Double Rosensweig instability in a ferrofluid sandwich structure

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We consider a horizontal ferrofluid layer sandwiched between two layers of immiscible nonmagnetic fluids. In a sufficiently strong vertical magnetic field the flat interfaces between magnetic and nonmagnetic fluids become unstable to the formation of peaks. We theoretically investigate the interplay between these two instabilities for different combinations of the parameters of the fluids and analyze the evolving interfacial patterns. We also estimate the critical magnetic field strength at which thin layers disintegrate into an ordered array of individual drops.

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I. INTRODUCTION

Ferrofluids are colloidal suspensions of nanosize ferromagnetic grains in a carrier liquidlike water or oil [1]. The dipole-dipole interaction between the ferromagnetic particles is for moderate volume concentrations rather small and ferrofluids hence behave magnetically as superparamagnets. Accordingly in the absence of an external magnetic field the magnetization of the fluid is zero. If a field is switched on the magnetic moments of the particles orient themselves along the field direction giving rise to a macroscopic magnetization. The notion *superparamagnets* refers to the unusually high value of the magnetic susceptibility, $\chi = 1, \ldots, 50$, to be compared with $\chi \simeq 10^{-4}$ for atomic paramagnets. Hydrodynamically diluted ferrofluids behave like ordinary Newtonian liquids with additional contributions in the bulk and surface force densities stemming from the interaction with the magnetic field.

Due to the unique interplay between hydrodynamic and magnetic degrees of freedom ferrofluids show a variety of instabilities and pattern formation processes. Among the most striking phenomena in this respect is the so-called Rosensweig instability in which the flat free surface of a ferrofluid becomes unstable when subjected to a sufficiently strong vertical magnetic field [2]. Although both gravity and surface tension favor a flat surface the decrease in magnetic energy for a periodic array of peaks and troughs can be large enough to overcompensate the increase in potential and surface energy. Both the linear instability and the details of the pattern formation as revealed by a weakly nonlinear analysis have been thoroughly studied [2–7].

In the present paper we investigate a sandwich structure in which a ferrofluid layer of given thickness is placed between two immiscible nonmagnetic liquids. The system is prepared such that in the absence of a magnetic field the layering is stable, i.e., the lower layers have larger densities than the upper ones in order to prevent the Rayleigh-Taylor instability. Applying a homogeneous external magnetic field perpendicular to the undisturbed interfaces gives rise to Rosensweig instabilities at *both the lower and the upper* interface of the ferrofluid layer. Due to the nonlocal character of the magnetic field energy these instabilities are *coupled* with each other. We first study the interplay and competition between these instabilities within the framework of the linear stability analysis. Depending on the parameters of the system one interface dominates and "slaves" the other one to its unstable wave number or both interfaces become unstable at rather similar values of the magnetic field giving rise to a competition between the corresponding wave numbers. This is similar to what occurs in Rayleigh-Bénard-Marangoni convection in systems of two superimposed fluids which are coupled viscously and thermally at their common interface [8–11].

In order to characterize the patterns evolving from the instability we perturbatively probe into the weakly nonlinear regime by expanding the free energy of the system in powers of the amplitude of the surface deflections generalizing the methods developed in Refs. [5,7,12]. When the amplitude of the surface deformations becomes comparable to the thickness of the ferrofluid layer itself the layer may be decomposed into disconnected parts. Within our nonlinear analysis we are able to estimate the field strength necessary for such a disintegration to occur. Finally by using experimentally relevant values for the parameters we point out interesting experimental realizations of our system.

The main difference between our sandwich system and the somewhat related problem of a ferrofluid *film* investigated in Ref. [13] is the thickness of the ferrofluid layer. For a film this thickness is by definition much smaller than the wavelength of the unstable mode. In the experiments reported in Ref. [13] the film thickness varied between 5 and 60 μm. The hydrodynamics of the film can then be very well described within the lubrication approximation. In our system the thickness of the ferrofluid layer is comparable to the unstable wavelength which is of the order of centimeters and correspondingly the full hydrodynamic equations have to be solved to describe its dynamics.

II. BASIC EQUATIONS

We consider a horizontally unbounded ferrofluid layer of thickness d and density $\rho^{(2)}$ sandwiched between two immis-

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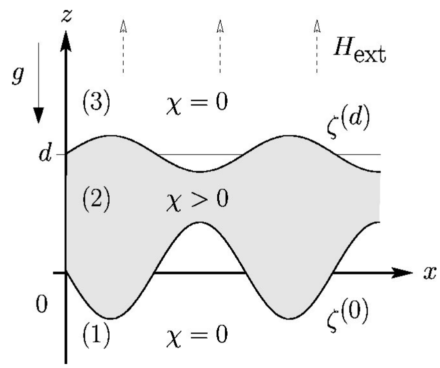

FIG. 1. Schematic two-dimensional plot of a ferrofluid layer of depth d with infinite horizontal extension sandwiched between two nonmagnetic liquids of infinite depth.

cible, nonmagnetic liquids with densities $\rho^{(1)}$ and $\rho^{(3)}$. The interfaces between the layers are parametrized by the functions $z=\zeta^{(d)}(x)$ and $z=\zeta^{(0)}(x)$ where for simplicity we will only consider one-dimensional interface modulations (see Fig. 1). It has recently been clarified that this situation can be realized experimentally by using an *oblique* magnetic field [14]. The interface tensions at the two interfaces are denoted by $\sigma^{(d)}$ and $\sigma^{(0)}$.

