Dynamics of freely suspended films with surface tension

Yatin Marathe, Sriram Ramaswamy

To cite this version:

Yatin Marathe, Sriram Ramaswamy. Dynamics of freely suspended films with surface tension. Journal de Physique, 1990, 51 (19), pp.2143-2152. <10.1051/jphys:0199000510190214300>. <jpa-00212518>

HAL Id: jpa-00212518
https://hal.archives-ouvertes.fr/jpa-00212518
Submitted on 1 Jan 1990

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Dynamics of freely suspended films with surface tension

Yatin Marathe (1) and Sriram Ramaswamy (1,2)

(1) Department of Physics, Indian Institute of Science, Bangalore 560 012, India
(2) Centre for Theoretical Studies, Indian Institute of Science, Bangalore 560 012, India

(Received on March 21, 1990, accepted in final form on June 18, 1990)

Abstract. — We have studied the hydrodynamics of freely suspended membranes, liquid as well as crystalline, with surface tension. We find that nonlinear coupling to thermally excited undulations gives a singular contribution to the kinetic coefficients of these systems at low frequency and wavenumber. Our results differ in some important respects from those of Katz and Lebedev on this problem, and can be tested in mechanical impedance as well as time-correlation studies.

1. In this paper, we study the dynamics of a single freely suspended membrane in vacuum (so that the surrounding medium does not damp the fluctuations), liquid or crystalline, using the methods of fluctuating hydrodynamics. These membranes are two dimensional objects which are allowed to fluctuate in the third (z) direction. The out-of-plane fluctuations of these membranes are governed by a surface tension which arises because these membranes are coupled to a reservoir. We argue below that, in linearised hydrodynamics, spatial variations in the z-component of the velocity field \( v_z \) of the membrane should enter the dissipation function only in the form \( \Gamma_0 (\nabla^2 v_z)^2 \), because a uniform tilt produces no relative motion of molecules in the bulk of the membrane. If fluctuations and nonlinearities are taken into account, the coefficient \( \Gamma_0 \), as well as the viscosities \( \eta \) and \( \zeta \) governing the damping of the in-plane components of \( v \), acquire frequency-dependent parts \( (\Delta \Gamma (\omega), \Delta \eta (\omega), \Delta \zeta (\omega)) \) which we have calculated. We find, to lowest order in a perturbative self-consistent scheme that

\[
\Delta \Gamma (\omega) \sim (k_B T)^{2/3} \rho_0^{1/3} a^{4/3} \omega^{-1/3}.
\]  

Here \( \rho_0 \) is the two-dimensional density, \( a \) is a microscopic length, \( \omega \) is the frequency, \( T \) is the temperature and \( k_B \) is Boltzmann's constant. Our results differ in an important way from those of Katz and Lebedev [1] on the same model. They calculate only the wavenumber dependences of the damping coefficients, and find (correctly) that \( \Gamma (q \neq 0, \omega = 0) \sim q^{-1} \). They take this to mean that the undulation mode has a damping rate \( \Gamma q^4 \sim q^3 \). We shall show that for small \( q \) and \( \omega \) near only hydrodynamic pole, \( \Gamma (q \neq 0, \omega \neq 0) \) is better approximated...
by \( \Gamma (q = 0, \omega \neq 0) \) (which is found to go as \( \omega^{-1/3} \)) than by \( \Gamma (q \neq 0, \omega = 0) \) \( (\sim q^{-1}) \). The consequences of this are discussed in section 5. The shear and bulk viscosities for two dimensional system go as

\[
\Delta \eta (\omega) \sim \omega^{-2/3} \\
\Delta \xi (\omega) \sim \omega^{-2/3}.
\]

The same results hold for a crystalline membrane with surface tension.

The paper is organised as follows: in section 2, we describe the elasticity theory of a membrane with surface tension and show that the static properties of membrane do not show any dramatic behaviour. In section 3 we develop the equations of fluctuating hydrodynamics for the membrane and calculate the mode structure at the linearised level. In section 4, we calculate the effective frequency- and wavenumber-dependent damping coefficients arising from nonlinear coupling to thermally excited undulations. We conclude, by explaining the disagreement between our results and those of Kats and Lebedev.

