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MAGNETIC INSTABILITY BETWEEN MISCIBLE FLUIDS IN A HELE–SHAW CELL

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We show experimentally the existence of a magnetic instability between two miscible fluids. The sample is an aqueous ferrofluid. At time $t = 0$, the ferrofluid and water are put into contact in a Hele–Shaw cell and then submitted to a homogeneous magnetic field perpendicular to the cell. Above a threshold value of the magnetic field a fingering instability is observed at the diffuse interface, with fingers growing with time. A wavelength can be defined as the mean distance between fingers. This wavelength in the linear regime is approximatively equal to the thickness of the cell.

1. Introduction. The interfacial phenomena between non-miscible magnetic fluids have been extensively studied and, in particular, the well-known "labyrinthine instability" [1–5]: it occurs when a magnetic fluid and an immiscible non-magnetic fluid are confined in a Hele–Shaw cell and subjected to a perpendicular magnetic field. This instability is governed by the magnetic Bond number, which is the ratio of the magnetic energy divided by the interfacial energy. Above a critical value of the magnetic Bond number, a fingering instability appears and the characteristic size of the pattern is limited to the thickness of the Hele–Shaw plate.

The miscible case is also of great interest, for example, in the field of biological and biomedical applications of ferrofluids [6, 7], where the mixing process of the ferrofluid and another biological constituent is crucial. The fingering instability at the diffuse interface under the effect of a magnetic field would indeed increase the total length of the interface and, therefore, speed up the mixing process. That is one reason why the evolution under the magnetic field at the frontier between a magnetic fluid and a miscible one is of great interest.

Moreover, the evolution of the frontier between two miscible fluids is of importance in fluid dynamics research. The instability induced by a difference in viscosity or by a difference in density has been extensively studied [9–15]. In the latter case, an interface separates a lighter fluid from a heavier one located above it in a gravitationnal field. This instability is driven by a buoyancy force, which is proportionnal to the difference in density of the two miscible liquids.

In the case of an interface between a magnetic liquid and a miscible non-magnetic fluid, the fingering instability induced by a magnetic field had been observed [16] and then studied numerically and theoretically [17, 18]. Due to the dipolar interactions between the nanoparticles, a demagnetizing field appears in the volume of the magnetic fluid, leading to a magnetic field larger outside the magnetic fluid than inside. Consequently, a gradient of magnetic field appears at the frontier: this gradient is at the origin of the destabilizing magnetic force.

In the following, we present experiments with a magnetic fluid and a miscible non-magnetic one in a Hele–Shaw cell in order to study this instability as a function of the two important following parameters: the cell thickness and the amplitude of the external magnetic field. We first describe the experimental setup. The experimental results are presented in the next section. Finally, we discuss the results and compare them to theoretical and numerical predictions.

2. Experimental system.

2.1. Sample. Experiments are performed with a magnetic fluid called a ferrofluid, which is chemically synthesized by the Massart's method [19]. It is an aqueous solution of cobalt ferrite nanoparticles. The nanoparticles of diameter about 10 nm are magnetic monodomains of magnetic moment $\mu = 10^4 \mu_{\mathrm{B}}$ (where μ_{B} is the Bohr magneton). The nanoparticles are in suspension in water at a volumic fraction about 10%. The stability of the solution is ensured through the adsorption of negatively charged citrate ions on the nanoparticles surface at a pH around 7.

It has been experimentally verified that in the range of the values of the magnetic field used here the ferrofluid follows a Langevin paramagnetism law. Up to $H \approx 3000\,\mathrm{A/m}$, the magnetization of the ferrofluid is proportionnal to the applied field $M = \chi H$ with the magnetic susceptibility $\chi \approx 2.3$.

2.2. Setup. The Hele–Shaw cell is composed of two microscope glass slides. They are spaced using mylar sheets of thickness between 12 and 250 microns. The cell is placed horizontally in the middle of an electric coil, which produces a homogeneous vertical magnetic field (perpendicular to the cell). Half of the cell is filled with a ferrofluid. At time t equals to zero the water is injected into the other half of the cell, and then the magnetic field is switched on. The experiments are recorded by a video camera.