The hydrodynamics of the system is quite generally described by the Navier-Stokes equation and the continuity equation. However, since the situation of interest is a static one these equations can be replaced by the pressure equilibrium at the two interfaces. This in turn is equivalent to the minimum condition for the total energy functional.

The energy per area in the x-y plane comprises three parts E_h, E_s, and E_m denoting the hydrostatic, the interfacial, and the magnetic energies, respectively. The first two parts are given by the well-known expressions

$$E_h = g\left\langle \rho^{(1)}\int_{-\infty}^{\zeta^{(0)}(x)} dz\ z + \rho^{(2)}\int_{\zeta^{(0)}(x)}^{\zeta^{(d)}(x)} dz\ z + \rho^{(3)}\int_{\zeta^{(d)}(x)}^{\infty} dz\ z \right\rangle \tag{1}$$

and

$$E_s = \langle \sigma^{(0)}\sqrt{1+[\partial_x\zeta^{(0)}(x)]^2} + \sigma^{(d)}\sqrt{1+[\partial_x\zeta^{(d)}(x)]^2}\rangle, \tag{2}$$

where g is the acceleration due to gravity. The brackets $\langle\cdots\rangle$ denote the spatial average along the x direction,

$$\langle\cdots\rangle = \lim_{L\to\infty}\frac{1}{2L}\int_{-L}^{L} dx\cdots. \tag{3}$$

The volume density of magnetic energy is of the general form [17]

$$e_m = -\mu_0\int_0^{\mathbf{H}_0} d\mathbf{H}'\cdot\mathbf{M}(\mathbf{H}'), \tag{4}$$

where $\mathbf{M}$ denotes the magnetization, $\mathbf{H}_0$ the magnetic field *in the absence* of any permeable material, and μ_0 is the permeability of free space. Assuming a linear magnetization law $\mathbf{M}=\chi\mathbf{H}$ of the ferrofluid with the susceptibility χ characterizing its magnetic properties we hence find in the present case for the magnetic energy per unit area

$$E_m = -\frac{\mu_0\,\chi}{2}\left\langle\int_{\zeta^{(0)}(x)}^{\zeta^{(d)}(x)} dz\ \mathbf{H}(x,z)\cdot\mathbf{H}_{ext}\right\rangle. \tag{5}$$

Here $\mathbf{H}_{\text{ext}}$ denotes the homogeneous external magnetic field produced by the experimental setup and in the absence of the ferrofluid and $\mathbf{H}(x,z)$ is the actual magnetic field in the ferrofluid.

Subtracting an irrelevant constant the complete energy functional of the system can hence be written as

$$\begin{aligned}E[\zeta^{(0)}(x),\zeta^{(d)}(x)] = \Bigg\langle &\frac{g}{2}[(\rho^{(1)}-\rho^{(2)})\zeta^{(0)^2}+(\rho^{(2)}-\rho^{(3)})\zeta^{(d)^2}] \\ &-\frac{\mu_0\,\chi}{2}\int_{\zeta^{(0)}(x)}^{\zeta^{(d)}(x)} dz\ \mathbf{H}(x,z)\cdot\mathbf{H}_{\text{ext}} \\ &+\sigma^{(0)}\sqrt{1+[\partial_x\zeta^{(0)}(x)]^2} \\ &+\sigma^{(d)}\sqrt{1+[\partial_x\zeta^{(d)}(x)]^2}\Bigg\rangle.\end{aligned} \tag{6}$$

The magnetic field has to obey the magnetostatic Maxwell equations

$$\nabla\cdot\mathbf{B}=0 \quad\text{and}\quad \nabla\times\mathbf{H}=0 \tag{7}$$

with $\mathbf{B}=\mu_0(1+\chi)\mathbf{H}$. These equations are completed by the following boundary conditions:

$$\lim_{z\to\pm\infty}\mathbf{H}(x,z) = H_{\text{ext}}\,\mathbf{e}_z \tag{8}$$

and

$$\begin{aligned}&[(\mathbf{B}^{(3)}-\mathbf{B}^{(2)})\cdot\mathbf{n}^{(d)}]_{z=\zeta^{(d)}}=0,\\ &[(\mathbf{H}^{(3)}-\mathbf{H}^{(2)})\times\mathbf{n}^{(d)}]_{z=\zeta^{(d)}}=0,\end{aligned} \tag{9}$$

