

## Stimulated Rayleigh-Brillouin gain spectroscopy

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Steady-state stimulated Rayleigh-Brillouin gain spectroscopy and its application are reported. The stimulated gain, which is derived in terms of the spectral function of density fluctuations, is shown to approach, in the limit of zero density, the gain derived from the individual-particle picture of Rayleigh scattering. Using a pulsed pump laser, experimental gain spectra of pure SF<sub>6</sub>, pure Ar, and a SF<sub>6</sub>- and He-gas mixture are obtained and explained. Other associated coherent Rayleigh-Brillouin spectroscopies and the possibility of using a cw pump laser for higher spectral resolution are discussed.

### I. INTRODUCTION

For about a decade after the discovery of stimulated Raman scattering<sup>1,2</sup> in 1962, the phenomenon of stimulated light scattering has been investigated extensively. Usually with a liquid medium and a single high-power pump-laser beam, nearly every mechanism of spontaneous scattering has been shown to yield stimulated scattering. Although with greater difficulty<sup>3,4</sup> than stimulated Raman scattering, stimulated Brillouin scattering<sup>5</sup> and Rayleigh scattering<sup>6</sup> were observed in 1964 and 1968, respectively. These phenomena of stimulated scattering are physically interesting and may be used for wavelength conversion of coherent radiation.<sup>7</sup> Owing to the competition in spectral gain, the spectrum of the stimulated light is much narrower, and in general very different from the intrinsic spectrum of the medium. Thus, the method of stimulated scattering is not useful for materials study.<sup>8</sup> This fact has long been recognized, and in the case of Raman spectroscopy, the technique of two-laser-beam CARS (coherent anti-Stokes Raman spectroscopy) was introduced<sup>9</sup> in 1974. Soon after, other types of coherent Raman spectroscopy,<sup>10</sup> SRGS (stimulated Raman gain spectroscopy), IRS (inverse Raman spectroscopy), and CSRS (coherent Stokes-Raman spectroscopy) have become popular. Because one-laser frequency is tunable in coherent Raman spectroscopy, the intrinsic spectrum of the medium can be obtained as the difference frequency of the two lasers is tuned across a Raman resonance of interest. With cw tunable lasers, the spectral resolution of coherent Raman spectroscopy has been improved<sup>11</sup> to better than  $10^{-4}$  cm<sup>-1</sup>, and these techniques have now been applied extensively.<sup>12</sup>

Rayleigh-Brillouin spectra also permit measurements of intrinsic parameters of the medium. These parameters include temperature, pressure, thermal conductivity, and shear and bulk viscosities. Some forms of coherent Rayleigh-Brillouin spectroscopy use two laser beams, in which one tunable laser scans in frequency across the other fixed-frequency laser. This way, a Rayleigh-Brillouin spectrum of the medium with high spectral resolution may be obtained, from which the above-mentioned pa-

rameters can be measured, and dynamical models of the medium may be tested. In this connection there has been some recent work with coherent polarization Brillouin spectroscopy<sup>13</sup> and near-zero-frequency gain spectroscopy,<sup>14</sup> both carried out in liquids. Very recently, we have performed the first stimulated Rayleigh-Brillouin gain spectroscopy in pure gases<sup>15</sup> with much higher spectral resolution. These works, in our opinion, will be modified and extended in the future to form a new type of spectroscopy, appropriately termed coherent Rayleigh-Brillouin spectroscopy. This new coherent spectroscopy will be complementary to and as useful as coherent Raman spectroscopy for materials study. Similar to Raman spectroscopy, the physical information of interest can also be obtained, in principle, from corresponding spontaneous light scattering experiments.<sup>16</sup> The advantages of coherent spectroscopy include high signal-to-noise, excellent discrimination from stray light and fluorescence backgrounds, and potential for much higher spectral resolution. These make coherent spectroscopy a more sensitive technique for probing low-frequency excitations and fluctuations with weak scatterers or with high background light.

The purpose of this paper is to give a more detailed account of stimulated Rayleigh-Brillouin gain spectroscopy than that described in a recent Letter.<sup>15</sup> In particular, we first derive the relationship between stimulated gain and spontaneous spectrum, along with a proof, which shows that our gain expression, which is derived in terms of density fluctuations, approaches the single-particle description of Rayleigh scattering in the single-particle limit when the collective effect is completely negligible. This proof, in our view, helps the understanding of the scattering process immensely. In Sec. II, comments on other types of coherent Rayleigh-Brillouin spectroscopy and their potential applications will be made. The experimental setup will be described briefly in Sec. III. The experimental results presented in Sec. IV, include comparisons of pure argon spectra to a better theoretical analysis than previously used,<sup>15</sup> pump-power dependence of Brillouin gain in pure SF<sub>6</sub>, and previously unpublished data of stimulated gain spectra of SF<sub>6</sub>-He mixtures. The paper ends with a brief conclusion in Sec. V.

