J. Fluid Mech. (1967), *vol.* 30, *part* 4, *pp.* 671–688
Printed in Great Britain

The interfacial stability of a ferromagnetic fluid

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(Received 11 April 1967)

A normal magnetic field has a destabilizing influence on a flat interface between a magnetizable and a non-magnetic fluid. Stabilizing influences are provided by interfacial tension and gravity if the lighter fluid is uppermost. The critical level of magnetization for onset of the instability is derived for a fluid having a non-linear relation between magnetization and magnetic induction. Experiments using a magnetizable fluid, which contains a colloidal suspension of ferromagnetic particles, at interfaces with air and water are made and cover a wide range of density differences. Measurements confirm the prediction for critical magnetization, and it was found that, after onset, the interface took a new form in which the elevation had a regular hexagonal pattern. The pattern was highly stable, and the measured spacing of peaks agreed reasonably with that derived from the critical wave-number for the instability of a flat interface.

1. Introduction

Fluids with ferromagnetic properties have recently been synthesized on a laboratory scale (Rosensweig, Nestor & Timmins 1965). They are formed by a colloidal suspension of solid ferrite particles in a parent liquid. To stop the particles coalescing in the well-known manner of iron filings in a magnetic field, their size is made small enough for thermal agitation to have a significant dispersive influence, and they are also coated with a layer of surfactant which provides short-range repulsion. The resulting material behaves like a normal fluid except that it can experience forces due to magnetic polarization. So far, the successful fluids have been good insulators, and forces due to the interaction of magnetic fields with currents of free charge, familiar in magnetohydrodynamics, can be taken as negligible. In the present work, we shall discuss some experiments on the stability of a flat interface between a ferromagnetic and a non-magnetic fluid in the presence of uniform, normal, magnetic and gravitational fields.

Our interest in the problem is due partly to the unexpected observation that an instability occurred readily on a laboratory scale, partly to its tech-

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nological importance because the instability sets a limit to some engineering applications, and partly to the fact that interfacial phenomena provide one area where the fluid mechanics of a ferromagnetic liquid differs from that of a non-magnetic material. As discussed in §2.2 and appendix B, the magnetic forces cannot induce circulation in the absence of temperature gradients, and it is therefore possible for them to be balanced by pressure forces alone. Incompressible flow patterns remain unaffected by the forces (only pressure distributions are altered) unless there is a boundary condition which involves the pressure directly—as at an interface. If the magnetic force is to have any engineering application to the control of fluid motion, there must be an interface or possibly temperature gradients. Some fluid-mechanic problems involving the latter have been discussed recently by Neuringer (1966).

It is clear from the work of Melcher (1963) that a flat interface between two stationary fluids of which one at least is ferromagnetic may be unstable when there is a normal magnetic field. If the surface is perturbed, the magnetic flux is concentrated at the peaks, and the resulting forces tend to drive the perturbation further, while surface tension and gravitational forces (assuming light fluid over heavy) have a stabilizing influence. The instability is very similar in nature to that found at the interface between conducting and dielectric fluids in the presence of electric fields, and recently described by Melcher (1963) and Taylor & McEwan (1965). One of the important differences which we find experimentally in the ferromagnetic case is that a stable interface with a regular periodic structure (see figure 5, plate 2) is formed when the flat one can no longer be supported. This feature allows us to measure the spacing of the interfacial perturbations for comparison with the prediction of the usual type of linear theory.

Before giving the results of the experiments in §3, we need to derive the stability criterion which will be valid for our ferromagnetic fluids, where the relation between the magnetic field **B** and magnetic induction **H** is non-linear, but no hysteresis effect has been observed, although this fact has not been stated explicitly in previous work. The successful prediction of magnetic behaviour by Langevin's classical theory (see Rosensweig *et al.* 1965) suggests that hysteresis is unlikely, and therefore that **B** and **H** can be assumed to be parallel. The criterion is found to be similar to that implicit in Melcher's (1963) work for a linear material.

2. Analysis

Using a Cartesian co-ordinate system (x, y, z), we suppose that for unperturbed conditions the region $z < 0$ is filled with a homogeneous ferromagnetic fluid, while the fluid in $z > 0$ is non-magnetic. A uniform magnetic field $\mathbf{B}_0$ lies in the z-direction. Gravity is considered as acting in the opposite direction when the region $z < 0$ contains the denser fluid and towards positive z when the denser fluid is in the region $z > 0$. Electromagnetic and fluid properties in the magnetic and non-magnetic media are distinguished by the subscripts 1 and 2. M.K.S. units are used in the analysis.

We then look for the condition of neutral stability of the interface, i.e. the condition which allows it to rest at a position $z = z_0(x, y)$, where z_0 is a periodic function of small amplitude in comparison to wavelength.

2.1. *The perturbed magnetic field*

We linearize the magnetic-field problem by writing

$$\mathbf{B} = \mathbf{B}_0 + \mathbf{b}, \quad \mathbf{H} = \mathbf{H}_0 + \mathbf{h}, \tag{1}$$

where the magnitudes of $\mathbf{b}$ and $\mathbf{h}$ are assumed small in comparison to H_0 and B_0. To first order, the change in the magnitude B is related to that in H by the slope of the (B, H)-curve, so that

$$\left.\begin{aligned} b_z &= \hat{\mu} h_z, \\ \hat{\mu} &= \partial B/\partial H \quad \text{at} \quad H = H_0. \end{aligned}\right\} \tag{2}$$

$\hat{\mu}$ is then an effective permeability for perturbations of the z-components. Assuming $\mathbf{B}$ and $\mathbf{H}$ are parallel, we also have

$$\left.\begin{aligned} b_x &= \mu h_x, \quad b_y = \mu h_y, \\ \mu &= B_0/H_0, \end{aligned}\right\} \tag{3}$$

and μ is the effective permeability for transverse perturbations.

Since there are no free currents, $\mathbf{h}$ is irrotational and can be expressed in terms of a magnetic perturbation potential ϕ, where

$$\mathbf{h} = \operatorname{grad} \phi. \tag{4}$$

Substituting from equation (4) in (2) and (3) and using the fact that $\mathbf{b}$ is solenoidal, we obtain

$$\mu \frac{\partial^2 \phi}{\partial x^2} + \mu \frac{\partial^2 \phi}{\partial y^2} + \hat{\mu} \frac{\partial^2 \phi}{\partial z^2} = 0. \tag{5}$$

In the non-magnetic medium, $\hat{\mu} = \mu = \mu_0$, the permeability of free space, and (5) naturally reduces to Laplace's equation.