2. Elasticity.

We now describe the static properties of freely suspended two-dimensional films with surface tension. More precisely, we construct the elastic free-energy functional for distortions of the membrane. The deformations to which the membrane responds elastically are (1) undulations of its surface (capillary waves); (2) intrinsic two-dimensional compressions or dilations in the plane of the membrane; and (3) (for crystalline membranes) shear in the plane of the membrane. We assume that an undulated configuration can be described by a height function \( h(r) \) which measures the local departure of the membrane from an undistorted flat reference state. Compressions and dilations are described by the two-dimensional density field \( \rho (r) \), with average value \( \rho_0 \). Finally, for crystalline sheets, distortions of the lattice are described by a lattice displacement field \( u(r) = (u_1(r), u_2(r)) \). We shall always treat our variables as functions of the coordinates \((x, y) = r\) on a flat reference plane. Therefore the area element in an integral of a physical quantity will be \( \sqrt{1 + (Vh)^2} \, d^2r \), where the Jacobian factor \( \sqrt{1 + (Vh)^2} \) relates the area of a piece of surface to the area \( d^2r \) of its projection onto \( D \). In particular, the total area of membrane in the experimental region is

\[
A = \int_D \sqrt{1 + (Vh)^2} \, d^2r
\]

and hence the total number of membrane molecules is

\[
N = nA = n \int \sqrt{1 + (Vh)^2} \, d^2r, \quad \text{where } n \text{ is the number of particles per unit intrinsic area of the membrane.}
\]

Let us now construct the free-energy functional or effective hamiltonian \( F \) for these variables. We begin by emphasising that the membrane is assumed to be coupled to a reservoir of molecules, so that the number of particles within the experimental region can fluctuate. These fluctuations are governed by a chemical potential, leading to a term

\[
\mu N = \mu n \int \sqrt{1 + (Vh)^2} \, d^2r = \sigma \int \sqrt{1 + (Vh)^2} \, d^2r \text{ in } F.
\]

Next, we know that the membrane resists changes in its intrinsic mass density (at constant \( N \)). This implies a term \( A/2 \int \sqrt{1 + (Vh)^2} \, (\delta \rho / \rho)^2 \, d^2r \) in \( F \), where \( A \) is the inverse isothermal compressibility of the membrane and \( \delta \rho = \rho - \rho_0 \). Finally, if there is crystalline order in the membrane then we must include an elastic energy cost for distortions of the two-dimensional lattice structure. We shall assume the crystal-structure has hexagonal symmetry, so that the elasticity is
isotropic, involving only two elastic constants. However, the strain tensor for a two
dimensional sheet suspended in three-dimensions involves both the in-plane displacement
field u and the height function h. The reason for this is simple, and lies in the definition of
u. Let us understand this for a one-dimensional chain imbedded in two dimensions. Begin
with a flat, undistorted chain. Then u = 0 and h = 0. Now tilt the chain uniformly, through a
small angle \( \theta \) so h \( \sim \) x\( \theta \). Keep in mind that the u-field is the departure of the x-coordinate of
each point from its mean position. A point originally at x will now be at x(\( \theta \))
\( 1 - \theta^2 /2 \), so that u(x) = x(\( \theta \)) - x \( \sim \) - x\( \theta^2 /2 \). Had we ignored the coupling of u to
h, we might have written down an elastic energy of the form \( \int (du/dx)^2 dx \), as a result of
which the pure tilt described above would have cost elastic energy. This is clearly unphysical.
We must therefore subtract, from du/dx, the appropriate function of derivatives of
h so that a pure tilt costs no energy.

