

Stochastic resonance in a suspension of magnetic dipoles under shear flow

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We show that a magnetic dipole in a shear flow under the action of an oscillating magnetic field displays stochastic resonance in the linear response regime. To this end, we compute the classical quantifiers of stochastic resonance, i.e., the signal to noise ratio, the escape time distribution, and the mean first passage time. We also discuss the limitations and role of the linear response theory in its applications to the theory of stochastic resonance.

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I. INTRODUCTION

The dynamics of periodically driven stochastic systems has been an active field of research in recent years [1]. This kind of system arises frequently in the fields of physics, chemistry, and biology. Examples are found in problems involving transport at the cellular level [2,3], optical and electronic devices [4], and signal transduction in neuronal tissue [5,6], to cite just a few.

A particularly interesting phenomenon, occurring in periodically driven nonlinear noisy systems, is stochastic resonance (SR) [7]. This term refers to the enhancement of the response of the system to a coherent signal when the intensity of the noise grows, instead of the degradation that one naively expects. The mechanism leading to this phenomenon is quite simple. Imagine a system that exhibits an energetic activation barrier. In the presence of noise, the system can be assumed to surmount this barrier with a rate proportional to $e^{-\Delta E/D}$, where ΔE is the height of the barrier and D is the intensity of the noise acting on the system. The inverse of this rate defines the average waiting time $T(D)$ between two noise-induced transitions. In the presence of a periodic forcing, the height of the barrier is periodically raised and lowered. When the period of the external force synchronizes with $2T(D)$, the barrier surmounting will be enhanced by the cooperative effect of the noise and the periodic forcing.

Although originally proposed for systems in a double-well potential, this original scheme has been extended. In fact, it is known that SR is exhibited by several classes of monostable system, among which one might mention excitable and threshold systems [8–11] or systems that do not follow an activated dynamics but a relaxational dynamics [12,13].

In this paper we will show that a magnetic dipole immersed in a shear flow exhibits stochastic resonance when a weak oscillating magnetic field is acting on it. The presence of this flow takes the system out of equilibrium causing certain peculiarities in the behavior of the system. In order to treat this problem we will analyze the response of the system in the linear regime. A previous study of the dynamics of a dipole under an oscillating magnetic field has revealed that linear response theory (LRT) predicts a monotonically decreasing behavior for the ratio between the output signal and the output noise or signal-to-noise ratio (SNR), i.e., for very

weak applied fields the dipole does not exhibit SR [13]. In the present case there is a new ingredient, absent in [13]: the presence of the shear flow, which is the determinant for many interesting aspects of the dynamics of this system. Additionally, although we show the existence of SR in the linear regime, we discuss the limitations and role of LRT in its application to the theory of SR, mainly related to questions concerning the fluctuation-dissipation theorem.

The paper is organized as follows. In Sec. II we analyze the dynamics of a dipole in a shear flow and find the fixed points. Section III is devoted to studying the response of the system to an oscillating magnetic field by computing the susceptibility. In Sec. IV we calculate the power spectrum and the signal-to-noise ratio. In Sec. V we compute the escape time distribution and from it the mean first passage time. Finally, in Sec. VI we discuss our results.

II. DYNAMICS OF A DIPOLE IN A SHEAR FLOW: FIXED POINTS AND THEIR STABILITY

We consider a dilute colloidal suspension of ferromagnetic dipolar spherical particles, with magnetic moment $\vec{m} = m_s \hat{R}$, where \hat{R} is a unit vector accounting for the orientation of the dipole; the magnetic moment is therefore rigidly attached to the particles. Each dipole is under the influence of a shear flow with vorticity $\vec{\Omega} = 2\omega_0 \hat{z}$, with \hat{z} being the unit vector along the z axis, and of an oscillating field $\vec{H} = H e^{-i\omega t} \hat{x}$, with \hat{x} being the unit vector along the x axis. The dynamics of these dipoles is governed by the following equation of motion:

$$I \frac{d\vec{\Omega}_p}{dt} = \vec{m} \times \vec{H} + \xi_r \left(\frac{1}{2} \vec{\Omega} - \vec{\Omega}_p \right), \quad (1)$$

where I is the moment of inertia of the particles, $\xi_r = 8\pi\eta_0 a^3$ is the rotational friction coefficient, η_0 the solvent viscosity, and a the radius of the particle. For $t \gg \tau_r$, with $\tau_r = I/\xi_r$ being the inertial time scale, the motion of the particle enters the overdamped regime. This time scale defines a cutoff frequency $\omega_r = \tau_r^{-1}$, such that the condition for overdamped motion is equivalent to $\omega \ll \omega_r$. In this case Eq.

(1) yields the balance condition between the magnetic and hydrodynamic torques acting on each particle,

$$\vec{m} \times \vec{H} + \xi_r \left(\frac{1}{2} \vec{\Omega} - \vec{\Omega}_p \right) = 0, \quad (2)$$

which, together with the rigid rotor evolution equation

$$\frac{d\hat{R}}{dt} = \vec{\Omega}_p \times \hat{R}, \quad (3)$$

leads to the dynamic equation for \hat{R} ,

$$\frac{d\hat{R}}{dt} = \omega_0 \{ \hat{z} + \lambda(t) (\hat{R} \times \hat{x}) \} \times \hat{R}. \quad (4)$$

Here $\lambda(t) \equiv (m_s H / \xi_r \omega_0) e^{-i\omega t}$, with $\vec{\Omega}_p$ being the angular velocity of the particle.

The computation of the fixed points of Eq. (4) when the magnetic field is held constant, i.e., $\lambda(t) = \lambda_0 = \text{const}$, and their linear stability analysis are given in detail in Appendix A. After some algebra Eq. (4) becomes

$$\frac{d\hat{R}}{dt} = \omega_0 [\hat{z} \times \hat{R} + \lambda \hat{x} - \lambda \hat{R} (\hat{R} \cdot \hat{x})]. \quad (5)$$

For $\lambda_0 \geq 1$, this equation has only a linearly stable stationary state. The orientation of the suspended particles is fixed to

$$\hat{R}_s = \sqrt{1 - \lambda^{-2}} \hat{x} + \lambda^{-1} \hat{y}. \quad (6)$$

This means that in this regime the hydrodynamic torque, which tends to cause the rotation of the particles, is insufficient to overcome the magnetic torque, which maintains their constant orientation.

For $\lambda_0 < 1$, which is the case we are interested in, the particles undergo a rotation around a fixed axis lying in the y - z plane, the director of this axis being given by

$$\hat{R}_s = \pm \sqrt{1 - \lambda^2} \hat{y} + \lambda \hat{z}. \quad (7)$$

In this case the hydrodynamic torque is strong enough to make the dipole precess around the orientation \hat{R}_s , Eq. (7) (see Appendix A).

III. RESPONSE TO AN OSCILLATING MAGNETIC FIELD

The analysis of Sec. II was carried out for the deterministic dynamics of a magnetic dipole in a shear flow. Fluctuations are introduced by means of a Brownian torque. The corresponding Langevin equation is

$$\frac{d\hat{R}}{dt} = \omega_0 \left\{ \lambda(t) (\hat{R} \times \hat{x}) + \hat{z} + \frac{1}{\xi_r \omega_0} (\hat{R} \times \vec{F}_B(t)) \right\} \times \hat{R}, \quad (8)$$

where $\vec{F}_B(t)$ is a Gaussian white noise of zero mean and correlation function

$$\langle \vec{F}_B(t) \vec{F}_B(t') \rangle = 2 \xi_r k_B T \delta(t - t'). \quad (9)$$

The Fokker-Planck equation corresponding to Eq. (8) is given by

$$\partial_t \Psi(\hat{R}, t) = [\mathcal{L}_0 + \lambda(t) \mathcal{L}_1] \Psi(\hat{R}, t), \quad (10)$$

where \mathcal{L}_0 and \mathcal{L}_1 are operators defined by

$$\mathcal{L}_0 = -\omega_0 \hat{z} \cdot \vec{\mathcal{R}} + D_r \vec{\mathcal{R}}^2, \quad (11a)$$

$$\mathcal{L}_1 = 2\omega_0 \hat{R} \cdot \hat{x} - \omega_0 (\hat{R} \times \hat{x}) \cdot \vec{\mathcal{R}}, \quad (11b)$$

with $D_r = k_B T / \xi_r$ being the rotational diffusion coefficient and $\vec{\mathcal{R}} = \hat{R} \times \partial / \partial \hat{R}$ the rotational operator. Notice that the first and second terms on the right hand side of Eq. (11a) correspond to convective and diffusive terms, respectively. Moreover, Eq. (10) which, according to Eq. (8), rules the Brownian dynamics in the case of overdamped motion, is valid in the diffusion regime. This regime is also characterized by the condition $t \gg \tau_r$, or equivalently $\omega \ll \omega_r$, which implicitly involves the white noise assumption.

