Effects of Phonon Lifetime on Stimulated Optical Scattering in Gases

E. E. HAGENLOCKER, R. W. MINCK, AND W. G. RADO
Scientific Laboratory, Ford Motor Company, Dearborn, Michigan
(Received 22 August 1966)

The effects of transient conditions on stimulated Brillouin and Raman scattering have been investigated by varying the lifetime of the scattered phonon over four orders of magnitude. The Stokes gain coefficients calculated from our experimental data are as much as several orders of magnitude less than those predicted using steady-state theory, but they are in excellent quantitative agreement with those predicted by a transient analysis which considers the buildup of the phonon population with time. The dependence of the Stokes intensity on laser power for stimulated Brillouin scattering is found to be in good agreement with the transient theory. However, the theory fails to predict properly the buildup of the Stokes intensity for either rotational or vibrational Raman transitions.

I. INTRODUCTION

EXPONENTIAL growth of the Stokes radiation with distance for stimulated Brillouin (SBS) and Raman (SRS) scattering has been predicted previously.[1] These predictions are based on the assumption that the electromagnetic fields and the phonon populations have achieved their steady-state distributions in a time short compared to the interaction time. Kroll[2] indicated that a transient analysis would be necessary if the phonon lifetime exceeds the pulse duration of the laser source, and that the resulting growth rate for the Stokes radiation would be reduced. Experimental observations of the growth of the Brillouin Stokes wave with cell length for a fixed phonon lifetime agree qualitatively with the predicted transient behavior.[3]

By changing the temperature and pressure of various gases, we have been able to vary the phonon lifetime over four orders of magnitude, and thereby more directly investigate the effects of transient conditions on stimulated optical scattering. It will be shown in this article that the gain coefficients calculated from our experimental data for SBS and SRS in gases are less than those predicted using steady-state theory by as much as several orders of magnitude, but that they are in excellent quantitative agreement with those predicted by a transient analysis which considers the build up of the phonon population with time. The evaluation of the phonon lifetimes include such details as the effects of thermodynamic properties on the acoustic phonons for SBS, and of Doppler broadening, motional narrowing, and pressure broadening on the lifetime of the optical phonons for SRS.

II. TRANSIENT CONDITIONS

To facilitate the interpretation of our data, Kroll's transient analysis[2] for SBS is repeated here with minor modifications which permit its extension to SRS. Both scattering processes will be described classically in terms of a wave formalism because the stimulated photon and phonon populations per mode are large. It is assumed that the waves describing the phonon populations satisfy the dispersion relationships appropriate to the acoustical and optical branches,[4] for SBS and SRS, respectively.

The transient rates of amplitude growth of the Stokes and acoustic waves for SBS (Stokes and optical phonon for SRS) are formulated assuming that the plane waves, propagating collinearly along the z axis, are stimulated by an undepleted plane-polarized monochromatic laser beam applied to the material at $t=0$. The linearized coupled growth equations for the wave amplitudes are obtained by assuming that the waves satisfy their normal dispersion relationships and by retaining only the first derivatives of their amplitudes.

$$\frac{\partial F(z,t)}{\partial t}+\frac{1}{2\tau}F(z,t)-v_1\frac{\partial F(z,t)}{\partial z}=\chi_1 G(z,t), \quad (1)$$

$$\frac{1}{v_2}\frac{\partial G(z,t)}{\partial t}+\frac{\partial G(z,t)}{\partial z}=\chi_2 F(z,t), \quad (2)$$

where $F(z,t)$, $G(z,t)$ are the slowly varying amplitudes of the acoustic and Stokes waves; v_1, v_2 are their velocities; positive z is taken as the direction of increasing Stokes amplitude; τ is the phonon lifetime; and χ_1, χ_2 are the coupling coefficients related to the laser power and the Pockels photoelastic constant[5] for SBS (the Raman polarizability[6] for SRS). In this article it will be assumed that the spatial rate of growth of the Stokes wave is much greater than its temporal rate; consequently, the term $[(1/v_2)(\partial G/\partial t)]$ in Eq. (2) will be neglected.[7]

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[4] C. Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, Inc., New York, 1957), 2nd ed., p. 111.
[5] See, for example, *American Institute of Physics Handbook* (McGraw-Hill Book Company, Inc., New York, 1957), 2nd ed., pp. 6–188 ff.
[6] H. W. Schrötter, H. J. Bernstein, J. Mol. Spectr. **7**, 464 (1961); J. P. Jesson, H. W. Thompson, Proc. Roy. Soc. (London) **A268**, 68 (1962).
[7] The term $[(1/v_2)(\partial G/\partial t)]$ cannot be neglected if the interaction length is comparable to the distance travelled by the Stokes wave during the interaction time.

If steady-state conditions exist for the phonon wave ($\partial F/\partial t=0$) and if the term ($v_1\partial F/\partial z$) can also be neglected,[8] then

$$\frac{G(z,t)}{G(0,t)}=\exp[2\tau\chi_1\chi_2 z]\equiv\exp[\tfrac{1}{2}gI_L z], \quad (3)$$

where g is the steady-state *intensity* gain coefficient (gain coefficient refers to the exponential factor normalized per unit laser intensity (I_L) per unit length). From Eq. (3) it is evident that the product $\chi_1\chi_2=gI_L/4\tau$.

