## Motion of an Asymmetric Ferrofluid Drop under a Homogeneous Time-Dependent Magnetic Field

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A magnetic fluid drop lying on a horizontal solid surface subjected to a vertical magnetic field  $H_0$  peaks with a cone shape above a threshold value  $H_c$ . If  $H_0$  is tilted, i.e., not parallel to the gravitational field, the shape becomes asymmetric. Using a tilted alternating magnetic field we observed the motion of the drop. We have measured this drift velocity, and we propose a simple two-level model to explain this new motion. Finally, we present another experiment with the same device concerning the free surface of a magnetic fluid: the so-called drifting instability. [S0031-9007(96)00688-6]

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The motion of drops has been known since Marangoni [1]. A theoretical explanation for the motion of (nonmagnetic) drops on a solid surface has been given by Brochard [2]: it can be induced by a thermal or a chemical gradient of the spreading coefficient. This drift motion has also been experimentally studied [3]. Although studies of the shape and instabilities of a magnetic fluid (MF) drop have been published in recent years [4–7], no drifting motion of a MF drop has as yet been reported. In this Letter, we present a new experiment where drift motion of a magnetic drop with an asymmetric shape (Fig. 1), is induced by an external homogeneous alternating magnetic field  $H_{\text{ext}} = H_0 \cos(2\pi t/T)$ , which is not parallel to the gravitational field. The motion is produced because the asymmetric drop relaxes in a direction other than  $H_{\text{ext}}$ .

This new type of motion is similar to the motion of particles induced by a periodic asymmetric potential without a macroscopic force [8], or of a mercury drop in an asymmetric structure [9]. In these experiments, the two conditions required for the motion to occur are the presence of a periodic asymmetric potential and of dissipation. In our case, the spatial symmetry is broken by the asymmetric shape of the drop, and the magnetic field time dependence involves a dissipation process because of the drop shape relaxations.

We present the experimental results, and propose a simple two-level model which explains qualitatively this motion. A quantitative prediction of the value of the drift velocity is also given. Finally, we present other experiments concerning a more extended MF system.

Drifting drop: experimental setup and results.—A MF is a colloidal suspension of magnetic particles. We use an ionic MF (a water and glycerol based mixture) with cobalt ferrite particles [10]. The viscosity of the MF is  $\eta_{\rm MF} \approx 0.4 \text{ kg m}^{-1} \text{ s}^{-1}$ . The cell, made of Altuglass, is filled with an organic liquid. The MF droplet is completely nonwetting because of the oil: the drop slides on a thin film of oil in a linear channel dug in the bottom of the cell. The oil viscosity is  $\eta_{\rm oil} \approx 10^{-3} \text{ kg m}^{-1} \text{ s}^{-1}$ .

The cell is placed between two Helmholtz type coils as sketched in Fig. 2; the magnetic field is alternating, linearly polarized, and can rotate around a horizontal axis in order to make an angle  $\alpha$  with the gravitational field  $\vec{g}$  (the frequency equals 1/T = 50 Hz). A time-space diagram is used to determine the drift velocity, and an image processing unit permits the measurements of the height and the area of the MF drop.

If we use a static vertical field, the magnetic dipoles point in the field direction. There is competition between the dipole-dipole interactions which tend to extend the drop in the field direction and the gravity and the surface energy which tend to return the drop to a hemispheric shape. Therefore, to minimize its total energy, the drop becomes a peak above a threshold value of the magnetic field. When the static field is tilted with  $\alpha \neq 0$ , the MF drop shape becomes asymmetric, but no motion is observed. This means that the total body force acting on the drop equals zero for each value of  $\alpha$ . If an alternating field is applied with  $\alpha = 0$ , oscillations of the top of the

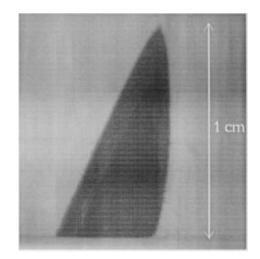


FIG. 1. Photograph of an asymmetric ferrofluid drop under the influence of a tilted alternating magnetic field.

