

our data is really quite good, and suggests that the interaction is hydrodynamic rather than electrostatic.⁴ The cross sections are consistently somewhat larger when the rings carry opposite charges than when the charges are the same. We suspect that this difference is accounted for by charge transfer (recombination).

The very low energy part of the data [see the low-voltage insert in Fig. 2(b)] requires separate discussion. Here V_1 is still fixed at some value chosen so that vortex rings reach collector 1. As V_2 is increased from zero, at some point the energy gained by a vortex becomes greater than that lost due to friction and rings enter the field-free region, where they experience only frictional effects and lose their energy. At the end of its range a charged ring will leave a free ion. As V_2 is increased, the range increases; for some value of V_2 free ions will be deposited at the position of the beam of vortex rings. When the polarities of the beams are opposite, the minimum in I_1 [Fig. 2(b)] occurs near the expected voltage, calculated by using values for the friction coefficient measured by Rayfield and Reif.¹ Evidently the free charge is recombining with the opposite charge on the ring, a process known to occur with vortex

lines.

As one might expect, on the basis of repulsive Coulomb forces, no capture seems to occur when the polarities of the two beams are the same. Ion capture on neutral rings should, however, be possible, offering interesting possibilities in probing the superfluid flow near orifices for ring vortices.

We wish to thank P. Hohenberg and G. Weinreich for helpful discussions and B. Brody for assistance in the reduction of data.

*This work was supported in part by the U. S. Atomic Energy Commission.

¹G. W. Rayfield and F. Reif, *Phys. Rev.* **136**, A1194 (1964).

²G. Gamota and T. M. Sanders, Jr., *Phys. Rev. Letters* **15**, 949 (1965).

³An early version of this experiment is described in G. Gamota, thesis, University of Michigan, 1966 (unpublished); see also, G. Gamota and T. M. Sanders, Jr., *Bull. Am. Phys. Soc.* **11**, 361 (1966).

⁴K. W. Schwartz, *Phys. Rev.* **165**, 323 (1968), describes an experiment related to the present one. He measures the interaction of vortex rings with vortex in rotating He II and obtains approximate agreement with hydrodynamic theory.

SPECTRUM OF LIGHT SCATTERING FROM THERMAL SHEAR WAVES IN LIQUIDS*

G. I. A. Stegeman and B. P. Stoicheff

Department of Physics, University of Toronto, Toronto, Canada

(Received 14 June 1968)

A depolarized doublet centered at the frequency of the exciting laser radiation has been observed in the light-scattering spectrum of several liquids, including nitrobenzene, aniline, quinoline, and *m*-nitrotoluene. The frequency separation, line shape, and polarization characteristics suggest that this doublet arises from thermally excited shear waves in these relatively viscous liquids.

In liquids with molecules having nonspherical polarizability, the spectrum of scattered light contains the usual central and Brillouin components and in addition, a depolarized component known as the Rayleigh "wing." This depolarized component is centered at zero frequency, is often very broad (sometimes extending as far as 10 or even 100 cm^{-1}), and arises from thermal fluctuations of anisotropy which are believed to originate from molecular rotational motions in liquids. The depolarized spectra of light scattering from liquids have long been known¹⁻³ and their intensities and linewidths have been stud-

ied by numerous authors, most recently with laser excitation.⁴⁻⁷ We have studied such depolarized spectra from a number of liquids, under the high resolution available with a He-Ne laser and a pressure-scanned Fabry-Perot interferometer. In some liquids, this spectrum consists of a doublet with an intensity minimum at the exciting frequency and with a peak separation of 1 to 3 Gc/sec, depending on the liquid. Similar spectra have been reported by Starunov, Tiganov, and Fabelinskii.^{8,9} These authors observed a variation in the doublet separation with k vector and on this basis proposed that the doublet is due

to shear deformation fluctuations or to transverse sound waves. We report here our observations of the polarization characteristics, line shape and intensity of the spectrum, along with measurements of the k -vector dependence of the doublet splitting. These results confirm in detail many of the features predicted in the theory of Rytov¹⁰ for light scattering by shear waves in liquids and thus lend strong support to the explanation that the depolarized doublet spectrum originates from scattering by thermally excited shear waves. It will also be shown that for those liquids for which the doublet splitting can be resolved, detailed analyses of the spectra yield values of the transverse sound velocity, the high-frequency shear modulus, and the shear relaxation time.

