Iodine spectrometer for low-shifted light scattering

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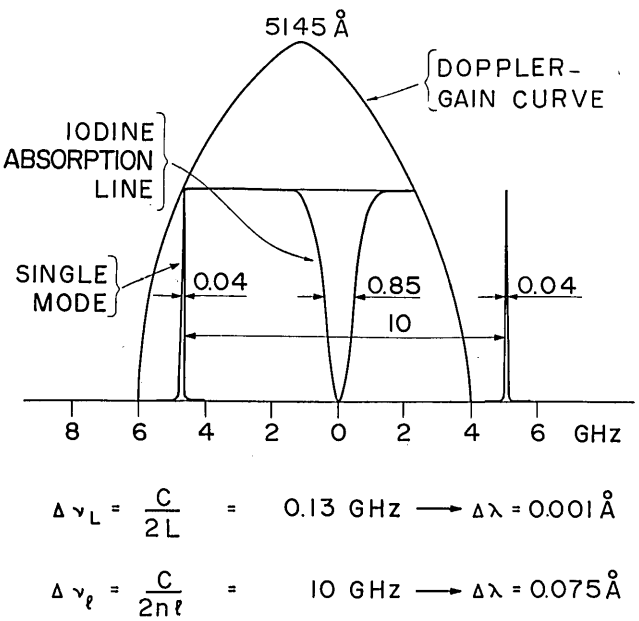
Using an iodine-vapor cell and a single-mode argon-ion laser, we have constructed a spectrometer for low-shifted light scattering, which has resolution better than 0.3 GHz (0.01 cm^{-1}). A method has been proposed to deconvolute the spectra.

Index Headings: Spectrometer; Laser; Brillouin scattering; Resolution.

Frequency tuning of the single-mode output of an ion laser is well known. A 1.15-m-long commercial argon-ion laser manufactured by Coherent Radiation Inc. has approximately 60 modes spaced at 0.13 GHz intervals along the gain curve of the 5145 Å excitation. The single mode is selected by use of an intracavity étalon 1 cm thick with a free spectral range of 10 GHz.

These details are shown in Fig. 1. The system stability is assured by maintaining the temperature of the étalon to an accuracy of ±0.01 K. The thermal contraction (expansion) of the étalon with decreasing (increasing) temperature produces a shift of the selected longitudinal single mode. This suggests the possibility of sweeping through a number of such modes. When a transition from one mode to another occurs, the light-power stabilizer is activated and a signal is extracted, which is fed to an event marker in the recorder to obtain a frequency interval of 0.13 GHz. This simple technique provides a tunable laser over a narrow frequency region (approximately 8 GHz).



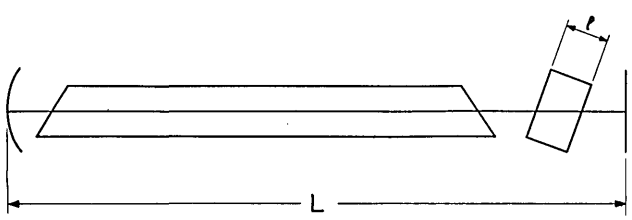


FIG. 1. Intensity of single mode of argon laser (5145 Å vs frequency.

The tunable laser scans over a small frequency range, compared to dye or parametric lasers, but it produces extremely narrow-band output of 0.04 GHz width and the iodine-gas absorption transition is well inside the tunable range.[1] This is particularly suited for studying phenomena such as Brillouin scattering. Details of the experimental set up are shown in Fig. 2. An iodine filter is introduced in the light scattered from the sample; subsequently the transmitted light is focused on a photomultiplier for measurement. When the laser is tuned through its available frequency range, a Brillouin-scattering spectrum is superimposed with the iodine absorption. The spectrum can then be deconvoluted by use of the theory described in the next section.

The advantages of Brillouin-scattering investigation by this technique are worth noting. We have a source of very-narrow half-width, and the whole apparatus is very inexpensive, because it does not need an interferometer to analyze the spectrum. The resolution is an order of magnitude better than is obtained with a typical Fabry–Perot interferometer.[2] In order to test our spectral system, we studied Brillouin scattering in carbon tetrachloride, benzene, methyl alcohol, and ethyl alcohol. The frequencies and the half-widths of the acoustic phonons obtained after deconvolution are listed in Table I.

THEORY

Each point in the spectrum obtained by the technique described represents the total output over the entire frequency range, part of which is absorbed by the iodine filter. In order to deconvolute it to obtain the real frequency of the acoustic phonon and its half-width we must express the intensity distribution of the phonon mathe-

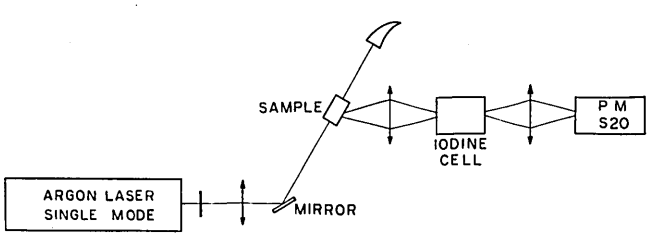


FIG. 2. Schematic diagram of the apparatus.

