Stimulated Rayleigh-Brillouin Gain Spectroscopy in Pure Gases

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Stimulated Rayleigh-Brillouin gain spectra of pure gases are reported. Stimulated Rayleigh scattering is now clearly distinguishable from stimulated Brillouin scattering without the aid of induced absorption. As examples, spectral line shapes of Ar and SF_6 at various pressures and the associated gain asymmetry are discussed. Potential applications of coherent Rayleigh-Brillouin spectroscopy are suggested.

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Using Ar and SF_6 at pressures ranging from 0.5 to 4.1 atm, we report the first study of stimulated Rayleigh-Brillouin gain spectroscopy in gases. This was done by crossing two single-frequency laser beams in the gas samples of interest. Although similar physical information can be obtained from corresponding spontaneous light-scattering experiments,[1] the present coherent method does not require a Fabry-Perot interferometer for analysis. Thus, the resolution of the experiment is limited only by the laser linewidths. As in Raman gain techniques,[2] the detected background light in stimulated Rayleigh-Brillouin gain spectroscopy is greatly reduced since the desired signal is emitted only along the probe beam. Thus, the stimulated Rayleigh-Brillouin gain spectroscopy reported here is a potentially sensitive technique for probing the low-frequency excitations and fluctuations in any system with weak scatterers or with high background light.

Experimentally, two mildly focused, counterpropagating beams are crossed at a small angle of $\sim 1^\circ$ in a Brewster-windowed gas cell. The probe beam, with wave vector $\vec{k}_s$ and frequency ω_s, is from a fixed-frequency, 30-mW, cw, single-mode dye laser operating at 5850 Å. The pump beam with $\vec{k}_l$, ω_l is the output of a single-frequency tunable dye laser amplified to yield pulses of 5 ns width and 1 MW peak power at 10 pulses per second. The linewidths of the pump and probe lasers are 150 and 1 MHz, respectively. After traversing the interaction cell containing pure gas at a preset pressure and temperature (296 K), the probe laser beam was detected by a fast photodiode and the signal was filtered and processed by a boxcar integrator. As the pump laser is frequency scanned across the probe-laser frequency, a stimulated Rayleigh-Brillouin (gain/loss) spectrum is obtained.

It is well known that the gain of stimulated scattering is directly proportional to the imaginary part of the associated third-order susceptibility $\chi^{(3)}$. Since the wave-vector–dependent spectral density function[3] $S(\vec{K}, \Omega)$, with $\vec{K} = \vec{k}_l - \vec{k}_s$ and $\Omega = \omega_l - \omega_s$, is often used to describe spontaneous light scattering experiments,[1] we may also relate $\mathrm{Im}\chi^{(3)}$ to $S(\vec{K}, \Omega)$ by the generalized fluctuation-dissipation theorem in statistical mechanics[4] and express the gain in terms of $S(\vec{K}, \Omega)$. In SI units, the stimulated Rayleigh-Brillouin gain per unit length is

$$g(\omega_s) = -[2\omega_s/(c\epsilon_0)^2] I_l [\mathrm{Im}\chi^{(3)}] \quad (1a)$$

$$= \frac{8\pi^3 c^2}{\omega_s^3} \frac{N}{k_B T} \frac{d\sigma}{d\Omega} [\Omega S(\vec{K}, \Omega)], \quad (1b)$$

where N, $d\sigma/d\Omega$, and I_l are, respectively, number density, spontaneous Rayleigh scattering cross section, and pump intensity. For pure gases, the form of the spectral density $S(\vec{K}, \Omega)$ depends on a dimensionless parameter[3] y. For convenience, we may refer to $y \gtrsim 3$ as the hydrodynamic regime and to $y < 3$ as the kinetic regime. In the hydrodynamic regime, the expression for $\mathrm{Im}\chi^{(3)}$ is given in terms of thermodynamic parameters by Herman and Gray[5] (with the absorption coefficient α set to zero). In the kinetic regime, we shall use Eq. (1b) and the $S(\vec{K}, \Omega)$ calculated, for simplicity, by the model of Yip and Nelkin.[3] For larger y values, the hydrodynamic model is more realistic because specific heats, thermal conductivity, and viscosity may enter into the model independently. For very dilute gas, as $y \to 0$, the individual gas molecules or atoms scatter light independently without collective effects, and the stimulated scattering gain should approach that obtained from the transition rate of individual particles in thermal equilibrium. One can also show that the gain obtained from the associated Rayleigh (two-photon) scattering transition rate due to individual moving atoms is identical to Eq. (1b) when y in $S(\vec{K}, \Omega)$ is set equal to zero.