Linearizing the boundary conditions, continuity of the tangential components of $\mathbf{H}$ requires

$$[h_x + H_0 \partial z_0/\partial x] = 0, \quad [h_y + H_0 \partial z_0/\partial y] = 0,$$

where the square brackets denote the jump in the quantity enclosed. Expressing h_x and h_y in terms of the magnetic potential and integrating, we obtain

$$[\phi] = -z_0[H_0], \tag{6}$$

having arbitrarily set the constant of integration to zero. Continuity of the normal component of $\mathbf{B}$ requires

$$[b_z] = 0. \tag{7}$$

In accordance with the linearization we apply the conditions at $z = 0$, and also it is convenient to express equation (6) in terms of the magnetization $\mathbf{M}$, where

$$\mathbf{B} = \mu_0(\mathbf{M} + \mathbf{H}). \tag{8}$$

Since $[B_0] = 0$, we have $[M_0] = -[H_0]$, and the conditions (6) and (7) may be given in their final form as

$$\left.\begin{aligned} \phi_1 - \phi_2 &= z_0 M_0, \\ \hat{\mu}\frac{\partial \phi_1}{\partial z} &= \mu_0 \frac{\partial \phi_2}{\partial z} \quad \text{at} \quad z = z_0, \end{aligned}\right\} \tag{9}$$

where we have dropped the subscript 1 on M_0, $\hat{\mu}$ and μ since only the one fluid is magnetized. Note that the first condition of (9) is equivalent to a distribution of magnetic sources with strength proportional to height of perturbation.

Seeking a solution which allows for periodic variation in z_0 and for which the perturbations decay with distance from the boundary, we find in the magnetic fluid

$$\phi_1 = \frac{z_0 M_0 \exp\{kz\sqrt{(\mu/\hat{\mu})}\}}{1+r}, \tag{10}$$

where r is a composite relative permeability $\sqrt{(\hat{\mu}\mu/\mu_0^2)}$, and in the non-magnetic material

$$\phi_2 = \frac{r z_0 M_0 \exp(-kz)}{1+r}. \tag{11}$$

k is the wave-number of the interfacial disturbance, which must satisfy

$$\frac{\partial^2 z_0}{\partial x^2} + \frac{\partial^2 z_0}{\partial y^2} + k^2 z_0 = 0. \tag{12}$$

The perturbation of the field magnitude at the interface is then

$$b_z = \frac{k z_0 \mu_0 M_0}{1 + 1/r}, \tag{13}$$

which implies a concentration of flux at the peaks of the disturbance with b_z a maximum where z_0 is maximum.

2.2. *Magnetostatics*

It is convenient to represent the stress tensor in a stationary ferromagnetic fluid by

$$\sigma_{ij} = -(p^* + \tfrac{1}{2}\mu_0 H^2)\,\delta_{ij} + H_i B_j. \tag{14}$$

This equation is believed to be valid for a fluid with a non-linear (B, H)-relation, and its derivation by the usual thermodynamic arguments is discussed in appendix A. Since **B** and **H** are parallel, the last term can also be written as $H_j B_i$ or $(B/H)\,H_i H_j = \mu H_i H_j$. The latter is the usual expression for the stress due to tension along field lines found in linear theory (see, for example, Landau & Lifschitz 1960). The isotropic part of the stress tensor is expressed merely for our convenience in terms of a particular magnetic pressure, while the remainder is defined as an effective pressure p^*. In continuum theory the definition of pressure is arbitrary, and, as Brown (1951) pointed out, this fact reconciles some of the conflicting expressions for magnetic forces which have been given in the past. In a compressible fluid further details of p^* would be necessary in order to relate it to the thermodynamic state, expressed, say, in terms of density, temperature, and magnetic induction (see equation (A 6)), but in the present incompressible problem the information is unnecessary.

The i-component of the force $\mathbf{f}$ per unit volume exerted on a fluid element is given by $\partial\sigma_{ij}/\partial x_j$, so that from (14)

$$\mathbf{f} = -\operatorname{grad} p^* - \mu_0 H \operatorname{grad} H + \mathbf{H} \operatorname{div} \mathbf{B} + (\mathbf{B}\,.\operatorname{grad})\,\mathbf{H},$$

and using a well-known vector identity we write

$$\begin{aligned}(\mathbf{B}\,.\operatorname{grad})\,\mathbf{H} &= (B/H)\,(\mathbf{H}\,.\operatorname{grad})\,\mathbf{H}\\ &= (\operatorname{curl}\mathbf{H}) \times \mathbf{B} + B \operatorname{grad} H.\end{aligned}$$

Substitution gives

$$\mathbf{f} = -\operatorname{grad} p^* + (\operatorname{curl}\mathbf{H}) \times \mathbf{B} + \mu_0 M \operatorname{grad} H, \tag{15}$$

where we have introduced the magnetization from equation (8), and set $\operatorname{div}\mathbf{B} = 0$. The second term in the expression for $\mathbf{f}$ is the usual force due to free currents and will be zero here. If we assume that M is a function of H and temperature T only (i.e. the fluid is homogeneous in composition), the last term can be expressed as

$$\operatorname{grad} \int \mu_0 M \, dH$$

in an isothermal fluid, and is irrotational. Then no circulation would be induced by the magnetic force in an incompressible fluid. For our linearized theory the term can be written as $\mu_0 M_0 \operatorname{grad} h_z$, and then, adding the effect of gravitational forces to (15), we find that under conditions of fluid equilibrium

$$p^* \pm \rho z - \mu_0 M_0 h_z = \text{const.}, \tag{16}$$

the positive and negative signs being taken for gravity acting in the directions of $-z$ and $+z$ respectively. The question arises as to whether our formulation is adequate in the face of small but finite compressibility. The elementary arguments of this section are supplemented by a more rigorous justification in appendix B.

From equation (14), the change in tangential stress across the interface $[H_{x,y} B_z]$ is zero, as it should be, since tangential H and normal B components are continuous. The change in normal stress is given by

$$\begin{aligned}[\sigma_{zz}] &= -[p^* + \tfrac{1}{2}\mu_0 H^2] + [H_z B_z]\\ &= -[p^*] + (1/\mu_0)\,[-\tfrac{1}{2}(B_z - \mu_0 M_z)^2 + B_z(B_z - \mu_0 M_z)].\end{aligned}$$

Hence

$$[\sigma_{zz}] = -[p^*] - [\tfrac{1}{2}\mu_0 M_z^2], \tag{17}$$

where again we have used the facts that tangential H and normal B components are continuous. A normal stress difference, as given by (17), can be supported by the interfacial tension T, where to the order of approximation required

$$\sigma_{zz2} - \sigma_{zz1} = -T\left(\frac{\partial^2 z_0}{\partial x^2} + \frac{\partial^2 z_0}{\partial y^2}\right) = k^2 z_0 T,$$

using equation (12). Substituting from (17), we replace M_z^2 by the linear approximation $M_0^2 + 2M_0 m_z$, where m_z is the perturbation magnetization, and use equation (16) to eliminate p_1^* and p_2^*, thus obtaining

$$\mu_0 M_0 m_z + \mu_0 M_0 h_z \pm \rho_2 g z_0 \mp \rho_1 g z_0 - k^2 T z_0 = \text{const.} - \tfrac{1}{2}\mu_0 M_0^2. \tag{18}$$

It is during the last step that any ambiguity introduced by the arbitrary definition of p^* is removed. Thus, if we re-define p^* by inserting an extra function of H and B in the isotropic part of the stress tensor (14), the magnetic body force in (15) and the interfacial stress difference (17) are both modified, but the effects cancel in deriving (18).

Equation (18) is satisfied by setting both sides equal to zero, and therefore

$$M_0 b_z = (g\,\Delta\rho + k^2 T)\,z_0, \tag{19}$$

where $b_z = \mu_0(m_z + h_z)$ is evaluated at $z = 0$ and $\Delta\rho$ is the density difference across the interface, defined to be positive.

