i = \frac{K}{N} \sum_{j=1}^N \sin(\phi_i - \phi_j) \mathbf{e}_z. \quad (8)$$

In the above expressions k_B denotes Boltzmann's constant, T temperature, and V the volume of the particles. The $\xi_i(t)$ are identical and independent Gaussian noise sources with zero mean and correlation $\langle \xi_i(t) \xi_j(t') \rangle = \delta_{ij} \delta(t-t')$. The interaction term (8) is of mean-field type as, e.g., also advocated in

[1,14]. Although the direct magnetic dipole-dipole interaction as well as the indirect hydrodynamic interaction between the particles are much more complicated than the simple assumption (8) several interesting implications of interactions will become apparent already in this mean-field description.

The stochastic dynamics of the system is described by the following set of Langevin equations:

$$\begin{aligned} \partial_t \phi_i = & \frac{1}{6\eta V} \left(m H_x + \frac{K}{N} \sum_{j=1}^N \cos \phi_j \right) \sin \phi_i \\ & - \frac{1}{6\eta V} \left(m H_y(t) + \frac{K}{N} \sum_{j=1}^N \sin \phi_j \right) \cos \phi_i + \sqrt{2D} \xi_i(t) \end{aligned} \quad (9)$$

with the diffusion coefficient D given by

$$D := \frac{k_B T}{6\eta V}. \quad (10)$$

It is convenient to introduce dimensionless quantities by performing the rescalings $t \rightarrow t/\Omega$, $\mathbf{H} \rightarrow 6\eta V \Omega / m \mathbf{H}$, $D \rightarrow \Omega D$, and $K \rightarrow 6\eta V \Omega K$ to obtain

$$\begin{aligned} \partial_t \phi_i = & \left(H_x + \frac{K}{N} \sum_{j=1}^N \cos \phi_j \right) \sin \phi_i \\ & - \left(H_y(t) + \frac{K}{N} \sum_{j=1}^N \sin \phi_j \right) \cos \phi_i + \sqrt{2D} \xi_i(t). \end{aligned} \quad (11)$$

Equivalent to this set of Langevin equations is a Fokker-Planck equation for the joint probability distribution $W(\phi_i, t)$ of the particle orientations.

As usual in a mean-field model we are interested in the limit $N \rightarrow \infty$. It is then useful to introduce the distribution function for particle orientations

$$P(\phi, t) = \frac{1}{N} \sum_{i=1}^N \delta[\phi - \phi_i(t)]. \quad (12)$$

As discussed in detail in [12,15–17] $P(\phi, t)$ becomes for $N \rightarrow \infty$ independent of the specific realization of the noise $\xi_i(t)$ (self-averaging property). Consequently, the same holds true for the collective orientation of the particles defined by

$$\mathbf{S}(t) = \frac{1}{N} \sum_{i=1}^N \mathbf{u}_i(t) = \int_0^{2\pi} d\phi \mathbf{u} P(\phi, t) = \begin{pmatrix} \langle \cos \phi \rangle \\ \langle \sin \phi \rangle \\ 0 \end{pmatrix}, \quad (13)$$

where $\langle \cdot \rangle$ denotes the average with $P(\phi, t)$. We therefore obtain from Eq. (11) N decoupled Langevin equations with the solution $W(\phi_i, t)$ of the equivalent Fokker-Planck equation factorizing, $W(\phi_i, t) = \prod_{i=1}^N w(\phi_i, t)$, where $w(\phi_i, t)$ obeys the single-particle Fokker-Planck equation [18,19]

$$\partial_t w(\phi, t) = \partial_\phi (w(\phi, t) \partial_\phi U(\phi, t)) + D \partial_\phi^2 w(\phi, t). \quad (14)$$

The potential U is defined by

$$\begin{aligned}
 U(\phi, t) &= -\mathbf{u} \cdot [\mathbf{H}(t) + K\mathbf{S}(t)] \\
 &= -[H_x + KS_x(t)]\cos\phi - [H_y(t) + KS_y(t)]\sin\phi,
 \end{aligned} \tag{15}$$

and depends parametrically on $\mathbf{H}(t)$ and $\mathbf{S}(t)$.

We may now average Eq. (12) over the realizations of the $\phi_i(t)$ to find that P obeys the same equation as w ,

$$\partial_t P(\phi, t) = \partial_{\phi} [P(\phi, t) \partial_{\phi} U(\phi, t; \mathbf{H}(t), \mathbf{S}(t))] + D \partial_{\phi}^2 P(\phi, t). \tag{16}$$

This equation is closed by Eq. (13). As characteristic for a mean-field system, we have thus reduced the dynamics of $N \rightarrow \infty$ coupled degrees of freedom to the dynamics of a single degree of freedom in an augmented field

$$\mathbf{H}_{mf}(t) = \mathbf{H}(t) + K\mathbf{S}(t), \tag{17}$$

to be determined *self-consistently*. For a related application of mean-field techniques to ferrofluids also see [20].

For a static field, $\mathbf{H}(t) \equiv \mathbf{H}$, Eq. (16) admits the stationary solution

$$P_{eq}(\phi) = \frac{1}{2\pi I_0(h_{mf})} \exp(h_{mf,x} \cos\phi + h_{mf,y} \sin\phi). \tag{18}$$

Here and in the following $I_n(x)$ denotes the n th Bessel function of complex arguments, $\mathbf{h}_{mf} = \mathbf{H}_{mf}/D$, and h_{mf} denotes the modulus of \mathbf{h}_{mf} . In the absence of an external field, $\mathbf{H} = 0$, the self-consistency condition (13) takes the form

$$\mathbf{S} = \begin{pmatrix} \langle \cos\phi \rangle \\ \langle \sin\phi \rangle \end{pmatrix} = B(kS) \frac{\mathbf{S}}{S}, \tag{19}$$

where $k = K/D$, and we introduced the function

$$B(x) = \frac{I_1(x)}{I_0(x)}, \tag{20}$$

which is monotonously increasing and satisfies

$$B(x) \sim \frac{x}{2} \quad \text{for } x \rightarrow 0, \tag{21}$$

$$B(x) \rightarrow 1 \quad \text{for } x \rightarrow \infty. \tag{22}$$

Equation (19) coincides with the self-consistent equation for the ferromagnetic mean-field x - y model. The dependence of the modulus S of the collective orientation \mathbf{S} on the ratio between interaction and fluctuation strengths is shown in Fig. 1. Note that the direction of \mathbf{S} in the ordered phase is arbitrary. Note also that for short-range interactions the situation is rather different [21,22].