3. Experimental results.

3.1. Diffusive law without magnetic field. If the magnetic field is equal to zero, the frontier between the ferrofluid and water evolves following a diffusive law. In order to measure the diffusion coefficient D between the ferrofluid and the water, we measured the intensity profile I in the x-direction normal to the diffuse interface. This intensity profile may be related to the ferrofluid concentration c thanks to the Beer–Lambert law $I(x) = I_0 \exp(-\kappa c(x))$, where the coefficient κ is proportional to the light absorbance of the suspension and to the thickness of the sample. Afterwards, the ferrofluid concentration may be fitted by an error function $c(x) = c_0 \mathrm{erf}(x/\delta)$, thus defining a length δ characteristic of the diffusion front, which evolves in time as $\delta = 2\sqrt{Dt}$. The evolution of δ with time allows to get an estimation of the diffusion coefficient D. We found $D = 6.10^{-10}\,\mathrm{m^2/s}$. This value is about ten times larger than the one predicted by the generalized Stokes–Einstein formula for the diffusion coefficient of hard spheres. This seems to indicate an important contribution of the repulsive interaction between the nanoparticles. Such an effect has already been observed in previous studies on ferrofluid (see [8]).

3.2. Development of fingers above a critical value of the magnetic field. The fingering instability of the diffuse interface is observed above a critical value of the magnetic field about $1.5 \pm 0.2\,\mathrm{mT}$. This destabilization appears less than one second after the field is switched on: fingers are clearly distinguished and then grow with time (see Fig. 1). The critical value of the magnetic field depends on different parameters like the initial conditions and the thickness of the cell.

3.3. Evolution of the fingers with time. The evolution with time of the fingers strongly depends on the value of the magnetic field. At a low field, the

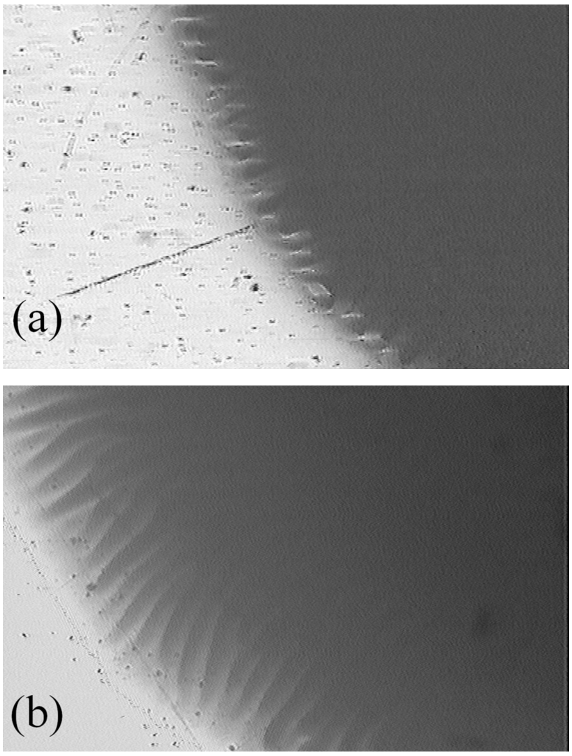

Fig. 1. Fingers are visible at the frontier between the ferrofluid (in grey on the photograph) and water (in white). The horizontal length of each photograph is 3 mm. In this experiment, the Hele–Shaw cell has a thickness of 100 microns. The magnetic field $B = 2.0\,\mathrm{mT}$ is perpendicular to the plane of the photographs. The first photograph (a) was recorded at $t = 4\,\mathrm{s}$ and the second one (b) at $t = 322\,\mathrm{s}$.

fingers may become very long (see Fig. 2). Such very long fingers have been observed for different values of a thickness from 75 to 250 microns. At higher values of the field, the fingers themselves are not stable and are always splitting (see Fig. 3). The minimum value of the magnetic field to enter this second regime is $2.4 \pm 0.2\,\mathrm{mT}$.

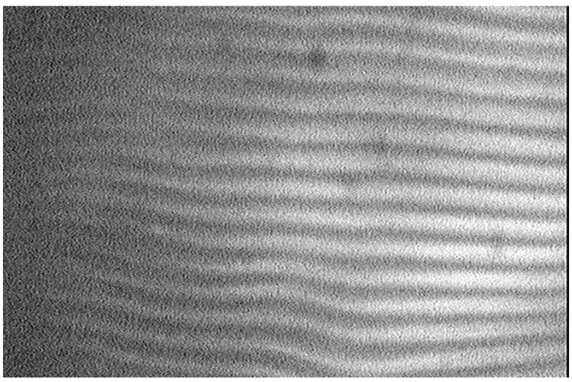

Fig. 2. Very long fingers under the field $B = 2.0\,\mathrm{mT}$. The cell is 75 microns thick and the picture was recorded at $t = 1099\,\mathrm{s}$. The picture is 3 mm long.

