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PHYSICAL REVIEW B VOLUME 2, NUMBER 6 15 SEPTEMBER 1970

Spontaneous-Raman-Scattering Efficiency and Stimulated Scattering in Silicon†

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(Received 5 March 1970)

The absolute spontaneous-Raman-scattering efficiency and linewidth of the 521-cm^{-1} optical mode of silicon have been measured at 77 K using a continuous laser (Nd in yttrium aluminum garnet) operating at 1.064 μm. The measured scattering efficiency (5.1×10^{-6}/cm sr for unpolarized forward scattering along the crystal [111] direction) and narrow linewidth yield a calculated value of the stimulated Raman gain coefficient which is considerably larger than those reported for other media, both solid and liquid. Stimulated Raman scattering in Si at 77 K has also been observed using a focused multimode *Q*-switched YAG : Nd laser. Inaccuracy in the measured stimulated gain resulted mainly from the uncertainty in the effective focal volume inside the silicon. Multiphoton absorption at the incident laser frequency has been considered and found to modify the measured stimulated gain by a significant amount. The estimated gain from the stimulated Raman effect was found to be in satisfactory agreement with that calculated from the absolute spontaneous-Raman-scattering efficiency.

I. INTRODUCTION

The near equality of the 1.064-μm laser photon energy (1.165 eV) to the indirect energy gap ($\Gamma'_{25}\rightarrow\Delta_1$) of silicon (1.17 eV at 0 K) suggests the possibility of resonant enhancement of the Raman scattering from the 521-cm^{-1} optic mode when the sample is cooled to 77 K and below. Previous investigators, using laser radiation to which silicon is opaque, have obtained values for the Raman-scattering efficiency of silicon relative to other media. Russell,[1] using reflection techniques and a 0.6328-μm laser, obtained a ratio of 35 for the scattering efficiency of silicon relative to diamond, but did not analyze his results in terms of crystal orientation and laser polarization. Parker *et al.*[2] have reported measurements of silicon's scattering intensity relative to that of germanium, using a 0.488-μm laser to which both crystals are opaque. Using general estimates for relevant parameters such as the deformation potential, Loudon[3] has suggested the scattering efficiency of homopolar semiconductors to be 10^{-6}–10^{-7}/cm sr. Most recently, Mooradian[4] has given a value of 5×10^{-6} $cm^{-1}sr^{-1}$ for the scattering efficiency of silicon using a 1.06-μm laser (Nd in yttrium aluminum garnet), and gallium arsenide as a reference medium.

We have measured with a continuous YAG: Nd laser and with due attention to crystal orientation and polarization, the spontaneous-Raman-scattering efficiency of silicon relative to liquids whose absolute scattering cross sections are known.[5] The fact that silicon at 77 K is transparent[6] to the YAG : Nd laser ($\alpha = 0.034$ cm^{-1}) and the Stokes radiation ($\alpha = 0.008$ cm^{-1}) allows an accurate measurement of the scattering efficiency. That is, the uncertain effect of surface condition on Raman-scattered intensity is not as important as in the case when the crystal strongly absorbs the incident radiation. We have also observed stimulated Raman scattering using a focused multimode *Q*-switched YAG : Nd laser. The experimental arrangements for the spontaneous and stimulated scattering will be discussed in Sec. II. In Sec. III, the stimulated Raman gain coefficient will be calculated from the spontaneous-Raman-scattering efficiency and compared with that estimated from the stimulated-Raman-scattering data.