A good approximation to the response under transient conditions can be obtained by retaining $\partial F/\partial t$ and neglecting the terms $[(1/v_2)(\partial G/\partial t)]$ and $(v_1\partial F/\partial z)$ in Eqs. (1) and (2). The resulting transient Stokes gain, $G(z,t)/G(0,t)$, is initially unity and grows with time to its steady-state value If $(gI_L lt/\tau)$ is much greater than unity, the time dependence of the transient Stokes gain for an interaction length (l) is approximately

$$\frac{G(l,t)}{G(0,t)}=\exp\left[\left(\frac{gI_L lt}{\tau}\right)^{1/2}-\frac{t}{2\tau}\right]. \quad (4)$$

Equation (4) is valid for times $t\leq gI_L l\tau$, after which the steady-state solution in Eq. (3) applies.

III. PHONON LIFETIME AND STEADY-STATE INTENSITY GAIN COEFFICIENT

For stimulated Brillouin scattering, the steady-state *intensity* gain coefficient (g_B) can be written (in esu units) as[9]

$$g_B=\frac{k_s[\rho(\partial\epsilon/\partial\rho)_s]^2\omega_B\tau_B}{2\epsilon_s cn[\rho(\partial P/\partial\rho)_s]}\times10^7\ (\mathrm{cm/W}), \quad (5)$$

where $(\partial\epsilon/\partial\rho)_s$ is the change in permittivity due to adiabatic density fluctuations, $\rho(\partial P/\partial\rho)_s$ is the adiabatic bulk modulus, k_s is the wave number of the Stokes wave, and ω_B and τ_B are the frequency and decay time (phonon lifetime) of the acoustic wave. Using the classical Navier-Stokes theory for the propagation of sound in a continuum, it can be shown that[9,10]

$$\tau_B^{-1}=\eta\left[V+\frac{\gamma-1}{P_r}\right]\frac{k_B^2}{\rho}, \quad (6)$$

where η is the shear viscosity, V is the viscosity number, γ is the ratio of specific heats, P_r is the Prandtl number, and k_B is the wave number of the acoustic wave. The dependence of g_B and τ_B on temperature and pressure can be calculated from thermodynamic data available in the literature[11] and is plotted for N_2 in Figs. 1 and 2.

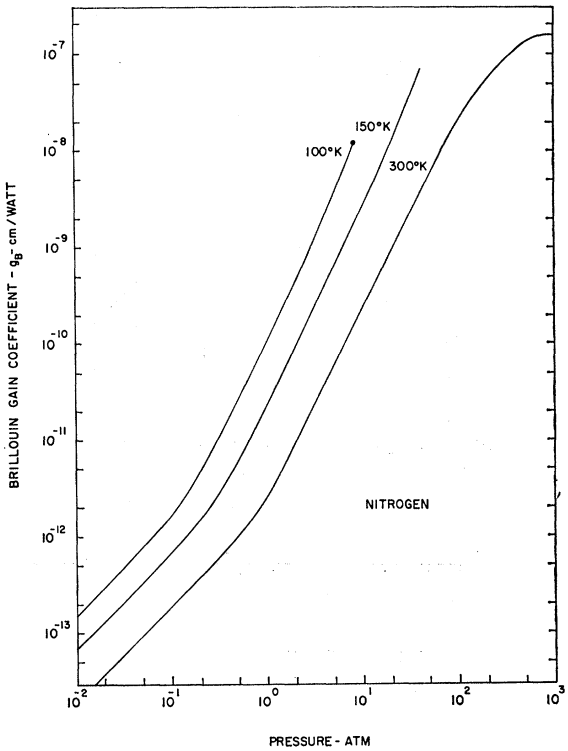

FIG. 1. Steady-state intensity gain coefficient for Brillouin scattering in nitrogen stimulated in the backward direction by a ruby laser.

For stimulated Raman scattering, g_R can be written as[12]

$$g_R=\frac{k_s[8\pi^2 L\Delta N\alpha^2]\tau_R}{\epsilon_s cnh}\times10^7\ (\mathrm{cm/W}), \quad (7)$$

where ΔN is the population difference between levels, α is the Raman polarizability, L is the local-field correction factor, and τ_R is the Raman phonon lifetime. The evaluation of τ_R for SRS is complicated since it is determined simultaneously by the perturbing influence of neighboring particles and by the thermal motion of the emitting atom itself. Using an analysis similar to that of Galatry,[13] the following expression for τ_R is obtained:

$$\tau_R=\frac{e^b}{\beta}\int_0^1 u^{-(1-a/\beta)}e^{-bu}du, \quad (8)$$

where $a=\alpha P+\beta b$, $b=(D/\beta)(2\pi/\lambda')^2$, $\beta=(kT/mD)$, D is the diffusion constant, λ' is the wavelength corresponding to the Raman transition, and α is the experimental pressure broadening coefficient in $\mathrm{cm^{-1}/atm}$ The solutions obtained transform smoothly from the low pressure

[8] In order to neglect $(v_1\partial F/\partial z)$ in Eq. (1), the gain for the Stokes wave, $\exp[gI_L v_1\tau]$, in the distance travelled $(v_1\tau)$ by a phonon during its lifetime τ has to be small, such that $(gI_L v_1\tau)$ is much less than unity. This condition is satisfied in all our SBS experiments as well as in SRS for which the optical phonon is assumed not to propagate.

