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Rosensweig Instability of Ferrogel Thin Films or Membranes

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Abstract. We derive the dispersion relation of surface waves for magnetic gel membranes or thin films at the interface between two fluids in the presence of an external magnetic field normal to the free surface. Above a critical field strength surface waves become linearly unstable with respect to a stationary pattern of surface protuberances. This linear stability criterion generalizes that of the Rosensweig instability for ferrofluid and ferrogel free surfaces to take into account bending elasticity and intrinsic elastic and magnetic surface properties of the film or membrane, additionally. The latter is of interest for uniaxial ferrogel film or membranes, which show a locked-in permanent magnetization.

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1 Introduction

Free surfaces of ferrofluids, stable colloidal solutions of single-domain magnetic nanoparticles in a carrier fluid, are known to undergo a transition from a flat state to a stationary pattern of surface spikes above a certain threshold of an applied normal magnetic field [1]. The instability mechanism is the focusing effect on the magnetization at the wave crests of an undulating surface, which has the tendency to increase the undulations. At the threshold this destabilizing effect is balanced by surface tension and gravity, which act towards a flat surface. In ferrogels, a polymer network crosslinked in the presence of a ferrofluid [2], elasticity of the network constitutes an additional stabilizing mechanism increasing the threshold value of the external magnetic field without, however, changing the characteristic wavelength of the most unstable linear mode [3].

These types of Rosensweig or normal field instabilities can be understood as the breakdown of the propagation of surface waves at the free surface. The effective softening of the surface by the normal field makes the wave speed (or frequency) vanish, which below the threshold is finite and describes propagating gravity or capillary waves in ordinary liquids and also modified transverse elastic waves in more complex systems like viscoelastic liquids or gels [4]. An important feature of this interpretation of the Rosensweig instability is the lack of any bulk magnetic force in ferrofluids and ferrogels (neglecting magnetostriction) with the result that the driving force is manifest in the boundary conditions only.

Here, we are interested in the situation of a thin layer of a magnetic gel sandwiched between two fluids in the presence of a normal field. The fluids above and below can be identical or different, in particular the former can be vapor/air or even vacuum. For simplicity we assume those fluids to be Newtonian, but generalization is straightforward. The fluids can also have paramagnetic properties. We do not consider gravitationally unstable configurations, so only the magnetic instability occurs. In contrast to the case of a free surface of a bulk ferrogel, a film or membrane can buckle as a whole thus bringing bending elasticity into the picture. Since the latter has a different wavevector dependence than ordinary elasticity, one can expect that in this case the critical wavevector at the onset of instability does depend on the elastic properties of the gel. Another reason for studying films is that generally film or surface attributes differ from bulk ones. The instabilities considered here probe specifically surface elastic and surface magnetic properties. We only consider very thin films, or finite thickness films that buckle as a whole neglecting peristaltic motions.

Besides isotropic ferrogels there are also uniaxial ones, where the latter are obtained by performing the crosslinking process in the presence of a magnetic field [5, 6]. In some cases their elastic anisotropy can be very small (if present at all) [5] and can safely be neglected. On the other hand, they show a residual, locked-in permanent magnetization, which will play a role, in particular if the paramagnetic properties of the bulk fluids above and below the film or membrane are identical. The case of hydrodynamically equivalent bulk fluids, where the gravitational

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influence on the film or membrane is reduced, will also be treated briefly.

The present treatment can also be applied to thermoreversible ferrogels [7] as long as the temperature is below the transition value, and it also contains the case of a simple ferrofluid or a viscoelastic ferrofluid (magnetorheological) film. The complete set of dynamic equations describing isotropic and uniaxial ferrogels has been given recently in [8] and [9], respectively, using the method of generalized hydrodynamics. There are various reversible and irreversible dynamic crosscouplings between flow, elasticity and magnetization, of which we only keep those that are presumably the relevant ones for the present problem.

2 Non-magnetic film modes

Some time ago the full dispersion relation for surface waves $\xi = \xi_0 \exp i(\omega t - kx)$, Fig.(1), of a nonmagnetic, viscoelastic thin film on top of a simple fluid has been derived [10] starting from basic hydrodynamic equations including a generalized Maxwell model for the viscoelastic properties. The fluid above the film is vapor/air and approximated as vacuum. It can be applied to the case of a permanently crosslinked (elastic) gel by putting the Maxwell relaxation time to infinity. In that case the dispersion relation between the frequency of the surface wave and its wavevector k reads implicitly $D(k,\omega) = 0$ with

$$D(k,\omega) = \left[\tilde{\mathcal{C}}^{(z)}(k,\omega)k^3 + i\eta k(q+k)\omega - \rho\omega^2\right] \times \left[\mathcal{C}^{(x)}(k,\omega)k^3 + i\eta k(q+k)\omega\right] + \eta^2 k^2 (q-k)^2 \omega^2, \quad (1)$$

where ρ and η are the density and viscosity of the underlying simple fluid. The surface disturbances decay exponentially inside the lower bulk fluid. For most of the variables or excitations involved the wavelength $1/k$ also acts as the decay length, except for the rotational part of the velocity field, whose decay length is $1/q$ with $q^2 = k^2 + i\rho\omega/\eta$. Equation (1) describes the well-known Lucassen mode spectrum [11–13]. In the case of a viscoelastic bulk fluid (or elastic gel), Eq. (1) remains valid [4], if η is replaced by $\eta + E_0\tau/(1+i\omega\tau)$ with τ the elastic relaxation time and E_0 the elastic plateau modulus ($\tau \to \infty$ in the gel case).