## II. THEORETICAL CONSIDERATIONS

Let us picture a finite box with volume  $V$  containing  $N$  molecules. The (number) density at a point  $r$  and time  $t$  is  $\rho(\mathbf{r}, t)$  with an averaged value  $\rho_0 = N/V$  and fluctuation  $\delta\rho(\mathbf{r}, t)$ . This density fluctuation may be decomposed into its allowed normal modes with wave vectors  $\mathbf{K}_j$ , angular frequencies  $\Omega_j$ , and amplitude

$$\delta\rho(\mathbf{K}_j, t) = \int \delta\rho(\mathbf{r}, t) e^{-i\mathbf{K}_j \cdot \mathbf{r}} d^3r, \quad (1)$$

as

$$\delta\rho(\mathbf{r}, t) = \frac{1}{V} \sum_j \delta\rho(\mathbf{K}_j) e^{-i\Omega_j t} e^{i\mathbf{K}_j \cdot \mathbf{r}}, \quad (2)$$

where

$$\delta\rho(\mathbf{K}_j, t) = \delta\rho(\mathbf{K}_j) e^{-i\Omega_j t}.$$

For a given  $\mathbf{K}$ , the autocorrelation function of  $\delta\rho(\mathbf{K})$  may be expressed in terms of its power spectral density  $\mathcal{S}(\mathbf{K}, \Omega)$  as

$$\langle \delta\rho(\mathbf{K}, t) \delta\rho^*(\mathbf{K}, 0) \rangle = \int e^{-i\Omega t} \mathcal{S}(\mathbf{K}, \Omega) d\Omega, \quad (3)$$

where  $\langle \rangle$  denotes an ensemble average. The normalized spectral density<sup>17</sup> per molecule,

$$S(\mathbf{K}, \Omega) = \frac{1}{N} \mathcal{S}(\mathbf{K}, \Omega), \quad (4)$$

is the space-time Fourier transform of the well-known van Hove's space-time correlation function<sup>18</sup>  $G(\mathbf{r}, t)$ . The excited density fluctuation scatters light spontaneously, which may be detected in a selected direction along  $\mathbf{k}_s$ . The scattered intensity as a function of  $\omega_s = \omega_l - \Omega$  is proportional to  $S(\mathbf{K}, \Omega)$  where  $\mathbf{K} = \mathbf{k}_l - \mathbf{k}_s$ . The knowledge of the spectral density  $S(\mathbf{K}, \Omega)$  reveals the dynamics of the density fluctuation.

In a light scattering experiment, the density fluctuation is excited by a pump field with amplitude  $E_l$ , wavevector  $\mathbf{k}_l$ , and frequency  $\omega_l$ . In coherent gain spectroscopy, on the other hand, the excited density fluctuation is probed by a probe field with  $\mathbf{E}_s$ ,  $\mathbf{k}_s$ , and  $\omega_s$ . The pump and probe fields select a normal mode with  $(\mathbf{K}, \Omega)$  for investigation via conservation of momentum,  $\mathbf{k}_l - \mathbf{k}_s = \mathbf{K}$  and energy  $\omega_l - \omega_s = \Omega$ . In this manner, the intensity of the probe beam is modified (stimulated gain or loss) by the density fluctuation  $\delta\rho(\mathbf{K}) e^{-i\Omega t}$ . To derive an expression for the stimulated gain, and to relate it to the spectral density function, we consider the effect of pump and probe field,

$$E(\mathbf{r}, t) = \frac{1}{2} [(E_l e^{i(\mathbf{k}_l \cdot \mathbf{r} - \omega_l t)} + E_s e^{i(\mathbf{k}_s \cdot \mathbf{r} - \omega_s t)} + \text{c.c.})] \quad (5)$$

on the molecules. These fields induce a density change which in turn modifies the permittivity of the medium. As a result, the molecules gain potential energy from the fields<sup>19</sup>

$$\Delta W = \frac{1}{2} \int \left[ \frac{\partial \epsilon}{\partial \rho} \right]_0 \delta\rho(\mathbf{r}, t) [E(\mathbf{r}, t)]^2 d^3r. \quad (6)$$

Substituting Eqs. (2) and (5) into (6), and neglecting all oscillatory terms, we obtain the energy of the excited normal mode as

$$\Delta W = \frac{1}{4} \left[ \frac{\partial \epsilon}{\partial \rho} \right]_0 E_l^* E_s \delta\rho(\mathbf{K}). \quad (7)$$