$$\begin{aligned}&[(\mathbf{B}^{(2)}-\mathbf{B}^{(1)})\cdot\mathbf{n}^{(0)}]_{z=\zeta^{(0)}}=0,\\ &[(\mathbf{H}^{(2)}-\mathbf{H}^{(1)})\times\mathbf{n}^{(0)}]_{z=\zeta^{(0)}}=0,\end{aligned} \tag{10}$$

where $\mathbf{n}^{(0)}$ and $\mathbf{n}^{(d)}$ denote the normal vectors on the lower and the upper interface, respectively. Note that the last four boundary conditions have to be fulfilled at the free interfaces of the ferrofluid layer. They hence describe the feedback of the interface modulations on the magnetic field. Note also that therefore the energy (6) depends in a complicated nonlocal way on the surface deflections $\zeta^{(0)}(x)$ and $\zeta^{(d)}(x)$

It is useful to introduce for each of the three liquid layers a scalar magnetic potential $\Phi^{(1)}$, $\Phi^{(2)}$, and $\Phi^{(3)}$, respectively. The potentials are related to the corresponding magnetic fields by

$$\mathbf{H}^{(i)} = -\nabla\Phi^{(i)} \tag{11}$$

and as a consequence of Eq. (7) they obey the Laplace equations

$$\Delta\Phi^{(i)} = 0. \tag{12}$$

The boundary conditions (9) and (10) for **H** and **B** translate in the well-known way into conditions for the continuity of the potentials themselves and jumps of their normal derivatives [17].

It is furthermore convenient to measure all distances in units of the inverse critical wave number

$$\frac{1}{k_{c,R}} = \sqrt{\frac{\sigma^{(d)}}{(\rho^{(2)} - \rho^{(3)})g}} \tag{13}$$

of the Rosensweig instability on an infinitely deep ferrofluid layer, all magnetic fields in units of the corresponding critical Rosensweig field

$$H_{c,R} = \left(\frac{(1+\chi)(2+\chi)2\sqrt{(\rho^{(2)} - \rho^{(3)})g\sigma^{(d)}}}{\chi^2\mu_0}\right)^{1/2}, \tag{14}$$

and energies per area in units of $\sigma^{(d)}$. Moreover we introduce the parameter ratios

$$\rho_1 = \frac{\rho^{(1)}}{\rho^{(2)}}, \quad \rho_3 = \frac{\rho^{(3)}}{\rho^{(2)}}, \qquad \sigma = \frac{\sigma^{(0)}}{\sigma^{(d)}}, \quad \eta = \frac{\chi}{\chi+2} \tag{15}$$

with η now characterizing the magnetic properties of the ferrofluid. After rescaling the magnetic potentials according to

$$-\frac{(2+\chi)}{\chi H_{\text{ext}}}\Phi^{(1)} \to \Phi^{(1)}, \tag{16}$$

$$-\frac{(1+\chi)(2+\chi)}{\chi H_{\text{ext}}}\Phi^{(2)} \to \Phi^{(2)}, \tag{17}$$

$$-\frac{(2+\chi)}{\chi H_{\text{ext}}}\Phi^{(3)} \to \Phi^{(3)}, \tag{18}$$

the energy (6) assumes the dimensionless form

$$E[\zeta^{(0)}(x),\zeta^{(d)}(x)] = \Bigg\langle \frac{1}{2}\left[\left(\frac{\rho_1-1}{1-\rho_3}\right)\zeta^{(0)^2} + \zeta^{(d)^2}\right] - H_{\text{ext}}^2(\Phi^{(2)}|_{z=\zeta^{(d)}(x)} - \Phi^{(2)}|_{z=\zeta^{(0)}(x)}) + \sigma\sqrt{1+[\partial_x\zeta^{(0)}(x)]^2} + \sqrt{1+[\partial_x\zeta^{(d)}(x)]^2}\Bigg\rangle. \tag{19}$$

The boundary conditions (9) and (10) translate into

$$[\partial_x(\Phi^{(3)} - \Phi^{(2)})\partial_x\zeta^{(d)} - \partial_z(\Phi^{(3)} - \Phi^{(2)})]_{z=\zeta^{(d)}} = 0,$$

$$\left[\frac{1+\eta}{1-\eta}\Phi^{(3)} - \Phi^{(2)}\right]_{z=\zeta^{(d)}} = 0, \tag{20}$$

and

$$[\partial_x(\Phi^{(2)} - \Phi^{(1)})\partial_x\zeta^{(0)} - \partial_z(\Phi^{(2)} - \Phi^{(1)})]_{z=\zeta^{(0)}} = 0,$$

$$\left[\Phi^{(2)} - \frac{1+\eta}{1-\eta}\Phi^{(1)}\right]_{z=\zeta^{(0)}} = 0, \tag{21}$$

respectively, whereas the asymptotic boundary conditions (8) acquire the form

$$\lim_{z\to+\infty}\partial_z\Phi^{(3)}(x,z) = \lim_{z\to-\infty}\partial_z\Phi^{(1)}(x,z) = \frac{1}{\eta}. \tag{22}$$

