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The spectrum of surface waves on viscoelastic liquids of arbitrary depth

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Abstract. The general formula for the surface-wave spectrum of an incompressible viscoelastic liquid of arbitrary depth is derived and discussed for several limiting cases. For liquids of high viscosity and long stress relaxation time, the formula includes the contribution of the elastic response at high frequencies, which is not obtained in the lubrication approximation for the shallow limit.

1. Introduction

The viscoelastic transition of a liquid of high viscosity from viscous flow at low frequencies to shear elasticity at high frequencies is reflected not only by the shear waves in the bulk, but also by surface waves [1, 2]. The properties of the latter, of course, depend on the depth of the liquid. For surface waves on shallow liquids, whose wavelength is much longer than the depth of the liquid, usually the lubrication approximation to the Navier–Stokes equations [3] is employed. However, the result obtained using the lubrication approximation lacks the contribution of elastic high-frequency waves in thin viscoelastic films. In general, it is desirable to have a formula describing the complete spectrum of surface waves on viscoelastic liquids *of arbitrary depth*. By comparison with this general formula, the validity and accuracy of approximate results obtained in various limits can be assessed. The derivation of the general result (section 2) and the discussion of several limiting cases (section 3) are the purposes of the present paper. Sections 3.1–3.3 are mostly pedagogical; the result of section 3.4 is new.

2. Derivation of the general formula

The spectrum of surface waves of small amplitude for a given wavevector $\boldsymbol{k}$ is derived from the dynamic susceptibility $\chi_{zz}(k, \omega)$ of the vertical surface displacement u_z (figure 1) with respect to an external force (per surface area) P_z acting vertically on the liquid surface. The dynamic susceptibility is obtained as the ratio of the amplitudes of u_z and P_z for monochromatic plane waves with wavevector $\boldsymbol{k}$ parallel to the surface (see equation (27) below). This ratio is calculated from the equations of the macroscopic hydrodynamic or viscoelastic theory. As follows from the theory of linear response [4], the power spectrum of thermal height fluctuations u_z of a free liquid surface is given by

$$2k_B T \chi''_{zz}(k, \omega)/\omega \tag{1}$$

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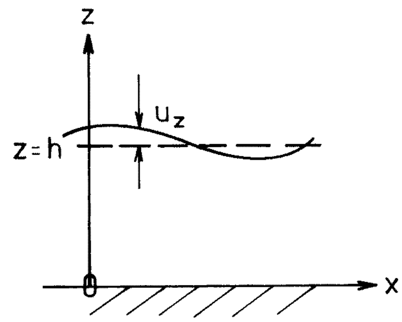

Figure 1. Geometry and notation.

where χ'' denotes the imaginary part of χ. On the other hand, for surface waves driven by an external vertical force field P_z, the expression for the absorbed power per surface area reads

$$\frac{1}{2}(\omega P_{z,0})^2 \chi''_{zz}(k,\omega)/\omega. \tag{2}$$

Therefore, in this paper we call the common factor $\chi''_{zz}(k,\omega)/\omega$ in (1) and (2) the 'surface-wave spectrum'. The calculation of $\chi_{zz}(k,\omega)$ proceeds as follows.

We start with the *ansatz* for monochromatic plane waves

$$\boldsymbol{v}(\boldsymbol{r},t) = (\bar{v}_x(z)\boldsymbol{e}_x + \bar{v}_z(z)\boldsymbol{e}_z)\exp\big[\mathrm{i}(kx-\omega t)\big] \tag{3a}$$

$$P(\boldsymbol{r},t) = \bar{P}(z)\exp\big[\mathrm{i}(kx-\omega t)\big] \tag{3b}$$

for the solution for the velocity and pressure of the linearized Navier–Stokes equation, which reads

$$\partial_t \boldsymbol{v} = -\frac{1}{\rho}\boldsymbol{\nabla} P + \nu\,\Delta\boldsymbol{v}. \tag{4}$$

$\nu = \eta/\rho$ is the kinematic viscosity of the liquid, which is treated as incompressible (with density ρ). The frequency dependence of the kinematic viscosity of a linear viscoelastic liquid can be taken into account with no difficulty. Inserting (3*a*) and (3*b*) into (4), we obtain the following equations for $\bar{v}_x(z)$ and $\bar{v}_z(z)$:

$$-\mathrm{i}\omega\bar{v}_x = -\mathrm{i}k(\bar{P}/\rho) + \nu(\partial_z^2 - k^2)\bar{v}_x \tag{5a}$$

$$-\mathrm{i}\omega\bar{v}_z = -\partial_z(\bar{P}/\rho) + \nu(\partial_z^2 - k^2)\bar{v}_z. \tag{5b}$$