Equation (7) suggests the existence of a force

$$f_{\mathbf{K}} = - \frac{d(\Delta W)}{d(\delta\rho)} = - \frac{1}{4} \left[ \frac{\partial \epsilon}{\partial \rho} \right]_0 E_l^* E_s, \quad (8)$$

which induces the density fluctuation

$$\delta\rho(\mathbf{K}) = R(\Omega) f_{\mathbf{K}}, \quad (9)$$

where  $R(\Omega)$  is the response function<sup>20</sup> depending on the dynamics of the medium. The amplitude of the associated third-order polarization can then be written,<sup>19</sup> for one normal mode, as

$$\begin{aligned} P^{(3)} &= \left[ \frac{\partial \epsilon}{\partial \rho} \right]_0 \delta\rho(\mathbf{r}) E_l \\ &= \left[ \frac{\partial \epsilon}{\partial \rho} \right]_0 \frac{\delta\rho(\mathbf{K})}{V} E_l \\ &= - \frac{1}{4V} \left[ \frac{\partial \epsilon}{\partial \rho} \right]_0^2 R(\Omega) |E_l|^2 E_s \\ &\equiv \chi^{(3)}(\omega_s) |E_l|^2 E_s, \end{aligned} \quad (10)$$

where Eqs. (8) and (9) have been used. Solving Maxwell's equations in the steady state under slowly varying wave approximation, the Stokes probe field grows according to

$$\frac{dE_s}{dz} = i \frac{\omega_s}{2c\epsilon_0} \chi E_s, \quad \chi = \chi^{(3)} |E_l|^2. \quad (11)$$

Equations (10) and (11) can be solved to yield the gain coefficient  $g \equiv (1/I)(\partial I / \partial z)$  for the probe (Stokes) beam,

$$g(\omega_s) = - [2\omega_s / (c\epsilon_0)^2] (\text{Im} \chi^{(3)}) I_l \quad (12a)$$

$$= - \frac{2\omega_s}{(c\epsilon_0)^2} \left[ - \frac{1}{4V} \left[ \frac{\partial \epsilon}{\partial \rho} \right]_0^2 \text{Im} R(\Omega) \right] I_l, \quad (12b)$$

where  $I_l$  is the intensity of the pump beam. The imaginary part of the response function  $R(\Omega)$  in Eq. (12) may be related to the spectral density function by the fluctuation dissipation theorem,<sup>20</sup> as

$$\frac{k_B T}{\pi \Omega} \text{Im} R(\Omega) = \mathcal{S}(\mathbf{K}, \Omega) = NS(\mathbf{K}, \Omega). \quad (13)$$

We note that  $(\partial \epsilon / \partial \rho)_0$  in Eq. (12) is the molecular polarizability  $\alpha$ ; it can be related to the Rayleigh cross section by electromagnetic theory<sup>19</sup> as

$$\left[ \frac{\partial \epsilon}{\partial \rho} \right]_0^2 = \alpha^2 = \frac{(4\pi\epsilon_0)^2}{k_s^4} \frac{d\sigma}{d\Omega}, \quad (14)$$

where  $d\sigma/d\Omega$  is the differential Rayleigh cross section, defined on the basis of power with the polarization of the stimulated radiation parallel to the pump field  $\mathbf{E}_l$ . Substituting Eqs. (13) and (14) into Eq. (12), we have the stimulated Raman gain coefficient

$$g(\omega_s) = \frac{8\pi^3 c^2}{\omega_s^3} \frac{\rho_0}{k_B T} [\Omega S(\mathbf{K}, \Omega)] \frac{d\sigma}{d\Omega} I_l. \quad (15)$$

This expression is identical to Eq. (1b) in our previous Letter<sup>15</sup> where  $I_l$  was unfortunately left out.

The shape of the spectral density function  $S(\mathbf{K}, \Omega)$  depends very much on the state of the medium which may be classified by the dimensionless parameter<sup>21</sup>  $y$ . This parameter is inversely proportional to  $Kl$  where  $l$  is the mean free path of the gas in question. Different<sup>10</sup> authors have defined  $y$  differently. For Maxwell potentials,<sup>22,23</sup> two popular definitions of  $y$  are given in Table I. For hard-sphere potentials,  $y$  is sometimes defined yet differently.<sup>24</sup> For  $y \gg 1$ , the medium is said to be in the hydrodynamic regime (or condensed phase) where collective effects are important. For  $y \sim 1$ , collective effects begin to be organized and the kinetic theory of gases must be used.<sup>21</sup> In the regime where  $y \ll 1$  (Knudsen regime), the medium consists basically of independent molecules in thermal equilibrium; the spectral density assumes a simple form resulting from Doppler broadening as

$$S(\mathbf{K}, \Omega) \rightarrow \mathcal{A}(\Omega) = \frac{1}{K} \left[ \frac{m}{2\pi k_B T} \right]^{1/2} e^{-m/2k_B T \left[ \frac{\Omega}{K} \right]^2} \quad \text{as } y \rightarrow 0. \quad (16)$$