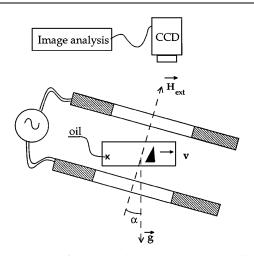


FIG. 2. Scheme of the experimental setup: two coils in the Helmholtz configuration are used to give a homogeneous alternating magnetic field. The coils can be easily tilted through an angle  $\alpha$ .

drop are observed, and a drifting motion occurs when  $\alpha \neq 0$ . Consequently, we deduce that the motion is due only to the relaxation of the asymmetric shape due to the peak top oscillations. This conclusion is the crucial ingredient for the following model.

We have performed many experiments for different angles  $\alpha = 10^{\circ}$ ,  $20^{\circ}$ ,  $30^{\circ}$ ,  $40^{\circ}$ , and for different values of  $H_0$ . The motion is observed even for values of  $H_0$  less than the threshold field  $H_c$  (in this case the drop has an asymmetric ellipsoidal shape). We have experimentally established that the peak base area  $S_0$  and the peak height P are only functions of  $H_0$  and do not depend on  $\alpha$ . The drift velocity  $v_x$  is plotted in Fig. 3:  $v_x$  varies roughly linearly with the product  $P(H_0) \tan(\alpha)$ ; the slope

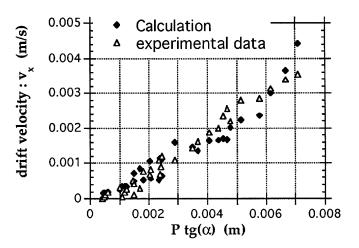


FIG. 3. Experimental data of the drift velocity; *P* is the maximal height of the peak, and  $\alpha$  the angle between the gravitational field and  $H_{\text{ext}}$ . The calculated values are obtained using the experimentally determinated oil film depth *e* under the drop. The slope of the calculated curve has been determined by a least squares method using expression (3).

of this curve defines a characteristic time of the order of  $\theta_{\rm exp}\approx 1.9~{\rm s}.$ 

Now we discuss the two-level model.

(A) Qualitative explanation of the drifting motion.— The magnetic energy of a MF drop, for the values of the magnetic field used here, depends on the square of  $H_{\text{ext}}$ . In this simple model, the time dependence of  $H_{\text{ext}}$ is approximated by a square function with a period T/2(Fig. 4). The phenomenon can thus be separated into two stages, depending on the presence or absence of  $H_{\text{ext}}$ . The drop motion can be explained by the relaxation process of the drop shape during these two stages, using the following assumptions. First, the drop shape is supposed to be conical (the surface curvature is neglected). Second, we assume that the cone generatrix tends to be parallel to the  $H_{\text{ext}}$  direction when  $H_{\text{ext}} = \pm H_0$ .

Suppose p(t) is the cone height, and  $p_{eq}$  the equilibrium height fixed by the field strength  $H_{ext} = \pm H_0$ . The center of gravity of the drop is situated on the cone generatrix, with a height equal to p(t)/4. Let us consider a MF cone with  $H_{ext} = \pm H_0$  (stage 0 in Fig. 4): When the field is switched off, the cone top drops vertically because of gravity (stage 1), with a relaxation time  $\tau_0$ . The center

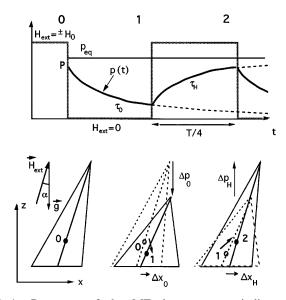


FIG. 4. Response of the MF drop to a periodic square magnetic field. On the upper graph, the hachured line represents the magnetic field square as a function of time. The full line shows the drop height evolution p(t) as a result of two relaxation processes towards equilibrium, with two time scales  $\tau_0$  and  $\tau_H$ . The dotted line represents the relaxation of p(t) towards equilibrium for  $t \to \infty$ . In the lower plot, the drop shape is sketched at different times: stage 0 represents the peak shape when  $H_{\text{ext}} = \pm H_0$ , just before the field switch off, stage 1 squares with the cone breaking down for  $H_{\text{ext}} = 0$ , and stage 2 represents a new state when  $H_{\text{ext}} \neq 0$ . In stage 1, the cone top falls vertically because of gravity, and in stage 2, it moves upwards following the direction of  $H_{\text{ext}}$ . Because of the asymmetric drop shape, the cone top motion generates a translational motion of the center of gravity in the horizontal direction.

