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Field-induced force-suppression in ferromagnetic colloids

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Abstract

We have analyzed the behavior of a colloidal ferromagnetic particle in a rotational flow and found a dynamic transition upon variation of a static applied magnetic field. Beyond the transition point the force exerted by the field on the particle does not increase, as usual, but decreases as the strength of the field is increased. Macroscopically, this behavior results in an effective viscosity that decreases as a function of the strength of the field. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

Suspensions of colloidal ferromagnetic particles, or ferrofluids, constitute a class of systems possessing the ability of undergoing significant changes in their properties upon the influence of an external field. In general, they can be viewed as formed of orientable particles suspended in a fluid phase, exhibiting a complex dynamics which is affected by the flow and by the imposed field [1–6]. These systems have been analyzed

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in many different situations, both theoretically and experimentally. In all of them, a trait which is always present is the increase of the effective viscosity as the strength of the static applied field is increased [7]. The ubiquity of this result relies in a rather common feature: the higher the strength of the field, the higher the force acting on the particle. In a ferromagnetic colloid, the action of a static field is to prevent the rotational motion of the particle which in turn prevents the motion of the surrounding fluid, in this way increasing the effective viscosity. The results obtained up to now are all fully supportive of this picture, although a complete analytical solution of the equations describing the dynamics of the system is far from being possible and only several approximations have been obtained.

In this paper, we show the existence of a regime in which the averaged force exerted on the particle does not increase with the strength of the applied field. This new regime emerges at sufficiently high vorticity and field, when the axis of the particle follows the rotation imposed by the vorticity, moving apart from the magnetic moment which remains oscillating around the field. The nonlinear interplay between the orientation and magnetic moment dynamics results in an effective viscosity, which decreases with the field.

The paper has been organized as follows. In Section 2 we present the model for a single ferromagnetic colloidal particle under the influence of both a rotational flow and a constant external magnetic field. In Section 3 we show analytically that, at sufficiently high vorticity, the viscosity is not a monotonous increasing function of the applied field. Numerical simulations presented in Section 4 confirm these results and elucidate the mechanism involved in the transition. Finally, in Section 5 we summarize our main results.

2. Model

To illustrate the essentials of the phenomenon we analyze the dynamics of a single ferromagnetic colloidal particle in a rotational flow under the influence of a constant external magnetic field. The energy of the particle is the sum of two contributions, one coming from the externally imposed field and the other from the anisotropy of the crystalline structure of the particle. For uniaxial crystals [8] it is given by

$$U = -\boldsymbol{m}\cdot\boldsymbol{H} - K_a V_p(\hat{\boldsymbol{n}}\cdot\hat{\boldsymbol{R}})^2 , \tag{1}$$

where $\boldsymbol{m} = m_0\hat{\boldsymbol{R}}$ is the magnetic moment of the particle, with m_0 the magnetic moment strength and $\hat{\boldsymbol{R}}$ its unit vector, $\boldsymbol{H}$ is the external magnetic field, K_a is the anisotropy constant, V_p is the magnetic volume of the particle, and $\hat{\boldsymbol{n}}$ is the unit vector along the direction of the axis of easy magnetization. The dynamics of $\hat{\boldsymbol{R}}$ and $\hat{\boldsymbol{n}}$ in a flow with vorticity $2\omega_0$ is described by [6]

$$\frac{d\hat{\boldsymbol{R}}}{dt} = \boldsymbol{\Omega}_R \times \hat{\boldsymbol{R}} \tag{2}$$

and

$$\frac{d\hat{\boldsymbol{n}}}{dt} = \boldsymbol{\Omega}_n \times \hat{\boldsymbol{n}}\,, \tag{3}$$

where the angular velocity of the magnetic moment and of the axis of the particle are given by

$$\boldsymbol{\Omega}_R = h\hat{\boldsymbol{R}} \times \frac{\partial U}{\partial \hat{\boldsymbol{R}}} + \boldsymbol{\omega}_L + \boldsymbol{\Omega}_n \tag{4}$$

and

$$\boldsymbol{\Omega}_n = \boldsymbol{\omega}_0 + \frac{1}{\xi_r}\boldsymbol{m} \times \boldsymbol{H}\,, \tag{5}$$

respectively. Here, $\boldsymbol{\omega}_L = -g\,\partial U/\partial\hat{\boldsymbol{R}}$ is the Larmor frequency, h and g are constants, and $\xi_r = 8\pi\eta_0 a^3$ is the rotational friction coefficient of the particle, with η_0 and a being the viscosity of the fluid phase and the hydrodynamic radius of the particles, respectively. To be explicit, we consider a field applied along the x-direction ($\boldsymbol{H} = H_0\hat{\boldsymbol{x}}$) and a vorticity perpendicular to it, along the y-direction ($\boldsymbol{\omega}_0 = \omega_0\hat{\boldsymbol{y}}$).

