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Surface Waves and Rosensweig Instability in Isotropic Ferrogels

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We derive the dispersion relation of surface waves for isotropic magnetic gels 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 ferrofluids by taking into account elasticity, additionally.

1. Introduction and results

Surface undulations of the free surface of viscous liquids are known to be able to propagate as gravity or capillary waves. In more complex systems like viscoelastic liquids or gels the transient or permanent elasticity allows for modified transverse elastic waves at free surfaces [1]. They are excited e.g. by thermal fluctuations or by imposed temperature patterns on the surface. In ferrofluids, colloidal solutions of magnetic nanoparticles in a carrier fluid, magnetic stresses at the surface come into play. In particular, in an external magnetic field normal to the surface there is a focusing effect on the magnetization at the wave crests of an undulating surface with the tendency to increase the undulations [2]. At a critical field strength no wave propagation is possible and the surface becomes unstable with respect to a stationary pattern of surface spikes (Rosensweig or normal field instability). Here, we combine the two aspects of elasticity and superparamagnetic response by dealing with (isotropic) ferrogels, a crosslinked polymer network swollen with a ferrofluid [3]. Using linearized dynamic equations and boundary conditions we get the general surface wave dispersion relation for ferrogels

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(in a normal external field), which contains as special cases those for ferrofluids and non-magnetic gels and can be generalized to viscoelastic ferrofluids and magnetorheological fluids. A linear stability analysis reveals the threshold condition, above which stationary surface spikes grow. This critical field depends on gravity, surface tension and on the elastic (shear) modulus of the gel, while the critical wavelength of the emerging spike pattern is independent of the latter. As in the case of ferrofluids neither the threshold nor the critical wavelength depends on the viscosity [4].

2. Model and basic equations

The complete set of dynamic equations describing isotropic ferrogels was given by *E. Jarkova et al.* [5] using the method of generalized hydrodynamics (for uniaxial ferrogels [6] cf. [7]). There are various reversible and irreversible dynamic crosscouplings between flow, elasticity and magnetization. For simplicity we only keep those of them, which are presumably the relevant ones for the present problem. In particular, we keep the magnetic Maxwell and the elastic and viscous contributions to the stress tensor

$$T_{ij} = \delta_{ij}(p + \frac{1}{2}\mathbf{B}\cdot\mathbf{H}) - \frac{1}{2}(B_i H_j + B_j H_i) - 2\mu_2 \epsilon_{ij} - \nu_2(\nabla_i v_j + \nabla_j v_i) \quad (1)$$

where p is the pressure, ϵ_{ij} is the strain tensor, $\boldsymbol{v}$ the velocity field, and $\boldsymbol{H}$ and $\boldsymbol{B}$ are the magnetic field and induction, respectively. The magnetization $\boldsymbol{M} \equiv \boldsymbol{B} - \boldsymbol{H}$ is assumed to have relaxed to its static value given by the magnetic field and magnetostatics, $\text{div}\boldsymbol{B} = 0$ and $\text{curl}\boldsymbol{H} = 0$, can be applied. (We use the rational Gaussian or Heaviside system of units [8].) Global incompressibility, $\text{div}\boldsymbol{v} = 0$, and incompressibility of the gel network, $\epsilon_{kk} = 0$, is employed and only the shear elastic modulus μ_2 and the shear viscosity ν_2 enter the stress tensor.

Neglecting the thermal degree of freedom, magnetostriction, and taking the simplest form for the dynamics of the elasticity we are left with the linearized dynamic equations

$$\frac{\partial}{\partial t}\rho v_i + \nabla_j T_{ij} = -\rho g \delta_{iz} \quad (2)$$

$$\frac{\partial}{\partial t}\epsilon_{ij} - \frac{1}{2}(\nabla_i v_j + \nabla_j v_i) = 0\,. \quad (3)$$

where the gravitational force ($\sim g$) is acting along the negative z axis.