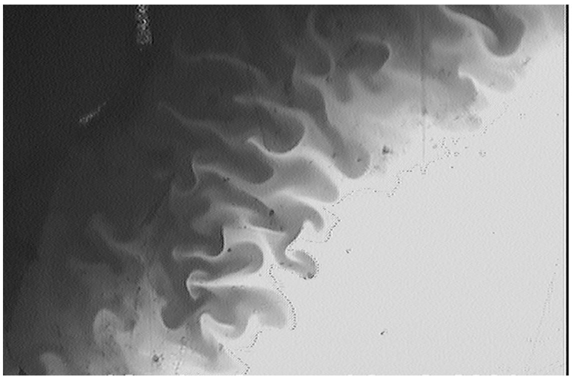

Fig. 3. Splitting fingers under the field $B = 3.5\,\mathrm{mT}$. The cell is 100 microns thick and the photograph was taken at time $t = 286\,\mathrm{s}$. The picture is 3 mm long.

In both cases when the field is switched off, the fingers disappear in a few seconds under the effects of diffusion. This is consistent with the measured value of the diffusion coefficient mentioned above: the characteristic time to delete such structures of characteristic size of 100 microns is found about $\lambda^2/D \approx (10^{-4})^2/6.10^{-10} \approx 17\,\mathrm{s}$.

3.4. Characterization of the wavelength. In order to characterize more quantitatively the fingering instability, we have defined the wavelength λ as the characteristic length between two fingers. As it can be seen in Fig. 4, the distance between the fingers is very dispersed, and we can only get a mean value for the characteristic length.

1. Wavelength as a function of time.
   The evolution with time of the wavelength depends on the thickness of the cell. At a low thickness, the wavelength always increases with time. Above a thickness of 70 microns, the wavelength is very dispersed around a mean value, which stabilizes in time. In the following, we define the wavelength in the "stationary regime" as the mean value for the long times. The wavelength "in the linear regime" is the first measurable distance between two fingers after the magnetic field is switched on (it was always measured less than two seconds after the magnetic field was switched on).

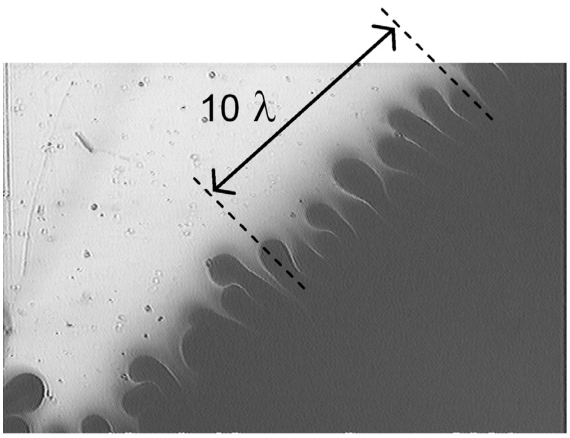

Fig. 4. The wavelength λ is defined at each time by the mean distance between fingers.

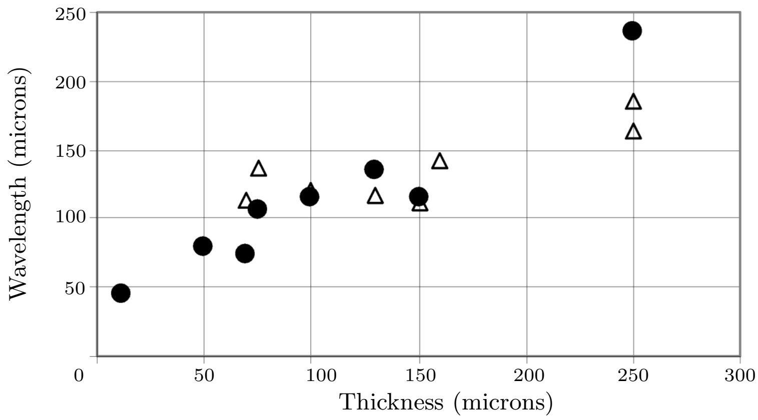

Fig. 5. The wavelength (● linear regime, △ stationary regime) as a function of the cell thickness. The magnetic field is $B = 2.0\,\text{mT}$.

2. Dependence on the thickness.
   We have studied the influence of the cell thickness on the wavelength. In Fig. 5 are plotted both the values of the wavelength in the linear regime and in the stationary one. As mentioned above, there is no stationary value below a thickness of 70 microns. Above it, the stationary wavelength seems to slightly increase when the thickness of the cell increases.

