

## XVI. PHYSICAL ACOUSTICS\*

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## RESEARCH OBJECTIVES

Our general objective involves the study of the emission, propagation, and absorption of acoustic waves in matter. Specific areas of present research include studies of (i) the interaction of waves with coherent light beams in fluids and solids, (ii) non-linear acoustics, and the generation, propagation, and (iii) amplification of sound waves in ionized gases.

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### A. LIGHT SCATTERING FROM A LIQUID-LIQUID INTERFACE

Our studies of thermally excited surface waves on liquids by means of light scattering, as described in earlier reports,<sup>1,2</sup> have been extended to the problem of surface waves on the interface between two liquids. As a specific example, we have considered the methanol-hexane interface. A laser beam is scattered off the interface, and the light scattered in a certain direction is analyzed by means of heterodyne spectroscopy, by using the specularly reflected light beam as a "local oscillator."

Examples of recorded beat spectra (actually the square root of the beam-power spectra) obtained at three different scattering angles are shown in Fig. XVI-1. It is interesting to note that as the scattering angle (and the wave number) increases, the scattered spectrum changes markedly in character from a Brillouin doublet to a single line. At low scattering angles corresponding to long wavelengths there is a distinct Brillouin doublet, but as the scattering angle increases (wavelength decreases), the doublet gradually turns into a single line. The same thing occurs at a given wavelength when the temperature is increased. Measurements of this sort were carried out at different temperatures ranging from 25°C to 35°C, and at each temperature several scattering angles corresponding to different ripple wavelengths were considered.

To interpret these experimental results, we again used the classical hydrodynamic theory of surface waves adapted to the present problem, assuming no slippage between the two liquids at the interface. The resulting dispersion relation is found to be of the form

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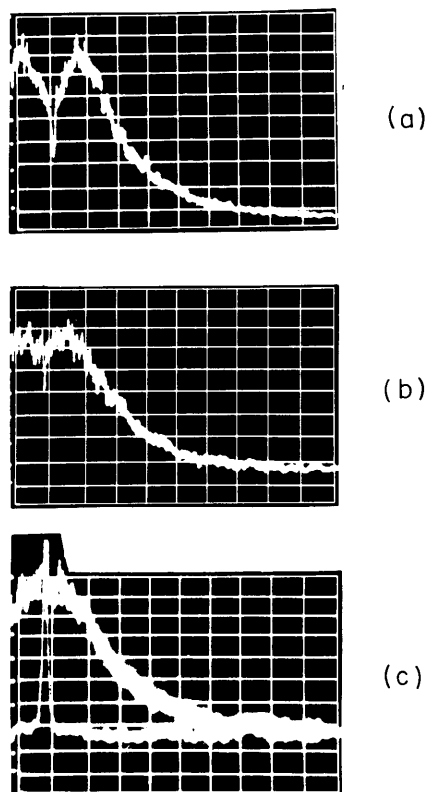


Fig. XVI-1. Examples of recorded spectra (the output is proportional to the square root of the intensity) of light scattered from thermally excited surface waves in a methanol-hexane interface at different scattering angles and corresponding wave numbers. (a)  $k = 750 \text{ cm}^{-1}$ ; (b)  $k = 1750 \text{ cm}^{-1}$ ; (c)  $k = 2330 \text{ cm}^{-1}$ .