2.3. *Stability criterion and discussion*

Substituting b_z at $z = 0$ from equation (13) in equation (19) we obtain the condition for $z_0 \neq 0$ as

$$\mu_0 M_0^2/(1+1/r) = g\,\Delta\rho/k + kT. \tag{20}$$

In general we might expect T to be a function of the thermodynamic state of the fluid and hence of the magnetization, but we have found no experimental evidence to suggest a significant dependence, and we assume it to be constant. The minimum value of the magnetization which can support a neutrally stable perturbation of the interface is given by

$$\left.\begin{aligned} \mu_0 M_{\text{crit}}^2(1+1/r) &= 2\sqrt{(g\Delta\rho T)},\\ r^2 &= \left(\frac{B}{\mu_0^2 H}\frac{\partial B}{\partial H}\right), \end{aligned}\right\} \tag{21}$$

and the wave-number under these conditions is

$$k_{\text{crit}} = \sqrt{(g\Delta\rho/T)}. \tag{22}$$

The expression (21) for M_{crit} can be related to the analysis of Melcher (1963), who works in terms of H and μ, for a linear material. The present form emphasizes the fact that it is the magnetization which is the important quantity. The range of $1/r$ lies between unity for a poor material (low effective permeability or well past saturation) and zero for infinite permeability. Hence, even if r is unknown, (21) predicts the critical magnetization within a factor $\sqrt{2}$. (Using the reasonable assumption $B/H \gg \partial B/\partial H \gg \mu_0$ closer limits can be specified in terms of B_0 and H_0.) Kerosene has been the usual parent liquid for the ferromagnetic fluids, and, inserting kerosene values of surface tension and density for T and $\Delta\rho$ in (21), we obtain $M_{\text{crit}} < 6{\cdot}7 \times 10^3\,\text{A/m}$ ($\mu_0 M_{\text{crit}} < 8{\cdot}4 \times 10^{-3}\,\text{Wb/m}^2 = 84\,\text{gauss}$). This value of magnetization is low by ferromagnetic standards, and it is not surprising that the interfacial instability became dramatically apparent in the early stages of the development of the fluids.

The limit of $r \to \infty$, infinite permeability, provides the exact analogue of the instability of the interface between conducting and dielectric fluids in the presence of electric fields (Melcher 1963; Taylor & McEwan 1965). The interface becomes a surface of constant magnetic potential (compare the condition of constant electric potential), and we have $M_0 = B_0/\mu_0, = H_0$ in the non-magnetic medium, which is the gradient of the magnetic potential and the analogue of the voltage gradient.

For the sake of simplicity in the above analysis, we have assumed infinite depth of ferromagnetic fluid. Finite depth would make no difference in the case of $r \to \infty$, but for our fluids r is less than 1·9. It is clear from equation (10) for the perturbation potential that the scale length is $1/k\sqrt{(\mu/\hat{\mu})}$, and the effect of finite depth d is negligible if $kd\sqrt{(\mu/\hat{\mu})} \gg 1$. A more detailed analysis has been made for the case of a solid boundary at $z = -d$, the infinite region $z < -d$ being non-magnetic. The resulting modification to (20) gives a correction factor α to the left-hand side, where

$$\alpha = \frac{1-\{(r-1)/(r+1)\}\exp\{-2kd\sqrt{(\mu/\hat{\mu})}\}}{1-\{(r-1)^2/(r+1)^2\}\exp\{-2kd\sqrt{(\mu/\hat{\mu})}\}}$$
$$\simeq 1-\{2(r-1)/(r+1)^2\}\exp\{-2kd\sqrt{(\mu/\hat{\mu})}\}. \tag{23}$$

It is interesting to note that no correction is necessary for a poor material, $r \to 1$. In this limit the field perturbation is that due to a sheet of sources of magnetic potential at $z = 0$ with strength $M_0 z_0$, while the surrounding regions are effectively non-magnetic everywhere; the presence of a solid boundary at $z = -d$ is then unimportant. For our recorded experiments, the minimum value of kd was 5·5, and the correction clearly has negligible influence on the critical values of M_0 and k.

Finally, we can relate the wave-number to a measurable quantity, such as spacing between peaks l if we assume a shape for the perturbed interface. In our experiments the patterns were predominantly hexagonal (see figure 5, plate 2) and could have been members of the family of solutions to equation (12):

$$z_0 = \epsilon\{\cos\tfrac{1}{2}k(\sqrt{3}x+y)+\cos\tfrac{1}{2}k(\sqrt{3}x-y)+\cos(ky+\theta)\}$$
$$= \epsilon\{\cos\tfrac{1}{2}\sqrt{3}\,kx\cos\tfrac{1}{2}ky+\cos(ky+\theta)\}, \tag{24}$$

where θ is a parameter which controls the height of peaks above mean to depth of troughs, as indicated in table 1. $\theta = 0$ represents high peaks above a plateau of shallow depressions, while for $\theta = \frac{1}{2}\pi$ the surface is symmetrically distributed about $z = 0$. These cases were discussed in detail by Christopherson (1940). For all values of θ, the spacing between one peak and the next nearest for the critical condition is given by

$$l_{\text{crit}} = 4\pi/(\sqrt{3}\,k_{\text{crit}}). \tag{25}$$

θ	Height of peak above mean	Depth of trough below mean
0	3ϵ	$3\epsilon/2$
$\frac{1}{2}\pi$	$3\sqrt{3}\,\epsilon/2$	$3\sqrt{3}\,\epsilon/2$
π	$3\epsilon/2$	3ϵ

TABLE 1

3. Experiment

3.1. *Fluid properties*

The ferromagnetic fluids used in the experiments were prepared by grinding magnetite to particles of submicron size in a parent fluid of kerosene to which 5·8 % by weight of oleic acid had been added. Oleic acid provides the surfactant which prevents flocculation of the particles, and it is believed that less than 10 %