To solve the Fokker-Planck equation (10) we will assume that $\lambda_0 \equiv |\lambda(t)|$ constitutes a small parameter such that this equation can be solved perturbatively. Thus up to first order in λ the solution of the Fokker-Planck equation (10) is

$$\Psi(\hat{R}, t) = e^{(t-t_0)\mathcal{L}_0} \Psi^0(t_0) + \int_{t_0}^t dt' \lambda(t') e^{(t-t')\mathcal{L}_0} \mathcal{L}_1 \Psi_0(t'). \quad (12)$$

Here $\Psi_0(t') = e^{(t'-t_0)\mathcal{L}_0} \Psi^0(t=t_0)$ is the zero order solution at time t' , and

$$\Psi^0(\hat{R}, t=t_0) = \delta(\hat{R} - \hat{R}_0), \quad (13)$$

with \hat{R}_0 being an arbitrary initial orientation. As follows from Eq. (11a), the unperturbed operator \mathcal{L}_0 is composed of the operators \mathcal{R}_z and \mathcal{R}^2 , which are proportional to the orbital angular momentum operators of quantum mechanics L_z and L^2 , respectively, and, therefore, their eigenfunctions are the spherical harmonics [14]

$$\mathcal{R}_z Y_{lm}(\hat{R}) = im Y_{lm}(\hat{R}), \quad (14a)$$

$$\mathcal{R}^2 Y_{lm}(\hat{R}) = -l(l+1) Y_{lm}(\hat{R}). \quad (14b)$$

Given that we know how $\vec{\mathcal{R}}$ acts on the spherical harmonics, it is convenient to expand the initial condition in series of these functions, since the spherical harmonics constitute a complete set of functions that are a basis in the Hilbert space of the integrable functions over the unit sphere [15]:

$$\Psi^0(\hat{R}, t_0) = \delta(\hat{R} - \hat{R}_0) = \sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\hat{R}_0) Y_{lm}(\hat{R}). \quad (15)$$

Using this expansion in Eq. (12), for the first order correction to the probability density $\Delta\Psi \equiv \Psi - \Psi_0$, we obtain

$$\Delta\Psi(\hat{R}, t) = \sum_{l=0}^{\infty} \sum_{m=-l}^l \int_{t_0}^t dt' \lambda(t') Y_{lm}^*(\hat{R}_0) \times e^{(t-t')\mathcal{L}_0} \mathcal{L}_1 e^{(t'-t_0)\mathcal{L}_0} Y_{lm}(\hat{R}). \quad (16)$$

Notice that the integral of $\Delta\Psi(\hat{R}, t)$ over the entire solid angle is zero, in agreement with the fact that the unperturbed solution $\Psi_0(\hat{R}, t)$ is normalized.

Since we are interested in the asymptotic behavior we will set $t_0 \rightarrow -\infty$. In this limit, Eq. (12) becomes

$$\Psi(\hat{R}, t) = \frac{1}{4\pi} \left\{ 1 + \int_{-\infty}^t dt' \lambda(t') e^{(t-t')\mathcal{L}_0} 2\hat{R} \cdot \hat{x} \right\}, \quad (17)$$

where now

$$\Delta\Psi(\hat{R}, t) = \frac{1}{4\pi} \int_{-\infty}^t dt' \lambda(t') e^{(t-t')\mathcal{L}_0} 2\hat{R} \cdot \hat{x}, \quad (18)$$

and

$$\Psi^0(\hat{R}, t) = \frac{1}{4\pi} \quad (19)$$

is the uniform distribution function on the unit sphere.

From Eq. (18) the contribution of the ac field to the mean value of the orientation vector \hat{R} can be obtained as

$$\hat{R}(t) = \int d\hat{R} \hat{R} \Delta\Psi = \frac{1}{4\pi} \int_{-\infty}^t dt' \lambda(t') \int d\hat{R} \hat{R} e^{(t-t')\mathcal{L}_0} 2\hat{R} \cdot \hat{x}. \quad (20)$$

This equation can be written in the more compact form

$$\hat{R}_i(t) = \int_{-\infty}^t dt' \lambda(t') \chi_i(t-t'), \quad (21)$$

where the response function [16] has been defined as

$$\chi_i(\tau) = \frac{1}{4\pi} \int d\hat{R} \hat{R}_i e^{\tau\mathcal{L}_0} 2\hat{R} \cdot \hat{x} \quad (22)$$

for $\tau > 0$.

By causality, we can write $t \rightarrow \infty$ in the upper limit of the integral in Eq. (21); hence, this equation becomes

$$\hat{R}_i(t) = \chi_i(\omega) \lambda(t), \quad (23)$$

where $\chi_i(\omega)$ is the generalized susceptibility, which is the Fourier transform of $\chi_i(\tau)$,

$$\chi_i(\omega) = \frac{1}{4\pi} \int_{-\infty}^{\infty} d\tau e^{i\omega\tau} \int d\hat{R} \hat{R}_i e^{\tau\mathcal{L}_0} 2\hat{R} \cdot \hat{x}. \quad (24)$$

From this equation we obtain the components of the susceptibility:

$$\chi_x(\omega) = \frac{1}{3} \left\{ \left[\frac{2D_r}{4D_r^2 + (\omega - \omega_0)^2} - i \frac{(\omega_0 - \omega)}{4D_r^2 + (\omega - \omega_0)^2} \right] + \left[\frac{2D_r}{4D_r^2 + (\omega + \omega_0)^2} + i \frac{(\omega_0 + \omega)}{4D_r^2 + (\omega + \omega_0)^2} \right] \right\}, \quad (25)$$

$$\chi_y(\omega) = \frac{1}{3} \left\{ \left[\frac{(\omega_0 - \omega)}{4D_r^2 + (\omega - \omega_0)^2} + i \frac{2D_r}{4D_r^2 + (\omega - \omega_0)^2} \right] + \left[\frac{(\omega_0 + \omega)}{4D_r^2 + (\omega + \omega_0)^2} - i \frac{2D_r}{4D_r^2 + (\omega + \omega_0)^2} \right] \right\}, \quad (26)$$

$$\chi_z(\omega) = 0. \quad (27)$$

The quantities χ_x and χ_y have poles at $\omega = \pm \omega_0 \pm 2D_r i$. The inverse of the imaginary part of these poles $(2D_r)^{-1}$ defines the Brownian relaxation time.

IV. POWER SPECTRUM

In order to discern whether or not SR is present in the relaxation process of the system under consideration we compute the power spectrum, which, following the Wiener-Khinchine theorem, is given by the Fourier transform of the correlation function [17,1]. Since we will take as output signal the projection of \hat{R} parallel to the magnetic field, i.e., \hat{R}_x , we compute only the correlation function of this quantity.

