ane, as given by Bartell.[16] The agreement of theory and experiment shows that the independent-atom model of the molecular scattering process is an excellent approximation with regard to the computation of cross sections and electron spin polarization for the two cases under investigation. Our calculation also showed that except at small angles (below 30°), the double sums in (5) are relatively unimportant. This suggests that the polarization and, hence, the intensity are insensitive to the atomic separation and depend primarily on the uncorrelated atomic terms for the cases studied. We also note that if enhancement of polarization by molecular scattering over atomic scattering is to occur it must occur through the molecular interference terms. This can only happen in molecules containing a large number of heavy atoms in close proximity. It might be expected that significant deviations from the proposed theory would occur if an appreciable number of electrons in the molecule were involved in chemical binding (~50%)[10] or if the atoms in the molecule were sufficiently large and in the appropriate geometry to favor strong intramolecular multiple scattering.[17]

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STIMULATED THERMAL RAYLEIGH SCATTERING IN GASES*

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Stimulated thermal Rayleigh scattering that has been seen previously in absorbing liquids has been observed in gaseous nitrogen and carbon dioxide using small amounts of NO_2 and I_2 to produce absorption. The anti-Stokes shift of half the half-intensity width of the incident light, thresholds, gain narrowing, and critical absorption coefficients were observed.

The stimulated scattering from localized thermal fluctuations in an absorbing medium, stimulated thermal Rayleigh scattering (STRS), was first predicted by Herman and Gray.[1] In absence of other competing stimulated effects which have lower power thresholds, the rapid thermalization of absorbed energy by the molecules enhances the thermal fluctuations and can produce an intense back-scattered wave. The signature of this stimulated effect is an anti-Stokes shift approximately equal to one half of the half-intensity width of the incident

light. Experimental evidence of this scattering in liquids was reported by Rank, Cho, Foltz, and Wiggins.[2] They confirmed the anti-Stokes shift in solutions of I_2 in CCl_4 and CS_2. In addition, gain narrowing and the presence of a threshold associated with a stimulated process were observed.

An investigation in connection with STRS was later reported by Zaitsev, Kyzylasov, Starunov, and Fabelinskii.[3] They studied STRS in benzene but were unable to verify their predicted Stokes shift of 10^{-3}-10^{-4} cm^{-1}. STRS has also been investigated by Gires and Mayer.[4]

The nonlinear gains for the stimulated Brillouin- and thermal Rayleigh-scattered light in an absorbing medium in the neighborhood of the laser frequency with the frequency shift $\Delta\omega$ are given by[1]

$$g_B = \frac{\alpha_{mol}}{M}\left(\frac{n^2-1}{2}\right)\left(\frac{n^2+2}{3}\right)^3 \frac{(\Delta\omega)_B}{2v^2} k_s |E_L|^2 \times \frac{\frac{1}{2}[\Gamma_L+\Gamma_B]}{[\Delta\omega-(\Delta\omega)_B]^2+\frac{1}{4}[\Gamma_L+\Gamma_B]^2} \quad (1)$$

and

$$g_R = -\frac{\alpha_{mol}}{M}\left(\frac{n^2+2}{3}\right)^2 \frac{\beta n c \alpha}{c_p} k_s |E_L|^2 \times \frac{\Delta\omega}{[\Delta\omega]^2+\frac{1}{4}[\Gamma_L+\Gamma_B]^2}, \quad (2)$$

respectively. Here the molecular polarizability α_{mol}, the molecular mass M, the refractive index n, thermal expansion coefficient β, and specific heat at constant pressure c_p refer to the scattering medium. The Brillouin shift $(\Delta\omega)_B$, the hypersonic sound speed v, and the Brillouin and the Rayleigh linewidths Γ_B and Γ_R, respectively, can be determined from spontaneous scattering data. $|E_L|^2$ is the usual laser intensity, c is the velocity of light in vacuum, k_S is the wave number for the scattered light, and α is the absorption coefficient of the medium at the laser frequency. The minus sign for g_R implies that the maximum gain for STRS occurs at an anti-Stokes shifted frequency, the shift being equal to $\frac{1}{2}(\Gamma_L + \Gamma_R)$. If Eqs. (1) and (2) are applied to a gaseous medium, it can easily be seen that, in a given gas, g_B is approximately proportional to density of the gas through the factor n^2-1,

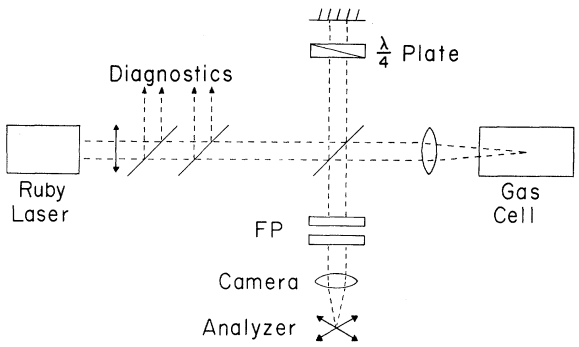

FIG. 1. Experimental arrangement for observation of stimulated thermal Rayleigh scattering in gases.