Indeed we model our system by an originally flat surface $z = 0$ dividing the magnetic gel ($z < 0$) from vacuum ($z > 0$), cf. Fig.1, where the applied external field ($\boldsymbol{B}^{vac} = B_0\boldsymbol{e}_z = \boldsymbol{H}^{vac}$) is normal to the surface. The dispersion relation of periodic undulations, described by the surface displacement $\xi(x, y, t)$, is derived, and its stability investigated in the following.

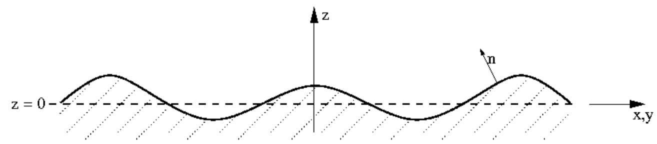

Figure 1: Small periodic perturbations $\xi(x, y, t)$ of the flat surface $z = 0$ between the ferrogel of susceptibility χ in the lower half space and vacuum in the upper half space.

To do this we need boundary conditions for our dynamic variables. First, there are the magnetic ones [8], the mechanical ones guaranteeing a stress-free surface, and the (linearized) kinematic one

$$\boldsymbol{n} \times \boldsymbol{H} = \boldsymbol{n} \times \boldsymbol{H}^{vac} \qquad \boldsymbol{n} \cdot \boldsymbol{B} = \boldsymbol{n} \cdot \boldsymbol{B}^{vac} \tag{4}$$

$$\boldsymbol{n} \times \mathsf{T}\cdot\boldsymbol{n} = \boldsymbol{n} \times \mathsf{T}^{vac}\cdot\boldsymbol{n} \tag{5}$$

$$\boldsymbol{n}\cdot\mathsf{T}\cdot\boldsymbol{n} - \boldsymbol{n}\cdot\mathsf{T}^{vac}\cdot\boldsymbol{n} = \rho\, g\, \xi + \sigma\, \mathrm{div}\boldsymbol{n} \tag{6}$$

$$\frac{\partial}{\partial t}\xi = v_z \tag{7}$$

all taken at the surface. The unit vector $\boldsymbol{n}$ is the surface normal, $\boldsymbol{n} = \boldsymbol{\nabla}(z - \xi)/|\boldsymbol{\nabla}(z - \xi)|$, and div$\boldsymbol{n}$ is twice the mean curvature. The vacuum stresses (superscript *vac*) are solely due to the magnetic fields (vacuum Maxwell stress tensor). The normal stress difference between the magnetic gel and the vacuum is balanced by gravity and the Laplace stress due to curvature of the surface and the surface tension σ.

3. Linear deviations from the ground state

The system of equations and boundary conditions (1-7) always has the trivial solution (ground state), where the surface is flat ($\xi = 0$, $\boldsymbol{n}_0 = \boldsymbol{e}_z$), flow and deformations are absent ($\boldsymbol{v} = 0$, $\epsilon_{ij} = 0$), and the fields are constant ($\boldsymbol{M}_0 = M_0\boldsymbol{e}_z$ with $M_0 = (1 - 1/\mu)B_0$, where μ is magnetic permeability). The boundary condition (6) requires a non-zero, constant stress contribution, $p_0 = -(1/2)(1 - 1/\mu)B_0^2$, which is of minor relevance, since in an incompressible system the pressure has no physical meaning anymore and merely serves as an auxiliary quantity that guarantees div$\boldsymbol{v} = 0$ for all times, if flow is present.

We now allow for periodic surface undulations with frequency $\omega = \omega_0 - i\lambda$ (ω_0 and λ real) and wavevector $\boldsymbol{k} = (k_x, k_y, 0)$

$$\xi(x, y, t) = \hat{\xi} e^{-ik_x x - ik_y y + i\omega t}\,. \tag{8}$$

describing propagating and damped surface waves. For $\omega = 0$ a stationary spatial pattern is obtained. Generally ω is a complex function of $\boldsymbol{k}$. In a linear theory the amplitude $\hat{\xi}$ is undetermined, Fourier modes of the type (8) can be superimposed as appropriate, and deviations from the ground state of all the other variables have to be proportional to $\xi(x, y, t)$. Linear deviations of the surface normal from the ground state due to undulations are given by $\boldsymbol{n}_1 \equiv \boldsymbol{n} - \boldsymbol{n}_0 = (-\nabla_x \xi, -\nabla_y \xi, 0)$.

