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Faraday instability in a linear viscoelastic fluid

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Abstract. – The onset of surface waves in a vibrated layer of a viscoelastic fluid is investigated theoretically. For vibration frequencies close to the inverse relaxation time of the non-Newtonian fluid, surface waves respond harmonically instead of being subharmonic as is usually the case in Newtonian fluids. This prediction has been made on the basis of the Maxwell model for viscoelastic fluids, but the result is robust and has been confirmed by using the empiric viscosity spectrum for a mixture of 2% polyisobutylene in primol. The parameter range is determined, where harmonic surface waves are favored.

Introduction. – Our understanding of pattern formation has experienced enormous progress during the recent decades [1]. Important insights were contributed by studies of Newtonian fluid systems, such as Rayleigh-Bénard convection [2], Taylor-Couette flow [3] and the Faraday instability [4-7]. Recently, pattern formation in viscoelastic fluids came into the focus of nonlinear science too. With the memory of viscoelastic fluids an additional time scale is introduced, which gives rise to a number of interesting phenomena. For example, the interplay between the viscoelastic relaxation time and the thermal diffusion time lead to an oscillatory onset of Rayleigh-Bénard convection [8], which exhibits also a multi-critical bifurcation behavior [9]. The competition between the elastic and inertial instability in Taylor-vortex flow leads to a number of novel phenomena [10], *e.g.* localized structures [11].

The well-founded theory of Newtonian fluids is one reason of the popularity of fluid systems in pattern formation. It allows *ab initio* calculations and hence quantitative comparisons between theoretical concepts in pattern formation and experimental measurements. For a number of examples, especially for Rayleigh-Bénard convection and Taylor-vortex flow, a very precise agreement has been achieved between both approaches. This accelerated the progress in pattern formation considerably.

For viscoelastic fluids the theoretical basis is less advanced. A generally accepted description for the large scale motion of viscoelastic fluids —as the Navier-Stokes equations for Newtonian fluids— is not available yet. However, for small displacement gradients the "general

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linear viscoelastic model" is broadly accepted as an appropriate description for incompressible viscoelastic fluids [12]. Nowadays quantitative measurements of the linear viscoelastic flow properties are available by standard rheological techniques. Accordingly, those experiments, for which the linear viscoelastic model applies, are expected to compare quantitatively with the theory.

The present paper deals with the onset of surface waves on a viscoelastic fluid layer subjected to a vertical vibration, the so-called Faraday instability. For Newtonian liquids Faraday waves are currently under intensive experimental and theoretical investigation [4, 6, 7]. This system is experimentally attractive because of its short time scales and the two available control parameters, namely the vibration amplitude and the vibration frequency. Usually, Faraday waves resonate *subharmonically*, *i.e.* with twice the period of the forcing, except in very thin layers, where the synchronous (harmonic) response can be observed [6, 7].

In viscoelastic fluids an inherent material specific frequency is defined by the inverse elastic relaxation time. On selecting the external vibration frequency close to this value, viscoelastic effects are expected to influence the onset of surface waves. Indeed, for a specified parameter range we predict that the surface waves resonate *harmonically* with the vibration.

System. – We consider an incompressible horizontal fluid layer with density ρ and with surface tension α, which is vertically vibrated. The vibration leads in the co-moving frame of the container to a modulation of the gravitational acceleration $g(t) = g_0 + a\cos(2\Omega t)$. In the quiescent basic state there is no flow in the fluid layer and no deformation of the surface. Therefore, the onset of Faraday waves can be calculated with equations linear in the velocity and surface deformation. The linearized Navier-Stokes equation is

$$\rho\frac{\partial \mathbf{v}}{\partial t} = -\rho\, g(t)\,\mathbf{e}_z + \nabla\cdot\sigma\,, \qquad \text{with} \qquad \sigma_{ij} = -p\,\delta_{ij} + \int_{-\infty}^{t} \mathrm{d}t' G(t-t')\,\dot{\gamma}_{ij}(t') \tag{1}$$