This dispersion relation reflects the coupling of transverse elastic and longitudinal sound bulk modes at the surface. It contains in-layer compressional and transverse (normal to the interface) deformations and flow of the gel layer. The in-layer shear mode is decoupled and does not take part in the surface waves. The material properties of the gel film are contained in the functions

$$\mathcal{C}^{(x)}(k,\omega) = \varepsilon + i\omega\nu_{\parallel} + c_{\parallel} \quad (2)$$

$$\mathcal{C}^{(z)}(k,\omega) = \gamma + i\omega(\nu_{\perp} + \nu_b k^2) + c_{\perp} + c_b k^2 \quad (3)$$

which appear on the r.h.s. of the transverse and normal stress boundary conditions [10]

$$\sigma^{(a)}_{\alpha z} - \sigma^{(b)}_{\alpha z} = \nabla_\beta\, \sigma^{(m)}_{\alpha\beta}, \quad (4)$$

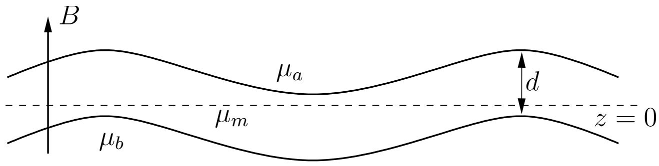

Fig. 1. Periodic lateral perturbations $\xi(x,y,t)$ with wavevector k of the flat ferrogel film around $z = 0$ between media of different magnetic permeabilities $\mu_b = 1 + \chi_b$ (below) and $\mu_a = 1 + \chi_a$ (above) in the thin film limit $kd \ll 1$. The magnetic field $\boldsymbol{B}$ and gravity $\boldsymbol{g} = -g\boldsymbol{e}_z$ are along the z-axis.

with $\{\alpha,\beta\} = \{x,y,z\}$ and the superscripts a, b referring to the media above and below the film (m), respectively. Here ε, γ, $c_{\parallel}$, $c_{\perp}$, c_b, $\nu_{\parallel}$, $\nu_{\perp}$, and ν_b are the film compressional (or Gibbs) modulus, the surface tension, the longitudinal and transverse elastic moduli, the bending elastic coefficient, and the appropriate viscosities, respectively. In contrast to ordinary 3D elastic moduli, the film elastic moduli have the same dimension as the surface tension. Therefore, to simplify notation, the combinations $\tilde{\varepsilon} = \varepsilon + c_{\parallel}$ and $\tilde{\gamma} = \gamma + c_{\perp}$ can be defined. Similarly, the abbreviation $\tilde{\mathcal{C}}^{(z)}(k,\omega) = \mathcal{C}^{(z)}(k,\omega) + \rho g/k^2$ in Eq. (1) already contains the gravity effect on the film.

In the case of a viscoelastic (rather than elastic) gel, $c_{\parallel}$ and $c_{\perp} + c_b k^2$ have to be replaced by $i\omega\tau_{\parallel} c_{\parallel}/(1 + i\omega\tau_{\parallel})$ and $i\omega\tau_{\perp}(c_{\perp} + c_b k^2)/(1 + i\omega\tau_{\perp})$, respectively, with $\tau_{\parallel}$ and $\tau_{\perp}$ being the longitudinal and transverse elastic relaxation times. For a liquid film, $c_{\parallel}$, $c_{\perp}$, and c_b are simply zero.

In the (hydrodynamically) symmetric case with fluids of the same density and viscosity below and above the elastic layer, the relevant dispersion relation [10] is much simpler than Eq. (1)

$$D^{sym}(k,\omega) = k^3(q-k)\mathcal{C}^{(z)}(k,\omega) - 2\rho q\omega^2. \quad (5)$$

In particular, there is no gravity force, as long as the inertia of the film itself can be neglected.

