

## Amplitude Equation for the Rosensweig Instability

Stefan BOHLIUS<sup>1</sup>, Helmut R. BRAND<sup>1,2</sup>, and Harald PLEINER<sup>1</sup>

<sup>1</sup> *Max Planck Institute for Polymer Research, Mainz, Germany*

<sup>2</sup> *Theoretical Physics, University of Bayreuth, Germany*

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The Rosensweig instability is a prominent example for a surface driven instability, where the deformation of the surface is amplified by an external generalized force, the normal magnetic field, and finally settles at a spatial pattern of spike deformations. This property has so far prohibited the derivation of an amplitude equation by means of the standard weakly nonlinear analysis. Here we give a derivation of the appropriate amplitude equation based on the hydrodynamic equations and the appropriate boundary conditions. We stress the fact, that even though the final pattern does not involve flow, the system has to be treated dynamically. The observed static pattern has to be interpreted as the limiting case of a frozen-in characteristic mode. The amplitude equation finally obtained contains first, for the ferrogel case also second order, time derivatives as well as quadratic (for the hexagonal case) and cubic nonlinearities in the amplitudes.

### §1. Introduction

The normal field or Rosensweig instability<sup>1)</sup> describes the transition of an initially flat ferrofluid surface to hexagonally ordered surface spikes as soon as an applied magnetic field exceeds a certain critical value. Ferrofluids are suspensions of magnetic nanoparticles dispersed in a suitable carrier liquid. Usually they are coated by polymers or charged in order to prevent coagulation and show various distinct material properties,<sup>2)</sup> in particular, the superparamagnetic behavior in external magnetic fields featuring a very large magnetic susceptibility and a high saturation magnetization in a rather low magnetic field. Ferrogels are obtained by cross-linking a mixture of a ferrofluid and a polymer solution.<sup>3)</sup> As in usual ferrofluids, the initially flat surface of ferrogels becomes linearly unstable beyond a critical magnetic field.<sup>4)</sup>

The linear stability analysis for ferrofluids<sup>1)</sup> was based on the freezing of surface waves, which is reached, when the stabilizing forces of gravity and surface tension are compensated by the destabilizing magnetic force. The latter arises due to the focusing effect of the local magnetic field at surface fluctuations of a permeable medium. The critical magnetic field and the characteristic wavelength do not depend on hydrodynamic parameters, like the viscosity, while the linear growth behavior does.<sup>5)-7)</sup> For magnetic gels the elastic force contributes as a stabilizing effect leading to an increase of the critical magnetic field, whereas the characteristic wavelength remains unchanged.<sup>4)</sup> For thermoreversible magnetic gels,<sup>8)</sup> which are viscoelastic rather than elastic, such a threshold shift does not occur.

A nonlinear analysis of the Rosensweig instability, however, turned out to be very complicated mainly due to the fact that the instability necessarily involves a deformable surface. Early attempts<sup>9)</sup> neglected viscous effects and discussed the

convexity of the surface energy density, including gravitational, surface tension, and magnetic energy. For a hexagonal spike pattern a transcritical instability was found, which becomes unstable to a quadratic pattern above a second threshold. Both transitions are accompanied by hysteretic regions. However, this method only works for (unrealistically) small magnetic susceptibilities.<sup>10)</sup> This energy method was extended<sup>11)</sup> to the Rosensweig instability in isotropic magnetic gels considering additionally the elastic surface energy density. The results are qualitatively similar to the ferrofluid case and the method faces similar restrictions. Of course, it cannot describe growth rates, since dissipative processes are ignored from the beginning. Another approach considering using a static description is given in Ref.,<sup>12)</sup> where only the normal stress boundary condition is considered for the nonlinear expansion.

The dynamics of the system has first been taken into account in Ref.<sup>13)</sup> postulating a Swift-Hohenberg model to describe fronts between hexagons and squares. This approach, however, lacks the connection to the material properties of the medium. What one would like to have is a systematic nonlinear expansion of the basic hydrodynamic equations in analogy to Ref.<sup>14)</sup> To use this method for the Rosensweig instability, the adjoint linear eigenvectors in the presence of a deformable surface have to be known (to apply Fredholm's theorem). To circumvent Fredholm's theorem, procedures restricted to potential flows were proposed,<sup>15),16)</sup> sacrificing the condition of a shear stress free boundary. Recently, the adjoint system of linear eigenvectors for the Rosensweig instability in isotropic magnetic gels in the presence of deformable surfaces was given by the present authors<sup>17)</sup> thus paving the way for accessing the nonlinear regime via a weakly nonlinear analysis.