and the pressure $p(\mathbf{r}, t)$. The right part in (1) makes use of the constitutive equation of linear viscoelasticity, which relates viscous stresses to the history of the rate of strain tensor $\dot{\gamma}_{ij} = (\nabla_i v_j + \nabla_j v_i)$. $G(t)$ is the relaxational modulus (see, *e.g.*, [12]). Since i) a linear stability analysis assumes infinitesimal perturbations and ii) Faraday waves bifurcate out of the motionless state, the linear viscoelastic model is fully sufficient for the present problem. More complex viscoelastic flow properties, such as normal stress differences, can be neglected here. They would enter the description beyond threshold in the nonlinear regime. Note, that the present situation is similar to the problem of thermal convection in viscoelastic fluids [8, 9], but differs from the Couette-Taylor instability, where the finite velocity gradients of the basic Couette-state [3] require knowledge of nonlinear material properties.

A complete description of surface waves requires boundary conditions (for details see, *e.g.*, [5]). As this paper is focused on the viscoelastic *bulk* effects, we consider a half-infinite liquid layer between $z = 0$ and $z \to -\infty$. Surface deformations are described by the Monge-function $z = \eta(\mathbf{r}_\perp, t)$, where $\mathbf{r}_\perp$ abbreviates the horizontal coordinates (x, y). With respect to $\mathbf{r}_\perp$, the linear equations for the velocity and the surface deformation $\eta(\mathbf{r}_\perp, t)$ can be Fourier-transformed and the z-dependence of the velocity can be integrated. Using a Floquet ansatz for the time dependence of both fields, one obtains

$$\eta(\mathbf{r}_\perp, t) = e^{(\sigma_g + i\beta\Omega)t} \sum_{n=-\infty}^{\infty} \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\mathbf{k} e^{i(\mathbf{k}\cdot\mathbf{r}_\perp + 2n\Omega t)}\hat{\eta}(\mathbf{k}, 2n\Omega)\,, \tag{2}$$

and a similar expansion for the velocity field. We always find real values for the Floquet parameter σ_g. Hence, the discrete Fourier sum is harmonic (H) with respect to the external

driving if $\beta = 0$ and subharmonic (S) if $\beta = 1$. Then the marginal stability criterion, $\sigma_g = 0$, leads to a tridiagonal system for the Fourier coefficients

$$\left[\omega_0^2 - \omega^2 + \omega^2 X(k,\omega)\right] \hat{\eta}(k,\omega) \; + \; a\,\frac{\mid k \mid}{2}\left[\hat{\eta}(k,\omega+2\Omega) + \hat{\eta}(k,\omega-2\Omega)\right] = 0,$$

$$\text{with} \;\; X(k,\omega) = 4i\left(\frac{\nu^\star k^2}{\omega}\right) \; + \; 4\left(\frac{\nu^\star k^2}{\omega}\right)^2 \left(\sqrt{1 + i\omega/(\nu^\star k^2)} - 1\right) \; . \tag{3}$$

The solvability condition determines the neutral vibration amplitude $a(k)$. A subsequent minimization with respect to the lateral wave vector $\mathbf{k}$ yields the threshold a_c. In eq. (3) we have $\omega = 2n\Omega$ for the harmonic response and $\omega = (2n+1)\Omega$ for the subharmonic one, where n runs form $-\infty$ to $+\infty$. Furthermore, $\omega_0^2 = g_0 \mid k \mid + (\alpha/\rho) \mid k \mid^3$ and the complex kinematic viscosity is

$$\nu^\star = \nu'(\omega) - i\nu''(\omega) = (1/\rho)\int_0^\infty G(t) e^{-i\omega t}\mathrm{d}t\,. \tag{4}$$

The real and imaginary parts of $\nu^\star$ account for the dissipative and the elastic character of the fluid. For $a = 0$, eq. (3) coincides with the complex dispersion relation for free (=unforced) viscoelastic surface waves studied in [13]. The last term on the right-hand side of eq. (3) reflects the parametric drive and couples different temporal Fourier modes.