III. LINEAR STABILITY ANALYSIS

In this section we study the linear stability of the reference state with flat interfaces $\zeta^{(0)} \equiv 0$ and $\zeta^{(d)} \equiv d$. To this end we use the ansatzes

$$\zeta^{(0)} = A_1\cos(kx), \qquad \zeta^{(d)} = d + B_1\cos(kx) \tag{23}$$

for the interface profiles. The corresponding forms of the magnetic potentials are then in view of Eqs. (12) and (22)

$$\Phi^{(3)} = \frac{z}{\eta} - \frac{2d}{1+\eta} + u_1e^{-kz}\cos(kx),$$

$$\Phi^{(2)} = \frac{z}{\eta} + (v_1^+e^{kz} + v_1^-e^{-kz})\cos(kx), \tag{24}$$

$$\Phi^{(1)} = \frac{z}{\eta} + w_1e^{kz}\cos(kx).$$

By using the linearized version of the boundary conditions (20) and (21) we can express the amplitudes u_1, v_1^+, v_1^-, and w_1 in terms of A_1 and B_1. This then allows us to expand the energy (19) up to second order in A_1 and B_1 with the result

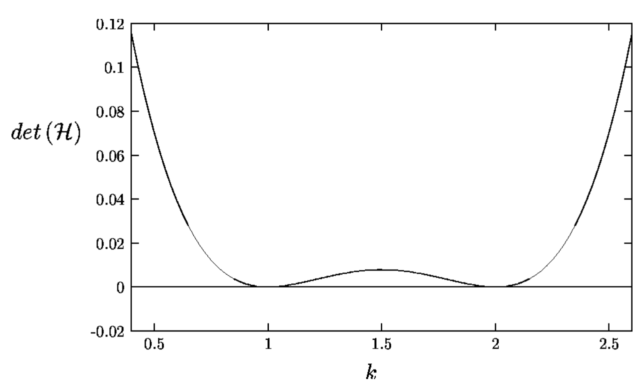

FIG. 2. Determinant of the Hessian for an infinitely thick ferrofluid layer as function of the dimensionless wave number k. Parameters $\rho_1=2$, $\rho_3=0.5$, $\sigma=0.5$, $\eta=1.5$ are chosen such that the two independent interfaces get unstable at the same value of the magnetic field (by definition $H_c=1$), but at different wave numbers $k_c^{(u)}=1$ for the upper interface and $k_c^{(l)}=2$ for the lower one.

$$E(A_1,B_1) = E(0,0) + \frac{1}{4}\left(\frac{\rho_1 - 1}{1-\rho_3} - 2\, H_{\text{ext}}^2\, k \frac{\eta\, e^{-2\ dk} - 1}{\eta^2\, e^{-2\ dk} - 1} + \sigma\, k^2\right) A_1^2 + \frac{1}{4}\left(1 - 2H_{\text{ext}}^2\, k \frac{\eta\, e^{-2\ dk} - 1}{\eta^2\, e^{-2\ dk} - 1} + k^2\right) B_1^2 + H_{\text{ext}}^2\, k(\eta - 1)\frac{e^{-dk}}{\eta^2\, e^{-2\ dk} - 1} A_1 B_1. \tag{25}$$

The energy has clearly a stationary point at $A_1 = B_1 = 0$. It is stable as long as the Hessian

$$\mathcal{H} = \begin{pmatrix} \frac{1}{2}\left(\frac{\rho_1 - 1}{1-\rho_3} - 2\, H_{\text{ext}}^2\, k \frac{\eta\, e^{-2\ dk} - 1}{\eta^2\, e^{-2\ dk} - 1} + \sigma\, k^2\right) & H_{\text{ext}}^2\, k(\eta - 1)\frac{e^{-dk}}{\eta^2\, e^{-2\ dk} - 1} \\ H_{\text{ext}}^2\, k(\eta - 1)\frac{e^{-dk}}{\eta^2\, e^{-2\ dk} - 1} & \frac{1}{2}\left(1 - 2H_{\text{ext}}^2\, k \frac{\eta\, e^{-2\ dk} - 1}{\eta^2\, e^{-2\ dk} - 1} + k^2\right) \end{pmatrix} \tag{26}$$

is positive definite. An instability is signaled by a vanishing determinant of $\mathcal{H}$.

We note that, of course, exactly the same condition results from the usual procedure of linear stability analysis. In this case one investigates the dispersion relation $\omega(k)$ of interface deformations of the form $\zeta^{(0)} = A_1 \exp[i(kx - \omega t)]$ and $\zeta^{(d)} = d + B_1 \exp[i(kx - \omega t)]$ resulting from a linearization of the equations of motion. An instability occurs if ω acquires a positive imaginary part. The linearized equation of motion corresponds to the quadratic approximation of the energy. An advantage of the energetic approach is that it applies equally well to inviscid and viscous fluids. On the other hand it does not give information on the linear growth rate of the unstable perturbation.