The central quantity of interest in connection with the ratchet effect in the present system is the time and ensemble averaged torque transferred from the magnetic field to the particles [8,9]

$$\overline{\langle \mathbf{N} \rangle} = \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T dt \langle \mathbf{u} \rangle \times \mathbf{H} = - \lim_{T \rightarrow \infty} \frac{1}{T} \int_0^T dt \langle \partial_t \phi \rangle \mathbf{e}_z. \tag{23}$$

In the absence of interactions between the particles the necessary conditions for the ratchet effect to operate, i.e., for

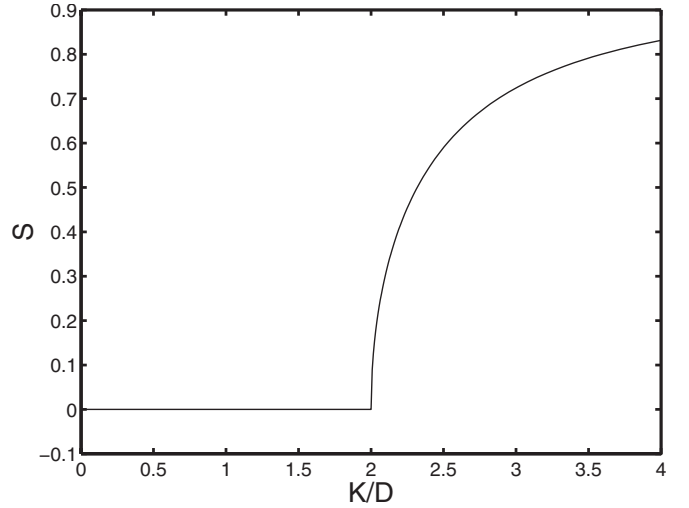


FIG. 1. Modulus S of the collective orientation \mathbf{S} defined in Eq. (13) as a function of the ratio K/D between interaction and disorder strength. In the disordered phase, $K/D < 2$, S is identically zero, for $K/D > 2$ a spontaneous ordering takes place in the mean-field model.

$\overline{\langle N_z \rangle} \neq 0$, have been discussed in detail in [8,23]. In particular it was shown by a symmetry argument that $H_x = 0$ implies $\overline{\langle N_z \rangle} = 0$. In the present mean-field model the role of \mathbf{H} is played by $\mathbf{H}_{mf} = \mathbf{H} + K\mathbf{S}$ and it might therefore be possible to find $\overline{\langle N_z \rangle} \neq 0$ whenever $H_{mf,x} \neq 0$. However, the latter condition may be fulfilled by $H_x = 0$ and $S_x \neq 0$. Hence we may suspect that rectification of fluctuations may take place in the interacting system even under conditions for which it would be impossible in a system without interactions. Note that there is no trivial mapping between the two cases since $\mathbf{H}_{mf,x}$ is time dependent and has to be determined self-consistently. Nevertheless the detailed investigations discussed below will show that the interactions between the particles may indeed give rise to a nonzero value of S_x , which in turn may drive the ratchet effect in the system even if $H_x = 0$.

III. NUMERICAL SOLUTION OF THE FOKKER-PLANCK EQUATION

In the present section we investigate some general features of the dynamics of the system under consideration with the help of a numerical solution of our central equation (16). To this end we expand $P(\phi, t)$ in Fourier modes with respect to ϕ

$$P(\phi, t) = \sum_{n=-\infty}^{\infty} a_n(t) \exp(in\phi), \tag{24}$$

with time-dependent complex expansion coefficients $a_n(t)$. From Eq. (13) we then have

$$S_x(t) = 2\pi \operatorname{Re} a_1(t), \tag{25}$$

$$S_y(t) = -2\pi \operatorname{Im} a_1(t), \tag{26}$$

whereas the average torque is given by

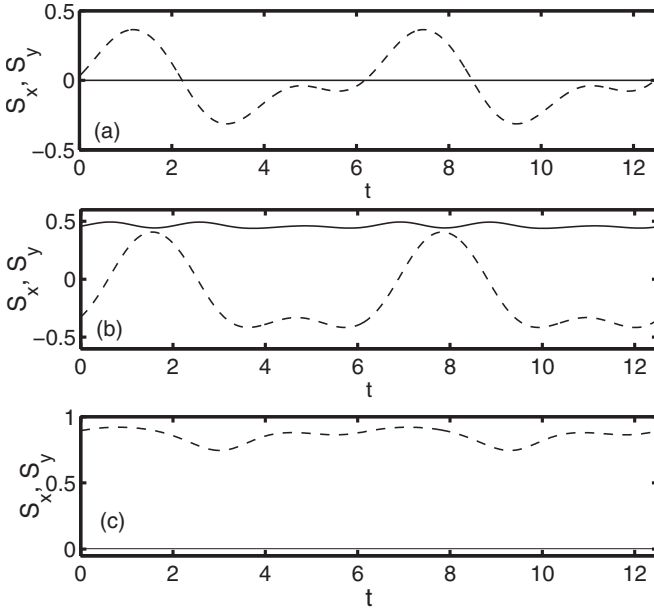


FIG. 2. Typical time dependence of the collective orientation \mathbf{S} in the three different regimes described in the text. Shown are $S_x(t)$ (dashed lines) and $S_y(t)$ (solid lines). The parameter values are $H_x = 0$, $\alpha = \beta = 1/\sqrt{2}$, and $K = 3$. (a) Regime 1: $D = 2.4$, $\overline{\langle N_z \rangle} = 0$. (b) Regime 2: $D = 1.2$, $\overline{\langle N_z \rangle} = 3.5 \times 10^{-5}$. (c) Regime 3: $D = 0.6$, $\overline{\langle N_z \rangle} = 0$.

$$\overline{\langle N_z \rangle} = \overline{H_x \text{Im } a_1 + H_y \text{Re } a_1}. \quad (27)$$

Using the Fourier expansion of $P(\phi, t)$ in Eq. (16), we obtain an infinite system of coupled ordinary differential equations for the coefficients $a_n(t)$ of the form

$$(\partial_t + Dn^2)a_n(t) = \frac{n}{2}[g(t)a_{n-1} - g^*(t)a_{n+1}], \quad (28)$$

where

$$g(t) = H_x - iH_y + 2\pi K a_1(t). \quad (29)$$

Starting with

$$a_0 = \frac{1}{2\pi} \quad (30)$$

as required by normalization of $P(\phi, t)$, we may solve the hierarchy of Eqs. (28) iteratively up to some value n_{max} of n . Using different values for n_{max} we have found that $n_{max} = 10$ is sufficient to obtain accurate numerical results.