The derivation of the stimulated gain coefficient in terms of density fluctuation is necessary if collective effects in the gaseous medium are to be accounted for. The description of Rayleigh scattering as given above, is nonetheless too complicated to be appreciated physically. In the single-particle limit ( $y \rightarrow 0$ ), the stimulated Rayleigh gain coefficient should also be derivable from the well-known two-photon transition<sup>25</sup> along with the principles of energy and momentum conservation.

This simpler approach, which does not seem to be given in the literature, should advance our understanding of the subject. Using quantum-mechanical perturbation theory, both the Rayleigh cross section  $d\sigma/d\Omega$  and its associated two-photon transition rate  $1/\tau$  may be expressed in terms of electric-dipole transition matrix elements.<sup>25</sup> The stimulated transition rate for Rayleigh scattering from a single molecule at rest may be related to the differential cross section as

$$\frac{1}{\tau} = \frac{8\pi^3 c^2}{\hbar^2 \omega_s^4} \frac{d\sigma}{d\Omega} \delta(\omega_l - \omega_s) I_l I_s, \quad (17)$$

where  $\omega_l$  and  $I_l$  are the frequency and intensity of the pump beam and  $\omega_s$  and  $I_s$  are the corresponding probe-beam parameters. The Dirac delta function amounts to a statement of energy conservation. In a light scattering experiment with preselected  $\Omega = \omega_l - \omega_s$  and  $\mathbf{K} = \mathbf{k}_l - \mathbf{k}_s$ , the light beams can interact with molecules moving with an average velocity  $v$  (before and after scattering) and a momentum transfer  $m \Delta v$  along  $\mathbf{K}$  direction.

$$\mathbf{K} \cdot \mathbf{v} = \Omega, \quad m \Delta v = \hbar K. \quad (18)$$

To satisfy these conditions, the initial velocity of the molecule may either be  $v - \Delta v/2$  or  $v + \Delta v/2$ , leading to stimulated Rayleigh gain and loss, respectively. Since the probability density, expressed in terms of  $\Omega$ , of a molecule having velocity  $v = \Omega/K$  is given by Eq. (16), the  $\delta(\omega_l - \omega_s)$  function of Eq. (17) should be replaced by

$$\mathcal{A}(K(v - \frac{1}{2}\Delta v)) - \mathcal{A}(K(v + \frac{1}{2}\Delta v)),$$

where  $\mathcal{A}(\Omega)$  is defined by Eq. (16).

Since  $\Delta v$  is very small compared to  $v$ , this difference may be written as

$$-\frac{\partial \mathcal{A}(\Omega)}{\partial \Omega} K \Delta v = \frac{\hbar \Omega}{k_B T} \mathcal{A}(\Omega).$$

The stimulated gain coefficient now becomes

$$g(\omega_s) = \frac{1}{I_s} \frac{N \hbar \omega_s}{V} \frac{1}{\tau} = \frac{8\pi^3 c^2}{\omega_s^3} \frac{\rho_0}{k_B T} [\Omega \mathcal{A}(\Omega)] \frac{d\sigma}{d\Omega} I_l. \quad (19)$$

The fact that Eq. (19) may be obtained from Eq. (15), shows that the Rayleigh gain derived from density fluctuations agrees with the gain of a two-photon transition in the limit  $y \rightarrow 0$ . Like in the spontaneous case,<sup>26</sup> we have demonstrated that stimulated Rayleigh scattering can also be described by scattering in terms of either individual particles or density fluctuations.

In practice, Gaussian beams are used for experiments. In this case, the total gain of the interaction can be shown to be

$$G = \eta_c \frac{4\pi^3 c \omega_l}{\omega_s^3} \frac{\rho_0}{k_B T} [\Omega S(\mathbf{K}, \Omega)] \frac{d\sigma}{d\Omega} P_l, \quad (20)$$

where  $\eta_c$  is the crossing efficiency of the experiment and  $P_l$  is the power of the pump beam. If two Gaussian beams overlap completely,  $\eta_c = 1$ .

Equations (15) and (20) are expressions for the gain coefficient and total gain in the steady state. As such,

TABLE I. Definition of the parameter  $y$  for Maxwell potential.

