## Roll-rectangle transition in the magnetic fluid Faraday instability

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At the surface of a magnetic fluid, parametric waves can be excited by an alternating magnetic field, parallel to the surface. With this anisotropic system, we observe a pattern transition from parallel rolls, perpendicular to the magnetic field, to a rectangular array of cross rolls. The neutral curve of the transition is well explained by a model of four coupled Landau-Ginzburg equations.

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Patterns have been and are strongly investigated in systems involving spatiotemporal disorder. The most wellknown example [1,2] is that of Rayleigh-Benard instability with a Busse balloon presentation of patterns in different regimes. If we believe in the properties of universality of the models, the description has to be independent of the microscopic level and should also apply to other systems. Another example frequently encountered in spatiotemporal chaos and turbulent structure studies is Faraday instability, which leads to capillary waves excited parametrically [3-7]. In a magnetic fluid (MF), Faraday instability can be excited by a horizontal and alternating magnetic field [8-10]. The system is thus anisotropic. As a function of the amplitude of the external magnetic field, magnetocapillary waves develop a rich series of patterns such as [1,2] rolls, rectangles, zig zag, oscillations, defects, etc. In this paper, we focus ourselves on the roll-rectangle transition, which is the equivalent of a "cross-roll" transition in the Rayleigh-Bénard experiment.

During the last decade, Faraday ripples on a free fluid surface in a vertically oscillating vessel have been intensively studied [3-7]. Strong magnetic properties of some magnetic fluids allow capillary waves to be simply excited by an external ac magnetic field [8]. In the case of an infinite depth of MF, the dispersion relationship of capillary-gravitational waves in a tangential magnetic fields is [11,12]

$$\omega^{2}(\mathbf{k}) = \frac{\sigma k^{3}}{\rho} + gk + \frac{\mu_{0}(\mu_{r} - 1)^{2} H^{2} k_{x}^{2}}{\rho(\mu_{r} + 1)} , \qquad (1)$$

 $\mu_r$  is the relative magnetic permeability of MF, and  $k_x$  is the wave vector component along the magnetic field. The "elasticity" of the surface in the ac field  $H = H_0 \cos(\omega t)$  is modulated and capillary waves can be excited in a parametric way [8]. The principal feature of this system is the anisotropic character of the interaction between the MF

and the tangential magnetic field. It is possible to excite one-dimensional surface patterns; the dispersion relationship for waves propagating transversely to the magnetic field is that of an ordinary liquid, unmodified by the field. Increasing supercriticality of the magnetic excitation, transversal modulations [4,5,7], and oblique and "chevron" patterns with "ziplike" defects [8,13-15] are observed. But in the vicinity of the roll threshold (Faraday instability), a roll-rectangle transition is always observed. The description of patterns arising by bifurcations and their various transformations in a weak nonlinear regime is associated with the formalism of the Ginzburg-Landau equations for the order parameters. In our case of parametric oscillations of the free surface under a tangential ac magnetic field, the order parameters of the excited pattern are the complex amplitudes of the surface waves traveling in opposite directions. They are coupled because of the ac character of the magnetic field; this coupling is responsible for the excitation of surface waves. If in the dispersion relationship the magnetic term is small with respect to the surface one,  $([\mu_0(\mu_r-1)^2/(\mu_r)]^2$  $+1)H^2 \ll \sigma k$ , the coupled system of equations for the amplitudes of the traveling MF surface waves [6,8,9] is, in a homogeneous situation,

$$\frac{\partial a_{\pm}}{\partial t} \pm i \Delta \omega a_{\pm} \pm 2i G a_{\pm} + \gamma a_{\pm}$$

$$= \mp i G a_{\pm} \pm i a_{+} (T|a_{+}|^{2} + S|a_{\mp}|^{2}), \quad (2)$$

the elevation of the free surface being

$$\xi(x,y,t) = \frac{1}{2}a_{+} \exp[i(kx - \omega t)] + \frac{1}{2}a_{-} \exp[i(kx + \omega t)] + \text{c.c.}$$

The dissipation coefficient is  $\gamma = 2\nu k^2$  ( $\nu$  is the kinetic viscosity of the fluid); the magnetic interaction parameter is  $G = [\mu_0(\mu_r - 1)^2 H_0^2 k^2]/[8\rho(\mu_r + 1)\omega]$ ; the interaction coefficients T, S are respectively equal to  $T = \frac{1}{16}\omega_0 k^2$  and  $S = \omega_0 k^2$ , with  $\omega_0(k)$  determined by the dispersion relationship of capillary waves in zero field; and  $\Delta\omega$  is the detuning term  $\Delta\omega = \omega_0(k) - \omega$  of the capillary wave in the ac field.