The correlation function of \hat{R}_x is defined by

$$\begin{aligned} \langle \hat{R}_x(t) \hat{R}_x(t+\tau) | \hat{R}_0(t_0) \rangle \\ = \int d\hat{v} \int d\hat{u} \hat{u}_x \hat{v}_x \Psi(\hat{v}, t; \hat{u}, t+\tau | \hat{R}_0, t_0), \end{aligned} \quad (28)$$

where the initial condition is taken as $\Psi(\hat{R}, t_0) = \delta(\hat{R} - \hat{R}_0)$. The above quantity can be calculated from the solution of the Fokker-Planck equation simply by recalling the following properties of a Markov process [17]:

$$\begin{aligned} \Psi(\hat{v}_1, t_1; \dots; \hat{v}_n, t_n) \\ = \Psi(\hat{v}_1, t_1) \Psi(\hat{v}_1, t_1 | \hat{v}_2, t_2; \dots; \hat{v}_n, t_n), \end{aligned}$$

$$\Psi(\hat{v}_1, t_1 | \hat{v}_2, t_2; \dots; \hat{v}_n, t_n) = \Psi(\hat{v}_1, t_1 | \hat{v}_2, t_2). \quad (29)$$

where $t_1 > t_2 > \dots > t_n$. By combination of these two properties Eq. (28) becomes

$$\begin{aligned}
& \langle \hat{R}_x(t) \hat{R}_x(t+\tau) | \hat{R}_0(t_0) \rangle \\
&= \int d\hat{v} \hat{v}_x \Psi(\hat{v}, t | \hat{R}_0, t_0) \int d\hat{u} \hat{u}_x \Psi(\hat{u}, t + \tau | \hat{v}, t).
\end{aligned} \tag{30}$$

To proceed further, we compute first the integral over \hat{u} in Eq. (30). From Eq. (12), if $\tau > 0$,

$$\begin{aligned}
\Psi(\hat{u}, t + \tau | \hat{v}, t) &= e^{\tau \mathcal{L}_0} \delta(\hat{u} - \hat{v}) + \int_t^{t+\tau} ds \lambda(s) \\
&\quad \times e^{(t+\tau-s)\mathcal{L}_0} \mathcal{L}_1 e^{s\mathcal{L}_0} \delta(\hat{u} - \hat{v}) \\
&= \Psi_0(\hat{u}, t + \tau | \hat{v}, t) + \Delta\Psi(\hat{u}, t + \tau | \hat{v}, t);
\end{aligned} \tag{31}$$

thus we have

$$\begin{aligned}
\int d\hat{u} \hat{u}_x \Psi(\hat{u}, t + \tau | \hat{v}, t) &= \int d\hat{u} \hat{u}_x \Psi_0(\hat{u}, t + \tau | \hat{v}, t) \\
&\quad + \int d\hat{u} \hat{u}_x \Delta\Psi(\hat{u}, t + \tau | \hat{v}, t).
\end{aligned} \tag{32}$$

The results of these integrals are

$$\begin{aligned}
\int d\hat{u} \hat{u}_x \Psi_0(\hat{u}, t + \tau | \hat{v}, t) &= -\sqrt{\frac{2\pi}{3}} [e^{-(2D_r - i\omega_0)\tau} Y_{11}(\hat{v}) \\
&\quad + e^{-(2D_r + i\omega_0)\tau} Y_{1-1}(\hat{v})],
\end{aligned} \tag{33a}$$

$$\begin{aligned}
& \int d\hat{u} \hat{u}_x \Delta\Psi(\hat{u}, t + \tau | \hat{v}, t) \\
&= -\sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\hat{v}) \int_t^{t+\tau} ds \lambda(s) \int d\hat{u} \\
&\quad \times \sqrt{\frac{2\pi}{3}} \{ [e^{-(2D_r - i\omega_0)(t+\tau-s)} Y_{11}(\hat{u}) \\
&\quad + e^{-(2D_r + i\omega_0)(t+\tau-s)} Y_{1-1}(\hat{u})] e^{-[l(l+1)D_r + im\omega_0]s} \\
&\quad \times \mathcal{L}_1 Y_{lm}(\hat{u}) \}.
\end{aligned} \tag{33b}$$

(for the detailed derivation, see Appendix B).

After introducing these expressions into Eq. (30) we obtain three terms corresponding to an expansion of the correlation function in powers of $\lambda(t)$, of zeroth, first, and second order, respectively. The presence of this driving yields an explicit dependence of the correlation function on the time t , instead of its depending only on the time difference, as occurs in the stationary case. The method for removing this dependence on the initial time is to average the correlation function over a period of the driving [1]. After doing this the

first order term vanishes, and consequently we do not worry about it and compute only those whose average gives a non-zero contribution, i.e., the zeroth and second order terms. Taking this into account, and by applying Eq. (12) to $\Psi(\hat{v}, t | \hat{R}_0, t_0)$,

$$\begin{aligned}
& \langle \hat{R}_x(t) \hat{R}_x(t+\tau) | \hat{R}_0(t_0) \rangle \\
&\sim \int d\hat{v} \int d\hat{u} \hat{u}_x \hat{v}_x \Psi_0(\hat{v}, t | \hat{R}_0, t_0) \Psi_0(\hat{u}, t + \tau | \hat{v}, t) \\
&\quad + \int d\hat{v} \int d\hat{u} \hat{u}_x \hat{v}_x \Delta\Psi(\hat{v}, t | \hat{R}_0, t_0) \Delta\Psi(\hat{u}, t + \tau | \hat{v}, t),
\end{aligned} \tag{34}$$

where the sign \sim indicates that the terms which vanish after averaging over the period of the driving have been neglected (although the average has not been performed yet). After introducing the corresponding expressions for $\Psi(\hat{v}, t | \hat{R}_0, t_0)$ and by using Eq. (33) we obtain

$$\begin{aligned}
\langle \hat{R}_x(t) \hat{R}_x(t+\tau) \rangle &\sim \left(\frac{4\pi}{3} \right)^2 2e^{-2D_r\tau} \cos(\omega_0\tau) + \left(\frac{2\pi}{3} \right)^3 \lambda^2(t) \\
&\quad \times e^{i\omega\tau} \left| \int_0^\infty dt' e^{i\omega t'} \chi_x(t') \right|^2
\end{aligned} \tag{35}$$

(the details of this computation are given in Appendix B), where we have defined

$$\langle \hat{R}_x(t) \hat{R}_x(t+\tau) \rangle \equiv \frac{1}{4\pi} \int d\hat{R}_0 \langle \hat{R}_x(t) \hat{R}_x(t+\tau) | \hat{R}_0(t_0) \rangle. \tag{36}$$

At this stage, and before applying the Fourier transform to the correlation function to obtain the power spectrum of the process $\hat{R}_x(t)$, we average Eq. (35) to obtain

$$\begin{aligned}
\overline{\langle \hat{R}_x(t) \hat{R}_x(t+\tau) \rangle} &= \frac{\omega}{2\pi} \int_0^{2\pi/\omega} dt \langle \hat{R}_x(t) \hat{R}_x(t+\tau) \rangle \\
&= \left(\frac{4\pi}{3} \right)^2 2e^{-2D_r\tau} \cos(\omega_0\tau) \\
&\quad + \left(\frac{2\pi}{3} \right)^3 \lambda_0^2 e^{i\omega\tau} \left| \int_0^\infty dt' e^{i\omega t'} \chi_x(t') \right|^2.
\end{aligned} \tag{37}$$

This computation has been carried out with the assumption that τ is a positive quantity. To extend our computation to $\tau < 0$ we have to use the backward Fokker-Planck equation. The operator that generates the backward evolution of the probability distribution is $-\mathcal{L}^\dagger$ [18], \mathcal{L} being the Fokker-Planck operator and \mathcal{L}^\dagger its adjoint operator. Consequently the formal solution of the backward Fokker-Planck, equivalent to Eq. (12), is given by ($t < t_0$)

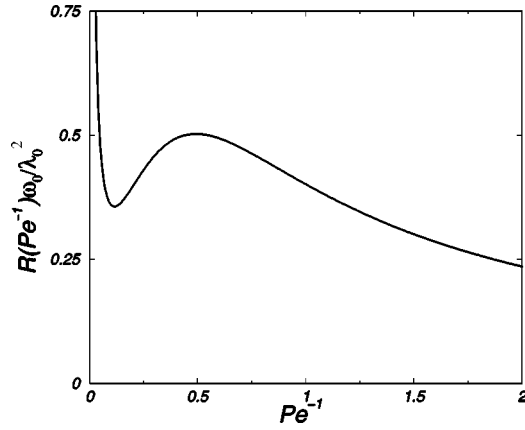


FIG. 1. Signal-to-noise ratio as a function of the inverse of the Péclet number $Pe^{-1} = D_r/\omega_0$. We have represented nondimensional quantities.

$$\Psi(\hat{R}, t) = e^{-(t-t_0)\mathcal{L}_0^\dagger} \Psi^0(t_0) + \int_{t_0}^t dt' \lambda(t') e^{-(t-t')\mathcal{L}_0^\dagger} \mathcal{L}_1 \Psi^0(t'). \quad (38)$$

As in the case of the operator \mathcal{L}_0 , the spherical harmonics are eigenfunctions of $-\mathcal{L}_0^\dagger$ with eigenvalues given by

$$-\mathcal{L}_0^\dagger Y_{lm}(\hat{R}) = [l(l+1)D_r - im\omega_0] Y_{lm}(\hat{R}). \quad (39)$$