In this communication we exhibit the starting hydrodynamic equations for ferrofluids and isotropic ferrogels and comment on the important and non-trivial intermediate steps that lead to the amplitude equations, which is discussed in more detail, finally. This work is based on the PhD thesis of S. Bohlius<sup>18)</sup> and Ref.<sup>19)</sup>

## §2. Basic equations and general approach

For the discussion of the Rosensweig instability in magnetic gels we use as starting point the hydrodynamic and magnetostatic equations derived for isotropic magnetic gels in Ref.<sup>20)</sup> They comprise the Navier-Stokes equations with  $T_{ij}$ , the stress tensor as current and gravity as external force, the balance of dynamic elasticity and flow

$$(\partial_t + v_k \partial_k) \epsilon_{ij} = \frac{1}{2} (\partial_i v_j + \partial_j v_i) - \epsilon_{kj} \partial_i v_k - \epsilon_{ki} \partial_j v_k \quad (2.1)$$

where  $\epsilon_{ij}$  is the (Eulerian) strain tensor and  $v_i$  the velocity field, and  $\partial_i B_i = 0$  and  $\epsilon_{ijk} \partial_j H_k = 0$ , where  $\mathbf{B}$  and  $\mathbf{H}$  are the magnetic induction and the magnetic field, respectively, and the magnetization is defined as usual by  $\mathbf{M} = \mathbf{B} - \mathbf{H}$ . Mass conservation will be replaced at the end by incompressibility of the total mass  $\rho = const.$  (and of the network  $\epsilon_{ii} = 0$ ). We completely neglect the thermal degree of freedom. We assume the magnetization to relax fast enough on the time scale considered in our discussion of the Rosensweig instability justifying the magnetostatic approximation. In addition, we neglect magnetostrictive effects as well as nonlinear elastic stresses

and strains. Although certainly present in polymeric systems, for a first treatment they seem to be less important. In the same spirit the permeability  $\mu$ , defined by  $B_i = \mu H_i$ , is taken to be constant (for the given value of the magnetic field) and all other material parameters are taken to be independent of the magnetic field. This leads to the stress tensor

$$T_{ij} = g_i v_j + p \delta_{ij} - B_i H_j + \frac{1}{2} B_k H_k \delta_{ij} - \mu_2 (\epsilon_{jkl} \epsilon_{kli} + \epsilon_{ikl} \epsilon_{klj}) - 2\mu_2 \epsilon_{ij} - \nu_2 (\partial_j v_i + \partial_i v_j) \quad (2.2)$$

with  $p$  the pressure, and  $\mu_2$  and  $\nu_2$  the shear elastic modulus and shear viscosity, respectively. The vacuum stresses are solely due to the magnetic field hence the stress tensor there reduces to the known vacuum Maxwell stress tensor<sup>21)</sup>  $\mathbf{T}^{\text{vac}}$ .

The hydrodynamic and magnetic bulk equations are supplemented by boundary conditions at the deformable surface defined by  $z = \xi$ . Aside from the usual magnetic boundary conditions the tangential components of the mechanical stress between the magnetic medium and the vacuum above is required to vanish at  $z = \xi$ , while the normal stress difference is balanced by gravity and surface tension.

$$\mathbf{n} \times \mathbf{T} \cdot \mathbf{n} = \mathbf{n} \times \mathbf{T}^{\text{vac}} \cdot \mathbf{n} \quad (2.3)$$

$$\mathbf{n} \cdot \mathbf{T} \cdot \mathbf{n} - \mathbf{n} \cdot \mathbf{T}^{\text{vac}} \cdot \mathbf{n} = \sigma_T \text{div} \mathbf{n} - \rho g \xi \quad (2.4)$$

$$\mathbf{n} \times \mathbf{H} = \mathbf{n} \times \mathbf{H}^{\text{vac}} \quad (2.5)$$

