ANISOTROPY EFFECTS IN FERROFLUID INSTABILITIES

H. BERCEGOL [1], E. CHARPENTIER [2], J.M. COURTY [1] and J.E. WESFREID
LHMP-ESPCI, UA 857 CNRS, 10 rue Vauquelin, 75231 Paris Cedex 05, France

Received 27 January 1987; accepted for publication 3 March 1987

We study the effects of a tangential component of a magnetic field on the instability of a layer of ferrofluid submitted to a vertical field (Rosensweig instability). This anisotropy produces a stabilizing effect with a shift of the critical value of the normal field, which we measured and explain here. The new axis of symmetry induces "rolls" (lines of deflection) in the direction of the horizontal field and influences the formation of the hexagonal pattern of peaks.

1. Introduction

Ferrofluids or magnetic liquids display a great variety of instabilities in magnetic fields [1]. The most typical of them is the instability of the free surface of the ferrofluid submitted to a normal magnetic field as was discovered by Cowley and Rosensweig [2]. In this case a transition from a flat surface to a corrugated one with peaks occurs when the applied normal magnetic field is higher than a critical value H_c. The peaks display triangular spatial symmetry at onset (each peak has six nearest neighbours) and their profile has approximately a conical form. The origin of this instability is the amplification of capillar waves in the ferrofluid interface.

In this note, we report our experimental study on the shift of the critical value H_c when a horizontal magnetic field is superposed, with stabilizing effect [3]. The expected increase in the value of the threshold as a function of the horizontal magnetic field is found to depend only on the value of the magnetic permeability μ of the magnetic fluid. We also describe the anisotropy in the formation of the instability caused by a horizontal magnetic field.

2. Theory

In what follows we recall the main results of the onset value calculation for this instability. The dispersion relation for capillar waves (as $e^{i(\omega t-qx)}$) excited in the interface of two fluids, in half-infinite systems, is given by [4]

$$2\rho_m\omega^2=\delta\rho\, gq+\sigma q^3, \tag{1}$$

where ρ_m, $\delta\rho$ and σ are the mean density of both fluids, the difference in density and the surface tension between the two fluids respectively, and g the value of the gravity acceleration.

When a normal magnetic field is applied across a horizontal layer of ferrofluid with free surface, the dispersion relation (1) is modified by the presence of a negative term proportional to the square of the field [3,5]. This is also the case of dielectric liquids submitted to normal electrical fields [6,7].

The modified relation then becomes

$$2\rho_m\omega^2=\delta\rho\, gq+\sigma q^3-AH_v^2q^2/\mu, \tag{2}$$

where $A=\mu_0(\mu-1)^2/(\mu+1)$ where μ_0 and μ are the vacuum and relative magnetic permeability of the magnetic liquid respectively.

When the magnetic field increases, the frequency diminishes until H_v reaches its critical value $H_{vc}(q)$ where ω^2 becomes negative. The imaginary part of ω implies that the neutral waves get amplified unboundedly, hence instability of the surface. Nonlinear mechanism, which is not present in this linear description, allows saturation and consequently the observation of the bounded stationary final structure in the form of peaks of deformation of the flat interface. The marginal state ($\omega=0$) gives the law for

[1] Magistère de Physique (M.I.P.)/ENS.
[2] D.E.A. Physique théorique/ENS.

$H_{vc}(q)$ as function of the wavenumber q of the spatial Fourier component of the deformation of the interface. The line

$$H_{vc}^2(q) = (\delta\rho\, g/q + \sigma q)\mu/A \tag{3}$$

gives the boundaries between the stable and unstable region, and the minimum of this line, occurring at

$$H_{vc}^2 = (4\delta\rho\, g\sigma)^{1/2}\mu/A$$

$$\text{and} \quad q_c = (\delta\rho\, g/\sigma)^{1/2}, \tag{4}$$

gives the critical value for the applied magnetic field and the critical wavenumber respectively.

When a horizontal magnetic field (parallel to the undeflected surface) is applied, this field has a stabilizing effect, as was demonstrated by Zelazo and Melcher [3]. They calculated the following dispersion relation:

$$2\rho_m\omega^2 = \delta\rho\, gq + \sigma q^3 - AH_v^2 q^2/\mu + AH_h^2 q^2 \cos^2\alpha, \tag{5}$$

where H_h is the horizontal magnetic field and α the angle between $\boldsymbol{H}_h$ and $\boldsymbol{q}$.