Here the gravitational potential is included in the pressure. Eliminating the pressure terms yields the equation

$$(\partial_z^2 - k^2)\bar{w} = \frac{-\mathrm{i}\omega}{\nu}\bar{w} \tag{6}$$

for

$$\bar{w}(z) = \partial_z\bar{v}_x(z) - \mathrm{i}k\bar{v}_z(z) \tag{7}$$

which is the rotation of the two-dimensional flow. The solution of equation (6) is given by

$$\bar{w}(z) = B\cosh(\kappa z) + C\sinh(\kappa z) \tag{8}$$

with

$$\kappa = \sqrt{k^2 - \mathrm{i}\omega/\nu} \tag{9}$$

and B and C are constants. In general, let (9) be the root with the positive real part. Combining the continuity equation

$$\mathrm{i}k\bar{v}_x + \partial_z \bar{v}_z = 0 \tag{10}$$

with (7) and (8), we find the decoupled equation for $\bar{v}_z(z)$ as follows:

$$(\partial_z^2 - k^2)\bar{v}_z = -\mathrm{i}k\bar{w}. \tag{11}$$

Using the Green function for $(\partial_z^2 - k^2)$

$$G(z) = k^{-1} \sinh(kz)\theta(z) \tag{12}$$

we can write the general solution of equation (11) as

$$\bar{v}_z(z) = \sum_{\pm} A_{\pm} \mathrm{e}^{\pm kz} - \mathrm{i} \int_0^z \mathrm{d}z' \ \sinh[k(z - z')]\bar{w}(z'). \tag{13}$$

From the no-slip boundary conditions at $z = 0$

$$\bar{v}_x(z = 0) = \bar{v}_z(z = 0) = 0 \tag{14}$$

it follows, using (10), that

$$A_{\pm} = 0. \tag{15}$$

For $\bar{v}_z(z)$ we thus obtain

$$\bar{v}_z(z) = -\mathrm{i}\left\{\hat{B}k\big(\cosh(kz) - \cosh(\kappa z)\big) + \hat{C}\big(\kappa \sinh(kz) - k \sinh(\kappa z)\big)\right\} \tag{16}$$

where

$$B/\hat{B} = C/\hat{C} = k^2 - \kappa^2. \tag{17}$$

For $\bar{v}_x(z)$, equation (10) yields

$$\bar{v}_x(z) = \hat{B}\big(k \sinh(kz) - \kappa \sinh(\kappa z)\big) + \hat{C}\kappa\big(\cosh(kz) - \cosh(\kappa z)\big). \tag{18}$$

With (16) and (18), using (5a) we obtain for the pressure

$$\bar{P}/\rho = -\mathrm{i}\nu\big(B \sinh(kz) + C(\kappa/k) \cosh(kz)\big). \tag{19}$$

The coefficients $\hat{B}$ and $\hat{C}$ need to be determined from the boundary conditions at the free surface at $z = h$, which read

$$\sigma_{xz} = \eta(\partial_z v_x + \partial_x v_z) = 0 \tag{20}$$

and

$$\sigma_{zz} = -P + 2\eta\, \partial_z v_z = \alpha\, \partial_x^2 u_z - g\rho u_z + P_z \tag{21}$$

where

$$u_z = (\mathrm{i}/\omega) v_z|_{z=h}$$

is the vertical surface displacement (figure 1) and

$$P_z(x, t) = P_{z,0} \exp[\mathrm{i}(kx - \omega t)] \tag{22}$$

is an external force per area acting on the surface in the z-direction. α denotes the surface tension, and g is the gravitational force per mass. With the *ansatz* (3), the boundary conditions take the form

$$\partial_z \bar{v}_x + \mathrm{i}k\bar{v}_z|_{z=h} = 0 \tag{23}$$

and

$$-(\bar{P}/\rho) + 2\nu\,\partial_z\bar{v}_z + \frac{\mathrm{i}}{\omega}\left(\frac{\alpha}{\rho}k^2 + g\right)\bar{v}_z\bigg|_{z=h} = P_{z,0}/\rho. \qquad (24)$$