   For the linear regime, it is possible to get values from a thickness of 12 microns, and we observe a quasi-linear increase of the wavelength with the thickness. The linear wavelength is actually of the order of the thickness of the cell for the values we studied from 12 to 250 microns.

3. Dependence on the magnetic field.
   In Fig. 6, the linear wavelength and the stationary one are plotted as a function of the magnitude of the magnetic field for a cell thickness of 100 microns. The values are dispersed and it is difficult to draw any clear dependence of the wavelength as a function of the magnetic field. However, the linear wavelength is very dispersed around a mean value, which is almost constant in this range and equal to the thickness of the cell.

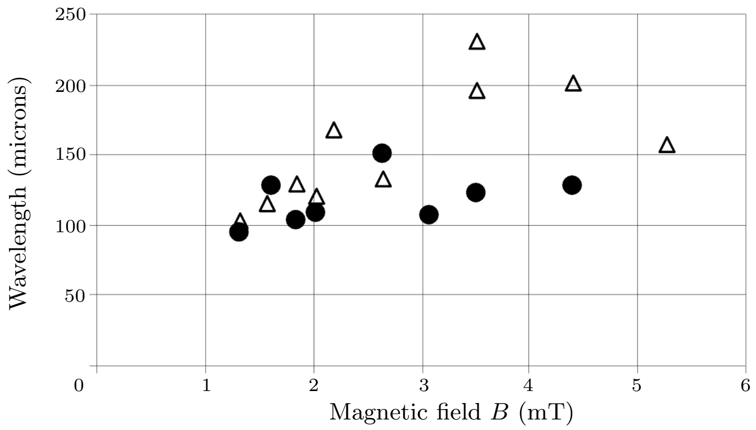

Fig. 6. The wavelength (● linear regime, △ stationary regime) as a function of the magnitude of the magnetic field. The cell is $100\,\mu$m thick.

4. Discussion. The fingering instability appears because of the existence of the demagnetizing field, which is due to the dipolar interactions between nanoparticles in the volume of the magnetic fluid. This demagnetizing field is responsible for the appearance of a gradient of magnetic field directed towards the non-magnetic fluid. Thus, a perturbation of the interface becomes unstable for a large enough value of the applied field. This perturbation grows into fingers, each finger acting as a magnet repelling its neighbours.

But in the case of miscible fluids, the perturbation is smeared by the diffusive effects. We define the following dimensionless parameter Cm as the ratio of the characteristic time $\tau_\mathrm{D} = h^2/D$ for the diffusion to act over a distance h (D is the diffusion coefficient of the magnetic nanoparticles) and of the characteristic time $\tau_\mathrm{m} = 12\eta/\mu_0 M^2$ of the magneto-convection (with η being the fluid viscosity, M its magnetization and μ_0 the permittivity of vacuum):

$$\mathrm{Cm} = \frac{\mu_0 M^2 h^2}{12\eta D}. \tag{1}$$

Note that this parameter is equivalent to the Peclet number $\mathrm{Pe} = uh/D$, where uh is associated with the convective transport, u being here the velocity due to the local gradient of the magnetic field.

The linear instability has been theoretically investigated in [18] by using the Brinkmann (Darcy–Stokes) equation. The stability diagram is reproduced in Fig. 7. The "marginal curve" separating the stable domain from the unstable one is plotted using a full line. The curve standing for the most unstable wavenumber for a given value of the parameter Cm is represented by a dashed line.

Due to the diffusion effects, the instability is time dependent. This implies that the marginal curve depends on a "waiting time" τ_w elapsed between the injection of water (formation of the diffuse interface) and the application of the magnetic field. As an example, in the same Fig. 7 the marginal curve and the curve of the most unstable wavenumber are plotted for $\tau_\mathrm{w} = 10^{-2}\tau_\mathrm{D}$, with τ_D being the characteristic time of diffusion. As it can be seen, there is a significant variation of the marginal curve (there is a slight increase of the critical value of Cm for the appearance of the instability, and a net increase of the corresponding wavelength).