Author	Definition <sup>a</sup>
Yip and Nelkin (Ref. 21)	$y = \sqrt{2/3} \frac{\rho_0 \sqrt{mk_B T}}{\eta K} = 0.89 \frac{1}{Kl}$
Tenti <i>et al.</i> (Ref. 22)	
Sugawara <i>et al.</i> (Ref. 23)	$y = \frac{\rho_0 \sqrt{mk_B T}}{\sqrt{2} \eta K} = 1.33 \frac{1}{Kl}$
Clark (Ref. 24)	

<sup>a</sup> $\rho_0$ ,  $m$ ,  $\langle v \rangle$ , and  $\eta$  are number density, mass, mean molecular speed, and shear viscosity, respectively. Note that  $l = 3/\rho_0 m \langle v \rangle$ .

they are linearly proportional to the pump intensity or power. If the interaction time is shorter than the characteristic time of the medium, the transient behavior of the interaction must be considered,<sup>19</sup> in which case the gain may be proportional to the square root of the pump intensity. Since the pulse width of our pump laser is about 5 nsec, and the Rayleigh or Brillouin linewidths are from 500 MHz to 2 GHz, we need to consider steady state only.

If in addition to the probe beam, two oppositely propagating pump beams are used, Rayleigh-Brillouin scattering can be used as process for nearly degenerate four-wave mixing experiments.<sup>27</sup> In analogy to CARS or CSRS, four-photon processes leading to coherent Rayleigh-Brillouin spectroscopy should also take place if two pump beams and one probe beam of nearly the same frequency are crossed under a three-dimensional phase-matching condition.<sup>28</sup> If the frequency of the probe beam used is very different from those of the pump beams, the four-photon interaction is usually referred to as forced Rayleigh scattering.<sup>29</sup> In this case, a coherent grating (collective motion) in the medium is first set up by the crossing of two pump beams, and then light scattered from this grating (usually at the different probe frequency) is enhanced in the momentum matched direction and may be easily detected.

### III. EXPERIMENTAL ARRANGEMENT

In principle, the pump beam and the probe beam may cross at any angle  $\theta$ , selectively exciting the normal mode with  $K \approx 2k_1 \sin(\theta/2)$ . For  $\theta$  near  $0^\circ$  or  $180^\circ$ , the two beams overlap considerably, leading to a longer interaction length and a larger stimulated gain. For our initial experiment, two mildly focused, counterpropagating beams are crossed at a small angle of  $\sim 1^\circ$  in a Brewster-windowed gas cell as shown in Fig. 1. We chose to use a counterpropagating geometry because our pump-laser linewidth is too broad to take advantage of the very narrow Rayleigh-Brillouin linewidth in the copropagating arrangement. In addition, the stray light from the pump-laser beam is weaker in the backward direction making the counterpropagating arrangement more attractive. As shown in Fig. 1, the probe beam, with wave vector  $\mathbf{k}_s$  and frequency  $\omega_s$ , is generated by a fixed-frequency, 30-mW, cw, single-mode dye laser operating at 5850 Å. The pump

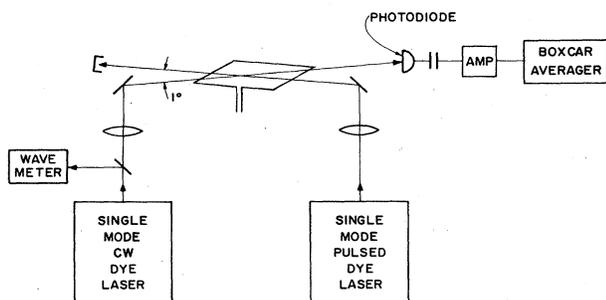


FIG. 1. Experimental setup for stimulated Rayleigh-Brillouin gain spectroscopy.

beam with  $\mathbf{k}_l, \omega_l$  is the output of a single-frequency tunable dye laser amplified to yield pulses of 5-nsec width and 1-MW peak power at 10 pulses per second. The linewidths of the pump and probe lasers are roughly 150 and 1 MHz, respectively. After traversing the interaction cell containing gas at a preset pressure and temperature (296 K), the probe-laser beam was differentially detected<sup>30</sup> by fast photodiodes 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.

### IV. RESULTS AND DISCUSSION

Using the setup shown in Fig. 1, sets of stimulated Rayleigh-Brillouin gain spectra in pure  $\text{SF}_6$ , pure Ar, and a  $\text{SF}_6$ - and He-gas mixture have been recorded. These quite different spectra may be understood in terms of dynamical behaviors of the gas systems in question.