If we choose \( du \) dx + \( 1 / 2 \) \( (dh/dx)^2 \) as the strain-field, we see that for a pure rotation,
du/dx \( \sim \) \( - \theta^2 /2 \) and dh/dx \( \sim \) - \( \theta \), so that the above combination is zero. That this works
without a small-angle assumption is shown in reference [2]. The generalisation of this to the
case of a two-dimensional sheet in three dimensions is also given there. They show that the
appropriate strain tensor

\[
E_{ij} = \frac{1}{2} (\nabla_i u_j + \nabla_j u_i + \nabla_i h \nabla_j h)
\]

for distortions of the lattice structure and the corresponding elastic energy is

\[
F_e = \frac{\mu}{2} \int E_{ij}^2 \sqrt{1 + (\nabla h)^2} d^2 r + \lambda \int E_{ii}^2 \sqrt{1 + (\nabla h)^2} d^2 r.
\]

Thus, for a fluid membrane, the effective Hamiltonian \( F \) is

\[
F_t = \int d^2 r \sqrt{1 + (\nabla h)^2} \left[ \sigma_0 + \frac{A_0}{2} \left( \frac{\delta \rho}{\rho_0} \right)^2 \right]
\]

and for a crystalline membrane, it is

\[
F_e = F_t + F_e + c \int d^2 r \sqrt{1 + (\nabla h)^2} E_{ii} \frac{\delta \rho}{\rho_0}.
\]

Of course, in writing \( F \), we have neglected terms of higher order in the gradient expansion,
since we know that these are less important, for long-wavelength modes, than the leading
terms. For example, we have chosen to ignore the curvature energy [3]

\[
\int (\nabla^2 h)^2 \sqrt{1 + (\nabla h)^2} d^2 r,
\]

in comparison to the surface tension term, which, when expanded
for small h, is \( \frac{\sigma_0}{2} \int (\nabla h)^2 d^2 r \). The former is of order \( \nabla^4 \) and hence is clearly smaller at long
wavelengths than the latter, which is of order \( \nabla^2 \).

Before embarking on a calculation of the hydrodynamical properties of our system, let us
ask if the static properties show any interesting features. Such features, if any, would arise
from the fact that the elastic free-energy for the membrane contains terms of higher than
quadratic order in h and u. Without a detailed calculation, we assert that in perturbation
theory, all fluctuation corrections to the elastic constants arise from graphs whose internal
lines are correlation functions of $V_h$, $V_u$ and $\delta \rho$. None of these is singular at long wavelengths. (For example, the Fourier transformed $\langle hh \rangle$ correlation function, at harmonic order, goes as $1/\sigma_0 q^2$ at small wavenumber $q$. This means that the Fourier transform of $\langle V_h V_h \rangle$ is $q$-independent at small $q$.)

As a result, the fluctuation corrections to the elastic constants $\mu$, $\lambda$, $\sigma$, $A$ have finite limits at zero wavenumber. We shall therefore ignore them in our calculations, and move on to a calculation of the damping coefficients of the modes of the membrane.

3. Hydrodynamics.

We now derive the equations of hydrodynamics, that is, the equations describing the dynamics of our membrane systems on large length and time-scales. First, let us decide on the variables for such a description [4]. The laws of conservation of momentum, mass and energy imply that the momentum density $q$, the mass density $\rho$ and the energy density $\varepsilon$ are hydrodynamic variables. The height variable also relaxes hydrodynamically because of the symmetry $h \rightarrow h + \text{constant}$. In addition for crystalline membranes the in-plane displacement field $u$, a « broken-symmetry » variable, must be included.

The equations of motion for the system are most conveniently derived using Poisson brackets. By analogy with earlier calculations in smectic liquid crystals [5] or in ordinary crystals [6], we may assert that the non-vanishing Poisson brackets are

$$\{g_i(\vec{r}), h(\vec{r}')\} = (\delta - \nabla h) \delta (\vec{r} - \vec{r}') \hspace{1cm} \{g_a(\vec{r}), u_b(\vec{r}')\} = (\delta_{ab} - \nabla_a u_b) \delta (\vec{r} - \vec{r}') \hspace{1cm} \{g(\vec{r}), \rho(\vec{r}')\} = -\rho(\vec{r}) \nabla \delta (\vec{r} - \vec{r}') \hspace{1cm} \{g_i(\vec{r}), g_j(\vec{r}')\} = -\nabla_i[\delta (\vec{r} - \vec{r}') g_i(\vec{r})] + \nabla_i[\delta (\vec{r} - \vec{r}') g_j(\vec{r}')].$$