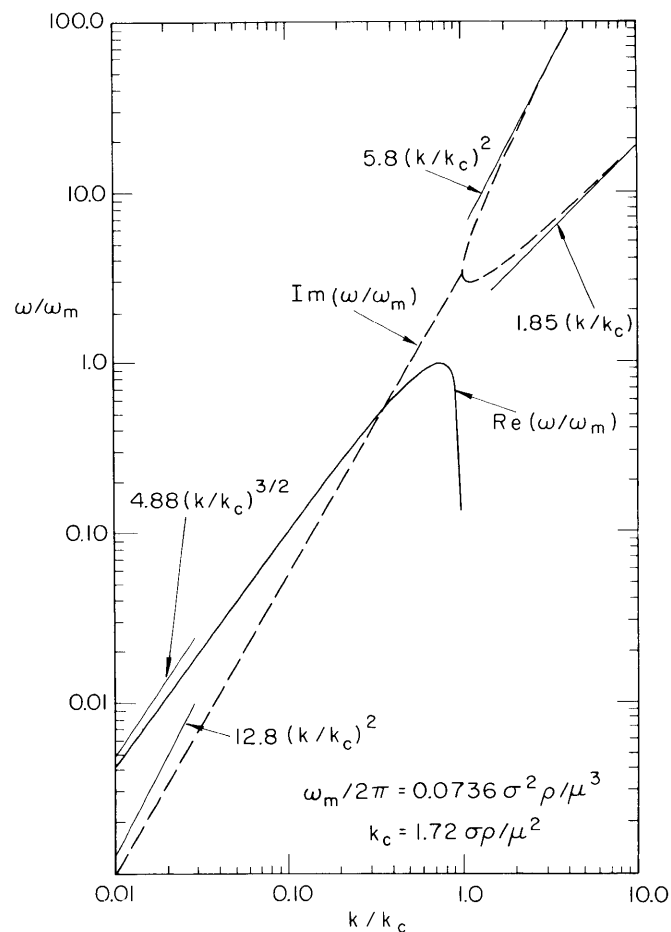


Fig. XVI-2. Dispersion relation for surface waves on a liquid  $\omega = \omega(k)$ .  $\omega_m = 0.4 \sigma^2 \rho / \mu^3$ ;  $k_c = 1.72 \sigma \rho / \mu^2$ .

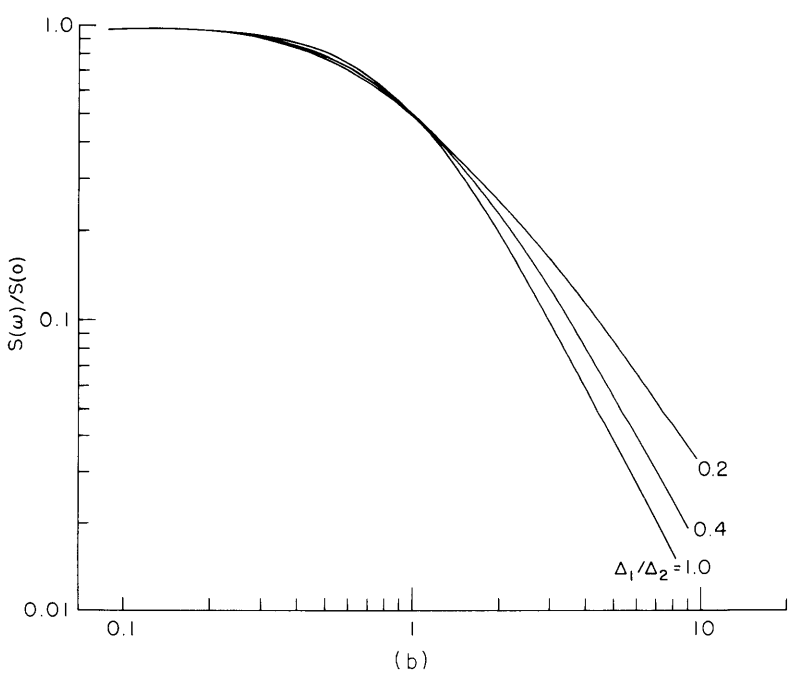
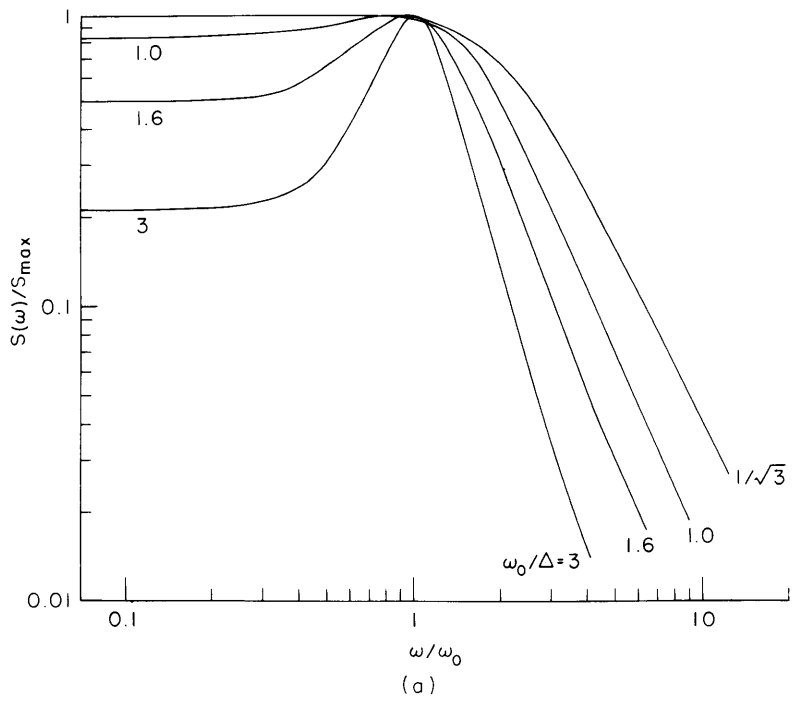


