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Topological instability of magnetic fluids

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Résumé. — On a étudié le comportement d'un volume limité de fluide magnétique reposant sur une base solide et placé dans un champ magnétique extérieur homogène. On a obtenu un critère d'instabilité topologique. L'existence de l'hystérésis des formes (transition de phase du 1^{er} ordre) a été prédite.

Abstract. — A finite volume of a magnetic fluid resting on a solid base in an external homogeneous magnetic field is considered. An approach to the theory of topological instability is proposed and the instability criterion is derived. The shape hysteresis (first order phase transition) for fluids with high magnetic susceptibility is predicted.

1. Introduction.

The breakup of the equilibrium shapes of isolated volumes resting on a solid base is observed in magnetic fluids (MF) when certain critical intensity H_0^c of the external homogeneous magnetic field is achieved. This phenomenon, due to topological changes of the free surface, is called topological instability [1]. It has been experimentally observed [1] that such instability induces a breakup of the MF drop into two approximately equal isolated volumes located at some finite distance from each other.

The physical reason for this phenomenon is that at external magnetic field intensity H_0^c the formation of two smaller isolated volumes V_i instead of a single larger one — V ($\sum_i V_i = V$) becomes energetically favourable. The competition among magnetic, gravitational and capillary forces results in the instability of the system relative to small perturbations, whose development leads to the breakup of volume V . The formulation of the mathematical model of the topological instability phenomenon is based on the principle of the total energy minimum. The model involves the Maxwell equations, the Navier-Stokes equations, boundary conditions on a free surface and on a solid base, the Dupré-Young condition for the three phase interface [2]. The stability analysis of a finite volume of MF is based on consideration of the second variation $\delta^2 F$ of the potential energy. If $\delta^2 F$ is positive for all perturbations N of the free surface which preserve the volume, then the equilibrium state is stable. Otherwise, instability will be observed. The characteristic feature of the stability analysis of MF is that a perturbation of the free surface implies a change in the magnetic field intensity $\delta \mathbf{H}$, and one cannot derive an explicit analytical relation between $\delta \mathbf{H}$ and arbitrary N . The full-scale theoretical study of the topological instability would entail great

computational difficulties. Instead of studying $\delta^2 F$, the approach based on comparison of the energy of the system before and after the breakup is suggested in the present paper.

This paper gives a theoretical description of the effect of topological instability in approximation (Sect. 2). The suggested approach enables us to obtain the relationship between H_0^c and the volume V of MF (Sect. 3), to predict the hysteresis effects (Sect. 4) and to investigate the interaction between the isolated volumes of MF (Sect. 5).

2. Model.

Let us consider an isolated volume of MF, resting on a solid base, made of ferromagnetic material. The system is placed in a homogeneous magnetic field of intensity $\mathbf{H}_0$ (see Fig. 1). The axis of symmetry of the drop, Z , is directed parallel to $\mathbf{H}_0$ and antiparallel to the gravity acceleration $\mathbf{g}$, and axes x and y of Cartesian coordinates are located in the plane of the solid base. We shall find the thermodynamic potential having a minimum in the equilibrium state, and investigate the conditions when the global minimum is achieved.

The following assumptions have been made : (i) variations of the intensity of the external homogeneous magnetic field H_0 are quasi-static ; (ii) internal flows are neglected ; (iii) magnetization $\mathbf{M}$ of the fluid is a linear function of the magnetic field intensity $\mathbf{H}$: $\mathbf{M} = \chi \mathbf{H}$; (iv) the solid base is made of ferromagnetic material of high magnetic permeability μ_s : $\mu_s \gg 1$; (v) the liquid-solid contact angle α_0 is in the vicinity of 90° ; (vi) the shape of the free surface is close to that of an ellipsoid ; (vii) temperature is uniform and constant (isothermality) ; (viii) evaporation is neglected.