The linear deviations of the magnetic field and induction from the ground state value, $\boldsymbol{b} \equiv \boldsymbol{B} - \boldsymbol{B}_0$ and $\boldsymbol{h} = \boldsymbol{H} - \boldsymbol{H}_0$, both for the ferrogel and the vacuum (superscript *vac*), still obey the linear electrostatic equations, $\boldsymbol{b} = \mu \boldsymbol{h}$, div$\boldsymbol{b} = 0 =$ curl$\boldsymbol{h}$. This allows for the introduction of a magnetic scalar potential [8] $\boldsymbol{h} = -\boldsymbol{\nabla} \Phi_m$ that has to be a potential function, $\Delta \Phi_m = 0$ (where $\Delta = \boldsymbol{\nabla}^2$), with the appropriate solutions

$$\Phi_m = \hat{\Phi}_m \, \xi(x, y, t) e^{kz} \tag{9}$$

$$\Phi_m^{vac} = \hat{\Phi}_m^{vac} \xi(x, y, t) e^{-kz} \, . \tag{10}$$

for the lower (ferrogel) and upper (vacuum) half plane, respectively and $k^2 = k_x^2 + k_y^2$. With the help of the magnetic boundary conditions (4) the amplitudes are found to be [2]

$$\hat{\Phi}_m = -\frac{1}{\mu} \hat{\Phi}_m^{vac} = -\frac{M_0}{1 + \mu} \tag{11}$$

For the velocity we make the usual partition into an irrotational (potential) and a rotational (source-free) part $\boldsymbol{v} = \boldsymbol{v}^{pot} + \boldsymbol{v}^{rot}$ with $\boldsymbol{v}^{pot} = \boldsymbol{\nabla} \phi$ and $\boldsymbol{v}^{rot} = \boldsymbol{\nabla} \times \boldsymbol{\Psi}$. Incompressibility requires $\Delta \phi = 0$ and leads to the ansatz

$$\phi = \hat{\phi} \, \xi(x, y, t) e^{kz} \tag{12}$$

for the scalar velocity potential. The vector velocity potential can be written as

$$\boldsymbol{\Psi} = \hat{\boldsymbol{\Psi}} \, \xi(x, y, t) e^{qz} \tag{13}$$

where the amplitudes $\hat{\phi}$ and $\hat{\boldsymbol{\Psi}}$ and the decay length q^{-1} are still undetermined. Since only two of the three amplitudes $\hat{\boldsymbol{\Psi}}$ can be independent, we set $\hat{\Psi}_z = 0$ without loss of generality resulting in $\boldsymbol{v}^{rot} = (-q\Psi_y, \, q\Psi_x, \, -ik_x \Psi_y + ik_y \Psi_x)$.

The strain ϵ_{ij} can be expressed by the velocity via Eq.(3) and the linear pressure deviation, $p_1 \equiv p - p_0$ is determined by Eq.(2). The latter can be simplified considerably by the observation that the divergence of the magnetic stress tensor vanishes, $\nabla_i (\boldsymbol{H} \cdot \boldsymbol{B}) - \nabla_j (H_i B_j + H_j B_i) = 0$, in linear order. With the help of Eq.(3), $i\omega \nabla_j \epsilon_{ij} = (1/2) \Delta v_i$, Eq.(2) takes the linear form

$$i\omega \rho v_i + \nabla_i p_1 - (\nu_2 + \frac{\mu_2}{i\omega}) \Delta v_i = -\rho g \delta_{iz} . \tag{14}$$

Taking div and curl of Eq.(14) we get [1]

$$p_1 = -i\omega\rho\phi + \text{const.} \tag{15}$$

and
$$q^2 = k^2 - \frac{\rho\omega^2}{\mu_2 + i\omega\nu_2} \tag{16}$$

respectively, where the unimportant constant in the pressure can be ignored.