Fig. XVI-3. Theoretically predicted shapes of the spectra of light scattered from (a) underdamped and (b) overdamped surface waves on liquid.  $\omega_0/2\pi =$  frequency at the maximum of the spectrum from underdamped waves.  $\Delta =$  linewidth ( $\Delta_1$  and  $\Delta_2$  are the linewidths of the two components in the spectrum from overdamped waves).

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$$(\gamma+1)^2 + (\omega_1 \tau)^2 - a \sqrt{1 + \frac{2a}{b} \gamma} - (1-a) \sqrt{1 + \frac{2(1-a)}{(1-b)} \gamma} = 0, \quad (1)$$

where  $\tau = (\rho_1 + \rho_2) / 2(\mu_1 + \mu_2)k^2$ ,  $\omega_1^2 = \sigma k^3 / (\rho_1 + \rho_2)$ ,  $a = \mu_1 / (\mu_1 + \mu_2)$ ,  $b = \rho_1 / (\rho_1 + \rho_2)$ , and  $\mu_{1,2}$  and  $\rho_{1,2}$  are the viscosities and densities of each phase. With this binary system we have  $a, b \approx 0.45-0.50$ , and the normalized form of the dispersion relation then does not greatly differ from that for free surface waves as shown in Fig. XVI-2.

It should be pointed out that in the analysis of the experimental spectra the frequency shift and the linewidth of each Brillouin line cannot be read directly from the recorded spectra unless the separation of the lines is large compared with the linewidth. Since in our experiment this is not always the case, these quantities can be obtained only by comparison of the scattered spectra with the spectral lines computed for different values of frequency shift and linewidth, or, in the overdamped case, for different values of the two linewidths  $\Delta_1$  and  $\Delta_2$ . Such computed spectra are shown in Fig. XVI-3. In the underdamped case a spectrum consists of a superposition of two Lorentzians of equal width  $\Delta$ , separated by a frequency  $2\omega$ , but two maxima in the resulting spectrum are obtained only if the frequency shift  $\omega$  is larger than  $\Delta/\sqrt{3}$ . In the overdamped case the scattered spectrum is a superposition of two unshifted Lorentzian lines of different linewidths  $\Delta_1$  and  $\Delta_2$  with peak values inversely proportional to the linewidths (since in thermal equilibrium each mode is excited with equal total energy).