With (16) and (18), the first of these boundary conditions (equation (23)) can be written as

$$A_{11}\hat{B} + A_{12}\hat{C} = 0 \qquad (25)$$

with coefficients

$$A_{11} = 2k^2\cosh(kh) - \left[k^2 + \kappa^2\right]\cosh(\kappa h) \qquad (25a)$$

$$A_{12} = 2k\kappa\sinh(kh) - \left[k^2 + \kappa^2\right]\sinh(\kappa h). \qquad (25b)$$

The second boundary condition (equation (24)) yields

$$A_{21}\hat{B} + A_{22}\hat{C} = P_{z,0}/\rho \qquad (26)$$

with coefficients

$$A_{21} = -\mathrm{i}\nu\left[k^2 + \kappa^2\right]\sinh(kh) + 2\mathrm{i}\nu k\kappa\sinh(\kappa h) + (k/\omega)\left[(\alpha/\rho)k^2 + g\right]\left(\cosh(kh) - \cosh(\kappa h)\right) \qquad (26a)$$

$$A_{22} = -\mathrm{i}\nu\left[k^2 + \kappa^2\right](\kappa/k)\cosh(kh) + 2\mathrm{i}\nu k\kappa\cosh(\kappa h) + (1/\omega)\left[(\alpha/\rho)k^2 + g\right]\left(\kappa\sinh(kh) - k\sinh(\kappa h)\right). \qquad (26b)$$

Solving the linear equations (25) and (26) for $\hat{B}$ and $\hat{C}$, we obtain the dynamical susceptibility

$$\chi_{zz}(k,\omega) = u_z/P_z = (\mathrm{i}/\omega)\bar{v}_z(z=h)/P_{z,0} \qquad (27)$$

as

$$\chi_{zz}(k,\omega) = \frac{1}{\rho\omega}\,\frac{A_{11}\left(\kappa\sinh(kh) - k\sinh(\kappa h)\right) - A_{12}k\left(\cosh(kh) - \cosh(\kappa h)\right)}{A_{11}A_{22} - A_{12}A_{21}} \qquad (28)$$

with the result

$$(\chi_{zz}(k,\omega))^{-1} = \alpha k^2 + g\rho + \rho\nu^2k^3\frac{N}{D} \qquad (29)$$

where

$$N = -4\zeta[1+\zeta^2] + \zeta(4 + [1+\zeta^2]^2)\cosh(kh)\cosh(\zeta kh) - (4\zeta^2 + [1+\zeta^2]^2)\sinh(kh)\sinh(\zeta kh) \qquad (29a)$$

and

$$D = \zeta\sinh(kh)\cosh(\zeta kh) - \sinh(\zeta kh)\cosh(kh) \qquad (29b)$$

where

$$\zeta = \kappa/k = (1 - \mathrm{i}\omega/(\nu k^2))^{1/2}. \qquad (29c)$$

Equation (29) is the desired general formula. Extracting the static susceptibility

$$\chi_0(k) = (\alpha k^2 + g\rho)^{-1} \qquad (30)$$

we can write the result (29) in a more compact form as

$$\chi_{zz}(k,\omega)/\chi_0(k) = \left(1 + \frac{(\nu k^2)^2}{(\omega_s(k))^2}\,\frac{N}{D}\right)^{-1} \qquad (31)$$

where

$$\omega_s(k) = ((\alpha/\rho)k^3 + gk)^{1/2} \tag{32}$$

is the dispersion relation for surface waves on an ideal liquid of infinite depth.

This general result applies to linear viscoelastic liquids, if the kinematic viscosity is chosen as an appropriate function $\nu(\omega)$ of frequency. Note that ν enters the dynamic susceptibility equation (29) not only explicitly but also via κ (equation (9)). A simple and convenient linear viscoelastic model is described by

$$\nu(\omega) = \nu_s + \nu_0/(1 - \mathrm{i}\omega\tau). \tag{33}$$

The frequency-dependent part of this expression corresponds to Maxwell's model. The constant part ν_s accounts for the effect of viscous damping at high frequencies $\omega\tau \gg 1$.

The frequency dependence of the general expression (29) for given k depends on a number of parameters and is very different in different parameter ranges. It is convenient to use dimensionless quantities. We consider the dimensionless ratio (31) of the dynamic and static susceptibility as a function of the dimensionless frequency variable $\omega/\omega_s(k)$, where $\omega_s(k)$ is given by equation (32). This function depends on the dimensionless parameter kh together with the three additional dimensionless parameters $\nu_0 k^2/\omega_s(k)$, $\omega_s(k)\tau$ and ν_s/ν_0, which are related to the viscoelastic model equation (33). We now consider several important limiting cases of these parameters for which very different spectra follow from the general formula (29).