4. Surface wave dispersion relation

We are left with three amplitudes, $\hat{\phi}, \hat{\Psi}_x, \hat{\Psi}_y$, that have to be related to the undulation amplitude, $\hat{\xi}$ by the stress boundary conditions (5,6). Without loss of generality we can choose the in-plane wavevector $\boldsymbol{k}$ to be along the x axis ($k_y = 0$). It is easy to show that then also $\hat{\Psi}_x = 0$, which means $v_y = 0$ and $\epsilon_{yz} = 0$.

For linear deviations from the ground state the normal stress boundary condition (6) can be written as

$$p_1 + \frac{\mu}{1+\mu} M_0^2 k\xi - \sigma k^2 \xi - 2\mu_2 \epsilon_{zz} - 2\nu_2 \nabla_z v_z - \rho g \xi = 0\,. \tag{17}$$

all taken at $z = 0$. In the shear stress boundary condition all magnetic contributions cancel in linear order leading to $\epsilon_{xz} = 0$ at $z = 0$. These two conditions translate into a set of linear homogeneous algebraic equations for the amplitudes $\hat{\phi}$ and $\hat{\Psi}_y$

$$\left[\omega^2 + \frac{\mu'}{1+\mu} M_0^2 k^2 - \sigma' k^3 - gk - 2\tilde{\mu}_2(\omega) k^2\right] \hat{\phi} + \left[\frac{\mu'}{1+\mu} M_0^2 k - \sigma' k^2 - g - 2\tilde{\mu}_2(\omega) q\right] (-ik_x \hat{\Psi}_y) = 0\,. \tag{18}$$

and

$$2k^3 \hat{\phi} + \left(k^2 + q^2\right)(-ik_x \hat{\Psi}_y) = 0\,. \tag{19}$$

with the frequency dependent $\tilde{\mu}_2(\omega) \equiv \mu_2' + i\omega\nu_2'$ describing (kinematic) elasticity and viscosity, and where we used the abbreviations $\mu_2' = \mu_2/\rho$, $\nu_2' = \nu_2/\rho$, $\sigma' = \sigma/\rho$, and $\mu' = \mu/\rho$.

To have a nontrivial solution for equations (18) and (19) the determinant of coefficients must vanish. This leads to the dispersion relation of surface waves for ferrogels

$$\omega^2 \left(2\tilde{\mu}_2(\omega) k^2 - \omega^2\right) + \omega^2 \left(\sigma' k^3 + gk + 2\tilde{\mu}_2(\omega) k^2 - \frac{\mu'}{1+\mu} M_0^2 k^2\right) - 4\tilde{\mu}_2^2(\omega)\, k^4 \left[1 - \left(1 - \frac{\omega^2}{\tilde{\mu}_2(\omega) k^2}\right)^{1/2}\right] = 0\,. \tag{20}$$

In the absence of an external magnetic field ($M_0 = 0$) Eq.(20) reduces to the dispersion relation for nonmagnetic gels [1]. It also contains as a special case the surface wave dispersion relation for ferrofluids (in an external field) by choosing $\tilde{\mu}_2 = i\omega\nu'_2$. It can be generalized to viscoelastic ferrofluids, whose elasticity relaxes on a time scale τ^{-1}, by replacing μ_2 with $i\omega\tau\mu_2/(1+i\omega\tau)$ [1], and to magnetorheological fluids by allowing μ_2, ν_2, and τ being functions of the external field.

