PHYSICAL REVIEW VOLUME 182, NUMBER 1 5 JUNE 1969

Density Dependence in Thermal Rayleigh Scattering in Gases*

D. R. Dietz,† C. W. Cho,‡ and T. A. Wiggins

Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802

(Received 7 March 1969)

Additional measurements of critical absorption coefficients and anti-Stokes shifts have been made in stimulated thermal Rayleigh scattering in CO_2 and CH_4. The threshold power for different absorption coefficients was determined as a function of density. The results are in qualitative agreement with the theoretical predictions of Gray and Herman.

INTRODUCTION

The phenomenon of stimulated thermal Rayleigh-scattering (STRS) in liquids was predicted by Herman and Gray.[1] The predictions of (1) the absorption coefficient required to produce equal gains for STRS and stimulated Brillouin scattering, (2) the anti-Stokes shift of half the incident linewidth, (3) the sharp threshold required, and (4) spectral and temporal narrowing were verified by Rank, Cho, Foltz, and Wiggins.[2] This work has been extended to additional liquids and a number of absorbers.[3]

The existence of STRS in gases was shown to exist in CO_2 and N_2 using NO_2 and I_2 as absorbing molecules.[4] The distinction between the production of this stimulated scattering in liquids and gases has been discussed by Gray[5] and Gray and Herman.[6] Introduction of the absorbing molecules would enhance the thermal fluctuation in a gas in a manner analogous to that in liquids and, consequently, STRS could be produced in the gas when the laser light is incident. In liquids it was assumed, however, that the excited molecules undergo rapid and localized thermalization. The assumption is well justified since the thermalization relaxation time is negligibly small compared with the characteristic time of the fluctuations. In gases, on the other hand, it is necessary to include the effects of diffusion of the excited molecules away from the point of excitation and those of thermalization processes with relaxation times which are now comparable to the characteristic time of the fluctuations. It has been concluded from the theoretical considerations[5,6] that the threshold power for STRS in gases should be dependent on density of the gas in addition to its dependence on the absorption coefficient.

It is the purpose of the present writing to present the experimental results on the dependence of threshold upon gas density and the amount of absorption.

EXPERIMENTAL

The apparatus is essentially the same as was used in previous work.[2-4] The laser system produced up to 0.7 J with a temporal half-width of 10 nsec. The divergence was ~7 mrad and the light was focused into the gas cell with a 10.6-cm focal-length lens. The beam was attenuated by glass plates placed before the diagnostics so that the incident power could be varied to determine the threshold power as a function of gas pressure and absorption coefficient. The absorbing molecule used in the threshold studies was NO_2. The absorption coefficient for a given amount of NO_2 in the gas cell was found not to depend strongly on the density of the other gas. The coefficient was measured using techniques previously described. The gases investigated were CO_2 and CH_4.

The spectral width of the incident light varied in the range of 0.017–0.026 cm^{-1}. Care was taken to exclude as much of the incident light as possible from the interferometer quadrants which detected the backscattered radiation. This was essential in getting a precise measure of the frequency shift and in determining the threshold as precisely as possible. Observations were made with a 101-mm Fabry-Perot interferometer.

RESULTS AND DISCUSSIONS

The results of measurements on the critical absorption coefficients and the anti-Stokes frequency shifts that have been determined to date are summarized in Table I. The expected frequency shift was calculated using a linewidth of 0.021 cm^{-1}. The precision of measurement of the observed frequency shift is ± 0.003 cm^{-1}.

The work on gases of Gray and Herman[5,6] predicts that the anti-Stokes shift is $\Gamma_L/2\sqrt{3}$, where Γ_L is the half-intensity width of the incident light. It appears that the measured shifts are somewhat larger than predicted. This is not understood since, as was noted previously,[3] the effects of stray light and saturation tend to reduce the amount of shift that is observed. The predicted increase in shift at lower densities is too small to be measured at the present time.