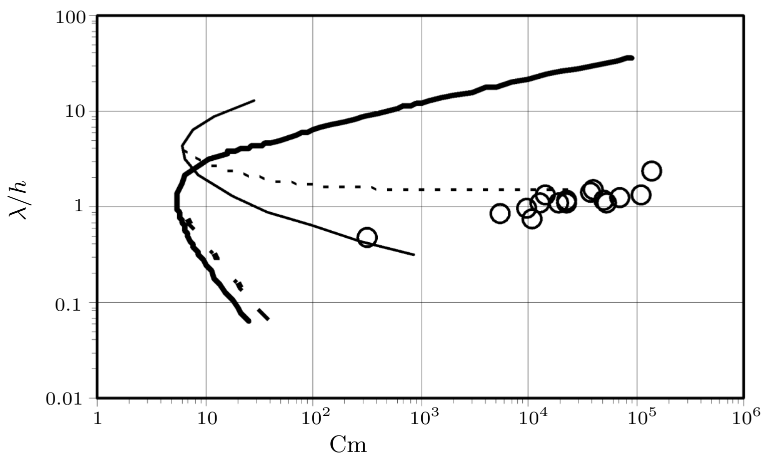

Fig. 7. The wavelength λ divided by the thickness h as a function of the dimensionless parameter Cm. Open circles – experimental points; full and dashed lines – theoretical predictions [18], respectively, for the marginal curve and for the most unstable wavenumber. Both are represented for $\tau_\mathrm{w} = 0$ (thick curves), and $\tau_\mathrm{w} = 10^{-2}\tau_\mathrm{D}$ (thin curves).

In our experiments, the value of the dimensionless parameter Cm can be estimated. As $D = 6.10^{-10}\,\mathrm{m}^2/\mathrm{s}$ and $M = \chi H$ with $\chi \approx 2.3$, one gets:

$$\mathrm{Cm} = 9.2 \cdot 10^5 h^2 H^2. \tag{2}$$

The experimental points are plotted in Fig. 7 for different values of the magnetic field and cell thickness.

The experimental points are consistent with the theoretical curves as they are well located in the unstable domain defined by the marginal curve. But we expect to measure experimentally the "most unstable wavelength", which would correspond to the theoretical dashed line. Concerning the time-dependence, the waiting time τ_{w} could not be accurately established experimentally, but it can be estimated to $\tau_{\mathrm{w}} \approx 0.5s$. The experimental value of τ_{D} is about $17s$ for a cell $100\mu m$ thick, which leads to $\tau_{\mathrm{w}}/\tau_{\mathrm{D}} \approx 3.10^{-2}$. As it can be seen in Fig. 7, the experimental points are rather well corresponding to the theoretical predictions for $\tau_{\mathrm{w}}/\tau_{\mathrm{D}} = 10^{-2}$. However, the experimental points correspond to different values of the cell thickness. Since $\tau_{\mathrm{D}} \propto h^2$, the experimental points should be compared to curves corresponding to different τ_{w}. At this stage, it is, therefore, difficult to compare the experimental data to the theory. Nevertheless, it appears that the waiting time τ_{w} plays an important role and may be carefully controlled in future experiments.

5. Conclusion. We have presented the first experimental study of the fingering instability appearing at the diffuse interface between a magnetic liquid and a miscible non-magnetic one under a perpendicular magnetic field, following the observation of [16].

Our results may be compared to the experiments [12] of the buoyancy driven instability for miscible fluids. In this case, the authors use the dimensionless Rayleigh number, which stands for the ratio of buoyancy-convective and diffusive transport ($\mathrm{Ra} = \Delta\rho g h^3/(12\eta D)$). At low $\mathrm{Ra} \leq 10$, they observe that λ/h scales as Ra^{-1}. For $\mathrm{Ra} \geq 100$ the wavelength reaches a constant value, roughly equal to five times the thickness of the cell. This is consistent with our results for magnetic instability, as our experiments were carried on for the analogous parameter Cm in the 300–200000 range and displayed $\lambda \approx h$.

Our results are consistent with the theoretical predictions for magnetic miscible fluids [18] as soon as we take into account the waiting time. But with the setup used here, it is very difficult to impose well defined initial conditions. Another geometry is needed, and we think that a microfluidic setup would be of great advantage: at this small scale the flow is laminar and in the absence of magnetic field the diffusion front between the ferrofluid and water is essentially controlled by the flow rate. Hence, there is an equivalence between the state of the experimental system in a Hele–Shaw cell at time t in the experiments described here and in a microchannel at a distance $d = ut$ (u is the flow velocity) from the junction, where the ferrofluid and water meet. For future experiments, this would allow a better control of the initial conditions before the magnetic field is switched on, and provide a very useful tool to go on with the study of such instability.

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