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Interactions of magnetic holes in ferrofluid layers

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Abstract Nonmagnetic microspheres in a ferrofluid layer interact as dipoles in an external magnetic field. With a constant field normal to the layer and an oscillating in-plane field, the spheres stabilize at fixed separations, and can thus be trapped at tunable finite distances. We show analytically how the susceptibility contrast at the system boundary is responsible for this secondary minimum of a pair interaction potential, obtain the effective interaction potential and equilibrium separation as a function of the applied field, and experimentally validate this theory.

Keywords Ferrofluids · Magnetic holes · Rotating magnetic fields · Effective interactions · Confined system

Introduction

Magnetic holes in a carrier ferrofluid are micrometric spherical nonmagnetic particles, whose size is orders of magnitudes above that of magnetites (0.01 μm), so the ferrofluids appears homogeneous on their scale. In the presence of an external magnetic field, they generate dipolar magnetic perturbations, whose moment is the opposite of that of the ferrofluid displaced [1, 2]. This induces dipolar interactions between them, which can be tuned through the type of imposed external field. This system, first invented by Skjeltorp [1], is confined between two nonmagnetic plates, spaced by a fixed distance ranging from one to ten particle diameters. The ability to design and modify the effective interactions in this system enables us to grow crystals of such holes and induce order/disorder transitions in them [1], to study aggregation dynamics of these particles [3], or to study the nonlinear dynamics of such magnetic holes in low-frequency rotating magnetic fields [4]. Understanding these systems is important for ferrofluid industrial applications [5], or their possible use in biomedicine [6]. Nonetheless, there was no satisfactory theoretical description of the effective interactions between holes in magnetic fields composed of a high-frequency rotating in-plane component and a constant normal one, and the existence of stable configurations of particles with a finite distance between them [2] remained unexplained.

Focusing on the boundary conditions of the magnetic fields along the confining plates, we will derive analytically the pair interaction potential in such oscillating fields, and demonstrate for a wide range of them the existence of a secondary minimum whose position depends continuously on the ratio between out-of-plane and in-plane field magnitudes. We will then compare this theory with experiments where the motion of a particle pair is followed.

System presentation and derivation of an effective pair interaction potential

In the presence of a far-range field $\vec{H}$ in a ferrofluid of susceptibility χ, each hole generates a dipolar perturbation of the dipolar moment equal and opposite to that of the displaced ferrofluid,$\vec{\sigma} = -V\chi_e \vec{H}$, where V is the volume of the particle, and $\chi_e = 3\chi/(3+2\chi)$ is the effective susceptibility including a demagnetization factor rendering for the boundary conditions of the magnetic field along the spherical particle boundary [1, 7]. The susceptibility contrast between the ferrofluid and the two planar nonmagnetic confining plates leads to a different dipolar field perturbation from that in the infinite medium expression. According to the image method [8], the boundary conditions for the magnetic field along the plates are fulfilled if in addition to the infinite space expression, the dipolar field emitted in an unbounded medium by an infinite series of dipole images is taken into account. This series consists of mirror images in the plane boundaries of the initial dipoles or of some previous image, by multiplying the magnitude of the dipole at each mirror symmetry operation by an attenuation factor $\kappa = \chi/(\chi+2)$ – see Fig. 1.

The instantaneous interaction potential between a pair of confined particles can then be expressed as [7]

$$U = \frac{\mu}{8\pi}\sum_{i\neq j}\left[\frac{\vec{\sigma}_i\cdot\vec{\sigma}_j}{r_{ij}^3} - 3\frac{\left(\vec{\sigma}_i\cdot\vec{r}_{ij}\right)\left(\vec{\sigma}_j\cdot\vec{r}_{ij}\right)}{r_{ij}^5}\right] \qquad (1)$$

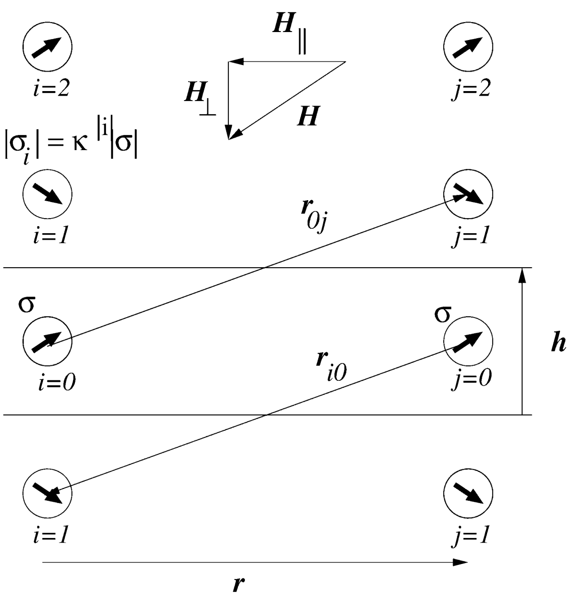

Fig. 1 Pair of nonmagnetic particles in a ferrofluid layer, viewed along the confinement plates, and a series of dipole images accounting for the boundary conditions of the magnetic field along the plates