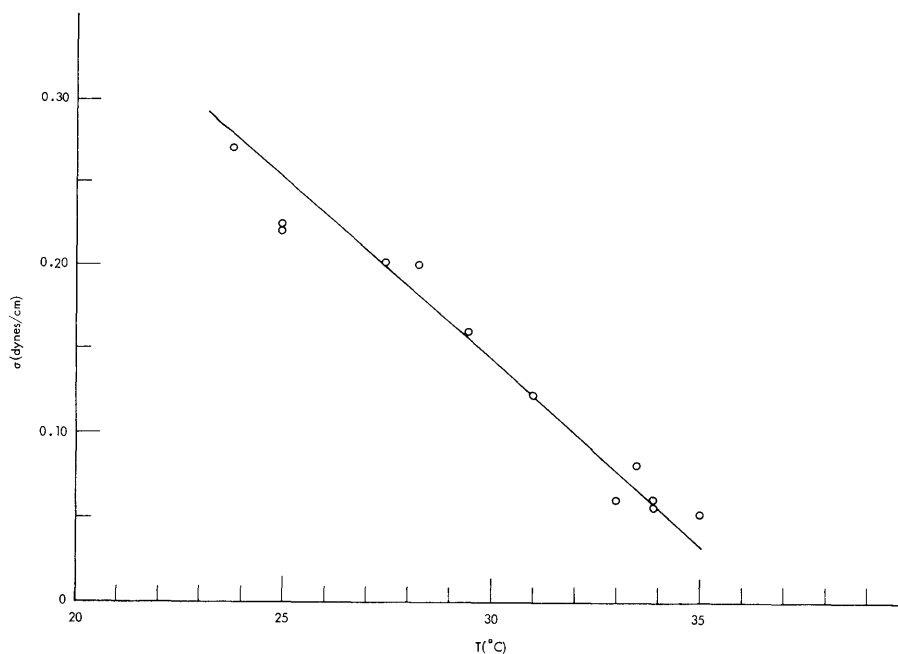


Fig. XVI-4. Experimental values of surface tension vs temperature for methanol-hexane interface.

In our experiment we encountered scattered spectra of both underdamped and overdamped cases, corresponding to Fig. XVI-3a and XVI-3b, respectively. For example, at a temperature of 25°C and a ripple wave number  $k = 750 \text{ cm}^{-1}$ , the scattered spectrum closely matched the shape of that corresponding to  $(\omega/\Delta) = 1.0$  in Fig. XVI-3a, whereas at  $k = 2330 \text{ cm}^{-1}$  the scattered spectrum was in closest agreement with that of an unshifted line composed of two Lorentzians with  $\Delta_1 = 0.5 \Delta_2$  as in Fig. XVI-3b.

Having obtained the real and imaginary parts of  $\gamma = -i\omega\tau$  in this manner, we can obtain the corresponding value of the interfacial surface tension  $\sigma$  from Eq. 1. In fact, for sufficiently large values of the ripple wavelength the surface tension is given simply by  $(\rho_1 + \rho_2)(\omega^2/k^3)$ , where  $\omega$  is the frequency shift.

The values of surface tension obtained from such measurements are shown as a function of temperature in Fig. XVI-4. The critical mixing temperature of the liquid is known to be 36.5°C, and it is interesting to note that our experiments indicate linear decrease of surface tension with temperature as the critical temperature is approached. It is important to point out in this context that our present method of measurement allows measurement of surface tension as low as 0.01 dyn/cm, which is an order of magnitude lower than most conventional techniques can give.

R. H. Katyl, U. Ingard

#### References

1. R. H. Katyl and U. Ingard, Quarterly Progress Report No. 90, Research Laboratory of Electronics, M.I.T., July 15, 1968, p. 53.
2. R. H. Katyl and U. Ingard, Quarterly Progress Report No. 88, Research Laboratory of Electronics, M.I.T., January 15, 1968, pp. 85-86.