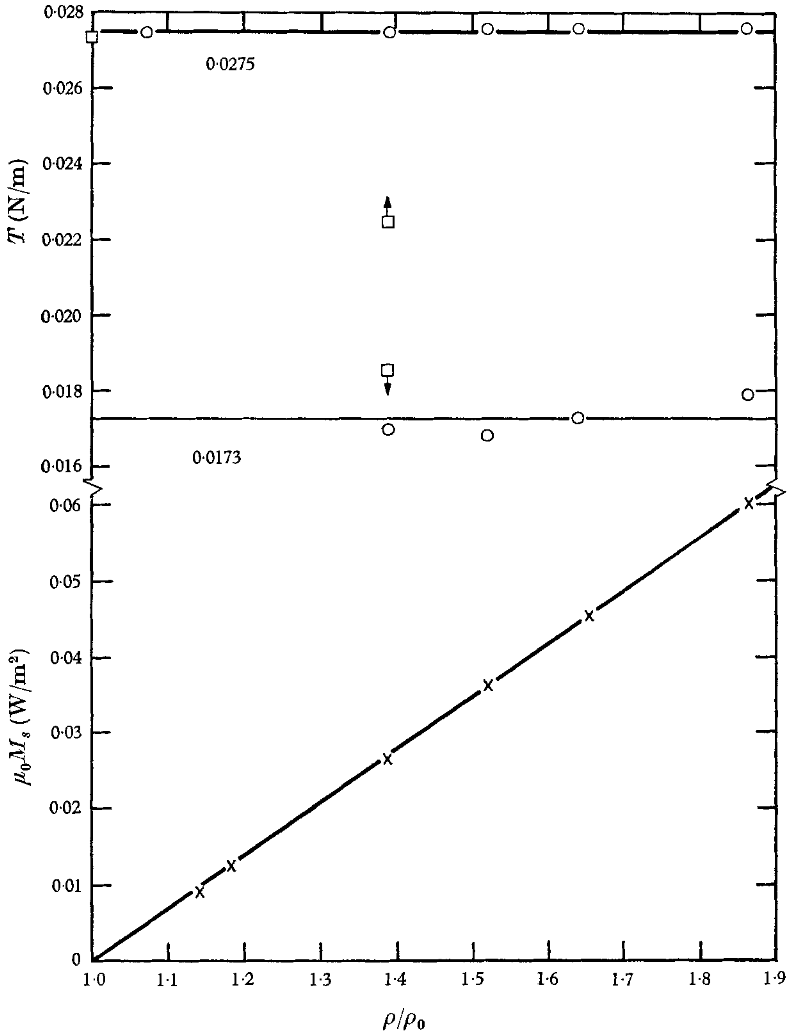

FIGURE 1. Fluid properties. Variation of interfacial tension T and saturation magnetization M_s with relative density ρ/ρ_0 where $\rho_0 = 792\,\mathrm{kg/m^3}$, the density of pure kerosene. ○, tensiometer measurements; □, drop-weight measurements.

of the surfactant remains unattached in the finished fluid. The solid magnetite has a saturation magnetization M_s of $4{\cdot}5 \times 10^5$ A/m ($\mu_0 M_s = 0{\cdot}566\,\mathrm{Wb/m^2}$) and density of $5000\,\mathrm{kg/m^3}$ compared to a density $\rho_0 = 792\,\mathrm{kg/m^3}$ for the kerosene.

For tests of an interface below either air or distilled water, a fluid of density $1{\cdot}863\rho_0$ was prepared, and subsequently diluted with pure kerosene to give a range of densities and magnetization curves. In the following discussion we shall refer to the fluids in terms of their relative density ρ/ρ_0.

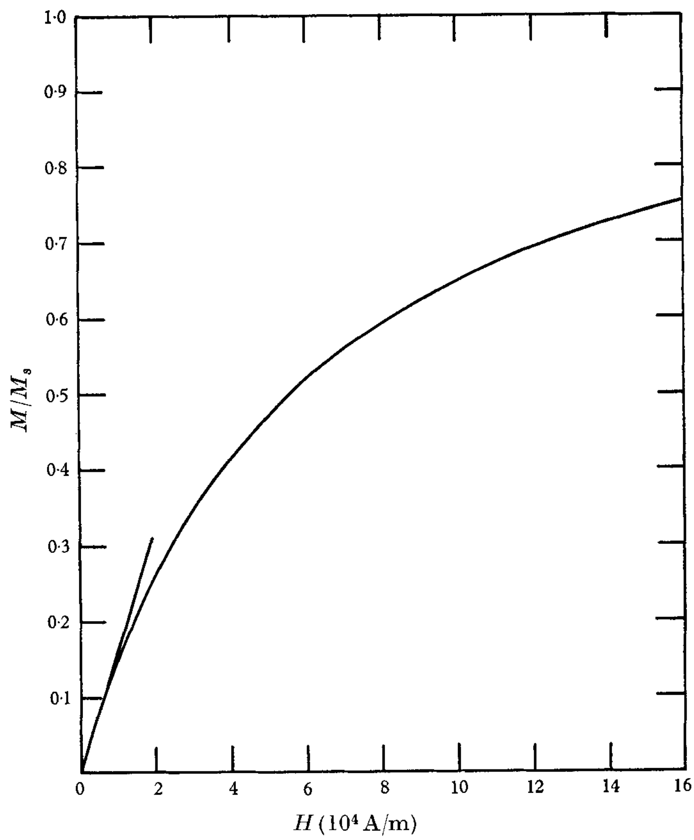

FIGURE 2. Universal magnetization curve. Initial slope $1{\cdot}66 \times 10^{-5}$ m/A.

Magnetization curves were determined by inserting a cylindrical sample of the fluid with a length to diameter ratio of four into a region of uniform field. A ballistic galvanometer connected to a search coil surrounding the region recorded the change in magnetic flux. The measured values of saturation magnetization shown in figure 1 were found to vary linearly with density of the fluid to the estimated accuracy of the experimental readings, $\pm 1{\cdot}5\,\%$. Extrapolation to the density of magnetite gives 65 % of the saturation value of the pure solid; the discrepancy is thought to be due to foreign particles introduced during the grinding. Similarly, magnetization at any value of H was found to vary linearly with density, and we give the general relation of M/M_s to H in figure 2.

Surface tension was measured by the drop-weight method and also with a Du Nouy tensiometer (see, for example, Davies & Rideal 1963). The former method gave reproducible results, whose values, however, depended on the size of tube used to form the drops, and we have more confidence in the results of the second method, in which a wire ring is pulled away from the interface. Results of the measurements are shown in figure 1. Each point represents an average of five determinations with standard deviation 0·5 % or less. Additional values given by the drop-weight method are included to show the possible uncertainty. The variation of the interfacial tension with density is not great (as measured by the tensiometer), and we have therefore chosen mean values of 0·0275 N/m for the air interface and 0·0173 N/m for the water interface. Since kerosene is not a pure compound, but a mixture of a number of components,

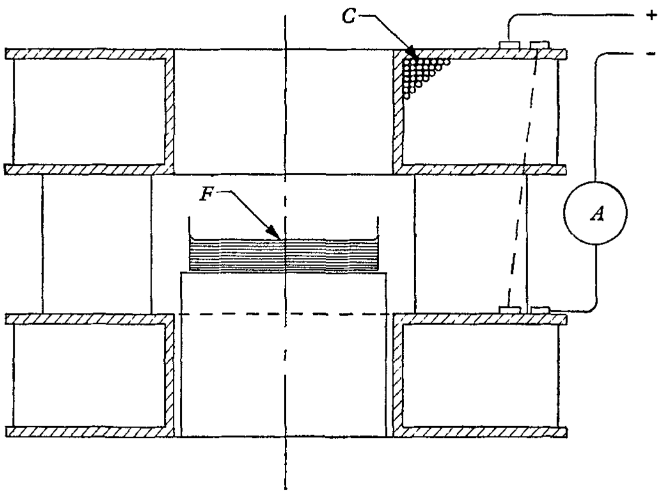

FIGURE 3. Experimental arrangement. The regulated power source supplies current through the ammeter A to the coils C and subjects the ferromagnetic fluid F to an approximately uniform normal, magnetic field.

published values for the surface tension vary between 0·023 and 0·030 N/m at 20 °C (Leslie & Geniesse 1933). Our measured value of 0·029 N/m for pure kerosene and air is within that range and is close to the one chosen for the ferromagnetic fluid, while pure kerosene and water gave 0·0374 N/m. In the latter case, the difference from 0·0173 N/m is due to the fact that the interfacial tension with water is dependent on the presence of oleic acid.

