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STIMULATED PURE ROTATIONAL RAMAN SCATTERING IN DEUTERIUM

R. W. Minck, E. E. Hagenlocker, and W. G. Rado
Scientific Laboratory, Ford Motor Company, Dearborn, Michigan
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This Letter reports the first observation of stimulated Raman scattering involving transitions between molecular rotational levels. In a diatomic gas, this scattering involves a $\Delta J = +2$ transition and has an intensity proportional to the square of the anisotropic part of the molecular polarizability $(\gamma_{00})^2$. The corresponding process in liquids, referred to as stimulated Rayleigh scattering,[1] arises from the anisotropic part of the Rayleigh line and produces a frequency shift corresponding to the peak of the unresolved rotational wing. In a gas the intensity-dependent change of refractive index at the laser frequency due to the dispersion associated with these rotational transitions is comparable to or greater than the index change resulting from the alignment of the anisotropic molecules.[2]

Polarization studies using a circularly polarized laser beam indicate that the observed rotational transitions are associated with $\Delta M = \pm 2$ as predicted. Threshold data show that various pure rotational and rotational-vibrational transitions compete and that different transitions dominate as the gas temperature and laser polarization are varied.

For pure rotational Raman scattering in a diatomic gas the nonlinear polarization at the Stokes frequency, $\vec{P}(\omega_S)$, can be written in complex notation as[3]

$$\vec{P}(\omega_S) \propto \tfrac{1}{2}(a+c)\vec{E}(\omega_l)[\vec{E}^*(\omega_l)\cdot\vec{E}(\omega_S)] + \tfrac{1}{2}(a-c)\vec{E}^*(\omega_l)[\vec{E}(\omega_l)\cdot\vec{E}(\omega_S)] + b\vec{E}(\omega_S)[\vec{E}^*(\omega_l)\cdot\vec{E}(\omega_l)], \quad (1)$$

where $\vec{E}(\omega_S)$ and $\vec{E}(\omega_l)$ are the electric fields at the Stokes and laser frequencies; and a, b, and c are constants related to the anisotropic polarizability γ_{00}. Because the rotational transitions involve a traceless symmetric scattering tensor, it can be shown that[4]

$$a = \tfrac{1}{3}b = -\tfrac{1}{5}c = (1/45)\gamma_{00}^2, \quad (2)$$

with

$$\gamma_{00} = \alpha_{\parallel} - \alpha_{\perp},$$

where $\alpha_{\parallel}$ and $\alpha_{\perp}$ are the polarizabilities along the molecular axis and perpendicular to it.

Figure 1 indicates that for pure rotational transitions the largest nonlinear polarization (and hence the highest gain) results from counter-rotating circularly polarized beams ($\Delta M = \pm 2$). The gain for parallel plane-polarized scattering ($\Delta M = 0$) is $\frac{2}{3}$ as large. For vibrational transitions which involve a large isotropic polarizability (scalar scattering[4]), the highest gain occurs for $\Delta M = 0$ and is essentially independent of laser polarization.

The experiment consisted of measuring the frequency and state of polarization of the Stokes

POLARIZATION	LASER	STOKES	FORM OF $P(\omega_S)$	ΔM
LINEAR	↑	↑	$P_Y(\omega_S) \propto 4/45(\gamma_{00})^2 E_Y(\omega_L) E_Y^*(\omega_L) E_Y(\omega_S)$	0
	↑	→	$P_X(\omega_S) \propto 3/45(\gamma_{00})^2 E_Y(\omega_L) E_Y^*(\omega_L) E_X(\omega_S)$	±1
CIRCULAR	↻	↻	$P_+(\omega_S) \propto 1/45(\gamma_{00})^2 E_+(\omega_L) E_+^*(\omega_L) E_+(\omega_S)$	0
	↺	↻	$P_+(\omega_S) \propto 6/45(\gamma_{00})^2 E_-(\omega_L) E_-^*(\omega_L) E_+(\omega_S)$	±2

FIG. 1. The form of the nonlinear polarization at the Stokes frequency $[P(\omega_S)]$ for pure rotational Raman scattering in a diatomic gas. The proportionality constant relating $P(\omega_S)$ to its expansion third order in electric field strength $[E(\omega)]$ is the same for all cases shown in the figure.

radiation emerging in both the forward and reverse directions from the deuterium cell, as shown schematically in Fig. 2. In order to obtain thermal equilibrium between para and ortho deuterium, a layer of activated charcoal was placed on the bottom of the 30-cm-long pressure cell before it was filled to 420 psi and sealed at room temperature. The properties of the Raman transitions were then studied at constant density as a function of gas temperature and laser polarization.

Using linear polarization, only vibrational transitions $[Q_1(J)]$ could be stimulated as is shown by part (a) of Fig. 3. The dominant transition switched from $Q_1(2)$ to $Q_1(1)$ to $Q_1(0)$ as the gas temperature was decreased from 300 to 80°K. Although the vibrational frequencies (~2990 cm^{-1}) were not measured, the frequency differences between adjacent lines were determined (4.24 and 2.11 cm^{-1}) and found to be in good agreement with measurements from spontaneous Raman spectra.[5]

When the laser was circularly polarized, two pure rotational transitions $[S_0(J)]$ were stimulated: $S_0(2)$ at 414 cm^{-1} (from room temperature to 200°K) and $S_0(0)$ at 170 cm^{-1} (below 160°K), as shown in Fig. 3(b). The Stokes radiation in the forward direction had the opposite sense of circular polarization while that propagating in the reverse direction had the same sense as the laser beam, as would be expected for a $\Delta M = \pm 2$ transition.