The dispersion relation (20) is very complicated and it is impossible to solve it analytically for $\omega(k)$. From non-magnetic gels it is known that there are basically three wave regimes (neglecting dissipation or damping): $\omega^2 = \sigma' k^3$ (capillary waves), $\omega^2 = \alpha\mu'_2 k^2$ (Rayleigh elastic waves), and $\omega^2 = gk$ (gravity water waves) for small wavelengths ($k \gg \mu_2/\sigma,\ \sqrt{g/\sigma'}$), intermediate ones ($g/\mu'_2 \ll k \ll \mu_2/\sigma$), and large ones ($k \ll g/\mu'_2,\ \sqrt{g/\sigma'}$), respectively, where α is a number of order unity. For typical material values ($\mu_2 \approx 1$ kPa, $\sigma \approx 0.02$ kg/sec^2) waves at wavelengths of 10^{-4} m and below (with frequencies of 50 kHz and above) are of purely capillary type, while for wavelengths above 1 m (and frequencies below 10 Hz) the gravity character dominates; this regime is, thus, irrelevant for usual ferrogel samples. In between, for typical wavelengths of 10^{-2} m and frequencies of 100 - 1000 Hz the elastic nature of the wave is prevailing. This scenario also applies to isotropic ferrogels in the absence of a field. The effect of a normal external magnetic field on the surface is a destabilizing one [2]. From Eq.(20) it is evident that an external field leads to an effective reduction of the surface stiffness (provided by surface tension, gravity or elasticity) and decreases the frequency (squared) of the propagating waves in all regimes by $\sim M_0^2 k^2$. If the field is large enough, this reduction is the dominating effect and can lead to $\omega = 0$ and thus, to the breakdown of propagating waves. In the next section we will show that this is indeed related to the Rosensweig instability.

5. Rosensweig instability

Eq.(20) can be slightly reinterpreted: It is an equation for that external field strength (or M_0), where a surface disturbance (8) with wavevector k and frequency ω_0 relaxes to zero or grows exponentially for λ negative or positive, respectively. For $\lambda = 0$ such a surface disturbance is marginally stable (or unstable) against infinitesimal disturbances, since Eq.(20) has been obtained by linearizing the dynamic equations and the boundary conditions about the ground state. The function M_0 still depends on ω_0 and k and has to be minimized with respect to these quantities in order to get the true instability threshold.

First, it can be shown analytically that for the two special cases, $\mu_2 = 0$ (ferrofluid) and $\nu_2 = 0$ (ferrorubber), only the stationary solution $\omega_0 = 0$ is possible at onset. For the general case numerical calculations show the absence of an oscillatory instability with $\omega_0 \neq 0$ at onset.

For $\omega_0 = 0$ the linear threshold condition is completely independent of ν_2 [9] and simplifies to

$$M_0^2 = \frac{1+\mu}{\mu} \left(\sigma k + \frac{\rho g}{k} + 2\mu_2 \right) . \tag{21}$$

Minimizing with respect to k leads to the critical wavevector

$$k_c = \sqrt{\frac{\rho g}{\sigma}} \tag{22}$$

and the critical field

$$M_c^2 = \frac{1+\mu}{\mu} \left(2\sqrt{\sigma \rho g} + 2\mu_2 \right) . \tag{23}$$

Obviously, k_c is identical to that in ferrofluids and not dependent on elasticity, but the critical field is enhanced by elasticity. The latter finding is no surprise, since elasticity increases the surface stiffness. For typical polymer networks with a shear elastic modulus of 1 kPa, the elastic contribution to M_c exceeds the surface tension contribution roughly by a factor of 5 and elasticity is the dominating factor. Critical values of 100 - 200 Gauss for M_0 have to be expected for typical samples. The critical wavelength is in the range of 1 cm, which for surface waves lies in the elasticity dominated regime. Thus, if the system cannot choose the optimal (critical) wavelength, but is fixed to a prescribed one like in many surface wave scattering experiments, the field necessary to destabilize the surface wave is about M_c in the elastic regime and higher in the other ones, where $k > k_c$ or $k < k_c$. For very soft gels with $\mu_2 < 10$ Pa, the influence of the elasticity is rather negligible and ordinary ferrofluid behavior is found.

The present linear stability analysis has various limitations. First, it cannot decide whether the instability is forward and continuous, or discontinuous and backward. Only in the former case the linear stability analysis gives the actual threshold. A linear treatment also cannot give the actual spatial pattern, since a linear superposition of different $\boldsymbol{k}$ orientations ($|\boldsymbol{k}| = k_c$) allows for various types of patterns. Finally, the amplitude of the spikes emerging just above threshold is undetermined by the linear stability analysis. All these questions have to be dealt with in future nonlinear considerations.

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[9] This justifies Rosensweig's procedure [2], who neglected viscosity from the beginning.