A Letters Journal Exploring the Frontiers of Physics

May 2007

EPL, **78** (2007) 44003
doi: 10.1209/0295-5075/78/44003

www.epljournal.org

Measuring the anomalous dispersion branch of surface waves on ferrofluids

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received 19 December 2006; accepted in final form 3 April 2007
published online 7 May 2007

PACS 47.50.Gj – Instabilities
PACS 75.50.Mm – Magnetic liquids
PACS 47.57.-s – Complex fluids and colloidal systems

Abstract – In this paper we report on the first experimental investigation of the anomalous dispersion branch of surface waves on ferrofluids in a magnetic field. The existence of such an anomalous dispersion branch has been predicted by several authors, but there has been no experimental evidence for its existence. In our experiments we used the well-known Faraday instability of parametrically excited surface waves on a ferrofluid in the presence of a magnetic field oriented perpendicular to the fluid's surface.

Cowley and Rosensweig [1] have investigated experimentally the instability of a flat ferrofluid surface in a static magnetic field H oriented perpendicular to the surface: above a certain critical value, H_C^R, the surface spontaneously deforms into regularly spaced peaks, that are usually arranged in a hexagonal pattern. At the critical field H_C^R of this so-called Rosensweig instability, the dispersion curve of surface waves touches the zero frequency axis at a finite wave number k_C^R, analogous *e.g.* to the soft mode behavior of phonons at structural phase transitions of solids. For magnetic fields slightly below H_C^R, the dispersion is non-monotonous with a section of negative slope (anomalous dispersion). There exists a number of papers dealing with theoretical modelling of the dispersion relation [2–5]. But up to now the anomalous dispersion has not been detected experimentally, although there is experimental evidence for a jump in wave number [6] that is expected to occur in the presence of anomalous dispersion when ramping the frequency up and down [7].

The excitation of surface waves has been realized by different mechanisms. Pétrélis *et al.* [8] have studied the influence of the Faraday instability mechanism of parametrically excited surface waves that arise in vertically shaken containers on the Rosensweig instability for driving frequencies $f \geqslant 30\,\mathrm{Hz}$. They have demonstrated experimentally that the Faraday instability inhibits the Rosensweig instability, a fact that was predicted by Müller in [7]. In [6] Mahr *et al.* have reported the existence of two different wave numbers occurring at the same excitation frequency in a V-shaped annular teflon channel. In order to generate surface waves they have directed an air jet towards the magnetic fluid's surface. Results of measurements at a magnetic field 3% below the threshold of the Rosensweig instability have been presented but the anomalous region of the dispersion has not been studied. Due to the finite geometry of the vessel a quantitative comparison of their experimental results with theoretical calculation was not possible. In [9,10] Mahr *et al.* have used a mechanical excitation of surface waves in the presence of a DC magnetic field. The authors have found so-called twin-peak patterns in the non-monotonic regime of the dispersion relation 2% below the critical field H_C^R and furthermore domain structures, oscillating defects and relaxational phase oscillations. These findings have been attributed to the non-monotonic dispersion relation of ferrofluids. The same group has presented in [11] experimental results on surface waves, that have been excited by an oscillating magnetic field. Both subharmonic and harmonic surface response have been detected. Higher-order resonance tongues were generated by means of tuning the viscous damping of the magnetic

