SCATTERING OF LIGHT BY THERMAL RIPPLONS*

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We report measurements of the spectrum of light scattered off a liquid surface by thermally excited surface modes of vibration. The quanta of these have been referred to as ripplons.[1] For underdamped surface vibrations,[2] the scattering process can be regarded as a photon-ripplon collision, in which energy and the component of momentum along the liquid surface are conserved. Energy conservation implies that the scattered light spectrum is a doublet of frequency $\Omega_0 \pm \omega$, where Ω_0 and ω are the frequencies of the incident light and the ripplon. If this spectrum is observed in a direction (θ, φ) below the surface, conservation of momentum implies that the wavelength Λ of the observed ripplon is given[3] by

$$\Lambda = (\lambda/n)(\sin^2\theta + \sin^2\theta_0 - 2\sin\theta\,\sin\theta_0\,\cos\varphi)^{-1/2},$$

where n is the index of refraction, λ is the free-space light wavelength, and θ_0 is the polar angle of the light beam incident from below the surface. The surface vibrations are damped by viscosity, and the shape of the lines of the doublet is Lorentzian, with a half-intensity width $\Delta\omega = 2/\tau$, where τ can be considered the ripplon lifetime. By spectral analysis of the scattered light at different values of the angles $(\theta_0, \theta, \varphi)$, we can determine the ripplon frequency ω and lifetime τ as functions of the wavelength Λ. Note, however, that considerable instrumental resolution is required, since the ripplon frequency $\omega/2\pi$ is usually less than 20 MHz.

In an earlier paper[3] we described a spectral analysis of the scattered light from surface vibrations of wavelength $\Lambda \simeq 1.2\lambda/n$ by means of a Fabry-Perot interferometer. The doublet could not be resolved, however, and only an over-all line broadening was found. We now report results in which the scattered light spectrum has been resolved by means of optical heterodyne spectroscopy.[4,5] These results unambiguously demonstrate scattering from thermal ripplons and permit the determination of the ripplon dispersion relation. Specific data are presented for methanol at wavelengths Λ between approximately $(1.84$ and $3.68) \times 10^{-3}$ cm.

The experimental arrangement is as follows:

Light from an argon ion laser, operating in the principal transverse mode at 4880 Å at a power of ~5 mW, is totally reflected from the liquid surface. The surface scattered light is collimated and recombined with a portion of the incident laser light at a beam splitter, and the combined beams are directed onto a photomultiplier. The resulting photocurrent, containing the difference frequencies between the two light beams, is filtered with an ultrasonic spectrum analyzer. To make the output of this analyzer proportional to the scattered-light spectrum (down-shifted by an amount Ω_0), a square-law detector (bolometer) is used at the i.f. output of the analyzer. To improve the signal-to-noise ratio, the scattered light is chopped before reaching the beam splitter, and the chopped output of the spectrum analyzer is fed to a synchronous detector and recorded.

Rather than attempt a direct measurement of the scattering angles, we found it convenient to calibrate the system by aligning the optics to the scattering from an artifically created surface wave of known wavelength. This surface wave was produced by a sound wave launched by an ultrasonic transducer (50-100 MHz) and reflected at the surface. If the wavelength of the sound wave is λ_S and the angle of incidence is θ_S, the wavelength of the corresponding surface wave is $\lambda_S/\sin\theta_S$.