II. EXPERIMENT

Spontaneous Scattering

A continuous YAG: Nd laser of approximately 2-W output of unpolarized radiation is weakly focused into the silicon sample, 1.3 cm in length, cooled to 77 K in a cold-finger Dewar. The silicon, of high purity (> 10 000 Ω cm), was mounted with the [111] direction parallel to the axis of laser propagation. Light is scattered from the sample at 90° to both the incident beam [111] and a crystal [100] direction, passes through an image-rotating Dove prism, and is focused on the entrance slit of a Spex 1400 double monochromator, equipped with 600-g/mm gratings blazed at 1.25 μm. Stokes radiation is detected by a Amperex XP1005 S-1 photomultiplier tube and photon-counting electronics. The relative response of the optics, monochromator, and phototube to wavelength and polarization has been calibrated with the aid of a G. E. quartz-iodine lamp of the type discussed by Stair *et al.*[7] and Polaroid HR film. Relative scattering efficiency measurements are made by interchanging silicon and media of known scattering efficiency. For this purpose carbon disulfide (CS_2) and benzene (C_6H_6) were used, contained in rectangular spectrocolorimetric cells. The directly observed scattered intensities must be corrected for instantaneous laser intensity, relative wavelength, and polarization response of the spectrometer and photomultiplier, effective solid angle of collection within the medium, and surface reflectivities. The indices of refraction at 1.064 μm assumed for these calculations were Si, $n = 3.56$; C_6H_6, $n = 1.50$; CS_2, $n = 1.62$. The vibration frequencies for Si, C_6H_6, and CS_2 were 521, 992, and 656 cm^{-1}, respectively. The instrument slits were set wide enough (10 cm^{-1}) to measure the full integrated scattering intensity. From reported[5] absolute values of the Raman-scattering cross section of CS_2 and C_6H_6 at 0.488 μm, we can put our relative measurements at 1.064 μm on an absolute basis by correcting for, in the case of the liquids, the ω^4_{Stokes} dependence of Raman intensity. Results of the Si absolute scattering efficiency are discussed in Sec. III. Our measurement of the relative scattering efficiencies of CS_2 and C_6H_6 made at 1.064 μm differed by about 5% from that of Skinner and Nilsen.[5]

Stimulated Scattering

The beam from a multimode Q-switched YAG : Nd laser is focused into the Si sample by an aspheric condensing lens of approximately 2.3-cm focal length. Owing to the high refractive index of silicon at 1.064 μm, a lens of such short focal length is required to keep sufficiently low intensity at the crystal surface, and still come to a focus within the 4-cm-long sample. Attempts to use a longer-focal-length lens or a narrow parallel beam incident on the crystal resulted in surface damage at the higher input intensities. Raman radiation scattered in the forward [111] direction is analyzed by the same system used to make our spontaneous measurements. The laser pulse intensity is monitored by an SGD-100 photodiode and varied by calibrated attenuators. The absolute laser power was calibrated with a TRG 100 Ballistic Thermopile. The stimulated gain coefficient can be obtained from a plot of relative Stokes intensity versus total incident laser power, once an estimate of effective focused spot diameter and interaction length within the sample is obtained. Figure 1 is such a plot, and is discussed in Sec. III.

III. ANALYSIS AND RESULTS

Spontaneous-Scattering Efficiency

Slightly modifying the notation of Loudon,[8] the Raman-scattering efficiency of silicon is written

$$S = A \sum_{l=1}^{3} [\hat{e}_i \cdot \overleftrightarrow{R}^l \cdot \hat{e}_s]^2 \quad . \tag{1}$$

Here the tensors $\overleftrightarrow{R}^l$ represent the three degenerate F_{2g} lattice vibrations contributing to the 521-cm^{-1} optical mode. In the coordinate system of the crystal's primitive (cubic) translation vectors, these $\overleftrightarrow{R}^l$ tensors are

$$\begin{bmatrix} & 1 & \\ 1 & & \\ & & \end{bmatrix} \begin{bmatrix} & & 1 \\ & & \\ 1 & & \end{bmatrix} \begin{bmatrix} & & \\ & & 1 \\ & 1 & \end{bmatrix} \quad .$$

The vectors $\hat{e}_i$ and $\hat{e}_s$ describe the polarization of incident and scattered radiation. It is evident that S has a strong dependence on the scattering geometry and sample crystallographic orientation. The quantity A has the same dimensions as the scattering efficiency S ($cm^{-1}sr^{-1}$) but has no angular dependence and thus can be used to totally characterize the Raman intensity.