If the layer thickness d tends to infinity it can be inferred from Eq. (26) that the off-diagonal elements of $\mathcal{H}$ tend to zero whereas the diagonal elements reduce to the well-known form of a usual Rosensweig instability on a infinitely deep layer of ferrofluid [1]. As expected we hence find in this limit two *uncoupled* interfaces showing *independent* Rosensweig instabilities of the usual kind. The situation is depicted in Fig. 2 where we have shown the determinant of the Hessian matrix $\mathcal{H}$ as function of the wave number k for the case in which the critical field of the two instabilities coincides but the respective critical wave numbers do not.

The situation changes if the layer thickness is reduced as shown in Fig. 3. Due to the interaction between the surface deformations mediated by the magnetic field the degeneracy observed in the case $d = \infty$ is lifted and the lower layer "slaves" the upper one to its critical wave number. At the same time the critical wave number is shifted somewhat, $k_c \neq 1$, from its "pure" value of the decoupled case. The same holds true for the critical magnetic field strength. Moreover, the two interface deflections accommodate to each other in an antiphase fashion. This manifests itself in different *signs* of A_1 and B_1 building the components of the eigenvector corresponding to the zero eigenvalue of $\mathcal{H}$. This anti phase orientation was to be expected intuitively since it allows the largest gain in magnetic energy (cf. Fig. 1).

Which interface dominates which depends on the parameter values of the system and accordingly a crossover can be observed when some parameter is changed. In Figs. 4–6 we give some examples of such crossover phenomena when the ratio σ between the two interface tensions is changed. Figure 4 displays the relative amplitude of the two surface deflections. The figure clearly indicates that for small values of σ, i.e., when $\sigma^{(0)} \ll \sigma^{(d)}$, the lower instability dominates, $A_1 \gg B_1$, whereas with increasing σ the amplitude of the lower interface deflection decreases and the coupled unstable modes get more and more dominated from the upper interface. Similarly Figs. 5 and 6 show the crossover of the critical wave number k_c and the critical field H_c, respectively, when σ is varied. In all cases the crossover gets sharper with increasing depth d of the ferrofluid layer as expected.

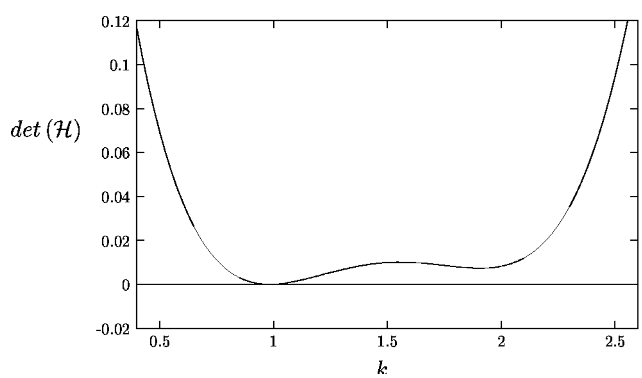

FIG. 3. Same as Fig. 2 for a layer thickness $d = 2$. The coupling between the two surfaces now lifts the degeneracy characteristic of Fig. 2 giving rise to new critical values for the wave vector $k_c = 0.96$, and the magnetic field $H_c = 0.98$.

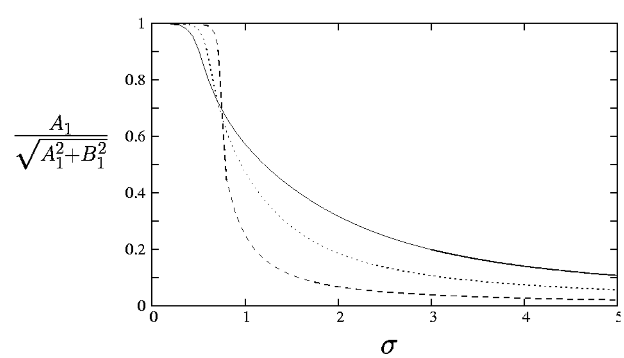

FIG. 4. Relative amplitudes of the unstable modes as function of the ratio σ of the interface tensions as defined in Eq. (15). The layer thickness is $d = 2$ (dashed), $d = 1$ (dotted), and $d = 0.5$ (solid). The other parameter values are $\eta = 0.66$, $\rho_1 = 1.2$, and $\rho_3 = 0.85$.

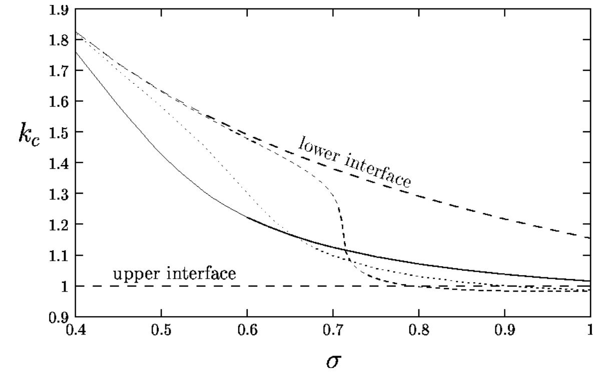

FIG. 5. Dimensionless critical wave number of the linear instability as function of the ratio σ of interface tensions for layer thickness $d=\infty$ (long dashed), $d=2$ (dashed), $d=1$ (dotted), $d=0.5$ (solid). Other parameters as in Fig. 4.