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The increment of growth of perturbations from the quiescent state  $(a_{\pm}=0)$  is deduced from Eq. (2):

$$\lambda = -\gamma \pm [G^2 - (\Delta\omega + 2G)^2]^{1/2}$$
.

As a result, at the threshold, the magnetic interaction parameter G is equal to

$$G^* = \gamma$$
,

with a detuning of the capillary waves  $\Delta \omega^* = -2G^*$ . This nonzero detuning at the threshold comes from the magnetic field dependence of the dispersion relationship of the magneto-capillary waves [Eq. (1)]. The neutral curve of Faraday instability is given by  $\lambda = 0$ , and we ob-

$$\frac{\Delta\omega}{G^*} = -\frac{2G}{G^*} \pm \left[ \left( \frac{G}{G^*} \right)^2 - 1 \right]^{1/2}. \tag{3}$$

For supercritical conditions  $(G > G^*)$ , traveling waves arise and are synchronized  $(a_{+} = a_{-}^{*})$ . According to (2), their amplitude A and temporal phase  $\varphi$  are determined by the following relations  $[a_{+} = A \exp(i\varphi), a_{-}]$  $= A \exp(-i\varphi)$ :

$$(T+S)A^2 = \Delta\omega + 2G + (G^2 - G^{*2})^{1/2}$$
,  
 $\sin(2\varphi) = -G^*/G$ . (4)

The arising pattern has the form of standing waves,

$$\xi(x, y, t) = 2A \cos(kx)\cos(\omega t - \varphi)$$
,

with the crests transverse to the exciting ac field.

Because of some additional nonlinear interaction terms in Eq. (2), which are small and negligible in the previous description, oscillations of the free surface induce some extra capillary waves, propagating transversely to the direction of the exciting magnetic field. The coupling arises from terms proportional to  $a_+a_-^*$  and  $a_+^*a_-$ . Standing cross waves can be parametrically excited, leading to rectangle patterns. The exact value of the amplitude of free surface oscillations A and the threshold  $G^{**}$ of the magnetic interaction parameter G can be deduced from the Ginzburg-Landau equations for the amplitudes of the cross waves  $b_{\pm}$ . We search the solution of the transverse problem, with a surface elevation of the form

$$\xi(x,y,t) = \frac{1}{2}b_{+} \exp[i(qy - \omega t)] + \frac{1}{2}b_{-} \exp[i(qy + \omega t)] + \text{c.c.},$$

with q the wave vector for the cross waves. As in the previous problem, we can derive a new system of Landau-Ginzburg (LG) equations for these cross waves:

$$\begin{split} \frac{\partial b_{+}}{\partial t} + i(\Delta\omega + \Delta\omega')b_{+} + \gamma b_{+} &= i\frac{3}{4}\omega_{0}(q)q^{2}b_{-}a_{+}a_{-}^{*} + \frac{i\omega_{0}(q)q^{2}(3 - 2\sqrt{2})b_{+}(|a_{+}|2_{+}|a_{-}|^{2})}{8(2 - \sqrt{2})} \;, \\ \frac{\partial b_{-}}{\partial t} - i(\Delta\omega + \Delta\omega')b_{-} + \gamma b_{-} &= -i\frac{3}{4}\omega_{0}(q)q^{2}b_{+}a_{-}a_{+}^{*} - \frac{i\omega_{0}(q)q^{2}(3 - 2\sqrt{2})b_{-}(|a_{+}|^{2} + |a_{-}|^{2})}{8(2 - \sqrt{2})} \end{split}$$

 $\Delta\omega'$  is the detuning between waves  $a_{\pm}$  and  $b_{\pm}$ ,

$$\Delta\omega' = \omega_0(q) - \omega_0(k)$$
.

We are interested in the smallest value  $A_c$  of the amplitude A of a "roll" pattern when the "cross" waves arise. The increment of growth  $(\lambda_1)$  of the amplitudes of cross waves is given by the LG  $[b_+ \sim \exp(\lambda_1 t)],$ 

$$\lambda_1 = -\gamma \pm \sqrt{|G_1|^2 - (T_1 - \Delta \widetilde{\omega})^2}$$
,

$$G_1 = \frac{3}{4}\omega_0(q)q^2A^2\exp(2i\varphi) = \delta A^2\exp(2i\varphi)$$

with 
$$\delta = \frac{3}{4}\omega_0(q)q^2$$
,

$$T_1 = \frac{\omega_0(q)q^2(3-2\sqrt{2})A^2}{4(2-\sqrt{2})} = \varepsilon A^2$$

with 
$$\varepsilon = \frac{\omega_0(q)q^2(3-2\sqrt{2})}{4(2-\sqrt{2})}$$

and  $\Delta \tilde{\omega} = \Delta \omega + \Delta \omega'$ . The marginal curve for the rollrectangle transition is given by the condition  $\lambda_1 = 0$ . It is dependent on the roll amplitude A and leads to a condition for the critical value of the amplitude of the roll

$$A_c^2 = \frac{-\Delta \widetilde{\omega} \varepsilon + \sqrt{(\Delta \widetilde{\omega}^2 v^2 + (\delta^2 - \varepsilon^2) \gamma^2)}}{(\delta^2 - \varepsilon^2)} .$$