TABLE I. Results of the Brillouin scattering (iodine half-width = 0.85 GHz).

Substance	Γ in GHz	ν_0 in GHz
CCl_4 $\theta = 63°52'$	0.580	3.088
C_6H_6 $\theta = 64°39'$	0.340	4.413
C_2H_5OH $\theta = 61°50'$	0.140	3.076
CH_3OH $\theta = 61°08'$	0.780	3.100

matically. In classical theory, this can be approximated by a damped harmonic oscillator

$$A(\nu) = \frac{x_0 \nu_0^2 \Gamma |\nu|}{(\nu_0^2 - \nu^2)^2 + \Gamma^2 \nu^2} n(\nu), \tag{1}$$

where Γ is the half-width and ν_0 is the frequency of the phonon. The function $n(\nu)$ represents the Bose distribution and can be expressed as

$$n(\nu) = \frac{1}{|1 - \exp(-h\nu/kT)|}$$

The total scattered intensity at frequency x, including

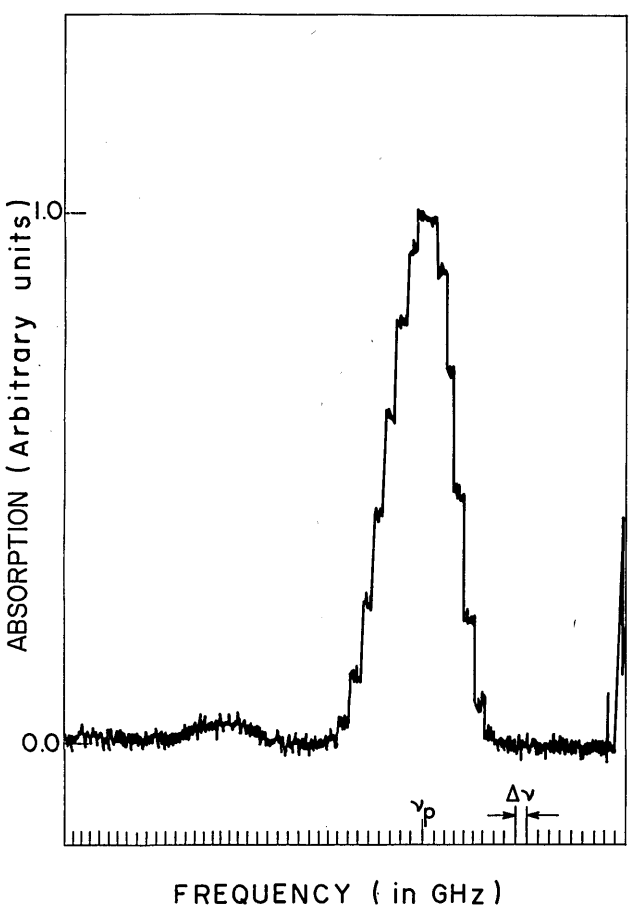


FIG. 3. Iodine absorption peaking at 5145.42 Å. Tick marks are separated by 0.13 GHz.

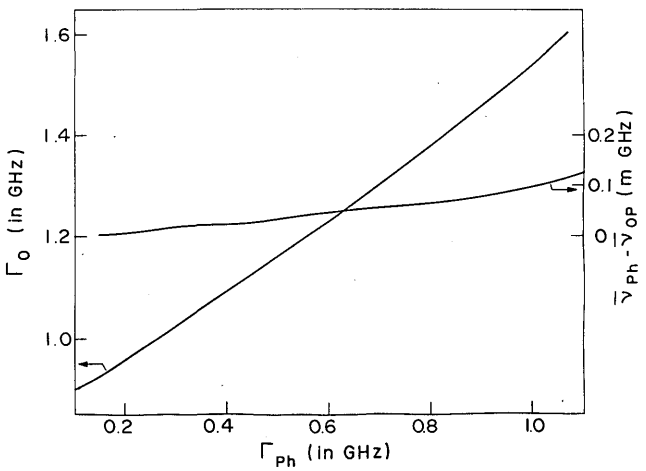


FIG. 4. Relation between real and observed half-width and frequency shift of the phonon with iodine absorption. Γ_0 is the observed half-width and ν_{op} is the observed peak position. Γ_{Ph} and ν_{Ph} are the real half-width and frequency, respectively.

iodine absorption, can then be expressed by the integral

$$f(x) = \int_{-\infty}^{\infty} A(x - x') G(x')\, dx', \tag{2}$$

where $G(x')$ represents the iodine absorption line. In order to know the exact form of the function $G(x')$, we studied the iodine absorption by scanning through the appropriate range with the laser in the single mode. The spectrum is shown in Fig. 3. Because the width of the input laser in single mode is just 0.04 GHz, it can