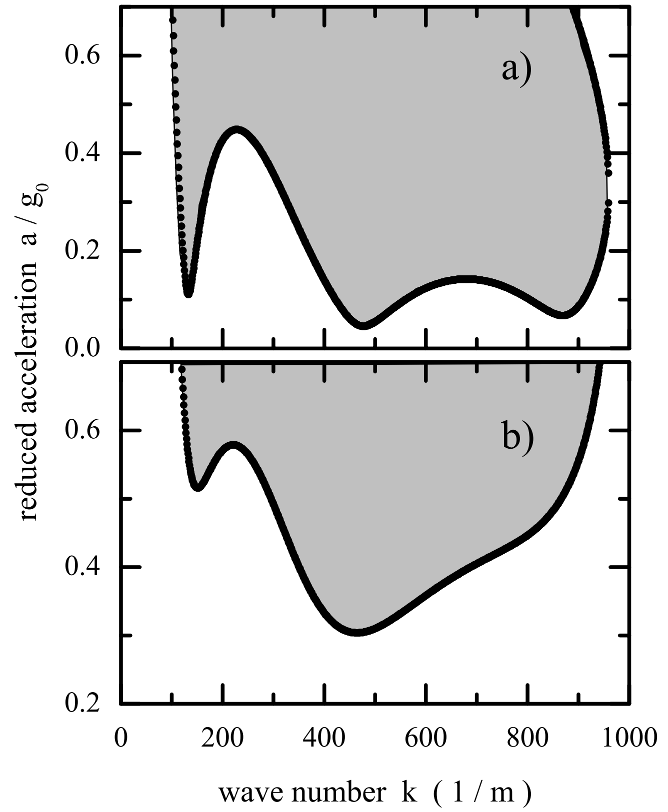

Fig. 1: Marginal stability curves of the flat surface against growth of standing waves for different values of the viscosity η ((a) $\eta = 6.66\,\mathrm{mPas}$, (b) $\eta = 53.3\,\mathrm{mPas}$) for the following parameters: $f = 7\,\mathrm{Hz}$, $h = 5\,\mathrm{mm}$, $\sigma = 0.022\,\mathrm{N/m}$, $\rho = 1110\,\mathrm{kg/m^3}$, $\chi = 0.8$ and $M = 0.999\,M_C^R$. Within the gray regions, the plane surface is unstable.

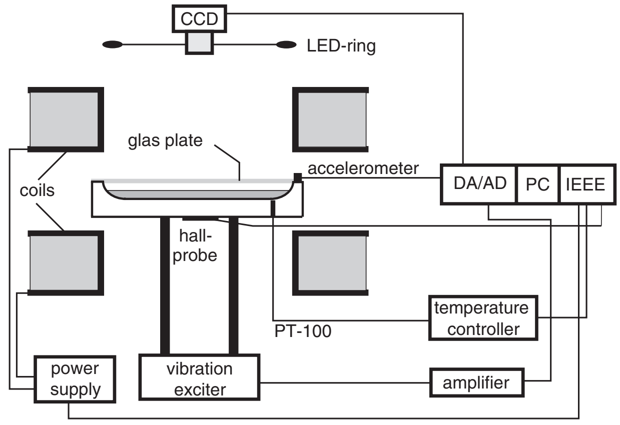

Fig. 2: Sketch of the experimental setup. For further details see the text.

fluid with an external magnetic field. This was made possible by a specific peculiarity of ferrofluids, namely a very weak damping of surface waves when the wave number is close to the critical one of the Rosensweig instability. Browaeys *et al.* [12] have used a small coil placed beneath the ferrofluid container in order to generate surface waves. With this method it was very difficult to excite surface waves with wavelengths small compared to the coils diameter. They limited the ratio H/H_C^R to 0.79 because above this ratio the peaks started to form at the boundary of the vessel.

A drawback of generating surface waves by modulating the magnetic field is that the time variation of the magnetization $M(t) = \chi H(t)$ enters quadratically into the dispersion relation so that the effective excitation signal is composed of two frequencies. Furthermore, the effective magnetic drive is proportional to k^2 while it is linear in k for the gravity modulation. Thus, shorter wavelength are favored by the magnetic drive. To avoid these complications the Faraday forcing is used, where surface waves are generated only beyond a critical excitation amplitude [13]. In this type of experiment the earth's gravity acceleration g_0 is modulated in the co-moving frame according to $g(t) = g_0 + a\cos(2\Omega t)$. As long as the amplitude a is smaller than a critical one, a_C, the surface of the ferrofluid remains flat. By increasing a beyond a_C, the flat surface becomes unstable and the system forms standing waves. The wave pattern depends on the mass density ρ, surface tension σ, viscosity η of the fluid and its filling level h. For small values of h, an experimental investigation of the anomalous part of the dispersion relation was predicted to be feasible [7].