The experimental arrangement is essentially that of Chiao and Stoicheff.¹¹ The exciting source was a He-Ne laser which produced 50 to 200 mW of optical power at 6328 Å. A polaroid and polarization rotator were used to obtain incident light polarized parallel and perpendicular to the scattering plane. Light scattered at 90° from the sample (contained in a cylindrical cell) was gathered in a solid angle of 0.004 sr and passed through a polaroid. The spectral analysis of the

scattered light was carried out with a pressure-scanned Fabry-Perot interferometer having a spectral free range of 15 Gc/sec. The total instrumental linewidth (laser + Fabry-Perot) was from 0.75 to 0.95 Gc/sec (full width at half-intensity). The detection system consisted of an RCA 7265 phototube followed by Ortec photon-counting equipment and a recorder. The scattering cell was enclosed in a temperature-controlled copper block with sample temperature maintained and measured to $\pm 1^\circ\text{C}$.

Of the many liquids whose spectra have been investigated with this apparatus only aniline, *m*-nitrotoluene, *o*-nitroanisole, ethylene bromide, nitrobenzene, and quinoline exhibited depolarized doublets; liquids which have a depolarized spectrum but without the doublet include benzene, chloroform, carbon disulfide, and ethyl ether. Quinoline exhibited the best resolved doublet and therefore its spectrum will be used to illustrate the characteristic features of the depolarized doublet which are common to the above liquids.

The total spectrum of light scattering from quinoline with incident plane polarized light consists of the central and Brillouin components superimposed on an extremely intense and broad background. The central and Brillouin compo-

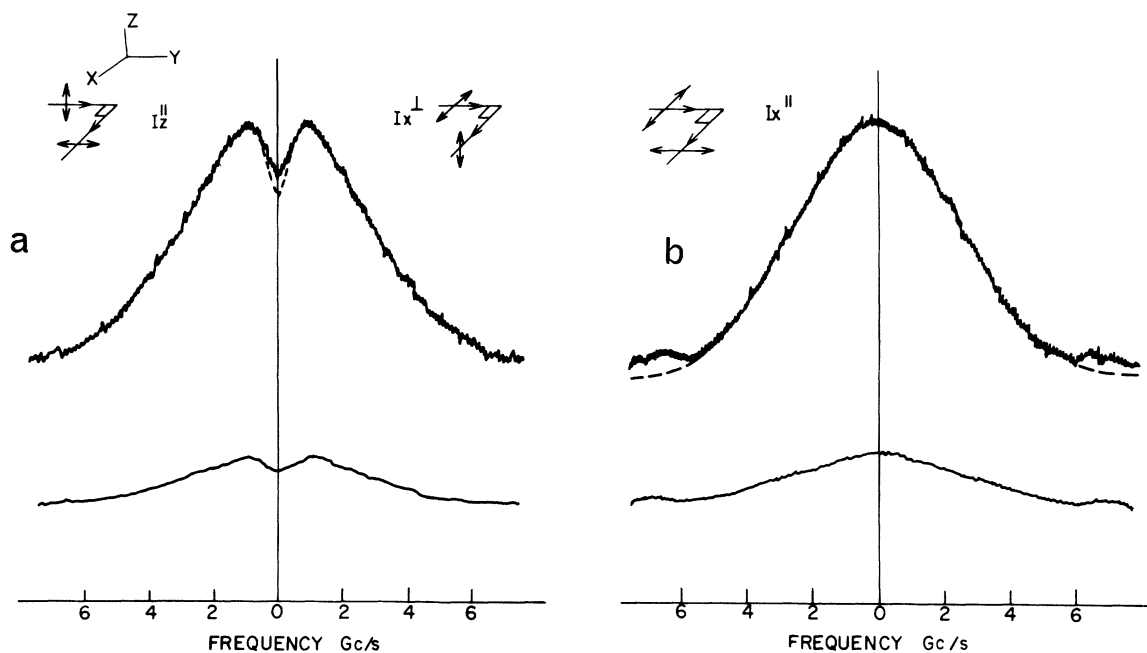


FIG. 1. Depolarized spectra of liquid quinoline (at 20°C) for a scattering angle of 90° to the incident radiation. (a) The doublet spectrum obtained with $I_{z,||}$ and $I_{x,\perp}$ polarization. (b) The Lorentzian line for $I_{z,||}$ polarization. In (a) and (b) the lower spectrum shows the relative intensity with respect to the broad background (baseline is zero intensity) and the upper spectrum is amplified by a factor of 5. The best fit to the theory is shown by the dashed lines.

nents are polarized and account for only 6% of the total intensity. The major part of the spectrum is depolarized, and it is shown in Fig. 1 for the various possible polarizations of the incident and scattered light (for scattering at 90°). Here, I_x and I_z represent the scattered light intensity for incident light polarized parallel and perpendicular to the scattering plane (x, y) and the subscripts \parallel and \perp refer to the state of polarization of the scattered light with respect to the plane (x, y). The $I_{z, \parallel}$ and $I_{x, \perp}$ spectra shown in Fig. 1(a) consist of a pronounced doublet, symmetrically displaced about the exciting frequency, and have a peak separation of the order of the breadth of the doublet. On the other hand, the $I_{x, \parallel}$ spectrum shown in Fig. 1(b) consists of a single broad component centered on the exciting frequency and has a weak Brillouin doublet in the wings. The integrated intensities $I_{x, \parallel}$ and $I_{x, \perp}$ are equal.