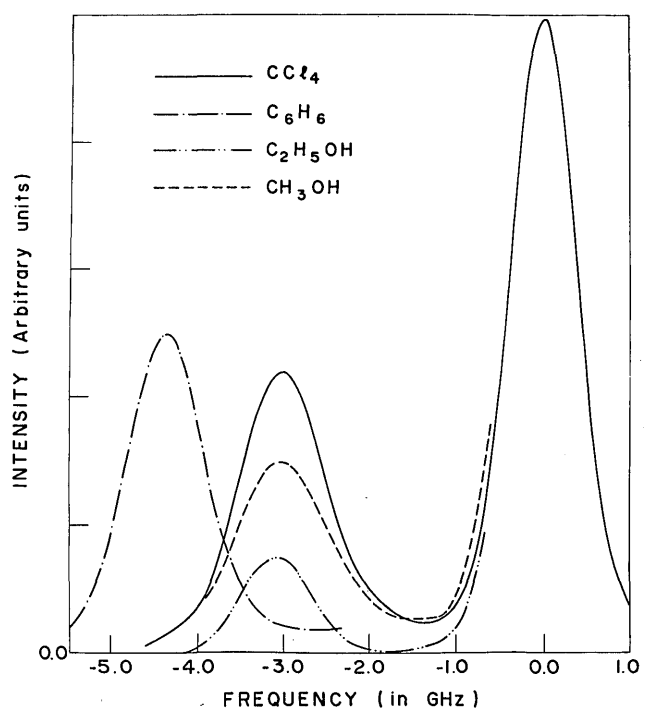


FIG. 5. Observed Brillouin spectra with iodine filter for various liquids. Intensities in various spectra are not to be compared.

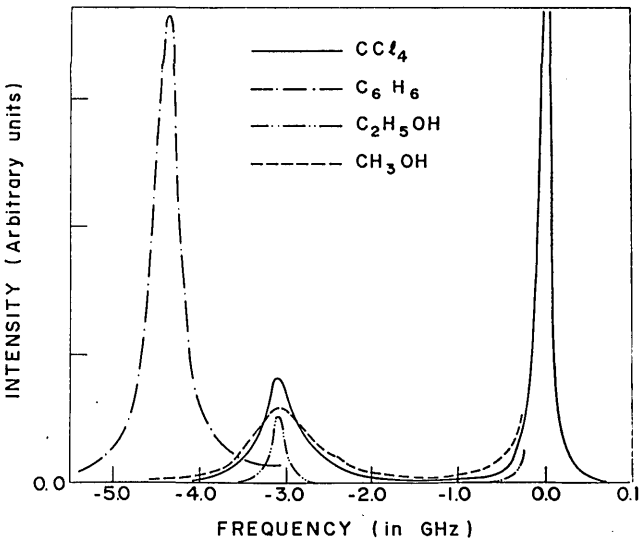


FIG. 6. Deconvoluted Brillouin spectra of CCl_4, C_6H_6, C_2H_5OH, and CH_3OH. The relative scattering efficiencies of various liquids are not to be compared.

be treated as a delta function; therefore the observed spectrum should represent the function $G(x')$. This function is gaussian with half-width 0.85 GHz.

The function $f(x)$ can now be fitted by a least-squares method to obtain Γ and ν_0 for the phonon. Conversely, for various values of Γ and ν_0, the apparent half-width and peak position of the phonon on the measured spectrum, i.e., $f(x)$, can be obtained. In Fig. 4, we have plotted such theoretical curves. These curves are useful for those working on similar experiments, to obtain the frequency and the half-width of the phonon without doing the numerical work involved in fitting.

In order to deconvolute the Rayleigh scattering, we need to replace the function $A(\nu)$ in Eq. (2) by a lorenzian function that represents Rayleigh scattering. The half-widths of the Rayleigh scattering obtained for the above liquids are typically of the order of 0.1 GHz. These are larger than those obtained from the expressions of Mountain.[3] The difference is presumably due to the fact that we are scanning with the frequency steps of 0.13 GHz. Table I summarizes the results of Brillouin scattering obtained by the technique described. The over-all accuracy is estimated to be of the order of 0.1 GHz.

Brillouin spectra observed with the iodine filter are plotted in Fig. 5. The intensity scale is arbitrary, and no attempt has been made to compare the scattering efficiencies of various liquids. Theoretical spectra obtained by deconvolution are plotted in Fig. 6. The liquids used in our experiments were not highly pure; therefore, the ratio of the intensity of the Rayleigh scattering to the intensity of the Brillouin scattering is very large compared to the theoretical predictions.[3]

ACKNOWLEDGMENT

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[2]Herman Z. Cummin and Robert W. Gammon, J. Chem. Phys. 44, 2785 (1966).
[3]R. D. Mountain, J. Res. Natl. Bur. Std. A70, 207 (1966).