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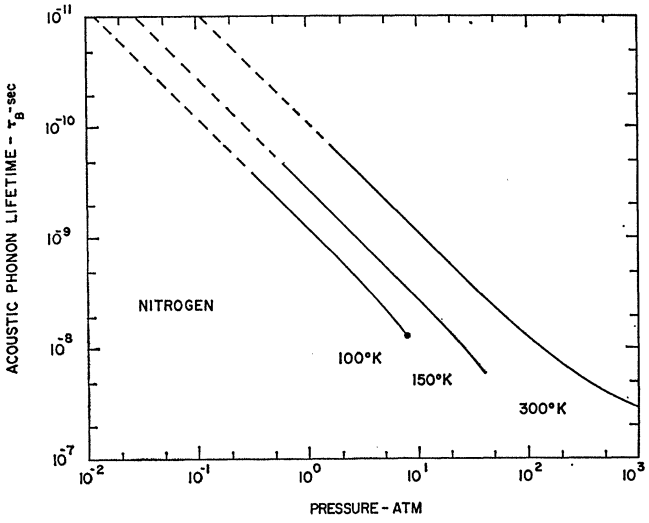

FIG. 2. Acoustic phonon lifetime for Brillouin scattering in nitrogen stimulated in the backward direction by a ruby laser. The dashed lines indicate that the acoustic behavior is uncertain at pressures for which the collision mean free path is comparable to the acoustic wavelength. (See Refs. 9 and 10.)

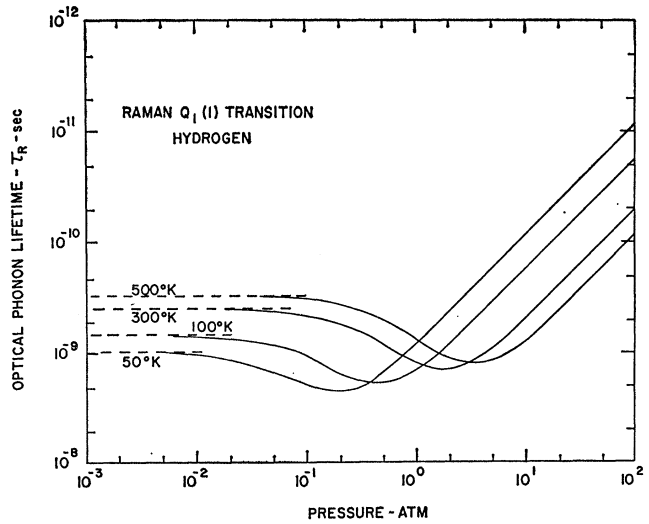

FIG. 3. Optical phonon lifetime for the Raman vibrational transition $Q_1(1)$ in hydrogen stimulated in the forward direction. The phonon lifetime in the low-pressure (or Doppler) limit is an equivalent Lorentzian lifetime calculated from Eq. (8).

Doppler limit through a motionally narrowed region[14] into the pressure broadened region at high pressures, as is shown for H_2 in Fig. 3. The pressure and temperature dependence of the gain coefficient (g_R) for one of the strong Raman transitions in hydrogen is shown in Fig. 4.

IV. ANALYSIS OF DATA

In this section the Stokes gain coefficients predicted from the transient analysis in Sec. II will be compared with those calculated from threshold data. The similarity of their dependence on the phonon lifetime suggests that the observed reduction of Stokes gain can be attributed to the transient buildup of the phonon wave.

The effects of transient conditions on the Stokes gain that one would expect to observe experimentally from a plane-wave interaction can be described by a ratio (g/g^*) where g^* is the effective *intensity* gain coefficient defined in terms of Eq. (4) by

$$\tfrac{1}{2}g^*I_L l \equiv \left(\frac{gI_L l t}{\tau}\right)^{1/2} - \frac{t}{2\tau}. \tag{9}$$

To eliminate the dependence of the gain ratio g/g^* on laser intensity and interaction length, it will be assumed that the Stokes intensity gain $\exp[g^*I_L l]$ is large and is a constant, $\exp(K)$, of the experiment. Subject to this assumption, the ratio g/g^* for an interaction time (T) is

$$g/g^* = \frac{[1+(K/T)\tau]^2}{4[(K/T)\tau]}. \tag{10}$$

Equation (10) describes a family of curves with the phonon lifetime (τ) as the independent variable and (K/T) as a parameter. These curves approach the

[14] Experimental evidence of motional narrowing is implied by the quadrupolar absorption spectrum in H_2, D. H. Rank and T. A. Wiggins, J. Chem. Phys. 39, 1348 (1963).

asymptotes $g/g^*=(K\tau/4T)$ for $(T/K)\ll\tau$ and $g/g^*\equiv 1$ for $(T/K)\geq\tau$.