In analogous fashion we define, for the liquid, an orthogonal coordinate system with axes parallel to the direction of incident laser propagation, 90° Raman observation, and the laboratory vertical. Then, neglecting the slight (≈ 5%) depolarization, the Raman scattering of CS_2 is described by

$$S(CS_2,\ 1.06\ \mu m) = A(CS_2,\ 1.06\ \mu m)[\hat{e}_i \cdot \overleftrightarrow{R} \cdot \hat{e}_s]^2 \ . \tag{2}$$

Here, S, A, $\hat{e}_i$, and $\hat{e}_s$ are as defined above, while $\overleftrightarrow{R}$ is simply

$$\begin{bmatrix} 1 & & \\ & 1 & \\ & & 1 \end{bmatrix} \ .$$

Our relative spontaneous measurements yield

$$\frac{A(\text{Si},\ 1.06\ \mu m)}{A(CS_2,\ 1.06\ \mu m)} = 3.0 \times 10^2 \ . \tag{3}$$

From the data of Ref. 6, we calculate (assuming a triangular line profile)

$A(CS_2,\ 0.488\ \mu m) = 2.69\times 10^{-7}/\text{cm sr}$.

Making an ω^4_{Stokes} correction for CS_2, we have

$A(CS_2,\ 1.06\ \mu m) = 1.02\times 10^{-8}/\text{cm sr}$,

which yields from Eq. (3)

$A(\text{Si},\ 1.06\ \mu m) = 3.05\times 10^{-6}/\text{cm sr}$.

Our measured value can be compared with that of Russell,[1] if we use the recent result on diamond of McQuillan *et al.*[9] and convert Russell's relative measurement on Si to absolute terms, S(Si, 0.6328 μm) $\approx 14\times 10^{-6}$ cm^{-1}sr. Since Russell did not consider the effect of laser polarization and crystal orientation, this value is only approximate and, unfortunately, no conclusions can presently be drawn about the dispersion of the Raman-scattering efficiency. Our measured value of the Raman-scattering efficiency was within the order of magnitude estimated by Loudon.[3]

Stimulated Gain Coefficient

The stimulated Raman gain coefficient can be obtained from a calculation based on the spontaneous-scattering efficiency and linewidth. The relation between the stimulated and spontaneous quantities is[10]

$$\gamma = \frac{8\pi^2 c^2}{\hbar\omega_s^3}\ \frac{S}{n_s^2(N_0+1)\Gamma} \quad . \tag{4}$$

Here $\gamma = g_s/I$, the stimulated Stokes gain per unit pump intensity. ω_s is the angular frequency of the Stokes emission. n_s is the refractive index at the Stokes wavelength (taken as 3.56). N_0 is the Bose factor (negligible at 77 K). Γ is one-half the full width at half-maximum of the Stokes line in units of angular frequency. S is the spontaneous-scattering efficiency in the forward direction, and is dependent on crystal orientation. Chandrasekharan[11] has shown that unpolarized forward Raman scattering is maximized for cubic crystals along the [111] direction. For a crystal so oriented, pumped by unpolarized light, $S = 0.83A$(Si, 1.06 μm) per plane of polarization of the Stokes radiation. Taking[12] $\Gamma = 0.8$ cm^{-1} at 77 K we calculate $\gamma = 0.19$ cm/MW. This is almost an order of magnitude larger than the gain coefficient calculated for Li_6NbO_3 by Johnston *et al.*[13] in a survey of calculated Raman gain at 1.06 μm for a variety of solids and liquids. It may also be compared with a recent calculation on diamond[9] of 6.9×10^{-3} cm/MW for stimulated scattering from the 1332-cm^{-1} mode using a 0.6943-μm pump.