In order to get access to the anomalous dispersion branch we took advantage of the dissipation in the bottom boundary layer [7] that is responsible for the suppression of the growth of waves with wave numbers below $k \sim 1/h = 200\,\mathrm{cm}^{-1}$ for $h = 5\,\mathrm{mm}$. Being strongly damped the onset of such waves in the region of linear dispersion is delayed. Furthermore, by choosing the viscosity appropriately the marginal stability curve $a(k)$ of the flat surface against the growth of standing waves can be tuned such that its absolute minimum k_c lies in the wave number region of anomalous dispersion. Then the growth rates of waves in this region are largest and get selected when increasing the acceleration above threshold. Besides the filling level h, the fluid's viscosity η influences the marginal curves in a way that can be used to get access to the anomalous dispersion branch. The influence of the viscosity on the marginal curves is shown in fig. 1. The upper panel of fig. 1 shows the marginal curves for a ferrofluid of low viscosity ($\eta = 6.66\,\mathrm{mPas}$), while in the lower panel the high-viscosity case ($\eta = 53.3\,\mathrm{mPas}$) is presented. In the latter, the high viscosity clearly favors the local minimum belonging to the anomalous dispersion branch. Thus, choosing an appropriate combination of filling level and fluid's viscosity enabled us to see in our experiment only the dispersion branch of interest, *i.e.* the anomalous one.

The experimental setup is sketched in fig. 2.

The cylindrical container filled with the ferrofluid is attached to a mechanical vibrator (Large Displacement

$$-\omega + \omega^2 X(k,\omega) = \frac{\nu^2}{q\coth(qh) - k\coth(kh)}$$
$$\times \left\{ q\left[4k^4 + (k^2+q^2)^2\right]\coth(qh) - k\left[4k^2 g^2 + (k^2+q^2)^2\right]\tanh(kh) - \frac{4k^2 q(k^2+q^2)}{\cosh(kh)\sinh(qh)} \right\}. \quad (5)$$

Shaker V617, Gearing & Watson Electronics Ltd.) connected to a 4 kW power amplifier (DSA-4k, Gearing & Watson Electronics Ltd.). The container is placed in the center of a pair of Helmholtz coils providing a magnetic field oriented perpendicular to the fluid surface. The homogeneity of the field across the liquid level is better than 5%. The sinusoidal signal for the power amplifier was synthesized by a digital-analog card installed in a Pentium PC. The vertical acceleration of the container is measured by a piezo-electric device (Brüel & Kjaer 4393). The signal of this device is amplified (Brüel & Kjaer 2525) and routed to the PC for data acquisition. The characteristics of the vibration exciter is rather non-linear for the low driving frequencies f that are of interest for the present study ($f \leqslant 10\,\mathrm{Hz}$). Therefore a continuous control of the excitation signal was necessary. In order to get a pure sinusoidal acceleration, the accelerometer signal was Fourier analyzed. Higher harmonics of Ω were eliminated by admixing Fourier components with appropriate inverse phases to the excitation signal. The amplitude of these components were determined by a proportional control loop. Using this method the power spectrum of the accelerometer signal was made monochromatic with a purity of 99%.

A heating foil was mounted to the container. Using a temperature controller the temperature measured by a Pt-100 resistor, embedded in the container core, was regulated to $T = (24.5 \pm 0.1)\,^\circ\mathrm{C}$. Thereby the viscosity and the surface tension of the liquid were held constant. In order to visualize the surface structure we used a full frame CCD camera (Hitachi KPF-1). The CCD camera was placed about 1 m above the interface ferrofluid/air in the center of a ring (with radius $r = 0.3\,\mathrm{m}$) of 120 LEDs illuminating the surface. Using this visualization technique only surface elements of appropriate slope reflect light into the camera, while the flat regions of the surface appear black. The spatial symmetry of the surface deformation $\zeta(x,y)$ was analyzed by using two-dimensional Fourier transformation. To that end the recorded light intensity $I(x,y)$ of a video image was convoluted with a Gaussian function and processed by a fast Fourier transformation algorithm. For the measurements we used a commercial ferrofluid (APG J12, FerroTec) with the following specifications: dynamic viscosity $\eta = 45\times10^{-3}\,\mathrm{Pas}$, mass density $\rho = 1110\,\mathrm{kg/m^3}$ and initial magnetic susceptibility $\chi = 0.8$. We choose this ferrofluid because its carrier liquid has a very low vapor pressure which ensures long time stability of all the fluid parameters. Note that changes in the parameters like viscosity and surface tension lead to a significant change of the dispersion curve. Furthermore, the large viscosity of APG J12 inhibits meniscus waves, that are usually emitted from the container walls and that disturb the Faraday waves.