3.2. *The interfacial phenomena*

Two coils, arranged approximately as a Helmholtz pair, provided the magnetic field for the tests (see figure 3). The current supply was measured by an ammeter which was calibrated against level of magnetic field between the coils using a Bell type 120 gaussmeter with Hall probe and standardizing magnets. The field was uniform to 2·3 % over the half radius at the test position of the interface and 11 % over-all, while the presence of the strongest ferromagnetic fluid altered the field calibration by 5 % (allowed for in the case of the strongest fluid only).

For the test recorded in §3.3 of an interface between air and ferromagnetic fluid, a glass Petri dish of 10 cm diameter was filled to a depth of 16–20 mm and placed between the coils. As the strength of the magnetic field was raised from zero, the interface remained flat initially except at the edges. There the curved surface due to capillary attraction and field fringing allows an interaction with the magnetic field (the fluid tended to climb up the glass wall). At a particular level of field, which was taken to be the critical B_{crit}, the pattern of light reflexion at the centre changed, indicating a small perturbation. Increasing the field by less than 2 % usually produced a clear central peak surrounded by six hexagonally arranged lesser peaks (see figure 4, plate 1). With further increase, the amplitude of the peaks grew, and more peaks were formed until, at a field level approximately 10 % above critical, the whole interface was covered except for a narrow band near the edges. The final pattern formed a remarkably regular hexagonal array, apart from a few dislocations, with approximately uniform amplitude throughout (see figure 5, plate 2). We do not know why the hexagonal pattern is preferred; possible reasons include not only second-order effects but also the circular boundary of the dish and the slight non-uniformity of the field, which retained axial symmetry.

The interface appeared to have the elevation pattern of the zero-θ member of the family given by (24) (see table 1), but no precise measurements of surface profile were taken.†

The spacing between peaks varied from 12 to 9·5 mm for the tests of an interface with air, and approximately fifty peaks would be formed at full coverage. In any one test the spacing appeared to remain constant from the first small deflexion of the interface to a point where the amplitude was sufficient for accurate measurement with a pair of non-magnetic dividers. With further growth no change could be measured until at a height which was much larger than the spacing a slight deviation could be detected (in one case the pattern changed to square array at a field 40 % above critical).

The interfacial pattern was highly stable and, if the fluid was stirred slowly, would re-form immediately behind the stirrer. When the field was raised enough to make the peaks grow into long spikes, attempts to chop them off with a non-magnetic blade were fruitless; the blade merely passed through.

Similar tests to the ones described above were run with a layer of distilled water over the ferromagnetic fluid in order to give smaller density differences and interfacial tension. A fluid of relative density 1·522 gives a critical spacing which is nearly twice that of the air tests (see figure 7), and consequently the size and shape of the container could be more important. However, at the onset of the instability, it formed a clear pattern of a central peak with six surrounding.

We also tried the following tests: air and a fluid of low magnetization at saturation, water above a fluid of only slightly greater density, and water below a fluid of slightly lower density. In these cases the interface deformed greatly before the instability occurred because non-uniformity of the field could cause large magnetic forces in comparison to gravitational. A more significant test was that of an interface between ferromagnetic fluid and a thin layer of mercury.

† The photographic technique of figure 5 picks out flats on the interface, i.e. peaks, troughs and saddles. The positions of the latter for $\theta = 0$ correspond well with the light pattern shown. $\theta = \frac{1}{2}\pi$ would give a measurably different pattern.

We observed the point at which the mercury layer was pierced since the ferromagnetic fluid is also opaque. The thinnest layer of mercury which would cover the bottom of the container corresponded to $2/k_{crit}$, where k_{crit} is the calculated value, so that the perturbation was not small at the point of observation. However, the test gave a reasonable value of magnetization at 30% above critical.

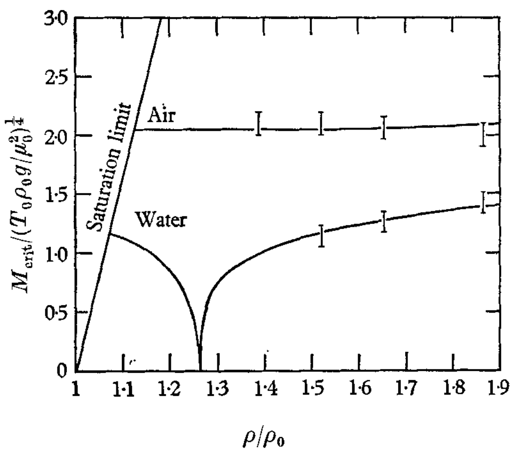

FIGURE 6. Comparison of the predicted critical magnetization with experiment.

3.3. *Results*

Since properties of the ferromagnetic fluids are expected to vary continuously with relative density, we have taken that as the experimental variable. The expression for the critical magnetization (21) can be written in non-dimensional form as

$$\frac{M_{crit}}{(g\rho_0 T_0/\mu_0^2)^{\frac{1}{4}}} = \left(4(1+1/r)^2 \frac{\Delta\rho}{\rho_0}\frac{T}{T_0}\right)^{\frac{1}{4}}, \tag{26}$$

where $\rho_0 = 792\,\text{kg/m}^3$ and $T_0 = 0{\cdot}0275\,\text{N/m}$, the values for pure kerosene. On the right-hand side r depends on the magnetization as well as the relative density. Similarly, spacing for hexagonal patterns from (22) and (25) can have the form

$$\frac{l_{crit}}{(16\pi^2 T_0/3\rho_0 g)^{\frac{1}{2}}} = \left(\frac{T}{T_0}\frac{\rho_0}{\Delta\rho}\right)^{\frac{1}{2}}. \tag{27}$$

The predicted curves for air and water, showing the variation with relative density ρ/ρ_0 of these non-dimensional forms of M_{crit} and l_{crit}, are given in figures 6 and 7. The critical magnetization for an air interface is nearly constant; although a slight density dependence might be expected from equation (26), it is offset by the increase in the effective permeability r. At a water interface the critica magnetization falls to zero for zero density difference and this point separates a part of the curve corresponding to water below from a part with water above. Experimental points are shown with error bars for the estimated uncertainty in deriving the magnetization from the measurement of magnetic field and the experimental determination of the magnetization curve (figure 6). In figure 7 bars indicate the estimated uncertainty in measurement of the spacing. Errors due to uncertainty in the interfacial tension are not included. If the tensiometer measurements are to be believed, the choice of a mean value of 0·0173 introduces an additional error of less than 1 % in magnetization and less than 2 % in spacing for the water interface, while errors for the air interface are negligible. However, if the drop-weight measurements truly indicate the uncertainty, additional errors for a water interface could be 2 % in magnetization and 4 % in spacing, while for the air interface they could be 5 and 10 %.