Although the linear analysis already reveals some aspects of the interplay between the two Rosensweig instabilities it is not able to yield information about the static pattern of interface deflections that will eventually emerge. In order to address this problem we need to extend our analysis to include nonlinear terms able to saturate the exponential growth predicted by the linear stability theory. This is the subject of the following section.

IV. WEAKLY NONLINEAR ANALYSIS

In our analysis of the energy of the surface deflections the instability of the flat surface corresponds to the minimum of the energy at $A_1=B_1=0$ turning into a saddle point. Within the quadratic approximation the energy hence decreases down to $-\infty$ with increasing amplitudes A_1 and B_1. In reality, however, already for moderate values of A_1 and B_1 higher-order terms in the expansion of the energy have to be included which cure this divergence. As a result the energy again *increases* with increasing amplitudes A_1 and B_1 and correspondingly a new minimum forms describing the new stationary surface profiles $\zeta^{(0)}(x)$ and $\zeta^{(d)}(x)$.

We assume that the susceptibility of the ferrofluid is sufficiently small such that an expansion of the energy (19) up

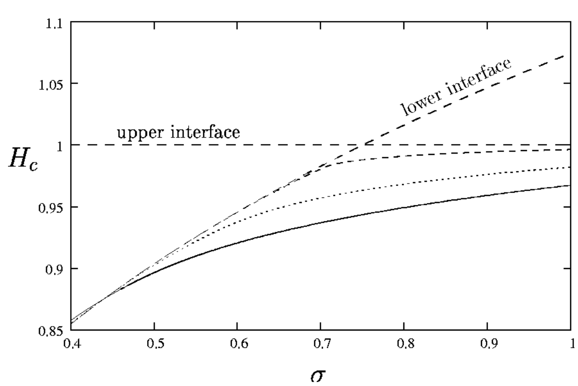

FIG. 6. Dimensionless critical magnetic field H_c of the linear instability as function of the ratio σ between the interface tensions for layer thickness $d=\infty$ (long dashed), $d=2$ (dashed), $d=1$ (dotted), and $d=0.5$ (solid). Other parameters as in Fig. 4.

to fourth order in the amplitudes of the surface deflection is sufficient to find the new stationary state. Such an expansion is equivalent to the derivation of a third-order amplitude equation for the unstable mode [15]. In order to obtain a consistent expansion the Fourier expansions (23) and (24) have to be extended according to

$$\zeta^{(d)}(x) = d + \sum_{n=1}^{2} B_n \cos(nkx),$$
$$\zeta^{(0)}(x) = \sum_{n=1}^{2} A_n \cos(nkx), \qquad (27)$$

and

$$\Phi^{(3)} = \frac{z}{\eta} - \frac{2d}{1+\eta} + \sum_{n=1}^{2} u_n e^{-nkz}\cos(nkx),$$
$$\Phi^{(2)} = \frac{z}{\eta} + \sum_{n=1}^{2} (v_n^{+} e^{nkz} + v_n^{-} e^{-nkz})\cos(nkx), \qquad (28)$$
$$\Phi^{(1)} = \frac{z}{\eta} + \sum_{n=1}^{2} w_n e^{nkz}\cos(nkx).$$

To explicitly perform the minimization of the free energy a variant of the computer algebra code documented in Ref. [12] is used. Fixing the desired order of the expansion (four in our case) this program selects in a first step those amplitude combinations which are compatible with the translational invariance of the problem in x direction. In our case only 17 of the originally 70 terms remain after this procedure. In a second step the ansatzes (27) and (28) are used in the boundary conditions (20) and (21) and the coefficients $u_n, v_n^{+}, v_n^{-}, w_n$ are determined as polynomials in the A_n and B_n. After this the energy (19) can be expanded up to fourth order in A_1 and B_1 and up to second order in A_2 and B_2. Several of the remaining terms disappear after the integration over x implicit in the horizontal average in Eq. (19). Minimizing the resulting expression in A_2 and B_2 we find that both are of order A_1^2, B_1^2 which proves the consistency of our expansion *a posteriori*. Finally the free energy is minimized in the amplitudes A_1 and B_1 of the main modes. The final expressions are explicit but too long to be displayed [16].

TABLE I. Magnetic fluid parameters used in Figs. 7–9.