Figure 1 shows recorded spectra of light scattered from thermal surface vibrations on methanol at three different wavelengths. The increase in frequency of vibration with decreasing wavelength is quite apparent, as well as the increase in spectral linewidth, thereby demonstrating a decrease in ripplon lifetime with frequency. It is interesting to compare these results with the predictions of the classical theory of surface vibrations from which we have $\omega/2\pi = (2\pi\sigma/\rho\Lambda^3)^{1/2}$, $\tau = \rho\Lambda^2/8\pi^2\mu S_r$, where σ is the surface tension, μ is the shear viscosity, ρ is the density of the liquid, and S_r is a quantity that appears in the numerical solution of the dispersion relation.[3] Using the static values for methanol at 25°C, $\sigma = 21$ erg cm^{-2} and $\mu = 5.50 \times 10^{-3}$ P, we obtain the values of $\omega/2\pi$ and $\Delta\omega/2\pi$ shown by the arrows and brackets in Fig. 1. As can
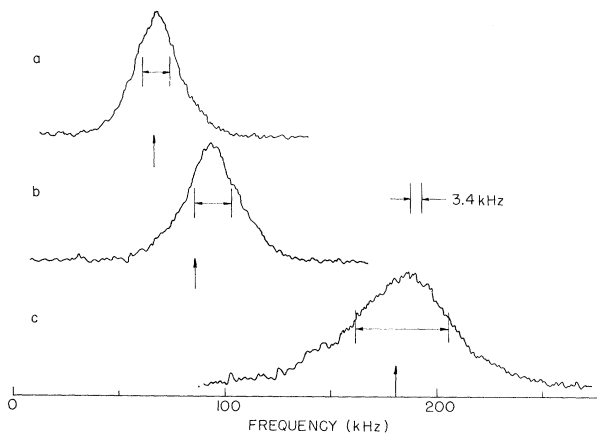
FIG. 1. Experimental spectra of light scattered from thermal surface vibrations on a liquid (methanol at 25°C) at three different surface wavelengths: (a) $\Lambda = 92\lambda/n$, (b) $\Lambda = 77\lambda/n$, and (c) $\Lambda = 47\lambda/n$, where $\lambda = 4880$ Å and $n = 1.33$. The arrows and brackets indicate the frequencies and linewidths calculated from classical theory of surface vibrations, with static values of surface tension and viscosity used. The instrumental bandwidth, including aperture broadening, is estimated to be 3.4 kHz, which is also indicated.

be seen, these values are in reasonably good agreement with the experimental values, and it appears that for this liquid (methanol at 25°C)

the classical dispersion relation and static values of σ and μ can be applied in the surface wavelength region considered here. A detailed discussion of experimental results in methanol and other liquids will be presented elsewhere.

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[1]W. Brouwer and R. K. Pathria, Phys. Rev. 163, 200 (1967).

[2]For wavelengths shorter than a critical wavelength $\Lambda_c = 1.16\pi\mu^2/\sigma\rho$, the surface vibrations are overdamped and exhibit nonoscillatory decay. For the liquid considered here, $\Lambda > \Lambda_c$, where $\Lambda_c \simeq 0.20\lambda/n$, and only underdamped, oscillatory vibrations occur, according to the classical theory. For further details see R. H. Katyl and U. Ingard, Phys. Rev. Letters 19, 64 (1967), and V. G. Levich, Physicochemical Hydrodynamics, translated by Scripta Technica, Inc. (Prentice-Hall, Inc., Englewood Cliffs, N. J., 1962), p. 603.

[3]Katyl and Ingard, Ref. 2.

[4]C. H. Townes, in Advances in Quantum Electronics, edited by J. P. Singer (Columbia University Press, New York, 1961), pp. 3-11.

[5]J. B. Lastovka and G. B. Benedek, in Physics of Quantum Electronics, edited by P. L. Kelley, B. Lax, and P. E. Tannenwald (McGraw-Hill Book Company, Inc., New York, 1966), p. 231; also see, Phys. Rev. Letters 17, 1039 (1966).

DIFFUSION OF EXCHANGE ENERGY IN SOLID He³ †

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We report a method for determining the rate of diffusion of exchange energy in solid He³. Over the temperature range 0.05-0.12°K, we obtain a temperature-independent diffusion constant $D_E = (0.75 \pm 0.15)Ja^2$, where a is the nearest-neighbor distance and J the exchange integral.

We report what we believe to be the first measurement of spatial diffusion of exchange energy in a paramagnet far above the magnetic transition temperature. The diffusion constant D_E, which is expected to be independent of temperature[1] and proportional to the exchange constant J, has been measured in the bcc phase of solid He³ using a new nmr technique. The experiment depended on the fact that the Zeeman system[2] (created by an external static magnetic field H_0) was in good thermal contact with the exchange system.

The constant D_E is to be distinguished from the Zeeman diffusion constant D_Z, which in

He³ was first determined by Reich[3] and has since been measured systematically by Thompson, Hunt, and Meyer.[4] Zeeman diffusion is a process wherein a temperature-independent transport of Zeeman energy results from the exchange mechanism and can be measured using well-known nmr techniques.[5] This differs from the present experiment since we determine the rate at which the total spin energy diffuses because of the exchange mechanism. The determination leads then to D_E.

The principle of the measurement is to heat the spin system locally and then watch this energy diffuse through the solid. In addition to