Ferrofluid drops in rotating magnetic fields

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Abstract. Drops of a ferrofluid floating in a non-magnetic liquid of the same density and spun by a rotating magnetic field are investigated experimentally and theoretically. The parameters for the experiment are chosen such that different \textit{stationary} drop shapes including \textit{non-axis-symmetric} configurations could be observed. Within an approximate theoretical analysis the character of the occurring shape bifurcations, the different stationary drop forms, as well as the slow rotational motion of the drop is investigated. The results are in qualitative, and often quantitative agreement, with the experimental findings. It is also shown that a small eccentricity of the rotating field may have a substantial impact on the rotational motion of the drop.

Contents

1. Introduction 2
2. Experimental results 3
3. Theoretical analysis 6
   3.1. Linear magnetization law ............................. 9
   3.2. Non-linear magnetization law ........................... 14
   3.3. Elliptically polarized field ............................. 16
4. Conclusion 18
Acknowledgment 19
References 19
1. Introduction

The equilibrium shapes of rotating fluid bodies are of importance in various fields of physics. Following the famous controversy between Newton and Cassini on whether the Earth has the form of an oblate or prolate rotational ellipsoid a series of ingenious investigations by Maupertius, MacLaurin, Jacobi, Riemann, Poincaré and others elucidated the equilibrium shapes of heavenly bodies [1]. Optimizing the efficiency of nuclear fission requires an analysis of the shapes of the participating nuclei [2]. Related problems of more recent interest concern rotating non-neutral plasmas, laser cooled in a Penning trap [3], and tank-treading elliptical membranes [4] in a shear flow which have been used to model the motion of human red blood cells [5]. Particularly interesting from the point of view of pattern formation is the possibility of stationary, non-axis-symmetric configurations as, for example, found theoretically by Jacobi for rotating stars [1] and realized experimentally in rotating plasmas [3].

To investigate the equilibrium shapes of rotating bodies in the laboratory one may create zero-gravity conditions by immersing fluid drops in another immiscible liquid of the same density. Rotating shafts [6] or acoustic torques [7] may then be used to spin up these drops. For drops made from either electrically or magnetically polarizable fluids rotating electric [3] or magnetic fields [8]–[10] are a convenient tool to set up a rotation drop.

In this paper we investigate ferrofluid drops in different types of rotating magnetic fields experimentally and theoretically. Ferrofluids are suspensions of ferromagnetic nano-particles in suitable carrier liquids combining the hydrodynamic behaviour of Newtonian liquids with the magnetic properties of super-paramagnets [11]. In the absence of an external magnetic field the magnetization is zero and in contrast to what the name ‘ferrofluid’ might suggest there is no spontaneous long-range order. For a non-zero external field the magnetization of the ferrofluid builds up with magnetic susceptibility values 4–7 orders of magnitude larger than for atomic paramagnets.

Many of the fascinating properties of ferrofluids result from the interaction between their hydrodynamic and magnetic degrees of freedom. A rotating external magnetic field induces a rotation of the ferromagnetic nano-particles. Due to the viscous coupling of the particles to the surrounding liquid the angular momentum is transferred to the whole drop. Depending on the rotation frequency of the magnetic field two different situations are conceivable. One is close to the well known situation of a static field in which a floating ferrofluid drop elongates along the field direction [12]. If the field rotates very slowly the elongated drop will follow the field rotation quasi-adiabatically with a small phase lag [13]–[15]. If, on the other hand, the field frequency is much higher than the inverse relaxation time of the drop shape the drop cannot follow the field rotation and will instead feel an averaged isotropic horizontal field. This in turn gives rise to an oblate spheroidal shape of the rotating drop [8, 16]. In this paper we will only investigate this regime of high field frequency. In particular, we will show that for suitable parameters the drop acquires a stationary non-axis-symmetric shape with reproducible shape bifurcations when the strength of the external field is altered.

The first systematic investigation of the behaviour of magnetic drops under the influence of a fast rotating magnetic field was performed by Bacri et al [8]. For both small and large magnetic fields an oblate spheroid was found to be the stationary shape of the drop. For large fields the spheroid is crowned by peaks at the periphery which are due to the normal field instability [11, 17] and induce a ‘spiny starfish’ appearance. For intermediate values of the magnetic field various transient shapes were found. Theoretically it was noted that for these values of the magnetic
field the axis-symmetric state may become unstable for sufficiently large susceptibility of the ferrofluid [18]. Due to the special preparation techniques used to synthesize the magnetic fluid the experiments in [8] were characterized by quite unusual parameters. The drops were rather small with a typical radius $R \simeq 10 \mu m$, the magnetic susceptibility was very high, $\chi \simeq 3, \ldots, 5.5$, the interface tension between the ferrofluid and the surrounding liquid was extremely small, $\alpha \simeq 10^{-3}$ dyn cm$^{-1}$, and the viscosity of the ferrofluid was very high, $\eta \sim 1$ P. These parameters give rise to very elongated and instationary shapes at intermediate field values which made a detailed comparison between theory and experiment in this regime impossible.

In contrast, our experiments are characterized by rather different parameters. We use much larger drops ($R \simeq 2, \ldots, 5$ mm) with smaller susceptibility values, $\chi \simeq 0.3, \ldots, 2$. The interface tension is about 3 dyn cm$^{-1}$ as for usual liquids and the viscosities of the ferrofluid and of the surrounding liquid are comparable. This enables us to experimentally produce well-defined transitions to stationary non-axis-symmetric shapes and back to rotationally invariant forms in quantitative agreement with our theoretical findings.

Part of our results have already been published in the short communication [10] where emphasis was placed on the influence of saturation effects on the shape and rotational frequency of the drop. Accordingly, in this paper saturation effects will be dealt with only in brief. On the other hand we include new experimental material shown in the attached movies, present much more details of our theoretical analysis, and extend our experimental and theoretical investigations to the case of an elliptically polarized field.

2. Experimental results

The experiments were performed with a kerosene-based magnetic fluid with dispersed magnetite particles of about 10 nm diameter stabilized with oleic acid. The original ferrofluid has a static magnetic susceptibility $\chi_s \approx 1.91$ and a density $\rho_m = 1.8$ g cm$^{-3}$. Different samples of magnetic fluid were prepared by diluting this fluid with pure kerosene resulting in susceptibilities between 0.31 and 1.54 and densities between 1.2 and 1.75 g cm$^{-3}$. The drops were immersed in 3-brome-1, 2-propandiol with a density of $\rho_p = 1.8$ g cm$^{-3}$. It can be diluted by pure water to reduce its density down to the desired value. A major advantage of 3-brome-1, 2-propandiol compared to other possible liquids is that the physical properties of its interface with the ferrofluid remain reproducible for several days.

The experiments started with the preparation of the surrounding fluid. To obtain the density and density gradient necessary to keep the ferrofluid drop floating near the centre of the container 3-brome-1, 2-propandiol is placed into a glass container and the appropriate amount of water is added to the top of the fluid. After 1–2 days the desired density and density gradient had developed by diffusion. Next the ferrofluid drop was placed near the centre of the container with a syringe. The drop radius value was between 0.2 and 0.5 cm.

