ly 2500 Å of aluminum. The aluminized LiF window of the uv detector was then bombarded directly with the proton beam. In this manner only those Lyman-α photons produced by de-excited protons in the LiF window could be detected. Figure 1 shows the Lyman-α photon counting rate as a function of proton energy. The cut-off point, corresponding to an energy of 22 keV, agrees closely with the range-energy data of Batzner[4] for protons in 2500 Å of aluminum.

The attenuation of 50-keV protons in 2500 Å of aluminum was measured to be at least 1000. This, together with the predetermined over-all sensitivity of the detector, yielded a minimum value for the production efficiency of 10^{-3} photons/proton. This value is of the order of magnitude of those obtained for surface-emitted photons (Table I) indicating that the majority of the observed Lyman-α photons originated within the target material.

The data were taken at four different angles of the surface relative to the proton beam as given in Fig. 2. The decrease in photon intensity with decreasing angles is attributed to a combination of increased attenuation in the aluminum filter and increased scattering.

To establish that the uv radiation observed in both the two previous experiments was in fact Lyman α, free of any uv fluorescence within the detector response range, a third experiment was conducted. A one-meter Seya-Namioka vacuum uv scanning spectrometer employing the tungsten photocathode multiplier was used to observe the uv radiation from both the aluminum target and the aluminized LiF target. In both cases, only the 1216-Å line of Lyman α was observed when protons impinged on the targets. Figure 3 shows the uv spectrum scan from the spectrometer. A beam of He$^+$ ions was also directed at the aluminized LiF target. A uv spectrum scan showed no observed uv lines within the response range of the tungsten photocathode detector.

This phenomenon may find application in mass analysis by providing a means of mass identification which is independent of charge-to-mass ratio; that is, by observation of the characteristic de-excitation spectra which is unique for a given ion. Also, it may prove to be a useful tool in the study of ion-impact phenomenon.

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SPECTRUM OF LIGHT SCATTERED QUASIELASTICALLY FROM A NORMAL LIQUID*

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We have observed and measured as a function of scattering angle $(0.33^\circ \leq \Theta \leq 3^\circ)$ the spectrum of light scattered quasielastically from entropy fluctuations in a *normal* fluid (toluene), using a laser light source and a high-sensitivity optical heterodyne spectometer. The magnitude and angular dependence of the linewidth are in excellent agreement with the predictions of the Leontovitch-Landau-Placzek theory.

We report measurements of the spectral distribution of the light scattered quasielastically from a normal fluid (toluene) at room temperature and atmospheric pressure. This feature of the spectrum of light scattered from thermal fluctuations was first observed by Gross,[1] who at the same time detected the Brillouin components. The latter components are produced by thermally excited propagating pressure fluctuations (sound waves) while the former "central" component (sometimes called the Rayleigh line) is produced by nonpropagating entropy fluctuations.[2,3]

A calculation of the scattered field shows that the light scattered at an angle Θ from the incident direction is produced by the spatial Fourier component of the fluctuations with wave vector $\vec{K}$ where $\vec{K} = \vec{k}_S - \vec{k}_0$;[4,5] $\vec{k}_S$ and $\vec{k}_0$ are, respectively, the wave vectors of the scattered and incident light. Since the frequency shifts are small ($|\vec{k}_s| \approx |\vec{k}_0|$), $|\vec{K}|$ is given by $|\vec{K}| = 2|\vec{k}_0|\sin\frac{1}{2}\Theta$. The spectrum of the central component is then determined by the time dependence of the entropy fluctuation with wave vector $\vec{K}$. Using the fact that these fluctuations decay according to the heat diffusion equation, Leontovitch[6] was able to show that this spectrum should be Lorentzian with a half-width at half-height of Γ radians, where

$$\Gamma = (\Lambda/\rho c_p)K^2 = [\Lambda/\rho c_p][4k_0^2 \sin^2\tfrac{1}{2}\Theta]. \qquad (1)$$

Here, Λ is the thermal conductivity, ρ is the density, and c_p is the specific heat per unit mass. The quantity $(\Lambda/\rho c_p)$ is called the thermal diffusivity. A more rigorous derivation of the spectrum recently obtained by Mountain[7] from an exact solution of the linearized hydrodynamic equations yields the same result. From Eq. (1) the central component linewidth goes to zero in the forward scattering direction and has its maximum value, $\Gamma_{\max}$, for backscattering. If we evaluate $\Gamma_{\max}$ for a number of liquids using the room-temperature values of Λ, ρ, and c_p, and assume the incident light to have a wavelength $\lambda \sim 6000$ Å, we find typically[5] $(\Gamma_{\max}/2\pi) \approx 10$ Mc/sec. Thus even the maximum broadening is so small that its observation requires an instrumental resolution $(\nu/\Delta\nu)$ exceeding 5×10^7. Furthermore, a study of the angular dependence of the spectrum of the scattered light toward the foward direction requires resolving powers of the order of 10^8 to 10^{14} corresponding to linewidths between 10 and 10^6 cps. This is far beyond the limits of conventional optical spectroscopy.