By force of assumptions (vii) and (viii) the equilibrium state is characterized by the minimum of the total Helmholtz free energy F of the system. Let us write out the functional for F excluding the energy of the applied external magnetic field in the total space in the absence of MF, as well as the energy of the surface tension (vapour-base) in the absence of MF (such definition of F excludes from consideration the quantities which are independent of the thermodynamic state and MF properties and therefore produce no effect on the equilibrium and the stability of the drop) :

$$F = \sigma S + (\sigma_{13} - \sigma_{23}) S_0 + \rho g \int_{\Omega} z \, dV + \int \left(F_m + \frac{1}{2} \mu_0 H_0^2 \right) dV \quad (1)$$

Here : σ , σ_{13} , and σ_{23} are coefficients of surface tension at the liquid-gas, liquid-solid and solid-gas interface, respectively ; S is the free surface area ; S_0 is the area of the base of the drop ; ρ is density of MF ; Ω is the region occupied by MF ; F_m is the « magnetic portion » of the free energy density ; μ_0 is the magnetic constant. Integration in the last term of the right-hand side of equation (1) is

Fig. 1. — Structure of the model.

carried out over the total space. We consider drops of macroscopic size ($L > 0.4$ cm), containing a great number ($\sim 10^{10}$) of ferromagnetic particles (diameter of the particle ~ 100 Å). Therefore, when calculating the magnetic energy we use a continuous media approximation, in which MF appears to be a superparamagnetic.

It follows from the Dupré-Young condition at the three phase interface $\sigma_{23} - \sigma_{13} = \sigma \cos \alpha_0$, and assumption (v) that $\sigma_{23} = \sigma_{13}$. By assuming the ellipsoid to be elongated along the Z axis (which corresponds to the experimental observations), we have :

$$S = \pi b^2 + \pi ab \arcsin e/e \quad (2)$$

where a and b are the larger and the smaller axes, respectively ; and $e = \left(1 - \frac{b^2}{a^2}\right)^{1/2}$ is the eccentricity of the ellipsoid. Let us consider the last term in the right-hand side of equation (1). By force of assumptions (iii) and (iv) one can obtain [3] :

$$\int \left(F_m + \frac{1}{2} \mu_0 H_0^2 \right) dV = - \frac{1}{2} \mu_0 \mathbf{H}_0 \int_{\Omega} \mathbf{M} dV.$$

Find the magnetization distribution inside Ω . The Maxwell equations $\text{rot } \mathbf{H} = 0$ and $\text{div}(\mathbf{H} + \mathbf{M}) = 0$ subject to (iii) lead to the Laplace equation for the magnetic field intensity potential φ ($\mathbf{H} = -\nabla \varphi$) : $\Delta \varphi = 0$ with ordinary conditions of continuity of $\mathbf{H}_t$ and B_n at the MF-gas interface and the condition $\varphi = 0$ on the solid base ($z = 0$), following from (iv). The solution of the boundary value problem for potential φ in the region Ω will be :

$$\varphi = - \frac{H_0}{1 + \chi n(e)} Z,$$

where $n(e)$ is the demagnetization factor [3],

$$n(e) = \frac{1 - e^2}{e^3} \left(\frac{1}{2} \ln \frac{1 + e}{1 - e} - e \right). \quad (3)$$

Thus the « magnetic portion » of F is given by :

$$\int \left(F_m + \frac{1}{2} \mu_0 H_0^2 \right) dV = - \frac{1}{2} \mu_0 \frac{\chi}{1 + n\chi} H_0^2 V \quad (4)$$

where $V = \frac{2}{3} \pi ab^2$ is the drop volume. We rewrite equation (1) by using equations (2) and (4) :

$$F = \pi \sigma (b^2 + ab \arcsin e/e) + \frac{1}{4} \pi \rho g a^2 b^2 - \frac{1}{3} \mu_0 \frac{\chi}{1 + n\chi} H_0^2 \pi ab^2. \quad (5)$$

Let us consider equation (5). For the similarity reasons it will be convenient to proceed to a dimensionless formulation of the problem. For this purpose we introduce dimensionless quantities $\bar{a}$, $\bar{b}$, $\bar{F}$:

$$\bar{a} = V^{-1/3} a, \quad \bar{b} = V^{-1/3} b, \quad \bar{F} = F/(\pi \sigma V^{2/3}).$$

The system behaviour will be determined by two dimensionless groups :

$$\begin{aligned} B_g &= \rho g V^{2/3} / \sigma && \text{— the gravitational Bond number and} \\ B_m &= (1/3) \mu_0 H_0^2 V^{1/3} / \sigma && \text{— the magnetic Bond number.} \end{aligned}$$

Free energy functional (5) will take the form (the bars above a , b and F are omitted) :

$$F = b^2 + ab \arcsin e/e + \frac{1}{4} B_g a^2 b^2 - B_m \frac{1}{\gamma + n} ab^2 \quad (6)$$

where $\gamma = 1/\chi$.