3. Limiting cases

3.1. Deep liquid ($h \to \infty$)

In this limit one obtains from (29) the result

$$\chi_{zz}(k, \omega) = \frac{k/\rho}{(\omega_s(k))^2 - \omega^2 - \mathrm{i}\omega\, 4\nu k^2 \kappa/(k + \kappa)} \tag{34}$$

which is equivalent to equation (21) of reference [2], but of simpler form. According to this new expression, the 'damping function' $\Gamma(k, \omega)$ is simply given by

$$\Gamma(k, \omega) = 4\nu k^2 \kappa/(k + \kappa). \tag{35}$$

We briefly summarize for the deep limit $h \to \infty$ the variation of the frequency spectrum $\chi''_{zz}(k, \omega)/\omega$ with increasing frequency, using the Maxwell model. (The constant background viscosity ν_s of expression (33) is not essential and only leads to some additional damping in that part of the spectrum for which $\omega\tau \gg 1$ holds.) The form of the spectrum at high viscosity and long relaxation time depends crucially on the value of kl_0, where the length l_0 is related to the surface tension α and the high-frequency shear modulus $G(\infty)$ by

$$l_0 = \alpha/G(\infty). \tag{36}$$

For the Maxwell model, $G(\infty)$ is given by relaxation time τ and hydrodynamic shear viscosity η_0 as

$$G(\infty) = \eta_0/\tau. \tag{37}$$

If $kl_0 \ll 1$ holds, the high-frequency response is dominated by $G(\infty)$ rather than by α. In this case, after the surface waves have first become overdamped for viscosities $2\nu_0 k^2 > \omega_s(k)$, elastic Rayleigh waves appear when the relaxation time exceeds their frequency, which is slightly lower than $c_t(\infty)k$. Here

$$c_t(\infty) = \left(G(\infty)/\rho\right)^{1/2} \tag{38}$$

denotes the high-frequency transverse sound velocity. The surface spectrum then consists of the quasi-elastic line due to the overdamped capillary waves, an inelastic part arising from the Rayleigh waves, plus a continuum contribution from bulk phonons [2]. For $kl_0 \gg 1$, on the other hand, the surface tension provides the dominant restoring force even in the slow-relaxation limit, and the viscoelastic effects on the spectrum are weak. This case may be realized with light scattering from polymeric solutions [1, 5]. In the intermediate case, $kl_0 \approx 1$, an interesting pattern of interference between the effect of surface tension and shear elasticity exists in the limit of slow relaxation [5].

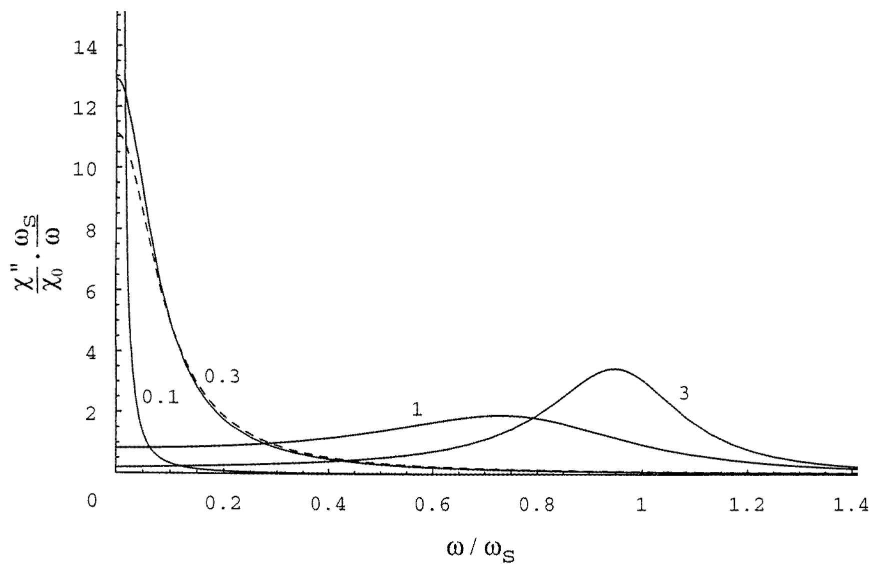

Figure 2. The surface-wave spectrum in dimensionless units for ordinary liquid (constant viscosity) for different values of the depth parameter: $kh = 0.1$, 0.3, 1 and 3, as indicated, and $\nu_0 k^2/\omega_s(k) = 0.1$. Dashed line: equation (40) for $kh = 0.3$. For $kh = 0.1$ the approximate result, equation (40), is indistinguishable from the exact result.

