Pattern Formation at Magnetophoretical Motion in the Self-Magnetic Field of Magnetic Colloid

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Abstract

The model with the magnetophoretical motion in the self-magnetic field of magnetic colloid is formulated. It is shown that growing microdrops due to the field induced demixing undergo the magnetostatic instabilities analogous to the macroscopic drops of the magnetic fluid.

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1. Introduction

There is a wide variety of systems which form modulated phases [1]. The generic models of these pattern formation processes in dependence on the physical nature of the system are formulated on the basis of the Ginzburg-Landau equations for conserved or non conserved order parameter. Here we illustrate that the model of the magnetophoresis in the self-magnetic field of magnetic colloid arising at field induced demixing in thin cells possess the features characteristic to the systems which form modulated phases.

2. Model

The mean field model of the magnetic colloid shows that for given concentration and temperature range exists the critical magnetic field strength exceeding which the demixing of colloid occurs [2, 3]. Expanding the thermodynamic potential near the critical point of demixing (n_c, H_c), taking into account the magnetic field perturbation $\delta \boldsymbol{H}$ arising at the modulation of the concentration, neglecting the concentration variation across the cell with the thickness h and the influence of the field perturbation on the magnetization the thermodynamic potential of the magnetic colloid under the action of the field normal to the cell reads

$$F = h\int\Big(-\frac{1}{2}\alpha(\delta n)^2 + \frac{1}{4}(\delta n)^4 + \frac{1}{2}\beta(\nabla_2\delta n)^2\Big)\,dS + \frac{1}{2}\int G(\rho-\rho')\delta n(\rho)\delta n(\rho')\,dS\,dS', \quad (1)$$

where $\alpha = \partial^2 M/\partial n^2(n_c, H_c)(H - H_c)$; $\gamma = \partial^3\phi/\partial n^3(n_c, H_c)$ (M is the magnetization of colloid, ϕ is the chemical potential of the particles). In the long-range dipolar interaction term

$$G(\rho-\rho') = = 2\Big(\frac{\partial M}{\partial n}(n_c, H_c)\Big)^2\int\Big(\frac{1}{|\rho-\rho|'} - \frac{1}{\sqrt{(\rho-\rho')^2+h^2}}\Big)\,dS'.$$

Here ρ is the radius vector parallel to the boundary of the cell. The term with the coefficient β takes into account the surface tension between the phases arising at demixing.

Scaling the concentration by $\sqrt{\alpha/\gamma}$, length by $l = \sqrt{\beta/\alpha}$ and time by $\tau = l^2/\zeta\alpha$, where ζ is the mobility of the particles the equation for the particle concentration

$$\frac{\partial\delta n}{\partial t} = \mathrm{div}\Big(\zeta\nabla\frac{\delta F}{\delta n}\Big)$$

put in dimensionless form reads

$$\frac{\partial\varphi}{\partial t} = \Delta\Big(-\varphi + \varphi^3 - \Delta\varphi + \frac{2Bm}{(h/l)^2}\int G(\rho-\rho')\varphi(\rho')\,dS'\Big). \quad (2)$$

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The magnetic Bond number $Bm = (\partial M/\partial n)^2 h/\alpha l$ characterizes the ratio of magnetic and capillary forces.

The thermodynamic potential (1) possess the generic property of the systems forming the modulated phases. Its minimal value is achieved for the concentration modulations with the finite wave number q_m determined by the minimum of $q^2 + 2BmJ(q)/(h/l)^2$ ($J(q) = 2\pi(1 - \exp(-qh/l))/q$). If the total increase of the surface and magnetodipolar energies is less than the condensation energy determined by the first term in (1) demixing of the colloid occurs. The critical magnetic Bond number Bm_c at $h/l >> 1$ is $Bm_c = (h/l)^2/6\pi\sqrt{3}$.

The phase diagram of the magnetic colloid in dependence on $Bm, h/l$ and φ_0 - the deviation of the mean dimensionless concentration from the critical is calculated in [4]. The phase diagram shows that at $Bm/Bm_c < 1$ there are stripe, hexagonal and homogeneous phases with the corresponding coexistence regions.

3. Numerical method

For the numerical simulation of the concentration dynamics described by the equation (2) large box with size $L_x \times L_y$ is taken and the periodicity $\varphi(x+nL_x, y+mL_y) = \varphi(x, y)$ assumed. Representing the concentration field by Fourier series

$$\varphi(\boldsymbol{r}) = \sum_k \varphi_k e^{i\boldsymbol{k}\cdot\boldsymbol{r}}$$

the equations for the Fourier amplitudes φ_k reads

$$\frac{\partial\varphi_k}{\partial t} = -k^4\varphi_k + N_k(\varphi_k)$$

$$N_k(\varphi_k) = k^2\Big[\Big(1 - \frac{Bm}{(h/l)^2}J(k)\Big)\varphi_k - \varphi_k^3\Big],$$

which are solved calculating φ_k^3 according to the pseudospectral rule. Time evolution with time step Δt is found applying the fourth-order Adams-Bashforth scheme:

$$\varphi_k^{n+1} = e^{-\Delta t k^4}\varphi_k^n + \Delta t\sum_{j=0}^{3} a_j e^{-(j+1)\Delta t k^4} N_k^{n-j},$$

where $\boldsymbol{a} = (55, -59, 37, -9)/24$.