As discussed at the end of the previous section, the case $H_x = 0$ is of particular interest. Depending on the ratio between coupling strength and noise intensity we find in this case three different regimes which are characterized by the time dependence of the average orientation $\mathbf{S}(t)$ as shown in Fig. 2. In the first regime, $k < 2$, we find $S_x(t) \equiv 0$ and $S_y(t)$ changes sign during one period of $H_y(t)$. In the second regime, $2 < k \lesssim 3.7$, S_x acquires nonzero values, whereas the behavior of $S_y(t)$ is qualitatively similar to the first case. In the third regime, $3.7 \lesssim k$, we have again $S_x \equiv 0$ but now $S_y(t)$

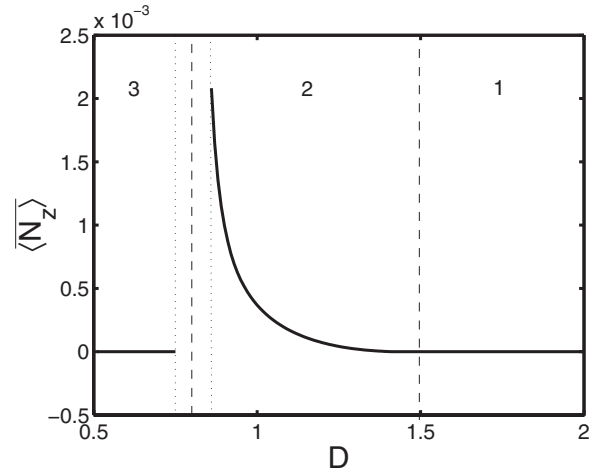


FIG. 3. Numerical results for the time averaged torque $\overline{\langle N_z \rangle}$ for $H_x = 0$ as a function of the disorder strength D for $K = 3$, $\alpha = \beta = 1/\sqrt{2}$, $H_x = 0$. The vertical dashed lines divide the different regimes from each other ($1.5 < D$, regime 1; $0.8 < D < 1.5$, regime 2; $D < 0.8$, regime 3). In the interval indicated by the dotted lines, $0.75 < D < 0.86$, no asymptotic value of $\overline{\langle N_z \rangle}$ could be obtained after 200 periods of driving.

is positive for all t . Similar regimes have been discussed also for the spherical model [24] and the anisotropic x - y model [25] in time-dependent external fields.

As discussed in the previous section the symmetry analysis performed in [8] implies that for $H_x = 0$ and $S_x = 0$ no ratchet effect is possible. Accordingly, using Eq. (27) we find $\overline{\langle N_z \rangle} = 0$ in regimes 1 and 3. In regime 2 we have $S_x \neq 0$ which allows nonzero values of $\overline{\langle N_z \rangle}$. This is indeed what we find with, however, two peculiarities. First, the values for $\overline{\langle N_z \rangle}$ are extremely small as long as k is still near to the value 2 separating region 2 from region 1 (cf. Fig. 3). Second, for values of k around the transition from region 2 to region 3 the system apparently relaxes very slowly to its asymptotic behavior and it becomes very difficult to extract reliable values for $\overline{\langle N_z \rangle}$ from the numerics. The dependence of $\overline{\langle N_z \rangle}$ on the ratio k as obtained numerically is summarized in Fig. 3.

IV. EFFECTIVE FIELD APPROXIMATION

In order to obtain an improved understanding of the numerical results obtained in the previous section we now turn to an approximate analytical analysis of the system. A valuable tool in this respect is the effective field approximation for ferrofluids as extensively reviewed in [13]. To apply this approximation in the present context the following steps are performed. From the central equation (16) we derive an exact equation for the time evolution of $\langle \mathbf{u} \rangle$ of the form

$$\partial_t \langle \mathbf{u} \rangle + D \langle \mathbf{u} \rangle = -D \langle \mathbf{u} (\mathbf{u} \cdot \mathbf{h}_{mf}) \rangle + D \mathbf{h}_{mf}, \quad (31)$$

where $\mathbf{h}_{mf} = \mathbf{H}_{mf}/D$ is determined by Eq. (17). In order to decouple the higher moment on the right-hand side of Eq. (31) we approximate the unknown distribution $P(\phi, t)$ by an *instantaneous equilibrium distribution* of the form (18) corresponding to a so far undetermined *effective magnetic field* $\mathbf{H}_e(t) = D \mathbf{h}_e$,

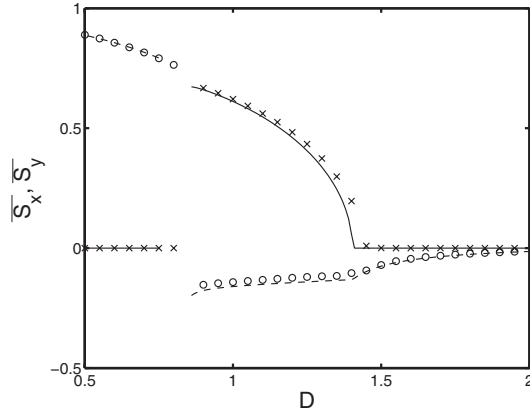


FIG. 4. Time averaged values of the x (solid lines and crosses) and y component (dashed lines and circles) of the average orientation \mathbf{S} as a function of the noise strength D . The other parameter values are as in Fig. 2. Lines are numerically exact results from the Fokker-Planck equation (16) whereas symbols denote the results obtained from the numerical solution of the effective field equations (36) and (37).

$$P(\phi, t) \simeq \frac{1}{2\pi I_0(h_e)} \exp[h_{e,x} \cos(\phi) + h_{e,y} \sin(\phi)]. \quad (32)$$

Using this distribution to calculate the averages in Eq. (31) and using [cf. Eq. (19)]

$$\mathbf{S} = B(h_e) \frac{\mathbf{h}_e}{h_e}, \quad (33)$$

the following evolution equation for the effective field $\mathbf{h}_e(t)$ may be derived

$$\begin{aligned} \partial_t \left[B(h_e) \frac{\mathbf{h}_e}{h_e} \right] = & - \frac{D[h_e - 2B(h_e)]}{h_e^3} \mathbf{h}_e \times (\mathbf{h}_e \times \mathbf{h}_{mf}) \\ & - D \frac{B(h_e)}{h_e} (\mathbf{h}_e - \mathbf{h}_{mf}). \end{aligned} \quad (34)$$