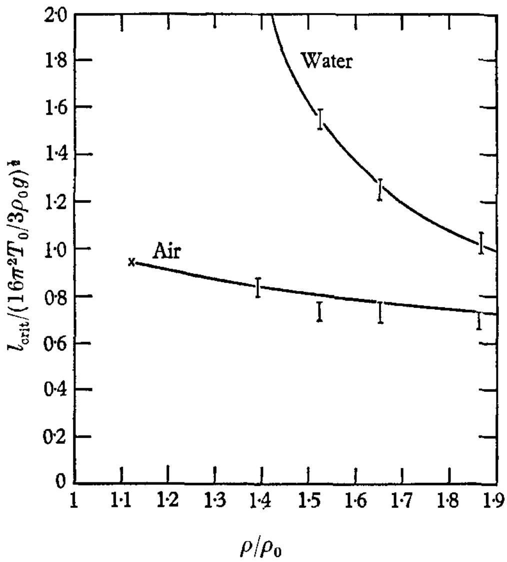

FIGURE 7. Comparison of the predicted critical length with experiment. ×, saturation limit.

The results for the critical magnetization at an air interface, for which the $\frac{1}{4}$-power density dependence is weak, confirm the prediction of (26) that it is the level of magnetization which is the important quantity for determining the onset of instability. It is worth recording that the critical magnetic field varied by a factor of 2·5 in the air-interface tests. Density dependence in the water-interface tests is more significant, but the variation is hardly beyond the range of estimated error. However, the relationship between the different curves is adequately predicted, and the density difference varied by a factor of 7·2 between the weakest ferromagnetic fluid with water and the strongest with air. Simultaneously the interfacial tension varied by a factor of 1·59 between water and air, so that the effect on critical magnetization was not great. However, the test with mercury described in §3.2 added a further factor of 8·2 to the density variation and 12·7 for the interfacial tension. Although the actual critical magnetization can only be said to be less than 1·3 times that predicted for mercury, there is a reasonable indication that the power law is being followed.

The results for critical spacing (figure 7) are remarkable for the type of phenomenon; of particular note is the agreement for the case of the water interface where the $\frac{1}{2}$-power density dependence gave a reasonable variation. The case of an air interface with relative density 1·522 was repeated, but in each case gave the result recorded.

4. Conclusions

From the experimental results we concluded that the observed instability was of the type suggested by Melcher (1963); i.e. it was governed by the stabilizing influences of gravity and interfacial tension. The theory for a non-linear material emphasizes the importance of magnetization as a critical field quantity, and this is confirmed although the degree of non-linearity was not great (M_{crit}/M_s ranged from 0·3 downwards—see figure 2). A more sensitive prediction is the dependence of equation (26) on the factor $(1+1/r)^{\frac{1}{2}}$, but this quantity only varied by 8% over the whole range of the experiments, and the order of accuracy required to show the effect is far greater than could be expected in an experiment involving interfacial tension.

The authors wish to thank Prof. J. A. Fay for his helpful criticism of this paper. The work was supported in part by NASA Research Grant NGR-22-009-052 and in part by the NASA Fluid Physics Branch, Research Division, Office of Advanced Research and Technology, under contract NASW-1219.

Appendix A. The stress tensor in a magnetizable fluid

To allow for a non-linear (B, H)-relation when hysteresis is absent, only a minor modification is necessary to the theory which gives the stress tensor in linear materials. The latter is derived by thermodynamic arguments in several text-books (see, for example, Landau & Lifschitz 1960). Since the topic is an unfamiliar one in fluid mechanics, we shall outline a complete derivation, choosing a method which is similar to that of Landau & Lifschitz, but more direct, since they work by analogy from the stress in an electrically polarizable material. The derivation is based on the fact that the change in free energy during an isothermal deformation is equal to the sum of the work done on the fluid by the unknown stresses and of the work delivered electromagnetically via field coils.

We consider a homogeneous layer of fluid which is confined between parallel planes (see figure 8). A uniform magnetic field can be generated by sheet currents in the planes and returned through an external path of zero reluctance. The angle of the field is determined by the current connexions in the following way. We can choose a closed loop, consisting of a straight line (e.g. PQ) across the layer of fluid and completed through the zero reluctance, which links no total current. Then the line integral $\oint \mathbf{H}.d\mathbf{l}$ is zero, and, since $\mathbf{H} = \mathbf{0}$ outside the fluid, there can be no component of $\mathbf{H}$ (and $\mathbf{B}$ if parallel) along PQ. We indicate this schematically in figure 8 by showing discrete wires, rather than sheet currents, connected at an angle ψ to the bounding planes, so that $\mathbf{H}$ and $\mathbf{B}$ are directed at $\frac{1}{2}\pi + \psi$. The exterior has to act also as a constant-temperature reservoir.

If the bounding planes are held rigidly (no deformation of the fluid) while H is increased under isothermal conditions, the change in free energy F per unit volume in the layer is just the electrical work $\int_0^B H\,dB$. The condition of zero reluctance outside implies that no field energy can be stored there. F is defined as $U-TS$, where U, T and S are internal energy, temperature and entropy. Hence we have

$$F(\rho,T,H) = F_0(\rho,T)+\int_0^B H\,dB$$

$$= F_0(\rho,T)+HB-\int_0^H B\,dH, \qquad (\mathrm{A}\,1)$$

where F_0 is the free energy at the same density and temperature, but zero H, and the integral is to be considered as the area under the (B,H)-curve for constant density and temperature.

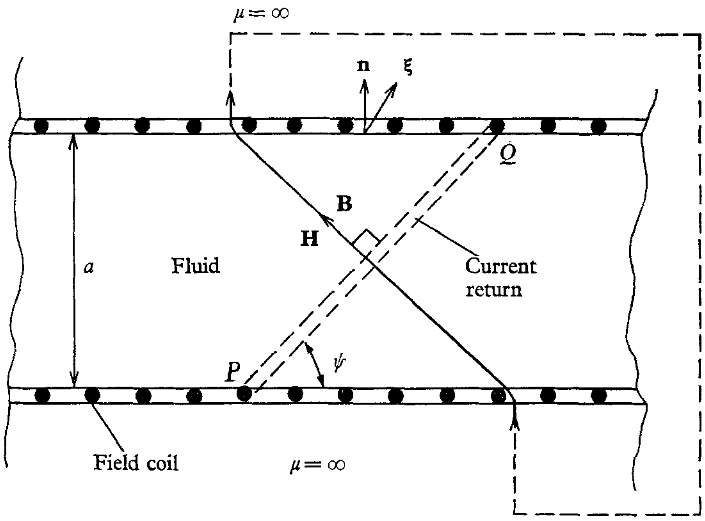

FIGURE 8

Suppose now that the fluid is deformed by giving one bounding plane an infinitesimal displacement $\boldsymbol{\xi}$, which is not necessarily parallel to the unit normal of the plane $\mathbf{n}$. The gap width a is increased by δa, where $\delta a = \boldsymbol{\xi}.\mathbf{n}$. During the deformation the temperature remains constant, and current to the field coils is adjusted so that the flux linked by the current does not change.† The latter condition implies that no electrical work is delivered, but from the surface stress σ_{ij} there is mechanical work per unit area

$$\sigma_{ij}\xi_j n_i = \delta(Fa). \qquad (\mathrm{A}\,2)$$

Using the fact that $\delta a/a = \delta v/v$, where v is the specific volume, and introducing (A 1), we obtain

$$\sigma_{ij}\xi_j n_i = \delta a\left\{F+v\left(\frac{\partial F_0}{\partial v}\right)_T-\int_0^H v\left(\frac{\partial B}{\partial v}\right)_{H,T} dH\right\}+aH\,\delta B. \qquad (\mathrm{A}\,3)$$

† The Landau & Lifschitz (1960) analysis is equivalent to taking a deformation at constant current.