The rotating magnetic field was produced by two perpendicular pairs of Helmholtz coils operated with an alternating current of frequency $f = 560$ Hz and a relative phase shift between the pairs of $\pi/2$. Since the typical relaxation time $\tau_S$ of shape deformations of the drops used was about 0.1 s the condition $\omega \tau_S = 2 \pi f \tau_S \gg 1$ for a fast rotating field is very well satisfied. The accuracy in the experimental control of the magnetic field strength $G$ was better than 1%, likewise the deviations from spatial homogeneity in the region of the drop was less than 1%. The field amplitude $G$ ranged between 0 and about 50 Oe.

Since we aim to compare the experimental results with our theoretical analysis the system
parameters were determined in independent measurements before the main experiment started. The complete static magnetization curve $M(H)$ of the ferrofluid was recorded from which in particular the initial static susceptibility $\chi_s$ and the saturation magnetization $M_\infty$ were extracted. Then the dynamic susceptibilities $\chi_1$ and $\chi_2$ forming the complex susceptibility $\chi = \chi_1 - i\chi_2$ were determined. This was carried out in an alternating magnetic field with the experimentally relevant frequency $f = 560$ Hz. In all cases $\chi_2$ was a factor of 7–10 smaller than $\chi_1$ such that the condition $(\chi_2/\chi_1)^2 \ll 1$ holds. However, since the rotation of the ferrofluid drop is a magneto-dissipative effect $\chi_2$ will nevertheless play an important role in the theoretical analysis. The dynamic viscosities $\eta(i)$ and $\eta(e)$ of the ferrofluid and the external fluid respectively were determined by standard rheological methods. For all viscosities measured the liquids behave as Newtonian fluids. Finally the interface tension $\alpha$ was determined by measuring the elongation of the drop in a static external field [12]. The values of $\alpha$ are between 2 and 5 dyn cm$^{-1}$. Whereas the accuracy in the determination of the susceptibilities $\chi_s$, $\chi_1$ and $\chi_2$ can be estimated to be better than 1% the determination of $\alpha$ could only be performed to within 3–5% for the diluted samples ($\chi_s \simeq 0.3$) and with an error of about 7–10% in the case of concentrated ferrofluids ($\chi_s \simeq 1.8$).

In the main experiment the behaviour of the drops in a rotating magnetic field of constant frequency and variable amplitude was investigated. To this end the amplitude of the field is increased in steps with sufficient time for the drop to adapt. The two top views of a typical experiment are shown in figure 1, the movie movie1 contains a complete experimental run using a time lapse factor of 5.

At zero field the shape is of course spherical. Increasing the field the drop remains circular when seen from the top but increases its radius. This corresponds to the flattening of the drop to a shape very well approximated by an oblate ellipsoid of revolution. At this stage the drop performs a hard-body rotation with very small angular velocity $\Omega$ which is difficult to discern. When the field strength reaches $G = 15.2$ Oe the axisymmetric shape becomes unstable and the drop assumes a form similar to a three-axis ellipsoid characterized by three semi-axes $a > b > c$. The ratio $\epsilon_b = a/b$ between the largest and the second largest semi-axis can be measured from the top view, the ratio $\epsilon_c = a/c$ between the largest and the smallest ones is extracted from the side view. It is these ratios that will be compared with the theory in what follows. The elongated shape in the top view makes the determination of the rotation velocity now easy. The value of $\Omega$ is about 0.2 s$^{-1}$, and the error in its experimental determination is about 1%. At $G = 30.4$ Oe peaks start to develop at the periphery of the drop. They are a manifestation of the normal-field or Rosensweig instability [11, 17]. The shape of the drop is now rather complicated and appears to be non-stationary. However, already for a slightly increased field, $G = 34.2$ Oe, a stationary shape is re-established. The basic drop form is now again axis-symmetric with several peaks at the periphery.

With increasing field the drop remains spheroidal. The number of peaks increases with their individual size simultaneously decreasing. For large field values the drop shape is hence again well approximated by an oblate ellipsoid of revolution, see the right-hand part of figure 1. On the other hand, and in contrast to what happened for small field values, the existence of the tiny peaks at the periphery allows the determination of the rotation frequency with the same accuracy of about 1% as for the three-axis ellipsoid. This will enable us to make a quantitative comparison between theory and experiment also for these large values of the external field. The rotation is now much faster than for the elongated form at intermediate field strength $\Omega \simeq 1$ s$^{-1}$.

The described scenario of shape bifurcations depends on the value of the magnetic
Figure 1. The top view of two typical shapes of the rotating drop. On the left (movie2) we show an elongated form well approximated by a three-axis ellipsoid. On the right (movie4) we show a flat spheroid with peaks at the periphery at large values of the magnetic field amplitude. The movies show the slow rotation of the drop in real time. The complete evolution of the drop shape for increasing field strength is available (movie1) with a time-lapse factor of 5. The amplitude $G$ of the magnetic field is shown at the bottom and also graphically displayed by the blue bar on the left. The parameters of this particular experiment are collected in table 1. A particularly intriguing phenomenon occurs after 55 s (4 min 33 s real time) when the external field is switched off. The relaxation to the spherical drop occurs via an intermediate non-axis-symmetrical shape. Switching on the field to the same value again the drop returns immediately to the ‘starfish’ configuration it showed before. This rather peculiar behaviour is shown in real time in movie3.

Table 1. Parameter values relevant to the experimental results reported in the movies related to figure 1 (first row), in figures 3 and 4 (second row) and in the movie related to figure 6 (third row) respectively.

| $R$ (cm) | $\chi_s$ | $\chi_1$ | $\chi_2$ | $M_\infty$ (G) | $\eta^{(i)}$ (P) | $\eta^{(e)}$ (P) | $\alpha$ (dyn cm$^{-1}$) |
|----------|----------|----------|----------|----------------|-----------------|----------------|------------------|
| 0.26     | 1.93     | 1.42     | 0.25     | 90             | 0.32            | 1.35           | 3.1              |
| 0.275    | 1.35     | 1.14     | 0.17     | 80             | 0.19            | 0.58           | 2.8              |
| 0.28     | 1.57     | 1.43     | 0.17     | 77             | 0.22            | 0.14           | 3.0              |

susceptibility. If the susceptibility is small, $\chi_1 \lesssim 0.32$ no transition to a non-axis-symmetric state occurs and we observe nothing but a continuous flattening of the drop with increasing field strength. If $0.32 \lesssim \chi_1 \lesssim 1.14$ the shape transitions are forward bifurcations, i.e. they occur for increasing and decreasing field at the same values of the field amplitude. For still higher susceptibilities both transitions show up as backward bifurcations and are hence accompanied by hysteresis. Measurements of the transition points for drops with different susceptibilities are denoted by the symbols in the shape diagram in figure 2.

For the detailed comparison with theory, as discussed in what follows, figure 3 shows the
dependence of the geometry ratios $\epsilon_b$ and $\epsilon_c$ on the magnetic bond number

$$B = \frac{G^2 R}{\alpha}$$

measuring the external field strength. Note that the uncertainty regarding the exact value of $\alpha$ translates into an error in the magnetic bond number of about 7\%. Figure 4 shows the experimental results for the rotation frequency of the elongated drop and the crowned spheroid at high field values respectively. The relevant parameters for these measurements are displayed in the second row of table 1.