3.2. Shallow liquid ($h \to 0$)

In this case we find, expanding in powers of h,

$$\chi_{zz}(k,\omega) = \frac{k^2h^3/3}{(\alpha k^2 + g\rho)k^2h^3/3 - \mathrm{i}\omega\eta(1 + (h^2/5)(9k^2 - 2\mathrm{i}\omega/\nu)) + \mathrm{O}(h^4)}. \tag{39}$$

To leading order in h this yields

$$\chi_{zz}(k,\omega) = \frac{k^2h^3/(3\eta)}{-\mathrm{i}\omega + ((\alpha/\rho)k^3 + gk)kh^3/(3\nu)} \tag{40}$$

which is the result obtained directly using the lubrication approximation [3]. It applies if both

$$kh \ll 1 \tag{41a}$$

and

$$\sqrt{\frac{\omega}{2\nu}}h \ll 1 \tag{41b}$$

hold. The first of these conditions requires that the depth h of the liquid must be small with respect to the wavelength in the lateral x-direction, the second that it is small compared with the penetration depth of oscillating shear flow.

Figure 2 illustrates the transition from the deep- to the shallow-liquid limit. The result (34) for $h = \infty$ applies already at $kh = 3$, while for $kh = 0.3$ the approximate result (40) (the dashed line in figure 2) deviates noticeably from the exact formula (29).

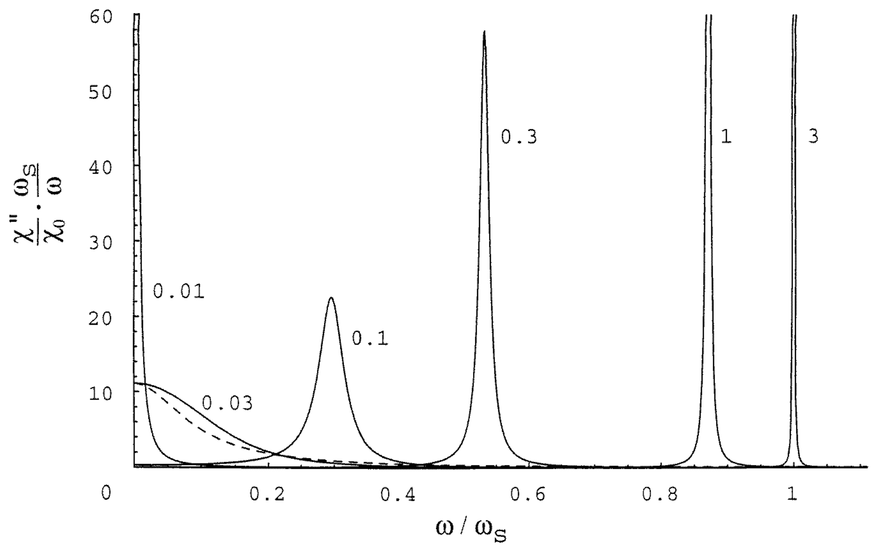

Figure 3. The surface-wave spectrum for nearly ideal liquid ($\nu_0 k^2/\omega_s(k) = 10^{-4}$) for $kh = 0.01, 0.03, 0.1, 0.3, 1$ and 3. Dashed line: equation (40) for $kh = 0.03$. For $kh = 0.01$ the approximate result, equation (40), is indistinguishable from the exact result. The curve for $kh = 3$ is indistinguishable from that for $kh = \infty$.

3.3. Nearly ideal liquid ($\nu \to 0$)

For very low viscosity ν, the viscous penetration depth becomes very short, and the reverse of condition (41*b*) may apply even if (41*a*) holds. For a shallow liquid, when (41*a*) holds, the second condition (41*b*) may be violated also in the opposite case of very high viscosity and long relaxation time τ for the high-frequency elastic response. Before treating the latter effect, we first consider the low-viscosity limit, for arbitrary values of the depths h. Specifically, we assume ν to be small enough for the condition

$$\omega \gg \nu k^2 \tag{42}$$

to hold (ν is assumed to be frequency independent in this case, naturally). We then get for κ (equation (9))

$$\kappa = (1 - \mathrm{i})\big(\omega/(2\nu)\big)^{1/2}\big(1 + \mathrm{O}(k^2/\kappa^2)\big). \tag{43}$$