4. Numerical simulation results

Numerical simulations where performed on 256×256 lattice, taking $L_x = L_y = 256l$. For all calculations $h/l = 10$. The parameters Bm/Bm_c and φ_0 are choosen such

Figure 1: Equilibrium microdroplets, obtained for parameters $h/L = 10$, $\varphi_0 = -0.5$. The values of Bm/Bm_c corresponds to 0.490, 0.473, 0.457, 0.441, 0.424, 0.408, 0.392 starting from left.

that they correspond to the unmodulate and hexagonal phase coexistence region near to the curve that separates this region from unmodulated phase region [4].

The calculations are started at $Bm/Bm_c = 0.490$ from random initial data and single microdroplets of concentrated phase are obtained, left in Fig. 1. It is important to note, that for a most of runs microdroplets are not observed and unmodulated phase is obtained. This illustrates the highly nonlinear nature of this phenomenon.

The second left picture in Fig. 1 is obtained taking the first solution for decreased value of $Bm/Bm_c = 0.473$. The process of decreasing of Bm/Bm_c is continued until the value 0.392 is reached. Obtained results show, that the size of microdroplet increases due to the condensation of the particles, but it remains stable on this parameter range.

When next step of calculation at $Bm/Bm_c = 0.375$ is performed, microdroplet increases its volume and undergoes the elliptical instability when its volume is enough large with the subsequent bending instability and the formation of the highly convoluted shape, as is shown in Fig. 2. Contrary to the elliptic instability of the macroscopic drops of magnetic fluid [5] the volume of the microdroplet is not conserved and the instability is due to the magnetophoresis in the self-magnetic field of the nonhomogeneous colloid. White region around the droplet show depletion of the particles due to their condensation on the growing region of the condensed phase. This phenomenon in some sense is similar to the development of the elliptic instability at the nucleation of the superconducting phase in the superconductors of type I at the decrease of the magnetic field [6].

It is interesting that if the size of the growing microdroplet is enough large the higher perturbation modes develop on the boundary of the microdroplet as is shown on last row of Fig. 2. This is done by taking the configuration of stable microdroplet at $Bm/Bm_c = 0.392$ as initial configuration. Sequence of the formation of branched structure with threefold vertexes is in correspondence to the behavior of macroscopic drops of magnetic fluid [5, 7].

Estimate of the equilibrium radius of the microdroplets similarly to the analysis of the nucleation of

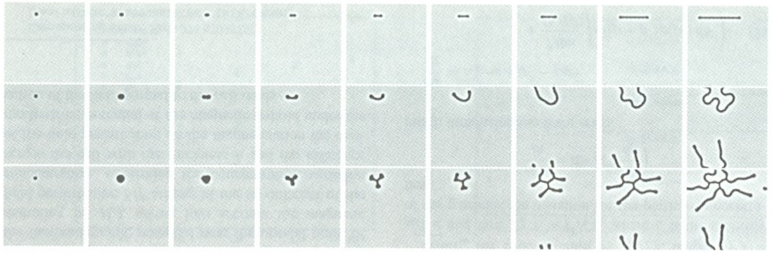

Figure 2: Sequence of snapshots obtained for parameters $h/L = 10$, $\varphi_0 = -0.5$. First row $Bm/Bm_c = 0.375$, for second $Bm/Bm_c = 0.310$ and for last $Bm/Bm_c = 0.278$. Time increases from left to right.

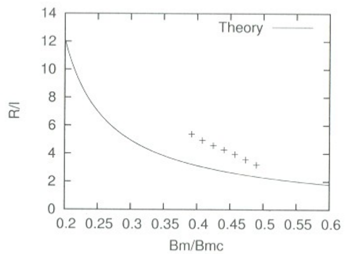

Figure 3: The equilibrium radius radius of microdroplet. Surface tension multiplier $\sigma_r = 0.1$ for theoretical curve.

the superconducting phase in [6] may be found by the calculation of the free energy of the microdroplet. Taking into account the variation of the free energy of the homogeneous phase due to the condensation of the particles, self-magnetic field energy for homogeneous distribution of the particles in the nucleus and adding the surface energy term for the thermodynamic potential in dependence on the radius of the microdroplet R we have

$$\Delta F = \pi R^2\varphi_0(1-\varphi_0)^2(1+\varphi_0)+\frac{8\pi R\sigma_r}{3\sqrt{2}}+\frac{(1-\varphi_0)^2 Bm}{3\sqrt{3}Bm_c}\times$$
$$\times\left(\pi h R^2+\frac{8R^3}{3}\left(1-\frac{(2k^2-1)E}{k^3}-\frac{(1-k^2)K}{k^3}\right)\right), \quad (4)$$

here K, E are elliptic integrals of type I and II respectively, $k^2 = (2R/h)^2/(1+(2R/h)^2)$. By multiplier σ_r we take into account that the surface tension of the nucleus is not equal to $4/3\sqrt{2}$ (in our dimensionless units) - surface tension between two homogeneous phases. Interest ing feature of the energy (4) is that for Bm/Bm_c less than the critical value there is energy minimum for the finite radius of the microdroplet which diminishes with the increase of Bm/Bm_c. At the critical Bm/Bm_c value the microdroplet collapses. Before the collapse due to the instabilities shown in Fig. 2 it tranforms to the branched configurations. Theoretical dependence of the radius of the microdroplet on Bm/Bm_c shown in Fig. 3 qualitatively corresponds to the results of the numerical calculation. Some quantitative difference should be caused by approximate estimate of the surface tension term.

5. Conclusions

It is found that the microdroplets of the magnetic colloids in thin cells arising at the magnetic field induced demixing undergo the magnetostatic instabilities characteristic to the macroscopic drops of the magnetic fluid. Contrary to the case of the macroscopic drops this instability is caused by the magnetophoresis of the magnetic particles in the self-magnetic field of the colloid.

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