#### A. Pure $\text{SF}_6$

In Fig. 2, the stimulated Rayleigh-Brillouin gain spectrum of  $\text{SF}_6$  at 4.1 atm, corresponding to  $\gamma = 7.2$  (hydrodynamic region) is shown. The solid curve corresponds to experimental results. 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  $v_s = 145$  m/sec as expected. Second, although the features near the unshifted frequency are much smaller than the corresponding features of Brillouin scattering,

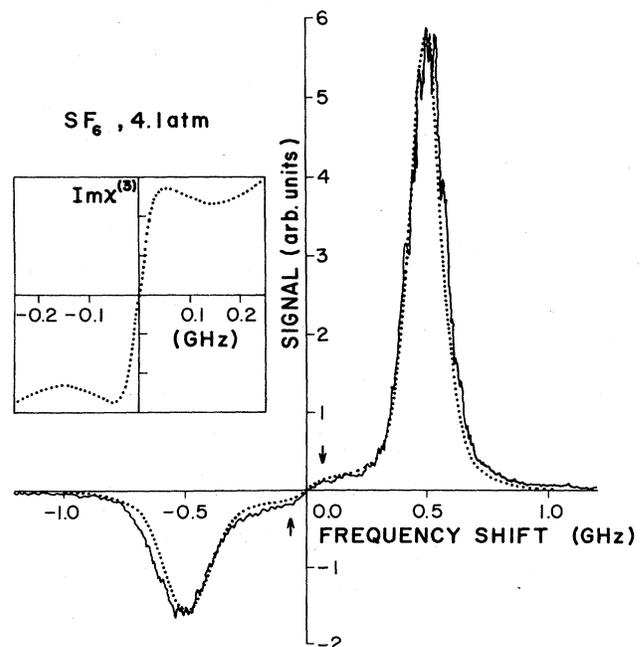


FIG. 2. Rayleigh-Brillouin gain spectrum of  $\text{SF}_6$  compared with the hydrodynamic model with  $G = 1.32$  at 4.1 atm and 296 K. The inset is a calculated curve with the pump linewidth ignored.

they are well separated in frequency and stimulated Rayleigh scattering (marked by arrows) can clearly be seen. Based on thermodynamic parameters<sup>31</sup> of SF<sub>6</sub>, a theoretical curve of  $\text{Im } \chi^{(3)}$  is shown in the inset in Fig. 2; since the pump-laser linewidth is ignored here, 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<sup>3</sup> or by laser-induced plasma.<sup>4</sup> There is no laser-induced plasma in our experiments because of the relatively low pump intensity ( $< 5 \text{ GW/cm}^2$ ) 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. Last, 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 recently in liquids<sup>13</sup> with coherent Brillouin spectroscopy. 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. 2 a theoretical spectrum normalized to

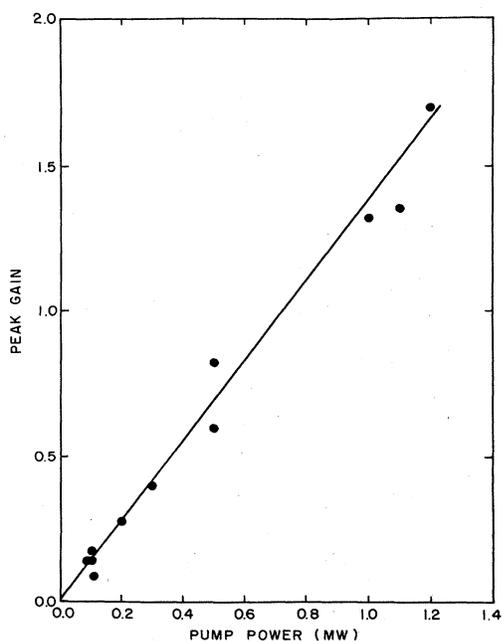


FIG. 3. Dependence of the stimulated Rayleigh-Brillouin gain on the pump power.

the loss peak. Agreement with experimental data is very good.

We have carried out a sequence of measurements with different pump powers while keeping other experimental parameters unchanged. For each pump power, the stimulated gain may be determined directly from the asymmetry of the Rayleigh-Brillouin gain spectrum. The dependence of the stimulated gain  $G$  on the pump power  $P_l$  is shown in Fig. 3. Within the experimental errors,  $G$  is linearly proportional to  $P_l$  or  $I_l$  as Eqs. (15) and (20) indicate. This linear dependence suggests, as expected, that the interaction is in the steady state.