All of the above intensity and polarization characteristics of the spectra agree with the general predictions of a phenomenological theory of light

scattering due to temperature and stress fluctuations by Rytov. Stress fluctuations are coupled to fluctuations in the refractive index by the photoelastic constants \bar{X} and \bar{Y} , which are complex functions of frequency. The constant \bar{Y} relates purely volume deformations to changes in the refractive index and gives rise to the isotropic Brillouin doublet. Depolarized spectra appear due to the constant \bar{X} , which couples pure shear deformations to fluctuations in the index of refraction. Starting from the equations of linearized hydrodynamics, Rytov used fluctuation theory to derive expressions for the spectral composition of scattered light. The expressions are simplified if the complex shear modulus $\bar{\mu}$ and the photoelastic constant $\bar{X} \propto \bar{\mu}$ are assumed to be of the form $\bar{\mu} = i\omega\tau\mu_\infty / (1 + i\omega\tau) = \mu(\omega^2) + i\omega\eta$. Here μ_∞ is the limiting high-frequency shear modulus, τ is the shear relaxation time and anisotropy relaxation time (assumed to be equal), and η is the frequency-dependent shear viscosity. The resulting expressions for the intensity of light scattering at an angle of 90° are

$$I_{z, \parallel} = I_{x, \perp} = C \left[\frac{\omega^2}{\omega^2 + \tau^2(\omega^2 - \omega_T^2)^2} + \frac{1}{1 + \omega^2\tau^2} \right],$$

$$I_{x, \parallel} = \frac{C}{2} \left[\frac{3(\omega^2 - \omega_L^2)^2}{(\omega^2 - \omega_L^2)^2 + \omega^2\tau^2(\omega^2 - \omega_L^2 - \frac{4}{3}\omega_T^2)^2} + \frac{1}{1 + \omega^2\tau^2} \right],$$

$$I_{z, \perp} = \frac{4}{3}I_{x, \parallel} + \text{Brillouin spectrum.}$$

Here C is a constant and ω_L is the usual Brillouin shift. ω_T is defined by the relation $\omega_T = (\mu_\infty/\rho_0)^{1/2}K$ and is identified as the frequency of transverse shear waves (in analogy to ω_L as the longitudinal wave frequency). ρ_0 is the fluid density, and K is the wave vector of the fluctuations, which satisfies the Brillouin equation $K = 2K_0 \sin \frac{1}{2}\theta$. K_0 is the wave vector of light (at the incident frequency) in the medium and θ is the scattering angle.

According to this theory, the $I_{z, \parallel}$ and $I_{x, \perp}$ spectra are expected to display a doublet structure with separation $\sim 2\omega_T$. If $\omega_L^2 \gg \omega_T^2$, as is normally the case, then $I_{x, \parallel}$ should reveal a Lorentzian line and in the wings, where $\omega^2 = \omega_L^2 \pm \frac{4}{3}\omega_T^2$, a Brillouin doublet is expected. Also the intensities $I_{x, \parallel}$ and $I_{x, \perp}$ should be equal. All of these expectations are in agreement with the observed spectra of Fig. 1.

A careful analysis of the observed intensity profiles resulted in a more conclusive test of

the theory. Since the breadth of the doublet [Fig. 1(a)] is so large and the frequency shift is of the order of the instrumental linewidth, a convolution of the theoretical line shape with the instrumental line shape was necessary. By an iterative procedure a "best fit" to each spectrum was obtained. In Fig. 1(a) is shown a "best fit" of the doublet term $I_{z, \parallel}$ to the observed spectrum: The agreement is quite good except in the region of zero frequency, where the theory predicts a larger intensity dip than is observed. In Fig. 1(b) is shown a "best" fit of the $I_{x, \parallel}$ term to the observed spectrum. A Lorentzian line centered at zero frequency provides a good fit to this unsplit component; however, a "best fit" was not attempted in the wings of this line where a weak Brillouin doublet appears. (This doublet occurs at the same frequency as that arising from longitudinal waves and is thought to occur because of a weak coupling of shear and volume deforma-