With the help of

$$\mathbf{h}_{mf} = \mathbf{h} + kB(h_e) \frac{\mathbf{h}_e}{h_e}, \quad (35)$$

which follows from Eqs. (17) and (33), and representing \mathbf{h}_e by modulus and phase according to $\mathbf{h}_e = h_e(\cos \varphi, \sin \varphi)$ we find the following closed set of nonlinear ordinary differential equations for the time evolution of the effective field:

$$\partial_t h_e = - \frac{DB(h_e)}{h_e - B(h_e) - h_e B^2(h_e)} [h_e - kB(h_e) - h_y(t) \sin \varphi], \quad (36)$$

$$\partial_t \varphi = \frac{D[h_e - B(h_e)]}{h_e B(h_e)} h_y(t) \cos \varphi. \quad (37)$$

These equations cannot be solved analytically, however their numerical solution is much simpler than the numerical solution of the Fokker-Planck equation (16). As shown in Fig. 4,

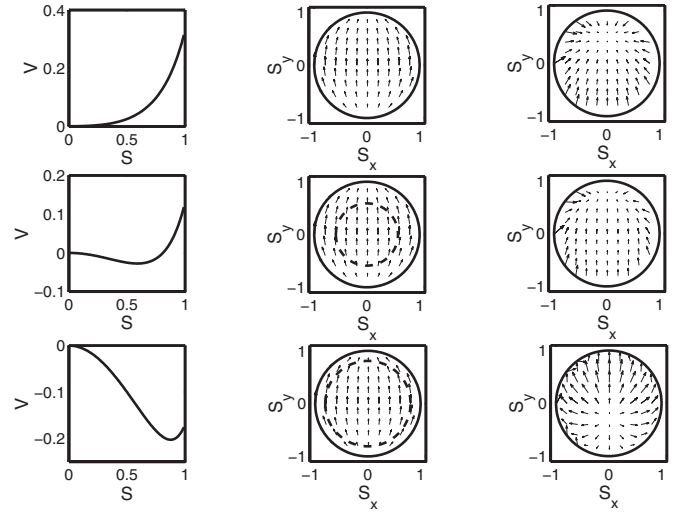


FIG. 5. Plot of the different terms contributing to the right-hand side of Eq. (38) for $H_x=0$, $K=3$ and a time t for which $H_y(t)=1$. The first line corresponds to $D=1.6$ (regime 1), the second one to $D=1.2$ (regime 2), and the third one to $D=0.6$ (regime 3). Shown are the potential $V(S)$ defined in Eq. (39) in the left column, the external force field \mathbf{F} defined in Eq. (39) in the middle column, and the total resulting force field in the right column. The dashed line in the middle column marks the equilibrium value S_{eq} .

the results provide rather accurate estimates for the relevant quantities.

V. DIFFERENT MANIFESTATIONS OF THE RATCHET EFFECT

With the help of the effective field approximation it is possible to gain a more intuitive understanding of the existence of different regimes of the system behavior as found in Sec. III. Moreover, their stability as well as the operation of the ratchet effect in the different regimes may be elucidated.

Using the evolution equation for the effective field, Eqs. (36) and (37), and the relation (33) between the average collective orientation \mathbf{S} and the effective field, we may write the time evolution of \mathbf{S} in the form

$$\partial_t \mathbf{S} = -\nabla V(\mathbf{S}) + \mathbf{F}(\mathbf{H}, \mathbf{S}). \quad (38)$$

The right-hand side of Eq. (38) was split into a central potential term incorporating the effects of diffusion and interaction and an external force field related to $\mathbf{H}(t)$. Note that $0 \leq S \leq 1$ always.

Let us first discuss the potential part. It is given by

$$V(S) = \frac{D}{2} S^2 - K \int_0^S dS' \frac{S'^2}{B^{-1}(S')}, \quad (39)$$

where B^{-1} denotes the inverse function of B defined in Eq. (20). Hence B^{-1} is linear for small values of S and tends to infinity for $S \rightarrow 1$ [cf. Eqs. (21) and (22)]. For small values of k the potential $V(S)$ therefore has a minimum at $S=0$, whereas for $k > 2$ it develops a nontrivial minimum at $S_{eq} > 0$ (cf. Fig. 5, left column). From Eqs. (35) and (37) the

corresponding equilibrium value of the effective field satisfying

$$B(h_e) = S_{eq} \quad (40)$$

is determined by

$$h_{eq} = kB(h_{eq}). \quad (41)$$

We note that with increasing k , S_{eq} also increases (see Fig. 5). In the absence of an external magnetic field, $H=0$, the stationary solution S_{eq} of the effective field equations coincides with the *exact* equilibrium solution of the Fokker-Planck equation (16).

For the external force field we obtain

$$\mathbf{F}(t) = \frac{S}{B^{-1}(S)} H_y(t) \sin \varphi \mathbf{e}_r + \left(1 - \frac{S}{B^{-1}(S)}\right) H_y(t) \cos \varphi \mathbf{e}_\varphi. \quad (42)$$

For small values of S this field is parallel to the external magnetic field \mathbf{H} . For $S \rightarrow 1$ on the other hand the radial component in \mathbf{F} is suppressed and only the tangential term survives (cf. Fig. 5, middle column). This is quite intuitive since for $S \rightarrow 1$ all magnetic moments are aligned and therefore the external field induces identical changes to their orientations giving rise to a change in the *orientation* of \mathbf{S} only.

For small values of the external magnetic field S will only slightly differ from its equilibrium value S_{eq} . We will hence find $S \approx 0$ for small values of k which was characteristic of regime 1. Larger values of k result in values of S differing substantially from zero. From Eq. (42) it is then clear that $\varphi = \pi/2$ is a possible steady state since in this case \mathbf{F} is parallel to \mathbf{e}_y and there is hence no systematic force deflecting \mathbf{S} from the orientation along the y axis. This situation corresponds to regime 3. Finally, there is also a steady-state solution with $S > 0$ and $\varphi \approx 0$ (regime 2), which is, however, not obvious from Eq. (42).

We will now give a simple argument for the transition between regime 2 and regime 3 which allows one to derive an estimate for the threshold value of k separating these two regimes. Since the potential part in Eq. (38) only influences the modulus of S , the time evolution of the angle φ is solely given by

$$\partial_t \varphi = \frac{1}{S} F_\varphi. \quad (43)$$

Assume now that at some given point of time we have $0 < \varphi(t) < \pi/2$. As long as $H_y(t)$ is positive (as in Fig. 5) \mathbf{S} will be pushed in positive y direction (cf. Fig. 5, right column) implying a slight increase of S beyond the value S_{eq} . Similarly, when somewhat later $H_y(t)$ changes sign and becomes negative \mathbf{S} will be pushed in negative y direction and S will slightly decrease. Simultaneously φ changes according to Eq. (43), i.e., increases when $H_y(t) > 0$ and decreases when $H_y(t) < 0$. However, the *rate* of change depends on $S(t)$. A rough estimate on whether the increase of φ dominates over its decrease or vice versa may be obtained from the quantity

$$J(\varphi) = \left. \frac{\partial}{\partial S} \left(\frac{F_\varphi}{H_y(t) S} \right) \right|_{S=S_{eq}}. \quad (44)$$