3. Theoretical analysis

To theoretically analyse the experiments presented above in full generality is a formidable task. The impact of the magnetic field on the ferrofluid drop is described by the magnetic stress tensor [19]

$$\sigma_{ik}^m = \frac{1}{4\pi}(H_i B_k - \frac{1}{2}H^2\delta_{ik}) + \frac{1}{2}(M_i H_k - M_k H_i)$$

where $B$, $H$ and $M$ denote the magnetic induction, the magnetic field and the magnetization respectively. A complete analysis would require one to simultaneously solve the free-boundary value problem for the hydrodynamics of both fluids and the magneto-static Maxwell equations.
Figure 3. The ratio $\epsilon_b = a/b$ between the two largest semi-axes (left) and ratio $\epsilon_c = a/c$ between the largest and smallest semi-axes (right) of a rotating ferrofluid drop as a function of the magnetic bond number $B$. The parameters are specified in table 1. The symbols represent the experimental results with filled symbols corresponding to increasing field strength and empty symbols to decreasing field strength. The red curves show the theoretical results for a linear magnetization law $M = \chi H$ and the green ones for the magnetization curve $M(H)$ as determined from an independent experiment respectively. Full curves correspond to stable configurations, dotted curves to unstable ones.

for the magnetic fields $B$ and $H$. This is, even numerically, highly demanding and we therefore propose several approximations which on the one hand describe the experimental situation to sufficient accuracy and on the other hand render the theoretical problem tractable. Our basic assumptions are as follows:

1. The shape of the drop is approximated by a three-axis ellipsoid. This assumption describes the experimental situation rather well except for the intermediate values of the magnetic field for which large peaks develop at the periphery of the drop. Within this approximation the shape of the drop is uniquely described by the values of the three semi-axes $a \geq b \geq c$. In fact, because of volume conservation, two parameters are sufficient which we choose to be the semi-axes ratios $\epsilon_b = a/b$ and $\epsilon_c = a/c$. The theoretical analysis simplifies considerably since the magneto-static problem of a magnetizable ellipsoid in a homogeneous external field can be solved analytically [20] and both the internal field $H$ and the magnetization $M$ are known to be homogeneous.

2. The shape is assumed to be determined solely from the balance between surface energy and magnetic energy. This assumption can be justified by estimating the different stresses relevant to the problem. The capillary stress can be estimated by $p_c \sim 2\alpha/R$, with $\alpha \simeq 2.5$ dyn cm$^{-1}$ and $R \simeq 0.25$ cm (cf table 1) we hence get $p_c \simeq 20$ dyn cm$^{-2}$. The magnetic normal stress is given by $p_m \sim 2\pi M_n^2$, where $M_n$ is the normal component of the fluid magnetization [11]. For a spherical drop we have $M = 3\chi/(3 + 4\pi \chi) G$ [20] and using $\chi \simeq 1.3$, $G \simeq 20$ Oe as well as $M_n \simeq M/2$ we end up with $p_m \simeq 13$ dyn cm$^{-2}$. The viscous stresses can be estimated as $p_v \sim \eta(\epsilon)\Omega$. With the experimental values $\eta(\epsilon) \simeq 1$ P and $\Omega \lesssim 1$ s$^{-1}$ we get $p_v \lesssim 1$ dyn cm$^{-2}$. Hence the capillary and the magnetic stresses are comparable whereas
Figure 4. The rotation frequency of the drop as function of the magnetic bond number for an elongated drop (left) and a disc-like spheroid (right). The parameters are given in table 1. The squares represent the experimental values, the red curves theoretical results using a linear magnetization law and the green ones using the magnetization curve $M(H)$ as determined in an independent experiment, respectively.

the viscous stress is at least one order of magnitude smaller. For the determination of the shape of the drop the viscous stress is therefore negligible. Nevertheless the viscous stresses will be crucial in the analysis of the motion of the drop.

(3) The hydrodynamic flow problem can be treated within the Stokes approximation. This assumption is reasonable since the Reynolds number can be estimated as $Re \sim \rho R^2 \Omega / \eta(e)$ and with $\rho \lesssim 1.8 \text{ g cm}^{-3}$ we find $Re \lesssim 0.9$. We are hence allowed to neglect the inertial term in the Navier–Stokes equation which makes the hydrodynamic equations linear. In our solution of the flow problem we will take advantage of this linearity by exploiting a superposition ansatz.

(4) The flow inside the drop in the co-rotating coordinate system is horizontal and of uniform vorticity. This final assumption builds on the last one, i.e. the dominance of the viscous terms in the flow problem. The stationary internal flow field $\mathbf{v}^{(i)}$ can then be well approximated by the two-dimensional elliptical form

$$\mathbf{v}^{(i)} = \left( -\zeta y \frac{a}{b}, \zeta x \frac{b}{a}, 0 \right)$$

matching the shape of the drop. The parameter $\zeta$ characterizing the vorticity of the flow remains to be determined. Note that this ansatz is hence more general than just describing a solid body rotation of the drop given by the special case $\zeta = 0$.

In view of the above assumptions the problem is now subdivided into two feasible subtasks which can be solved one after the other. In the first one the shape of the ferrofluid drop is determined by minimizing the sum of the surface energy and of the time-averaged magnetic energy. In the second one the rotation frequency of the drop is calculated assuming that the shape is known. In the following we will work out this program assuming a linear relationship between the magnetization and magnetic field, i.e. $M = \chi H$. 

New Journal of Physics 5 (2003) 57.1–57.20 (http://www.njp.org/)
3.1. Linear magnetization law

The equilibrium between the two normal stresses due to capillarity and magnetization can be rewritten as an extremum condition for the sum of the surface and magnetic energies

\[ E = \alpha S + \overline{E_m}. \]  

where the time-dependent magnetic energy has to be averaged over one period of the applied magnetic field as indicated by the overbar. \( S \) simply denotes the surface area of the drop. For a three-axis ellipsoid with volume \( V = 4\pi R^3/3 \) and semi-axis ratios \( \epsilon_b = a/b \) and \( \epsilon_c = a/c \) it is given by

\[ S = 2\pi R^2 \epsilon_b^{2/3} \epsilon_c^{-4/3} \left[ 1 + \frac{\epsilon_c}{\epsilon_b \sqrt{\epsilon_c^2 - 1}} \left( F(m, \kappa) + (\epsilon_c^2 - 1) E(m, \kappa) \right) \right]. \]  

Here \( F \) and \( E \) are elliptic integrals of the first and second kind, respectively [21], and

\[ m = \frac{\sqrt{\epsilon_c^2 - 1}}{\epsilon_c} \quad \text{and} \quad \kappa = \sqrt{\frac{\epsilon_c^2 - \epsilon_b^2}{\epsilon_c^2 - 1}}. \]  

The time-dependent magnetic energy is given by [20]

\[ E_m(t) = -\int_V \int_0^{G(t)} \, dG' \, M(r, G') \]  

where the first integral is over the volume of the drop. Since the magnetization of an ellipsoid in a spatially homogeneous field is also homogeneous this integral can be trivially performed. The complete solution of the magneto-static problem also yields that the following relation holds between the magnetization \( M \), the external field \( G \) and the internal field \( H \) in the ferrofluid [20]:

\[ G_x = H_x + 4\pi n_1 M_x, \quad G_y = H_y + 4\pi n_2 M_y. \]  

Here \( n_1 \) and \( n_2 \) denote the demagnetizing factors along the \( x \) and \( y \) axes, respectively, which are also known functions of \( \epsilon_b \) and \( \epsilon_c \), and can be expressed in terms of elliptic integrals [20].