3 Ferrogel film surface modes

As discussed in the Introduction and shown in [3], the influence of an external magnetic field on a ferrogel (and ferrofluid) surface deformation is manifest only in the boundary conditions (magnetostriction neglected), in particular in the normal stress boundary condition. Here, the stabilizing contributions of surface tension γ, gravity g, and elasticity μ_2 are amended by a destabilizing addition due to the external magnetic field $\boldsymbol{B}$ by the replacement

$$\gamma k^2 + \rho g + \mu_2 k \;\longrightarrow\; \gamma k^2 + \rho g + \mu_2 k - \kappa B^2 k, \quad (6)$$

as can be seen from Eq. (17) of [3] (the surface tension is denoted as σ there). Here $\kappa = \chi^2(1+\chi)^{-1}(2+\chi)^{-1}$ with χ the magnetic susceptibility of the ferrogel. The external magnetic field induces a magnetization $\boldsymbol{M} = \chi\boldsymbol{B}/(1+\chi)$ in the ferrogel. The magnetic field effect is quadratic meaning that the orientation of the field (parallel or antiparallel) with respect to gravity or to the surface normal does not matter.

For a (magnetic) film the magnetic influence on surface deformations comes from two surfaces, an upper and lower one to fluid a and b with magnetic susceptibilities χ_a and χ_b, respectively. For a very thin film or a film with equal deformations at both surfaces (disregarding peristaltic motions) the magnetic properties of the bulk fluids enter only via the l.h.s. of the normal stress boundary condition (4). Therefore, the magnetic destabilizing influence from the two surfaces leads to a contribution with $\kappa \to \kappa_1 \sim (\chi_a - \chi_b)^2$ in Eq. (6) independent of the magnetic properties of the film. If the two bulk fluids are magnetically equivalent (magnetically symmetric case), there is no destabilizing effect of a normal magnetic field coming from the boundaries. A rigorous and complete derivation of κ_1 is given in the Appendix with the result

$$\kappa_1 = \frac{(\chi_a - \chi_b)^2}{(\mu_a + \mu_b)\mu_a\mu_b}, \tag{7}$$

with permeabilities $\mu = 1+\chi$. In the case $\chi_a = 0$ (vacuum) the expression for χ_1 of Ref. [3] is regained.

However, as is the case for viscous and elastic film properties, the magnetic properties of the film itself enter the $\mathcal{C}^{(z)}$ function (3) via the right hand side of Eq. (4). A uniaxial ferrogel film does have a permanent (surface) magnetization, while in an isotropic one a considerably large surface magnetization can be induced by an external field. This induced magnetization is always parallel to the external field and has a stabilizing effect on surface waves (cf. Appendix). The frozen-in surface magnetization $\tilde{M}_0$, however, deforms with the membrane or film and produces a stabilizing (destabilizing) effect, if it is parallel (antiparallel) to an external field (cf. Appendix). This influence of a permanent surface magnetization on surface deformations is of the same k-order as the film elasticity and the surface tension and can be described by the replacement

$$\tilde{\gamma} \to \tilde{\gamma} \pm M_0' B, \tag{8}$$

where we are interested in the destabilizing case, only. Here, M_0', the magnetic moment density per unit area (rather than volume) [14] is used in the same spirit as has been done above for the elastic and viscous properties of the film. Some surface contributions to the magnetic free energy are discussed within a different approach in [15].

Taking together both magnetic contributions to the normal stress boundary condition (4) the $\mathcal{C}^{(z)}$ function

$$\mathcal{C}^{(z)}(k,\omega) = \tilde{\gamma} - M_0' B + i\omega(\nu_\perp + \nu_b k^2) + c_b k^2 - \kappa_1 B^2 k^{-1} \tag{9}$$

replaces Eq. (3), while Eq. (2) remains the same. Using these two functions in the dispersion relation (1) for a half-space surface, or in Eq. (5) for the hydrodynamically symmetric interface (or in Eq. (B24) of Ref. [10] for the general case) describes propagating, weakly damped surface waves at magnetic films that can be excited and maintained by thermal fluctuations, external mechanical (acoustic) forces, or other means. The wave propagation speed is clearly reduced due to the action of the magnetic field, which "softens" the stiffness of the film or membrane. Non-propagating modes are also possible.

4 Rosensweig instability

Eqs. (1,2), and (9) can be slightly reinterpreted: These are conditions for an external field strength B, at which a surface perturbation ξ with wavevector k and (real) frequency ω_0 relaxes to zero or grows exponentially for λ negative or positive, respectively ($\omega = \omega_0 - i\lambda$). For $\lambda = 0$ such a surface perturbation is marginally stable (or unstable) against infinitesimal disturbances, since Eq. (1) has been obtained by linearizing the dynamic equations and the boundary conditions about the ground state. The functions ω_0 and B still depend on k and the latter has to be minimized with respect to k in order to get the true linear instability threshold. There is no guarantee that a threshold exists for a finite frequency due to the additional requirement $\omega_0^2 > 0$. We therefore discuss first the stationary case. Assuming $\omega_0 = 0$ the threshold condition $\lambda = 0$ leads to $\tilde{\mathcal{C}}^{(z)}(k, \omega{=}0) = 0$. We will further analyze this condition for the special cases, where the surface magnetism can be either neglected or has only a small influence in Sec. 4.1, a permanently magnetized film with no magnetic contrast of the surrounding fluids in Sec. 4.2, while the general case, when both destabilizing magnetic field effects are present, is discussed in Sec. 4.3. The possibility of an oscillatory instability and the case of hydrodynamically symmetric configurations is discussed in the final subsection Sec. 4.4.