As regards the depth h, we assume that it is much larger than the viscous penetration depth; that is,

$$h \gg (2\nu/\omega)^{1/2}. \tag{44}$$

Neglecting all terms containing a factor $\exp(-\kappa h)$ and keeping only the terms of leading and next-to-leading order in (κ/k), we obtain from (29)

$$\chi_{zz}(k,\omega) = \frac{1}{\alpha k^2 + g\rho + \rho\nu^2(\kappa^4/k)\,\mathrm{cotanh}(kh)(1 + 2k/(\kappa\sinh(2kh)))}$$
$$\approx \frac{(k/\rho)\tanh(kh)}{(\omega_s(k))^2\tanh(kh) - \omega^2 - (1+\mathrm{i})\sqrt{2\omega^3\nu k^2}/\sinh(2kh)}. \tag{45}$$

According to this result, the frequency of surface waves is reduced by a factor $(\tanh(kh))^{1/2}$ compared with the case of large depth [6] (see figure 3). For the damping coefficient one reads off from equation (45) the peculiar result

$$\beta = \sqrt{2\omega\nu k^2}/\sinh(2kh). \tag{46}$$

For (45) to be consistent with assumption (42), the condition

$$\omega_s(k)\big(\tanh(kh)\big)^{1/2} \gg \nu k^2 \tag{47}$$

must be fulfilled. The origin of the damping (46) is the dissipation in the boundary layer at the bottom of the liquid [7], the width of which is equal to the viscous penetration depth. For deep liquids $h \to \infty$, only the dissipation in the bulk is left, giving an attenuation coefficient $\beta = 4\nu k^2$, corresponding to expression (34) for $k/\kappa \to 0$ [8, 9].

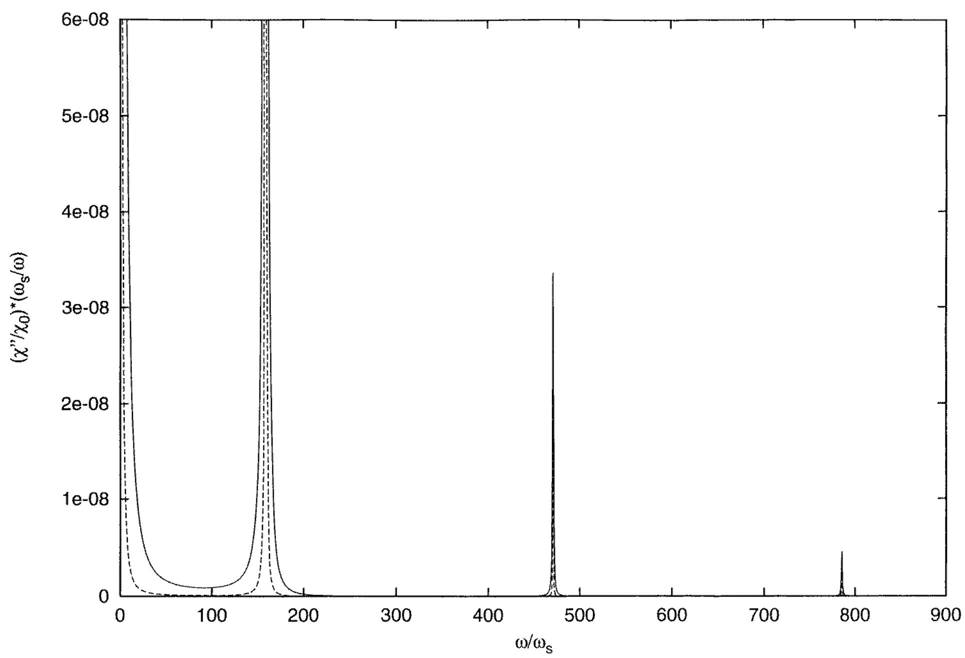

Figure 4. The surface-wave spectrum for shallow viscoelastic liquid for $kh = 0.1$, $\nu_0 k^2/\omega_s(k) = 10^2$ (10^3) and $\omega_s(k)\tau = 1$ (10). (The numbers in brackets are for the dashed line.) Apart from the quasi-elastic contribution, three elastic resonances are seen, which correspond to equation (54) for $n = 0, 1$ and 2.