Truncated solution. – The thresholds for the harmonic and the subharmonic branch are fairly good approximated by truncations of the system of equations (3). For the subharmonic branch a two-mode truncation ($n = -1, 0$) leads to a driving force $a^{(\mathrm{S})}(k)$, which is minimal at the wave number k_S:

$$a_\mathrm{c}^{(\mathrm{S})} = a^{(\mathrm{S})}(k_\mathrm{S}) \simeq \frac{2\Omega^2}{\mid k_\mathrm{S} \mid}\Im[X(k_\mathrm{S},\Omega)]\,, \quad \text{where } k_\mathrm{S} \text{ solves} \quad \omega_0^2(k_\mathrm{S})/\Omega^2 + \Re[X(k_\mathrm{S},\Omega)] \simeq 1\,. \tag{5}$$

$\Re[X]$ and $\Im[X]$ are the real and imaginary part of $X(k,\Omega)$, respectively. For the harmonic response a three-mode truncation ($n = -1, 0, 1$) yields

$$a_\mathrm{c}^{(\mathrm{H})} = a^{(\mathrm{H})}(k_\mathrm{H}) \simeq \sqrt{\frac{40}{\sqrt{6}}}\,\frac{\Omega\,\omega_0(k_\mathrm{H})}{\mid k_\mathrm{H} \mid}\sqrt{\Im[X(k_\mathrm{H},2\Omega)]}\,, \tag{6}$$

with the critical wave number k_H solving

$$\frac{\omega_0^2}{4\Omega^2} + \left(\Re[X(k_\mathrm{H},2\Omega)] - \sqrt{\frac{2}{3}}\Im[X(k_\mathrm{H},2\Omega)]\right) \simeq 1. \tag{7}$$

Linear viscoelastic materials. – The complex kinematic viscosity $\nu^\star$ in many polymeric solutions exhibits an extended power-law region with ν', $\nu'' \propto \Omega^{-\gamma}$ and γ around unity [12]. The dissipative real part ν' starts at the zero-shear rate viscosity $\nu_0 = \nu'(\Omega \to 0)$ and decays by orders of magnitude to a saturation value at large frequencies. For low frequencies the imaginary part is proportional to Ω but vanishes proportionally to Ω^{-1} at large frequencies. Hence the strongest influence of the fading memory is expected at intermediate frequencies, around the maximum of $\nu''(\Omega)$. This is the frequency range we are mostly interested in. A familiar model which mimics the viscosity spectra of ν' and ν'' is the Maxwell model, which adopts an exponentially decaying memory, $G(t) = (\nu_0/\lambda)e^{-t/\lambda}$. With eq. (4) one gets

$$\nu^\star(\Omega) = \frac{\nu_0}{1 + i\Omega\lambda} = \frac{\nu_0}{1 + (\Omega\lambda)^2} - i\Omega\lambda\frac{\nu_0}{1 + (\Omega\lambda)^2}\;, \tag{8}$$

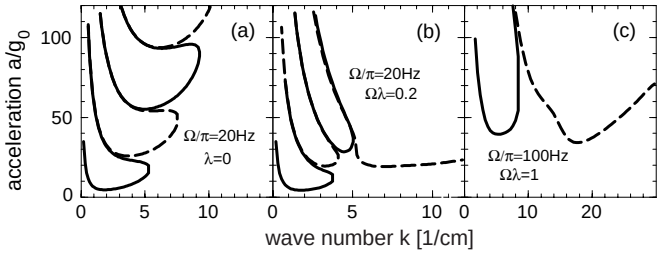

Fig. 1. – The neutral curves $a(k)$ of Faraday waves are given for the subharmonic (solid) and the harmonic (dashed) branches at different vibration frequencies, $f = \Omega/\pi$. In part (a) a Newtonian fluid is considered for the same viscosity $\nu_0 = 6 \times 10^{-4}\,\mathrm{m}^2/\mathrm{s}$ and surface tension $\alpha = 0.02$ N/m as for the Maxwell model with the relaxation time $\lambda = (0.01/\pi)$ s in parts (b), (c).

giving an easy two-parameter approach for the complex viscosity $\nu^\star$. For real polymeric solutions the Maxwell model applies only qualitatively. However, it covers the principal features of viscoelastic liquids, which are important for the effect considered here. Moreover, our results turn out to be insensitive to the actual power laws. For a special class of aqueous surfactant solutions [15] the Maxwell model applies even quantitatively.