At a macroscopic level, the presence of the particles in the host fluid changes the effective viscosity, which in the diluted regime is expressed as $\eta_{eff} = \eta_0 + \eta_p + \eta_r$. Here, η_0 is the viscosity of the host fluid, $\eta_p = 5/2\phi\eta_0$ is the contribution due to the mere presence of the particles, with volume fraction ϕ, and η_r is the rotational viscosity, arising from the difference between the mean angular velocity of the particles and that of the fluid: explicitly [6],

$$\eta_r = \frac{3}{2}\eta_0\phi\left(1 - \frac{\langle\Omega_n\rangle}{\omega_0}\right)\,. \tag{6}$$

Notice that Eq. (5) expresses the balance between the hydrodynamic $[\xi_r(\boldsymbol{\omega}_0 - \boldsymbol{\Omega}_n)]$ and magnetic $[\boldsymbol{m} \times \boldsymbol{H}]$ torques. Thereby, Eqs. (5) and (6) make the relationship between force and viscosity explicit, showing that the effective viscosity is an increasing function of the strength of the magnetic torque. The typical form in which the viscosity increases with the field is exemplified by the limiting case of rigid dipole ($\hat{\boldsymbol{R}} \equiv \hat{\boldsymbol{n}}$), where the rotational viscosity is given by $\eta_r = \frac{3}{2}\eta_0\phi(m_0H_0/\xi_r\omega_0)^2$, for $\xi_r\omega_0 > m_0H_0$, and by $\eta_r = \frac{3}{2}\eta_0\phi$, for $\xi_r\omega_0 \leqslant m_0H_0$ [6]. Similar results are also obtained beyond this approximation for the case of low vorticity [9,10], where the rotational viscosity increases with the field reaching a saturation value, as in the case of rigid dipole.

The form of the energy of the particle, however, suggests that the behavior described by Eqs. (1)–(5) can be richer than the ones analyzed up to now. Figs. 1(a) and (b) show the typical form of this energy for low and high intensity of the applied field, respectively. Since under most experimental conditions (for constant applied field) the relaxation of the magnetic moment is much faster than any other time scale entering the system [10,11], the magnetic moment remains always in its energy minimum, following the effective field, $\boldsymbol{H}_{eff} = -\,\partial U/\partial\hat{\boldsymbol{R}}$. Therefore, for low field, the magnetic moment and the easy axis of magnetization have both the same direction. For high field, in contrast, the magnetic moment points around the applied field, irrespective of the direction of the easy axis of magnetization.

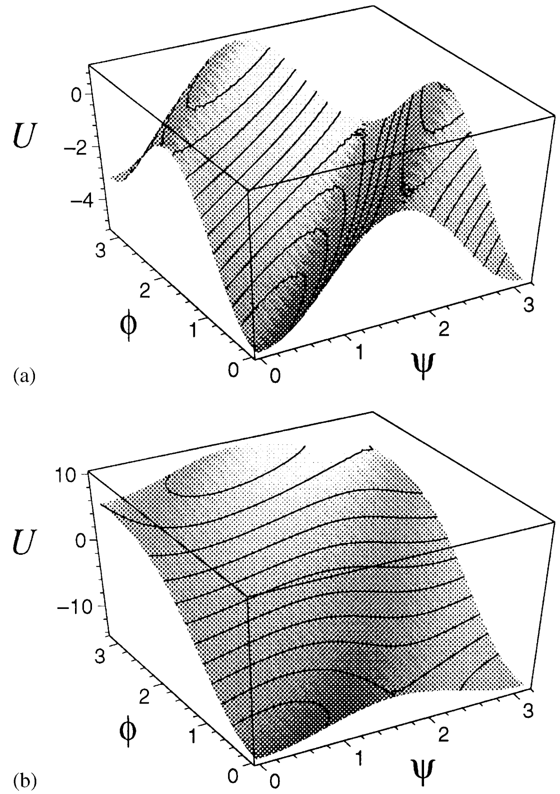

Fig. 1. Energy of the particle [Eq. (1)] as a function of the angle ϕ between the applied field and the magnetic moment $[\cos(\phi) = \hat{\boldsymbol{H}} \cdot \hat{\boldsymbol{R}}]$ and the angle ψ between the magnetic moment and the easy axis of magnetization $[\cos(\psi) = \hat{\boldsymbol{R}} \cdot \hat{\boldsymbol{n}}]$ for (a) $H_0 = 2$ and (b) $H_0 = 10$. The values of the remaining parameters are $K_a V_p = 1$ and $m_0 = 1$.