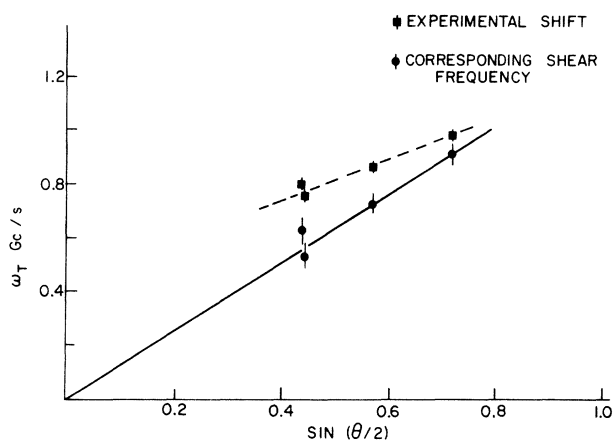


FIG. 2. Graph of observed doublet shift and corresponding shear-wave frequency ω_T in quinoline (from best fit to theory) showing dependence on scattering angle.

tions.) From the "best fit" of theoretical profiles to observed spectra values of the parameters ω_T and τ were obtained. For example, from the spectrum of the doublet ($I_{z, \parallel}$), $\omega_T = 0.91 \pm 0.04$ Gc/sec, and $\tau = 3.7 \times 10^{-11}$ sec; from the Lorentzian line ($I_{x, \parallel}$) essentially the same value, $\tau = 4.0 \times 10^{-11}$ sec, was obtained.

A further test of the correct interpretation of the origin of these spectra was made by investigating the dependence of the spectra on scattering angle. In particular, the variation of the frequency splitting of the doublet with angle was noted (although this variation could only be followed over a limited range of angle). As shown in Fig. 2, the direct measurement of the frequency shift (or splitting) did not exhibit a $\sin^{\frac{1}{2}}\theta$ dependence, contrary to the results of Ref. 8. However, with the "best-fit" procedure, the values of ω_T do show such a dependence. Thus we conclude that the depolarized spectra of Fig. 1 have their origin in fluctuations of anisotropy and in transverse sound waves. From the parameters ω_T and τ , a transverse sound velocity of 415 m/sec, a high-frequency shear modulus of 1.88×10^9 dyn/cm², and a shear relaxation time of 4.0×10^{-11} sec are determined for liquid quinoline at 20°C.

The temperature dependence of the doublet splitting was also investigated. For temperatures above 20°C, ω_T is approximately constant; below 20°C, ω_T (as well as the observed splitting) decreases with decreasing temperature. This behavior is contrary to theory and is not

Table I. Values of frequency shift ω_T , velocity of shear waves, and shear modulus derived from depolarized spectra of liquids.

Liquid	ω_T	v_T	μ_∞
	(Gc/sec)		
	($\theta = 90^\circ$)	(m/sec)	(10^9 dyn/cm ²)
Quinoline	0.95	415	1.88
Aniline	1.25	560	3.21
Nitrobenzene	0.46	212	0.54
<i>m</i> -Nitrotoluene	0.77	365	1.54

yet understood. Only in the temperature region where ω_T is constant can the parameters ω_T and τ be used to evaluate the shear velocity and high-frequency shear modulus. Values of these quantities for aniline, *m*-nitrotoluene, nitrobenzene, and quinoline derived from these spectra are given in Table I.

In conclusion, the results of this investigation confirm the suggestion made by Starunov, Tiganov, and Fabelinskii that these depolarized spectra are due to light scattering from shear waves in liquids, and establish the validity of Rytov's theory. Also, it has been shown that detailed analyses of such spectra lead to the determination of the velocity and attenuation of shear waves, and of the shear modulus in liquids.

We are very grateful to Mr. W. S. Gornall for experimental assistance during the early stages of this investigation and to Dr. V. Volterra for helpful discussions on the theory.

*Research supported in part by the National Research Council of Canada.

¹S. Bhagavantam, *Scattering of Light and the Raman Effect* (Chemical Publishing Company, Brooklyn, N. Y., 1942), pp. 61 and 265.

²J. Frenkel, *Kinetic Theory of Liquids* (Oxford University Press, Oxford, England, 1946), p. 294.

³I. L. Fabelinskii, *Molecular Scattering of Light* (Plenum Press, Inc., New York, 1968), p. 132.

⁴D. H. Rank, A. Hollinger, and D. P. Eastman, *J. Opt. Soc. Am.* **56**, 1057 (1966).