Assuming no loss at the Stokes frequency, a value of the gain coefficient γ can, in principle, be obtained by fitting an equation of the form $I_{\text{Stokes}} = (\text{const})\, I e^{\gamma I l}$, to the stimulated-Raman-scattering data of Fig. 1. Here I is the effective laser intensity acting over an effective interaction length l. Note that this equation is readily transformed to

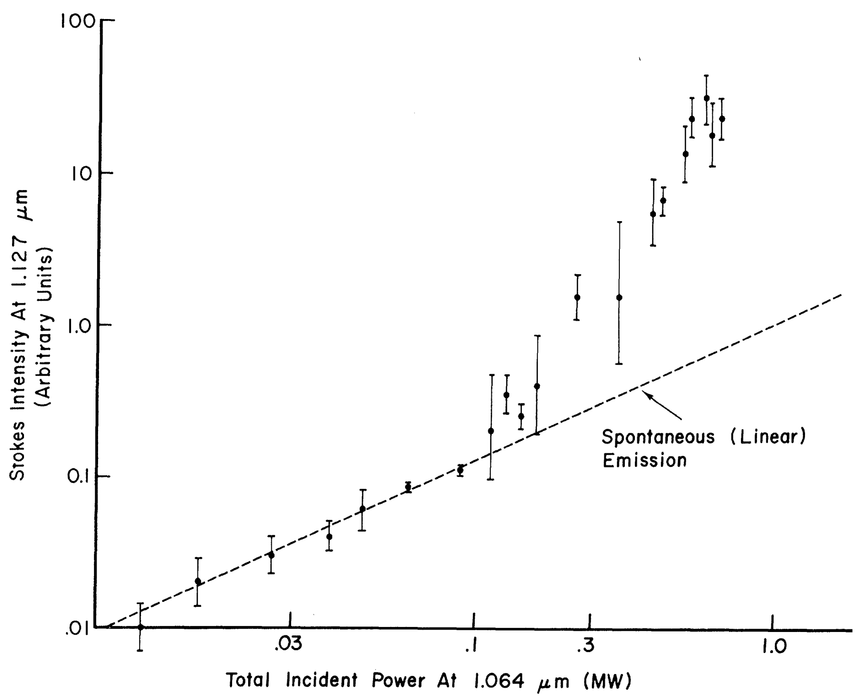

FIG. 1. Forward-scattered Stokes intensity (relative units) versus total incident power. The dotted line corresponds to spontaneous Raman scattering. Each data point represents an average of 2–5 individual pulses.