They also performed dynamic experiments with excitation of capillary waves, verifying this relation for fields smaller than the critical field. From expression (5) it is clear that the last term (with the plus sign) gives a shift to upper values of the critical magnetic field (when $\omega=0$). A simple expression for this shift may be derived, as an expansion of (5), around the critical value of the wavenumber q as a consequence of the applied horizontal magnetic field. So, the expression for the change in the critical value of the vertical magnetic field (control parameter of the instability) as function of the applied horizontal field reads

$$\delta H_{vc} = \mu \cos^2\alpha\, H_h^2/2H_{vc}. \tag{6}$$

This correction to H_{vc} is a function of observable variables and depends on the initial magnetic permeability as unique parameter. This expression is only valid for linear ferrofluid, working far from saturation.

As is known, the spatial distribution of the peaks has a hexagonal array produced by three wavevectors of magnitude $q_i = q_c$ ($i = 1, 2, 3$) with the condition $\boldsymbol{q}_1 + \boldsymbol{q}_2 + \boldsymbol{q}_3 = 0$. In this case the elevation of the deflected surface, at the onset, may be represented by the function

$$h = \sum_{i=1}^{3} h_i \cos(\boldsymbol{q}_i \cdot \boldsymbol{x}) \quad \text{with} \quad h_i = h_0. \tag{7}$$

This is the typical function describing a hexagonal pattern in thermal convection [8]. As for the onset calculation there only exists interest in the linear coupling between the three modes at 120°, so the threshold condition may be written as

$$\omega_t^2 = \omega_1^2 + \omega_2^2 + \omega_3^2 = 0,$$

where ω^2 is given by formula (5) with the respective projections of each $\boldsymbol{q}$-vector on the direction of H_h ($q_{1x} = q$, $q_{2x} = -q_{3x} = q\sqrt{3/2}$ due to the triangular symmetry of the pattern, where x is the $\boldsymbol{H}_h$ direction).

Operating in the same way as for only one mode ("rolls") and developing around the critical value for H_{vc} and q_c (these values for the onset, with $H_h = 0$, for the hexagonal pattern are the same as in the case with only one mode, as is trivial) we obtain the following result:

$$\delta H_{vc} = \mu H_h^2/4H_{vc}. \tag{8}$$

One should notice the fact that the correction of the value of the onset (1/2 in the place of $\cos^2\alpha$) now is not a function of the orientation of the hexagonal pattern with respect to the applied horizontal field. In addition this geometrical prefactor corresponds to an orientation of 45° with respect to a fictitious roll.

3. Experiments

3.1. Experimental arrangement

Our experiments were performed with two kinds of different available commercial ferrofluids: the first one (FF1) with $\mu = 2.4$ is produced by surfactant as stabilizing agent [9] and the second one (FF2) with $\mu = 6.9$ is stabilized by ionic forces [10]. The sample containing the magnetic liquid is a cylindrical container 160 mm in diameter placed in the center of two Helmholtz coils of 600 mm in diameter, ensuring a vertical field homogeneity better than 3%. The values of the critical magnetic field (onset of the instability without horizontal field) for each ferrofluid are $H_{vcFF1}=157\pm3$ G and $H_{vcFF2}=130\pm5$ G. Another pair of coils produces a horizontal magnetic field with less homogeneity (10%). The depth of the layer is 10 mm, so the bond number B (or non-dimensional height, related to the wavelength of the periodical structure $\lambda=10$ mm) is $B=1$ so there is no correction in the dispersion relation related to the finite height of the sample [11].

In absence of the horizontal field, the deformed surface at the onset begins in the form of concentric rings ("rolls") growing from the boundaries toward the center (fig. 2a), reflecting, in our situation, the imperfect case of the bifurcation related to the magnetic enhanced meniscus wetting the side wall.

This is the typical configuration in containers within extended magnetic field homogeneity, as opposed to the case with initial development of the peaks in the center related to locally higher magnetic fields, when the diameter of the coils is not very large.

As all the peaks do not appear together we defined the critical field as the value of the vertical magnetic field as the last peak appears (i.e. as the peaks completely fill the container). In all our experiments we begin in fixing the horizontal field and we increase (or decrease) the vertical field to study the behaviour of the fluid at the onset. The hysteresis in the value of the onset, if finite, is very small in our case.