Within the approximation of a linear magnetization law \( M = \chi H \) we hence also find a linear relation between \( M \) and \( G \) and the magnetic energy (7) can be further simplified:

\[ E_m(t) = -V \int_0^{G(t)} \, dG' \, M(G') = -\frac{V}{2} M \cdot G. \]  

It is now most convenient to use a complex notation for the magnetic fields and to describe the reactive and dissipative magnetic response of the ferrofluid by a complex susceptibility \( \chi = \chi_1 - i\chi_2 \). Accordingly the external magnetic field is specified by \( G = \text{Re} \{ \hat{G} \} \) where

\[ \hat{G} = Ge^{\text{tot}}(1, -i, 0). \]  

We then find from (8) for the magnetization \( M = \text{Re} \{ \hat{M} \} \) with

\[ \hat{M} = G\chi \exp(\text{tot})(\frac{1}{1 + 4\pi \chi n_1}, -i, \frac{1 + 4\pi \chi n_2}{1 + 4\pi \chi n_2}, 0). \]  

To perform the time average of the magnetic energy over one period of the rotating field we use

\[ M \cdot G = \frac{1}{4} (\hat{M} + \hat{M}^*) \cdot (\hat{G} + \hat{G}^*) = \frac{1}{4} (\hat{M} \cdot \hat{G} + \hat{M}^* \cdot \hat{G}^* + \hat{M}^* \cdot \hat{G} + \hat{M} \cdot \hat{G}^*). \]
The first and last terms in this sum disappear after the time average, the other two are constant in time and remain. In this way we obtain for the averaged magnetic energy

$$
\tilde{E}_m = -\frac{V G^2}{8} \left( \frac{\chi + \chi^* + 8\pi |\chi|^2 n_1}{|1 + 4\pi \chi n_1|^2} + \frac{\chi + \chi^* + 8\pi |\chi|^2 n_2}{|1 + 4\pi \chi n_2|^2} \right).
$$

If we replace $\chi = \chi_1 - i\chi_2$ in this expression by $\chi_1$ the resulting error is of order $(\chi_2/\chi_1)^2$ which is about one per cent for the data relevant to our experiment. The shape of the drop is hence indeed scarcely influenced by magneto-dissipative effects as already anticipated above. The averaged magnetic energy hence simplifies to

$$
\tilde{E}_m = -\frac{V G^2 \chi_1}{4} \left( \frac{1}{1 + 4\pi \chi_1 n_1} + \frac{1}{1 + 4\pi \chi_1 n_2} \right),
$$

and we find for the dimensionless total energy

$$
\frac{E}{2\pi R^2} = \epsilon_c^{2/3} \epsilon_{-4/3} \left[ 1 + \frac{\epsilon_c}{\epsilon_b \sqrt{\epsilon_c^2 - 1}} \right] \left( F(m, \kappa) + (\epsilon_c^2 - 1)E(m, \kappa) \right) - \frac{\chi_1}{6} B \left( \frac{1}{1 + 4\pi \chi_1 n_1} + \frac{1}{1 + 4\pi \chi_1 n_2} \right),
$$

with $B$ defined in (1). The equilibrium shape is given by the minimum of this function with respect to the geometry parameters $\epsilon_b$ and $\epsilon_c$, which in general has to be determined numerically.

One extremum of (13) corresponds to an oblate spheroid characterized by $a = b > c$. In this case $n_1 = n_2 = n$ and the expression for the total energy simplifies to

$$
\frac{E}{2\pi R^2} = \epsilon_c^{2/3} + \epsilon_c^{-4/3} \ln \left( \frac{1 + m}{1 - m} \right) - \frac{B \chi_1}{3} \frac{1}{1 + 4\pi \chi_1 n}.
$$

Moreover, $n$ can be expressed in terms of elementary functions [20] and the derivative with respect to $\epsilon_c$ can be explicitly calculated. As a result we find

$$
B = 4\pi \left( \frac{1}{4\pi \chi_1} + n \right) \frac{2\epsilon_c^2 - 1}{\epsilon_c^{4/3}} \frac{2\epsilon_c^2 - 1 - \hat{n}(4\epsilon_c^2 - 1)}{n(\epsilon_c^2 + 2) - 1}
$$

giving an explicit relation between the magnetic bond number $B$ and the semi-axis ratio $\epsilon_c$. Here $\hat{n}$ denotes the demagnetizing factor along the axis $\hat{a}$ of the so-called additional ellipsoid with semi-axes $\hat{a} = a$, $\hat{b} = ac/b$ and $\hat{c} = c$. The axis-symmetric solution defined by (15) describes the continuous flattening of the drop with increasing field strength.

Although this solution exists for all values of $\chi_1$ and $B$ it may become unstable for intermediate values of $B$ if $\chi_1$ is sufficiently large. More precisely we find that for $\chi_1 > \chi_1^* = 0.325$ the axis-symmetric solution loses its stability within a bounded window of intermediate values of $B$. Here a non-axis-symmetric solution with $a \neq b$ becomes the stable stationary shape. With increasing magnetic field the drop hence transforms from a spheroid to a three-axes ellipsoid and back to a rather flat spheroid at high values of $B$. From the numerical analysis of (13) we find that the first bifurcation is supercritical for $\chi_1^* < \chi_1 < \chi_1^A = 0.843$, and the second one for $\chi_1^b < \chi_1 < \chi_1^B = 1.02$. For larger values of $\chi_1$ these bifurcations become subcritical and the transitions are accompanied by hysteresis. These theoretical findings are displayed in figure 2 and compare well with the experimental results. Note that no fit parameters were used; all parameters of the system were determined in independent experiments.

Whereas the transition lines between the different shapes determined within the approximation of a linear magnetization law reproduce the experimental findings rather well

New Journal of Physics 5 (2003) 57.1–57.20 (http://www.njp.org/)
the actual values for the semi-axis ratios are overestimated. This can be inferred from figure 3 in which the theoretical results for $\epsilon_b$ and $\epsilon_c$ are shown together with the experimental values. In view of the fact that a magnetic bond number of 50 corresponds to an external magnetic field of the order of the saturation magnetization $M_\infty$ of the ferrofluid these differences are no real surprise. The linear magnetization law systematically overestimates the magnetic field energy giving rise to overly large values of the eccentricities, in particular for large field values. We will briefly discuss in the next section whether the situation can be improved by using more realistic magnetization laws including saturation.

