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## LETTER TO THE EDITOR

# The undulation mode of freely suspended liquid films

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**Abstract.** We argue that for real, freely suspended liquid films the undulation mode (transverse sound mode) has a linear dispersion relation of the usual sound-like structure, i.e. a damping contribution proportional to the wavevector squared ( $k^2$ ). Thus, there is no need to introduce non-linear fluctuation effects, which would be necessary if the regular damping mechanism ( $\sim k^2$ ) were absent, and which would lead to singular contributions in the damping.

In recent years there has been growing interest in the physical properties of liquid films. They are studied because of their special material properties, because of the novel structures and phases they can form, or in order to investigate the influence of dimensionality on the fundamental behaviour of liquids. Very widespread are thin (at least bilayer) smectic liquid crystal films, bilayer membranes of amphiphiles in lyotropic systems, monolayer interfaces in micellar systems, Langmuir-Blodgett films on a liquid or solid support [1], or soap films [2].

Here we discuss the transverse dynamics (undulation mode or transverse sound mode) of those real, liquid films in the hydrodynamic limit. In contrast to two recent publications we argue that there *are* internal dissipative processes within the liquid films, which provide the usual sound-like damping ( $\text{Im } \omega \sim k^2$ ) of the undulation mode with frequency  $\omega$  and wavevector  $k$ , even in the absence of any surrounding medium. If there were no damping  $\sim k^2$  present, the non-linearities in the hydrodynamic equations would provide the coupling to (or scattering at) thermal fluctuations giving rise to an anomalous, renormalized dispersion relation ( $\text{Im } \omega \sim k^3$  [3] or  $\sim k^4 \omega^{-1/3}$  [4]). However, the presence of the ordinary damping mechanism in real films makes it very unlikely that such damping processes due to non-linear fluctuations are of any importance.

On the other hand, for mathematical, i.e. ideally two-dimensional surfaces, the arguments in [3] and [4] apply and there is no damping of the order  $k^2$ . But in addition, there is no sound-like propagation ( $\text{Re } \omega \sim k$ ) in such mathematical surfaces due to the lack of surface tension. In the next section we argue in detail, why in real liquid films the undulation mode has the ordinary sound-like dispersion relation.

Apart from the usual hydrodynamic variables (density  $\rho$ , momentum density  $\rho v$  and energy density  $\epsilon$ ) necessary to describe a simple fluid, a film is additionally

characterized by a displacement variable  $u$  describing the non-equilibrium position of the film. In equilibrium  $u \equiv 0$  and the film is assumed to be flat (e.g. given by  $z = \text{constant}$ ) and to have thickness  $d_0$ . In the incompressible limit the transverse momentum conservation is then expressed by the linearized equation (gravity neglected)

$$\rho \dot{v}_z - \nabla_{\perp} \Phi = \nu_2 \Delta_{\perp} v_z \quad (1)$$

where the subscript  $\perp$  denotes components transverse to the film. Since we are dealing with thin films, any dependence on the position across the film can be neglected; for the same reason one displacement field  $u$  is sufficient for the description. In equation (1)  $\nabla_{\perp} \Phi = (\delta/\delta u_x) \int \epsilon \, dV$ , with  $\epsilon$  the elastic free energy density, describes the elastic response of the film to shear flow. Shearing the film, however, also means stretching and compressing the film (figure 1), thus increasing the surface and decreasing the thickness. In this non-equilibrium state an elastic restoring force occurs due to compressional elasticity (in smectic systems), due to curvature elasticity (in smectic films and amphiphilic membranes), and due to surface tension (in any liquid film). Since compression due to shear is a secondary effect, i.e. the change in thickness is quadratic ( $\sim [\nabla_{\perp} u_x]^2$ ), the appropriate compressional elastic energy is then quartic in the shear and does not give rise to an elastic restoring force in a linearized theory [5]. On the other hand the change in surface energy due to shear is proportional to the change of surface area [6] (i.e. quadratic in the shear) thus giving rise to a linear elastic force  $\nabla_{\perp} \Phi_s = \gamma \nabla_{\perp}^2 u_x$ , where  $\gamma d_0$  is the surface tension. In addition, bending gives a linear restoring force  $\nabla_{\perp} \Phi_b = -K \Delta_{\perp}^2 u_x$ , where  $K$  is the splay elastic constant  $K_1$  in smectic systems [7] or the bending coefficient  $\kappa$  in amphiphilic bilayers [8].

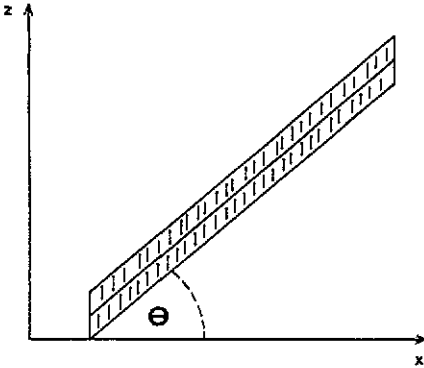


Figure 1. Cross section of a bilayer smectic film after constant shearing ( $\nabla_x u_x = \text{constant}$ ) showing stretching (length  $l = l_{eq} / \cos \Theta$ ) and compression (thickness  $d = d_{eq} \cos \Theta$ ) with  $\tan \Theta = \nabla_x u_x$ , where subscripts eq refer to equilibrium quantities.