Numerical results. – The static viscosities $\nu_0 = \nu'(\Omega = 0)$ of polymer solutions often exceed those of their Newtonian solvents by several orders of magnitude. In our numerical example we use $\nu_0 = 6 \times 10^{-4}\,\mathrm{m}^2/\mathrm{s}$ (600 times that of pure water). Systematic investigations [16,11] reveal that the relaxation time λ of surfactant or polymeric solutions can be varied considerably by the salt concentration, the viscosity of the solvent, or the temperature. For our calculation we choose the value $\lambda = (0.01/\pi)$ s, which can be conveniently probed in a Faraday experiment. The values $\alpha = 0.02$ N/m for the surface tension and $\rho = 1\ \mathrm{g/cm}^3$ for the density are typical for most fluids. In our frequency–wave number range the following inequalities hold: $\alpha k^3/\rho \ll \nu_0^2 k^4$ and $\alpha k^3/\rho \ll 2\nu_0 k^2/\lambda$. This parameter regime of negligible surface tension is also addressed by Pleiner *et al.* [13]. They investigate surface waves *without driving* in the transition range between elastic Rayleigh waves at $\Omega\lambda \gg 1$ and overdamped surface modes at $\Omega\lambda \ll 1$. Here however, the supercritical parametric drive guarantees standing surface waves over the whole parameter range, even in the overdamped region.

In Newtonian fluids ($\lambda = 0$) as in fig. 1a the neutral stability chart exhibits an alternating succession of neutral subharmonic and harmonic resonance tongues [5]. Viscoelasticity with a finite relaxation time $\lambda = (0.01/\pi)$s reduces the vibration amplitudes along the neutral curves (cf. fig. 1b). Simultaneously, a new harmonic tongue develops. On increasing the drive frequency to $\Omega \propto 1/\lambda$ (while keeping the rest of the parameters fixed) the new harmonic branch falls down below the neighboring subharmonic one and determines the primary threshold (see figs. 1c and 2a). Further elevation of the frequency beyond $\Omega \sim 1/\lambda$ makes the subharmonic tongue re-overtake the primary stability onset.

The harmonic branch as the first instability is intimately related to the visco*elasticity* of the material. For Newtonian viscous fluids the broad harmonic instability tongue as shown in fig. 1b,c never exists.

The frequency dependence of the minima of the neutral curves of the S- and the H-branch are shown in fig. 2a. They indicate that the frequency window with the dominating harmonic response is located around $\Omega \simeq 1/\lambda$. The thin lines in fig. 2a are obtained by the truncated solutions (5)-(7), which are fairly good approximation of the full solution in this parameter range.