In Fig. 1, the stimulated Rayleigh-Brillouin gain spectrum of SF_6 at 4.1 atm, corresponding to $y = 7.2$, in the hydrodynamic region is shown. The solid curve corresponds to experimental result. Several features may be noticed. First, the dominance of Brillouin scattering is evident and the location of these peaks corresponds to a sound velocity of $v_s = 1.45 \times 10^3$ m/s as expected. Second, although the features near the unshifted frequency are much smaller than the corresponding features of Brillouin scattering, they are well separated in frequency and stimulated Rayleigh scattering (marked by arrows) can clearly be seen. Based on thermodynamic parameters[6] of SF_6, a theoretical curve of $\mathrm{Im}\chi^{(3)}$ is shown in the inset in Fig. 1; since the pump laser linewidth is ignored, the Rayleigh peaks are more evident. Similar to the Brillouin peaks, the Rayleigh features exhibit gain on the Stokes side with $\Omega = \omega_l - \omega_s > 0$ and negative gain or loss on the anti-Stokes side where $\Omega < 0$. This is contrary to earlier experiments of absorption-induced stimulated Rayleigh scattering where the maximum gain occurs on the anti-Stokes side. In those experiments, one laser beam was used and there was the need to overcome a threshold in order to observe stimulated scattering. The competition from Brillouin scattering prevented the weak gain of Rayleigh scattering from reaching the desired threshold unless the temperature fluctuations of the medium were enhanced by impurity absorption[7] or by laser-induced plasma.[8] There is no laser-induced plasma in our experiments because of the relatively low pump intensity (< 5 GW/cm^2) used. To our knowledge, this represents the first clear observation of stimulated Rayleigh scattering in both the Stokes and anti-Stokes sides from a pure substance.[9] Lastly, there exists an anomalous asymmetry between the gain and loss portions of the Brillouin spectrum. Since the signal of the gain spectroscopy is proportional to $\exp G - 1$, this interesting gain asymmetry is to be expected when the absolute value of the total gain $|G|$ is greater than 0.1. Similar asymmetric behavior has also been observed in recent coherent Brillouin spectroscopy of liquids.[10] Taking into account the gain asymmetry with measured $G = 1.32$ and the finite pump laser linewidth, we show as the dotted curve in Fig. 1 a theoretical spectrum normalized to the loss peak. Agreement with experimental data is very good.

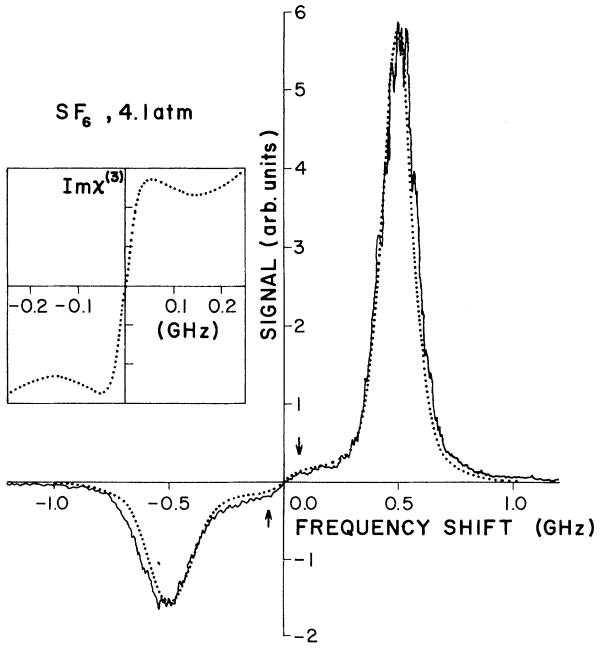

FIG. 1. Rayleigh-Brillouin gain spectrum of SF_6 compared with the hydrodynamic model with $G = 1.32$ at 4.1 atm and 296 K. The inset is a calculated curve with the pump linewidth ignored.