The minimum of  $A_c$  as a function of the detuning  $\Delta \tilde{\omega}$ gives another relation:

$$\frac{dA_c^2}{d\Delta\widetilde{\omega}} = 0 \ . \tag{5}$$

The minimal amplitude of rolls to observe the excitation of cross waves is then

$$A_c^2 = \frac{\gamma}{\delta}$$
,

leading to the neutral curve of the roll-rectangle transi-

with 
$$\varepsilon = \frac{\omega_0(q)q^2(3-2\sqrt{2})}{4(2-\sqrt{2})}$$
,  $\frac{\Delta\omega}{G^*} + \frac{2G}{G^*} + \left[\frac{G^2}{G^{*2}} - 1\right]^{1/2} = \frac{17\omega_0(k)k^2}{12\omega_0(q)q^2}$ . (6)

Assuming that, for these capillary waves, the gravity term is negligible in the zero-field dispersion relationship, the ratio k/q is equal to  $[\omega_0(k)/\omega_0(q)]^{2/3}$ . The neutral curve (6) becomes

$$\frac{\Delta\omega}{G^*} + \frac{2G}{G^*} + \left[\frac{G^2}{G^{*2}} - 1\right]^{1/2} = \frac{17\left[\frac{\omega}{G^*} + \frac{\Delta\omega}{G^*}\right]^{7/3}}{12\left[\frac{\omega}{G^*} + \frac{\varepsilon}{\delta}\right]^{7/3}}.$$
 (7)

The parameter  $\omega/G^*$ , which enters in Eq. (7), measures the ratio of the relaxation time  $\tau=\gamma^1$  of the surface elevation to the field period  $(T=2\pi/\omega)$ . It is large,  $\omega/G^*=2\pi\tau/T$ . In Fig. 1, we have plotted the neutral curve (curve 1) of the Faraday instability [Eq. (3)] and the neutral curve (curve 3) of the roll-rectangle transition [Eq. (7)] if  $\omega/G^* >> \Delta\omega/G^*$ ,  $\varepsilon/\delta$ . Curve 2 represents the dispersion equation of the magnetocapillary waves. The crossing of curves 1 and 2 is the Faraday threshold for the rolls  $(G=G^*)$ . The intersection between curves 2 and 3 characterizes the roll-rectangle transition  $G^{**}$ :  $G^{**}/G^*=1.73$ .

A magnetic fluid is a colloidal suspension of nanometric particles; the grains are made of Cobalt ferrite and behave as rigid dipoles [16]. We use a MF obtained through a chemical synthesis [17,18] without surfactant. The important feature is that the magnetization curve of the MF as a function of the applied field is that of a giant-paramagnetic material. The volume fraction of magnetic particles is 15%, and the initial magnetic susceptibility is equal to 3; the surface tension  $\sigma$  is  $48\text{mJ/m}^2$ ,  $\rho = 1850 \text{ kg/m}^3$ , and cinematic viscosity  $v=2\times10^{-6} \text{ m}^2/\text{s}$ .

In a previous paper [10], we have shown that the threshold of the onset of rolls is very sensitive to the geometry of the vessel containing the magnetic fluid. In this experiment, we have kept a constant depth (10 mm) for the fluid and used different box sizes. With a moving wall, the length L of the box (dimension along the field) is varied (experiment 1) from 20 to 100 mm with a constant width D=65 mm (dimension perpendicular to the field); or the width D is varied (experiment 2) from 5 to 65 mm with a constant length L=100 mm. Two coils in the

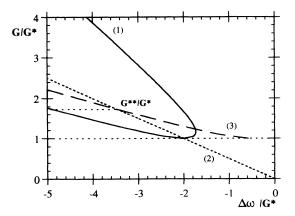


FIG. 1  $G/G^*$  versus  $\Delta\omega/G^*$ . Curve 1: neutral curve of Faraday instability. Curve 2: dispersion curve for the magnetocapillary waves; the experimental points are in the vicinity of this curve. Curve 3: neutral curve of rectangle instability.