To study the spectrum of such narrow lines it is necessary to employ laser light sources combined with optical heterodyne[8-12] or self-beating[9,13,14] spectrometers. Alternatively, one could determine the spectrum, with comparable sensitivity and resolving power, by measuring the statistics of the time distribution of the photoelectrons emitted when a photosurface is illuminated with the scattered light.[15] The former two spectrometers have in fact[13,14,16] been used to study the width of the central component in fluids near their critical point. However, in those experiments the scattering cross section is 10^4 to 10^5 times greater than that from a normal fluid.

We have developed for the present study an optical heterodyne spectrometer using a multimode helium-neon laser ($\lambda = 6328$ Å) as a source and have achieved resolving powers from 5×10^{11} to 2×10^{14}. The sensitivity of this spectrometer is sufficient to detect 10^{-14} W of scattered power. The experimental arrangement is shown in Fig. 1. The sample of liquid, contained in a Brewster's angle cell, is located inside the laser cavity. In the absence of the sample the power in the laser cavity was typically 1 W. Insertion of the Brewster's angle sample cell, however, produced sufficient loss through the thermal lens effect[17] that the incident power was typically 50 mW. Light scattered away from the incident direction is collected and passed through an aperture which defines the angle Θ. The position of the aperture was fixed by deflecting part of the incident laser beam by the desired angle using the Debye-Sears effect.[18] The angle of deflection, and hence Θ, was determined to within ±0.002° by measuring the frequency of the sound waves temporarily injected for this purpose. Also passing through the aperture is light elastically scattered from "dust" or imperfections on the cell windows. This light at frequency ω_0,

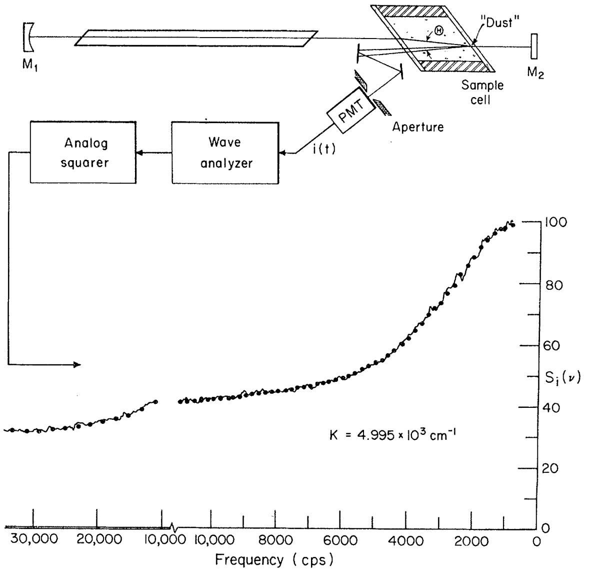

FIG. 1. A schematic representation of the optical heterodyne spectrometer used to study the spectrum of light scattered from toluene. The Brewster's angle sample cell is located inside the laser cavity formed by mirrors M_1 and M_2. Shown at the output is a typical photocurrent power spectrum, $S_i(\nu)$, obtained at a scattering angle, Θ, of about 2°, and with a wave-analyzer resolution of 300 cps. The heavy dots represent a fit to a Lorentzian of half-width 3374 ± 75 cps, plus a constant due to the shot noise.

which is generally ~1000 times as intense as the quasielastically scattered light, is used as the "local oscillator" of the spectrometer. Mixing between the signal and this local oscillator takes place on the surface of an RCA-7326 photomultiplier tube. For small-angle scattering and an aperture size of about one coherence area in the scattered field, the depth of field of the aperture-source system is such that the two wave fronts are accurately collinear over the photosurface, resulting in high mixing efficiency. The resulting photocurrent, $i(t)$, thus contains beat notes $(\Delta\omega)$ between all the spectral components of the two fields. In fact, we may show that the power spectrum of the photocurrent, $S_i(\Delta\omega)$, is related to the local oscillator power, P_{LO}, the power in the scattered light, P_S, and the normalized power spectrum of the scattered field, $S_E(\omega_0+\Delta_\omega)$, by

$$S_i(\Delta\omega)\propto P_{LO}P_SS_E(\Delta\omega). \tag{2}$$

The power spectrum of the photocurrent is thus identical with the power spectrum of the scattered light except that $S_i(\Delta\omega)$ is centered at $\omega=0$ rather than $\omega=\omega_0$. $S_i(\Delta\omega)$ also contains a frequency-independent contribution from the shot noise in the photocurrent. This shift of the center frequency is produced by the heterodyning process. The spectrum of the current is examined using a wave analyzer; a General Radio Model 1900 A was used for linewidths less than 2500 cps, and a Hewlett-Packard Model 310 A was used for larger widths. The output of both analyzers is a dc signal proportional to the root-mean-square photocurrent present in a small range of frequencies (the analyzer bandwidth) around the center frequency to which the analyzer is tuned. In order to obtain a signal proportional to the power spectrum of the scattered light, we square this dc signal prior to recording.