For a set of « slow » variables $\{\phi_a\}$, the equations of motion, including dissipation and thermal fluctuations are

$$\frac{\partial \phi_a}{\partial t} + \{\phi_a, \phi_b\} \frac{\partial H}{\partial \phi_b} + \Gamma_{ab} \frac{\partial H}{\partial \phi_b} + k_B T \frac{\partial}{\partial \phi_b} \{\phi_a, \phi_b\} = f_a(t).$$

where repeated indices are summed over. Here $H$ is the effective Hamiltonian governing the $\{\phi_a\}$ ($H = -k_B T \ln P_{eq}$ where $P_{eq}$ is the equilibrium probability distribution for the $\{\phi_a\}$), $(\Gamma_{ab})$ is a matrix of kinetic coefficients, and the effects of thermal fluctuations are contained in the noise $f$ which satisfies

$$\langle f_a(t) \rangle = 0 \hspace{1cm} \langle f_a(t) f_b(t') \rangle = 2k_B T \delta_{ab} \delta(t-t').$$

For the case at hand, the free-energy functional is simply that for the variables which entered the statics (see Eqs. (2.3) and (2.4)) together with a kinetic energy term :

$$H = \int d^2 r \frac{g^2}{2 \rho} + F$$

where $F$ is $F_t$ (Eq. (2.3)) for liquid membranes and $F_c$ (Eq. (2.4)) for crystalline membranes.
For both fluid and crystalline membranes, the equations of motion for \( h \) and \( g_z \) are

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot g = 0
\]

\[
\frac{\partial h}{\partial t} - V_z + \nabla \cdot \nabla h = 0
\]

\[
\frac{\partial g_z}{\partial t} + \nabla (g_z \nabla) + \Gamma_0 \nabla^4 V_z + \frac{\delta F}{\delta h} = f_z(\mathbf{r}, t).
\] (3.5)

For crystalline membranes, the equation of motion for the in-plane components \( g_i \) of the momentum density is (keeping in mind that the system is two-dimensional)

\[
\frac{\partial g_i}{\partial t} = \nabla (g_i \nabla) - \eta_0 \nabla^2 V_i - \zeta_0 \nabla (\nabla \cdot \nabla V_i) + \rho \nabla \frac{\delta F}{\delta \rho} + (\delta_{ij} - \nabla_i \nabla_j) \frac{\delta F}{\delta u_j} - \nabla_i h \frac{\delta F}{\delta h} = f_i(\mathbf{r}, t)
\]

\[
\frac{\partial u_i}{\partial t} + \nabla \cdot \nabla u_i - V_i + \gamma \frac{\delta F}{\delta u_i} = \theta_i(\mathbf{r}, t)
\] (3.6)

while for fluid membranes, the last equation as well as the last term on the left-hand side of (3.5) are absent since there is no in-plane displacement field. In the above, \( g \) is the in-plane momentum density and \( v = g / \rho \) the in-plane velocity field, \( F \) is \( F \) for fluid membranes and \( F \) for crystalline membranes, \( \nabla \) is the gradient operator in the \( x-y \) plane, and all subscripts \((i,j)\) run over \( x \) and \( y \) only, \( \eta_0 \) and \( \zeta_0 \) are the bare viscosity coefficients damping in-plane gradients in \( v \), and \( \gamma \) is a bare damping coefficient for \( u \). There is no \((\nabla^2)\) damping for \( v \) for the reasons stated in section 1. The thermal noise sources \( f_i \) and \( f_z \) are Gaussian with zero mean and satisfy

\[
\langle f_i(F, t) f_j(F', t') \rangle = \{ 2 k_B T \delta_{ij} \nabla^2 - (\rho_0 + \eta_0 / 2) \nabla_i \nabla_j \delta(F - F') \delta(t - t') \}
\]

\[
\langle f_z(\mathbf{r}, t) f_z(F', t') \rangle = 0
\]

\[
\langle f_z(\mathbf{r}, t) f_z(F', t') \rangle = 2 k_B T \Gamma_0 \nabla^4 \delta(F - F') \delta(t - t').
\] (3.7)