The magnetic field decreases in proportion to the increase in distance between connected wires of the field coils. Since the direction of the line which measures this distance is parallel to $(\mathbf{B}\times\mathbf{n})\times\mathbf{B}$, we have

$$\delta B/B = -\{\mathbf{B}\times\mathbf{n}\}\times\mathbf{B}\}.\boldsymbol{\xi}/B^2a,$$

or

$$a\delta B = -B\boldsymbol{\xi}.\mathbf{n}+(\boldsymbol{\xi}.\mathbf{B})(\mathbf{B}.\mathbf{n})/B.$$

Substitution in (A 3) gives

$$\sigma_{ij}\xi_j n_i = \left\{\left(\frac{\partial(vF_0)}{\partial v}\right)_T - \int_0^H \left(\frac{\partial(vB)}{\partial v}\right)_{H,T} dH\right\}\boldsymbol{\xi}.\mathbf{n}+(\boldsymbol{\xi}.\mathbf{B})(\mathbf{H}.\mathbf{n}), \quad \text{(A 4)}$$

which is easily shown to be equivalent to the expression derived by Landau & Lifschitz (1960), and will be true for all $\boldsymbol{\xi}$ if

$$\sigma_{ij} = \left\{\left(\frac{\partial(vF_0)}{\partial v}\right)_T - \int_0^H \left(\frac{\partial(vB)}{\partial v}\right)_{H,T} dH\right\}\delta_{ij}+H_iB_j. \quad \text{(A 5)}$$

Finally we use a well-known thermodynamic relation which shows the first term in the braces to be $-p_0$, where p_0 is the pressure at the same density and temperature, but for zero H, and introduce the magnetization

$$\sigma_{ij} = -\left\{p_0(\rho,T)+\int_0^H \mu_0\left(\frac{\partial(vM)}{\partial v}\right)_{H,T} dH + \tfrac{1}{2}\mu_0H^2\right\}\delta_{ij}+H_iB_j, \quad \text{(A 6)}$$

for which we emphasize that the integration is to be carried out at constant density and temperature.

If we combine the integral in (A 6) with p_0 and define the sum as an effective pressure p^*, we obtain equation (14). However, it is worth noting that vM is the magnetization per unit mass, which may not vary significantly with density in the colloids, since the bulk modulus of the parent fluid, kerosene, is less than that of magnetite by a factor of more than 100. The compressibility of the kerosene may be the dominant effect in changes of volume, while the evidence of §3.1 suggests that the magnetizing effect of the solid particles may depend more on the mass present than the spacing. Hence there is the possibility, which needs further investigation, that the integral can be taken as zero to a first approximatio n.

Appendix B. Circulation and compressibility

In this appendix we seek to clarify the assumption of incompressibility in ferromagnetic fluids. The change in circulation Γ on a closed fluid loop is given by

$$\frac{D\Gamma}{Dt} = \frac{D}{Dt}\oint \mathbf{u}.d\mathbf{l} = \oint\left\{-\frac{\operatorname{grad} p^*}{\rho}+\mathbf{g}+\frac{\mu_0 M \operatorname{grad} H}{\rho}\right\}.d\mathbf{l}, \quad \text{(B 1)}$$

where we have neglected the effect of viscosity and free currents, and we have introduced the force expression (15). The analysis of appendix A gives

$$p^* = p_0+\int_0^H \mu_0\left(\frac{\partial(vM)}{\partial v}\right)_{H,T} dH.$$

If the magnetization per unit mass $vM = M/\rho$ were to be a function of H and T only, both the first and last terms in the braces of (B 1) can be reduced to pure gradients under isothermal conditions. In general we have

$$-\frac{\operatorname{grad} p^*}{\rho}+\frac{\mu_0 M \operatorname{grad} H}{\rho}$$
$$= -v \operatorname{grad} p_0 - \mu_0 v \frac{\partial(vM)}{\partial v} \operatorname{grad} H - \left(\int_0^H \mu_0 v \frac{\partial}{\partial v}\left(\frac{\partial(vM)}{\partial v}\right) dH\right) \operatorname{grad} v$$
$$- \left(\int_0^H \mu_0 \frac{\partial}{\partial T}\left(v \frac{\partial(vM)}{\partial v}\right) dH\right) \operatorname{grad} T + \mu_0 vM \operatorname{grad} H,$$

which after some manipulation gives

$$-\frac{\operatorname{grad} p^*}{\rho}+\frac{\mu_0 M \operatorname{grad} H}{\rho}$$
$$= \frac{\operatorname{grad} p_0}{\rho} - \operatorname{grad}\left(\int_0^H \mu_0 v^2 \left(\frac{\partial M}{\partial v}\right)_{H,T} dH\right) - \left(\int_0^H \mu_0 \left(\frac{\partial(vM)}{\partial T}\right)_{H,v} dH\right) \operatorname{grad} T. \tag{B2}$$

Substitution from (B 2) in (B 1) shows that the change in circulation is precisely zero in the absence of temperature gradients, since p_0 is a function of density only at constant temperature (and fortunately since it is not too difficult to imagine a scheme which directly contravenes the second law of thermodynamics without this condition on circulation).

It is now a trivial matter to justify the elementary formulation given in §2.2. We can assume a stationary isothermal fluid irrespective of compressibility effects. The problem only requires an adequate approximation to the static force balance. We have

$$dp^* + \rho g\, dz + \mu_0 M\, dH = 0, \tag{B3}$$

and here z is assumed to be directed upwards. We write $\rho = \tilde{\rho} + \delta\rho$, where $\tilde{\rho}$ is a standard density, define $\tilde{M} = M(\tilde{\rho}, H, T)$, and substitute in (B 3) to give

$$dp^* + \tilde{\rho} g\, dz + \mu_0 \tilde{M}\, dH = -\frac{\delta\rho}{\tilde{\rho}}\left\{\tilde{\rho} g\, dz + \mu_0 \left(\frac{\partial M}{\partial \rho}\right)_{H,T} \tilde{\rho}\, dH\right\}$$
$$= -\frac{\delta\rho}{\rho}\{\tilde{\rho} g\, dz + O(\mu_0 \tilde{M}\, dH)\}. \tag{B4}$$

The terms on the right-hand side are clearly negligible in comparison to the corresponding ones on the left for a liquid. Since $\tilde{M}(H)$ is usually only known to the same order of approximation, no special condition on the density need be applied in interpretation of the magnetization curves. We therefore obtain equation (16) of §2.2.