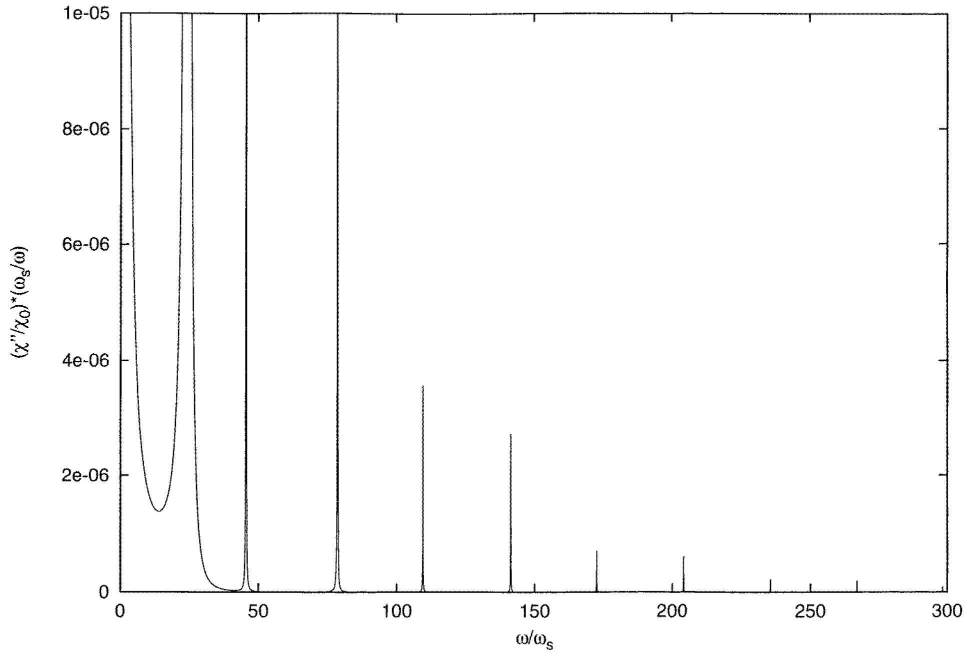

Figure 5. The surface-wave spectrum for viscoelastic liquid of intermediate depth $kh = 1$ with $\nu_0 k^2/\omega_s(k) = 10^3$ and $\omega_s(k)\tau = 10$.

3.4. Shallow liquid; the elastic limit ($\omega\tau \gg 1$)

We finally discuss the case of high viscosity and long relaxation time τ in the shallow limit (41a) for the Maxwell model ($\nu_s = 0$). We assume that the high-frequency transverse sound velocity $c_t(\infty)$, equation (38), is sufficiently large for the condition

$$\omega_s(k)(kh)^{3/2} \ll c_t(\infty)k \tag{48}$$

to hold. Under this condition, the result (40), with ν replaced by its hydrodynamic value ν_0, represents the low-frequency response for the Maxwell model. This replacement is self-consistent since it leads to a relaxation frequency in (40), which is much smaller than the viscoelastic relaxation rate τ^{-1}, due to condition (48):

$$\omega_s^2(k)kh^3/(3\nu_0) \ll \tau^{-1}. \tag{49}$$

However, the result (40), which can be obtained by using the lubrication approximation, is not valid at high frequencies $\omega\tau \gg 1$. The elastic high-frequency part of the response must be derived directly from (29). As is now shown, it occurs on a frequency scale of the order of $(c_t(\infty)/h)$. For the response at such frequencies to be elastic we must assume that the stress relaxation rate is far below this frequency range; that is,

$$\tau^{-1} \ll c_t(\infty)/h. \tag{50}$$

With the high-frequency form

$$\nu(\omega) = \frac{(c_t(\infty))^2}{-\mathrm{i}\omega} \tag{51}$$

of the kinematic viscosity of the Maxwell model, we then obtain for κ, using condition (41*a*),

$$\kappa = \mathrm{i}q \qquad \text{with } q \approx \omega/c_t(\infty). \tag{52}$$

Keeping only the terms of leading order in the expansion of the expression (29) with respect to kh leads to an elastic part of the dynamical susceptibility given by

$$\chi_{zz}^{(el)}(k,\omega) = (k^2h/\rho) \bigg/ \left(\omega_s^2(k)kh - \omega^2 \frac{qh\cos(qh)}{qh\cos(qh) - \sin(qh)}\right). \tag{53}$$

Under the condition (48), this expression has resonances near

$$\omega = \big(c_t(\infty)/h\big)(\pi/2)(2n+1) \qquad \text{for } n = 0, 1, 2, \ldots \tag{54}$$

in agreement with the assumption on the frequency scale made above. These resonances correspond to shear oscillations of an elastic film with a free surface on a rigid substrate, but with a lateral modulation of long wavelength superimposed. The strength of the resonances decreases rapidly with increasing order n (figure 4).