and on the other hand the gain for the stimulated thermal Rayleigh per unit absorption coefficient, g_R/α, is nearly independent of density. In order to examine the possibility of observing STRS in gases, one may compare the values of g_R in gases with the corresponding values in liquids, such as CCl_4 and CS_2 in which the effect was observed. This value can also be compared with g_B, the competing stimulated Brillouin gain. An inspection of Eq. (1) reveals that g_B for a gas at a density of 1 amagat is at least 3 orders of magnitude smaller than that for liquids. A simple calculation for g_R/α also shows that the value for CO_2 is almost identical to that for CCl_4.

These considerations lead to the interesting conclusion that the value of g_R in a gas may be large enough to produce STRS with a very small value of α compared with the value required to produce the effect in liquids because of the small Brillouin gain. It may be postulated that rapidity of the thermalization process involved would decrease considerably at small densities (<10 amagat), and such a phenomenon would prevent observation of STRS with available incident laser power.

It is the purpose of this Letter to report observation of stimulated thermal Rayleigh scattering (STRS) in gases. The experimental arrangements are similar to that used in the previous work.[2] Because of the lower index of refraction of gases compared with liquids, even more effort was required to suppress stray light in order to observe the small frequency shift predicted. In addition, the smaller size of the Brillouin shift in gases restricts the choice of interferometer spacers that may be employed. The experimental arrangements are shown in Fig. 1. The Korad laser system

produced pulses of 60 MW in a time of 10 nsec with a divergence of 6 mrad and a spectral width of 0.026 cm^{-1}. After suitable attenuation the power was monitored for each experimental condition. Attenuating the beam rather than adjusting the cryptocyanine dye or flashlamp voltage insured that the spatial, spectral, and temporal characteristics of the beam were unchanged. The beam was focused into a 5-cm high-pressure gas cell with a 10-cm focal-length lens. The temperature of the gas cell could be varied from room temperature to 150°C. A beam splitter reflected part of the back-scattered light into a Fabry-Perot interferometer and camera. Spacers of 64 and 100 mm were used. This beam splitter also reflected a portion of the incident light through a quarter-wave plate and attenuators. The light was returned to the interferometer through the quarter-wave plate so that its plane of polarization was rotated 90° with respect to the incident beam. Analyzers in the focal plane of the camera allowed the separation of the light beams so that the incident light appears in two opposite quadrants and the back-scattered light in the remaining two.

Figure 2 shows an interferogram using the 100-mm spacer obtained with CO_2 at a density of 71 amagat at 26°C to which NO_2 as an absorbing gas was added. The absorption coefficient measured using low light intensity was 0.025 cm^{-1}. The incident power was 3 MW. The measured anti-Stokes shift in Fig. 2 is 0.012 cm^{-1}, slightly less than one-half the half-intensity width of the laser light. Stimulated Brillouin scattering was not observed in pure CO_2 at these conditions. The total power of STRS observed was approximately 15% of the incident power. STRS in the gas was observed at various incident powers and it was found that the frequency shift decreased if the incident power is well above threshold. This may be attributed to saturation in the STRS process permitting an asymmetric gain due to the presence of high-intensity light at frequencies between the STRS line and the center of the laser line. A similar effect has been observed in N_2 and in some liquids. It has also been found that with lower CO_2 densities a higher input power is required to exceed the STRS threshold using the same amount of absorbing gas. Although this dependence of the STRS threshold on density cannot be explained by means of Eq. (2), it may arise from the thermalization process in the gas. Further investigation is being carried out for the quantitative determination of this dependence.

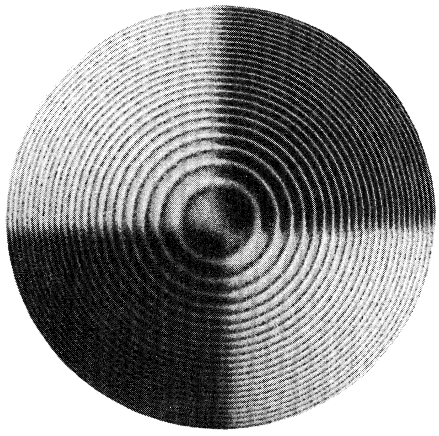

FIG. 2. Interferogram of stimulated thermal Rayleigh-scattered light from a CO_2+NO_2 gaseous mixture at the density of 71 amagat. Measured $\alpha = 0.025$ cm^{-1} at $\lambda = 6943$ Å, and interferometer spacer of 100 mm was used. The lighter sectors are due to STRS light and the darker sectors the incident laser light.