$$\mathbf{n} \cdot \mathbf{B} = \mathbf{n} \cdot \mathbf{B}^{\text{vac}} \quad (2.6)$$

with  $\mathbf{G} = -g \mathbf{e}_z$ , the acceleration due to gravity along the negative z-axis, and  $\sigma_T$ , the surface tension coefficient at the free boundary. Further,  $\mathbf{H}^{\text{vac}}$  and  $\mathbf{B}^{\text{vac}}$  denote the magnetic field and the magnetic flux density in the vacuum, respectively, and the surface normal is  $\mathbf{n} = \partial(z - \xi) / |\partial(z - \xi)|$ . Additionally, we have to consider the kinematic boundary condition

$$(\partial_t + \mathbf{v}_\perp \cdot \partial_\perp) \xi = v_z \quad (2.7)$$

modeling the dynamics of the deformable surface at  $z = \xi$ .

### §3. Procedure

For the weakly nonlinear analysis of the stationary state above the linear threshold, we have to expand the macroscopic variables in terms of  $\epsilon$ , the normalized difference of the actual applied magnetic field to the critical one

$$\{p, \mathbf{B}, \mathbf{H}, \mathbf{M}\} = \{p_0, \mathbf{B}_c, \mathbf{H}_c, \mathbf{M}_c\} + \epsilon \{p^{(1)}, \mathbf{B}^{(1)}, \mathbf{H}^{(1)}, \mathbf{M}^{(1)}\} + \dots \quad (3.1)$$

$$\{\mathbf{v}, \epsilon_{ij}, \xi\} = 0 + \epsilon \{\mathbf{v}^{(1)}, \epsilon_{ij}^{(1)}, \xi^{(1)}\} + \dots \quad (3.2)$$

The magnetic field, however, is an externally given parameter acting as the control parameter, the series expansion of  $\mathbf{H}$  can therefore be reinterpreted as the definition of  $\epsilon$ .

Writing this expansion as  $|\Psi\rangle = |\Psi^{(0)}\rangle + \epsilon |\Psi^{(1)}\rangle + \epsilon^2 |\Psi^{(2)}\rangle + \dots$  with  $|\Psi\rangle = (v_x, v_y, v_z, p, \epsilon_{xx}, \epsilon_{xy}, \dots)$  the fundamental hydrodynamic equations take the form

$$\mathcal{L}_0 |\Psi^{(1)}\rangle = 0 \quad (3.3)$$

$$\mathcal{L}_0 |\Psi^{(2)}\rangle = -\mathcal{L}_1 |\Psi^{(1)}\rangle + \mathcal{N}(\Psi^{(1)}, \Psi^{(1)}) \quad (3.4)$$

$$\mathcal{L}_0 |\Psi^{(3)}\rangle = \dots \quad (3.5)$$

where the first order renders the results for the linear critical threshold  $\mu M_c^2 = 2(1 + \mu)(\sqrt{\sigma_T \rho g} + \mu_2)$ .<sup>4)</sup> This instability can be interpreted as the breakdown of surface waves ( $\omega = 0$ ) at the critical wave vector  $k_c^2 = \rho g / \sigma_T$ . The unstable mode  $\xi = \hat{\xi} \exp(i\Phi)$  is characterized by an undetermined amplitude  $\hat{\xi}$  of the surface deflection and a phase  $\Phi = \omega t - kx$  (with an arbitrary transverse direction  $x$ ), to which all the other variables are proportional. In addition, the bulk variables decay into the material by either  $\exp(-kz)$  or  $\exp(-qz)$ , for longitudinal and transverse variables, respectively. The wave numbers  $k$  and  $q = q(k, \omega)$  are defined by the dispersion relation  $\omega = \omega(k)$  obtained from the normal stress boundary condition.