⁵V. S. Starunov, E. V. Tiganov, and I. L. Fabelinskii *Zh. Eksperim. i Teor. Fiz.—Pis'ma Redakt.* **4**, 262 (1966) [translation: *JETP Letters* **4**, 176 (1966)].

⁶S. L. Shapiro and H. P. Broida, *Phys. Rev.* **154**, 129 (1967).

⁷A. Szöke, E. Courtens, and A. Ben-Reuven, *Chem. Phys. Letters* **1**, 87 (1967).

⁸V. S. Starunov, E. V. Tiganov, and I. L. Fabelinskii, *Zh. Eksperim. i Teor. Fiz.—Pis'ma Redakt.* **5**, 317

(1967) [translation: JETP Letters 5, 260 (1967)].

⁹I. L. Fabelinskii and V. S. Starunov, Appl. Opt. 6, 1793 (1967).

¹⁰S. M. Rytov, Zh. Eksperim. i Teor. Fiz. 33, 514,

669 (1957) [translations: Soviet Phys.-JETP 6, 401, 513 (1958)].

¹¹R. Y. Chiao and B. P. Stoicheff, J. Opt. Soc. Am. 54, 1286 (1964).

OBSERVATIONS OF NONLINEAR DAMPING OF TONKS-DATTNER RESONANCES

J. F. Decker and C. W. Mendel, Jr.

Bell Telephone Laboratories, Whippany, New Jersey

(Received 3 June 1968)

A strong decrease in the damping of Tonks-Dattner resonances with increasing strength of the exciting microwave field has been observed in low-pressure mercury and argon dc discharges. Order-of-magnitude estimates indicate that a nonlinear effect involving a balance between wave-particle and particle-particle collisions may be responsible.

We have measured the amplitudes and widths of Tonks-Dattner resonances as a function of the magnitude of the exciting microwave field. For a certain range of applied fields a nonlinear increase in the amplitude, a decrease in the width, and no change in resonant frequency are observed with increasing field strengths. We have attributed this behavior to a change in Landau damping of the excited longitudinal plasma wave, the change being produced by a flattening of the steady-state electron velocity distribution. Recently, observations of nonlinear spatial Landau damping of both electron plasma waves¹ and ion acoustic waves² propagating along magnetized plasma columns have been reported. The results of those experiments were explained in terms of collisionless theories. However, for the present observations, we believe that the balance between the wave-particle interaction and particle-particle collisions is important in determining the field strengths at which the nonlinear effect is observed.

The Tonks-Dattner resonances are observed by monitoring the microwave power reflected, P_R , from a discharge tube inserted through the narrow side of RG 95/U waveguide (1.872 in. \times 0.872 in., inside dimensions). As is commonly done we keep the incident microwave frequency constant and vary the discharge current. In contrast to other experiments where the incident microwave power is held constant, we keep the power transmitted past the discharge tube, P_T , constant. It can be shown that if $2a/\lambda_g \ll 1$, where a is the tube radius and λ_g is the wavelength of the radiation in the waveguide, this is equivalent to maintaining a constant amplitude of the microwave electric field at the discharge tube as the plasma goes through a resonance. Such a method

leads to a simple interpretation of the resonances observed in the reflected power without making impedance measurements.

A schematic diagram of the apparatus is shown in Fig. 1. P_T is leveled by controlling the signal source output power with a negative feedback signal proportional to the difference between the power past attenuator No. 1 and a preset value. Since the leveling circuit keeps the power past the attenuator A_1 constant, changing A_1 by Δ dB changes P_T by the same amount. Measurements are made as a function of P_T by changing both A_1 and A_2 by the same amount. A_3 is changed by $-\Delta$ dB to maintain a constant signal-to-noise ratio from the signal source and to insure that the feedback circuit operates at a constant power level. If the reflection process is linear with power, then the signal incident on the receiver is constant. The microwave system itself was found to be linear to within ± 0.15 dB over the full range of 50 dB.

The first few thermal resonances³ in the Hg discharge are well separated and at low power levels are observed to obey the following rela-

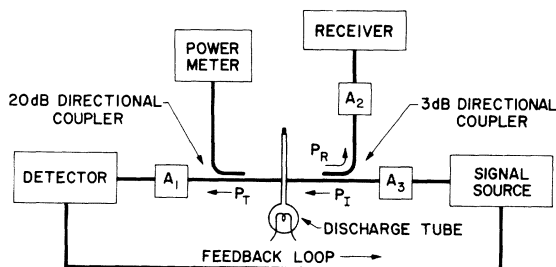


FIG. 1. Schematic diagram of apparatus.