3.1 LINEARISED THEORY : DISPERSION RELATIONS. — If we linearise the above equations and Fourier-transform than in space and time, the resulting dynamical matrix for fluid membranes will have eigenvalues

\[
\omega_{\pm}(q) = \pm c_L q - \frac{i(\xi_0 + \eta_0)}{2 \rho_0} q^2
\] (3.8)

which is in-plane longitudinal sound,

\[
\omega_h(q) = \pm c_h q - i (\Gamma_0 / \rho_0) q^4
\] (3.9)

which is the out-of-plane undulation mode, and

\[
\omega_s(q) = - i (\eta_0 / \rho_0) q^2
\] (3.10)

which describes the diffusive transport of shear within the layer. For a crystalline layer, the form of (3.8) to (3.9) is unchanged but (3.10) is replaced by

\[
\omega_t(q) = \pm c_t q - i (\eta_0 / \rho_0) q^2
\] (3.11)
because the layer can support shear waves. In addition there is an in-plane vacancy diffusion mode

\[ \omega_D(q) = -i \gamma (\lambda + 2 \mu) q^2 \]  \hspace{1cm} (3.12)

which will not play a significant role in this work. In the above equations,

\[ c_h = \sqrt{\frac{\sigma}{\rho_0}} \]
\[ c_t = \sqrt{\frac{\mu}{\rho_0}} \]
\[ c_L = \sqrt{\frac{A/\rho_0}{A + \lambda + 2 \mu - 2 c}} \]  \hspace{1cm} (3.13)

Note the form of the damping in (3.9). There is no contribution to \( \text{Im} (\omega_h(q)) \) at order \( q^2 \) for the reasons stated in section 1.

3.2 LINEARISED THEORY : CORRELATION FUNCTIONS. — The equations (3.5) to (3.6) can also be used to calculate the correlation functions of the hydrodynamic variables. When Fourier transformed in space and time, these are

\[ \langle h(q, \omega) h(-q_1 - \omega) \rangle = \frac{2 k_B T(\Gamma_0/\rho_0) q^4}{\omega^2 - \left( \frac{\sigma_0}{\rho_0} \right)^2 q^2 + \omega^2 \left( \frac{\Gamma_0}{\rho_0} \right)^2} \]
\[ \langle g_z(q, \omega) g_z(-q, -\omega) \rangle = \frac{2 k_B T(\Gamma_0/\rho_0) q^4 \omega^2}{\omega^2 - \left( \frac{\sigma_0}{\rho_0} \right)^2 q^2 + \omega^2 \left( \frac{\Gamma_0}{\rho_0} \right)^2} \]  \hspace{1cm} (3.14)

Note several features of these correlation functions.

(i) They are all singular at \( \omega = q = 0 \), a sign of the hydrodynamic nature of the variables.

(ii) When integrated over all frequencies, they yield the equal-time or static correlation functions.

(iii) As an inevitable consequence of the linearised treatment their decay in time is exponential.

In the next section, we shall see how these correlation functions are changed when the nonlinear terms in the equations of motion are taken into account.

4. Effect of nonlinearities.

4.1 GENERALITIES. — The presence of nonlinear terms in the hydrodynamic equations means, of course, that the various Fourier components are in fact coupled to each other. We can therefore no longer calculate dispersion relations and correlation functions as simply as for the linearised problem. However, the effect of the nonlinearities can be summarised very simply. The correlation functions and dispersion relations are given by the expressions (3.8-3.12) and (3.14) with one difference: the kinetic coefficients \( \eta_0, \rho_0 \) and \( \Gamma_0 \) must be replaced by frequency and wavenumber dependent quantities \( \eta(q, \omega), \rho(q, \omega) \) and \( \Gamma(q, \omega) \). These generalised kinetic coefficients must be calculated by solving the full nonlinear equations of motion but one statement can already be made: the fact that they are frequency dependent.
means that the decay in time is not exponential. We now outline the calculation of the kinetic\ncoefficients.