STRS has also been observed in N_2 using NO_2 and I_2 as absorbers. With NO_2 a value of critical absorption could be determined since the stimulated Brillouin component could be observed for low values of absorption. At a density of 47 amagat the threshold for STRS was 10 MW for absorption coefficients in the range between 0.007 and 0.056 cm^{-1}. No STRS was observed for lower absorption at this density for incident power up to 60 MW. However, at an absorption of 0.007 cm^{-1}, as the power was increased the stimulated Brillouin component appeared and was of intensity equal to the STRS at an input power of 30 MW. The value of α, 0.007 cm^{-1}, can then be defined as the critical absorption coefficient, α_{cr}, for which the stimulated Brillouin gain is equal to the STRS gain. The expression for α_{cr} can be derived from Eqs. (1) and (2) and is given by[1]

$$\alpha_{cr} = \left(\frac{n^2-1}{2}\right)\left(\frac{n^2+2}{3}\right)\frac{cp(\Delta\omega)_B}{\beta n v^2 c}\,\frac{\Gamma_L+\Gamma_R}{\Gamma_L+\Gamma_B}. \qquad (3)$$

In a gas, α_{cr} is approximately proportional to the density of the gas. The calculated value of α_{cr} for N_2 at 47 amagat is 0.0069 cm^{-1}, showing an excellent agreement with the observed value. It is noted that α_{cr} seems to depend on incident power as well as the density of the gas.

Table I. Gases in which stimulated thermal Rayleigh scattering was investigated. Frequency shifts, thresholds, and critical absorption coefficients α_{cr} are given.

Gases	Density (amagat)	α (cm^{-1})	Anti-Stokes shift (cm^{-1}) Calc.	Obs.	Threshold (MW)	α_{cr} (cm^{-1}) Calc.	Obs.
CO_2+NO_2	71	0.025	0.013	0.012	3.2	0.012	a
N_2+NO_2	47	0.007	0.013	0.008	10	0.0069	0.007
$He+NO_2$	47	0.050	0.013	STRS not observed		0.0018	a
N_2+I_2	47	0.060	0.013	0.004	<6	0.0069	<0.05

[a]No stimulated Brillouin scattering observed.

A similar result was obtained when iodine crystals were placed in the gas cell with N_2 gas at 47 amagat. At room temperature with input power of 30 MW, the back-scattered light corresponded to the Brillouin shift. As the temperature was raised the Brillouin gradually weakened and disappeared at 95°C, $\alpha = 0.02$ cm^{-1}. At 115°C, the I_2 absorption was great enough ($\alpha = 0.05$ cm^{-1}) to produce STRS.

The above results are summarized in Table I. The calculated values of α_{cr} were obtained from Eq. (3), assuming complete thermalization and using available material parameters.[5] It was also assumed that $(\Gamma_L+\Gamma_R)/(\Gamma_L+\Gamma_B) = 1$ in gases. STRS was sought in He with NO_2 for absorptions up to 0.056 cm^{-1}. None was seen for input powers up to 60 MW, presumably because of the small value of α_{mol}.

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KAPITZA EFFECT IN GASEOUS HELIUM*

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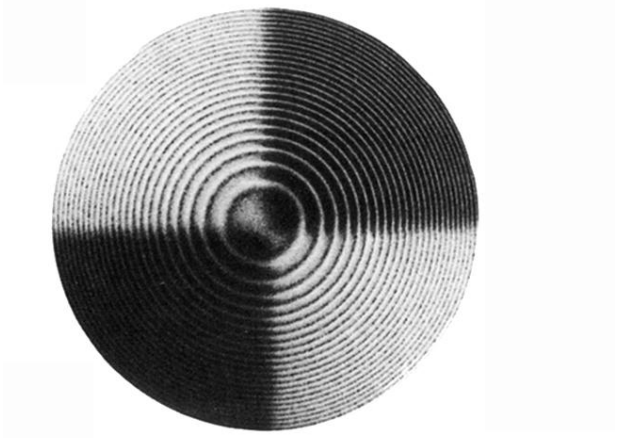
An adequate account of the thermal boundary resistance between solids and liquid helium II, first observed by Kapitza,[1] has not yet been found either by extension[2,3] of Khalatnikov's[4] discussion or by the consideration of a variety of heat-transfer mechanisms.[5] Apparently, discussion of the Kapitza effect requires a new point of departure. Here we report some measurements relevant to the suggestion that the kinetics of adsorbed helium atoms play an important part in heat transfer between helium and a solid boundary.[5] If this is so, the Kapitza resistance should not depend greatly on the density, or for that matter the phase, of the bulk helium.

We have investigated the heat transfer between helium gas and a copper surface at low temperatures. In order to be able to distinguish between surface resistances and bulk resistive effects, we constructed a conductance cell in which the width of the sample space is very small, nominally 10 μ. The cell, shown in Fig. 1, consisted of two oxygen-free, high-conductivity copper cylinders (2 cm diam) mount-



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