Thus the process to follow in the calculation of the correlation function for $\tau < 0$ is identical to the corresponding computation for $\tau > 0$ but changing the eigenvalues of the operator \mathcal{L}_0 to those of $-\mathcal{L}_0^\dagger$, which yields

$$R = \frac{S(\omega)}{N(\omega)} = \lambda_0^2 \frac{6}{\pi} \times \frac{\{2D_r/[4D_r^2 + (\omega + \omega_0)^2] + 2D_r/[4D_r^2 + (\omega - \omega_0)^2]\}^2 + \{(\omega + \omega_0)/[4D_r^2 + (\omega + \omega_0)^2] + (\omega - \omega_0)/[4D_r^2 + (\omega - \omega_0)^2]\}^2}{2D_r/[4D_r^2 + (\omega + \omega_0)^2] + 2D_r/[4D_r^2 + (\omega - \omega_0)^2]}. \quad (42)$$

This quantity has been plotted in Fig. 1 as a function of the inverse of the Péclet number $Pe^{-1} = D_r/\omega_0$, which measures the ratio between the time scales associated with diffusion (thermal noise) and flow. The presence of a maximum in R for a nonzero value of this parameter shows the existence of stochastic resonance in the relaxation process of a dipole in a shear flow. In addition to the slow relaxation to the single attractor of the dynamics, our model includes another effect, which hides, to some extent, the SR profile. To understand this, note that even though the signal is too weak, it nevertheless causes the position of the attractor of the dynamics to vary, and so the output will always have a nonzero compo-

$$\overline{\langle \hat{R}_x(t) \hat{R}_x(t + \tau) \rangle} = \left(\frac{4\pi}{3} \right)^2 2e^{2D_r\tau} \cos(\omega_0\tau) + \left(\frac{2\pi}{3} \right)^3 \lambda_0^2 e^{i\omega\tau} \left| \int_0^\infty dt' e^{i\omega t'} \chi_x(t') \right|^2 \quad (40)$$

for $\tau < 0$.

We now apply to this averaged correlation function (now defined for $-\infty < \tau < \infty$) the Wiener-Khinchine theorem, which states that the power spectrum and the correlation function are related through a Fourier transform. Thus,

$$Q(\Omega) = \int_{-\infty}^\infty d\tau \overline{\langle \hat{R}_x(t) \hat{R}_x(t + \tau) \rangle} e^{i\Omega\tau} = N(\Omega) + S(\omega) \delta(\Omega - \omega), \quad (41a)$$

$$N(\Omega) = \left(\frac{4\pi}{3} \right)^2 \left[\frac{2D_r}{4D_r^2 + (\Omega + \omega_0)^2} + \frac{2D_r}{4D_r^2 + (\Omega - \omega_0)^2} \right], \quad (41b)$$

$$S(\omega) = \left(\frac{2\pi}{3} \right)^3 \lambda_0^2 |\chi_x(\omega)|^2. \quad (41c)$$

Since our purpose is to discern whether or not SR is present in the relaxation process of the quantity $\hat{R}_x(t)$, we proceed to compute the signal-to-noise ratio R , i.e., the ratio between the weight of the δ function in Eq. (41a) and the noisy part of $Q(\Omega)$ computed at the frequency of the driving. From Eqs. (25) and (41) we achieve

nent at the signal frequency. This fact causes the SNR to go to infinity in the zero noise limit [19].

V. MEAN FIRST PASSAGE TIME

In this section we study the behavior of the escape time distribution and the mean first passage time of the magnetic dipole immersed in a shear flow. To this end, we have to account for the fixed point orientations of Eq. (4) in the case $\lambda_0 < 1$. In this situation there is a single fixed point corresponding to an orientation contained in the plane $x=0$ or, equivalently, $\phi = \pi/2$. However, when $\lambda(t) > 0$ this station-

ary orientation is in the subspace $z > 0$ ($\cos \theta > 0$) and in the subspace $z < 0$ ($\cos \theta < 0$) if $\lambda(t) < 0$. Therefore, we are going to study the escape from the region $z > 0$ ($\cos \theta > 0$) assuming that the initial orientation of the dipole is contained in this region. Consequently, we have to solve the Fokker-Planck equation (10) with absorbing boundary conditions in the plane $\cos \theta = 0$ [20,21], i.e.,

$$\Psi(\cos \theta = 0, \phi, t) = 0. \quad (43)$$

Since this escape problem will be treated perturbatively, the first step is to analyze the eigenvalue problem of the operator \mathcal{L}_0 [Eq. (11)] under the boundary condition (43). It is easy to check that the eigenfunctions and the eigenvalues are the same with the restriction that only those spherical harmonics that vanish at $\cos \theta = 0$ are solutions of this eigenvalue problem. From the parity properties of the associated Legendre functions [15], one can see that Eq. (43) selects only the spherical harmonics such that $l + m = 2n + 1$ with $n = 0, 1, \dots$. Thus, we have

$$\Psi(\hat{R}, t) = \sum_{\langle l, m \rangle} a_{lm}(t) Y_{lm}(\hat{R}), \quad (44)$$

where $\langle l, m \rangle$ denotes that the sum is carried out over $0 \leq l < \infty$ and $-l \leq m \leq l$ restricted by $l + m = 2n + 1$.

In order to evaluate the mean first passage time (MFPT), we have to compute first the survival probability $S(\hat{R}_0, t)$ and the escape time distribution (ETD), which are related through

$$\rho(\hat{R}_0, t) = - \frac{d S(\hat{R}_0, t)}{dt}, \quad (45)$$

where $S(\hat{R}_0, t)$ is defined by

$$\begin{aligned} S(\hat{R}_0, t) &= \int_{\mathcal{R}} d\hat{R} \Psi(\hat{R}, t | \hat{R}_0) \\ &= \int_0^{2\pi} d\phi \int_0^1 d(\cos \theta) \Psi(\cos \theta, \phi, t | \hat{R}_0), \end{aligned} \quad (46)$$

with \mathcal{R} the region from which we are studying the escape problem (in the present case $\cos \theta > 0$), and $\hat{R}_0 \in \mathcal{R}$ the initial orientation of the dipole. The probability distribution $\Psi(\hat{R}, t | \hat{R}_0)$ is obtained from Eq. (12) with the boundary conditions (43) and the initial condition

$$\Psi(\hat{R}, t = 0) = \delta(\hat{R} - \hat{R}_0) = \sum_{\langle l, m \rangle} Y_{lm}^*(\hat{R}_0) Y_{lm}(\hat{R}). \quad (47)$$

Before proceeding to obtain the survival probability, there are some facts to consider that will facilitate further computation. Looking at Eq. (46), one can see that, due to the integration over the azimuthal angle, only terms with $m = 0$ contribute to $S(\hat{R}_0, t)$. Consequently, the selection rule l

$+m = 2n + 1$ reduces to keeping only the odd values of l . In addition, we are interested only in the modes with greater relaxation times. Therefore, from the whole series Eq. (44) we are interested only in the term $l = 1$, $m = 0$. Thus, our purpose is to obtain the coefficient $a_{10}(t)$ up to first order in $\lambda(t)$ from Eqs. (10) and (43). Up to zeroth order, we have

$$a_{10}^{(0)}(t) = e^{-2D_r t} Y_{10}(\hat{R}_0). \quad (48)$$

Obtaining the first order contribution $a_{10}^{(1)}(t)$ requires somewhat more elaborate calculation. To proceed further with this computation, the operator \mathcal{L}_1 acting on $Y_{lm}(\hat{R})$ yields

$$\begin{aligned} \mathcal{L}_1 Y_{lm}(\hat{R}) &= -2\omega_0 \sqrt{\frac{2\pi}{3}} [Y_{11}(\hat{R}) + Y_{1-1}(\hat{R})] Y_{lm}(\hat{R}) \\ &\quad - \omega_0 \left[\sqrt{\frac{4\pi}{3}} Y_{10}(\hat{R}) \mathcal{R}_y \right. \\ &\quad \left. - i \sqrt{\frac{2\pi}{3}} [Y_{11}(\hat{R}) - Y_{1-1}(\hat{R})] \mathcal{R}_z \right] Y_{lm}(\hat{R}), \end{aligned} \quad (49)$$

where the action of \mathcal{R}_z on $Y_{lm}(\hat{R})$ is given by Eq. (14a) and

$$\begin{aligned} \mathcal{R}_y Y_{lm}(\hat{R}) &= -\frac{1}{2} \{ \sqrt{(l-m)(l+m+1)} Y_{l, m+1}(\hat{R}) \\ &\quad - \sqrt{(l+m)(l-m+1)} Y_{l, m-1}(\hat{R}) \}. \end{aligned} \quad (50)$$