A similar ratio (g/g_{exp}) can be formed where g_{exp} is the experimental gain coefficient calculated from the power levels of the Stokes and laser beams. For nonplane waves, this gain coefficient is defined to relate the output Stokes *power* to that which would result from an equivalent plane wave interaction,

$$g_{\text{exp}}P_L l/A\equiv K', \tag{11}$$

where P_L is the laser power required to observe a Stokes power gain of $\exp(K')$ and l/A is the length to cross-sectional area ratio of the equivalent interaction volume. In our threshold experiments, the laser power

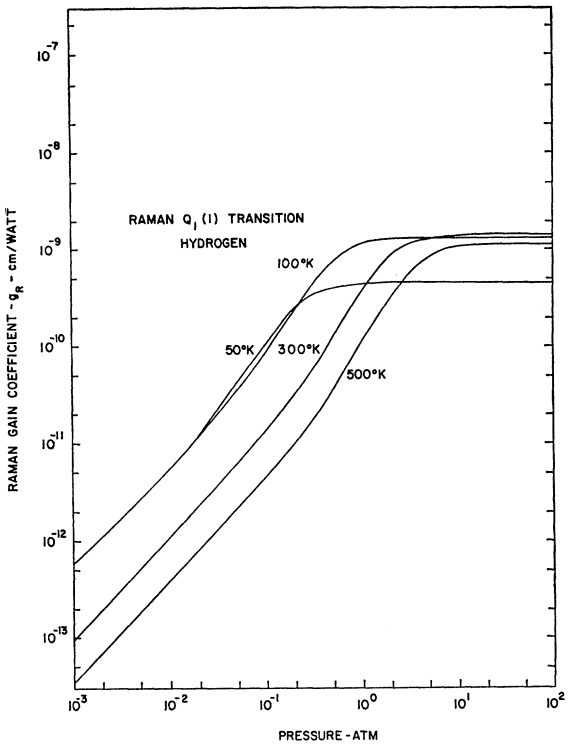

FIG. 4. Steady-state intensity gain coefficient for the Raman vibrational transition $Q_1(1)$ in hydrogen stimulated in the forward direction.

required to observe a large Stokes gain, (i.e., $K'\approx 30$), will be referred to as the threshold power (P_T).

The evaluation of l/A for finite-sized beams involves several approximations since the radial distribution of the Stokes intensity is sharply peaked because of its exponential dependence on the laser intensity. This peaking is modified by the effects of diffraction as the Stokes wave propagates through the interaction volume. Consequently, the l/A ratio is obtained by estimating the region of overlap of the Stokes and laser beams which results in the highest gain for the Stokes wave in the presence of diffraction losses. Under *steady-state* conditions, this estimate for parallel or softly focused

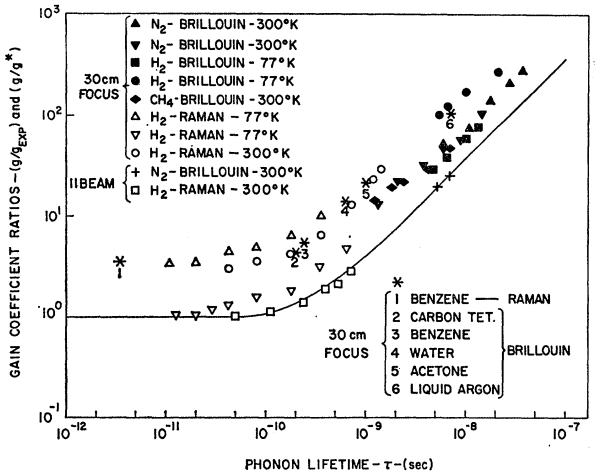

FIG. 5. Comparison of steady-state, transient, and experimental gain coefficients. The data points represent the ratio of the predicted plane wave steady-state intensity gain coefficient (g) to the nonplane wave experimental gain coefficient (g_{exp}) calculated from threshold data in a series of experiments in gases involving phonon lifetimes which vary from 10^{-7} to 10^{-11} sec. The solid curve is a plot of the ratio of the steady-state intensity gain coefficient (g) to the transient intensity gain coefficient (g^*) for plane waves as given by Eq. (10) with $K/T=1.5\times 10^{10}$ sec^{-1}. The data for SBS and SRS in liquids, taken under focused conditions to minimize the possibility of beam trapping, have been included to extend the range of phonon lifetimes. A repeateds et of data under the same conditions indicates a modification of the properties of the laser beam.

beam experiments is easily performed since the incremental Stokes gain[15] at each point has a linear dependence on the intensity of the laser at that point. Under *transient* conditions, the estimate for beams of nonuniform intensity is further complicated by the fact that at a given time the incremental Stokes gain at any point deviates from its steady state value by an amount which varies throughout the interaction volume. How-

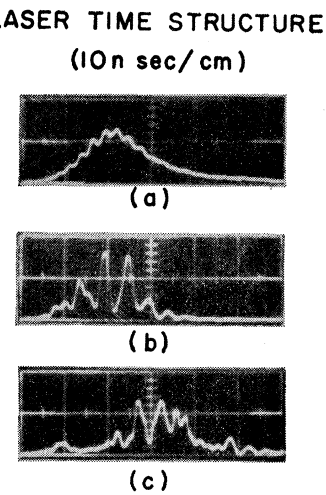

FIG. 6. Time structure of the laser pulse before (a) and after a 100 μ aperture placed (b) at the focus of a 30 cm lens, and (c) in the parallel beam one meter from the laser. Trace (a) shows the typical symmetric, almost unmodulated, power of the laser beam before the aperture. Traces (b) and (c) show the typical erratic, highly modulated, time behavior after the aperture.