4.1 Stationary, asymmetric case without surface magnetism

Dealing with the case of a strong magnetic contrast between the upper and lower bulk fluid (*e.g.* vacuum and a ferrofluid, respectively), we neglect the surface magnetic effect. Experimentally, this case can be realized by a ferrogel (or a non-magnetic gel) on top of a ferrofluid and vapor or vacuum above the film. In that case the threshold magnetic field is

$$\kappa_1 B^2(k) = \tilde{\gamma} k + \frac{\rho g}{k} + c_b k^3 \tag{10}$$

and is finite for a non-zero magnetic contrast, $\chi_a \neq \chi_b$, of the bulk fluids, only. Minimizing with respect to k leads to the critical wavevector

$$k_c^2 = \frac{1}{6c_b}\left(\sqrt{\tilde{\gamma}^2 + 12\rho g c_b} - \tilde{\gamma}\right) \tag{11}$$

and the critical magnetic field $B_c = B(k = k_c)$. Slightly above the minimum, the curvature of the marginal stability curve is given by $\kappa_1(B(k)^2 - B_c^2) = (1/k_c)\sqrt{\tilde{\gamma}^2 + 12\rho g c_b}(k - k_c)^2$.

The linear threshold conditions for this stationary instability are completely independent of the viscosities of both, the underlying fluid as well as the film itself, resembling the case of bulk free surface Rosensweig instabilities in ferrofluid and ferrogels [3]. In contrast to the latter case, here the critical wavevector does depend on the transverse elastic properties ($c_\perp$) of the ferrogel (through $\tilde{\gamma}$) as well as on the bending elastic modulus c_b. The reason is that both effects enter the normal stress boundary condition with a k-dependence different from that of the magnetic field [cf. Eq. (9)], or to phrase it differently, the magnetic field deformations do not introduce a specific internal length scale compared to ordinary 3 D elasticity, but they do in relation with surface elasticity.

On the other hand, the linear growth rate λ of the most unstable mode is completely determined by the (transverse) viscous properties of the film and the bulk fluid

$$\lambda = \frac{\kappa_1(B^2 - B_c^2)}{\nu_\perp k_c + \nu_b k_c^3 + 2\eta}, \tag{12}$$

where the wave vector of the most unstable mode $k_u = k_c(1 - \delta)$ with

$$\delta = \frac{\kappa_1(B^2 - B_c^2)}{2\tilde{\gamma} + 12 c_b k_c^2} \, \frac{\nu_\perp + 3\nu_b k_c^2}{\nu_\perp k_c + \nu_b k_c^3 + 2\eta} \tag{13}$$

is slightly smaller than the critical one. If the dissipation in the film or membrane can be neglected, the growth rate is given by the bulk fluid viscosity, only, $\lambda = \kappa_1(B^2 - B_c^2)/(2\eta)$ as in the case of a bulk ferrofluid or ferrogel, and the most unstable mode is the critical one, $k_u = k_c$ in linear order [16].

The linear threshold conditions for the stationary instability are also independent of the longitudinal material properties (ϵ, $c_\parallel$) of the film and therefore indistinguishable from those of an incompressible film.

Since we are operating in the long wavelength limit, usually the bending elasticity is less important than ordinary elasticity, except for very thin films, where $c_\perp$ and γ are zero or can be neglected. In the former case, in particular for $\rho g c_b \ll \tilde{\gamma}^2$ the critical quantities can be simplified to

$$k_c^2 = \frac{\rho g}{\tilde{\gamma}} \left(1 - 3\frac{\rho g c_b}{\tilde{\gamma}^2}\right) \tag{14}$$

$$\kappa_1 B_c^2 = 2\sqrt{\rho g \tilde{\gamma}} \left(1 + \frac{1}{2}\frac{\rho g c_b}{\tilde{\gamma}^2}\right) \tag{15}$$

Of course, the critical wavelength and field increase with increasing elasticity and scale at onset with the relevant elastic modulus of the ferrogel $c_\perp$ with exponents 1/2 and 1/4, respectively. In the pure ferrofluid case, $c_\perp = 0 = c_b$, the critical values are identical to those of the usual Rosensweig instability, *i.e.* there is no difference between a bulk free surface and a film, except for a possible difference in the surface tension γ in the two cases.