Strong anti-Stokes lines were observed in the forward direction, although none were expected since the nonlinear polarization at the anti-Stokes frequency is zero for a circularly polarized laser beam. However, the $\lambda/4$ plate in the incident laser beam was not perfect; consequently, the laser was elliptically polarized allowing the anti-Stokes lines to occur.

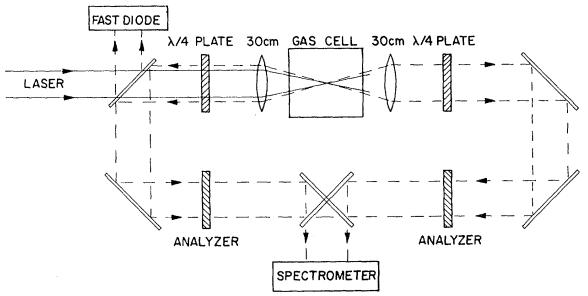

FIG. 2. Schematic of experimental agreement. The output beam of the Q-switched $\frac{1}{4} \times 2$-in.^{2} ruby exhibited a beam divergence of less than 1 mrad and produced 10 MW in two resolvable axial modes separated by about 0.01 cm^{-1}.

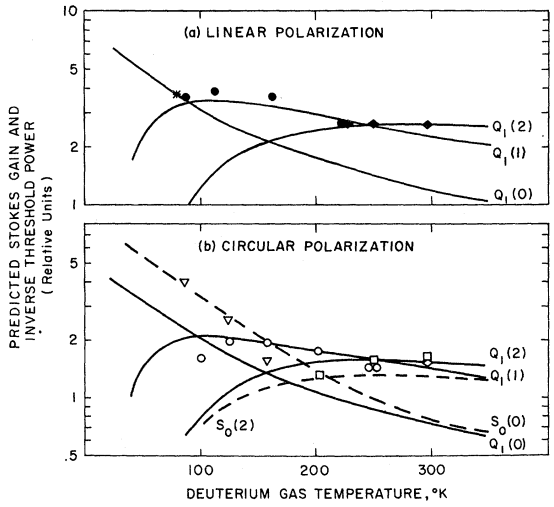

FIG. 3. Comparison of the inverse of measured threshold laser power with the predicted Stokes power gain curves (see text). In part (a) for a linearly polarized laser, only pure vibrational transitions [asterisk, $Q_1(0)$; circle, $Q_1(1)$; diamond, $Q_1(2)$] are stimulated. The data point for $Q_1(2)$ at 300°K was fitted to its gain curve and all other data were plotted relative to it. The gain curves were calculated assuming that the linewidths of the transitions were in the ratios 2:1:2. In part (b) for a circularly polarized laser beam, pure rotational transitions [inverted triangle, $S_0(0)$, and square, $S_0(2)$] are also seen in addition to $Q_1(1)$ and $Q_1(2)$. The data point for $Q_1(1)$ at 200°K was fitted to its curve and all data were plotted relative to it. The gain curves were calculated assuming that the linewidths of the rotational transitions were equal to that of $Q_1(1)$ which is half that of $Q_1(0)$ or $Q_1(2)$. Two data points at the same temperature indicate that two transitions could be stimulated simultaneously. In this case it is probable that the threshold level for the less likely transition is modified by the presence of the dominant transition.

In Fig. 3, the inverse of laser power necessary to achieve stimulated scattering is compared to the calculated Stokes gain[6] which is a function of the polarizability matrix element for the transition, the population difference between the transition levels, and the transition linewidth. Because the linewidths of the transitions have not been measured with sufficient resolution, they are considered as an adjustable parameter to fit the threshold data. Reasonably consistent agreement can be achieved by assuming that the vibrational transitions for even J had linewidths a factor of 2 larger than all other transitions.[7]

The rotational Raman transition $S_0(0)$ has also been stimulated in hydrogen (1 atm, 80°K) using a laser beam of either linear or circular polarization. Because in both D_2 and H_2 the vibrational transitions were always stimulated when the laser was plane polarized, it was not possible to measure the polarization dependence of threshold power for the rotational transitions.

During these and other studies, large pulses of Stokes radiation were observed in the reverse direction. The time duration of these pulses were instrument limited (<1 nsec) while the forward Stokes radiation followed the modulation in the laser source (~4 nsec). The ratio of peak powers front to back varied with experimental conditions, but was typically 3:1 for both the rotational and vibrational lines. Stimulated Brillouin scattering was neither expected nor observed during these experiments.[8]

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REFLECTION OF ATOMS FROM STANDING LIGHT WAVES

S. Altshuler and L. M. Frantz
TRW Systems, Redondo Beach, California

and

R. Braunstein
University of California, Los Angeles, California

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It was predicted in 1933 by Dirac and Kapitza[1] that electrons could undergo Bragg reflection from standing light waves. This phenomenon of electron-wave scattering is analogous to the ordinary scattering of light waves by periodic structures such as diffraction gratings and crystals. Recent interest[2] has been stimulated by the increased possibility of observing the effect using the intense light beams now available as a result of laser technology.

We wish to point out that Bragg reflection by standing light waves is not restricted to electrons, but should occur for all particles capable of scattering photons, including neutral atoms. The Dirac-Kapitza analysis results in the expression for the scattering probability per unit length k of the electron traversing the region occupied by the standing waves,

$$k = \frac{4\pi^3 n^2 c^4}{\omega^2 \gamma v} \frac{d\sigma(\pi)}{d\Omega}, \tag{1}$$

where n is the photon number density, ω is the angular frequency, γ is the spectral width of the electromagnetic radiation, v is the electron