of gravity is thus shifted of a quantity  $\Delta x_0$  corresponding to the displacement of the cone generatrix. After a time T/4,  $H_{\text{ext}}$  is switched on: The cone top moves up towards  $p_{\text{eq}}$  following the generatrix direction, without reaching the equilibrium state because the relaxation time  $\tau_H$  is longer than T/4 (stage 2). Let us call P the maximum value reached by p(t); P depends on the period and the relaxation times. A displacement,  $\Delta x_H$ , of the center of gravity is produced in this stage.

(B) Drifting velocity formula.—The expression for the center of mass coordinates  $\vec{r} = (x, z = p/4)$ is given by the balance of the forces acting on the drop. It can be demonstrated that the motion is overdamped; thus the inertial term is neglected. We obtain  $\vec{F}_{0/H}(\vec{r}) = \eta_{\text{oil}}(S_0/e)v_x\vec{e}_x + \eta_{\text{MF}}f(p_{\text{eq}})V_0^{1/3}\vec{v}$ , where  $f(p_{\text{eq}})$  is a dimensionless function of the drop geometry,  $V_0 = 25 \text{ mm}^3$  the drop volume, *e* the oil film depth under the cone, and  $\vec{v} = \vec{r}$ . The term on the left-hand side is the total force acting on the drop, respectively, without (0) and with (H) an external magnetic field. On the right-hand side, the first term represents the dissipation due to the oil shear under the drop [11], and the second term is the Stokes force [11], which describes the viscous dissipation inside the drop. Let us call  $R = (P \tan(\alpha)/4, P/4)$  the maximum position of the center of mass (we assume that the cone generatrix is parallel to the field direction when  $\vec{r} = \vec{R}$ ).  $F_{0/H}(\vec{r})$  can be developed to the first order since the drop displacement  $\delta \vec{r} = \vec{r} - \vec{R}$ remains small (in our experiment,  $\delta p/p \approx 5 \times 10^{-2}$ ):  $\vec{F}_{0/H}(\vec{r}) \approx \vec{F}_{0/H}(\vec{R}) - K_{0/H}\delta\vec{r}$ . In the horizontal direction the Stokes force can be neglected regarding the oil shear under the drop; it means that the drop is considered in the horizontal direction as a rigid object. We have

$$\eta_{\rm MF} f(p_{\rm eq}) V_0^{1/3} \delta \dot{z} + K_{0/H} \delta z - \vec{F}_{0/H}(\vec{R}) \cdot \vec{e}_z = 0, \eta_{\rm oil} \frac{S_0}{e} \delta \dot{x} + K_{0/H} \delta x - \vec{F}_{0/H}(\vec{R}) \cdot \vec{e}_x = 0,$$
<sup>(1)</sup>

with  $x = z \tan(\alpha)$ . We thus obtain exponential relaxations for vertical motions with relaxation times  $\tau_0$  and  $\tau_H$  related to the unknown constant  $K_{0/H}$  by  $\tau_{0/H} = \eta_{\text{MF}} f(p_{\text{eq}}) V_0^{1/3} / K_{0/H}$ . Solving (1) during a period T/2 and using the continuity condition  $\vec{r}_0(T/4) = \vec{r}_H(T/4)$ , both displacements of the center of mass are given by

$$\Delta x_0(t) = \frac{3}{4} P \tan(\alpha) [1 - e^{-t/\theta_0}]$$
  

$$\Delta x_H(t) = \frac{1}{4} P \tan(\alpha) \frac{1 - e^{-T/4\tau_0}}{e^{T/4\tau_H} - 1} e^{T/4\theta_H}$$
  

$$\times [1 - e^{-(t - T/4)/\theta_H}]$$
(2)

with

$$heta_{0/H} = rac{\eta_{
m oil}}{\eta_{
m MF}} rac{S_0}{eV_0^{1/3}} rac{ au_{0/H}}{f(p_{
m eq})}.$$

The drifting velocity of the drop is given by  $v_x = [\Delta \dot{x}_0(t) + \Delta \dot{x}_H(t)]/2$ .