[15] The incremental Stokes gain describes the rate of increase of Stokes intensity at a point and is given by $\chi_2[F(z,t)/G(z,t)]\Delta z$. For steady-state conditions, $[F(z,t)/G(z,t)]\rightarrow 2\tau\chi_1$ for all z, t, so that the incremental Stokes gain is $2\tau\chi_1\chi_2\Delta z=\frac{1}{2}gI_L\Delta z$.

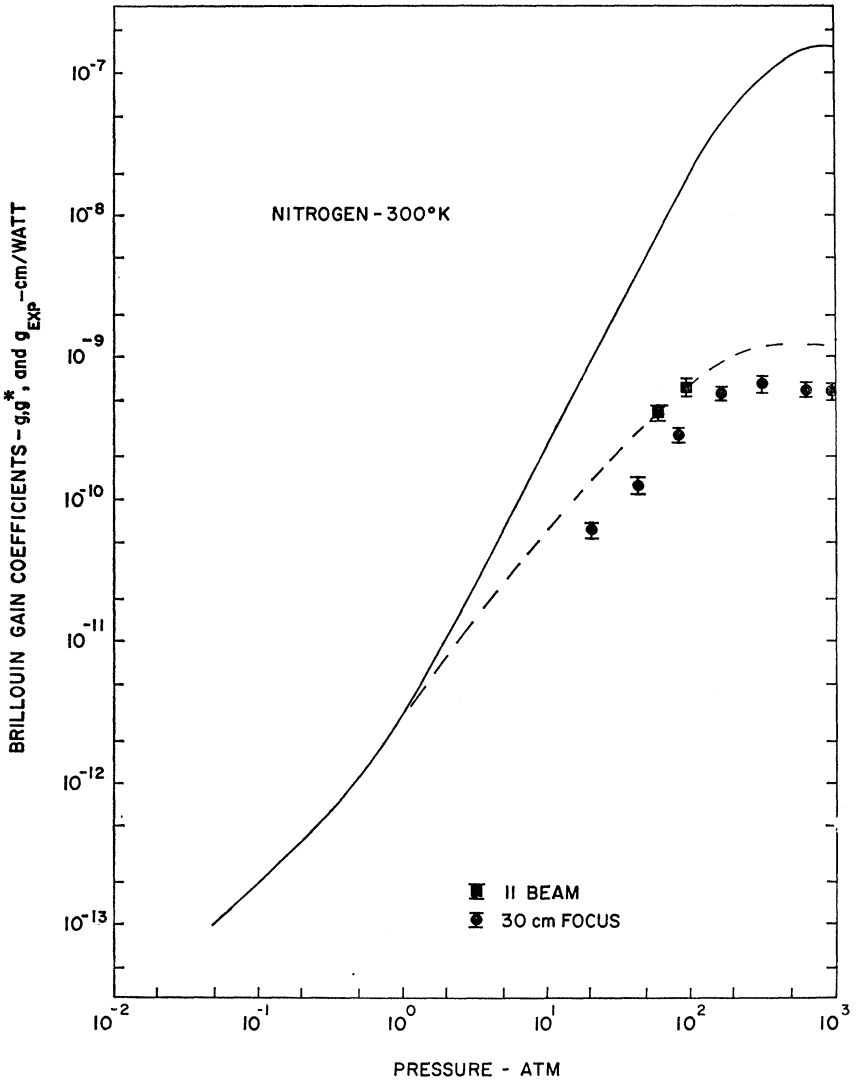

FIG. 7. Brillouin gain coefficients in nitrogen at 300°K. The solid curve is a plot of the steady-state intensity gain coefficient (g) given by Eq. (5). The dashed curve is a plot of the transient intensity gain (g^*) given by Eq. (10) with $K=30$ and $T=2\times10^{-9}$ sec. The data points were calculated from threshold data by using Eq. (11) with $K'=30$.

ever, even in our focused experiments (typically $f/60$), the change in cross-sectional area is small; consequently the l/A ratio evaluated for steady-state conditions is assumed to remain valid for transient conditions.

For a focused beam, l/A is taken equal to $5n/\lambda$, the value determined from the contour for a diffraction limited focus enclosing 15% of the total power.[16] For the parallel beam data, the l/A ratio was estimated from direct measurements of the laser-beam cross section. In contrast, if self focusing[17] and beam trapping[18] of the laser were occurring in either the parallel or focused beam experiments, the l/A ratio would be much higher than that stated above with a resulting decrease in threshold laser power. Self focusing was not expected at the laser power levels used in these experiments, nor were there any indications that self focusing had occurred.

In Fig. 5 the gain ratios (g/g_{exp}) calculated from threshold data for SBS and SRS in several gases at various temperatures and pressures are plotted assuming K' equals 30 in Eq. (11). It is apparent from Fig. 5 that for the longest phonon lifetimes shown, the experimental gain coefficient (g_{exp}) is almost three orders of magnitude less than the steady-state intensity gain coefficient (g). The slope one asymptote of the solid curve is a fit of Eq. (10) to the parallel beam data. All of the focused data fall slightly above this theoretical curve as expected for a nondiffraction limited laser beam. Since the theoretical transient curve and the threshold data in Fig. 5 are in agreement over many orders of magnitude of phonon lifetime, we attribute the observed reduction of Stokes gain to the transient buildup of the phonon population.