For the higher orders there exists a solution only, if the inhomogeneous parts are orthogonal to the first order (homogeneous) solution (Fredholm's theorem). In second order the following identity must hold

$$\langle \Psi^{(1)} | \mathcal{N}(\Psi^{(1)}, \Psi^{(1)}) \rangle - \langle \Psi^{(1)} | \mathcal{L}_1 \Psi^{(1)} \rangle = 0 \quad (3.6)$$

where  $\langle \dots \rangle$  denotes the conventional integral over space and time. To make use of that condition, however, the adjoint eigenvectors  $\langle \Psi^{(1)} |$  have to be known

$$\langle \Psi^{(1)} | \mathcal{L}_0^\dagger = 0 \quad (3.7)$$

with  $\mathcal{L}_0^\dagger$  denoting the adjoint linear operator. This has been achieved in Ref.<sup>17)</sup> using a full dynamic description (because of the kinematic boundary condition), compressibility (to preserve the explicit symmetry of the stress tensor), and taking into account the adjoint boundary conditions as well. As a result, right traveling surface waves transform into left traveling waves in the adjoint space and vice versa. For the explicit first order expressions of the variables and their adjoints see Ref.<sup>17)</sup>

The most general ansatz as a starting point to solve the equations in higher  $\epsilon$  order is to superimpose  $N$  characteristic modes, e.g. linear eigenvectors, with different orientations. Each of these modes  $n$  consists of a right and left traveling wave (subscripts  $R$  and  $L$ , respectively), e.g.

$$\xi^{(1)} = \sum_n^N \xi_n \equiv \sum_n^N (\xi_{nR} + \xi_{nL} + c.c.) \equiv \sum_n^N (\hat{\xi}_{nR} e^{i\omega_n t - i\mathbf{k}_n \cdot \mathbf{r}} + \hat{\xi}_{nL} e^{-i\omega_n t - i\mathbf{k}_n \cdot \mathbf{r}} + c.c.) \quad (3.8)$$

where c.c. (or an asterisk) means the complex conjugate and  $\mathbf{k}_n$  characterizes the direction of the  $n$ -th mode. The wave number  $k = |\mathbf{k}| = |\mathbf{k}_n|$  is the same for all modes. As can be expected from symmetry considerations, three patterns are important, hexagons, squares and rolls (or stripes). They are described by six critical wave vectors, a set of three ( $\xi_1 = \xi_2 = \xi_3$ ) with the  $\mathbf{k}_n$   $120^\circ$  apart from each other, and another set of three ( $n = 4, 5, 6$ ) rotated by  $90^\circ$  with respect to the first one.

While performing a weakly nonlinear analysis, we have to specify the scales in space and time. In a first approach we will assume a surface pattern that arises homogeneously in space, which allows us, not to rescale the spatial degrees of freedom.

Time, however, will be rescaled in the following manner

$$t^{(1)} = \epsilon t \quad \text{and} \quad t^{(2)} = \epsilon^2 t \quad (3.9)$$

which will lead to the substitution for the time derivative

$$\partial_t \longrightarrow \partial_t^{(0)} + \epsilon \partial_t^{(1)} + \epsilon^2 \partial_t^{(2)} + \dots \quad (3.10)$$

This scaling in time means the dynamics of the amplitudes to take place on the slower time scales,  $\xi_{n,\{R,L\}} \rightarrow \xi_{n,\{R,L\}}(t^{(1)}, t^{(2)}, \dots)$ .

Generally, the procedure of obtaining the nonlinear amplitude equation is then straightforward, but rather cumbersome: Solving Fredholm's condition in second order (3.6) allows for determining the second order eigenfunctions, which are needed to get Fredholm's condition in third order. The latter is then combined with the second order one (if that is not trivially fulfilled, anyhow) making use of the scaling (3.9) to give the final amplitude equation for the nonlinear surface deflection. However, in the present case the matter is even more complicated. Since the bulk equations do not contain the control parameter, Fredholm's conditions do not lead to the desired relation between the amplitude and the magnetic field or magnetization. Instead, one has to fulfill additionally the normal stress boundary conditions in second and third order (in first order the linear dispersion relation is obtained). These boundary conditions consist of two parts each. One is proportional to higher harmonics of the fundamental mode and merely add to the hydrostatic pressure in the medium. The other one is proportional to the fundamental mode and serves as an additional solvability condition providing the connection between the growth rate and the external control parameter. They have to be combined in a reasonable way with Fredholm's conditions leading finally to the amplitude equation. Unfortunately, the normal stress boundary condition requires the knowledge of (at least some parts) of the third order eigenfunctions making the calculations even more involved.

This whole procedure has been carried out in detail in the PhD thesis of S. Bohlius<sup>18)</sup> and in Ref.<sup>19)</sup> and will not be repeated here. Only some important steps will be commented upon and the final result will be discussed.