#### B. MEASUREMENTS OF DISPERSION RELATION FOR SURFACE WAVES ON PURE LIQUIDS

The experimental procedure of studying surface waves on a liquid, as described in a previous report,<sup>1</sup> has been applied to the study of a number of liquids. The results are shown in Fig. XVI-5, where the real and imaginary parts of the frequency have been plotted as a function of the wave number  $k = 2\pi/\lambda$ , where  $\lambda$  is the wavelength of the surface waves. The characteristic frequency  $\omega_m$ , used for normalization, is given by

$$\frac{\omega_m}{2\pi} = f_m \approx 0.074 \frac{\sigma^2 \rho}{\mu^3}, \quad (1)$$

where  $\sigma$  = surface tension,  $\rho$  = liquid density, and  $\mu$  = coefficient of shear viscosity. Similarly, the quantity  $k_c = 2\pi/\lambda_c$ , used in the normalization of the wave number,

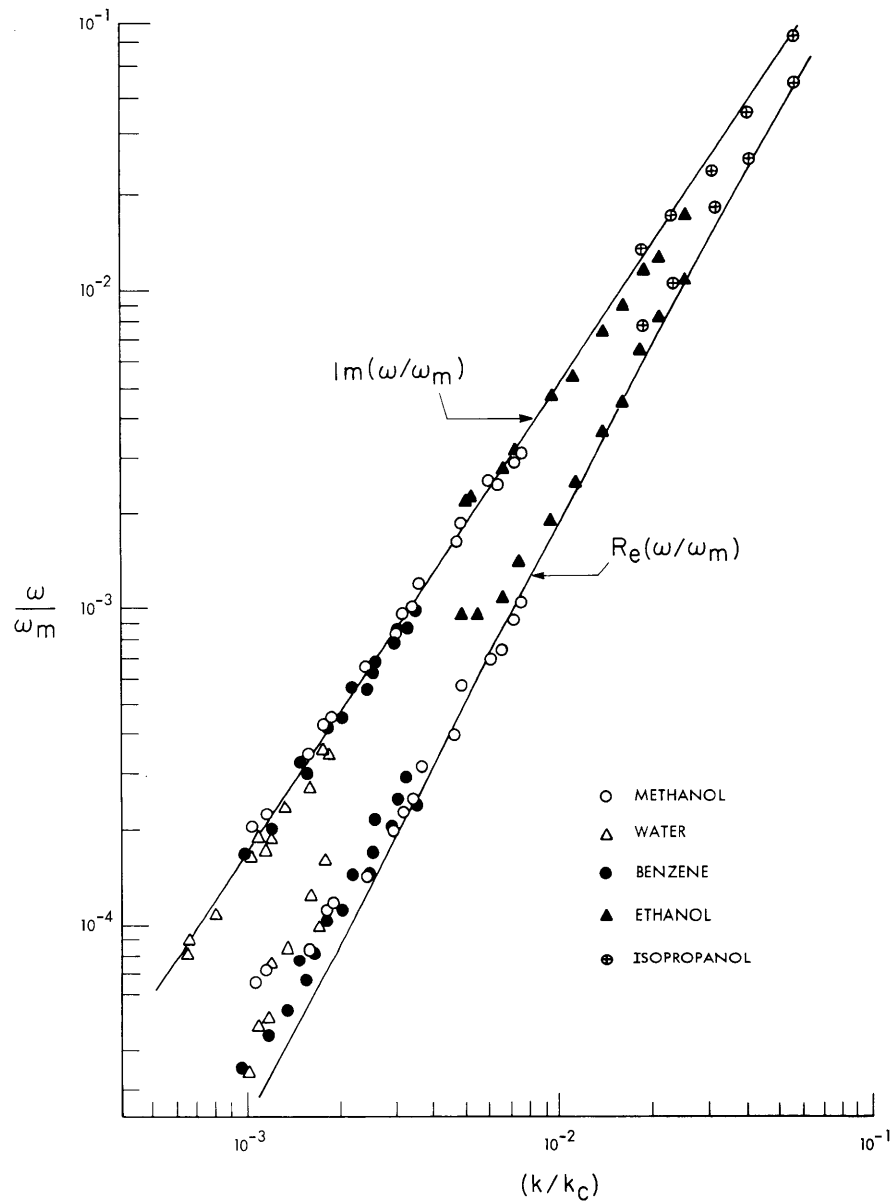


Fig. XVI-5. Experimental values of (normalized) frequency of oscillation  $Re(\omega/\omega_m)$  and decay rate  $Im(\omega/\omega_m)$  for thermally excited surface waves in some liquid surfaces as a function of (normalized) wave number  $k/k_c$  ( $k = 2\pi/\lambda$ ).  
 $k_c = 1.72 \sigma/\mu^2$ ;  $\omega_m = 0.44 \sigma^2/\mu^3$ .

is given by

$$\frac{2\pi}{k_c} = \lambda_c = 8\pi y_c \frac{\mu^2}{\sigma\rho} \approx 3.65 \frac{\mu^2}{\sigma\rho}. \quad (2)$$

As can be seen, the results obtained for these different liquids are in good agreement with the theoretically predicted dispersion relation obtained from classical hydrodynamics. A comprehensive discussion of these results is being prepared for publication.

R. H. Katyl, U. Ingard

#### References

1. R. H. Katyl and U. Ingard, Quarterly Progress Report No. 88, Research Laboratory of Electronics, M. I. T., January 15, 1968, pp. 85-86.

#### C. ABSORPTION CHARACTERISTICS OF A NONLINEAR OSCILLATOR

Absorption characteristics of nonlinear acoustic resonators of the Helmholtz type have been computed for a large number of resonator parameters. The results have recently been published.<sup>1</sup>