If $J(\varphi) < 0$ the decrease of φ will dominate, if $J(\varphi) > 0$ its increase. Remarkably, although J depends on φ its *sign* depends only on the ratio k . Hence either φ decreases all the time until it reaches $\varphi=0$ and we are in regime 2 or it increases up to $\varphi=\pi/2$ corresponding to regime 3. An explicit calculation yields

$$J = \frac{k^2(h_{eq}^2 + k + 1 - k^2)}{h_{eq}^2(k^2 - k - h_{eq}^2)}. \quad (45)$$

As shown in the Appendix, the denominator is positive for all values $k > 2$. The condition for the stability of regime 2, $J < 0$, is hence

$$h_{eq}^2 + k + 1 - k^2 < 0, \quad (46)$$

which together with Eq. (41) yields that regime 2 is stable if

$$B(\sqrt{k^2 - k - 1}) < \frac{\sqrt{k^2 - k - 1}}{k}, \quad (47)$$

or equivalently,

$$k < 3.75 \dots \quad (48)$$

As we will see in Secs. V B and V C a more systematic investigation of the stability of regimes 2 and 3 will give rise to the same result [cf. Eqs. (82) and (97)].

A. Regime 1

Regime 1, characterized by $k < 2$, is the most relevant for ferrofluids. There is no spontaneous collective orientation, i.e., $S=0$, and, accordingly, we must have $H_x \neq 0$ in order to find a noise-induced rotation. The situation is hence similar to the case without interaction, nevertheless we will show that the interactions bring about a strongly reinforced ratchet effect.

To obtain a quantitative estimate of this reinforcement we use a variant of the perturbation theory for small values of the external field introduced in [8]. To keep track of the different orders in the expansion it is convenient to use the rescaling

$$\mathbf{H} \rightarrow \epsilon \mathbf{H}. \quad (49)$$

We then solve the Fokker-Planck equation (16) using the expansion

$$P = P^{(0)} + \epsilon P^{(1)} + \epsilon^2 P^{(2)} + \epsilon^3 P^{(3)} + \dots, \quad (50)$$

with the unperturbed solution given by

$$P^{(0)} = \frac{1}{2\pi}. \quad (51)$$

Using Eq. (24) this *Ansatz* gives rise to a similar expansion for the coefficients a_n with $n > 0$,

$$a_n = \epsilon a_n^{(1)} + \epsilon^2 a_n^{(2)} + \dots, \quad (52)$$

whereas a_0 is to all orders in ϵ fixed by the normalization condition to

$$a_0 = \frac{1}{2\pi}. \quad (53)$$

The peculiarity of the first regime is that $S \rightarrow 0$ when $H \rightarrow 0$. We may therefore consistently employ a similar expansion for the collective orientation \mathbf{S} ,

$$\mathbf{S} = \epsilon \mathbf{S}^1 + \epsilon^2 \mathbf{S}^2 + \dots. \quad (54)$$

The first nonzero result for $\overline{\langle N_z \rangle}$ is obtained in fourth order in ϵ . From Eq. (27) we infer that we hence need a_1 up to third order. Similar to [8] we find to first order in ϵ

$$\left(\partial_t + D - \frac{K}{2} \right) a_1^{(1)} = \frac{H_x - iH_y(t)}{4\pi}, \quad (55)$$

and to second order

$$\left(\partial_t + D - \frac{K}{2} \right) a_1^{(2)} = 0, \quad (56)$$

$$(\partial_t + 4D)a_2^{(2)} = (H_x - iH_y)a_1^{(1)} + 2\pi K a_1^{(1)} a_1^{(1)}. \quad (57)$$

In the first regime we have $D - K/2 > 0$. From Eq. (56) we then obtain $a_1^{(2)}(t) \rightarrow 0$ for $t \rightarrow \infty$. To third order we find

$$\left(\partial_t + D - \frac{K}{2} \right) a_1^{(3)} = -\frac{H_x + iH_y}{2} a_2^{(2)} - \pi K a_{-1}^{(1)} a_2^{(2)}. \quad (58)$$

Using the special time dependence (2) and solving for the asymptotic behavior of $a_1^{(1)}$ and $a_1^{(3)}$, we find for the torque (27) after some algebra,

$$\begin{aligned} \overline{\langle N_z \rangle} = 6H_x \alpha^2 \beta \left\{ \frac{(128D^5 K - 64D^4 K^2 + 40D^3 K - 20D^2 K^2 + 8DK - 2K^2) \sin \delta}{(16D^2 + 1)(4D^2 - 4DK + K^2 + 4)^2 (2D - K)(4D^2 - 4DK + K^2 + 16)(4D^2 + 1)} \right. \\ \left. + \frac{(184D^5 + 68D^4 K + 58D^3 K^2 + 200D^3 + 8D^2 K - 3D^2 K^3 + 16D + 6DK^2 + 4K) \cos \delta}{(16D^2 + 1)(4D^2 - 4DK + K^2 + 4)^2 (2D - K)(4D^2 - 4DK + K^2 + 16)(4D^2 + 1)} \right\}. \quad (59) \end{aligned}$$

For $K=0$ this expression simplifies to [8]

$$\overline{\langle N_z \rangle} = \frac{3H_x \alpha^2 \beta}{8} \frac{(23D^2 + 2) \cos \delta}{(4D^2 + 1)(D^2 + 4)(D^2 + 1)(16D^2 + 1)}, \quad (60)$$

which coincides with the result found in [8] when specialized to the x - y plane, i.e., to $\theta \equiv \pi/2$. Figure 6 shows the torque as a function of the phase angle δ for different values of the interaction strength K . It is clearly seen that although the interaction is in regime 1 too weak to induce qualitative changes in the behavior the value of the noise-induced torque is greatly enhanced by the interaction between the particles.

B. Regime 2

In regime 2 the interactions between the particles give rise to a nonzero value of the x component S_x of the collective orientation \mathbf{S} . This breaks the symmetry $x \mapsto -x$ even in the absence of an x component of the external field, $H_x=0$, and results in an interaction sustained ratchet effect in the system. In the present section we derive an approximate solution of the effective field equations (36) and (37) characteristic for this regime and discuss its stability.