We now turn to the theoretical analysis of the rotational motion of the drop. According to our assumption of an elliptical shape of the drop the magnetic field inside the drop is homogeneous and therefore the magnetic force density $(\mathbf{M} \cdot \nabla) \mathbf{H}$ is zero. The stationary flow fields $\mathbf{v}^{(i)}$ and $\mathbf{v}^{(e)}$ of the internal and external fluid, respectively, have hence to satisfy the simple Stokes equations

$$\eta^{(i,e)} \Delta \mathbf{v}^{(i,e)} = \nabla p^{(i,e)}$$

(16)

where $p^{(i)}$ and $p^{(e)}$ denote the pressures inside and outside the drop, respectively. It is convenient to work in the coordinate system in which the drop shape is at rest. For the internal flow we then assume the simple form

$$\mathbf{v}^{(i)} = \left(-\zeta \frac{a}{b}, \zeta \frac{b}{a}, 0\right)$$

(17)

with uniform vorticity

$$\frac{1}{2}(\nabla \times \mathbf{v}^{(i)}) = \frac{\zeta}{2} \left(0, 0, \frac{a}{b} - \frac{b}{a}\right),$$

(18)

implying $p^{(i)} = \text{const.}$

The external flow has the asymptotic behaviour

$$\mathbf{v}^{(e)} \to -\Omega \times r = (y\Omega, -x\Omega, 0) \text{ for } r \to \infty.$$  

(19)

It is very useful to write the external flow field in the form

$$\mathbf{v}^{(e)} = \mathbf{v}^{(i)} + \mathbf{v}^{(J)}.$$  

(20)

Exploiting the linearity of the Stokes equation we infer that $\mathbf{v}^{(J)} = (u^{(J)}, v^{(J)}, w^{(J)})$ describes the Stokes flow around a stationary, rigid ellipsoid with no-slip boundary conditions at the surface of the ellipsoid and asymptotics

$$\mathbf{v}^{(J)} \to \left(y\left(\Omega + \frac{a}{b}\Omega \zeta\right), -x\left(\Omega + \frac{b}{a}\Omega \zeta\right), 0\right) \text{ for } r \to \infty.$$  

(21)

The determination of $\mathbf{v}^{(J)}$ is a classical problem in hydrodynamics solved by Jeffrey many years ago [22]. The solution is parametrized in terms of the asymptotic velocity-gradient tensor

$$\lim_{r \to \infty} \frac{\partial \mathbf{v}^{(J)}}{\partial x_k} = \gamma_{ik} + \omega_{ik}$$

(22)

decomposed into its symmetric and antisymmetric parts $\gamma_{ik}$ and $\omega_{ik}$ respectively. In our case we find from (21) for the relevant components of this tensor

$$\gamma_{12} = \gamma_{21} = \frac{\zeta}{2} \left(\frac{a}{b} - \frac{b}{a}\right),$$

(23)

$$\omega_{12} = -\omega_{21} = \Omega + \frac{\zeta}{2} \left(\frac{a}{b} + \frac{b}{a}\right).$$

(24)
The solution of the rotation problem is hence reduced to the determination of the so far unspecified rotation frequency $\Omega$ of the drop and the vorticity parameter $\zeta$ of the internal flow. To this end two equations are needed. One of these is the balance between the viscous and time-averaged magnetic torque on the drop which is clearly mandatory for a stationary rotation:

$$L^v + L^m = 0.$$  \hfill (25)

The second equation is the integrated balance of tangential stresses at the surface of the drop:

$$\int_{\partial V} dS \nu^i \sigma^{(i)}_{ik} v_k = \int_{\partial V} dS \nu^i \sigma^{(e)}_{ik} v_k.$$  \hfill (26)

Here $\sigma^{(i)}_{ik}$ and $\sigma^{(e)}_{ik}$ denote the stress tensors inside and outside the drop respectively, and $\nu$ is the surface normal. Note that due to the various simplifying assumptions in our theoretical treatment it is impossible to satisfy the continuity of tangential stresses at the surface point-wise. However, it should be fulfilled on average. It can be shown that this requirement is equivalent to the energy dissipation balance in the system stating that per time unit the energy fed into the system by the magnetic field must be equal to the energy dissipated in the viscous flows and the periodic re-magnetization of the ferrofluid [23].

Let us start with the first condition concerning the balance of torques. For symmetry reasons only their $z$-component is different from zero. For the viscous torque it is given by

$$L^v_z = \int_{\partial V} dS (x F_y - y F_x),$$  \hfill (27)

where $F$ denotes the force on an element $dS$ of the drop surface and the integral is over the complete surface of the drop. From Jeffrey’s solution we find for the contribution of $v^{(J)}$ to the force [22]

$$F_i^{(J)} = -p \nu_i + \eta^{(e)} A_{ik} v_k,$$  \hfill (28)

where $p$ is a constant and $A_{ik}$ are linear functions of the velocity-gradient tensor of the external flow. The elements relevant to our geometry are

$$A_{12} = 2 n_1 \gamma_{12} + b^2 n_2^a \omega_{12} \frac{(a^2 n_1 + b^2 n_2)n_3'}{n_3'},$$  \hfill (29)

where $n_3' = (n_2 - n_1)/(a^2 - b^2)$. The value of $A_{21}$ is obtained from $A_{12}$ by the replacements $1 \leftrightarrow 2$ and $a \leftrightarrow b$. Performing the integral over the surface of the ellipsoid we get for the $z$-component of the viscous torque

$$L^v_z = V \eta^{(e)} (A_{21} - A_{12}).$$  \hfill (30)

The magnetic torque is generally given by [20]

$$L^m = \int_V d^3 r \ M \times G$$  \hfill (31)

and for a homogeneously magnetized drop we find for the $z$-component

$$L^m_z = V (M_x G_y - M_y G_x).$$  \hfill (32)

Using again (8) and (9) and averaging over one period of the external field we find by similar calculations as performed for the determination of the averaged magnetic energy

$$\tilde{L}^m_z = \frac{V G^2}{2} \frac{1}{(1 + 4 \pi \chi_1 n_1)^2} + \frac{1}{(1 + 4 \pi \chi_2 n_2)^2}.$$  \hfill (33)
As anticipated we hence find that the time-averaged magnetic torque $\bar{L}_m$ is proportional to the imaginary part $\chi_2$ of the susceptibility and is hence a magneto-dissipative effect. Combining (30) and (33) we find the first equation between $\Omega$ and $\zeta$ in the form

$$A_{12} - A_{21} = \frac{\chi_2 G^2}{2\eta^{(e)}} \left[ \frac{1}{(1 + 4\pi \chi_1 n_1)^2} + \frac{1}{(1 + 4\pi \chi_1 n_2)^2} \right].$$

(34)

The dependence on $\Omega$ and $\zeta$ is here via $A_{ij}$ as defined in (29).