In order to estimate the characteristic relaxation times  $\tau_0$  and  $\tau_H$ , we have performed measurement of p(t)with a stroboscopic apparatus. The results show the exponential decay of the droplet height as suggested above. We obtain that  $\tau_0$  and  $\tau_H$  are of the order of 1 s. The variation of the oil film depth e with respect to the parameters  $H_0$  and  $\alpha$  must also be determined. On the theoretical side, *e* is a solution of the balance of magnetic, hydrostatic, curvature, and disjoining pressures on the cone base [12]. However, the complex geometry of the drop makes the problem somewhat difficult and it is impossible to determine e ab initio. Therefore, we obtain the values of e by measuring the sliding velocity  $v_s$  of the drop along an inclined plane. The balance between gravity and oil-shearing stress gives an expression for e:  $\eta_{\rm oil} S_0/e = {\rm mg sin}(\beta)/v_s$ , where  $\beta$  is the plane tilting angle. In these experiments, a tilted static magnetic field is applied in order to reproduce the asymmetric shape of the drop; the field is static to avoid the motion induced by the asymmetric shape relaxation.

A first order development of (2) in  $T/\theta$  gives (3):  $v_x/P \tan(\alpha) = (2\theta_0)^{-1}$ . Using the experimental slope  $\theta_{\exp} \approx 1.9$  s and determination of  $\theta_0$  (2) we can deduce the value of the quantity  $f(p_{eq})/\tau_0 \approx 1.4 \text{ s}^{-1}$ , which is reasonable since  $f(p_{eq})$  and  $\tau_0$  are of the order of unity. We can use this determination in order to compare the calculated value and the experimental data as shown in Fig. 3. The agreement is quite good if we consider the simplicity of our model: we consider only an idealized geometry and study the response to a square signal instead of a cosine signal.

Beyond the drop: the drifting surface instability.— Using an elongated MF drop several peaks are nucleated above  $H_c$ . The distance between two peaks is fixed by

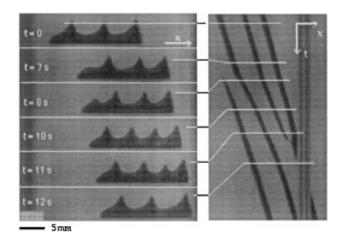


FIG. 5. Photographs (on the left) of the motion of a train made by several peaks, and a time-space diagram (on the right) of this motion. The top line of the peaks is recorded as a function of time (the vertical line is a static defect of the cell).

the capillary length [13]. If the external field is tilted and alternating, we observe the motion of the entire drop as shown in Fig. 5, with a group velocity  $V_x$ . The macroscopic angles of the drop are different at the head and the tail of the drop because of the drift. The shape of the drop is more elongated at the end of the drop and thus becomes unstable with respect to the magnetic field: A new peak is nucleated at the tail of the drop. Since the volume and the distance between two peaks are fixed, this peak has to disappear. In fact, the second peak, starting from the head, disappears. This mechanism (creation of a new peak followed by the annihilation of another peak) occurs with a phase velocity  $v_x$ , which is naturally greater than the group velocity. We have measured  $V_x = 1.26 \text{ mm s}^{-1}$  and  $v_x = 1.72 \text{ mm s}^{-1}$ , for  $\alpha = 10^{\circ}$  and  $H_0 = 4.7 \text{ kA m}^{-1}$ .

If the channel of the cell is completely filled with MF, we obtain a free interface which becomes unstable above  $H_c$ : A periodic line of peaks is formed with a wavelength equal to the capillary length. When the external alternating field is tilted, we can observe the asymmetric peaks which drift at the surface of the MF (corresponding to a phase velocity) without motion of the MF (i.e., the group velocity equals zero). The drifting peak instability is similar to the primary ink instability [14]. The addition of a static magnetic field may allow the obtention of a nonlinear period doubling [15] and, consequently, secondary instabilities; the second field plays the same role than the second roller in the ink instability.

To summarize, we have presented a new experiment where the relaxation of an asymmetric magnetic fluid drop, subjected to a homogeneous alternating magnetic field, gives rise to translational motion. This new kind of mechanism could be used to build liquid motors.

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