Since the drop volume is constant (viii), we have :

$$ab^2 = 3/(2\pi) = q. \quad (7)$$

The equilibrium at specified values of B_g and B_m is determined by $\frac{dF}{de} = 0$. From equation (6) we have :

$$\begin{aligned} -\frac{2}{3} q^{\frac{2}{3}} e d^{-\frac{2}{3}} + \frac{1}{3} q^{\frac{4}{3}} d^{-\frac{7}{6}} p + q^{\frac{2}{3}} d^{-\frac{1}{6}} \frac{d^{-\frac{1}{2}} e - p}{e^2} + \frac{1}{6} B_g q^{\frac{4}{3}} e d^{-\frac{4}{3}} - \\ - B_m q \frac{1}{(\gamma + n)^2} \left[\frac{3 - e^2}{e^4} \left(\frac{1}{2} \ln \frac{1 + e}{1 - e} - e \right) \right] = 0 \end{aligned} \quad (8)$$

where $p = \arcsin e$, $d = 1 - e^2$. A numerical study of equation (8) shows that at given B_g and B_m the unique solution $e = e(B_g, B_m)$ exists when the magnetic susceptibility of MF χ is not too high : $\chi < 20$ (Fig. 3a). At $\chi > 20$ equation (8) can have multiple roots (see Sect. 4). In what follows we consider the case $\chi < 20$. By force of a probabilistic character of fluctuations causing the drop breakup, the two pieces formed may, generally speaking, be of different volumes. But the most probable is the situation when these volumes are approximately equal : $V_1 \approx V_2 \approx \frac{V}{2}$. By using the principle of the total energy minimum the breakup condition can be written as :

$$F \geq 2 F' + F_{12} \quad (9)$$

where F' is the total energy of a solitary drop of volume V_1 ; F_{12} is the interaction energy of the two volumes of MF formed. The equality sign in equation (9) corresponds to the critical values of B_g and B_m , and determines the topological instability curve $f(B_g^c, B_m^c) = 0$. In order to determine the equilibrium value of F' one should solve equation (8) for the same values of B_g and B_m by substituting $q_1 = q/2$ ($\equiv 3/(4\pi)$) for q . Then the obtained value of eccentricity must be used to calculate F' according to equation (6) where q_1 must be substituted for q .

The value of the interaction energy F_{12} is small as compared to the magnetic self-energy F' and can be neglected, the estimates and behaviour of F_{12} being considered in section 5.

Thus, the topological instability criterion can be written as :

$$\begin{aligned} q^{\frac{2}{3}} d^{\frac{1}{3}} + q^{\frac{2}{3}} d^{-\frac{1}{6}} p/e + \frac{1}{4} B_g^c q^{\frac{4}{3}} d^{-\frac{1}{3}} - B_m^c q/(\gamma + n(e)) - \\ - 2 \left[q_1^{\frac{2}{3}} d_1^{\frac{1}{3}} + q_1^{\frac{2}{3}} d_1^{-\frac{1}{6}} p_1/e_1 + \frac{1}{4} B_g^c q_1^{\frac{4}{3}} d_1^{-\frac{1}{3}} - B_m^c q_1/(\gamma + n(e_1)) \right] = 0 \end{aligned} \quad (10)$$

where $q_1 = q/2$, $d_1 = 1 - e_1^2$, $p_1 = \arcsin e_1$.

Equations (8) and (10) present the dimensionless formulation of the problem.

3. Results.

Solutions to the set of equations (8)-(10) are given in figure 2, the values of B_g^c being plotted as abscissa and the logarithmical values of B_m^c as ordinate. The elongations $k = a/b$ of the critical

equilibrium shapes are given in table I. From the results obtained it follows that the small values of B_g^c ($B_g^c \lesssim 8$) correspond to the high values of B_m^c ($B_m^c \gg B_g^c$). In other words, the breakup of small volume drops involves high magnetic fields H_0^c . The value of H_0^c decreases rapidly as the drop volume increases. Also, the computation results suggest that there exists a limiting value of the gravitational Bond number $\tilde{B}_g \cong 5$, so that for $B_g < \tilde{B}_g$ no breakup occurs. The experimental results presented in reference [1] tentatively suggest the existence of $\tilde{B}_g$. Figure 2 shows the topological instability curves for some typical values of χ .