In the opposite, bending dominated regime, $\rho g c_b \gg \tilde{\gamma}^2$ the critical values are

$$k_c^4 = \frac{\rho g}{3 c_b} \left(1 - \frac{\tilde{\gamma}}{\sqrt{3\rho g c_b}}\right) \tag{16}$$

$$\kappa_1^2 B_c^4 = \frac{16}{9}\rho g \sqrt{3\rho g c_b} \left(1 + \frac{3}{2}\frac{\tilde{\gamma}}{\sqrt{3\rho g c_b}}\right) \tag{17}$$

Here, the critical wavelength and field scale at onset with the bending elastic modulus of the ferrogel film c_b with exponents 1/4 and 1/8, respectively.

4.2 Permanent-magnetic, symmetric case

We now consider a film consisting of a permanent-magnetic gel with the intrinsic (surface) magnetization M_0' to be rigidly anchored to the elastic degrees of freedom. In particular we choose it to be always antiparallel to the external field B. In this section we just discuss the case of magnetic symmetry between the bulk fluids a and b being either non-magnetic or having the same magnetic susceptibility. For this case the magnetic contribution stemming from the left hand side of Eq. (4) cancels (κ_1 in Eq. (7) is zero) and only the divergence of the magnetic membrane stress tensor gives a field dependent contribution to the threshold condition for a stationary instability

$$\tilde{\mathcal{C}}^{(z)}(k) = \rho g k^{-2} + \tilde{\gamma} - M_0' B + c_b k^2 = 0 \tag{18}$$

Here, ρ is the density difference between the medium above and below the film or membrane. Eq. (18) leads to an instability with a characteristic mode

$$k_c^4 = \frac{\rho g}{c_b} \tag{19}$$

when the applied critical field reaches the threshold value

$$B_c = \frac{1}{M_0'}\Big(\tilde{\gamma} + 2\sqrt{c_b \rho g}\Big). \tag{20}$$

Note that the critical wavevector is independent of M_0', dominated by the bending elastic coefficient, and rather similar to Eq. (16). The threshold field is inversely proportional to the magnitude of the intrinsic permanent magnetization.

4.3 The general case

We now discuss the general case, where both destabilizing magnetic field effects are present, *i.e.* a uniaxial film with the permanent magnetization opposite to the field a magnetic contrast between the two surrounding fluids. The condition for marginal stability against a stationary convection, Eq. (9),

$$\tilde{\mathcal{C}}^{(z)}(k) = \rho g k^{-2} + \tilde{\gamma} + c_b k^2 - M_0' B - \kappa_1 B^2 k^{-1} = 0 \tag{21}$$

leads to the neutral curve $B = B(k)$. In principle, one could expect a competition between the two different instabilities described in the two preceding subsections, *i.e.*

a transition from a stationary instability with a wavevector like that of Eq. (11) to one like that of Eq. (19).

The minimum threshold condition $dB/dk = 0$ allows us the calculate the critical wave vector k_c as a real root of

$$\kappa_1(3c_b k_c^4 + \tilde{\gamma} k_c^2 - \rho g)^2 + 2M_0'^2(c_b k_c^4 - \rho g)k_c^3 = 0. \quad (22)$$

In dimensionless form Eq. (22) contains three relevant numbers $R_B = c_b/(\rho g d^4)$, $R_E = \tilde{\gamma}/(\rho g d^2)$, and $R_M = M_0'^2/(\rho g \kappa_1 d^3)$, if the wavevector is scaled by the film thickness d. For $R_M > R_B, R_E$ there are two different minimum solutions, k_{c1} and k_{c2}, possible. However, the critical fields associated with these wavevectors, $B_{c1} = B(k_{c1})$ and $B_{c2} = B(k_{c2})$, are never equal, except in the limit $R_M \to \infty$, where $k_{c1} = -k_{c2}$ and the case of Sec. 4.2 is reached. For $R_M \lesssim R_B, R_E$ there is only one minimum solution of Eq. (22), which tends for smaller R_M to the solution of Sec. 4.1. Thus, for a given set of material parameters there is always one definite instability at a minimum B_c , and never a competition between instabilities of different k_c.

4.4 Additional remarks

Finally we will explore the possibility of an oscillatory instability. If we assume that the film compressional modulus, $\tilde{\varepsilon}$, and the longitudinal elastic modulus $c_{\parallel}$ and viscosity $\nu_{\parallel}$ can be neglected (incompressible film), it is straightforward to show that the curve of marginal stability, $B = B(k,\omega)$ has its minimum at $\omega_0 = 0$, and thus any oscillatory state would have a higher threshold than the stationary one. In the general case, the proof of the non-existence of an oscillatory instability is much more involved. One can show (under the proviso that $\nu_{\perp} + \nu_b k^2$ and $\nu_{\parallel}$ are of the same order of magnitude) that there is no finite frequency possible if $\tilde{\epsilon} k^2 \leq \sqrt{3}(\tilde{\gamma} k^2 + \rho g + c_b k^4)$. In the opposite case the threshold of an oscillatory instability (if it exists) is higher than that for the stationary one.