#### §4. Amplitude equation

The second order solvability condition (3.6) explicitly reads

$$\begin{aligned} \langle \bar{v}_i | -\rho \partial_t^{(1)} v_i^{(1)} - \partial_j (\rho v_i^{(1)} v_j^{(1)} - 2\mu_2 \epsilon_{jk}^{(1)} \epsilon_{ki}^{(1)}) \rangle \\ + \langle \bar{\epsilon}_{ij} | -\partial_t^{(1)} \epsilon_{ij}^{(1)} - v_k^{(1)} \partial_k \epsilon_{ij}^{(1)} \rangle = 0 \end{aligned} \quad (4.1)$$

where the barred functions are the adjoint ones. For a hexagonal lattice this implies a nonlinear relation among the amplitudes ( $\hat{\xi}_n \equiv \hat{\xi}_{nR} = \hat{\xi}_{nL}$  for  $n = 1, 2, 3$ )

$$\sigma^{(1)} \hat{\xi}_1 = -\sigma^{(0)} k_c \hat{\xi}_2^* \hat{\xi}_3^* \quad \text{and} \quad |\hat{\xi}_1|^2 = |\hat{\xi}_2|^2 = |\hat{\xi}_3|^2 \quad (4.2)$$

for all cyclic permutations  $1 \rightarrow 2 \rightarrow 3 \rightarrow 1$  and complex conjugates. Here, the time derivatives  $\partial_t^{(i)} \equiv i\omega^{(i)} + \sigma^{(i)}$  have been Fourier transformed. Equation (4.2)

shows, that the  $\sigma^{(1)}$  scales in the bulk with  $\sigma^{(0)}$ , indicating that  $\sigma^{(1)}/\sigma^{(0)}$  stays finite in the stationary limit. This ratio will be used as a dimensionless time derivative  $\tilde{\partial}_T^{(1)} = \sigma^{(1)}/\sigma^{(0)}$  for the bulk hydrodynamic equations (in second order).

For other regular patterns, quadratic or stripes, Eq.(4.1) does not lead to any relation between the amplitudes.

As discussed above, we need the normal stress boundary condition to relate the control parameter  $M^{(1)}$  to the appropriate growth rate. As a result one finds for all types of lattices

$$\sigma^{(1)} = \frac{\mu M^{(1)} M_c}{\nu_2(1+\mu)} \quad \text{and} \quad \omega^{(1)} = 0 \quad (4.3)$$

The first order growth rate  $\sigma^{(1)}$  increases when going further beyond the threshold and decreases the more viscous the medium under consideration is. As expected, elasticity does not influence  $\sigma^{(1)}$  and Eq. (4.3) applies to ferrofluids and ferrogels, alike. Comparing with Eq. (4.2) we realize that the boundary behaves qualitatively different compared to the bulk, since it does not scale with  $\sigma^{(0)}$ . This is already manifest in the kinematic boundary condition, which connects the velocity field to the temporal change of the amplitude. It is therefore reasonable to compare the scaled time derivative from the bulk with that from the boundary. To get the latter we multiply Eq.(4.3) with the typical (linear) time scale  $\tau_0 = \nu_2 k_c (\rho g + \mu_2 k_c)^{-1}$  and assume that the dimensionless time scales are the same in the bulk and at the boundary, i.e.  $\tau_0 \sigma^{(1)} = \tilde{\partial}_T^{(1)}$ .

Adding up both second order conditions we finally get a rudimentary form of an amplitude equation

$$\tilde{\partial}_T^{(1)} \hat{\xi}_i = \frac{k_c \mu M^{(1)} M_c}{2(1+\mu)(\rho g + \mu_2 k_c)} \hat{\xi}_i - \frac{k_c}{4} \sum_{j,k}^{i \neq j \neq k} \hat{\xi}_j^* \hat{\xi}_k^* \quad (4.4)$$

In addition,  $\omega^{(1)} = 0$ , excludes an oscillatory behavior in this order.