### B. Pure Ar

The spectral density function  $S(\mathbf{K}, \Omega)$  of rare-gas atoms, Ar included, has been the subject of experimentation<sup>32,33</sup> and theoretical calculation<sup>34</sup> since the early 1960's. This problem has attracted much attention because of the advances in neutron scattering and light scattering and the possibility of theoretically solving the collision problem of atoms. The theoretical spectral density function may be calculated by solving the linearized Boltzman equation containing the collision integral which depends on a prescribed potential characteristic of the gas system. The linearized Boltzman equation was initially solved analytically, but approximately by Yip and Nelkin.<sup>21</sup> Although providing physical insights for the dynamics of gases, their model disagrees with measured spectra<sup>33</sup> for the transition region of  $y \sim 1$ . The linearized Boltzman equation was later solved numerically, but nearly exactly for both a Maxwell potential and a hard-sphere potential by Sugawara *et al.*,<sup>23</sup> eliminating the discrepancy with the experiment.<sup>35</sup> Clark later performed more accurate light scattering experiments<sup>24</sup> and compared his results to the kinetic theory using the computer program of Sugawara *et al.* Clark showed that his room-temperature data are insensitive to various potentials, which amount to a 3–5% effect, and that the spectra, predicted by the kinetic model, are valid for all values of  $y$ . 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. 4. 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<sup>21</sup> (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 spectrum. In the transition region,  $y = 1.5$ , the hydrodynamic model (dashed curve) using the given thermodynamic parameters<sup>36</sup> still fails, although it begins to work for  $y = 2.5$ . The soundwave propagation now begins to take place and the experimental data show a minor but discernible discrepancy with the early kinetic model.<sup>21</sup> This discrepancy can be reconciled with an improved numerical procedure for solving the Boltzman equation as mentioned. With the calculated spectral density function, kindly provided to us by Clark, our experimental gain

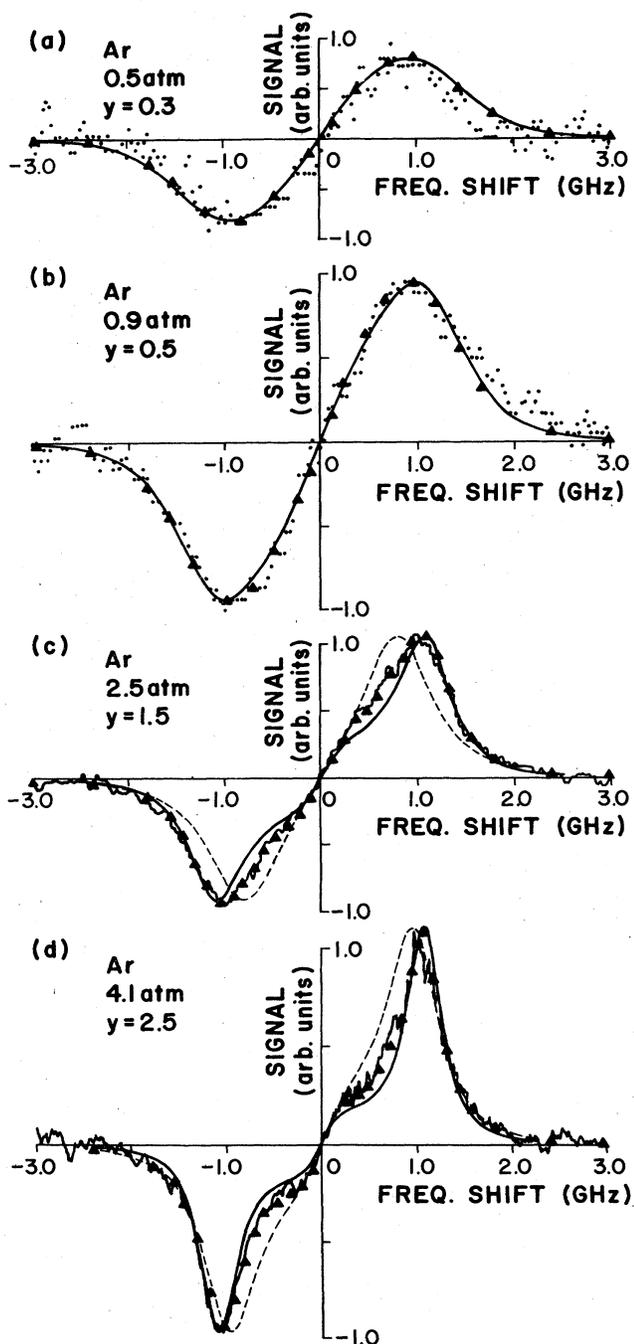


FIG. 4. Rayleigh-Brillouin gain spectra of Ar at different  $y$  values compared to the hydrodynamic theory (dashed lines) and the kinetic theory of Yip and Nelkin (smooth solid lines). Improved theoretical results by Clark are indicated by triangles. The experimental curves are shown as dots in (a) and (b) and as wiggly solid lines in (c) and (d).

spectra are in agreement with the calculated points indicated by triangles in Fig. 4. Our measured spectra are not accurate enough to provide a comparison between different collision model potentials.