4.2 RELEVANT NONLINEARITIES. — The mechanism whereby nonlinearities give rise to\nfrequency and wavenumber-dependent contributions (« renormalisations ») to transport\ncoefficients is scattering from thermal fluctuations. Let us understand this process. Suppose we\ntry to measure the shear viscosity by propagating in the membrane a sound wave with\nfrequency $\omega$ and wavevector $q$. The membrane is at some nonzero temperature and,\ntherefore, pairs of thermally excited modes with equal and opposite wavevectors and\nfrequencies are constantly appearing and recombining. The sound wave (provided $g$ is coupled nonlinearly to the mode in question, in equations (3.6) and (3.7)) will be\nscattered by these thermal excitations. The longer the lifetime of the modes, the more\npronounced this scattering. This scattering will ultimately give rise to renormalised transport\ncoefficients. Now of all the hydrodynamic mode in our system, the undulations decay most\nslowly (lifetime $\sim q^{-4}$). We need therefore include only the effect of the undulation\n($h$) modes. Looking at equations (3.6) and (3.7), we see that $g_z$ couples to $h$ through the term\n$\delta F / \delta h$, whose leading nonlinear part is

$$\sigma \frac{1}{2} \nabla_j (\nabla h (\nabla h)^2).$$  \hspace{1cm} (4.1)

Similarly in the equations for $g_i$, nonlinear couplings to $h$ arise

$$- \nabla_j h \frac{\delta F}{\delta h} = - \sigma \nabla_j h \nabla^2 h + \text{higher order}$$  \hspace{1cm} (4.2)

and the $h$-dependent nonlinear part of $\delta F / \delta u_i$

$$- \delta F / \delta u_i = \mu \nabla_j (\nabla h \nabla h) + \frac{\lambda}{2} \nabla_j (\nabla h)^2.$$  \hspace{1cm} (4.3)

We can calculate the effect of these nonlinearities by generalising the Kubo formula.\nEssentially, $\Gamma_0$ is the order $q^2$ part of a viscosity (the order-1 part being zero for the reasons\nstated in Sect. 1). Therefore we should be able to calculate corrections to $\Gamma_0$ arising from $h$-\nfluctuations by taking the contribution of those $h$-fluctuations to the stress tensor,\'autocorrelating it and extract the order $q^2$ part. The nonlinear term can be written as

$$\nabla_j \sigma_{ji} = \nabla_j \left[ \frac{\sigma}{2} \nabla_j (\nabla h)^2 \right]$$  \hspace{1cm} (4.4)

in other words,

$$\sigma_{ji} = \frac{\sigma}{2} \nabla_j (\nabla h)^2.$$  \hspace{1cm} (4.5)

Therefore,

$$\Delta \Gamma(q, \omega) = \frac{1}{Ak_B T} \int \frac{d^2}{dq^2} \int \langle \sigma_{ji}(q, 0) \sigma_{ji}(-q, t) \rangle e^{i\omega t} dt.$$  \hspace{1cm} (4.6)

What about the order-1 term? Clearly it does arise. However, the rotation-invariance\nargument of section 1 means that we must renormalise the theory in such a way that that\ncoefficient is zero. This is analogous to the situation that arises in the problem of the statics of a\ncrystalline membrane with zero surface tension. There, fluctuation corrections arising from anharmonic couplings apparently generate in the Hamiltonian a term proportional to
However, we insist that the renormalised value of the coefficient of that term be zero. Similar and perhaps more familiar is the case of $T_c$-renormalisation in ordinary critical phenomena. There, the Hamiltonian is

$$
\int dr \left[ \frac{r_0}{2} \phi^2 + (\nabla \phi)^2 + \frac{\lambda}{4} \phi^4 \right]
$$

and the critical point is defined by a vanishing coefficient for the $\phi^2$ term. Clearly, if we begin with a Hamiltonian with $r_0 = 0$, the fluctuation connections will, by way of the $\phi^4$ term, generate terms quadratic in $\phi$. This does not mean that the critical point does not exist: it means only that at the critical point, the renormalised (fluctuation-corrected) value of $r$ is zero. Likewise, our model of a membrane with no order $q^2$ damping for $g_z$ is consistent, and all calculations must be carried out with the renormalised value of the order $-q^2$ damping equal to 0.