The third order can be treated rather similarly. Fredholm's theorem for the bulk hydrodynamic equations leads to the condition, involving three modes  $120^\circ$  apart for hexagons

$$\tilde{\partial}_T^{(2)} \hat{\xi}_1 = -\frac{A'}{16\mu_2 k_c} |\hat{\xi}_1|^2 \hat{\xi}_1 - \frac{B'(120)}{32\mu_2 k_c} (|\hat{\xi}_2|^2 + |\hat{\xi}_3|^2) \hat{\xi}_1 \quad (4.5)$$

and cyclic permutations  $1 \rightarrow 2 \rightarrow 3 \rightarrow 1$ . The second order growth rate is  $\tilde{\partial}_T^{(2)} = \sigma^{(2)}/\sigma^{(0)}$ . For the case of the square pattern the cubic cross-coupling (with a different coefficient  $B'(90)$ ) is to the mode  $\hat{\xi}_5$ , which is under an angle of  $90^\circ$ . The pre-factors  $A'$  and  $B'$  will be discussed below.

The normal stress boundary condition leads in third order to the condition

$$\sigma^{(2)} + \frac{\mu_2 [\sigma^{(1)}]^2}{\nu_2 [\sigma^{(0)}]^2} = \frac{\mu(2M^{(2)} M_c + [M^{(1)}]^2)}{2\nu_2(1+\mu)} \quad \text{and} \quad \omega^{(2)} = 0 \quad (4.6)$$

Again we scale this equation by the linear time scale  $\tau_0$  and assume that the dimensionless time scale is the same in the bulk and at the surface ( $\tau_0\sigma^{(2)} = \tilde{\partial}_T^{(2)}$ ) and combine both third order conditions into

$$\begin{aligned} \tilde{\partial}_T^{(2)} \hat{\xi}_1 + \frac{\mu_2 k_c}{2(\rho g + \mu_2 k_c)} [\tilde{\partial}_T^{(1)}]^2 \hat{\xi}_1 &= \frac{k_c \mu (2M^{(2)} M_c + [M^{(1)}]^2)}{4(1+\mu)(\rho g + \mu_2 k_c)} \hat{\xi}_1 \\ &\quad - \frac{A'}{32\mu_2 k_c} |\hat{\xi}_1|^2 \hat{\xi}_1 - \frac{B'(120)}{64\mu_2 k_c} (|\hat{\xi}_2|^2 + |\hat{\xi}_3|^2) \hat{\xi}_1 \end{aligned} \quad (4.7)$$

for the hexagonal patterns. Following standard methods and multiplying the third order equation (4.7) by  $\epsilon^3$  and the second order equation (4.4) by  $\epsilon^2$ , we obtain the amplitude equation

$$\partial_T \xi_1 + \frac{\delta}{2} \partial_T^2 \xi_1 = \frac{1}{2} \tilde{\epsilon} \xi_1 - \frac{1}{2\sqrt{A}} \xi_2^* \xi_3^* - |\xi_1|^2 \xi_1 - \frac{B_{120}}{A} (|\xi_2|^2 + |\xi_3|^2) \xi_1 \quad (4.8)$$

with the dimensionless parameter  $\delta = \mu_2 k_c (\rho g + \mu_2 k_c)^{-1}$  and cyclic permutations  $1 \rightarrow 2 \rightarrow 3 \rightarrow 1$ . The (new) control parameter  $\tilde{\epsilon}$  is defined in the familiar way

$$(M^2 - M_c^2) = M_c^2 \tilde{\epsilon}. \quad (4.9)$$

The standard scaling of time, according to Eq. (3.9), and of the amplitude

$$\epsilon \tilde{\partial}_T^{(1)} + \epsilon^2 \tilde{\partial}_T^{(2)} \longrightarrow \partial_T \quad \text{or} \quad [\epsilon^1 \tilde{\partial}_T^{(1)}]^2 \longrightarrow \partial_T^2 \quad \text{and} \quad \epsilon k_c \sqrt{A} \hat{\xi}_n \longrightarrow \xi_n \quad (4.10)$$

is employed and the coefficients  $A \approx 5.750$  and  $B_{120} \approx 3.544$  are now dimensionless parameters. The corresponding amplitude equations for the square pattern read

$$\partial_T \xi_1 + \frac{\delta}{2} \partial_T^2 \xi_1 = \frac{1}{2} \tilde{\epsilon} \xi_1 - |\xi_1|^2 \xi_1 - \frac{B_{90}}{A} |\xi_5|^2 \xi_1 \quad (4.11)$$

with cyclic permutations  $1 \rightarrow 5 \rightarrow 1$  and  $B_{90} \approx 4.021$ .