To find the viscous contribution to the integrated balance of tangential stresses (26) we have to calculate

$$\Pi^{(v)} = \int_{\partial V} dS \left( v_i^{(i)} \sigma_{ik}^{(i)} - v_i^{(e)} \sigma_{ik}^{(e)} \right) v_k,$$

where $\sigma_{ik}^{(i)}$ and $\sigma_{ik}^{(e)}$ are the viscous stress tensors inside and outside the drop. Using (17) and (20) we find for the relevant components of the viscous stress tensors of the internal and external flows at the surface of the drop

$$\sigma_{12}^{(i)} = \eta^{(i)} \left( \frac{\partial v_1^{(i)}}{\partial x_2} + \frac{\partial v_2^{(i)}}{\partial x_1} \right) = \eta^{(i)} \zeta \left( \frac{b}{a} - \frac{a}{b} \right),$$

(36)

$$\sigma_{12}^{(e)} = \eta^{(e)} \left( \frac{\partial v_1^{(e)}}{\partial x_2} + \frac{\partial v_2^{(e)}}{\partial x_1} \right) = \eta^{(e)} \left( \zeta \left( \frac{b}{a} - \frac{a}{b} \right) + A_{12} \right).$$

(37)

Performing the integral over the drop surface we eventually get [23]

$$\Pi^{(v)} = \left( \eta^{(i)} - \eta^{(e)} \right) \zeta^2 V \left( \frac{b}{a} - \frac{a}{b} \right)^2 + \eta^{(e)} \zeta V \left( \frac{a}{b} A_{12} - \frac{b}{a} A_{21} \right).$$

(38)

The magnetic contributions to the tangential stresses

$$\Pi^{(m)} = \int_{\partial V} dS \left( v_i^{(i)} \sigma_{ik}^{(m,i)} - v_i^{(e)} \sigma_{ik}^{(m,e)} \right) v_k$$

follow from the magnetic stress tensor (2). Due to the boundary conditions for the magnetic fields $H$ and $B$ the contributions from the Maxwell term $H_i B_k/(4\pi)$ inside and outside the drop cancel each other. The only non-zero contribution comes from the Shliomis term $(M_i H_k - M_k H_i)/2$ describing the non-equilibrium character of the drop magnetization. Using a similar calculation as employed for the determination of the averaged magnetic energy we get

$$\bar{\Pi}^{(m)} = \frac{V G^2}{2} \zeta \chi_2 \left( \frac{a}{b} \right)^2 \left( \frac{b}{a} \right)^2 \left( \frac{1}{1 + 4\pi \chi_1 n_1} \right) \left( \frac{1}{1 + 4\pi \chi_1 n_2} \right).$$

(40)

Combining (38) and (40) we hence find the second equation for the determination of $\Omega$ and $\zeta$:

$$\frac{a}{b} A_{12} - \frac{b}{a} A_{21} - \zeta \left( 1 - \frac{\eta^{(i)}}{\eta^{(e)}} \right) \left( \frac{b}{a} - \frac{a}{b} \right)^2 = \frac{\chi_2 G^2}{2\eta^{(e)}} \left( \frac{a}{b} + \frac{b}{a} \right) \left( \frac{1}{1 + 4\pi \chi_1 n_1} \right) \left( \frac{1}{1 + 4\pi \chi_1 n_2} \right) \left( \frac{1}{(1 + 4\pi \chi_1 n_1)^2} + \frac{1}{(1 + 4\pi \chi_1 n_2)^2} \right).$$

(41)

For a given geometry and material parameters and fixed magnetic bond number equations (34) and (41) give a linear system of equations to determine $\zeta$ and $\Omega$. For the parameters given in the second row of table 1 the results for $\Omega$ are displayed in figure 4. A visualization of the drop motion and the associated internal flow is given in figure 5 and the corresponding animation.

Compared with the experimental results the theoretical values for the rotation frequency are too large. It is tempting to relate the corresponding overestimation of the magnetic torque in the theoretical treatment again to saturation effects which were ignored when assuming a linear magnetization law $M = \chi H$. 

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\[ \varepsilon_b = 1.73 \quad \varepsilon_c = 27.95 \quad \zeta = 0.96 \text{ 1/s} \quad \Omega = 0.47 \text{ 1/s} \]

**Figure 5.** An animated version of the theoretical results for the rotation frequency and the internal flow is shown in movie5. The parameters are the same as in figure 4, the magnetic bond number is \( B = 66 \). The movie shows clearly the difference between the rotation of the drop shape and the vorticity of the internal flow of the fluid. The mass elements of the ferrofluid are coded by the colour and exhibit a rotational flow in the rest frame of the shape.

### 3.2. Non-linear magnetization law

In order to improve the quantitative comparison between experiment and theory the first step is to use a more realistic magnetization law \( M(H) \). Naturally this makes the theoretical analysis more complicated and less accessible to analytical techniques. A detailed theoretical discussion of the implications of a non-linear magnetization law on the problem at hand can be found in [23]. Here we will only mention the main points and present some of the results which can be obtained by using the complete magnetization function \( M = M(H) \) as determined in an independent experiment. Of central importance remains equation (8) which is also valid for non-linear magnetization laws as well as the fact that the internal field is homogeneous.

The simplest case is the shape problem for a spheroid \( a = b > c \). Barring magnetodissipative effects \( G, H \) and \( M \) are then all parallel. This implies from (8) that

\[ H + 4\pi nM(H) = G. \tag{42} \]

For given the magnetization law \( M(H) \) and given geometry factor \( \varepsilon_c \) this equation can be used to determine \( M \) numerically. Combining the result with the generalization of (15) which is of the form

\[ \frac{4\pi M^2 R}{\sigma} = \frac{\varepsilon_c^2 - 1}{n(4\varepsilon_c^2 + 2) - 1} - \tilde{n}(4\varepsilon_c^2 - 1) \tag{43} \]

we again find an equation between \( \varepsilon_c \) and the magnetic bond number \( B \). The results obtained in this way for the experimentally determined \( M(H) \) are included in the right-hand part of figure 3. The agreement between the theoretical and experimental results is markedly improved, in particular for large values of the magnetic bond number \( B \).
For a three-axis ellipsoid the situation is somewhat more complicated since, even neglecting magneto-dissipation, \( G \) and \( H \) are no longer parallel. The relation between \( G, H \) and \( M \) following from (8) is now time dependent:

\[
\frac{1}{G^2} = \frac{\cos^2(\omega t)}{(H + 4\pi n_1 M(H))^2} + \frac{\sin^2(\omega t)}{(H + 4\pi n_2 M(H))^2}
\]

(44)

and accordingly it has to be solved numerically for every time moment. Starting from (7) the time-dependent magnetic energy can be written as

\[
\frac{E_m(t)}{V} = -\left[\frac{G^2 M(H)}{2} \left( \frac{\cos^2(\omega t)}{H + 4\pi n_1 M(H)} + \frac{\sin^2(\omega t)}{H + 4\pi n_2 M(H)} \right) \right.
\]

\[
-\frac{HM(H)}{2} + \int_0^H dH' M(H') \left. \right]\]

(45)

For the linear case \( M = \chi H \) the last two terms cancel and after averaging over one period of the external driving we recover (12). For the general case the time averaging also has to be performed numerically. Proceeding in this way by using the experimentally determined \( M(H) \) values the lower curves in figure 3 were obtained. The reduction for \( \epsilon_b \) is now even too large whereas for \( \epsilon_c \) the agreement with experiment is now rather good.

The analysis of the rotation problem on the basis of a non-linear magnetization law is still more involved than the above-sketched shape determination. This is due to the fact that the generalization of the linear law \( M = \chi H \) with complex susceptibility \( \chi \) to a non-linear dependence \( M(H) \) including magneto-dissipation is a complicated issue.