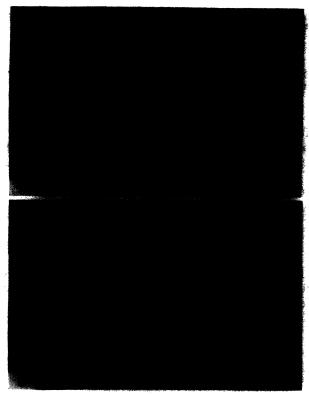
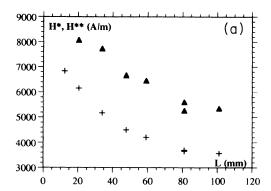


FIG. 2. (a) roll pattern  $H^* < H < H^{**}$ ; (b) rectangular pattern  $H > H^{**}$ .



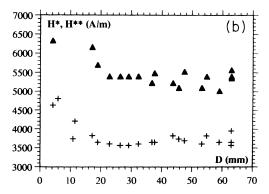


FIG. 3. (a) Roll threshold  $H^*$  (+) and roll-rectangle threshold  $H^{**}$  ( $\triangle$ ) versus the length L of the box (D=65 mm). (b) Roll threshold  $H^*$  (+) and roll-rectangle threshold  $H^{**}(\triangle)$  versus the width D of the box (L=100 mm).

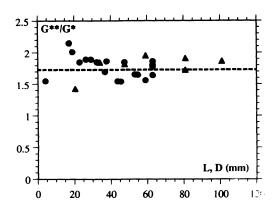


FIG. 4.  $G^{**}/G^*$  versus L (with D=65 mm) and versus D (with L=100 mm). The dashed line presents the theoretical value  $G^{**}/G^*=1.73$  for an infinite geometry.

Helmholtz position supply uniform horizontal magnetic field at 50 Hz. A video camera records the surface deformations. Figures 2(a) and 2(b) present the surface of the magnetic fluid for a roll pattern [1(a)] and for a rectangular pattern [1(b)]. Figures 3(a) and 3(b) present, respectively, the magnetic field thresholds (onset of rolls)  $H^*$ and roll-rectangle transition  $H^{**}$  as a function of L and D in experiments 1 and 2. Finite size effects [4,10] are clearly observed on both  $H^*$  and  $H^{**}$ . For a detailed study of this  $H^*$  dependence see Ref. [10]. In Fig. 4, the ratio  $G^{**}/G^{*}$  is presented as a function of the two variable dimensions of the vessel. In spite of the influence of the geometry on the thresholds  $H^*$  and  $H^{**}$ , the ratio  $G^{**}/G^*$  is almost constant and its mean value 1.8 is in accordance with the theoretical one, independent of the geometric constants in our model,  $G^{**}/G^{*}=1.73$ .

At the roll-rectangle transition, the ratio of the wavelength perpendicular to the magnetic field  $\lambda_{\perp}$  and the wavelength parallel to the magnetic field  $\lambda_{\parallel}$  is equal to the ratio k/q and given by

$$\lambda_{1}/\lambda_{\parallel} = \frac{(\omega/G^{*} - 2G^{**}/G^{*})^{2/3}}{(\omega/G^{*} + \varepsilon/\delta)^{2/3}} . \tag{8}$$

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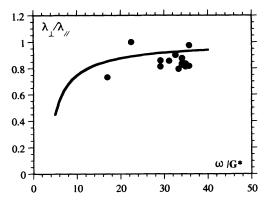


FIG. 5.  $\lambda_1/\lambda_{\parallel}$  versus  $\omega/G^*$ ; the experimental points correspond to different geometries of the box. The full line is the theoretical curve deduced from expression (8).

This expression reflects the dependence of the magnetocapillary waves on the angle between the magnetic field and the wave vector. Figure 5 presents the theoretical curve  $\lambda_{\perp}/\lambda_{\parallel}$  versus  $\omega/G^*$  with the theoretical value for  $G^{**}/G^*$ . In the same graph, the experimental data are presented, they are in good agreement with expression (8). This fact also supports our theoretical model.

A MF surface submitted to horizontal and alternating magnetic field is an anisotropic system that gives rise to a unidimensional parametric Faraday instability. For supercritical conditions, other transitions occur, leading to some symmetry breaks. The roll-rectangle transition studied here is the first one to arise. The present model, developed with a set of four coupled Ginzburg-Landau equations, perfectly accounts for the experimental data. With this MF system, the next steps of symmetry breaking will be tilts with drift, oscillations, chevrons, and zigzag patterns, etc.

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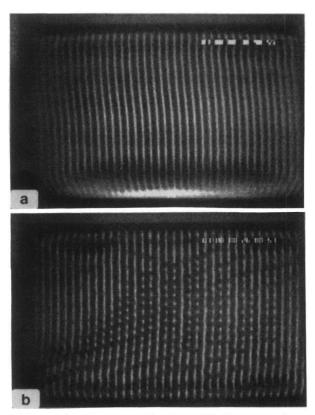


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