For the in-plane viscosities $\eta$ and $\zeta$, the cases of fluid and crystalline membranes must be handled separately. In the fluid case, the only coupling of $g$ to $h$ is the term $-\nabla_j h \delta F/\delta h = -\sigma \nabla_j h \nabla^2 h + \text{higher order in } (3.5)$. For crystalline membranes, this term is accompanied by the terms

$$\mu \nabla_j (\nabla_j h \nabla_j h) + \lambda \nabla_i (\nabla h)^2$$

arising from $\delta F/\delta u_i$. Thus, the nonlinear contribution of $h$ to the stress tensor is

$$\sigma_{ij}^F = \sigma_0 \nabla_i h \nabla_j h$$

for the fluid case and

$$\sigma_{ij}^c = \sigma_0 \nabla_i h \nabla_j h + \mu \nabla_i h \nabla_j h$$

for the crystalline case. Now $\Delta \eta$ and $\Delta \zeta$ are extracted simply by applying the usual Kubo formula [7] with equation (4.7) and equation (4.8) for the stresses:

$$\Delta \zeta = \frac{\delta_{ij} \delta_{kl}}{4 Ak_B T} \int_0^\infty dt e^{i\omega t} \langle \sigma_{ij}(q, t) \sigma_{kl}(0, 0) \rangle$$

$$\Delta \eta = \frac{\delta_{il} \delta_{jk} + \delta_{ik} \delta_{jl}}{8 Ak_B T} \int_0^\infty dt e^{i\omega t} \langle \sigma_{ij}(q, t) \sigma_{kl}(0, 0) \rangle.$$ (4.9)

4.3 PERTURBATION THEORY AND SELF-CONSISTENT APPROXIMATION. — When we rewrite equations (4.6) and (4.9) by replacing the stresses by their explicit forms in terms of $h$ (Eqs. (4.5), (4.7) and (4.8)) the resulting expressions are

$$\Delta \eta(q, \omega) = \frac{\sigma_0^2}{Ak_B T} \int dt e^{i\omega t} \langle [h(\nabla h)^2]_{q, t} [h(\nabla h)^2]_{-q, 0} \rangle$$

$$\Delta \zeta(q, \omega) = \frac{\delta_{il} \delta_{jk} + \delta_{ik} \delta_{jl}}{8 k_B TA} \sigma_0^2 \int dt e^{i\omega t} \langle (\nabla_i h \nabla_j h)_{q, t} (\nabla k h \nabla l h)_{-q, 0} \rangle$$

$$\Delta \zeta(q, \omega) = \frac{\delta_{ij} \delta_{kl}}{4 Ak_B T} \sigma_0^2 \int dt e^{i\omega t} \langle (\nabla_i h \nabla_j h)_{q, t} (\nabla k h \nabla l h)_{-q, 0} \rangle.$$ (4.10)

for the fluid membrane. A similar expression can be written down for the crystalline membrane using equations (4.8) and (4.9). These can be turned into relatively tractable
integral equations only if we can rewrite the right-hand sides in terms of products of two-point \( h \)-correlation functions. Then, since the pair correlation function for \( h \) is itself related to \( \Gamma(q, \omega) \)

\[
\langle h(q, \omega) h(q', \omega') \rangle = \frac{2 k_B T \Gamma(q, \omega)}{\rho_0^2} q^4 \delta(q + q') \delta(\omega + \omega') \left( \omega^2 - \frac{\sigma_0}{\rho_0} q^2 \right)^2 + \frac{\Gamma^2(q, \omega)}{\rho_0^2} \omega^2 q^8.
\]  

We see that (4.10) and (4.11) give an integral equation

\[
\Delta \Gamma(q, \omega) = \frac{\sigma_0^2}{k_B T} \int d\omega' q' q'' \left( \frac{2 k_B T \Gamma}{\rho_0} \right)^3 q^6 q^8 |\bar{q} - q' - \bar{q}''|^4 \int R
\]