Let us consider an example of estimating H_0^c , when the drop of the MV-27 fluid has $V = 0.35 \text{ cm}^3$, the characteristics of the MV-27 fluid being $\rho = 1.37 \times 10^3 \text{ kg/m}^3$, $\sigma = 0.05 \text{ N/m}$, $\chi \cong 0.51$. Hence $B_g = 13$ [1]. The results of computations show that $B_m^c = 53.4$, which corresponds to the intensity of external magnetic field

$$H_0^c = (3 \sigma B_m^c / (\mu_0 V^{2/3}))^{1/2} = 31 \times 10^3 \text{ A/m}.$$

Fig. 2. — Topological instability curves.

Table I. — Critical elongations for topological instability effect. k_1 is the elongation of the drops formed after the breakup.

B_g	6	7	8	9	10	11	12	13	14
B_m	730.6	327.2	189.1	137.2	89.9	71.4	60.4	53.4	47.7
k	10.5	6.5	4.5	3.5	2.5	2.0	1.7	1.4	1.2
k_1	10.9	6.8	4.8	3.7	2.7	2.2	1.9	1.6	1.4

It's interesting to note that the drops formed after the breakup are more elongated than the original one.

4. Phase transition.

At $\chi > 20$ (Fig. 3b) the equilibrium curves have regions which correspond to unstable and metastable states. In figure 3, the values of the magnetic Bond number B_m are plotted as abscissa and the drop elongations $k = a/b$ as ordinate. Let us consider one of the equilibrium curves, for example, the one corresponding to $B_g = 13$. CE is stable, BD unstable, AB and CD are metastable portions of the curves. The elongation process displays hysteresis properties : when B_m is increased, k increases as well. Initially k increases continuously but at $B_m = B_{m_1}$ (point B of the curve of Fig. 3b) k elongates abruptly up to point C, after which the elongation again becomes continuous. When B_m decreases, k undergoes an abrupt change in point D where $B_m = B_{m_2} < B_{m_1}$, the elongated ellipsoidal shape of the drop ($k > 1$) becoming flattened ellipsoidal one ($k < 1$). In the limit of $B_g \rightarrow 0$ abrupt changes keep the drop in the class of elongated ellipsoids. The curve, corresponding to the case of $B_g = 0$, coincides with the equilibrium curve for the free drop obtained in reference [4], where such abrupt change of the shape is called a first order phase transition. It seems this definition may be used for a more general case $B_g \neq 0$, described in the present paper.

Fig. 3. — Equilibrium curves : (a) low magnetic susceptibility : $\chi = 10$; (b) high magnetic susceptibility : $\chi = 30$.

Besides, we must note that with B_g increasing hysteresis becomes more distinct, i.e. the difference $(B_{m_1} - B_{m_2})$ increases.

Thus, a finite volume of MF of high magnetic properties ($\chi > 20$), resting on a solid base, can undergo a first order phase transition. This transition occurs in low magnetic fields (as compared to H_0). Thus, for the above case $V=0.35 \text{ cm}^3$ at $\chi=30$ the phase transition occurs at $H_0=10^3 \text{ A/m}$.

5. Interaction energy.

In section 1, when the conditions of topological instability were considered, we neglected the interaction energy F_{12} of the two isolated volumes Ω_1 and Ω_2 produced by the breakup. In order that this approach be correct one has to estimate the value of F_{12} .

Let there be two point dipoles $\mathbf{m}_1$ and $\mathbf{m}_2$ located at a distance R from each other, R being much greater than the size of the dipoles, and let $\mathbf{m}_1 \uparrow \uparrow \mathbf{m}_2$. Then their interaction energy is :

$$f_{12} = -\mu_0 \frac{m_1 m_2 (3 \cos^2 \theta - 1)}{R^3}, \quad (11)$$

where θ is the angle between the direction of the dipoles and the radius vector $\mathbf{R}$. We assume that volumes Ω_1 and Ω_2 consist of point dipoles directed along the z -axis and uniformly distributed over the volume.

Proceeding to the continuous media approximation and taking into account the contribution made by each dipole couple only once, we get the total interaction energy from equation (11) :

$$F_{12} = -\frac{1}{2} \mu_0 M^2 \int_{\Omega_1} \int_{\Omega_2} \frac{3 \cos^2 \theta - 1}{R^3} dV_1 dV_2. \quad (12)$$

By using the dimensionless quantities introduced in section 2, this expression can be written as

$$F_{12} = -B_m \frac{q}{(\gamma + n)^2} F_{MC}, \quad (13)$$

$$F_{MC} = \int_{\Omega_1} \int_{\Omega_2} \frac{3 \cos^2 \theta - 1}{R^3} dV_1 dV_2 \quad (14)$$