A series of measurements was made to study the threshold for STRS in CH_4 and CO_2 as a func-

TABLE I. Measured values of the critical absorption coefficient for several gases at the indicated densities. The measured anti-Stokes frequency shifts are also given. The expected shift is 0.006 cm^{-1}.

Gas	ρ (amagats)	Dye	$\alpha_{crit}(cm^{-1})$	$\Delta\omega_{meas}(cm^{-1})$
N_2	47	NO_2	0.007	0.008
N_2	47	I_2	0.02–0.05	0.008
CO_2	69	NO_2	0.018	0.012
CH_4	30	NO_2	•••	0.005

tion of density. This was done for each gas using two different values of absorption coefficient. The backscattering showed an abrupt appearance when the power density at the focus of the lens was raised sufficiently. The results are shown in Figs. 1 and 2. The relative values of the threshold power are repeatable to 10%. The results of Gray and Herman indicate that in gases the threshold should be inversely proportional to the density and to the absorption coefficient. This follows from the result that the gain in the stimulated process depends upon the heat deposited in the gas and upon the thermalization time which depends on the density. They predict that the gain, and hence the input power required to produce a given minimum detectable intensity of backscattered light, is given by

$$G_R(\rho) \sim \frac{1}{P_{th}} \frac{\beta\alpha}{C_p} \frac{\rho}{[1+(\omega_r' + \Gamma_R/2)/\rho\Gamma_L]^2}$$

$$= \frac{\beta\alpha}{C_p} \frac{\rho}{(1+\rho_0/\rho)^2} ,$$

where β is the coefficient of volume expansion, α is the absorption coefficient, C_p is the specific heat at constant pressure, ρ is the density, ω_r' is the diffusion rate, and Γ_R is the half-intensity width of the Rayleigh line. Only the terms which are important in this experiment are retained from the complete expression. Recognizing that the ratio β/C_p can vary, at least for some gases, over the density range investigated, we can plot

$$(\beta\alpha/C_p)(P_{th}) \text{ versus } [\rho/(1+\rho_0/\rho)^2]^{-1}$$

and expect a straight line of zero intercept, the slope of which depends upon geometrical factors associated with the focused beam as well as molecular parameters.

The results are shown in Figs. 3 and 4. The calculated values of ρ_0 are given in the figures. The fit of the data to a straight line is considered satisfactory except for the lowest densities where, although the relative measurements of the threshold are as precise as at higher density, the absolute values are not as well determined. They indicate a conservative measurement of the threshold power and indicate that a somewhat lower power input would have been sufficient. The data are not sufficiently precise to determine the value of ρ_0 to better than an order of magnitude.

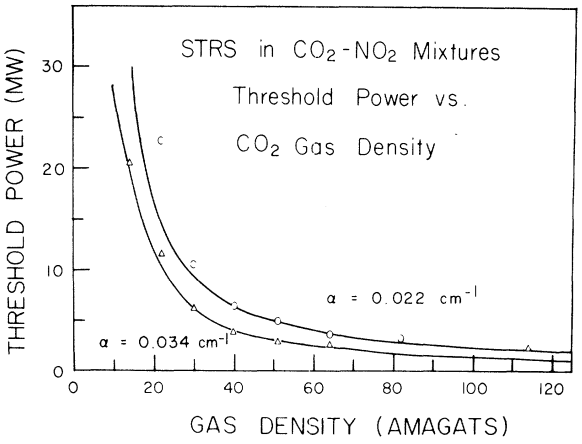

FIG. 1. Dependence of threshold power on gas density in a CO_2–NO_2 mixture for two absorption coefficients. The solid lines are drawn using the values of slope and ρ_0 from Fig. 3.