From Eqs. (49) and (50) together with the rules for the addition of angular momenta familiar from quantum mechanics [14] and the selection rule $l + m = 2n + 1$ imposed by the boundary condition (43), one can deduce that only the term $Y_{2\pm 1}(\hat{R})$ contributes to $a_{10}^{(1)}(t)$. The rules of addition of angular momenta imply that the product of two spherical harmonics $Y_{lm} Y_{pq}$ has a projection onto a third spherical harmonic Y_{rs} only when $m + q = s$. On the other hand, these same rules impose the restriction that the product $Y_{lm} Y_{pq}$ projects only onto subspaces such that $|l - p| \leq r \leq l + p$. By using these restrictions one can see that when one takes $l = 1$ and $m = 0$ in Eq. (49) one obtains a vanishing contribution and only when $l = 2$ and $m = \pm 1$ is the contribution to $a_{10}^{(1)}(t)$ different from zero. All other contribution of higher values l are explicitly excluded by the rule $|l - p| \leq r \leq l + p$.

Taking these considerations into account and by using the results

$$\begin{aligned}
& \frac{3}{4\pi} \int_0^{2\pi} d\phi \int_0^1 d(\cos \theta) Y_{11}(\hat{R}) Y_{2-1}(\hat{R}) Y_{10}(\hat{R}) \\
&= \frac{9}{80\pi} \sqrt{\frac{5}{3\pi}}, \\
& \frac{3}{4\pi} \int_0^{2\pi} d\phi \int_0^1 d(\cos \theta) Y_{1-1}(\hat{R}) Y_{21}(\hat{R}) Y_{10}(\hat{R}) \\
&= \frac{9}{80\pi} \sqrt{\frac{5}{3\pi}}, \\
& \frac{3}{4\pi} \int_0^{2\pi} d\phi \int_0^1 d(\cos \theta) Y_{10}(\hat{R}) Y_{20}(\hat{R}) Y_{10}(\hat{R}) \\
&= \frac{3}{20\pi} \sqrt{\frac{5}{4\pi}}. \tag{51}
\end{aligned}$$

we obtain the first order correction to the coefficient $a_{10}(t)$:

$$\begin{aligned}
a_{10}^{(1)}(t) &= -\omega_0 \frac{\lambda_0}{40} \sqrt{\frac{15}{4\pi}} \left(\frac{\sqrt{2}}{2} + 2\sqrt{3} \right) \\
&\times \left\{ Y_{21}(\hat{R}_0) \frac{4D_r + i(\omega - \omega_0)}{16D_r^2 + (\omega - \omega_0)^2} \right. \\
&\left. + Y_{2-1}(\hat{R}_0) \frac{4D_r + i(\omega + \omega_0)}{16D_r^2 + (\omega + \omega_0)^2} \right\}. \tag{52}
\end{aligned}$$

Equations (48) and (52) together with Eqs. (44) and (46) allows us to obtain the survival probability $S(\hat{R}_0, t)$, which is given by

$$\begin{aligned}
S(\hat{R}_0, t) &= \pi \sqrt{\frac{3}{4\pi}} Y_{10}(\hat{R}_0) e^{-2D_r t} - \frac{\omega_0}{40} \sqrt{\frac{15}{4\pi}} \left(\frac{\sqrt{2}}{2} + 2\sqrt{3} \right) \\
&\times \left\{ Y_{21}(\hat{R}_0) e^{-2D_r t} \int_0^t d\tau \lambda(\tau) e^{-(4D_r + i\omega_0)\tau} \right. \\
&\left. + Y_{2-1}(\hat{R}_0) e^{-2D_r t} \int_0^t d\tau \lambda(\tau) e^{-(4D_r - i\omega_0)\tau} \right\}. \tag{53}
\end{aligned}$$

This quantity is directly related to the MFPT, since

$$T(\hat{R}_0) = \int_0^\infty dt t \rho(\hat{R}_0, t) = \int_0^\infty dt S(\hat{R}_0, t), \tag{54}$$

where we have used Eq. (45). Consequently, the MFPT is given by

$$\begin{aligned}
T(\hat{R}_0) &\equiv T_0(\hat{R}_0) + \Delta T(\hat{R}_0), \\
T_0(\hat{R}_0) &= \pi \sqrt{\frac{3}{4\pi}} \frac{\sqrt{1-\lambda^2}}{2D_r}, \\
\Delta T(\hat{R}_0) &= T_0 \left[\frac{\lambda_0^2}{40} \sqrt{\frac{15}{4\pi}} \left(\frac{\sqrt{2}}{2} + 2\sqrt{3} \right) \right. \\
&\times 3\omega_0 \sqrt{\frac{5}{24\pi}} \left\{ \frac{(\omega - \omega_0)}{16D_r^2 + (\omega - \omega_0)^2} \right. \\
&\left. \left. + \frac{(\omega + \omega_0)}{16D_r^2 + (\omega + \omega_0)^2} \right\} \right], \tag{55}
\end{aligned}$$

where we have taken $\hat{R}_0 = \hat{R}_s$. In Fig. 2 we have plotted the quantity $\Delta T/T_0$. The figure shows that this quantity exhibits a minimum, as required for the appearance of SR.

The knowledge of the survival probability allows us to obtain the ETD $\rho(\hat{R}_0, t)$. From Eqs. (45) and (53) the ETD is given by

$$\begin{aligned}
\rho(\hat{R}_0, t) &= e^{-2D_r t} \left[2D_r \pi \sqrt{\frac{3}{4\pi}} Y_{10}(\hat{R}_0) - \lambda_0 \frac{2D_r}{40} \sqrt{\frac{15}{4\pi}} \left(\frac{\sqrt{2}}{2} + 2\sqrt{3} \right) \right. \\
&\times \left\{ Y_{21}(\hat{R}_0) \frac{2D_r + i(\omega - \omega_0)}{16D_r^2 + (\omega - \omega_0)^2} Y_{2-1}(\hat{R}_0) \frac{2D_r + i(\omega + \omega_0)}{16D_r^2 + (\omega + \omega_0)^2} \right\} + \lambda_0 \frac{1}{40} \sqrt{\frac{15}{4\pi}} \left(\frac{\sqrt{2}}{2} + 2\sqrt{3} \right) \\
&\left. \times \{ Y_{21}(\hat{R}_0) e^{-[4D_r + i(\omega - \omega_0)]t} Y_{2-1}(\hat{R}_0) e^{-[4D_r + i(\omega + \omega_0)]t} \} \right] \tag{56}
\end{aligned}$$

By taking $\hat{R}_0 = \hat{R}_s$, we finally obtain

$$\begin{aligned}
\frac{e^{2D_r t}}{2D_r \sqrt{1-\lambda_0^2}} \Delta \rho &= \frac{\omega_0 \lambda_0^2}{40\pi} \sqrt{75} \left(\frac{\sqrt{2}}{2} + 2\sqrt{3} \right) \left\{ \frac{e^{-4D_r t}}{2} \{ \sin[(\omega - \omega_0)t] + \sin[(\omega + \omega_0)t] \} \right. \\
&\left. - \left[\frac{(\omega - \omega_0)}{16D_r^2 + (\omega - \omega_0)^2} + \frac{(\omega + \omega_0)}{16D_r^2 + (\omega + \omega_0)^2} \right] \right\}, \tag{57}
\end{aligned}$$

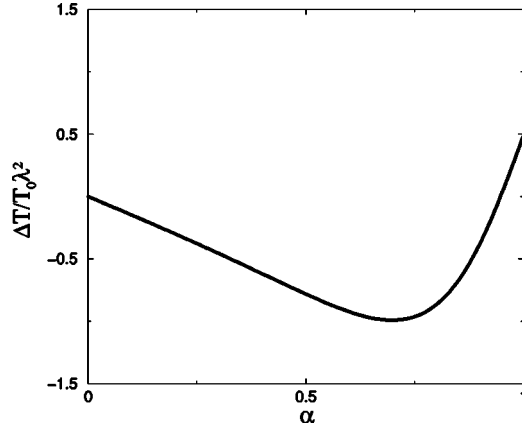


FIG. 2. Mean first passage time as a function of the parameter $\alpha = \omega/\omega_0$. The presence of a minimum reveals the existence of stochastic resonance. $Pe^{-1} = D_r/\omega_0 = 0.08$. We have represented nondimensional quantities.

where $\Delta\rho \equiv \rho - \rho_0$, ρ_0 being the corresponding ETD when the amplitude of the oscillating field is set to zero.