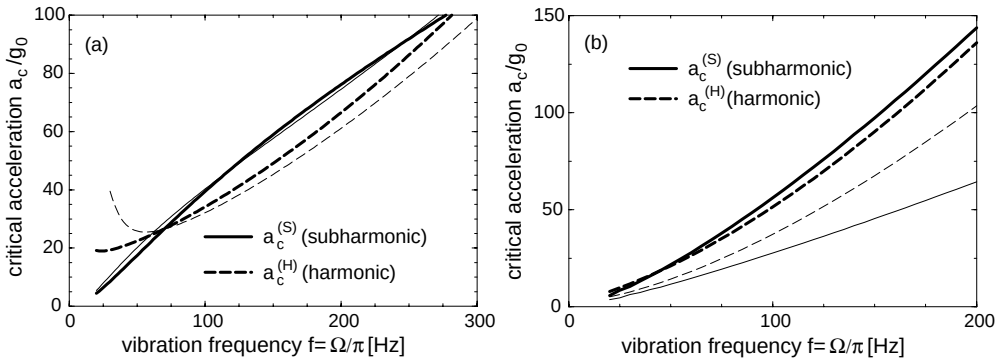

Fig. 2. – In part (a) the critical vibration amplitudes $a_c^{(S)}$ and $a_c^{(H)}$ for the subharmonic (thick solid line) and harmonic (dashed) Faraday instability in a Maxwell fluid are shown as a function of the vibration frequency (parameters as in fig. 1). Thin lines are the respective truncated solutions according to eqs. (5)-(7). Part (b) shows the onset amplitude for the harmonic and subharmonic Faraday instability calculated for the viscosity spectra of 1.5% polyacrylamide in a 50/50 mixture of water and glycerin (thin line) and for a mixture of 2% polyisobutylene in primol, a pharmaceutical grade white oil with $\nu = 1.5$ P (thick line). Viscometric data are taken from ref. [14].

Instead of using the complex viscosity for the Maxwell model, cf. eq. (8), any experimentally measured spectra for $\nu'(\Omega)$ and $\nu''(\Omega)$ may be used in our calculations. The corresponding result for two polymeric solutions is given in fig. 2b. For the polyisobutylene mixture (thick line) the harmonic instability is found to preempt the subharmonic as predicted by the Maxwell model. Experimental results which support the present theoretical results will be presented in a following paper [17].

Figure 3a indicates that the harmonic response is preferred in a limited range of the (λ, Ω)-plane around the curve $\Omega\lambda = 1$. For given surface tension α the frequency window of the harmonic response may shrink to zero by changing the static viscosity or the relaxation time. The critical values where this happens are shown in the (λ, ν_0)-plane in fig. 3b for

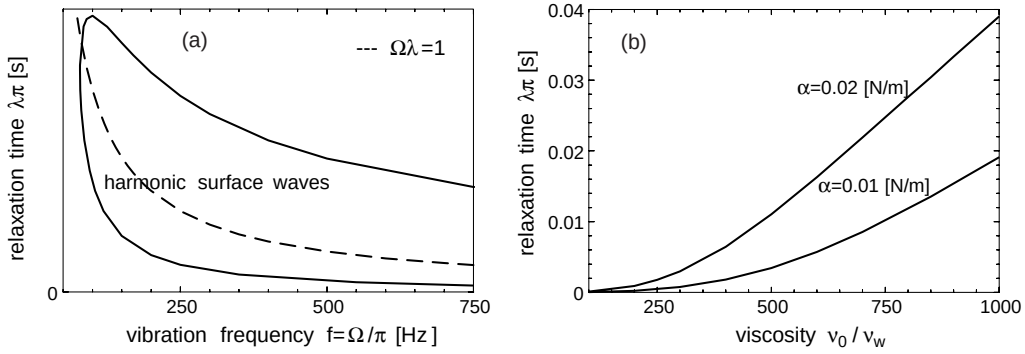

Fig. 3. – In part (a) the range is shown where the harmonic branch determines the primary instability (between the solid lines). Parameters for the Maxwell model: $\nu_0 = 5 \times 10^{-4}\,\mathrm{m}^2/\mathrm{s}$, others as in fig. 1. Along the dashed line one has $\Omega\lambda = 1$. Below each curve in part (b) there is a finite frequency range where the harmonic response is preferred for the Maxwell model. $\nu_w = 10^{-6}\,\mathrm{m}^2/\mathrm{s}$ is the viscosity of water.

different values of α. Since harmonic waves are preferred below each curve in fig. 3b, large static viscosities and small relaxation times are favorable for a harmonic response.