A useful starting point is the quite general relaxation equation for the magnetization [24]–[26]

\[
\partial_t M = -\frac{1}{\tau_\parallel H^2} [H \cdot (M - M_s(H))] H - \frac{1}{\tau_\perp H^2} H \times (M \times H).
\]

(46)

Here \( M_s(H) \) denotes the equilibrium magnetization corresponding to the magnetic field \( H \), and \( \tau_\parallel \) and \( \tau_\perp \) are the relaxation times of the magnetization parallel and perpendicular to the field \( H \) respectively. Equation (46) can be derived from an approximate solution of the Fokker–Planck equation describing the orientational Brownian motion of the ferromagnetic particles [24, 25] and therefore includes both relaxation and saturation. Combining this equation with (8) a system of ordinary differential equations for the time dependence of the internal magnetic field can be derived which are to be solved numerically. From the solution of these equations for one period of the external field the magnetic torque (32) and the magnetic contribution to the averaged balance of tangential stresses (39) can be calculated. The remaining time average is finally performed numerically. In this way we find again two linear equations for \( \Omega_1 \) and \( \zeta \). The results for \( \Omega(B) \) obtained for the magnetization curve as determined in an independent experiment are included in figure 4.

From the left-hand part of figure 4 it is can be seen that despite the significant reduction of the magnetic torque the results for \( \Omega \) are almost the same as for a linear magnetization law. The formal reason is that due to the smaller eccentricity of the drop (cf figure 3) the viscous torque is also reduced. We therefore expect that other effects than the saturation of the magnetization are responsible for the reduction of the experimental values for the rotation frequency when compared with the theoretical results. Possible candidates are the multi-dispersedness of the ferrofluid reducing the magnetic torque, deviations from the elliptic shape increasing the viscous one, and a small ellipticity in the external magnetic field (see section 3.3 in what follows).
The situation is somewhat more gratifying for a spheroid in large fields. As explained in section 2 the small peaks at the periphery then allow an accurate determination of the rotation frequency. On the other hand the two equations (34) and (41) coincide and give rise to one equation for the sum $\zeta + \Omega$ describing the rigid-body rotation of the drop in the laboratory frame. As can be seen in the right-hand part of figure 4 the theoretical results for the rotation of the spheroidal drop at high field values compare well with experiment. For smaller values of $B$ the experimental rotation frequency is still below the theoretical values which is presumably due to the few and large peaks at the periphery of the drop increasing the viscous torque. For a larger bond number our theoretical model of a spheroid becomes yet more realistic and the theoretical results for the rotation are in good agreement with experiment.

3.3. Elliptically polarized field

It is interesting to study the behaviour of a ferrofluid drop in an elliptically polarized field of the form

$$G = (G_{0x} \cos(\omega t), G_{0y} \sin(\omega t), 0),$$

(47)

where for definiteness we assume $G_{0x} \geq G_{0y}$. On the one hand an elliptically polarized field interpolates between a rotating field as studied above and a linearly polarized field which should in general not induce a rotation of the drop (see, however [27]). On the other hand experimental realizations of a rotating field will always show small eccentricities and it is important to understand their possible implications. We will study this case only within the approximation of a linear magnetization law for which it is again convenient to use $G = \text{Re} \{\hat{G}\}$ with

$$\hat{G} = e^{i\omega t} (G_{0x}, -iG_{0y}, 0).$$

(48)

The ellipticity of the external field will influence both the shape and the rotational motion of the drop. Since the set-up is no longer axis-symmetric the results for the magnetic energy and the magnetic torque will depend on the angle $\theta$ between the $x$-axis of the laboratory coordinate system and the $x'$-axis of the rotating coordinate system defined as the direction of the largest semi-axis $a$ of the rotating drop. As in the case of a circularly polarized field the solution of the shape problem should be the first step of the investigation since it fixes the geometry parameters which are a necessary input for the analysis of the rotational motion. It will turn out, however, that the rotation is much more severely influenced by a small eccentricity of the field than the shape. We will therefore first study the case of the rotation of a ferrofluid drop with fixed geometry in an elliptically polarized field. Afterwards we show that the resulting scenario is hardly modified by the shape modulations induced by the field ellipticity.

Of central importance is the determination of the magnetic torque which is most easily calculated in the rotating coordinate system. The magnetization is then again determined by (8) and we find

$$\dot{M}'_x = \frac{\chi \hat{G}'_x}{1 + 4\pi \chi n_1},$$

(49)

$$\dot{M}'_y = \frac{\chi \hat{G}'_y}{1 + 4\pi \chi n_2},$$

(50)

where the field in the rotating frame is given by

$$\hat{G}' = e^{i\omega t} (G_{0x} \cos \theta - iG_{0y} \sin \theta, -G_{0x} \sin \theta - iG_{0y} \cos \theta, 0).$$

(51)
Figure 6. The top view of a non-axis-symmetric drop in an elliptically polarized magnetic field. The relevant parameter values are collected in the third row of table 1. The relative strength of the field in the $x$ (horizontal in the figure) and $y$ directions, respectively, is shown by the bars on the left. For the values given in the figure the drop had just stopped its rotation. The complete experiment is shown in movie6. Note that in the experiment $G_{0y}$ is negative. The drop hence rotates clockwise and the stopping angle is $\theta = 3\pi/4$ or equivalently $\theta = 7\pi/4$.

The magnetic torque is given by (32) in the co-moving coordinate system. Note that $\partial_t \theta$ is of the order of $\Omega$ and hence small compared to $\omega$. It is hence reasonable to perform the time average over one period of the external field at a fixed value of $\theta$. This gives rise to

$$\bar{L}'_z = \frac{V}{4} (\hat{M}_x \hat{G}_y + \hat{M}_y \hat{G}_x - \hat{M}'_x \hat{G}'_y - \hat{M}'_y \hat{G}'_x).$$

With the help of (49), (50), and (51) and neglecting again terms of order $(\chi^2/\chi_1)^2$ we finally find

$$\bar{L}'_z = \frac{V}{2} \chi_2 G_{0x} G_{0y} \left( \frac{1}{1 + 4\pi \chi_1 n_1} + \frac{1}{1 + 4\pi \chi_1 n_2} \right)$$

$$- \frac{\chi_1}{2} (G_{0x}^2 - G_{0y}^2) \sin 2\theta \left( \frac{1}{1 + 4\pi \chi_1 n_1} - \frac{1}{1 + 4\pi \chi_1 n_2} \right).$$

This result is quite interesting. The first term in the torque is due to magneto-dissipation since it is proportional to $\chi_2$. This is similar to the case of a circularly polarized field and in fact for $G_{0x} = G_{0y} = G$ this term reproduces (33). The second term stems from a reactive response of the fluid as shown by the proportionality to $\chi_1$. It disappears if $G_{0x} = G_{0y}$ or $n_1 = n_2$. Moreover, it depends on the relative orientation $\theta$ between the long axis of the ellipsoid and the direction of the larger magnetic field component $G_{0x}$. Since $n_1 < n_2$ as a result of $a > b$ this term hence describes the tendency of the field to align the ellipsoid with its long axis along the $x$-axis of the laboratory frame.