Experimental parameters	Dimensionless values
$\rho^{(1)}=1.69$ g/cm^3	
$\rho^{(2)}=1.12$ g/cm^3	$\rho_1=1.51$
$\rho^{(3)}=0.0013$ g/cm^3	$\rho_3=0.001$
$\sigma^{(0)}=16.6$ mN/m	
$\sigma^{(d)}=25.9$ mN/m	$\sigma=0.64$
$d=1.54$ mm	$d=1.0$
$\chi=0.8$	$\eta=0.29$

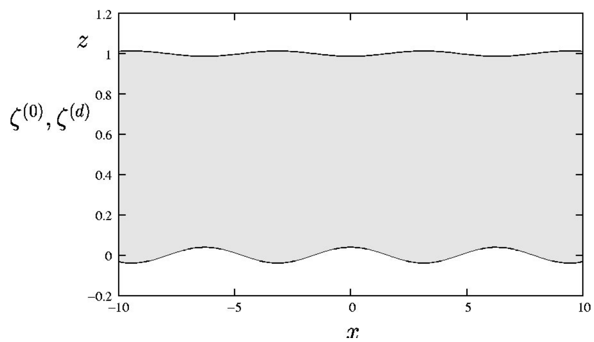

FIG. 7. Stationary pattern of coupled surface deflections that evolve after the instability of the state with flat interfaces. The ferrofluid layer is shown in gray. Parameters are given in Table I, the value of the external magnetic field is $H_{\mathrm{ext}}=1.0001H_c$. The figure uses dimensionless units.

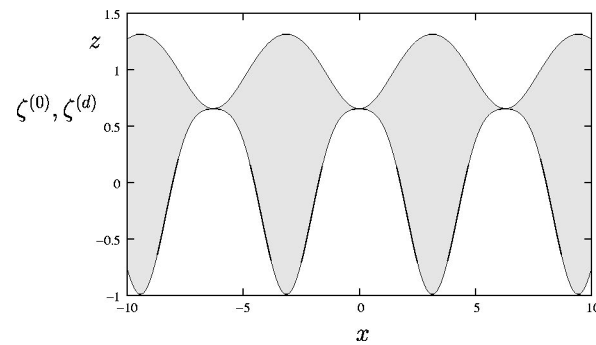

FIG. 9. Same as Fig. 7 for $H_{\mathrm{ext}}=1.04H_c$.

For $d=\infty$ we again reproduce the results obtained for the standard Rosensweig instability on a layer of infinite depth [5,7]. For $d<\infty$ the two interfaces couple and the two surface deflections arrange in a stable, antiphase pattern.

To elucidate this final structure in detail we consider the case of experimentally realistic parameters collected in Table I. From the linear stability analysis we find for the dimensionless wave number $k_c=0.84$ and for the corresponding dimensionless field $H_c=0.75$. The stationary interface profiles resulting from the weakly nonlinear analysis are displayed in Figs. 7–9. As can be seen the lower interface is the dominating one. For slightly overcritical magnetic field the lower interface already shows an array of developed Rosensweig ridges whereas the upper one is just gently curved by the inhomogeneous magnetic pressure resulting from the field modulation induced by the lower interface (Fig. 7). With increasing field both deformations grow (Fig. 8). A particular interesting case is shown in Fig. 9 where the amplitudes of the surface deflections have increased to such an extent that the two interfaces touch each other. Correspondingly the ferrofluid layer stays no longer connected but disintegrates. In our two-dimensional (x,z) model this gives rise to the formation of parallel slices. In a more realistic three-dimensional setting, including surface variations in y direction as well, the layer would evolve into a regular array of disconnected *islands*. A similar phenomenon occurs in the usual Rosensweig instability on very shallow layers of ferrofluid [7].

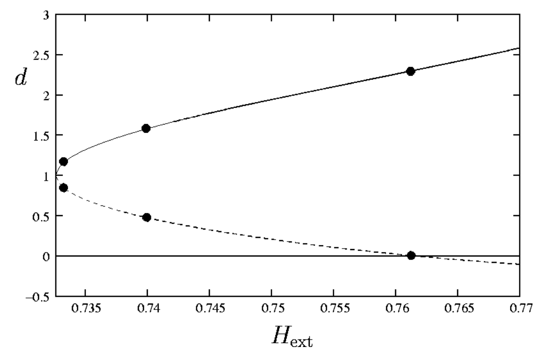

FIG. 10. Maximum (full line) and minimum (dashed line) dimensionless thickness of the ferrofluid layer as function of the dimensionless external field strength for the parameters given in Table I. The dots correspond to the situations displayed in Figs. 7–9, respectively. For the field at which the minimum distance between the interfaces shrinks to zero the layer disintegrates into an array of disconnected rolls.