where μ is the ferrofluid's permeability, the i-index runs over both the source and image dipoles, and the j-index runs only over the two source particles. A detailed analysis shows that the dominant effect for the components of the forces normal to the plates is the interaction between a particle and its own mirror images, which stabilizes the particles midway between the plates. This constraint is therefore adopted throughout this paper and in Fig. 1. Buoyancy forces have also been checked to be negligible for the ferrofluid and particles used in this work.

Decomposing the instantaneous far-range field into its in-plane and normal components $\vec{H}_\perp$ and $\vec{H}_\parallel$, we define the ratio of their magnitudes as $\beta = H_\perp/H_\parallel$, the angle between in-plane component and separation vector as φ, the particle diameter and interplate separation, respectively, as a and h, and the scaled separation as $x = r/h$.

A straightforward analysis of Eq. (1) in the configuration of Fig. 1 leads up to an additive constant to

$$\frac{144h^3U}{\mu\pi a^6\chi_e^2H_\parallel^2} = \sum_{l=-\infty}^{+\infty}\kappa^{|l|}\left[\frac{1+(-1)^{|l|}\beta^2}{\left(x^2+l^2\right)^{3/2}} - 3\frac{(y\cos\varphi + l\beta)\left(y\cos\varphi + (-1)^{|l|}l\beta\right)}{\left(x^2+l^2\right)^{5/2}}\right]. \qquad (2)$$

The term $l = 0$ corresponds to the source–source interaction term, already used in previous studies [1, 2], the other terms correspond to interactions between a particle and the images of the other one. For all existing ferrofluids, κ is sufficiently smaller than unity, so the prefactor ensures that the three first images are enough to get a relative precision better than 1% for the potential and its derivatives.

The imposed fields considered consist of a constant component $\vec{H}_\perp$, while $\vec{H}_\parallel$ rotates uniformly at sufficiently high frequency (10–100 Hz in this work). For the micrometric particles considered here (a = 50 μm), inertial terms can be neglected, and a characteristic viscous relaxation time can be obtained as the time to separate two particles initially in contact by their size, balancing the Stokes drag with the magnetic interaction forces derived from the potential. Retaining the main term $l = 0$ in Eq. (1) leads to an estimate of $T_c = 144\eta/(\mu\chi_e^2\mathrm{H}^2) \approx 5\mathrm{s}$, for the ferrofluid [$\eta = 9\times10^{-3}$ Pa s, $\mu = \mu_0(1+\chi)$, $\chi = 1.9$, $\mu_0 = 4\pi\times10^{-7}$ H m^{-1}] and the typical field $H = 10$ Oe considered here. At field rotation frequencies significantly exceeding the inverse relaxation time, the particle's motion can be neglected during a field oscillation, and an effective interaction potential averaged over time can be obtained by replacing simply the φ-dependent terms in Eq. (2) by their average over an oscillation, while the slowly varying separation vector is maintained constant, which leads to the dimensionless effective interaction potential:

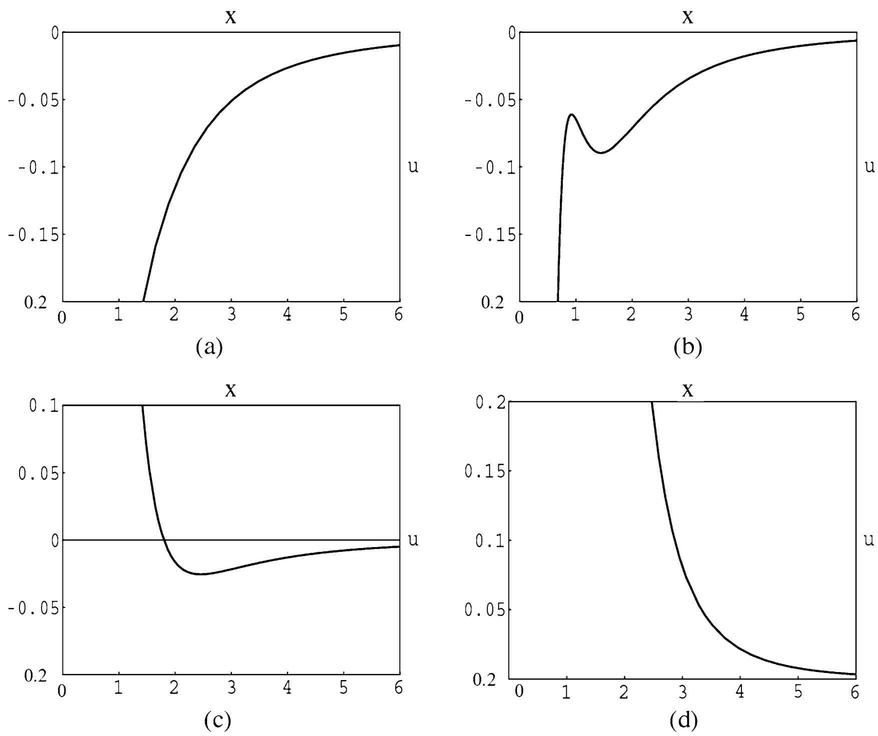

Fig. 2 Four possible types of interaction potentials: **a** $\beta < \beta_m$ purely attractive interactions, **b** $\beta_m < \beta < \beta_c$ interactions with a secondary minimum, **c** $\beta_c < \beta < \beta_u$ interactions with a single equilibrium position at a finite distance, **d** $\beta_u < \beta$ purely repulsive interactions

$$u(x) \equiv \frac{144h^3\overline{U}}{\mu\pi a^6\chi_e^2H_\parallel^2} = \sum_{l=-\infty}^{+\infty}\kappa^{|l|}\left[\frac{1+(-1)^{|l|}\beta^2}{(x^2+l^2)^{3/2}} - 3\frac{(-1)^{|l|}l^2\beta^2+y^2/2}{(x^2+l^2)^{5/2}}\right]. \quad (3)$$

For a given ferrofluid and field, this central potential can be of four possible types as illustrated in Fig. 2.

At low normal field $\beta < \beta_m$, the interactions are purely attractive up to contact; at higher fields $\beta_m < \beta < \beta_c$, a secondary minimum at finite distance appears, later on in the regime $\beta_c < \beta < \beta_u$ this minimum becomes the only one, and ultimately interactions are purely repulsive for $\beta_u < \beta$. For the ferrofluid used, the equilibrium separation for these effective interactions as a function of β are displayed as a continuous line in Fig. 3.

From Eq. (3), the separating values can be shown to be $\beta_C = 1/\sqrt{2}$, $\beta_u = \beta_c(1+\kappa)/(1-\kappa)$, which is a growing function of the susceptibility diverging to infinity when the susceptibility does, and β_m a function of the susceptibility decreasing regularly from β_c at zero susceptibility to 0 at infinite susceptibility – for the ferrofluid used here, $\beta_m \approx 0.55$ and $\beta_u \approx 2.05$. Neglecting the susceptibility contrast along the plates – terms $l \neq 0$ in Eq. (3) – would correspond to the limiting case $\kappa = 0$, where these three separating values merge, and the interactions are either purely attractive or repulsive. The presence of this susceptibility contrast is thus essential to trap the particles at a given equilibrium distance in this type of field.

In order to confront this theory with experiments, the motion of particle pairs initially in contact in a given field was recorded through time. The dynamical equation ruling the particles in this overdamped regime is obtained by balancing the Stokes drag from the embedding fluid with the magnetic interactions, which leads to $\dot{x} = -u'(x)$, where the dot refers to derivation with respect to $t' = t/T$, the dimensionless time, with $T = 3T_c h^5/a^5$, the unit time. The function $t'(x)$ was then

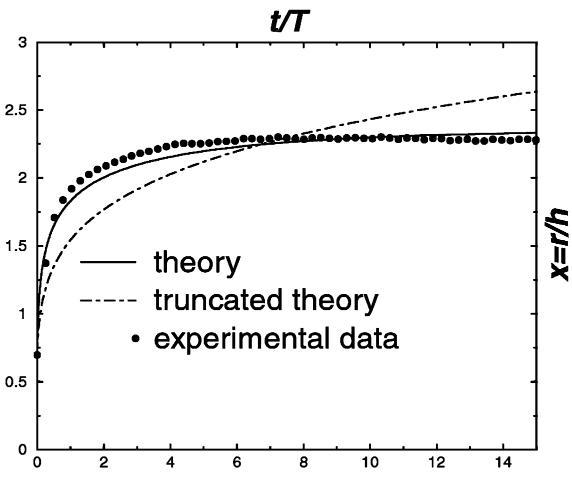

Fig. 3 Scaled separation between a pair of particles as a function of the scaled time, comparison between theory and experiment obtained with an oscillating in-plane field of magnitude $H_\parallel = 14.2$ Oe and a constant normal field $H_\perp = 0.8H_\parallel$

numerically evaluated as the integral of $-1/u'$ from the initial a/h to the actual x value of the scaled separation, to obtain the dashed lines of Fig. 3 and the full line of Fig. 4.