3.2. *Description of the influence of a horizontal magnetic field*

With a non-zero horizontal field we observe new phenomena: first the pattern does not grow initially with axisymmetry during the creation of the first peaks; second, with a strong horizontal field, we can see "rolls" (lines of deflection) appear before the peaks (1% under the critical value) (fig. 2b); third, for lower values of the horizontal field, there is no roll but we can observe that one direction of the hexagonal pattern is parallel to the horizontal field ($\boldsymbol{q}_1\perp\boldsymbol{H}_h$) (fig. 2c), and fourth, the onset value is higher.

It is possible to explain simply the dissymmetry of the formation of the peaks. As we have seen, the surface of the fluid is not flat: there is an enhanced meniscus caused by the vertical field, along the glass side walls; this meniscus has almost a circular symmetry, but the magnetic field is not vertical. So its normal (to the fluid surface) component has no circular symmetry. The side where the peaks appear first is the one where the local normal field is more intense. Experimentally we have checked this fact in observing the direction of the field lines and the side where the peaks appear first.

With a horizontal magnetic field above approximately a sixth of the vertical field we obtain rolls (we cannot observe an onset value). With $H_h\sim H_{vc}/4$, the rolls are as long as four wavelengths of the hexagonal pattern. They appear near the boundary of the cylinder (always on the side where the local normal field is higher) as described above. As H_v increases, and reaches close to its critical value, peaks appear on the rolls near the boundary. The peaks extend toward the other side, always preceded by the same length of roll. When the cell is full, one direction of the hexagonal pattern is perfectly parallel to the horizontal component of the magnetic field.

This particular arrangement of the peaks also exists for lower values of the horizontal field, even when there is no appreciable roll before the appearance of the peaks. When H_h is decreased (i.e. we perform new experiments with a lower value of H_h) the value of the critical field H_{vc} diminishes, and the rolls disappear, progressively replaced by the annuli; but the peaks – which appear in one side, on the annuli – go on appearing along directions parallel to H_h. There is a competition between the rolls parallel to the field and the rolls in annulus. And finally, when H_h is low (under $H_{cv}/15$) there is no apparent influence of the direction of the horizontal magnetic field on the disposition of the peaks.

3.3. *Study of the shift of the onset value*

We have observed the dependence of the value of the critical vertical field H_c on the value of the horizontal field H_h. We have checked that the relation between the square of the horizontal field and the shift of the critical value is linear as we predicted in (8) for low values of H_h (fig. 1).

We have also observed that the ratio of the slopes of the lines obtained for FF1 and FF2 is almost in quantitative agreement with our prediction: the observed value for the ratio of the slopes gives $\mu_{FF2}/\mu_{FF1}=3.8\pm0.9$, while the ratio of the values of μ given by the suppliers is 2.9. But the absolute slopes

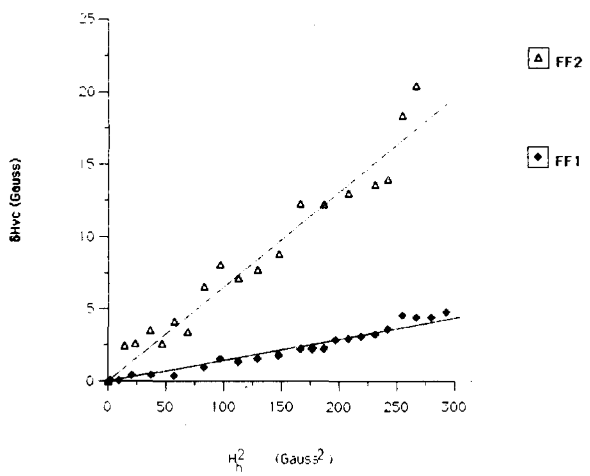

Fig. 1. Shift of the critical vertical magnetic field as function of the square of the horizontal field.

are greater than expected. One source of this disagreement may come from the fact that we have worked in the non-linear region of the magnetization curve, not far from saturation, without the necessary corrections with the values of the tangential permeability [1].

3.4. The different kinds of rolls (fig. 2)

The first one comes from imperfect bifurcation: before the onset the vertical magnetic field enhances the meniscus at the wall. This deformation of the surface is the cause of rolls. These rolls are parallel to the boundaries of the container. In our case, we use a cylindrical cell and we obtain concentric circular rolls separated by a distance equal to the wavelength of the hexagonal pattern.