The dispersion relation of surface waves for an ideal inviscid fluid in a perpendicular magnetic field,

$$0 = -\omega^2 + \omega_0^2(k) \quad (1)$$

$$\omega_0^2(k) = \tanh(kh)\left\{ g_0 k + \frac{\sigma}{\rho}k^3 - \frac{\mu_0}{\rho}\left(\frac{1+\chi}{2+\chi}\right) \times \left(1 - \frac{2\chi}{(2+\chi)^2 e^{2kh} - \chi^2}\right) M^2 k^2 \right\} \quad (2)$$

depends on h, g_0, σ, χ and the magnetization M. For high filling levels, $kh \gg 1$, the critical magnetization M_C^R and the critical wave number k_C^R of the Rosensweig instability read

$$\left(M_C^R\right)^2 = \frac{2}{\mu_0}\left(\frac{2+\chi}{1+\chi}\right)\sqrt{\rho g_0 \sigma}, \quad k_C^R = \sqrt{\rho g_0/\sigma}. \quad (3)$$

There is a second characteristic value of the magnetization $\widetilde{M} < M_C^R$ above which the dispersion curve is no longer monotonic. A simple calculation gives: $\widetilde{M} = \sqrt[4]{3/4}\, M_C^R$, *i.e.*, $\widetilde{M} \approx 0.93\, M_C^R$. For $M > \widetilde{M}$ the dispersion relation $\omega_0^2(k)$ shows a minimum so that there is a range of k-values in which the group velocity is negative; this is referred to as anomalous dispersion. Thus, for M approaching M_C^R there is a band of frequencies in which the dispersion relation eqs. (1), (2) have solutions with three different k-values. For real fluids the viscosity of the fluid must be taken into account. Using the expression of Kumar and Tuckerman [13] the dispersion relation including the viscosity (and appropriate boundary conditions) reads now (see eq. (5))

$$-\omega^2 + \omega^2 X(k,\omega) + \omega_0^2(k) = 0 \quad (4)$$

with $q = \sqrt{k^2 + i\omega/\nu}$, where ν denotes the kinematic viscosity. The magnetization enters via the inviscid dispersion relation $\omega_0(k)$ eqs. (2).

In order to determine the critical parameters (a_C and k_C) of the Faraday instability for a given field value H and a given driving frequency f, we used a special version of the algorithm proposed by Kumar and Tuckerman. The

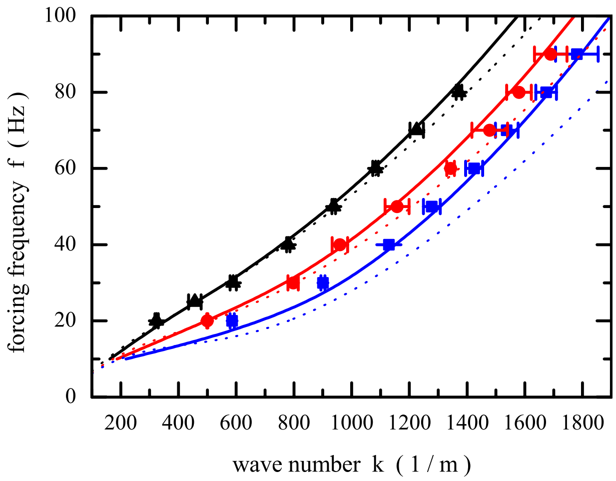

Fig. 3: (Color online) Forcing frequency f *vs.* wave number k of the standing wave pattern at slightly supercritical acceleration amplitudes. The solid lines show critical values resulting from eqs. (4)–(7) with the parameters $\rho = 1110\,\mathrm{kg/m^3}$, $\chi = 0.8$, $\sigma = 0.02\,\mathrm{N/m}$ and $\eta = 45.77 \times 10^{-3}\,\mathrm{Pas}$. The dashed lines are calculated for $\eta = 0$. The magnetization was $M = 0$, $0.7\,M_C^R$, and $0.87\,M_C^R$ from top to bottom, respectively.