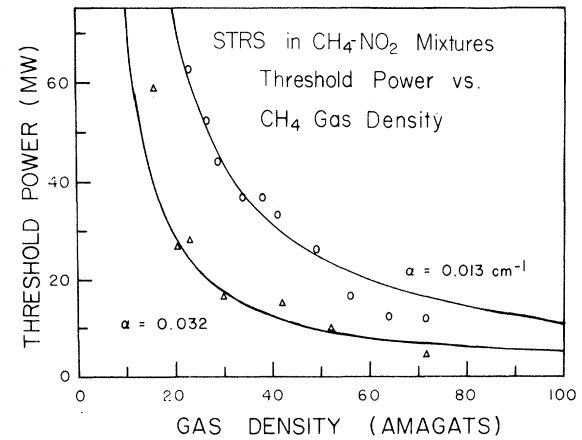

FIG. 2. Dependence of threshold power on gas density in a CH_4–NO_2 mixture for two absorption coefficients. The solid lines are drawn using the values of slope and ρ_0 from Fig. 4.

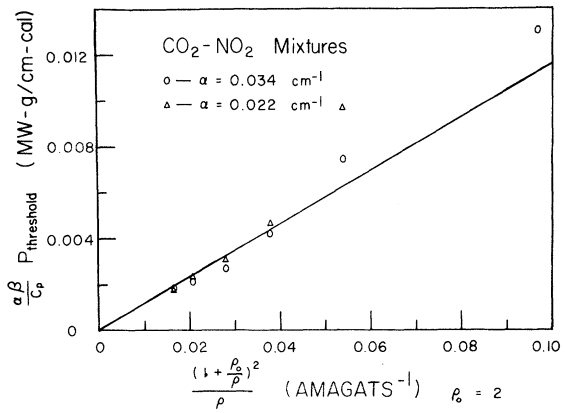

FIG. 3. Comparison of experimental results with predicted density dependence in a CO_2-NO_2 mixture for two absorption coefficients.

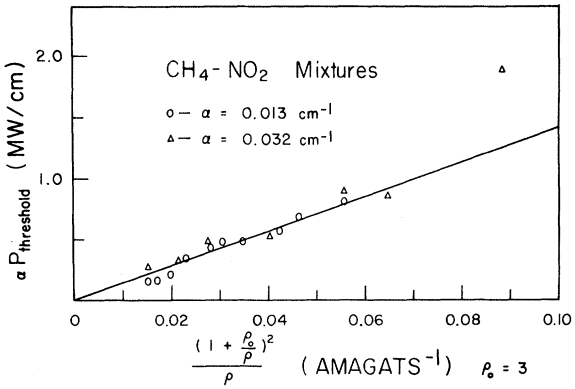

FIG. 4. Comparison of experimental results with predicted density dependence in a CH_4-NO_2 mixture for two absorption coefficients.

The critical absorption coefficient is not as easily compared with theory as it is in the case of liquids since in gases it involves the thermal relaxation time τ_1, the time for the absorbed laser light energy to be deposited as heat in the bulk gas. The value of τ_1 reflects the quenching ability of the bulk gas. From the experimental results it is possible to use the definition of α_{cr} and solve for a value of this parameter. From the work of Dietz[7] and Gray and Herman[6]

$$\tau_1 \approx 3^{3/2} \frac{n\rho}{n^2-1} \frac{cc_0^2\beta}{\omega_B C_p \Gamma_L} \alpha_{cr} \ .$$

Here c is the speed of light, c_0 is the speed of sound, and ω_B is the Brillouin frequency shift.

From the results given in Table I values of $\tau_1 = 6\times10^{-8}$ sec for CO_2 and 1.0×10^{-7} sec for N_2 when NO_2 is the absorber molecule can be calculated. There are no known values of this parameter with which to make comparison. However, they are short compared with radiative lifetimes.[8]

ACKNOWLEDGMENTS

The authors wish to thank Dr. Herman and Dr. Gray for numerous helpful discussions and Dr. A. H. Guenther, Weapons Laboratory, Kirtland Air Force Base, for the use of the laser system.

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*This work was supported by the Office of Naval Research and the National Science Foundation.

†Submitted in partial fulfillment of requirement for M.S. degree, Pennsylvania State University, December 1968.

‡On leave from Memorial University of Newfoundland, St. John's, Newfoundland, Canada.

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