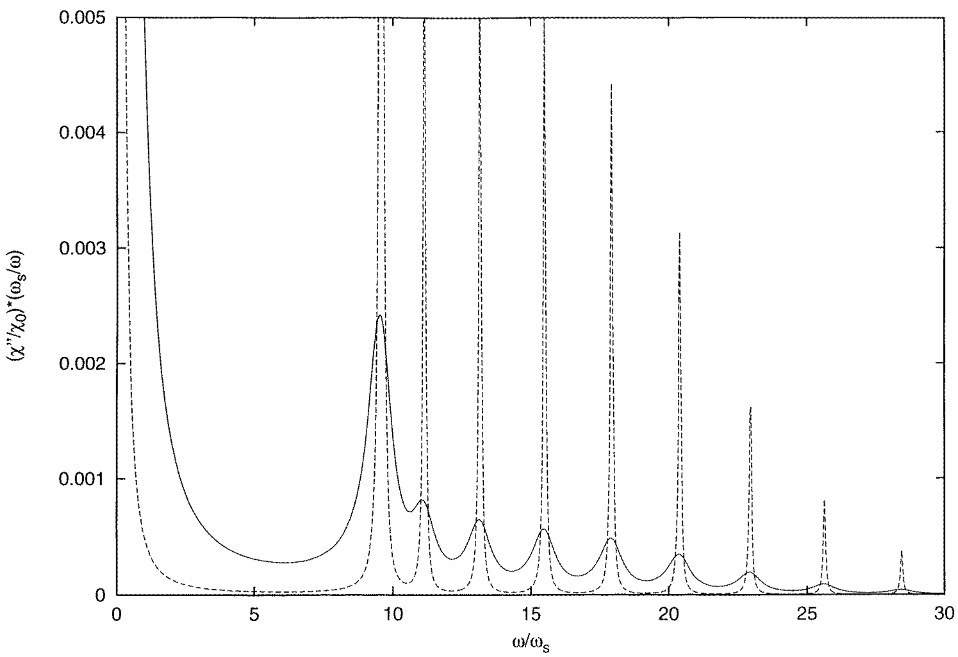

Figure 6. As figure 4, but with $kh = 10$. The strong line below $\omega/\omega_s(k) = 10$ is the precursor of the elastic Rayleigh line which develops for $kh \to \infty$.

The elastic parts of the surface-wave spectrum look completely different in the shallow limit (figure 4) and in the deep liquid (figure 1 of reference [2]). It is interesting to see how the elastic part of the spectrum changes between these limits when the depth h of the liquid is varied. Figures 5 and 6 show the cases where $kh = 1$ and $kh = 10$. (The other parameters are as in figure 4.) With increasing depth the number of elastic resonances increases. For $kh = 10$ (figure 6), there is a strong lowest resonance near $\omega = 0.95c_t(\infty)k$, which for $h \to \infty$ eventually develops into the contribution of the elastic Rayleigh surface wave. The higher resonances go over into a continuum.

4. Conclusions

To summarize, the general formula for the surface-wave spectrum $\chi''_{zz}(\omega)/\omega$ of a viscoelastic liquid of arbitrary depth h has been derived (equation (29)). For an ordinary liquid with constant viscosity, the result from using the lubrication approximation is obtained in the shallow limit, where the depth h is small both compared with the lateral wavelength $2\pi/k$ and compared with the viscous penetration depth $(2\nu/\omega)^{1/2}$ (figure 2). The formula can also be applied to nearly ideal liquids of low viscosity, for which it reproduces the known results for the depth dependence of the surface-wave frequency (figure 3) and damping. For shallow viscoelastic liquids of high viscosity and long stress relaxation time, in addition to the quasi-elastic part obtained with the lubrication approximation, the new result for the contribution from the elastic high-frequency response of the liquid is found. It consists of resonances corresponding to elastic waves in a thin plate (figures 4–6). It would be of interest to study the depth dependence of the spectrum of thermal surface fluctuations of viscoelastic liquids experimentally by means of inelastic surface light scattering [10].

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