The succession of maxima in the ETD (see Fig. 3) indicates that the dynamics of a magnetic dipole suspended in a shear flow under a periodic field exhibits SR.

VI. DISCUSSION

We have shown that the relaxation process of a dipole immersed in a shear flow exhibits SR upon application of a weak periodic field. To this end we have computed three quantities typically used to characterize SR, namely, the signal-to-noise ratio, the escape time distribution, and the mean first passage time. All of them behave as expected for a process in which SR occurs.

Previous work devoted to analyzing whether or not SR is present in the relaxation process of an overdamped dipole in a fluid at rest has shown that this phenomenon does not occur in the linear regime [13]. Effectively, linear response theory predicts a maximum in the signal, i.e., in the susceptibility, as a function of the noise level. However, the SNR decreases monotonically with the noise level. This behavior can be easily understood. In the limit of zero noise the output of the system has a small component (proportional to the applied field) at the frequency of the signal whereas the background noise vanishes at zero noise level, this behavior being responsible for the monotonic dependence of the SNR on the noise intensity.

In our case, the situation is completely different. When the fluid in which the dipole is suspended is submitted to a pure rotation (vortex flow), both output signal and output background noise exhibit a peak at the same value of D_r (see Fig. 4). Consequently, although the background noise vanishes when D_r goes to zero, the characteristic SR profile of the SNR cannot be completely hidden, as shown in Fig. 1. This feature arises as a consequence of the presence of shear acting on the suspension; thus, the appearance of SR in the system studied in this paper is a nonequilibrium feature.

In a sense, the mechanism yielding SR in this system is similar to the one operating in SR in threshold devices

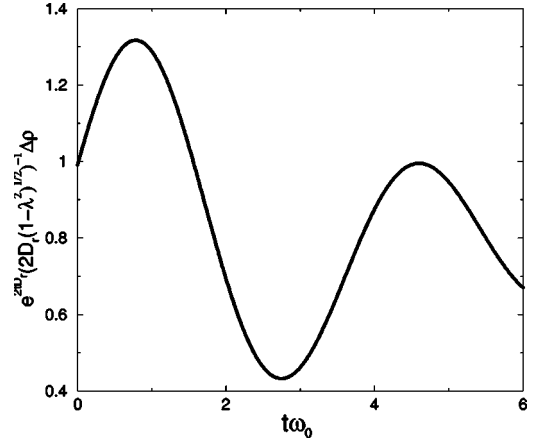


FIG. 3. Escape time distribution for a dipole in a shear flow under an oscillating magnetic field. The succession of maxima is a signature of the presence of stochastic resonance in this system. $Pe^{-1} = D_r/\omega_0 = 0.08$ and $\alpha = \omega/\omega_0 = 0.7$. We have represented nondimensional quantities.

[9,10]. Due to the presence of noise, the dipole can eventually acquire enough energy to get out from its stable orientation by crossing the absorbing barrier (the threshold) $\cos\theta = 0$. After this, the system is driven to its stable position. This process produces a short spike in the magnetization. Of course, the time that the system takes to return to the fixed point has to be smaller than the semiperiod of the oscillating magnetic field. Thus, SR in this system can be understood in the same way as, for example, the SR in level crossing detectors [9].

LRT has been one of the most widely used tools in the study of stochastic resonance [22]. When the system is in thermal equilibrium in the absence of the external periodic force, a very adequate way of describing stochastic resonance is in terms of the susceptibility. This is because the noisy part of the power spectrum is given directly by the susceptibility through the fluctuation-dissipation theorem,

$$\text{Im}\chi(\Omega) = \frac{\Omega}{2D_r} N(\Omega). \quad (58)$$

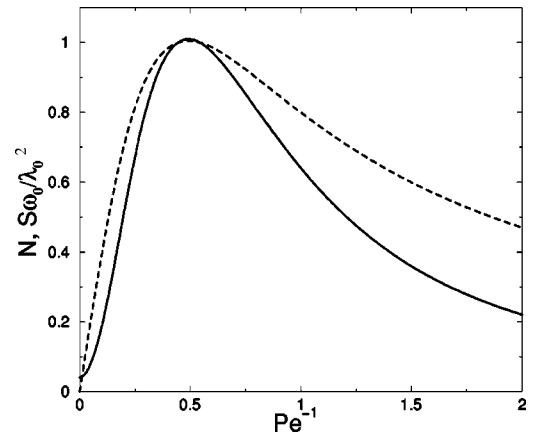


FIG. 4. Output signal and output background signal as functions of the inverse of the Péclet number. Solid line represents the quantity $S(Pe^{-1})\omega_0/\lambda_0^2$ whereas dashed line represents $N(Pe^{-1})$. We have taken $\alpha = 0.1$. We have represented nondimensional quantities.

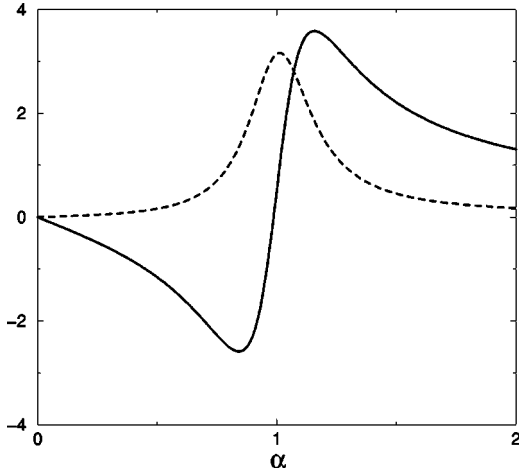


FIG. 5. Comparison between the imaginary part of the susceptibility of the signal \hat{R}_x (solid line) and the noisy part of the spectrum computed from the Fokker-Planck equation (dashed line). $Pe^{-1} = D_r/\omega_0 = 0.08$. We have represented nondimensional quantities.

This result is correct when the fluctuations whose spectral density is given by $N(\Omega)$ have the thermal equilibrium state as reference state [16].

However, in the present case we are dealing with a system that is maintained in an out-of-equilibrium steady state due to the presence of a shear flow. It is evident from Fig. 5, where we have plotted the imaginary part of the susceptibility corresponding to \hat{R}_x and the noisy part of the power spectrum, that these two quantities are clearly different. Note that, if $\omega_0 = 0$, i.e., the system in the absence of the periodic field is in equilibrium, the relation (58) is fulfilled. Thus we have shown that, although we can define a susceptibility that describes the response of our system to a small perturbation, we cannot describe SR by means of LRT. The reason can be found in the fact that due to the nonequilibrium nature of the attractor of the dynamics the fluctuation-dissipation theorem fails to be valid.

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APPENDIX A: LINEAR STABILITY ANALYSIS OF THE FIXED POINTS OF EQ. (4)

From Eqs. (3) and (4) one can see that the time derivative of \hat{R} vanishes either when $\vec{\Omega}_p = \vec{0}$ or when $\vec{\Omega}_p \times \hat{R} = \vec{0}$. In the first case we have

$$\vec{\Omega}_p = \omega_0 \{ \hat{z} + \lambda_0 (\hat{R}_s \times \hat{x}) \} = 0 \Rightarrow \hat{z} = -\lambda_0 \hat{R}_s \times \hat{x}. \quad (\text{A1})$$

From this equation, and taking into account that $|\hat{R}_s| = 1$, we obtain that the stationary orientation is

$$\hat{R}_s = \pm \sqrt{1 - \lambda_0^{-2} \hat{x}^2} + \lambda_0^{-1} \hat{y}. \quad (\text{A2})$$

This solution exists only when $\lambda_0 \geq 1$ and corresponds to a fixed orientation of the dipoles, given that the intensity of the magnetic field is high enough to maintain this fixed direction.