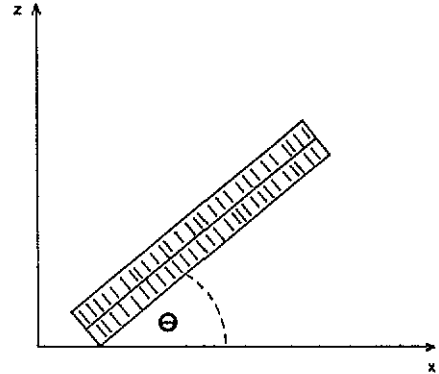


Figure 2. Cross section of a bilayer smectic film after a constant rotation leading to an equilibrium state.

The viscous contribution ( $\nu_2$ ) in equation (1) describes dissipative processes within the film during shearing. It has been argued in [3] and [4] that those dissipative processes have to be absent, because for films, shearing and rotating would be equivalent

processes. Obviously, this is not true for real films. Figure 2 shows the film after a constant rotation in a perfect equilibrium state, where no elastic or dissipative restoring forces arise. The state obtained after shearing (figure 1), however, is a non-equilibrium state and dissipation occurs during the relaxation into any equilibrium state. Thus, shearing and rotation of a real film lead to fundamentally different states. This holds for smectic systems as well as for soap films and amphiphilic membranes. As usual the shear viscosity can be attributed to relative motions of the molecules on the molecular length scale and is an intrinsic and unavoidable property of any real liquid film. It is not related to changes of the surface area or of the film thickness, but is proportional to the shear flow.

For liquid films the (diffusional) motion within the film is decoupled from the transverse film motion and need not be considered here. Then equation (1) is sufficient to describe momentum conservation and reads ( $\Phi = \Phi_s + \Phi_b$ )

$$\rho \dot{v}_z - \gamma \Delta_{\perp} u_z + K \Delta_{\perp}^2 u_z = \nu_2 \Delta_{\perp} v_z. \quad (2)$$

In smectic systems there is an additional dissipative process, permeation, the diffusion of molecules across the layers [9]. It is related to the deformation of the layers. A similar process occurs in soap films, where soap molecules dissolved in the water layer diffuse to the amphiphilic monolayers (or vice versa), if the film is distorted (Gibbs diffusion [10]). As discussed above, layer compression is a quadratic effect and thus only bending can lead to permeation or Gibbs diffusion in the linear regime. Since we have already considered a dissipative process (shear diffusion), which is of lower order in the gradients, and since we are dealing with thin films, we will neglect permeation and Gibbs diffusion due to bending. Then we are left with the simple kinematic relation between the transverse film velocity and the displacement

$$\dot{u}_z = v_z. \quad (3)$$

Equations (2) and (3) lead in the long wavelength limit to a transverse (undulation) mode of the usual sound-like dispersion relation

$$\omega = \pm \left( \frac{\gamma}{\rho} \right)^{1/2} k_{\perp} + \frac{i}{2} \frac{\nu_2}{\rho} k_{\perp}^2. \quad (4)$$

Of course, any friction between the film and the surrounding medium (air, water etc) gives rise to additional damping contributions of up to order  $k_{\perp}^2$ . The considerations presented here apply not only to isotropic films but equally well to anisotropic ones, e.g. films of tilted smectics. In this case there are more elastic and viscous coefficients involved.

Thus non-linear fluctuation effects are not *a priori* necessary to be considered for real films, only in the special case that  $\nu_2$  is extraordinarily small.

For mathematical films, which are infinitesimally thin (and thus also of laterally infinite extent in order to have a non-vanishing total mass), there is no compression and no stretching during shear and thus no compressional elasticity and no surface tension ( $\gamma \equiv 0$ ). But such a system can have curvature elasticity ( $K \neq 0$ ).

Since shearing and rotating are identical operations for mathematical films, it is clear that shear viscosity cannot occur as a dissipative process, because (homogeneous) rotations must not lead to any entropy production (a homogeneously rotated system

is still in an equilibrium state). Thus,  $\nu_2 \equiv 0$  and in the momentum conservation equation only inhomogeneous shear diffusion is allowed—a process also present in physical systems, but usually neglected against ordinary shear viscosity. Therefore, in the case of mathematical films equation (2) is replaced by

$$\rho \dot{v}_z + K \Delta_{\perp}^2 u_z = -\xi_2 \Delta_{\perp}^2 v_z. \quad (5)$$

This leads together with equation (3) to the dispersion relation

$$\omega = \pm \left( \frac{K}{\rho} \right)^{1/2} k_{\perp}^2 + \frac{i}{2} \frac{\xi_2}{\rho} k_{\perp}^4 \quad (6)$$

which is not a sound-like excitation. Thus, in the limit of vanishing film thickness the sound-like behaviour (4) is replaced by (6). Whether one can reach this transition in a real system and, if so, at what thickness, depends on the specific nature of the film considered (soap, membrane, smectic etc), since the thickness dependence of the relevant parameters ( $\gamma$ ,  $K$ ,  $\nu_2$ ,  $\xi_2$ ) is not universal. In addition, such a transition also depends on the wavelength of the excitation considered and could be expected for e.g.  $k_{\perp} \approx (\gamma/K)^{1/2}$  or  $k_{\perp} \approx (\nu_2/\xi_2)^{1/2}$ .

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