The second one is obtained when a strong horizontal magnetic field is added to the vertical field. The straight rolls are parallel to the horizontal field and separated by a critical wavelength. This case was also observed by Barkov and Bashtovoi [12] for very strong horizontal magnetic fields and in the particular case of a square array of peaks, where the influence of the external rectangular cell is not discussed [13].

The third kind is created by an artificially inhomogeneous vertical field and is also related to imperfect bifurcation. In our case we create the inhomogeneity by placing metallic strips under the container, the rolls appear exactly above the strip (fig. 3).

A general property of the rolls is that, as the vertical field reaches the onset value, the first peaks always appear on the rolls.

We recall that the region where rolls exist is very short in the space of H_v ($\Delta H_v \sim 5$ G $\sim 0.05 H_{vc}$).

4. Conclusions

(1) We may explain the existence of rolls by saying that since q_x do not "see" H_h ($=H_x$), it is the first to appear and then we have rolls in the direction of the field before hexagons. Once the field is higher, the field restores the coupling between modes and hexagonal array appears, but at a higher onset. Perhaps a more elaborated experiment (measurement of the profile of the peaks) may resolve the spatial unequal contributions of each of the modes q_1, q_2, q_3 more than our visual experiment (indeed, probably $h_3 = h_2 \neq h_1$ in formula (7), with $\boldsymbol{q}_1 \perp \boldsymbol{H}_h$). This point must be explored in future work.

We have observed that the field of asymmetry modifies the onset of instability as in other cellular structures, where the general dispersion relation may be written as [14]

$$-(-1)^n (\omega/\omega_0)^n = \epsilon - \xi_0^2 (\delta q)^2 - \Gamma^2 ,$$

where $n=1$ in dissipative (diffusive) instabilities, as the Rayleigh–Bénard case, and $n=2$ in elastic or propagative instabilities, as in our case; ϵ is a relative distance to the onset; in this instability $\epsilon = (H_v^2 - H_{vc}^2)/H_{vc}^2$; $\omega_0^2 = \text{const} = \sigma q_c^3/\rho_m$ in this problem; ξ_0^2 is the square of the amplitude of the correlation length, given by the concavity of the marginal linear curve of stability, in our case this value is $\xi_0^2 = 1/2q_c^2$; and Γ^2 is the magnitude of the anisotrope field (up to a constant factor). When a horizontal magnetic field is applied we obtain $\Gamma^2 = \mu (H_h/H_{vc})^2 \cos^2\alpha$.

All these values are obtained by expansion of ω^2 around q_c and H_{vc} from eq. (5) with $(H_h/H_{vc})^2 \sim (\delta q/q_c)^2 \sim \epsilon$.

The interest now is to know the non-linear contributions once the onset is exceeded. It is clear that this analysis may explain the interval between rolls and (perhaps deformed) hexagons.