If the densities of the two bulk fluids above and below the film or membrane are identical, their gravitational influence on the interface undulations cancels. The thin film itself is not sensitive to gravity, since its volume is going to zero in the two-dimensional limit. Therefore, the gravity term is absent in the normal stress boundary condition and the linear instability criterion in the stationary case is $\mathcal{C}^{(z)} = 0$ (instead of $\tilde{\mathcal{C}}^{(z)} = 0$). The general marginal stability curve $B = B(k)$ then has a minimum for a vanishing $k_c^2 \sim \rho g \to 0$ leading to a vanishing threshold $B_c^4 \sim \rho g \to 0$ [17]. The lowest wavevector for a finite experimental set-up of horizontal dimension L, $k_c = 2\pi/L$ gives $\kappa_1 B_c^2 \approx 2\pi\tilde{\gamma}/L$, since effects of bending and surface magnetization are negligible for large L. This means there is only one surface excitation (spike) in the whole sample, governed by the (effective) surface tension. This is a very well known scenario, theoretically and experimentally [18], for ordinary ferrofluid free surfaces under strongly reduced gravity conditions.

5 Discussion

The driving force of the Rosensweig instability manifests itself in the boundary conditions, only, for ferrofluids as well as ferrogels (if magnetostriction is neglected). The question arises, how will the characteristics of the onset of the instability change, if the elastic medium itself is very thin so that it can be considered as a film or a membrane. In the present article we have addressed this question by extending previously obtained dispersion relations of surface waves at a half-space ferrogel boundary to those of the membrane surfaces. The very thin membrane is surrounded by two Newtonian fluids that can be ferrofluids with different magnetic properties. Possible generalizations to viscoelastic surrounding fluids and to viscoelastic (rather than elastic) membranes have been sketched. The magnetic film itself can be either a superparamagnetic isotropic magnetic gel, or an anisotropic ferromagnetic one having a finite intrinsic magnetization.

Apart from the material properties of the surrounding fluids, the derivation of dispersion relations in thin films makes use of certain effective (frequency and wavevector dependent) surface material parameters that describe the internal film properties. For surface waves an effective elastic surface modulus is introduced that contains the intralayer elastic and viscous properties. In the same manner we introduce in our discussion an effective surface permeability for the magnetic film describing the induced or permanent magnetic film properties, which generally are different from the bulk quantities. In recent experiments [19] this kind of difference between bulk and surface behavior in the magnetic properties has been seen when spin coating a ferrofluid.

In our discussion we have restricted ourselves to modes where the upper and the lower surface of the membrane move in phase, resulting in a buckled membrane of constant thickness (in linear approximation). This is complementary to a previous discussion of films of finite thickness, where just peristaltic motions where taken into account [20]. For superparamagnetic films we get two different additional contributions to the dispersion relation. One is due to the magnetic asymmetry between the surrounding liquids. This contribution is of the same character as the magnetic part of surface waves in the half-space case and vanishes in the symmetric case (no magnetic contrast between the two surrounding fluids). The second contribution comes from the magnetizability of the thin film itself. This last contribution, however, acts always stabilizing and effectively stiffens the membrane. Thus, a (symmetric) superparamagnetic membrane in air, for instance, will never become unstable to undulations. An intuitive reason for this is the fact, that in the symmetric case the magnetic field is not distorted in the limit of an infinitely thin membrane even if the membrane itself is subject to small perturbations. As a result, no destabilizing force acts on the magnetic dipoles in the film. In the present discussion we therefore focus on the case of high magnetic contrast between the surrounding fluids discussing the influence of the surface elastic properties to the characteristics of the Rosensweig instability. Due to the elastic and bending elastic surface properties, the characteristic mode at onset is shifted to higher wavelengths and the critical magnetic field towards higher field strengths. We can distinguish the limiting cases of a bending dominated regime and the regime where surface elasticity plays the important role.

For an anisotropic magnetic thin film or membrane, its permanent magnetization can lead to the Rosensweig instability, if the applied field is strong enough and oriented antiparallel to it. In this case the magnetic asymmetry between the surrounding liquids is not needed and such a magnetic film surrounded by air can become unstable.

Finally, the general case of an anisotropic magnetic membrane separating two liquids of different magnetic properties has been discussed. In principle, there is a competition between the previously discussed instability mechanisms (either based on the magnetic contrast or on the permanent film magnetization), which generally occur at a different wavelength. However, it turns out that such a pattern competition does not occur in the system under consideration, because the critical magnetic field according to one of the mechanisms is always smaller than the other one. Only in the limiting case of infinitely high intrinsic magnetization (infinitely low magnetic contrast) both critical fields can be equal. In this case, however, the different characteristic modes at onset are of the same magnitude, but of opposite sign, and no competition of two different spatial modes arises.

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Appendix A. Magnetic fields and potentials

Appendix A.1 The superparamagnetic case

In this Appendix we derive the magnetic fields for the geometry depicted in Fig. 1, to get the coefficient κ_1, Eq. (7), used in the discussions above.