Experiments and results

The experiments were carried using $a = 50$-μm diameter neutral nonmagnetic polystyrene particles designed by Ugelstad's technique [9] and produced under the trademark Dynospheres by Dyno Particles, Lillestrøm, Norway, in a kerozene-based ferrofluid (type EMG905, Ferrofluidics, Nashua, NH, USA). The confining cell consisted of two glass plates 70 μm from each other, and was obtained by slightly pressing the cells together with a few $h = 70$-μm-diameter spacers in-between (of the same composition as the holes). The cell was placed in three pairs of Helmholtz coils, and the particle motion was recorded and digitized from optical microscopy data. The particles were initially brought into contact by applying a fast oscillating purely in-plane field, after which the constant normal field component was added at time zero. The particle pair observed was separated from any other particle or spacer by more than 20 diameters, so as to avoid perturbations.

A typical record of the scaled distance as a function of the scaled time, obtained for a field $H_{||} = 14.2$ Oe, at $\beta = 0.8$, is shown in Fig. 3.

In this case, the unit time is $T = 32$ s, and particles come to the predicted equilibrium distance $r = 2.35h$ after a few minutes. Comparing this data with the present theory (full line) and with the preexisting expression [1, 2] ignoring the effect of the boundary conditions along the nonmagnetic plates (dashed line) we conclusively show the primary importance of this susceptibility contrast in explaining the existence of this secondary minimum.

A range of geometric β parameters of the applied field was explored in a series of experiments with the same setup, and the resulting scaled separation as a function of β, for various characteristic scaled times, is displayed in Fig. 4.

The error bars correspond to an experimental error in β reflecting a possible angle up to 2.5° of the constant field over the normal direction, which is the maximum variation of the field orientation over the cell, calculated from its precise geometry with respect to the external coils. This error effect was calculated to be the major one owing to the slight inhomogeneity of the field along the experimental cell. Once again, in the range $\beta > \beta_m \approx 0.55$ experiments and theory agree fairly well.

The main discrepancy lies in the small but nonzero separation ($x > 50/70$) for $0.3 < \beta < \beta_m \approx 0.55$. This is believed to be due to the effect of the non-point-like character of the magnetic holes for the magnetic field, which should generate higher-order terms in a multipolar expansion at moderate separations r/a – qualitatively, the dipolar perturbation field of a hole does not fulfill proper boundary conditions along the surface of another hole that is close enough, and a repulsive term corresponding qualitatively to taking into account images of one sphere in the other one, similar to the repulsive effect of images in the plane boundaries on its source particle, becomes sensitive at these short distances.

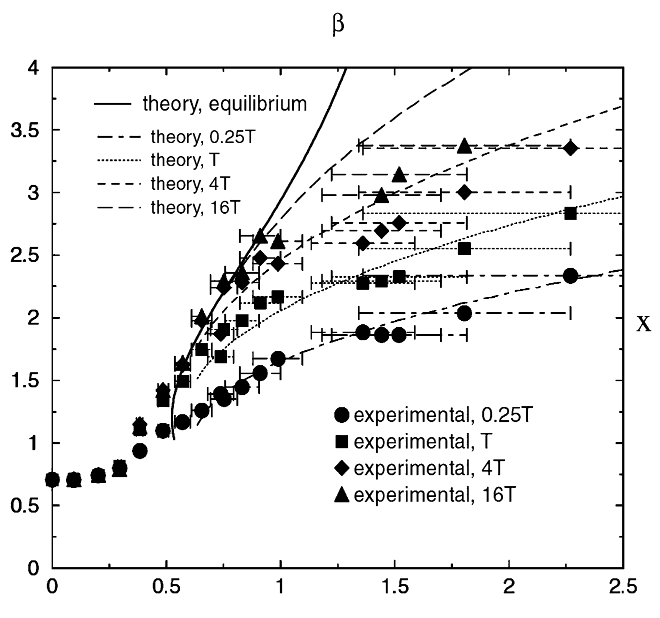

Fig. 4 Scaled distance as a function of the field geometric parameter β, at various values of the scaled time

Conclusions

We have established the effective pair interactions of nonmagnetic particles embedded in a ferrofluid layer confined between two nonmagnetic plates, submitted to magnetic fields including constant normal components and high-frequency oscillating in-plane components. Owing to the susceptibility contrast along the glass plates, a family of potentials displaying a secondary minimum at a finite separation distance has been proven to exist, which should allow us to trap nonmagnetic bodies at tunable distances via the external field.

A system with interactions such as those described here should be relevant for any colloidal suspension of electrically or magnetically polarizable particles constrained in layers. The realization of the detailed effective interaction potentials of this system also makes it a good candidate as an analog model system to study phase transitions [1], aggregation phenomena in complex fluids [4], or fracture phenomena in coupled granular/fluid systems.

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