[16] M. Born and E. Wolf, *Principles of Optics* (Pergamon Press, Inc., New York, 1959), p. 434.
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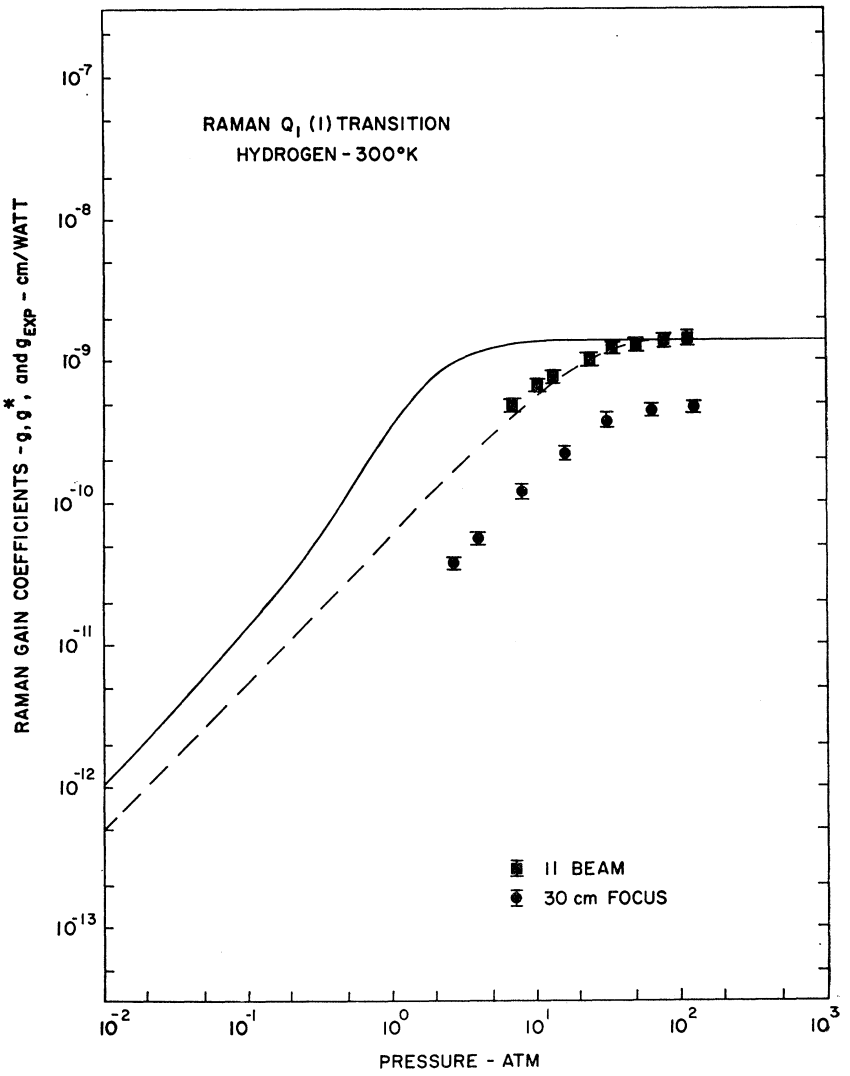


FIG. 8. Raman gain coefficients for the vibrational transition $Q_1(1)$ in hydrogen at 300°K. The solid curve is a plot of the steady-state intensity gain coefficient (g) given by Eq. (7). The dashed curve is a plot of the transient intensity gain (g^*) given by Eq. (10) with $K=30$ and $T=2\times10^{-9}$ sec. The data points were calculated from threshold data by using Eq. (11) with $K'=30$.

If the Stokes intensity gain in Eq. (10) is also taken as exp(30), the value of the parameter (K/T) which best fits the parallel beam data in Fig. 5 corresponds to an interaction time (T) of 2 nsec, much shorter than the laser pulse duration (30 nsec). The physical origin of this time has not been established, but it is felt that spatial wander of the laser beam could cause each small interaction volume to experience the peak laser intensity for a time short compared to the laser pulse duration. Figure 6 is an attempt to demonstrate this spatial variation by observing the time structure of the laser intensity before and after a 100-μ aperture placed in the laser beam. Although the total laser power produces a slightly modulated pulse, the apertured intensity shows an erratic spiking behavior.

Because of their slow velocity, the phonons created in one region cannot reach the adjoining region during the laser pulse time. Therefore each interaction volume acts independently and experiences transients appropriate to the variation of laser intensity in that region. It is not surprising, therefore, that traces (b) and (c) of Fig. 6 lead to an effective interaction time much less than the pulse duration (30 nsec). However, it is surprising that the effective interaction time should always be 2 nsec, as implied by the consistency of the various data in Fig. 5. An alternative method of presenting threshold data which better demonstrates the quantitative agreement with the transient analysis for $T=2$ nsec is shown in Fig. 7 for SBS in N_2 and Fig. 8 for SRS in H_2.