The second possibility leads to

$$\hat{z} \times \hat{R}_s = -\lambda_0 (\hat{R}_s \times \hat{x}) \times \hat{R}_s = -\lambda_0 [\hat{x} - (\hat{x} \cdot \hat{R}_s) \hat{R}_s]. \quad (\text{A3})$$

Equation (A3) provides two equations for three unknowns. If one sets $\hat{R}_z = 0$ one recovers Eq. (A2). If, by contrast one makes $\hat{R}_x = 0$ then a different stationary orientation is obtained,

$$\hat{R}_s = \lambda_0 \hat{y} \pm \sqrt{1 - \lambda_0^2} \hat{z}, \quad (\text{A4})$$

which exists only when $\lambda_0 \leq 1$. This orientation gives rise to a rotation of the dipoles with angular velocity

$$\vec{\Omega}_s = \omega_0 \sqrt{1 - \lambda_0^2} \{ \sqrt{1 - \lambda_0^2} \hat{z} \pm \lambda_0 \hat{y} \}, \quad (\text{A5})$$

since, in this case, the field is not strong enough to inhibit the rotation caused by the shear flow.

The linear stability of these fixed points is better analyzed in spherical coordinates. Taking into account that

$$(\hat{R} \times \hat{x}) \times \hat{R} = \hat{x} - \hat{R}(\hat{R} \cdot \hat{x}), \quad (\text{A6})$$

we obtain

$$\begin{aligned} \frac{1}{\omega_0} \frac{d\hat{R}_x}{dt} &= \lambda_0 (1 - \hat{R}_x^2) - \hat{R}_y, \\ \frac{1}{\omega_0} \frac{d\hat{R}_y}{dt} &= -\lambda_0 \hat{R}_x \hat{R}_y + \hat{R}_x, \\ \frac{1}{\omega_0} \frac{d\hat{R}_z}{dt} &= -\lambda_0 \hat{R}_x \hat{R}_z. \end{aligned} \quad (\text{A7})$$

After expressing the components of \hat{R} in spherical coordinates, we obtain the following bidimensional dynamical system:

$$\begin{aligned} \frac{1}{\omega_0} \frac{d\theta}{dt} &= \lambda_0 \cos \theta \cos \phi, \\ \frac{1}{\omega_0} \frac{d\phi}{dt} &= -\lambda_0 \frac{\sin \phi}{\sin \theta} + 1, \end{aligned} \quad (\text{A8})$$

where θ and ϕ are the polar and azimuthal angles, respectively. By linearization of Eqs. (A8) around the $\lambda_0 \geq 1$ fixed points we obtain the matrix

$$A(\lambda_0 \geq 1) = \begin{pmatrix} \mp \lambda_0 \sqrt{1 - \left(\frac{1}{\lambda_0}\right)^2} & 0 \\ 0 & \mp \lambda_0 \sqrt{1 - \left(\frac{1}{\lambda_0}\right)^2} \end{pmatrix}, \quad (\text{A9})$$

which implies that, if λ_0 is positive, the orientation corresponding to choosing the sign $+$ in Eq. (A2) is stable, while the other one is unstable.

The same linearization procedure carried out around the $\lambda < 1$ fixed points leads to

$$A(\lambda_0 < 1) = \begin{pmatrix} 0 & \lambda_0^2 \sqrt{1 - \lambda_0^2} \\ -\lambda_0^2 \sqrt{1 - \lambda_0^2} & 0 \end{pmatrix}. \quad (\text{A10})$$

The eigenvalues of this matrix are given by

$$\alpha = \pm i \lambda_0^2 \sqrt{1 - \lambda_0^2}. \quad (\text{A11})$$

APPENDIX B: COMPUTATION OF EQS. (33) AND (35)

In this Appendix we work out in detail some steps of the computation of the power spectrum corresponding to the relaxation process of a dipole under an oscillating magnetic field in a shear flow; in particular, we calculate the integrals that yield Eqs. (33) and (35). From Eqs. (31) and (32),

$$\int d\hat{u} \hat{u}_x \Psi(\hat{u}, t + \tau | \hat{v}, t) = I_1 + I_2, \quad (\text{B1a})$$

$$I_1 = \int d\hat{u} \hat{u}_x \Psi_0(\hat{u}, t + \tau | \hat{v}, t) = \int d\hat{u} \hat{u}_x e^{\tau \mathcal{L}_0} \delta(\hat{u} - \hat{v}), \quad (\text{B1b})$$

$$\begin{aligned} I_2 &= \int d\hat{u} \hat{u}_x \Delta \Psi(\hat{u}, t + \tau | \hat{v}, t) \\ &= \int d\hat{u} \hat{u}_x \int_t^{t+\tau} ds \lambda(s) e^{(t+\tau-s)\mathcal{L}_0} \mathcal{L}_1 e^{s\mathcal{L}_0} \delta(\hat{u} - \hat{v}). \end{aligned} \quad (\text{B1c})$$

To begin with we focus on the integral I_1 , which can be rewritten as

$$I_1 = \int d\hat{u} (e^{\tau \mathcal{L}_0^\dagger \hat{u}_x}) \delta(\hat{u} - \hat{v}), \quad (\text{B2})$$

where \mathcal{L}_0^\dagger is the adjoint operator of \mathcal{L}_0 defined by

$$\int d\hat{u} \hat{A} (\mathcal{L}_0 \hat{B}) = \int d\hat{u} (\mathcal{L}_0^\dagger \hat{A}) \hat{B} \quad (\text{B3})$$

with \hat{A} and \hat{B} two arbitrary observables. Explicitly, \mathcal{L}_0^\dagger is given by

$$\begin{aligned} \mathcal{L}_0^\dagger &= \omega_0 \hat{R}_0 \cdot \vec{\mathcal{R}} + D_r \mathcal{R}^2, \\ \mathcal{L}_0^\dagger Y_{lm}(\hat{R}) &= [-l(l+1)D_r + i\omega_0 m] Y_{lm}(\hat{R}), \end{aligned} \quad (\text{B4})$$

and therefore Eq. (B2) reads

$$\begin{aligned} I_1 &= -\sqrt{\frac{2\pi}{3}} \int d\hat{u} [e^{-(2D_r - i\omega_0)\tau} Y_{11}(\hat{u}) \\ &\quad + e^{-(2D_r + i\omega_0)\tau} Y_{1-1}(\hat{u})] \delta(\hat{u} - \hat{v}) \\ &= -\sqrt{\frac{2\pi}{3}} [e^{-(2D_r - i\omega_0)\tau} Y_{11}(\hat{v}) + e^{-(2D_r + i\omega_0)\tau} Y_{1-1}(\hat{v})], \end{aligned} \quad (\text{B5})$$

leading to Eq. (33a). In Eq. (B5) we have used the relation

$$\hat{u}_x = -\sqrt{\frac{2\pi}{3}} [Y_{11}(\hat{u}) + Y_{1-1}(\hat{u})]. \quad (\text{B6})$$

To compute the integral I_2 , we have to use the following representation of the δ function:

$$\delta(\hat{u} - \hat{v}) = \sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\hat{v}) Y_{lm}(\hat{u}). \quad (\text{B7})$$

After introducing this expression into Eq. (B1c) we obtain

$$\begin{aligned} I_2 &= \sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\hat{v}) \int_t^{t+\tau} ds \lambda(s) \int d\hat{u} (e^{(t+\tau-s)\mathcal{L}_0^\dagger \hat{u}_x}) \\ &\quad \times \mathcal{L}_1 e^{s\mathcal{L}_0} Y_{lm}(\hat{u}) \\ &= \sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\hat{v}) \int_t^{t+\tau} ds \lambda(s) e^{-[l(l+1)D_r + im\omega_0]s} \\ &\quad \times \int d\hat{u} (e^{(t+\tau-s)\mathcal{L}_0^\dagger \hat{u}_x}) \mathcal{L}_1 Y_{lm}(\hat{u}), \end{aligned} \quad (\text{B8})$$

and, by using Eq. (B6), Eq. (B8) yields Eq. (33), i.e.,

$$\begin{aligned} I_2 &= -\sqrt{\frac{2\pi}{3}} \sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\hat{v}) \int_t^{t+\tau} ds \lambda(s) \\ &\quad \times \int d\hat{u} \{ [e^{-(2D_r - i\omega_0)(t+\tau-s)} Y_{11}(\hat{u}) \\ &\quad + e^{-(2D_r + i\omega_0)(t+\tau-s)} Y_{1-1}(\hat{u})] \\ &\quad \times e^{-[l(l+1)D_r + im\omega_0]s} \mathcal{L}_1 Y_{lm}(\hat{u}) \}. \end{aligned} \quad (\text{B9})$$