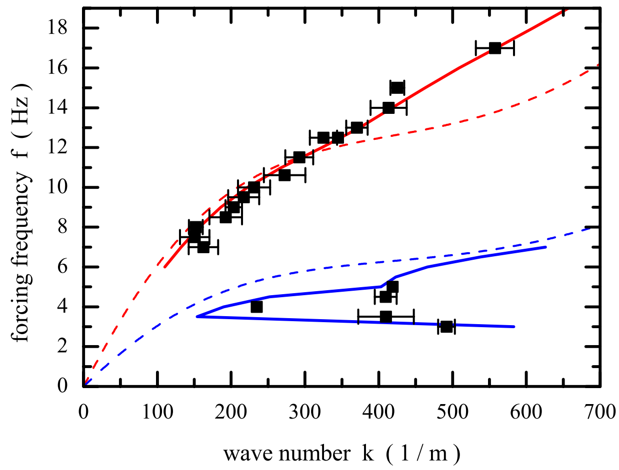

Fig. 4: (Color online) Forcing frequency f *vs.* wave number k as in fig. 3; here, however, for $M = 0.9\,M_C^R$. For $3\,\mathrm{Hz} \leqslant f \leqslant 5.5\,\mathrm{Hz}$ the surface response is harmonic in time, while for $f \geqslant 6\,\mathrm{Hz}$ it is subharmonic. Full lines show critical values as in fig. 3; dashed curves refer to the idealized case $\eta = 0$.

surface deformation is described by a Floquet-ansatz

$$\zeta \propto e^{i\beta\Omega t} \cdot \sum_{n=-\infty}^{\infty} \zeta_n e^{2in\Omega}. \tag{6}$$

Here $\beta = 1$ ($\beta = 0$) corresponds to a subharmonic (harmonic) response of the surface deformation, respectively. The Floquet-ansatz leads to tridiagonal system of equations

$$0 = \{-\omega^2 + \omega^2 X(\omega, k) + \omega_0^2(k)\}\zeta_n + \frac{ak\tanh(kh)}{2}(\zeta_{n+1} + \zeta_{n-1}) \tag{7}$$

with $\omega = (2n + \beta)\Omega$. Equations (7) can be numerically approximated by an appropriate cutoff. The solvability condition yields the neutral stability curves $a^{SH}(k)$ and $a^H(k)$ for the subharmonic (SH) and harmonic (H) surface response, respectively. A minimization of these curves with respect to the wave number k determines the critical acceleration a_C^{SH} (a_C^H) and the critical wave number k_C^{SH} (k_C^H).

In what follows we present results of experiments performed at a fixed filling level of $h = 5\,\mathrm{mm}$. Knowing the mass density of the ferrofluid under investigation, the filling height was determined by weighing the fluid. We measured the mass density ρ with a calibrated pycnometer and the value of $\rho = 1110\,\mathrm{kg/m^3}$ agrees with the specification provided by the manufacturer. The surface tension σ of the ferrofluid was not known, therefore we first performed measurements at $M = 0$, $M = 0.7\,M_C^R$ and $M = 0.87 M_C^R$. The data of these measurements are presented in fig. 3. As demonstrated in [13,14], the Faraday instability provides a versatile method to measure the surface tension. Within our investigations we measured the critical acceleration a_C as a function of the driving frequency. Furthermore the critical wave number k_C was determined from video images recorded close to the threshold of the Faraday instability.

Our experimental results are well described by numerical calculations using eqs. (4)–(7) and the following parameters: $\eta = 45.77 \times 10^{-3}\,\mathrm{Pas}$, $\chi = 0.8$ and $\sigma = 0.02\,\mathrm{N/m}$. So the surface tension $\sigma = 0.02\,\mathrm{N/m}$ is a direct outcome of our experiments. In order to study the anomalous dispersion branch the external field strength was chosen to be shortly below the critical value. The experiments have been performed in the following way: with M and f set to the desired values, the acceleration of the shaker has been slowly increased up to the critical value of the Faraday instability where the initially flat surface transforms into a standing wave pattern. In order to map the surface deformation we had to increase the acceleration further to about 3–5% above a_c.