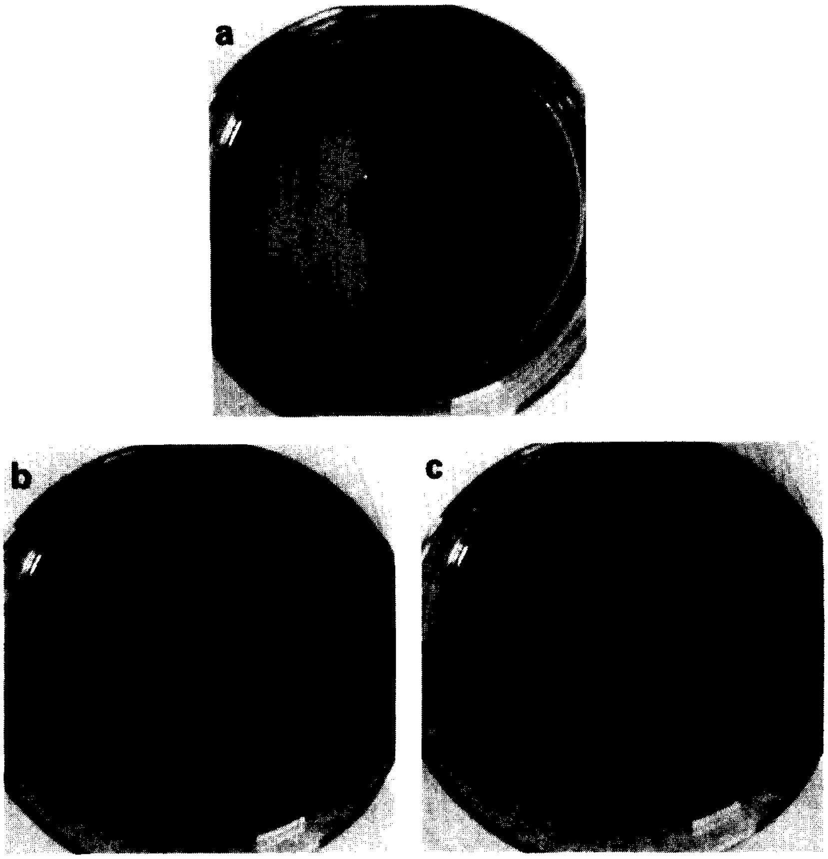

Fig. 2. (a) Concentric rolls in absence of horizontal field. (b) Rolls in the direction of the applied horizontal magnetic field. (c) Formation of peaks on the rolls.

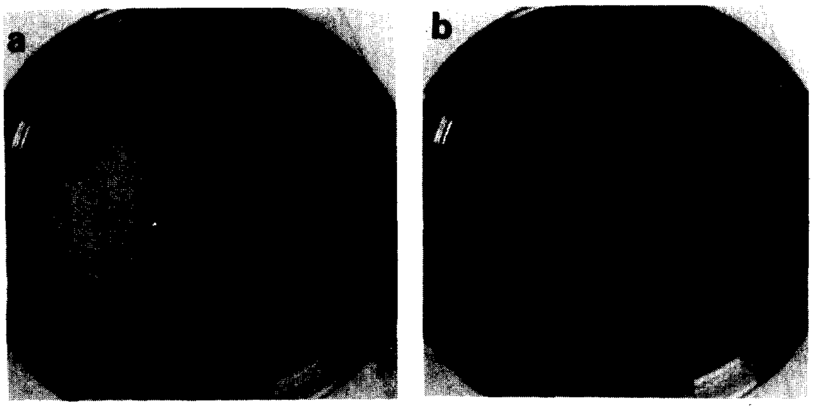

Fig. 3. (a) Roll induced by strip for H_v smaller than H_{vc} (in the direction of the strip). (b) Formation of peaks on the roll.

(2) In many other hydrodynamical instabilities the observation of the onset value was used for the determination of physical constants of the fluid [15]. In our problem the point of interest is the fact that the relation we have derived giving the shift of the onset value due to the stabilizing horizontal magnetic field (9) is a function of only one physical parameter of the fluid: the relative magnetic permeability μ. We have used this result for comparison between the magnetic permeabilities of ferrofluids FF1 and FF2.

Acknowledgement

We wish to thank the Laboratoire d'Electricité Générale (ESPCI), M. Martinet and D. Allais for their assistance.

References

[1] R.E. Rosensweig, Ferrohydrodynamics (Cambridge Univ. Press, Cambridge, 1985).
[2] M.D. Cowley and R.E. Rosensweig, J. Fluid Mech. 38 (1967) 671.
[3] R.E. Zelazo and J.R. Melcher, J. Fluid Mech. 39 (1969) 1.
[4] L. Landau and E. Lifshitz, Mécanique des fluides (Mir, Moscow).
[5] J.-P. Brancher and A. Sero Guillaume, J. Mec. Theor. Appl. 2 (1983) 265.
[6] Ya.I. Frenkel, Collection of selected works (Dover, New York, 1955).
[7] G.I. Taylor and A.D. McEvans, J. Fluid Mech. 22 (1965) 1.
[8] D.G. Christopherson, Q. J. Math. 11 (1940) 63.
[9] Ferrofluidics SARL (Palaiseau, France).
[10] Union Chimique Européenne (St. Fromond, France).
[11] V. Bashtovoi, M. Krakov and A. Reks, Magn. Gidrodin. 1 (1985) 19.
[12] Y. Barkov and V. Bashtovoi, Magn. Gidrodin. 4 (1977) 137.
[13] D. Allais and J.E. Wesfreid, preprint (1986).
[14] E. Guazelli, D. Walgraef and P. Dewel, J. Phys. (Paris), to be published.
[15] D.R. Caldwell, Rev. Sci. Instrum. 41 (1970) 1856.