Having in mind that we are interested in the limit $kd \to 0$, we assume the two boundaries at $z = -d/2$ and $z = +d/2$ to be distorted in-phase from their initially flat position by $\xi_\pm = \xi \equiv \xi_0 \exp i(\omega t - kx)$. We only consider the magnetostatic limit, since the surface wave frequencies involved are much smaller than the electrodynamic ones. The magnetizations are assumed to follow instantaneously (on the time scale of the surface waves) the external fields. This leads to the Laplace equation for the magnetic potentials of the field-distortions $\boldsymbol{h}^{(i)} = -\boldsymbol{\nabla}\varphi^{(i)}$ from the initially homogenous fields $\boldsymbol{H}^{(i)}_{\rm hom} = \boldsymbol{B}/\mu_i$ in the three regions $i = \{a, m, b\}$

$$\Delta\varphi^{(i)} = 0 \qquad \text{(A.1)}$$

The solution of these three equations can be written as

$$\varphi^{(a)} = \hat\varphi^{(a)}\xi e^{-kz}, \qquad \text{(A.2)}$$
$$\varphi^{(m)} = \hat\varphi^{(m)}_a \xi e^{kz}, +\hat\varphi^{(m)}_b \xi e^{-kz} \qquad \text{(A.3)}$$
$$\varphi^{(b)} = \hat\varphi^{(b)}\xi e^{kz}, \qquad \text{(A.4)}$$

defining the amplitudes $\hat\varphi^{(a)}$, $\hat\varphi^{(m)}_a$, $\hat\varphi^{(m)}_b$, and $\hat\varphi^{(b)}$ with $k^2 = k_x^2 + k_y^2$. These functions have to fulfill the usual magnetic boundary conditions, therefore the amplitudes are related to the external magnetic field strength B by

$$\hat\varphi^{(a)} = \frac{\mu_b - \mu_a}{\mu_b + \mu_a}\,\frac{B}{\mu_a}, \qquad \text{(A.5)}$$
$$\hat\varphi^{(b)} = \frac{\mu_a - \mu_b}{\mu_b + \mu_a}\,\frac{B}{\mu_b}, \qquad \text{(A.6)}$$

where the limit $kd \to 0$ has already been taken. The magnetic contributions to the stress tensor

$$\sigma_{\alpha\beta} = -\frac{1}{2}\boldsymbol{B}\cdot\boldsymbol{H}\,\delta_{\alpha\beta} + \frac{1}{2}(B_\alpha H_\beta + B_\beta H_\alpha) \qquad \text{(A.7)}$$

enter the l.h.s. of the normal stress boundary condition (4) as

$$\sigma^{(a)}_{zz} - \sigma^{(b)}_{zz} = -B\,\nabla_z(\varphi^{(a)} - \varphi^{(b)}) \qquad \text{(A.8)}$$
$$= B^2 k\frac{(\mu_a - \mu_b)^2}{\mu_a\mu_b(\mu_a + \mu_b)}\xi, \qquad \text{(A.9)}$$

which immediately leads to the magnetic contribution in Eq. (6) with the coefficient κ_1 given in Eq. (7).

As we take the limit towards infinitely thin films, the magnetization $\boldsymbol{M}'$ in the membrane becomes a density per unit area. We therefore introduce the effective surface permeability of the infinitely thin film μ' within the same framework as done for the in-plane elastic moduli in Ref. [10]. It is then useful to take

$$\boldsymbol{H}' = \frac{1}{\mu'}\boldsymbol{B}, \qquad \text{(A.10)}$$

with $\boldsymbol{H}' = \boldsymbol{H}^{(m)}d$ and the effective permeability of the membrane μ' given by $\mu' = \mu_m/d$. Obviously, this surface quantity has a dimension different from the bulk one.

Expanding the magnetic potential in the membrane $\varphi^{(m)}$ (at $z = 0$) in terms of the film thickness d, gives

$$\varphi^{(m)} = \frac{\mu_a + \mu_b - 2\mu_m}{\mu_a + \mu_b}\,\frac{B}{\mu_m}\xi + \mathcal{O}(d). \qquad \text{(A.11)}$$

Substituting (A.11) into the expression of the membrane stress tensor $\sigma^{(m)}_{\alpha\beta}$ yields, after integration over the film thickness and substitution of the expression for the effective membrane permeability, a distortion linear in ξ to the effective film stress tensor $\sigma^{(m)}_{\alpha\beta}$

$$\sigma^{(m)}_{z\beta} = iB^2\mu'^{-1}k_\beta\xi + \mathcal{O}(d), \qquad \text{(A.12)}$$

Taking the divergence of $\sigma^{(m)}_{z\beta}$ results in the source of normal stress due to the presence of the magnetic membrane in the effective boundary condition (4)

$$\nabla_\beta \sigma^{(m)}_{z\beta} = B^2 k^2 \mu'^{-1} \xi. \qquad (A.13)$$

This contribution is always stabilizing. For the case of a large magnetic contrast between the fluids a and b the contribution (A.9) dominates in the limit of vanishing kd and is therefore used in the main text.