In addition to the spectra of Ar and SF<sub>6</sub>, we have also

obtained and analyzed spectra of N<sub>2</sub> and CO<sub>2</sub> at different pressures. We note from Eq. (20) that for the same density and pump intensity, the stimulated Rayleigh gain is directly proportional to the spontaneous Rayleigh cross section. This is indeed the case when the gains of Ar, N<sub>2</sub>, and CO<sub>2</sub> at the same pressure (1 atom) are compared.

### C. SF<sub>6</sub>-He mixture

When a system of two gases with different atomic masses and polarizabilities is considered, in addition to the density fluctuation (entropy fluctuations and pressure fluctuations), the concentration fluctuation of the gas mixture plays an important role in its dynamic behavior. In the hydrodynamic regime, there exist both theoretical analysis<sup>37</sup> and experimental verifications with both spontaneous light scattering<sup>38</sup> and the single-beam stimulated scattering.<sup>31</sup> In the kinetic regime, there is also a theoretical treatment<sup>39</sup> but only limited experimental results dealing with dilute concentrations.<sup>40</sup> It is clear that more theoretical and experimental work would be necessary to elucidate the behavior of concentration fluctuations for different  $y$  values and different concentration ratios.

We feel that the development of stimulated Rayleigh-Brillouin gain spectroscopy will provide a sensitive and convenient tool for evaluating light scattering spectra due to concentration fluctuations under different conditions and that good experimental data will stimulate better theoretical studies. Using the same setup shown in Fig. 1, we have taken preliminary gain spectra of SF<sub>6</sub>-He mixtures. In Fig. 5, the gain spectrum of a gas mixture with 17-psi SF<sub>6</sub> and 15-psi He is shown along with a spectrum of 17-psi pure SF<sub>6</sub>. Even though the pump-laser linewidth ( $\sim 150$  MHz) is wider than desirable, the concentration fluctuation is seen to provide an added relaxation mechanism which smears out Brillouin and Rayleigh spectra and merges them into each other. This preliminary experimental result suggests that the Rayleigh-Brillouin gain spectroscopy after improvements in laser linewidth should be adequate for studying the dynamics of gas mixtures.

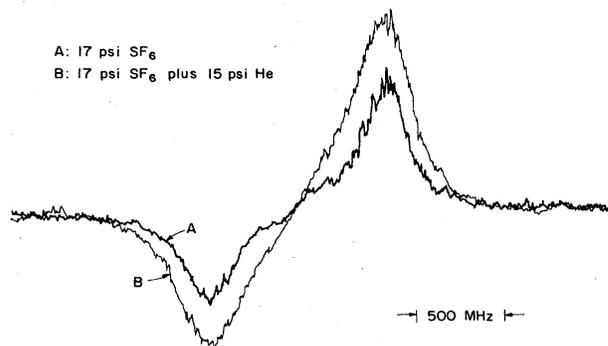


FIG. 5. Rayleigh-Brillouin gain spectra of 17-psi pure SF<sub>6</sub> (A) and a gas mixture with 17-psi SF<sub>6</sub> and 15-psi He (B).

## V. CONCLUSION

The Rayleigh-Brillouin gain spectroscopy reported here is only one branch of a larger family of coherent spectroscopies. With the 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 shifts in liquids and solid surfaces can also be studied this way without the interference of stray 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.

Rayleigh-Brillouin gain spectroscopy and the associated coherent spectroscopies with beams crossing at arbitrary angles (thus probing the different scattering wave vectors  $\mathbf{K}$ ) should be possible. For probing small  $K$  values, much higher spectral resolution is required, due to the linear  $K$  dependence of the Rayleigh-Brillouin linewidth. In spite

of small signal levels, Rayleigh-Brillouin gain spectroscopy with a cw pump laser is possible and should be pursued. The feasibility of such a cw experiment may be estimated as follows. With a 1-MW pump beam crossing at  $\theta = 179.5^\circ$  (counterpropagating) to the probe beam in 4.1-atm  $\text{SF}_6$ , we have obtained a measured gain of  $G = 2.5$  and a Brillouin linewidth of about 200 MHz. Crossing at  $5^\circ$ , the Brillouin linewidth will be narrower than 10 MHz. If a 1 W stabilized cw pump laser is used, the stimulated Brillouin gain should be  $5 \times 10^{-5}$  which is measurable with lock-in detection. If the pump and probe lasers can be stabilized to widths much narrower than 10 MHz, then the stimulated gain can be further increased by using a smaller crossing angle or by using a higher gas pressure to take advantage of the quadratic pressure dependence<sup>31</sup> of the Brillouin signal.

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