3. Analytical results

For sufficiently high vorticity, the dynamics still remains unexplored; we now consider explicitly the situation in which the magnetic energy is much higher than the energy of anisotropy. Under these circumstances, the motion of the particle, except perhaps an initial transient, takes place in the plane perpendicular to the vorticity. This enables us to study the dynamics in terms of the angle θ between the applied field and the axis of the particle $[\cos(\theta) = \hat{\boldsymbol{H}} \cdot \hat{\boldsymbol{n}}]$:

$$\frac{\mathrm{d}\theta}{\mathrm{d}t} = \omega_0 \left(1 - \frac{m_0 H_0}{\xi_r \omega_0} R_z \right) . \tag{7}$$

Since the magnetic moment relaxes fast to the effective field, as explained previously, its orientation is given by the minimum of Eq. (1), which for high field leads to

$$R_z = \frac{K_a V_p}{m_0 H_0} \sin(2\theta) . \tag{8}$$

The mean angular velocity then follows from integration of Eq. (7) with (8). For $\xi_r\omega_0 \leqslant K_a V_p$ the angular velocity is zero and the viscosity has the same value as the saturation one obtained for the case of rigid dipole. For $\xi_r\omega_0 > K_a V_p$ the mean angular velocity is given by

$$\frac{1}{\langle \Omega_n \rangle} = \frac{1}{2\pi\omega_0} \int_0^{2\pi} \left(1 - \frac{m_0 H_0}{\xi_r \omega_0} R_z\right)^{-1} d\theta \,, \tag{9}$$

which leads to

$$\eta_r = \tfrac{3}{2}\eta_0\phi(1 - \sqrt{1-\alpha^2}) \,, \tag{10}$$

where $\alpha = K_a V_p/\xi_r\omega_0$ Therefore, for high field the rotational viscosity reaches a constant value. How this constant value is approached indicates whether the viscosity decreases or not as the applied field is increased. In this regard, it is worth studying the case for $m_0 H_0 = K_a V_p$ when the motion of the particle takes place in the plane perpendicular to the vorticity. The minimum of Eq. (1) is then given by

$$R_z = \begin{cases} \sin(2\theta/3) & \text{if } -\pi/4 < \theta < 3\pi/4 \,, \\ \sin(2\theta/3 - 2\pi/3) & \text{if } 3\pi/4 < \theta < 7\pi/4 \,. \end{cases} \tag{11}$$

By substituting this in Eq. (9), we obtain

$$\eta_r = \frac{3}{2}\eta_0\phi \left[1 - \frac{\pi\sqrt{1-\alpha^2}}{3\arctan(\sqrt{3(1+\alpha)/(1-\alpha)})}\right] . \tag{12}$$

The difference between the rotational viscosity at $H_0 = K_a V_p/m_0$ and $H_0 = \infty$,

$$\Delta\eta_r = \frac{3}{2}\eta_0\phi\sqrt{1-\alpha^2} \left[1 - \frac{\pi}{3\arctan(\sqrt{3(1+\alpha)/(1-\alpha)})}\right] , \tag{13}$$

is always positive. Therefore, when $\xi_r\omega_0 > K_a V_p$, the rotational viscosity is not an increasing function of the applied field. Notice that the higher the vorticity, the more pronounced the effect is.

4. Numerical simulations

The situations envisaged previously exhibit a markedly different behavior: for low field the viscosity increases as the field is increased, as usual, whereas for high field the dependence is just the opposite. To elucidate how this change in the behavior of the system takes place we have numerically integrated the corresponding equations following a standard procedure [12,13]. In Fig. 2, we display the rotational viscosity as a function of the intensity of the applied field for different values of the vorticity. For low vorticity, the rotational viscosity saturates to its maximum value at high field following a monotonous increase as for a rigid dipole. Increasing the vorticity leads to the appearance of hysteresis. Two different states exist for intermediate values of the applied field: one with the magnetic moment close to the easy axis of magnetization (rigid-dipole-like) and the other with the magnetic moment oscillating around the

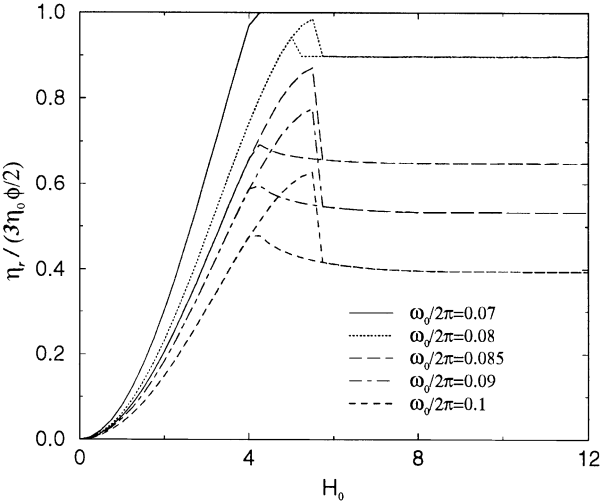

Fig. 2. Normalized rotational viscosity, $\eta_r/(\frac{3}{2}\eta_0\phi)$, as a function of the intensity of an increasing (upper branches) and a decreasing (lower branches) applied field for different values of the vorticity. The values of the remaining parameters are $h = 20$, $g = 1$, $K_a V_p = 4$, $m_0 = 1$, and $\xi_r = 8$.