Therefore, as long as the field ellipticity is not too large such that

$$2\chi_2 G_{0x} G_{0y} \left( \frac{1}{1 + 4\pi \chi_1 n_1} + \frac{1}{1 + 4\pi \chi_1 n_2} \right)$$
\[ \chi_1 (G_{0x}^2 - G_{0y}^2) \left( \frac{1}{1 + 4\pi \chi_1 n_1} - \frac{1}{1 + 4\pi \chi_1 n_2} \right) \]  

(53)

the magnetic torque is positive for all values of \( \theta \) and the drop rotates. With an increasing difference between \( G_{0x} \) and \( G_{0y} \), the torque and therefore also the angular velocity of the drop become very small if \( \theta \) is near to either \( \pi/4 \) or equivalently \( 5\pi/4 \). Finally, if (53) is fulfilled as equality the motion of the drop stops at either \( \theta = \pi/4 \) or equivalently \( \theta = 5\pi/4 \). Increasing the ellipticity of the field further the equilibrium orientation of the drop will be given by the solution of

\[ \sin 2\theta = \frac{\chi_2}{\chi_1} \frac{2G_{0x}G_{0y}}{G_{0x}^2 - G_{0y}^2} \left( \frac{4\pi \chi_1 (n_2 - n_1)}{(1 + 4\pi \chi_1 n_1)(1 + 4\pi \chi_1 n_2)} + \frac{1}{2\pi \chi_1 (n_2 - n_1)} \right) \]  

(54)

until in the case of a linearly polarized field, \( G_{0y} = 0 \), there is complete alignment of the long axis with the external field direction, i.e. \( \theta = 0 \) or equivalently \( \theta = \pi \).

Let us finally return to the shape problem in an elliptically polarized field. Using a similar calculation as above for the torque the magnetic energy can be shown to be of the form

\[ \bar{E}_m = -\frac{V \chi_1}{8} \left[ (G_{0x}^2 + G_{0y}^2) \left( \frac{1}{1 + 4\pi \chi_1 n_1} + \frac{1}{1 + 4\pi \chi_1 n_2} \right) 
\right. 
\left. + (G_{0x}^2 - G_{0y}^2) \cos 2\theta \left( \frac{1}{1 + 4\pi \chi_1 n_1} - \frac{1}{1 + 4\pi \chi_1 n_2} \right) \right]. \]  

(55)

The first term is independent of the orientation \( \theta \) and is simply the generalization of (12). The second term comprises the dependence on \( \theta \). Since both the magnetic and viscous torque values depend on the geometry factors \( \epsilon_b \) and \( \epsilon_c \) a complete solution of the problem would require one to first determine \( \epsilon_b(\theta) \) and \( \epsilon_c(\theta) \), and then to calculate the two torques. However, in view of our findings concerning the two parts in the magnetic torque (52) it is clear that the angle-dependent part of the magnetic energy is by the factor \( \chi_2/\chi_1 \) smaller than the first part. In other words, before the \( \theta \)-dependent modulation of the shape becomes noticeable the rotation of the drop has already stopped. The shape of the static drop is then described by (55) with \( \theta \) given by the solution of (54). Without performing the analysis explicitly here it is clear that for \( G_{0y} \to 0 \) we will reproduce the results found in [12]: in particular we will obtain \( \epsilon_b = \epsilon_c \).

4. Conclusion

Drops of ferrofluid floating in a non-magnetic liquid of the same density and spun by a rotating magnetic field of high frequency show a variety of interesting phenomena. Upon variations of the amplitude of the external field several bifurcations in the stationary shape of the drop occur including transitions to a non axis-symmetric form which can often be well approximated by a three-axis ellipsoid. The occurrence and character of these shape bifurcations depend on the magnetic susceptibility \( \chi \) of the ferrofluid.

In this paper we have reported the results of experiments performed in a region of the parameter space where all occurring drop shapes are stationary. This made a detailed comparison between the experimental and theoretical results possible. To this end all parameters of our
Experimental set-up were determined in independent experiments so that there remain no fit parameters.

Whereas the experimentally determined transition lines for the shape bifurcations shown in the phase diagram (figure 2) are already well reproduced by a theoretical analysis based on a linear magnetization law for the ferrofluid the detailed reproduction of the experimentally observed eccentricities must take into account saturation effects in the magnetization behaviour, cf figure 3.

The rotational motion of the drop is most easily studied experimentally for the non-axisymmetric shape. However, since the spheroidal shape is ‘crowned’ by peaks due to the normal field instability for large field strength an experimental determination of the rotation frequency is also possible in this case. The theoretical analysis of the rotation of the drop building on Jeffrey’s classical solution for the viscous flow around a rigid ellipsoid shows only in this latter case quantitative agreement with the experiment.

The case of an elliptically polarized field interpolates between a rotating and an oscillating magnetic field. The theoretical analysis of this case reveals that already a small eccentricity of the rotating field, as it always occurs in experiment, may substantially slow down the rotation of the drop. This effect may contribute to the difference in the experimental and theoretical results for the rotation frequency of the elongated drop.

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References

[1] Chandrasekhar S 1987 Ellipsoidal Figures of Equilibrium (New York: Dover)
[2] Bohr N and Wheeler J A 1939 Phys. Rev. 56 426
[3] Huang X-P, Bollinger J J, Mitchell T B and Itano W M 1998 Phys. Rev. Lett. 80 73
[4] Seifert U 1997 Adv. Phys. 46 13
[5] Keller S R and Skalak R 1982 J. Fluid Mech. 120 27
[6] Ohsaka K and Trinh E H 2000 Phys. Rev. Lett. 84 1700
[7] Wang T G, Trinh E H, Croonquist A P and Elleman D D 1986 Phys. Rev. Lett. 56 452
[8] Bacri J-C, Cebers A and Perzynski R 1994 Phys. Rev. Lett. 72 2705
[9] Bratukhin Yu K, Lebedev A V and Pshenichnikov A F 2000 Fluid Dyn. 35 17
[10] Morozov K I, Engel A and Lebedev A V 2002 Europhys. Lett. 58 229
[11] Rosensweig R E 1985 Ferrohydrodynamics (Cambridge: Cambridge University Press)
[12] Bacri J-C and Salin D 1982 J. Physique 43 L649
[13] Lebedev A V and Morozov K I 1997 JETP Lett. 65 160
[14] Morozov K I 1997 JETP 85 728
[15] Sandre O, Browaeys J, Perzynski R, Bacri J-C, Cabuil V and Rosensweig R E 1999 Phys. Rev. E 59 1736
[16] Morozov K I and Lebedev A V 2000 JETP 91 1029
[17] Cowley M D and Rosensweig R E 1967 J. Fluid Mech. 30 671
[18] Cebers A and Lacis S 1995 Brazil. J. Phys. 25 101
[19] Shliomis M I 1974 Sov. Phys.–Usp. 17 153
[20] Landau L D and Lifshitz E M 1984 Electrodynamics of Continuous Media 2nd edn (New York: Pergamon)
[21] Abramowitz M and Stegun I (ed) 1964 Handbook of Mathematical Functions (Washington DC: National Bureau of Standards)
[22] Jeffrey G B 1922 Proc. R. Soc. A 102 161
[23] Engel A, Morozov K I and Lebedev A V 2003 Rotating ferrofluid drops Phys. Fluids, submitted
[24] Martsenyuk M A, Raikher Y L and Shliomis M I 1974 Sov. Phys.–JETP 38 413
[25] Raikher Y L and Shliomis M I 1994 Adv. Chem. Phys. 87 595
[26] Müller H W and Liu M 2001 Phys. Rev. E 64 061405
[27] Engel A, Müller H W, Reimann P and Jung A 2003 Ferrofluids as thermal ratchets Phys. Rev. Lett. , submitted