In all the experiments described above, the threshold laser power required to observe a large constant Stokes power gain was recorded as a function of the phonon lifetime. The inverse experiment can also be performed in which the phonon lifetime is held constant and the Stokes gain is recorded as a function of laser power. Under transient conditions the expected dependence of Stokes gain on laser intensity for plane waves is given

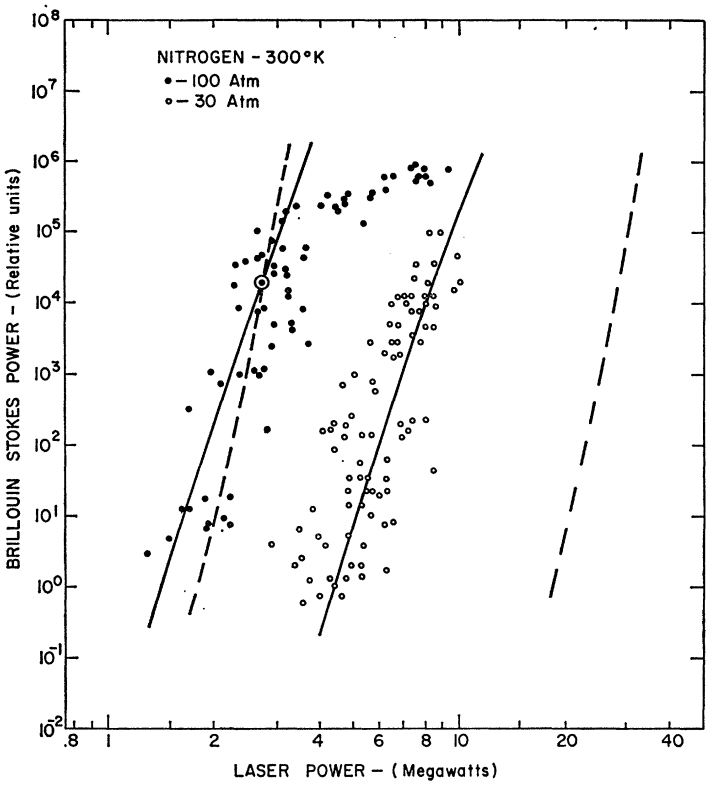


FIG. 9. The dependence of Brillouin Stokes power on incident laser power. The data points indicate the typical scatter in the actual Stokes power levels. The solid curves are plots of Eq. (12) using the appropriate values of g for the two pressures. The set of curves are fitted to the circled point in the high-pressure data to account for the nonideal properties of the focused laser and the undetermined spontaneous scattering levels. The dashed curves, also fitted to the circled point, give the dependence that would have been expected under steady-state conditions. A circularly polarized laser beam was used to improve the discrimination between the Stokes and reflected laser beams and to prevent repeated Stokes components (Ref. 9). The relative Stokes power units correspond approximately to watts.

by Eq. (4). For $gI_Ll\tau \gg t$, the relationship between two Stokes intensities $G_1(l,t)$ and $G_2(l,t)$ is approximately,

$$\frac{G_1(l,t)}{G_2(l,t)} = \exp\left(\frac{glt}{\tau}\right)^{1/2} (I_{L_1}^{1/2} - I_{L_2}^{1/2}). \qquad (12)$$

The data points in Fig. 9 for Brillouin scattering in N_2 are in good agreement with this relationship. The buildup from the spontaneous scattering level could not be observed because of the high-background level resulting from the inability to eliminate reflections of laser light from cell windows and optical components.

However, the observed dependence of the Stokes power on laser power for Raman scattering in H_2 is not in agreement with simple theory for either the rotational or vibrational transitions. This anomaly is demonstrated by Fig. 10 for the Raman $S_0(1)$ rotational transition in H_2 under both nearly steady state (7 atm) and transient (1 atm) conditions. Both of these curves indicate a dependence which varies more like $\exp(AI_L^2)$ rather than as the expected $\exp(gI_Ll)$ or $\exp[(gI_Llt/\tau)^{1/2} - (t/2\tau)]$. In Fig. 11 the buildup of the Raman $Q_1(1)$ vibrational transition under slightly transient conditions (10 atm), also appears to be like $\exp(AI_L^2)$. However, under steady-state conditions (100 atm) the buildup appears to have an abrupt onset from the spontaneous level which is even faster than $\exp(AI_L^2)$. This abrupt onset has been seen previously by Bret *et al.*[19] At present there is no explanation for this anomalous behavior.

V. CONCLUSIONS

The magnitudes of the Stokes power gain coefficients calculated from our threshold data for SBS and SRS and the buildup of Brillouin Stokes with laser power are in good absolute agreement with those predicted by a transient analysis. These experiments indicate that transient conditions must be considered even when the phonon lifetimes are two or three orders of magnitude less than the pulse duration of the laser.

Neither the steady-state nor the transient theory properly predicts the dependence of the intensity of Raman Stokes on laser intensity, implying that the agreement of our Raman threshold data with the nearly steady-state portion of the transient curve in Fig. 5 was fortuitous. This anomalous behavior raises serious questions concerning the validity of the normal Raman theory and hence our extension of the theory to transient conditions.