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#### References

1. U. Ingard, "Absorption Characteristics of Nonlinear Acoustic Resonators," J. Acoust. Soc. Am. 44, 1155-1156 (1968).

#### D. CRYSTAL STRUCTURE OF $\text{LaF}_3$

There have been many conflicting studies of the crystal structure of  $\text{LaF}_3$ . In a recent paper, Bauman and Porto<sup>1</sup> reviewed the published data suggesting or corroborating the following symmetries:  $D_{6h}^3$ ,  $D_{6h}^4$ , and  $D_{3d}^4$ . Their Raman spectra data are consistent with the  $D_{3d}^4$  symmetry.

Using Bragg diffraction of light,<sup>2</sup> we have obtained sound-velocity data that are consistent with a trigonal structure ( $D_{3d}$ ) and exclude the possibility of a hexagonal structure ( $D_{6h}$ ). Hexagonal crystals possess elastic isotropy in the basal plane; we found that the velocities  $v(k\parallel x; u\parallel y)$  and  $v(k\parallel y; u\parallel x)$  differed by 0.12%. A detailed description of this work has been published.<sup>3</sup>

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References

1. R. P. Bauman and S. P. S. Porto, Phys. Rev. 161, 842-847 (1967).
2. C. Krischer, Quarterly Progress Report No. 88, Research Laboratory of Electronics, M.I.T., January 15, 1968, pp. 86-87.
3. C. Krischer, "Measurement of Sound Velocities in Crystals Using Bragg Diffraction of Light and Applications to Lanthanum Fluoride" Appl. Phys. Letters 13, 310-311 (1968).

E. LIGHT SCATTERING FROM SURFACE WAVES  
ON CARBON DIOXIDE

An experiment designed to study light scattering from thermally excited surface waves on carbon dioxide in the neighborhood of the critical point has been started. A scattering cell has been designed, and means for providing liquid samples are now being explored.

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