In fig. 4 we present the experimental results obtained at $M = 0.9\,M_C^R$. For forcing frequencies $3\,\mathrm{Hz} \leqslant f \leqslant 5.5\,\mathrm{Hz}$ the system responds with a frequency equal to the forcing frequency (harmonic response), while for $f \geqslant 6\,\mathrm{Hz}$ the response frequency is half the driving frequency (subharmonic response). In contrast to the subharmonic dispersion curve, that shows normal behavior, the harmonic one in the lower part of fig. 4 shows more complex behavior which is well described by the numerical results. The later one shows a sharp turning point around wave number $150\,\mathrm{m^{-1}}$. Note that for higher frequencies it is essential to include the effects of the viscosity as demonstrated by the comparison of the experimental data with the viscid and inviscid model. We stress that in non-magnetizable fluids temporal harmonic

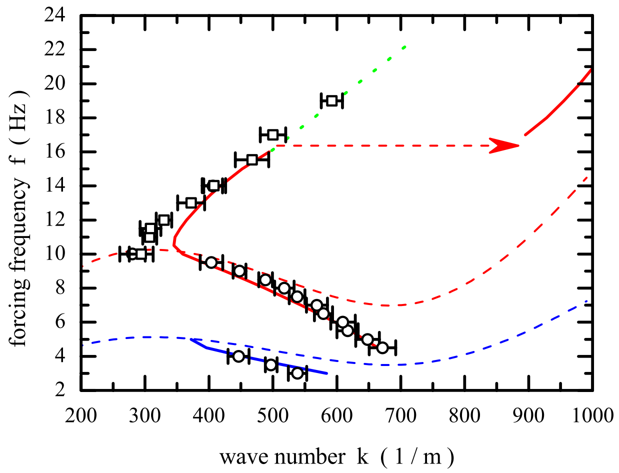

Fig. 5: (Color online) Forcing frequency f *vs.* wave number k as in fig. 3 for $M = 0.987\, M_C^R$. Open circles denote experimental data on the anomalous dispersion branch, while open squares refer to the normal dispersion branch. The dotted (green) line is a guide for the eye, following the subharmonic-dispersion relation. The arrow is explained in the text. Full lines show critical values as in fig. 3; dashed curves refer to the idealized case $\eta = 0$.

surface waves are realizable only either at very extreme experimental conditions, *i.e.*, low excitation frequencies and very low filling levels (usually below $h = 1\,\mathrm{mm}$) [15,16] or by using non-Newtonian liquids [17].

In a second experiment M was set 1.3% below the critical Rosensweig value M_C^R. The results of the measurements are presented in fig. 5.

Both the temporal harmonic and the temporal subharmonic dispersion relation now show anomalous behavior for forcing frequencies $f \leqslant 10\,\mathrm{Hz}$. The experimental findings are well described with the numerical results obtained with the same parameters for η, σ and χ as in the experiments at $M = 0$ and $M < \widetilde{M}$. For forcing frequencies $f \geqslant 16\,\mathrm{Hz}$ the numerical results predict a jump in wave number from $k \approx 500\,\mathrm{m}^{-1}$ to $k \approx 900\,\mathrm{m}^{-1}$ as indicated by the arrow in fig. 5. However, this jump does not appear in the experiment. The data points rather follow the continuation of the low-frequency dispersion branch (as indicated by the dotted line in fig. 5 as a guide for the eye). This finding is up to now not fully clear, though the topology of the neutral stability curves could explain this result: for frequencies $f \geqslant 16\,\mathrm{Hz}$ and $\varepsilon_A \equiv a/a_C - 1 = 0.05$ the band of possible wave numbers k is about one order of magnitude larger than for driving frequencies within the range of the anomalous dispersion relation. Since the experiments have to be performed at finite supercritical ε_A there exists the possibility for the system to select a wave number different from k_C. With the aid of appropriate amplitude equations it might be possible to determine the wave number that will be selected for $\varepsilon > 0$ but this procedure requires the knowledge of the coupling coefficients appearing in the amplitude equations which are still unknown.

In conclusion, we present an experimental setup that enables the investigation of the dispersion relation of surface waves on ferrofluids. To our knowledge, these are the first experimental results within the anomalous range of the dispersion curve. Furthermore, the experiments provide a simple way to generate temporal harmonic surface response at moderate filling levels.

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This work was supported by DFG (SFB 277). Stimulating discussions with H. W. Müller, R. Richter, and V. V. Mekhonoshin are appreciated.

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