\[
R = \left\{ \left[ \left( \omega^2 - \frac{\sigma_0}{\rho_0} q^2 \right)^2 + \frac{\omega^2}{\rho_0^2} \Gamma^2 q^8 \right] \right\} \times \left[ \left( \omega^2 - \frac{\sigma_0}{\rho_0} q^2 \right)^2 + \frac{\omega^2}{\rho_0^2} \Gamma^2 q^8 \right] \times \left[ \left( \omega^2 - \omega' - \omega'' \right)^2 - \frac{\sigma_0}{\rho_0} \left( \bar{q} - q' - \bar{q}'' \right)^2 \right] \times \left[ \left( \omega^2 - \omega' - \omega'' \right)^2 + \frac{\Gamma^2}{\rho_0^2} (\omega - \omega' - \omega'')^2 (q - q' - q'')^4 \right].
\]  

for \( \Gamma(q, \omega) \). If we solve this, the resulting \( \Gamma(q, \omega) \) can be inserted into (4.10) which can then be integrated to give \( \Gamma(q, \omega) \). Rather than carry out the calculation in such detail, we shall assume that \( \Gamma, \eta \) and \( \zeta \) have power-law divergences at small \( q \) and \( \omega \), and calculate those powers.

To be precise, we assume

\[
\Gamma(q, \omega) \sim \begin{cases} q^{-a} & \omega = 0 \\ \omega^{-b} & q = 0 \end{cases}
\]

If we then take (4.10), replace the left-hand-side by (4.13a) and, on the right, assume that the frequency dependence of \( \Gamma \) is negligible within the integrals, so that (4.13a) can be used again, then we get an equation for the wavenumber-dependence of \( \Gamma(q, \omega = 0) \), so that

\[
\Gamma(q) \sim (k_B T \rho_0)^{1/2} a q^{-1}, \quad \omega = 0.
\]

Similarly, using equation (4.13b), we can get

\[
\Delta \Gamma(\omega) \sim (k_B T)^{2/3} \rho_0^{1/3} a^{4/3} \omega^{-1/3}.
\]

Which form is a better approximation to \( \Gamma(q, \omega) \)? Well, in general, we extract \( \Gamma \) by propagating an undulation mode and measuring its attenuation. If the mode has frequency \( \omega \), then it has wavenumber \( q_w = \omega / \sqrt{\sigma / \rho_0} \). The damping will thus tell us \( \Gamma(q_w, \omega) \), which is of course smaller than both \( \Gamma(q = 0, \omega) \) and \( \Gamma(q_w, \omega = 0) \). Therefore the smaller of (4.14) and (4.13b) is the estimate we are looking for. Now

\[
\frac{\Gamma(q = 0, \omega)}{\Gamma(q_w, \omega = 0)} \sim \frac{\omega^{-1/3}}{q_w^{-1}} \sim \omega^{2/3} \quad \text{which goes to zero as } \omega \to 0.
\]

Thus for low frequencies, \( \Gamma(\omega) \) is always smaller than \( \Gamma(q_w) \) and is hence the better estimate. Thus, for our purposes, fluctuations give rise to a predominantly frequency dependent \( \Gamma \), and hence to an undulation mode whose damping is
$\Gamma(\omega) q^4 \sim \omega^{-1/3} q^4$, not $\Gamma(q) q^4 \sim q^3$, as Kats and Lebedev suggest. The two answers are very different, as (4.15) makes clear.

Finally, the frequency-dependences of $\eta$ and $\rho$ can be extracted by inserting $\Gamma(\omega)$ into (4.9). Here again, it is the frequency-dependence that dominates. We note that the dependence of the stress tensor on the $h$ field is similar for fluid and crystalline membranes (see Eqs. (4.6) and (4.7)), and when inserted in the Kubo formula gives similar answers for the in-plane viscosities for liquid and crystalline membranes. That is,

$$\eta(\omega) \sim \omega^{-2/3}. \quad (4.17)$$

**Conclusion.**

We have studied theoretically the dynamical properties of freely suspended two-dimensional films, both fluid and crystalline, with surface tension. We find that their damping coefficients diverge strongly at low frequencies. These singularities can be measured in mechanical impedance experiments. These systems can be realised in the Laboratory in the form of freely suspended films peeled off from smectic A liquid crystals [8].

**Acknowledgements.**

SR thanks T. C. Lubensky, P. Pershan and J. Prost for useful discussions and the University Grants Commission, India, for support. YM is supported by the Indian Institute of Science.

**References**