The static solution for the hexagonal pattern takes the form  $\xi_n = -|\xi_n| \exp(i\Phi_n)$  for  $n \in \{1, 2, 3\}$  and  $\sum_n \Phi_n = 0$ , with the magnitude of the amplitudes

$$|\xi_n| = \frac{1}{4} \sqrt{A} \frac{1 + \sqrt{1 + 8(A + 2B_{120}) \tilde{\epsilon}}}{A + 2B_{120}}. \quad (4.12)$$

Thus, hexagons exist, if  $-(A + 2B_{120})^{-1} < 8\tilde{\epsilon}$ , i.e. already for magnetic fields below the critical one, thus defining a bistable regime. This transcritical behavior is due to the quadratic term in the amplitude equation.<sup>22)</sup> The square pattern is supercritical with amplitudes  $|\xi_n|^2 = \frac{1}{2} \tilde{\epsilon} A / (A + B_{90})$  for  $n = 1, 5$ .

The stability of the patterns is discussed in general terms in Ref.<sup>23)</sup> Hexagons are losing stability with respect to squares at the critical control parameter  $\tilde{\epsilon}_B$ , while the square pattern is stable for control parameters larger than  $\tilde{\epsilon}_S$ . With the values for  $A$ ,  $B_{120}$ , and  $B_{90}$  derived above (and  $B_{30} \approx 4.188$ ),  $\tilde{\epsilon}_S \approx 1.17 < \tilde{\epsilon}_B \approx 31.9$ , and a bistable regime exists also between hexagons and squares.

The dynamical behavior of the patterns beyond the linear threshold can also be inferred from the amplitude equations. Assuming a spatially homogeneous hexagonal

pattern with the amplitude  $|\xi_n| + X(t)$ , where  $|\xi_n|$  is the stationary solution Eq. (4.12), the linearized dynamic equation for the disturbance  $X$  takes the form of a damped harmonic oscillator equation

$$\partial_T^2 X + \gamma \partial_T X + \omega_0^2 X = 0 \quad (4.13)$$

with the damping rate constant  $1/\gamma = \delta/2$  and frequency  $\omega_0^2 = (\tilde{\epsilon} + |\xi_n|/\sqrt{A})/\delta$  (in dimensionless form). Thus, over the whole existence range, the hexagon pattern relaxes by damped oscillations to the stationary state, if disturbed homogeneously. Returning to physical time units the damping rate constant is  $\nu_2/(\mu_2 + \sqrt{\sigma_T \rho g})$ , while the frequency (squared) is proportional to  $\mu_2(\mu_2 + \sqrt{\sigma_T \rho g})$ , and in addition increases with the external field and the pattern amplitude. Disturbances of the square pattern behave dynamically quite similarly and are described by Eq.(4.13) with  $\omega_0^2 = \tilde{\epsilon}/\delta$ .

### §5. Usual Ferrofluids

In the case of ferrofluids, there is no elasticity and the shear elastic modulus  $\mu_2$  vanishes. This immediately eliminates the second order time derivative from the amplitude equations indicating that the stationary state is reached by a pure relaxation. The critical threshold  $\mu[M_c^{\text{fl}}]^2 = 2(1 + \mu)\sqrt{\sigma_T \rho g}$  regains its familiar ferrofluid value changing implicitly  $\tilde{\epsilon}$  in Eq.(4.9) to  $\tilde{\epsilon}^{\text{fl}}$ , while the critical wave number  $k_c^2 = \rho g/\sigma_T$  is unchanged. Although the condition  $\mu_2 = 0$  is necessary, since it removes elasticity from the static part of the fundamental equations, it is not sufficient to extract the ferrofluid case from the ferrogel one. The kinematic boundary condition (2.7), crucial for the description of a deformable free surface, is the same for gels and fluids, and it is necessary to treat the Rosensweig instability fully dynamically, even for ferrofluids. However, the dynamic equation for the strain field (2.1), which has no physical meaning for ferrofluids, has to be absent. Thus, the derivation of the amplitude equations sketched above has to be redone without that equation.