To obtain an approximate analytical solution valid in the long-time limit we resort again to perturbation theory and assume that $h=H/D$ is much smaller than the equilibrium value h_{eq} of the effective field h_e . This assumption can easily be met if we are sufficiently far from the boundary of regime 2, $k=2$, at which h_{eq} tends to zero. To formally organize the perturbation expansion it is again convenient to employ the

rescaling (49) and to write the solution of the effective field equations (36) and (37) as power series in ϵ

$$h_e = h_e^{(0)} + \epsilon h_e^{(1)} + \epsilon^2 h_e^{(2)} + \dots, \quad (61)$$

$$\varphi = \varphi^{(0)} + \epsilon \varphi^{(1)} + \epsilon^2 \varphi^{(2)} + \dots, \quad (62)$$

where $h_e^{(0)} = h_{eq}$ and $\varphi^{(0)}$ is not specified at this stage. Using these *Ansätze* in Eqs. (36) and (37) and matching powers of ϵ we find to first order

$$\partial_t h_e^{(1)} = D \frac{k^2 - h_{eq}^2 - 2k}{k^2 - h_{eq}^2 - k} h_e^{(1)} + Dk \frac{\sin \varphi_0}{k^2 - h_{eq}^2 - k} h_y(t), \quad (63)$$

$$\partial_t \varphi^{(1)} = D \frac{(k-1) \cos(\varphi_0)}{h_{eq}} h_y(t). \quad (64)$$

These equations can be solved for arbitrary φ_0 . The second-order equations are rather long and will not be displayed in full generality. However, as typical for degenerate perturbation theory, the second-order equations may contain secular terms. In the present case this happens unless either $\varphi_0 = \pm \pi/2$ or $\int_0^{2\pi} h_y(t) h_e^{(1)}(t) dt = 0$. The first condition corresponds to regime 3 to be discussed in the next section. The second one is equivalent to $\varphi_0=0$ as follows from Eq. (63) and is therefore the condition appropriate for the present investigation of regime 2. Using $\varphi_0=0$ and the specific form (2) of the time dependence of $H_y(t)$, the first-order solution tends for large t to

$$h_e^{(1)} = 0, \quad (65)$$

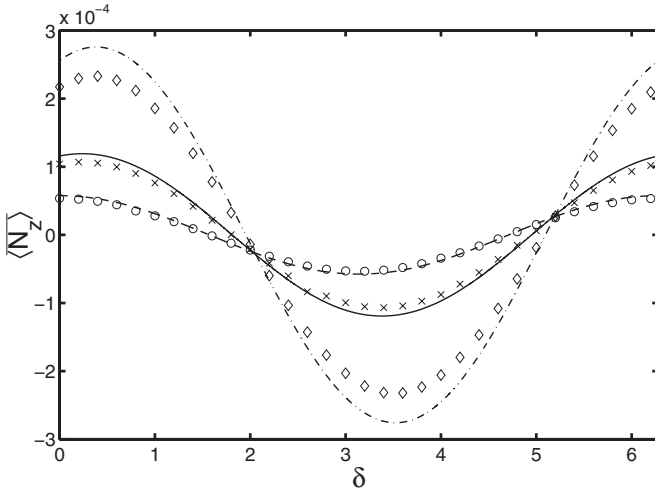


FIG. 6. The time averaged torque in regime 1 as function of the phase δ in the time dependence (2) for different values of the coupling strength K . The symbols are results from the numerical solution of the Fokker-Planck equation (16), the lines show the results of the perturbation theory outlined above. The parameter values are $H_x = \alpha = \beta = 1/\sqrt{6}$, $D = 1.6$, and $K = 0$ (dashed line and circles), $K = 0.5$ (solid line and crosses), and $K = 1$ (dashed-dotted line and diamonds), respectively.

$$\varphi^{(1)} = \frac{k-1}{h_{eq}} \left(\alpha \sin(t) - \frac{\beta}{2} \cos(2t) \right) + \varphi_m, \quad (66)$$

where φ_m is an integration constant. Using these results the second-order equations greatly simplify and take the form

$$\partial_t h_e^{(2)} = -A h_e^{(2)} + B h_y(t) \varphi^{(1)}, \quad (67)$$

$$\partial_t \varphi^{(2)} = 0, \quad (68)$$

with

$$A = D \frac{k^2 - 2k - h_{eq}^2}{h_{eq}^2 + k - k^2}, \quad (69)$$

$$B = -\frac{Dk}{h_{eq}^2 + k - k^2}. \quad (70)$$

Hence $\varphi^{(2)}$ is a constant and $h_e^{(2)}$ may easily be calculated for all values of φ_m . However, this solution produces secular terms in the third-order equations unless

$$\int_0^{2\pi} dt h_y(t) h_e^{(2)}(t) = 0, \quad (71)$$

implying

$$\varphi_m = \frac{3\beta\alpha^2(A^2 + 2)}{4(\alpha^2 A^2 + \beta^2 A^2 + 4\alpha^2 + \beta^2)} \frac{k-1}{h_{eq}}. \quad (72)$$

This result for φ_m completes the first-order solution of the effective field equations. Higher orders may be obtained along the same lines but will not be considered here.

For the collective orientation we then obtain to first order in the external field

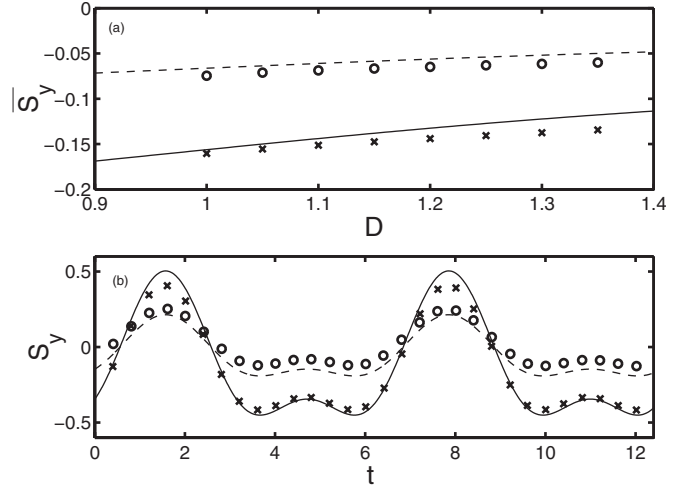


FIG. 7. (a) Time average of the y component S_y of the collective orientation S in regime 2 as a function of the noise strength D for interaction parameter $K=3$. Symbols are numerical results from the Fokker-Planck equation, lines show perturbative results from the effective field equations. The solid line and the crosses are for $\alpha = \beta = 1/\sqrt{2}$, the dashed line and the circles are for $\alpha = \beta = 0.3$. (b) Time evolution of the y component of the orientation vector S in regime 2. The noise intensity is $D = 1.2$, all other parameter values are as in (a).

$$S_x = \frac{h_{eq}}{k}, \quad (73)$$

$$S_y = \frac{h_{eq}\varphi^{(1)}}{k}. \quad (74)$$

From Eq. (66) we hence infer that the time average of S_y is directly related to φ_m ,

$$\overline{S_y} = \frac{h_{eq}\varphi_m}{k}. \quad (75)$$

Figure 7 compares the first-order results obtained above with numerically exact results from the Fokker-Planck equation (16) for different parameter values. The agreement is rather good showing that both parameter sets for α and β correspond to external field strengths well within the region of validity of the perturbation theory.