The experimental Rayleigh-Brillouin gain spectra of Ar at pressures 0.5, 0.9, 2.5, and 4.1 atm, corresponding respectively to $y = 0.3$, 0.5, 1.5, and 2.5, are shown in Fig. 2. For low pressures ($y = 0.3$ and 0.5), there is no apparent gain asymmetry because the total gain G is smaller than 0.1. Within the experimental errors, the simple kinetic model[3] (solid curves) with pump laser linewidth accounted for yields results in agreement with the experimental spectra. At these pressures, the Brillouin doublet is highly suppressed and one sees only a combined scattering feature which is commonly referred to as Rayleigh scattering. In the transition region, $y = 1.5$, the hydrodynamic model (dashed curve) using the given thermodynamic parameters[11] still fails, although it begins to work for $y = 2.5$. The sound-wave propagation now begins to take place and the experimental data show a minor but discernible discrepancy with the early kinetic model.[3] This discrepancy can be reconciled with an improved numerical procedure for solving the Boltzmann equation.[12] Since gas dynamics is not the subject of this paper, we used a simpler kinetic model to compare with our experimental spectra.

In addition to the spectra of Ar and SF_6, we have also obtained and analyzed spectra of N_2 and CO_2 at different pressures. We note from Eq. (1b) that for the same density and pump in-

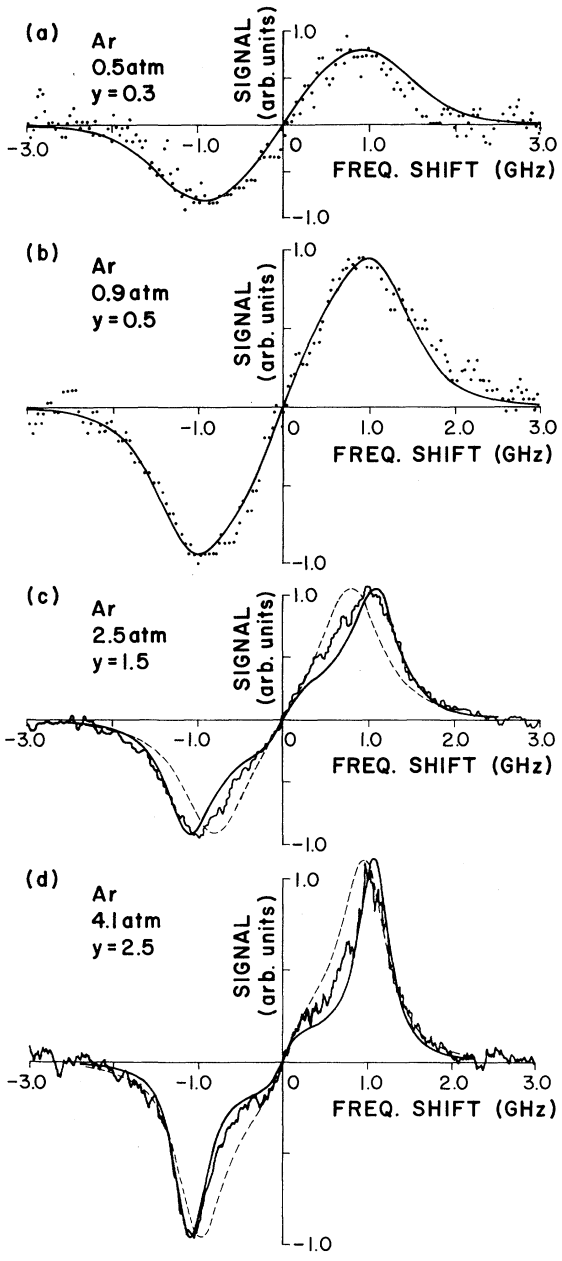

FIG. 2. Rayleigh-Brillouin gain spectra of Ar at different y values compared to the hydrodynamic theory (dashed lines) and the kinetic theory of Yip and Nelkin (smooth solid lines). The experimental curves are shown as dots in (a) and (b) and as wiggly solid lines in (c) and (d).