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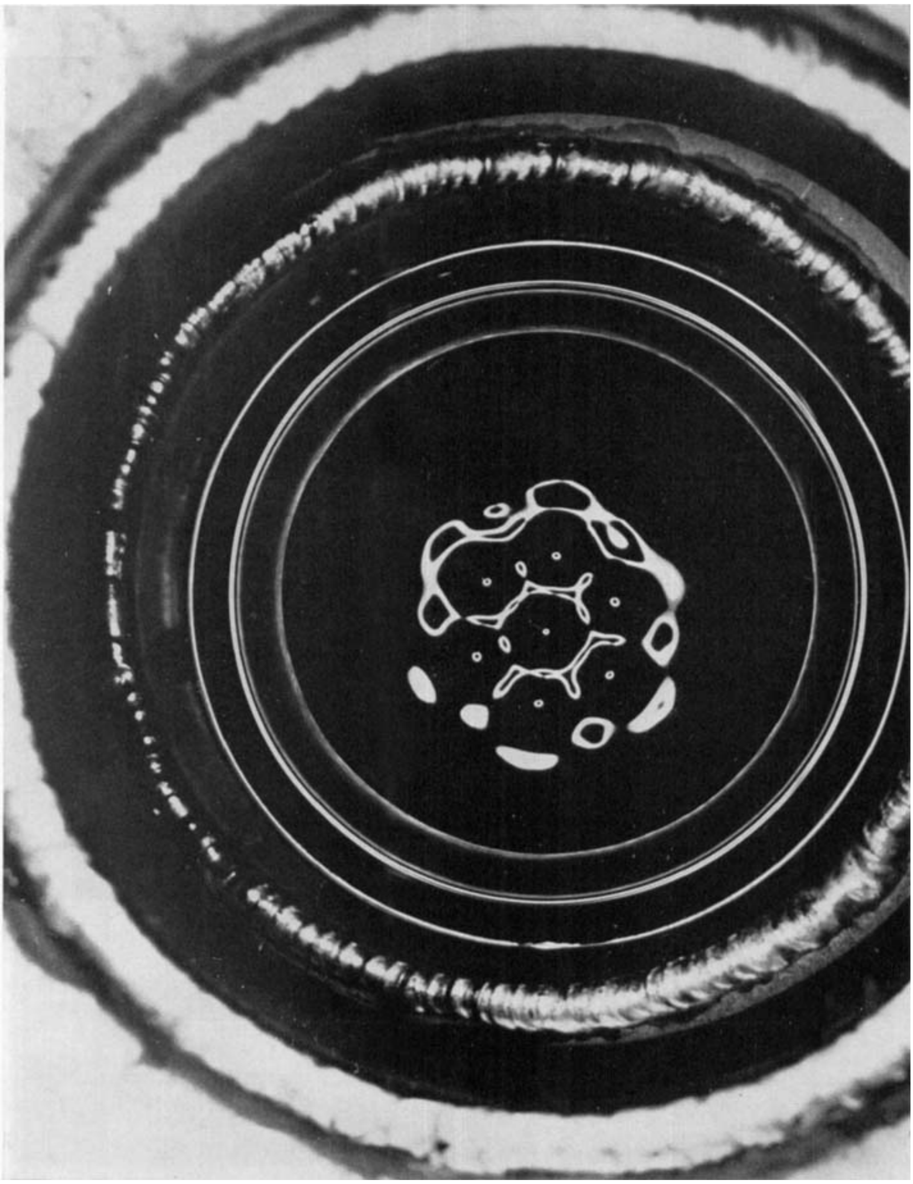

FIGURE 4. Interface between ferromagnetic fluid and air for a magnetization approximately 0·8 % above critical with $\rho/\rho_0 = 1{\cdot}388$. Exposure time 1 msec using a ring source of flash illumination concentric with the lens. Highlights indicate where the surface was flat enough for direct reflexion of the light.

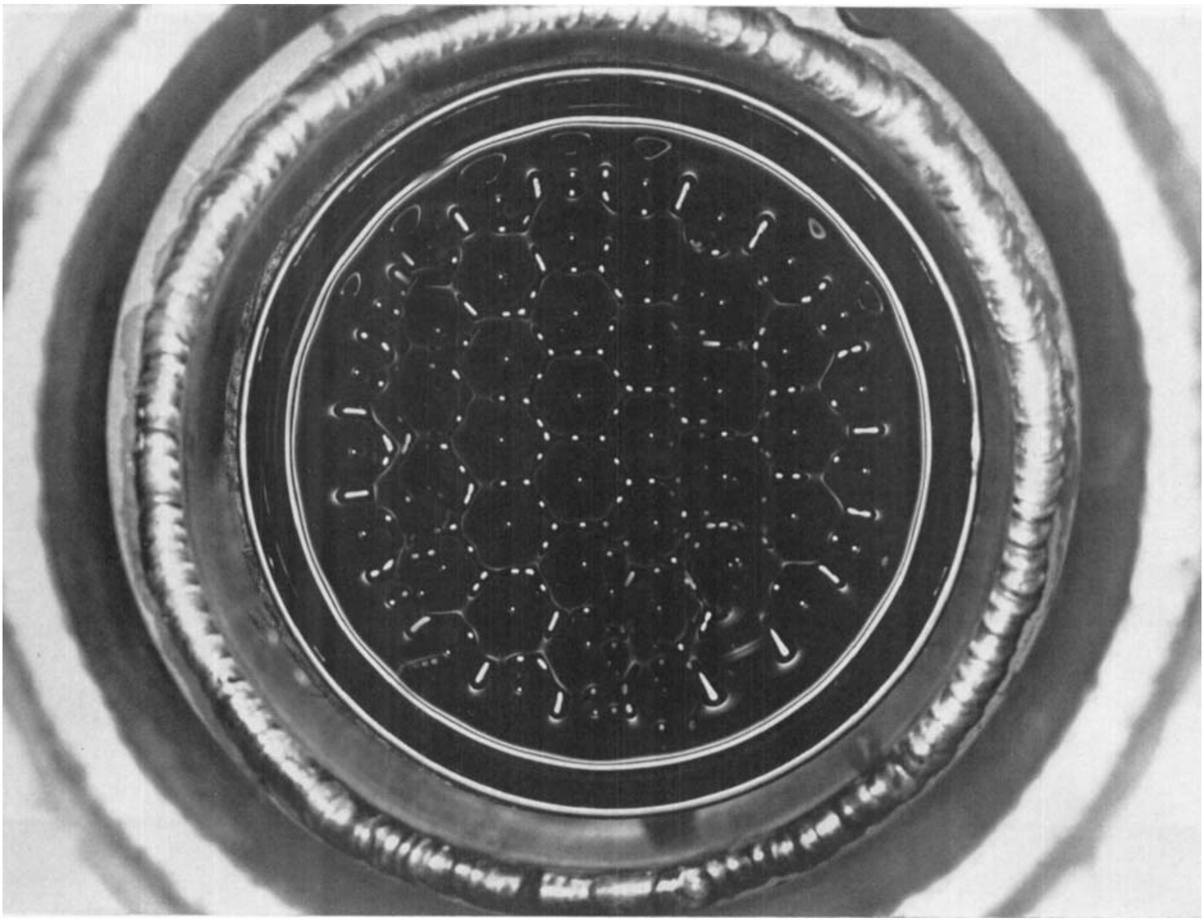

FIGURE 5. Interface between ferromagnetic fluid and air for a magnetization approximately 3 % above critical with $\rho/\rho_0 = 1{\cdot}388$. Photography as in figure 4. The more isolated highlights represent peaks.