Once these expressions have been obtained we can compute the correlation function given by Eq. (34),

$$\begin{aligned} \langle \hat{R}_x(t) \hat{R}_x(t + \tau) | \hat{R}_0(t_0) \rangle &\sim \int d\hat{v} \hat{v}_x \Psi_0(\hat{v}, t | \hat{R}_0, t_0) I_1 \\ &\quad + \int d\hat{v} \hat{v}_x \Delta \Psi(\hat{v}, t | \hat{R}_0, t_0) I_2, \end{aligned} \quad (\text{B10})$$

$\Psi_0(\hat{v}, t | \hat{R}_0, t_0)$ and $\Delta\Psi(\hat{v}, t | \hat{R}_0, t_0)$ being given by

$$\begin{aligned} \Psi_0(\hat{v}, t | \hat{R}_0) &= e^{t\mathcal{L}_0} \delta(\hat{v} - \hat{R}_0) \\ &= \sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\hat{R}_0) e^{-[l(l+1)D_r + im\omega_0]t} Y_{lm}(\hat{v}), \\ \Delta\Psi(\hat{v}, t | \hat{R}_0) &= \int_0^t dr \lambda(r) e^{(t-r)\mathcal{L}_0} \mathcal{L}_1 e^{r\mathcal{L}_0} \delta(\hat{v} - \hat{R}_0) \\ &= \sum_{l=0}^{\infty} \sum_{m=-l}^l Y_{lm}^*(\hat{R}_0) \int_0^t dr \lambda(r) \\ &\quad \times e^{-[l(l+1)D_r + im\omega_0]r} e^{(t-r)\mathcal{L}_0} \mathcal{L}_1 Y_{lm}(\hat{v}), \end{aligned} \quad (\text{B11})$$

where the initial time t_0 has been fixed to zero and Eq. (B7) has been used.

Equations (B11) provide the evolution of the probability distribution under the condition of the system being initially in the state \hat{R}_0 . Since *a priori* nothing is known about this initial condition, we assume that \hat{R}_0 is a random variable uniformly distributed over the orientation space; consequently we average the correlation function over the distribution of initial states [Eq. (36)]. Taking into account that

$$\frac{1}{4\pi} \int d\hat{R}_0 Y_{lm}^*(\hat{R}_0) = \delta_{l,0} \delta_{m,0}, \quad (\text{B12})$$

the average of the correlation function over initial conditions is given by

$$\langle \hat{R}_x(t) \hat{R}_x(t+\tau) \rangle \sim I_3 + I_4, \quad (\text{B13a})$$

$$I_3 \equiv \frac{1}{4\pi} \int d\hat{v} \hat{v}_x I_1, \quad (\text{B13b})$$

$$I_4 \equiv \frac{1}{\sqrt{4\pi}} \int d\hat{v} \hat{v}_x \int_0^t dr \lambda(r) e^{(t-r)\mathcal{L}_0} \mathcal{L}_1 Y_{00}(\hat{v}) I_2. \quad (\text{B13c})$$

After introducing Eqs. (B5) and (B6) into (B13b), we obtain

$$\begin{aligned} I_3 &= \frac{1}{4\pi} \int d\hat{v} \frac{2\pi}{3} [Y_{11}(\hat{v}) + Y_{1-1}(\hat{v})] \\ &\quad \times [e^{-(2D_r - i\omega_0)\tau} Y_{11}(\hat{v}) + e^{-(2D_r + i\omega_0)\tau} Y_{1-1}(\hat{v})] \\ &= \frac{4\pi}{9} e^{-2D_r\tau} \cos(\omega_0\tau), \end{aligned} \quad (\text{B14})$$

where the orthogonality relation for the spherical harmonics,

$$\int d\hat{v} Y_{pq}^*(\hat{v}) Y_{lm}(\hat{v}) = \frac{4\pi}{2l+1} \frac{(l+m)!}{(l-m)!} \delta_{l,p} \delta_{m,q}, \quad (\text{B15})$$

has been used. On the other hand, from Eqs. (B9) and (B13c),

$$\begin{aligned} I_4 &= -\frac{1}{3} \sqrt{\frac{2\pi}{3}} \sum_{l=0}^{\infty} \sum_{m=-l}^l \int_0^t dr \lambda(r) \int_t^{t+\tau} ds \lambda(s) \int d\hat{v} \left\{ [Y_{11}(\hat{v}) + Y_{1-1}(\hat{v})] [e^{-(2D_r - i\omega_0)(t-r)} Y_{11}(\hat{v}) \right. \\ &\quad \left. + e^{-(2D_r + i\omega_0)(t-r)} Y_{1-1}(\hat{v})] Y_{lm}^*(\hat{v}) \int d\hat{u} \{ [e^{-(2D_r - i\omega_0)(t+\tau-s)} Y_{11}(\hat{u}) + e^{-(2D_r + i\omega_0)(t+\tau-s)} Y_{1-1}(\hat{u})] \right. \\ &\quad \left. \times e^{-[l(l+1)D_r + im\omega_0]s} \mathcal{L}_1 Y_{lm}(\hat{u}) \right\}. \end{aligned} \quad (\text{B16})$$

Let us focus our attention on the integral over \hat{v} :

$$\begin{aligned} &\int d\hat{v} [Y_{11}(\hat{v}) + Y_{1-1}(\hat{v})] [e^{-(2D_r - i\omega_0)(t-r)} Y_{11}(\hat{v}) + e^{-(2D_r + i\omega_0)(t-r)} Y_{1-1}(\hat{v})] Y_{lm}^*(\hat{v}) \\ &= \int d\hat{v} Y_{11}(\hat{v}) e^{-(2D_r - i\omega_0)(t-r)} Y_{11} Y_{lm}^*(\hat{v}) + \int d\hat{v} Y_{1-1}(\hat{v}) e^{-(2D_r + i\omega_0)(t-r)} Y_{1-1}(\hat{v}) Y_{lm}^*(\hat{v}) \\ &\quad + \int d\hat{v} Y_{1-1}(\hat{v}) e^{-(2D_r - i\omega_0)(t-r)} Y_{11}(\hat{v}) Y_{lm}^*(\hat{v}) + \int d\hat{v} Y_{11}(\hat{v}) e^{-(2D_r + i\omega_0)(t-r)} Y_{1-1}(\hat{v}) Y_{lm}^*(\hat{v}). \end{aligned} \quad (\text{B17})$$

From the rules of addition of angular momenta familiar from quantum mechanics, which imply that the product of two spherical harmonics $Y_{pq}Y_{rs}$ has a nonvanishing projection over a third spherical harmonic Y_{lm} only when the relations $|p-r| \leq l \leq r+p$ and $q+s=m$ are fulfilled, it is easy to see that these integrals will give a nonzero result only when $l=0,1,2$ [14]. In addition, for the integrals containing the products $Y_{1\pm 1}(\hat{v})Y_{1\pm 1}(\hat{v})$ the parameter m has to be $m=\pm 2$ whereas it must be $m=0$ for the integrals with $Y_{1\pm 1}(\hat{v})Y_{1\mp 1}(\hat{v})$ to yield a nonzero contribution. However, although these integrals give a nonvanishing contribution in principle, note that when we perform the integral over the variable \hat{u} in Eq. (B16) the terms introduced by these contributions finally yield, by the orthogonality property of the spherical harmonics, a vanishing result. Thus, only $l=0$ and $m=0$ contributes to I_3 . Taking this into account and using Eq. (B15),

$$I_4 = \left(\frac{2\pi}{3}\right)^3 \int_0^t dr \lambda(r) \{e^{-(2D_r - i\omega_0)(t-r)} + e^{-(2D_r + i\omega_0)(t-r)}\} \\ \times \int_t^{t+\tau} ds \lambda(s) \{e^{-(2D_r - i\omega_0)(t+\tau-s)} \\ + e^{-(2D_r + i\omega_0)(t+\tau-s)}\}. \quad (\text{B18})$$

Finally, performing the changes of variables $t' = t - r$ and $t'' = t + \tau - s$ and using Eq. (25) we obtain

$$I_4 = \left(\frac{2\pi}{3}\right)^3 \lambda^2(t) e^{-i\omega\tau} \left(\int_0^\infty dt e^{-i\omega t} \chi_x(t) \right)^2. \quad (\text{B19})$$

In this integral the upper limit goes to infinity by causality.

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