We now turn to the investigation of the stability of the first-order solution given by Eqs. (65), (66), and (72). To this end we add small deviations δh_e and $\delta\varphi$ to this solution and study their time evolution by linearizing Eqs. (36) and (37) around $h_e^{(1)}(t), \varphi^{(1)}(t)$ [26]. In this way we find

$$\partial_t \begin{pmatrix} \delta h_e \\ \delta\varphi \end{pmatrix} = \begin{pmatrix} a_{11} & -a_{12}h_y(t) \\ a_{21}h_y(t) & 0 \end{pmatrix} \begin{pmatrix} \delta h_e \\ \delta\varphi \end{pmatrix}, \quad (76)$$

where

$$a_{11} = D \frac{h_{eq}^2 + 2k - k^2}{h_{eq}^2 + k - k^2} = -A, \quad (77)$$

$$a_{12} = \frac{Dk}{h_{eq}^2 + k - k^2} = -B, \quad (78)$$

$$a_{21} = D \frac{k + h_{eq}^2 + 1 - k^2}{h_{eq}^2}. \quad (79)$$

We are interested in the stability of the fixed point $\delta h_e = \delta\varphi = 0$ of this system. Because of the periodic time dependence of the coefficient matrix in Eq. (76), a general analysis requires Floquet theory [26]. However, the special form of Eq. (76) allows us to use a more direct method.

We first note that one can show (see the Appendix) $a_{11} < 0$ within regime 2. We next define the function

$$V(\delta h_e, \delta\varphi) = \frac{a_{21}}{2} (a_{21} \delta h_e^2 + a_{12} \delta\varphi^2). \quad (80)$$

For its time derivative we find

$$\frac{dV}{dt} = a_{21} a_{11} h_e^2 < 0. \quad (81)$$

Moreover if

$$a_{21} a_{12} > 0, \quad (82)$$

we have for all values of δh_e and $\delta\varphi$

$$V(\delta h_e, \delta\varphi) \geq 0, \quad (83)$$

with equality being valid only for $\delta h_e = \delta\varphi = 0$. $V(\delta h_e, \delta\varphi)$ is hence a Lyapunov function [27] of the system (76) and the point $\delta h_e = \delta\varphi = 0$ is stable as long as Eq. (82) holds. From Eq. (78) one finds $a_{12} < 0$ (see the Appendix) within regime 2 and hence the first-order solution $h_e^{(1)}(t), \varphi^{(1)}(t)$ is stable if $a_{21} < 0$. From Eq. (79) it is seen that this condition is identical with Eq. (46). We hence find again that regime 2 is stable if

$$2 < k \leq 3.75. \quad (84)$$

Note that the entire stability analysis performed above is valid only to first order in the external field strength.

C. Regime 3

Similarly to the previous section we are now using the effective field equations (36) and (37) to derive an approximate analytical solution typical for regime 3 and to discuss its stability. In regime 3 we have $\varphi = \pi/2$ (the case $\varphi = -\pi/2$ can be dealt with similarly) and we find from Eqs. (36) and (37),

$$\partial_t h_e = - \frac{DB(h_e)}{h_e - B(h_e) - h_e B^2(h_e)} [h_e - kB(h_e) - h_y(t)], \quad (85)$$

$$\partial_t \varphi = 0. \quad (86)$$

From the second equation we obtain immediately $\varphi \equiv \pi/2$. The first equation determines $h_e(t)$ and cannot be solved in closed form. Focusing again on the situation of small exter-

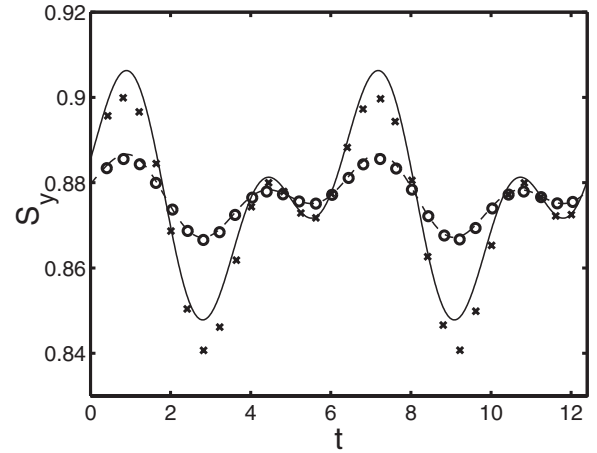


FIG. 8. Time evolution of the y component of the orientation vector S in regime 3. Symbols are numerical results from the Fokker-Planck equation, lines show perturbative results from the effective field equations. The parameters are $D=0.6$ and $K=3$, as well as $\alpha=\beta=0.3$ (crosses and full line) and $\alpha=\beta=0.1$ (circles and dashed line), respectively.

nal fields we use Eq. (61) and obtain for the first-order term the equation

$$\partial_t h_e^{(1)} = -Ah_e^{(1)} + Bh_y(t), \quad (87)$$

with A and B defined by Eqs. (69) and (70), respectively. The solution is given by

$$h_e^{(1)} = B e^{-At} \int dt h_y(t) e^{At}. \quad (88)$$

For the special time dependence (2) we find for large t

$$h_e^{(1)} = B\alpha \frac{A \cos(t) + \sin(t)}{D(A^2 + 1)} + B\beta \frac{A \sin(2t) - \cos(2t)}{D(A^2 + 4)}. \quad (89)$$

In Fig. 8 this result is compared with the solution of the Fokker-Planck equation showing again good agreement for the parameter values chosen.