Appendix A.2 The permanent-magnetic case

Things slightly change, when assuming a membrane made of an anisotropic magnetic gel. Think of the intrinsic permanent magnetization $\boldsymbol{M}_0$ to be oriented always antiparallel to the normal vector of the membrane. We assume the same geometry as done for the paramagnetic case (see Fig. 1). However, the magnetization in the membrane material is fixed and shall not change its magnitude while applying an external magnetic field.

The ground state for the unperturbed flat case with an intrinsic membrane magnetization M_0 and an externally applied field B is given for the surrounding media a and b by ($i \in \{a, b\}$)

$$B^{(i)}_z = B - M_0, \qquad (A.14)$$

$$H^{(i)}_z = \frac{1}{\mu_i} B - \frac{1}{\mu_i} M_0, \qquad (A.15)$$

$$M^{(i)}_z = \left(1 - \frac{1}{\mu_i}\right)(B - M_0), \qquad (A.16)$$

while the situation in the film is defined by

$$B_z = B - M_0, \qquad (A.17)$$

$$H_z = B. \qquad (A.18)$$

The intrinsic magnetization $\boldsymbol{M}_0$ is anchored rigidly to the membrane, therefore while deforming the film, the magnetization follows as

$$\boldsymbol{M} = -M_0 \boldsymbol{e}_z + M_0 \boldsymbol{\nabla} \xi \qquad (A.19)$$

The magnetic flux density $\boldsymbol{B}^{(m)}$ in the membrane is then given by

$$\boldsymbol{B}^{(m)} = -M_0 \boldsymbol{e}_z + M_0 \boldsymbol{\nabla} \xi + \boldsymbol{H}^{(m)} + B \boldsymbol{e}_z. \qquad (A.20)$$

We can split the field $\boldsymbol{H}^{(m)}$ in the membrane again into a constant undisturbed part, given by Eq. (A.18), and a part proportional to the surface deflection ξ. Due to the latter part the static Maxwell equations can be fulfilled, which correspond to the following Poisson equation for the potential $\varphi^{(m)}$ defined by $\boldsymbol{h}^{(m)} = -\nabla \varphi^{(m)}$

$$\Delta \varphi^{(m)} = M_0 \Delta \xi, \qquad (A.21)$$

whose general solution is given by

$$\varphi^{(m)} = \hat{\varphi}_a e^{kz} \xi + \hat{\varphi}_b e^{-kz} \xi + M_0 \xi. \qquad (A.22)$$

For the distortions in the bulk fluids a and b we assume the same structure as in the superparamagnetic case, fulfilling the Laplace equation. Matching the field disturbances according to the magnetic boundary conditions at the two surfaces $z = -d/2$ and $z = d/2$, fixes the amplitudes of the magnetic potential in the three regions.

When performing the limit towards thin films, we have to consider a permanent magnetization with respect to unit area M_0' rather than with respect to unit volume. Both quantities are related by $M_0 = M_0'/d$ when assuming a homogeneously magnetized bulk material. However, for the actual calculations it is convenient to introduce an effective membrane permeability μ_0'. Due to the definition of M_0', this effective membrane permeability is given by $\mu_0' = 1/d$. Within the limit of vanishing film thickness we then obtain

$$\varphi^{(a)} = -\frac{(B_0 - \mu_0' M_0')(\mu_a - \mu_b)}{\mu_a(\mu_a + \mu_b)} e^{-kz} \xi, \qquad (A.23)$$

$$\varphi^{(m)} = B(\mu_0')^{-1} \xi, \qquad (A.24)$$

$$\varphi^{(b)} = \frac{(B_0 - \mu_0' M_0')(\mu_a - \mu_b)}{\mu_b(\mu_a + \mu_b)} e^{kz} \xi, \qquad (A.25)$$

Evaluation of the right hand side of the boundary condition (4) in case of a permanent magnetic film material leads to

$$\nabla_\beta \sigma^{(m)}_{\alpha\beta} = (B - \mu_0' M_0') B (\mu_0')^{-1} k^2 \xi. \qquad (A.26)$$

In the limit $d \to 0$ this expression simplifies to $-M_0' B k^2 \xi$, which has been used in the main text, Eqs. (8,9).

The usual magnetic normal stress difference arising from the left hand side of Eq. (4) turns out to be

$$\sigma^{(a)}_{zz} - \sigma^{(b)}_{zz} = (B_0 - \mu_0' M_0')^2 k \frac{(\mu_a - \mu_b)^2}{\mu_a \mu_b (\mu_a + \mu_b)} \xi, \qquad (A.27)$$

which again is only present in case of a magnetic contrast between the media a and b. Furthermore it is essentially the same contribution as in Eq. (A.9) with the combined field $B_0 - \mu_0' M_0'$.

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