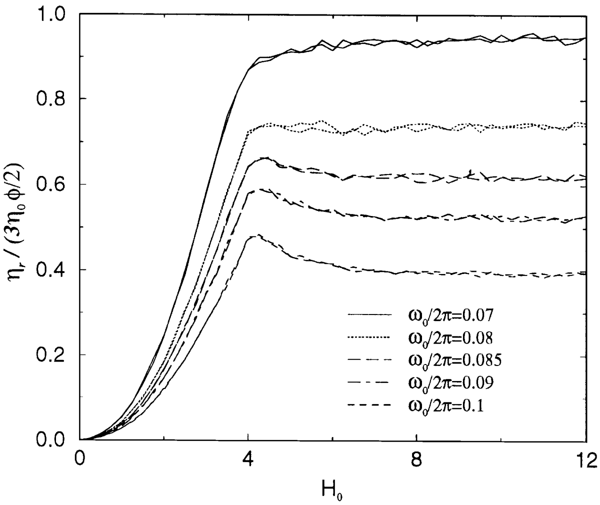

Fig. 3. Same situation as in Fig. 2 but including Brownian motion in the axis of easy magnetization [12]. The rotational diffusion coefficient is $D_r = 1$.

magnetic field. Continuously varying the field, the former is reached from low values, whereas the latter is reached from high values. Notice that for low field the viscosity increases quadratically as a function of the applied field, as shown by the result for a rigid dipole. For high field, in contrast, the viscosity saturates to a constant value as given by Eq. (10). Usually, there is also a stochastic contribution coming from thermal fluctuations [10]. In Fig. 3 we show the same situation as in Fig. 2 but now

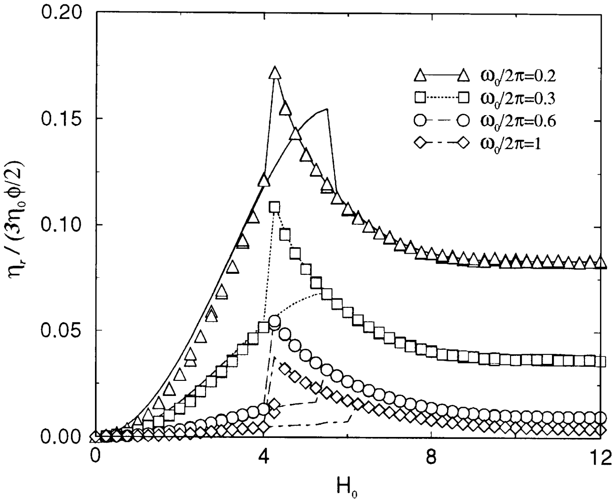

Fig. 4. Same situation as in Figs. 2 and 3 but for different values of the vorticity. Symbols and lines stand for $D_r = 1$ and $D_r = 0$, respectively.

with allowance of Brownian motion in the axis of the particle [12]. It can be seen that a moderate amount of noise does not significantly change the behavior of the particle. However, hysteresis does not appear; the most stable branch is selected. For higher values of the vorticity, the decrease of the rotational viscosity as the field increases is more pronounced, as shown in Fig. 4. This behavior is in agreement with Eq. (13). Again, hysteresis appears in the noiseless case but fades away when Brownian effects take place.

5. Conclusions

To summarize, we have analyzed the behavior of a colloidal ferromagnetic particle in a rotational flow. Beyond a threshold value of the applied magnetic field, for sufficiently high vorticity, the increase of the strength of the field leads to a decrease of its effects: the increase of the field induces the suppression of the force it is responsible for. At a macroscopic level, the monotonous character of the viscosity as a function of the static field reported in the situations analyzed up to now, involving low vorticity, does not occur. Conversely, upon increasing the field, the system passes from a state in which the viscosity increases to another in which the behavior of the viscosity is just the opposite, decreasing and exhibiting hysteresis. These findings indicate that even in apparently simple situations nonlinearities may lead to unexpected changes in the response of the system to an applied field. Our results then open new perspectives about the role played by external fields in the control of the properties of colloidal systems.

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