Conclusion. – The Faraday instability in viscoelastic surface waves occurs in harmonic resonance with the external drive if the vibration frequency comes close to the inverse elastic relaxation time. We have shown this in detail for Maxwell fluids, but it is not a specific property of this model. Calculations with empiric viscosity data confirm this observation. A large static viscosity, often met in polymeric solutions, and a small relaxation time are favorable for the observation of the harmonic response. It should be emphasized that the predicted harmonic response is a viscoelastic bulk effect. This is in contrast to a similar observation in a thin layer of a Newtonian fluid due to increasing boundary layer dissipation [7].

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REFERENCES

[1] WESFREID J. E., BRAND H. R., MANNEVILLE P., ALBINET G. and BOCCARA N., *Propagation in Systems Far from Equilibrium* (Springer, Berlin) 1988; CROSS M. C. and HOHENBERG P. C., *Rev. Mod. Phys.*, **65** (1993) 851.

[2] BUSSE F. H., in *Hydrodynamic Instabilities and the Transition to Turbulence*, edited by H. L. SWINNEY and J. P. GOLLUB (Springer, New York) 1981.

[3] DIPRIMA R. C. and SWINNEY H. L., in *Hydrodynamic Instabilities and the Transition to Turbulence*, edited by H. L. SWINNEY and J. P. GOLLUB (Springer, New York) 1981.

[4] KUDROLLI A. and GOLLUB J. P., *Physica D*, **97** (1996) 133; BINKS D. and VAN DE WATER W., *Phys. Rev. Lett.*, **78** (1997) 4043; CHEN P. and VINALS J., *Phys. Rev. Lett.*, **79** (1997) 2670; ZHANG W. and VINALS J., *J. Fluid Mech.*, **336** (1997) 301; **341** (1997) 225.

[5] KUMAR K. and TUCKERMAN L. S., *J. Fluid Mech.*, **279** (1994) 49; KUMAR K., *Proc. R. Soc. London, Ser. A*, **452** (1996) 1113.

[6] CERDA E. and TIRAPEGUI E., *Phys. Rev. Lett.*, **78** (1997) 859.

[7] MÜLLER H. W., WITTMER H., WAGNER C., ALBERS J. and KNORR K., *Phys. Rev. Lett.*, **78** (1997) 2357.

[8] VEST C. M. and ARPACI V. S., *J. Fluid Mech.*, **36** (1969) 613; SOKOLOV M. and TANNER R. I., *Phys. Fluids*, **15** (1972) 534.

[9] BRAND H. R. and ZIELINSKA B. J. A., *Phys. Rev. Lett.*, **57** (1986) 3167; ZIELINSKA B. J. A., MUKAMEL D. and STEINBERG V., *Phys. Rev. E*, **33** (1986) 1454.

[10] LARSON R. G., SHAQFEH E. S. and MULLER S. J., *J. Fluid Mech.*, **218** (1990) 537; LARSON R. G., *Rheol. Acta*, **31** (1992) 213.

[11] GROISMAN A. and STEINBERG V., *Phys. Rev. Lett.*, **77** (1996) 1480; **78** (1997) 1460; *Elastic vs. inertial instability in Couette-Taylor flow of a polymer solution: Review*, preprint.

[12] BIRD R. B., ARMSTRONG R. C. and HASSAGER O., *Dynamics of Polymeric Liquids*, Vol. **I** (Wiley, New York) 1987.

[13] PLEINER H., HARDEN J. L. and PINCUS P., *Europhys. Lett.*, **7** (1988) 383.

[14] HUPPLER J. D., ASHARE E. and HOLMES L. A., *Trans. Soc. Rheol.*, **11** (1967) 159. The viscometric data are also presented on p. 118 of ref. [12].

[15] REHAGE H. and HOFFMANN H., *J. Phys. Chem.*, **92** (1988) 4712; *Mol. Phys.*, **74** (1991) 933.

[16] FISCHER P. and REHAGE H., *Langmuir*, **13** (1997) 7012.

[17] WAGNER C., MÜLLER H. W. and KNORR K., in preparation.