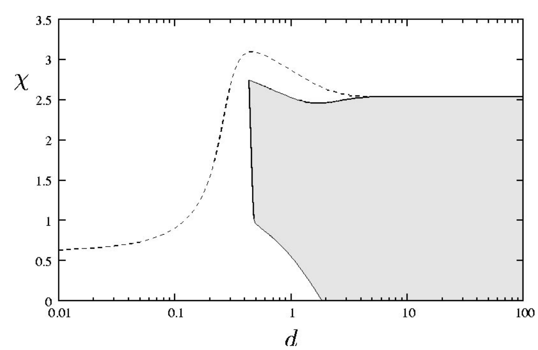

FIG. 11. Region of consistency of our nonlinear treatment of the pattern formation in the plane spanned by the dimensionless layer thickness d and the susceptibility χ. Outside the shaded region the fourth-order terms in the expansion of the energy are not sufficient to saturate the linear instability and higher-order terms are needed to get finite results for the amplitudes of interface deflections when minimizing the energy. The dashed line is the result of Ref. [7] for a ferrofluid layer with rigid bottom. Parameter values are from Table I.

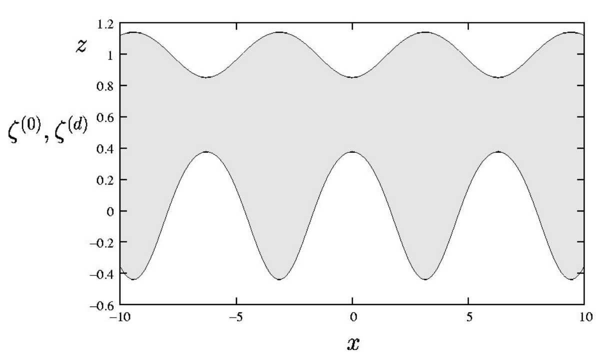

FIG. 8. Same as Fig. 7 for $H_{\mathrm{ext}}=1.01\ H_c$.

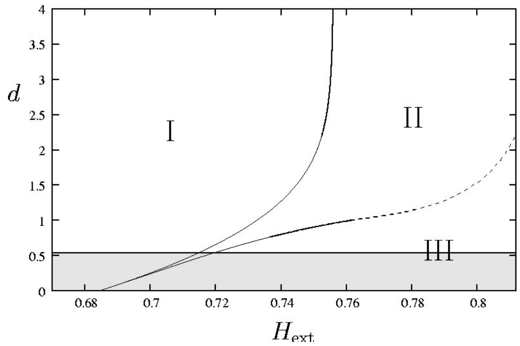

FIG. 12. Phase diagram in the plane spanned by the dimensionless external field H_{ext} and layer thickness d for a ferrofluid sandwich structure with the parameters given in Table I. In region I both interfaces are flat, in region II, ridges occur and in region III, the layer disintegrates into an array of disconnected rolls. The dashed line indicates that for larger values of the magnetic field higher-order terms are necessary to accurately determine the location of the transition line. In the shaded region the fourth-order terms in the energy are not sufficient to saturate the linear instability and higher-order terms are mandatory, cf. Fig. 11.

In Fig. 10 we have shown the maximum and the minimum layer thickness as function of the magnetic field. The formation of islands occurs when the lower branch intersects with the horizontal axis. Note that, at least for the parameters of Table I, this happens already for a field exceeding the critical one by only 4%.

With the appearance of such an island structure our theoretical model breaks down. To study the future evolution of the structure when increasing the field still further it is more appropriate to start from a model of independent ferrofluid drops [18].

In omitting higher orders of the expansion of the energy in the amplitudes of surface deflection in our nonlinear analysis we have tacitly assumed that the fourth-order terms are sufficient to saturate the linear instability, i.e., to make $E[\zeta^{(0)},\zeta^{(d)}]\to\infty$ for $[(\zeta^{(0)})^2+(\zeta^{(d)})^2]\to\infty$. This, however, is correct only if the susceptibility χ of the ferrofluid is not too large and the thickness d of the layer is not too small. In Fig. 11 we have displayed the region in the d-χ plane, in which our treatment is consistent.

Finally Fig. 12 gives the phase diagram of the ferrofluid sandwich structure showing the transition lines from flat interfaces to antiphase interface modulations and further to disconnected regions. It would be interesting to compare the location of these theoretical lines with experimental results.

V. CONCLUSION

In the present paper we have investigated the linear and weakly nonlinear theory of two coupled Rosensweig instabilities in a ferrofluid sandwich structure. To this end an approximate expression for the energy of the system was minimized in the deflection amplitudes of the two interfaces between magnetic and nonmagnetic liquids. The approximate expression for the free energy was obtained from a fourth-order perturbative expansion in these interface deflections. At the onset of instability the two individual Rosensweig instabilities compete and depending on the concrete values of the parameters one is able to "slave" the other one to its unstable wave number. As a result, a stable antiphase pattern of two interacting modulated interfaces arises. For sufficiently thin layers and sufficiently large magnetic fields the two curved interfaces may touch each other which brings about the disintegration of the layer and gives rise to disconnected rolls or islands. Using realistic parameter values we gave estimates of the required layer thicknesses and magnetic fields necessary to observe this phenomenon in an experiment. Being perturbative in nature our theoretical analysis has a limited range of validity which we quantified by estimating the contributions of higher-order terms. It is possible though tedious to push the expansion to higherorders in a systematic way.

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