[19] G. Bret, F. Gires, and G. Mayer, IEEE—J. Quant. Electron. 2, 18 (1966).

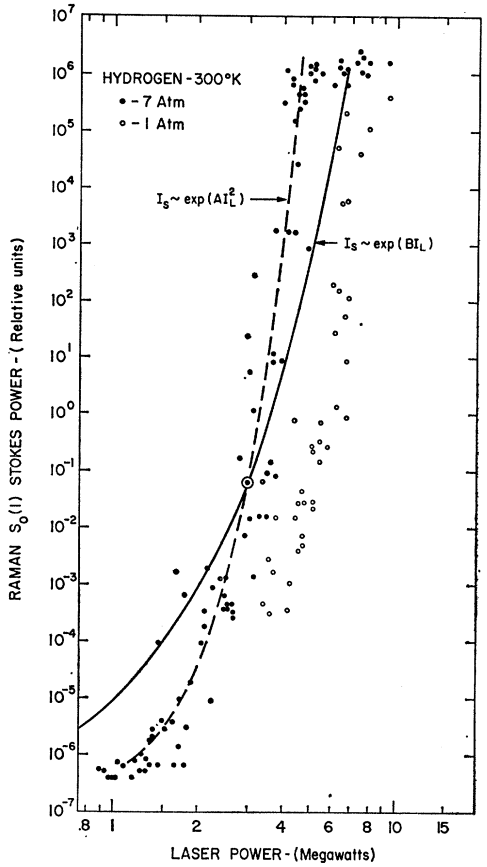


FIG. 10. The dependence of rotational Raman $S_0(1)$ Stokes power on the incident circularly polarized laser power. The data points indicate the typical scatter in the actual Stokes power levels collected with $f/25$ optics which contained the $f/30$ cone of the focused laser beam. The dashed curve, fitted to the circled point, shows a Stokes build up of $\exp(AI_L^2)$. The solid curve, also fitted to the circled point, gives the dependence that would have been expected under steady state conditions. The relative power units correspond approximately to watts.

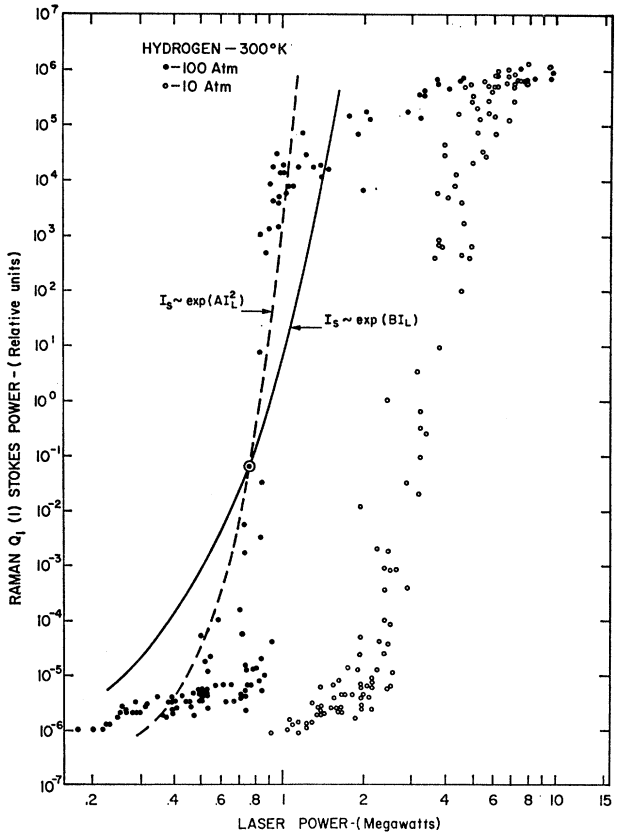
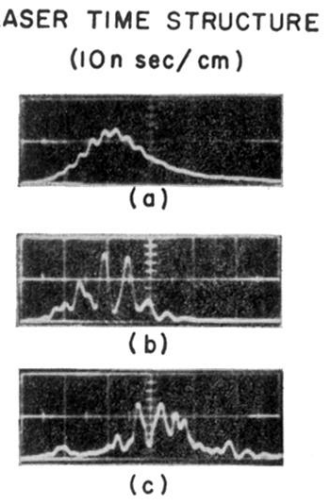


FIG. 11. The dependence of vibrational Raman $Q_1(1)$ Stokes power on the incident plane polarized laser power. The data points indicate typical scatter in the actual Stokes power levels collected with $f/25$ optics which contained the $f/30$ cone of the focused laser beam. The dashed curve, fitted to the circled point, shows a Stokes buildup of $\exp(AI_L^2)$. The solid curve, also fitted to the circled point, gives the dependence that would have been expected under steady-state conditions. The relative power units correspond approximately to watts.

When measuring large Stokes gains as was done in these experiments, several problems are encountered (i.e., the uncertainty in the initiating spontaneous level, the determination of the l/A ratio, and the possibility of oscillation due to feedback). To overcome these experimental problems, smaller Stokes gains could be measured in the second cell of a two-cell parallel beam experiment.

ACKNOWLEDGMENT

The authors are indebted to P. D. Maker for his helpful discussions.



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