tensity, the stimulated Rayleigh gain is directly proportional to the spontaneous Rayleigh cross section. This is indeed the case when the gain of Ar, N_2, and CO_2 at the same pressure (1 atm) are compared. Theory further indicates that the Rayleigh-scattering gain increases linearly with pressure and the Brillouin-scattering gain increases quadratically with pressure.[6] Analysis of our SF_6 data confirms the Brillouin case, although our present signal-to-noise ratio does not allow a similar determination for the Rayleigh case. In evaluating the 4.1-atm SF_6 spectra, pump powers ranging from 0.05 to 1.2 MW were used. Analysis of this set of data shows that the measured total gain G is linearly proportional to the pump power. Furthermore, the measured absolute total gain of our experiments is also consistent with the expressions given in Eq. (1).

The Rayleigh-Brillouin gain spectroscopy reported here is only one branch of a larger family of coherent spectroscopies. With improved lasers, e.g., with use of a cw single-frequency pump laser, spectra with beams crossing at arbitrary angles (thus probing the different scattering wave vectors $\vec{K}$) with higher spectral resolution and better signal-to-noise ratios should be possible. With use of polarization spectroscopy and three-dimensional phase matching a variety of coherent Rayleigh-Brillouin spectroscopies can be implemented and the respective tensor elements of $\chi^{(3)}$ can be measured. Although gases are used as examples to demonstrate the potential of these methods, features with very low frequency shift in liquids and solid surfaces can also be studied this way without the interference of elastic scattering. Thermodynamic properties of matter, including concentration fluctuations and critical fluctuations in systems with weak scatterers or high background light, can be more easily measured. In addition, since one laser is frequency scanned across the other, this technique is also ideal for probing closely spaced ($\Delta\nu \lesssim 1$ GHz) energy levels or low-frequency excitations.

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[1]T. J. Greytak and G. B. Benedek, Phys. Rev. Lett. 17, 179 (1966).

[2]P. Esherick and A. Owyoung, in *Advances in Infrared and Raman Spectroscopy*, edited by R. J. H. Clark and R. E. Hester (Heydon and Son, London, 1982), Vol. 9, p. 130.

[3]S. Yip and M. Nelkin, Phys. Rev. 135, A1241 (1964).

[4]A. S. Barker and R. Loudon, Rev. Mod. Phys. 44,

18 (1972).

[5]R. M. Herman and M. A. Gray, Phys. Rev. Lett. 19, 824 (1967).

[6]W. H. Lowdermilk and N. Bloembergen, Phys. Rev. A 5, 1423 (1972).

[7]T. A. Wiggins, C. W. Cho, D. R. Dietz, and N. D. Foltz, Phys. Rev. Lett. 20, 831 (1968).

[8]C. J. Hsu, N. D. Foltz, and C. W. Cho, Phys. Rev. A 9, 2365 (1974).

[9]I. L. Fabelinskii, D. I. Mash, V. V. Morozov, and V. S. Starunov, Phys. Lett. 27A, 253 (1968). These authors had observed stimulated Rayleigh scattering in H_2 on the Stokes side. No information on spectral shape was obtained.

[10]A. G. Jacobson and Y. R. Shen, Appl. Phys. Lett. 34, 464 (1979).

[11]S. Chapman and T. G. Cowling, *The Mathematical Theory of Non-Uniform Gases* (Cambridge Univ. Press, Cambridge, England, 1952).

[12]N. A. Clark, Phys. Rev. A 12, 232 (1975).