We now turn to the investigation of the stability of the first-order solution. In view of the form of the potential $V(S)$ in regime 3 (see Fig. 5, first column, last line) it is reasonable to assume that the state will be destabilized by perturbations in φ . Restricting ourselves to this case we use $h_e(t)$ as determined above, set $\varphi(t) = \pi/2 + \delta\varphi(t)$, and determine the linearized time evolution of $\delta\varphi(t)$. From Eq. (37) we obtain

$$\partial_t \delta\varphi = -D \frac{h_e - B(h_e)}{h_e B(h_e)} h_y(t) \delta\varphi, \quad (90)$$

with solution

$$\delta\varphi(t) = \delta\varphi_0 \exp\left(-D \int_0^t dt' \frac{h_e - B(h_e)}{h_e B(h_e)} h_y\right). \quad (91)$$

Since both h_y and h_e are 2π -periodic functions of time this solution may be written in the form

$$\delta\varphi(t) = \delta\varphi_0 e^{-a_0 t} f(t), \quad (92)$$

where $f(t)$ is also 2π periodic and a_0 is the constant term in the Fourier expansion of the exponent in Eq. (91) and therefore given by

$$a_0 = \frac{D}{2\pi} \int_0^{2\pi} dt \frac{h_e - B(h_e)}{h_e B(h_e)} h_y. \quad (93)$$

Using the first-order solution $h_e(t) = h_{eq} + h_e^{(1)}(t)$, we find

$$a_0 = D \frac{(k+1 - k^2 + h_{eq}^2)}{2\pi h_{eq}^2} \int_0^{2\pi} dt' h_e^{(1)}(t') h_y(t'). \quad (94)$$

The integral in this expression is in regime 2 and in regime 3 always positive as can be shown by expanding $H_y(t)$ in a Fourier series

$$h_y = \sum_{n=-\infty}^{\infty} b_n e^{int} \quad (95)$$

and using Eq. (88). We then find

$$\frac{1}{2\pi} \int_0^{2\pi} dt h_e^{(1)}(t) h_y(t) = \sum_{n=0}^{\infty} \frac{B}{n^2 + A^2} |b_n|^2 \geq 0, \quad (96)$$

since $B > 0$ as demonstrated in the Appendix. Hence the sign of a_0 as given by Eq. (94) depends solely on the prefactor and the solution is unstable if

$$k + 1 - k^2 + h_{eq}^2 < 0. \quad (97)$$

This condition is again identical with Eq. (47) and we hence find that regime 3 becomes unstable at the same value of k at which regime 2 becomes stable.

VI. CONCLUSIONS

In the present paper we have theoretically analyzed the influence of particle-particle interactions of the mean-field type on the ratchet effect in ferrofluids. We have used the simplifying mean-field model because the details of the realistic dipole-dipole and hydrodynamic interactions are too complicated for a general discussion. On the other hand, several qualitative effects which may be expected to show up in case of these realistic interactions can already be discussed within the mean-field approach.

Quite generally we have found that interactions that favor the parallel alignment of the magnetizations of the ferrofluid particles reinforce the ratchet effect. In the present situation this means that the magnetic torque per volume of the ferrofluid is enhanced. Although intuitive this result is by no means trivial since the coupling between the ferromagnetic grains is also likely to reduce their orientational fluctuations, which are the driving force of the ratchet effect. This reinforcement of the ratchet effect may be expected for general interactions and is no artefact of the mean-field approximation. The corresponding results derived in Sec. V A will be used for the comparison between our theory with ongoing experimental work, in particular for the discussion of the dependence of the transferred torque on the volume fraction

of ferromagnetic particles in the ferrofluid [28].

For sufficiently strong coupling our model shows a spontaneous symmetry breaking with respect to the collective orientation of the grains. This in turn may bring about a ratchet effect for situations in which it could not operate in the absence of interactions. In the present system this happens for a purely oscillating external magnetic field without constant component in the x direction. In this case and without interactions no ratchet effect is possible due to symmetry reasons whereas a spontaneous breaking of the relevant symmetry caused by the interactions may induce a rectification of fluctuations.

For even stronger coupling strength between the particles this self-sustained ratchet effect again disappears. The particle orientations are now highly aligned and closely follow the direction of the external field. Fluctuations are therefore suppressed and no rectification is possible anymore.

The possibility of ferromagnetic order in ferrofluids is presently debated [20]. It may therefore be possible that the above effects resulting from the spontaneous symmetry breaking are due to our implementation of the mean-field approximation and do not show up in models with a more realistic description of the interactions. On the other hand the investigation of a self-sustained ratchet effect in ferrofluids may hence contribute to decide whether orientational long-range order in ferrofluids is possible or not.

Our analysis builds on the nonlinear Fokker-Planck equation (16) for the collective orientation (13) of the ferrofluid particles. A first insight into the behavior of the system is gained from a numerical solution of this equation. We then use a variant of the effective field method, which is a well-known tool in the theory of ferrofluids, to obtain approximate analytical results for the transitions between and stability of the different regimes of operation of the ferrofluid ratchet.

The central parameter distinguishing the different regimes is the ratio k between the dimensionless coupling constant K and the intensity of the fluctuations D . For $k < 2$ no self-sustained ratchet effect is possible since the interactions are too weak to induce a spontaneous symmetry breaking. For $2 < k < 3.75$, a self-sustained ratchet effect may be observed with a wide spectrum of values for the transferred angular momentum. For $3.75 < k$ the self-sustained ratchet effect disappears again due to an instability in the steady-state solution for the collective orientation of the particles.

ACKNOWLEDGMENT

We would like to thank Dirk Rannacher for clarifying discussions.

APPENDIX

In this appendix we determine the signs of some expressions needed to determine the stability of regimes 2 and 3. In these regimes we have $k > 2$ the value of S_{eq} is determined by the nontrivial minimum of the potential $V(S)$. From Eq. (39) we find by differentiation

$$k = \frac{B^{-1}(S_{eq})}{S_{eq}}, \quad (\text{A1})$$

verifying that S_{eq} depends only on k . Using the properties of the function $B(x)$ defined in Eq. (20) one can show that for all $S \in [0, 1]$,

$$1 - \frac{S}{B^{-1}(S)} \geq S^2. \quad (\text{A2})$$

This in turn implies

$$k^2 - k - k^2 S_{eq}^2 \geq 0 \quad (\text{A3})$$

and using Eq. (40) we find

$$k^2 - k - h_{eq}^2 \geq 0. \quad (\text{A4})$$

Due to Eqs. (70) and (78) this immediately implies

$$B = -a_{12} = -\frac{Dk}{h_{eq}^2 + k - k^2} \geq 0. \quad (\text{A5})$$

Using again the properties of the function $B(x)$ one can also show

$$1 - 2\frac{S}{B^{-1}(S)} \leq S^2. \quad (\text{A6})$$

Similar manipulations as used above then yield for $k > 2$,

$$k^2 - 2k - h_{eq}^2 \leq 0, \quad (\text{A7})$$

from which by using Eqs. (69) and (77) we find

$$A = -a_{11} = \frac{D(k^2 - 2k - h_{eq}^2)}{h_{eq}^2 + k - k^2} \geq 0. \quad (\text{A8})$$

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