Using this spectrometer we have determined the spectrum of the central component in toluene liquid at room temperature and atmospheric pressure in the range $0.33°\leq\Theta\leq 3°$. The spectrum in this range was found to be accurately Lorentzian with a half-width at half-height $(\Gamma/2\pi)$ varying from 75 to 7500 cps. A typical spectrum and a least-squares fit to a Lorentzian plus a constant (the shot-noise part) is shown in Fig. 1. At each angle the half-width can be established to between 1 and 12%.

In Fig. 2 we show the results for the linewidth plotted versus K^2. The narrowest line $(\Gamma/2\pi=75$ cps) observed at $\Theta=0.33°$ corresponds to scattering from an entropy fluctuation with a wavelength $\lambda_f\cong 0.1$ mm, while for $\Theta=3°$ $(\Gamma/2\pi\cong 7500$ cps) we have $\lambda_f\cong 0.01$ mm. All measurements were taken with an acceptance angle $\delta\Theta$ such that $\delta\Theta/\Theta<0.1$. Each datum point has been corrected to $T=20.0°\mathrm{C}$ using the known temperature dependences of Λ, ρ, c_p, and the index of refraction. These data accurately follow the K^2 dependence, as predicted by Eq. (1), over a range of two orders of magnitude in K^2.

The slope of the best straight-line fit to the data, as shown in Fig. 2, gives

$$(\tfrac{1}{2}\pi)(\Lambda/\rho c_p)_{\text{exptl.}}=(1.38\pm 0.04)\times 10^{-4}\ \mathrm{cm^2/sec}.$$

Conventional $(K=0)$ thermodynamic measurements of the parameters entering into Eq. (1) give $\rho=0.8669$ g/cc,[19] $c_p=(1.6725\pm 0.016)$ J/g,[20] and $\Lambda=(1.38\pm 0.07)$ mW/cm °C,[20] from which we compute

$$(\tfrac{1}{2}\pi)(\Lambda/\rho c_p)_{\text{static}}=(1.52\pm 0.09)\times 10^{-4}\ \mathrm{cm^2/sec},$$

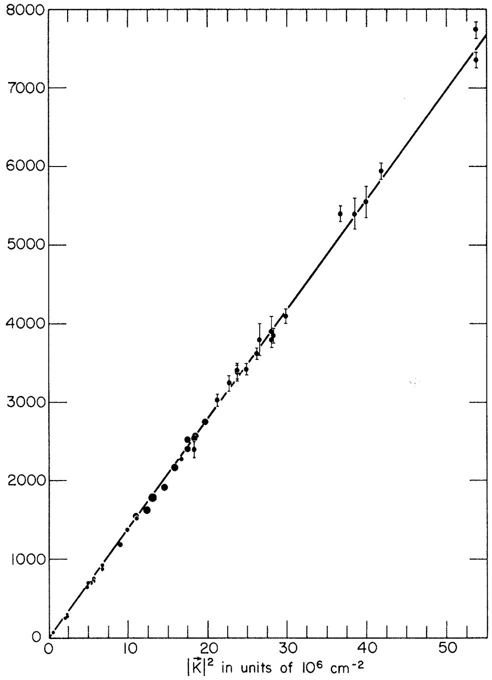

FIG. 2. Measurements of the half-width at half-height $(\Gamma/2\pi)$ of the power spectrum of the light scattered quasielastically from toluene as a function of K^2, the square of the magnitude of the scattering vector. Each datum point has been corrected to $T=20.0°\mathrm{C}$. The straight line demonstrates that the linewidth is accurately proportional to K^2.

which is in satisfactory agreement with our result. In addition, an ultrahigh-resolution optical measurement by Greytak[21] of the linewidth in the backward direction ($K = 2.94\times 10^5$ cm^{-1}, where $\lambda_f \cong 2\times 10^{-4}$ mm) gives a half-width of 10.5 ± 2 Mc/sec. Extrapolation of our low-K measurements to this value of K gives a width of 12.00 ± 0.35 Mc/sec.

These results (1) demonstrate that the Leontovitch-Landau-Placzek theory is correct for this liquid, i.e., that the heat-flow equation accurately describes both the temporal and spatial behavior of entropy fluctuations whose wavelengths range from $\lambda_f \cong 0.1$ mm to $\lambda_f \cong 2\times 10^{-4}$ mm; (2) support the recent application of this theory to the interpretation of the spectrum of light scattered from fluids near the gas-liquid critical point[13,14,16]; and (3) show that the spectrum of the quasielastic scattering provides a sensitive and accurate means of measuring the thermal diffusivity, not at infinite wavelength, as is the case in conventional measurements, but for finite wavelength. This enables a study of fluids whose thermal diffusivity may be a function of the wavelength[22] or the frequency[23] of the thermal fluctuation.

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