(Here we have used the expression for $\mathbf{M}$: $\mathbf{M} = \frac{\chi}{1 + n\chi} \mathbf{H}_0$, obtained in Sect. 2). The error in calculation of the total energy F due to neglecting the value of F_{12} is

$$\Delta = \left| \frac{F_{12}}{2 F'_m} \right|,$$

where $F'_m = -B_m \frac{q}{2(\gamma + n)}$ is the magnetic portion of F' . Then Δ can be written as

$$\Delta = \frac{\chi}{1 + n\chi} |F_{MC}| \leq \chi |F_{MC}|.$$

Thus, in order to estimate the upper limit of Δ one should calculate F_{MC} , which depends on the aspect ratio $k = \frac{a}{b}$ and the distance L between the centres of the bases of the drops.

The Monte Carlo method was used to evaluate F_{MC} . Let us introduce the Cartesian coordinate systems (x_i, y_i, z_i) into each ellipsoid ($i = 1$ or 2 stands for the number of ellipsoid) as shown in figure 1. The x_i axes contain a straight line connecting the centres of the drops bases. After the transformation

$$x_i = bx'_i, \quad y_i = by'_i, \quad z_i = az'_i$$

we proceed, in equation (14), from the ellipsoids to the unit spheres :

$$F_{MC} = \int_{\Omega_1^S} \int_{\Omega_2^S} \frac{K(3 \cos^2 \theta - 1)}{(R')^3} dV'_1 dV'_2 \quad (15)$$

where Ω_i^S are the halvespheres of a unit radius,

$$dV'_i = dx'_i dy'_i dz'_i \quad (i = 1, 2)$$

$$R' = [(x'_1 - x'_2 + l')^2 + (y'_1 - y'_2)^2 + k^2(z'_1 - z'_2)^2]^{1/2}$$

$$\cos \theta = \frac{k(z'_1 - z'_2)}{R'}, \quad \text{and} \quad l' = \frac{L}{b}.$$

The Monte Carlo procedure reduces to sampling the N couples of random points $\{(x'_1, y'_1, z'_1), (x'_2, y'_2, z'_2)\}$ uniformly distributed in Ω_1^S and Ω_2^S , respectively, and to expressing F_{MC} in the form :

$$F_{MC} \approx \bar{F}_{MC} = \frac{1}{N} \sum_{n=1}^N \varphi_n$$

Fig. 4. — Error calculation : F_{MC} as a function of separation $l = \frac{L}{b}$ for several values of aspect ratio k .

① $k = 1.5$; ② $k = 2.5$; ③ $k = 4$.

where φ_n is the integrand (15) for the n -th couple. The error of calculation is found from the expression

$$\sigma_F = \left\{ \left[\frac{1}{N} \sum_{n=1}^N \varphi_n^2 - (\overline{F}_{MC})^2 \right] / N \right\}^{\frac{1}{2}}.$$

Calculations were carried out on the HP-3000 computer system with the use of the URAND random number generator [5]. The results of calculations, presented in figure 4, show that as the distance L between the drops increases, F_{12} tends monotonically to zero being positive everywhere, i.e. there exists dipole-dipole repulsion between Ω_1 and Ω_2 ; for high values of L : $F_{12} \sim 1/L^3$.

The error is $\Delta \lesssim 10^{-3}$. (The relative error of calculation $\frac{\sigma_F}{\overline{F}_{MC}} \sim 20\%$ for $N = 1000$).

In the experiments [1] the drops formed after the breakup were settled at a finite distance L from each other. It can be explained by non-uniformity of magnetic field at the base edge which results in the magnetic force $M \nabla H$ directed inside the region of the base. This non-uniformity was neglected in the present paper.

6. Conclusions.

1) The phenomenon of topological instability in the model suggested is fully described by the 3 dimensionless parameters: B_g , B_m , χ . It has been found that critical values of the magnetic Bond number B_m^c , corresponding to topological instability, decrease rapidly as the gravitational Bond number B_g^c increases, in the region $B_g^c \lesssim 8$.

2) It has been shown that there exists the limiting value $\tilde{B}_g = 5$, at which the topological instability is impossible.

3) For the finite volume of a strongly polarizable magnetic fluid ($\chi > 20$), resting on a solid base the existence of shape hysteresis has been predicted (first-order phase transition).

4) The interaction energy of the volumes produced by a breakup has been investigated. It has been shown that this energy is of the order of 10^{-3} - 10^{-4} of the magnetic self-energy of the system.

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