The resulting amplitude equations for hexagons and squares, respectively, are of the same general form as those for ferrogels, Eqs. (4.8) and (4.11) with  $\delta \equiv 0$  of course,

$$\partial_T \xi_1 = \frac{1}{2} \tilde{\epsilon}^{\text{fl}} \xi_1 - \frac{2}{3\sqrt{A^{\text{fl}}}} \xi_2^* \xi_3^* - |\xi_1|^2 \xi_1 - \frac{B_{120}^{\text{fl}}}{A^{\text{fl}}} (|\xi_2|^2 + |\xi_3|^2) \xi_1 \quad (5.1)$$

$$\partial_T \xi_1 = \frac{1}{2} \tilde{\epsilon}^{\text{fl}} \xi_1 - |\xi_1|^2 \xi_1 - \frac{B_{90}^{\text{fl}}}{A^{\text{fl}}} |\xi_5|^2 \xi_1 \quad (5.2)$$

but have different coefficients for the quadratic and cubic amplitude terms  $A^{\text{fl}} \approx 8.625$ ,  $B_{120}^{\text{fl}} \approx 3.150$ , and  $B_{90}^{\text{fl}} \approx 4.266$ .

The discussion of the stability of the different patterns shows qualitatively the same results as for ferrogels, despite the somewhat different numerical values of the cubic amplitude coefficients. The dynamics of homogeneous disturbances of those patterns is purely relaxational with a damping rate constant of  $\nu_2/\sqrt{\sigma_T \rho g}$ .

## §6. Discussion

In this article we showed how to derive an amplitude equation for the Rosensweig instability in isotropic magnetic gels and fluids based on the fundamental hydrodynamic equations. The most important step was to treat the Rosensweig instability fully dynamically and take the zero frequency limit at the end, only. The decoupling of the magnetic bulk equations from the hydrodynamic ones, required Fredholm's conditions to be amended by the normal stress boundary condition. While combining those two contributions there was some freedom in choosing the relative weight between them. It seemed reasonable to weigh these single contributions equally with respect to each other, which was implicitly also done, for example, in the nonlinear discussions using an extended scalar product.<sup>24)</sup>

The results for the stationary patterns in this article are in qualitative accordance with the bifurcation scenario obtained with the energy method.<sup>11)</sup> Hexagons are the stable surface pattern at the linear instability point, and the bifurcation from the flat surface to the hexagonal pattern is transcritical. For high magnetic field strengths, hexagons become unstable and a square pattern develops. Also this transitions involves a bistable region. We obtained qualitatively similar results in the case of ferrofluids for the statics of the patterns.

The analysis in this article provided us with nonlinear dynamical processes, too. We obtained the typical first order time derivative describing, above the linear threshold, the growth of the surface spikes or the relaxation to their stationary state. The typical time scale of the growth (or relaxation) processes was proportional to the viscosity and became smaller for increasing surface tension and shear moduli. Additionally, however, we found a second order time derivative in the case of magnetic gels giving rise to an internal frequency proportional to the elastic modulus (and depending on the surface tension and the actual amplitude or external field). Comparing our derivation of the amplitude equation with that for oscillatory instabilities,<sup>25)</sup> we found that not only the critical frequency at the linear threshold is zero, but also the first and second order corrections to it, thus excluding traveling or standing oscillatory patterns.

The nonlinear behavior of the Rosensweig instability was based on the nonlinearity of the Maxwell stress tensor, the nonlinear transport derivatives of the dynamic variables including the strain tensor, and on the explicit and implicit nonlinearities that arise due to the deformation of the surface, at which the boundary conditions are taken. We neglected other nonlinearities like magnetostriction, nonlinear elasticity and nonlinear magnetization (in the sense that deviations of the magnetization from its value given by the actual field are treated linearly). As a consequence of that the coefficients of the cubic amplitude terms are independent of the elastic shear modulus and the magnetic susceptibility. Using the energy method to describe the nonlinear behavior, however, the results depended on those parameters and, in particular, were useful for very small values of the magnetic susceptibility and the elastic moduli, only. Since in the energy method case the same basic assumptions have been made, this classifies the intrinsic procedure of that method to be unsystematic.

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