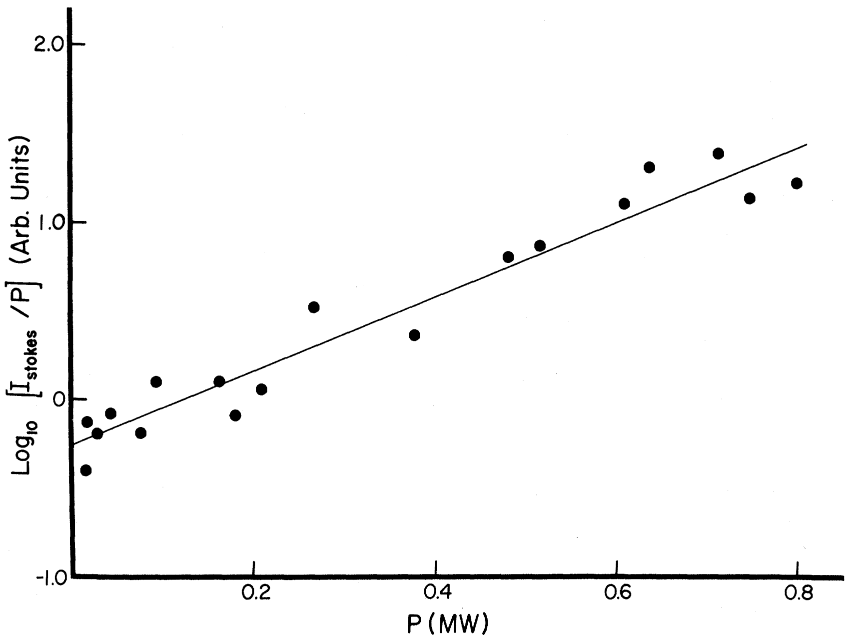

FIG. 2. Data of Fig. 1 transformed and replotted to make use of Eq. (5). A line of slope 2.1 MW^{-1} is drawn through the experimental points.

$$\log_{10}(I_{\text{Stokes}}/P) = (0.434)\gamma lP/a + \log_{10}(\text{const}). \quad (5)$$

Here I_{Stokes} is the uncalibrated Stokes intensity in arbitrary units, P is the total focused laser power, and a is the effective focused area over the interaction length l. In Fig. 2, the stimulated data previously shown in Fig. 1 are replotted in this form, together with a line of slope $2.1(\text{MW})^{-1}$. The spherical aberration resulting from focusing into a high-index medium and the inherent large aberrations of the aspheric condenser used make it difficult to estimate the effective focused area within the sample. We estimate an area, $a = 0.001$ cm^2. The solid angle subtended by the cone of focused rays in the sample is $\Omega = 0.014$ sr, giving an estimated effective interaction length of

$$l \approx (0.001 \text{ cm}^2/0.014)^{1/2} = 0.26 \text{ cm}.$$

Recognizing that the power per plane of polarization is one-half that indicated on the abscissa of Fig. 2, we obtain

$$\gamma = \frac{(2)(2.1 \text{ MW}^{-1})(0.001 \text{ cm}^2)}{(0.434)(0.26 \text{ cm})} \approx 0.04 \text{ cm/MW} .$$

The disparity of the gain coefficient estimated from the stimulated scattering and that predicted from the absolute spontaneous efficiency ($\gamma = 0.19$ cm/MW) is not disturbing considering the gross approximations made in estimating the effective focal area and interaction length.

In view of the large power densities present at the focal volume employed in this experiment (600 MW/cm^2), we must consider the effect of multiphoton absorption at the pump frequency in Si. At 77 K these are well described by a transmission equation of the form[14,15]

$$I(z) = I_0/(1 + BzI_0) . \quad (6)$$

I_0 is the laser intensity at the beginning of the interaction volume, $I(z)$ the intensity at position z within the interaction volume, and B the two-photon absorption coefficient. We substitute Eq. (6) into the differential equation describing the growth of the Stokes intensity[16] I_s,

$$\frac{dI_s(z)}{dz} = \gamma I_s(z)I(z) \quad (7)$$

and integrate Eq. (7) over the effective interaction length l,

$$I_s(l) = I_s(0)\exp[(\gamma/B)\ln(1 + BlI_0)] . \quad (8)$$

Thus the effective gain coefficient will be somewhat smaller than otherwise anticipated,

$$\gamma_{\text{eff}} = [\ln(1 + BlI_0)/BlI_0]\,\gamma . \quad (9)$$

Based on the above estimate of the interaction volume, $l = 0.26$ cm, $a = 0.001$ cm^2, I_0 reaches a maximum of 600 MW/cm^2. B is 0.01 cm/MW.[14,15] Substitution yields

$$\gamma_{\text{eff}} = 0.6\gamma .$$

This correction, albeit large, is not overly important in view of the gross approximation involved in our estimate of the interaction volume.

In the above analysis, the effect of Stokes feedback from the ends of the sample is not considered.

From Fig. 2 it is apparent that no sharp threshold of oscillation has been observed in our experiment, in contrast with the results of McQuillan *et al.*[9] in diamond, despite the large (32%) reflectivity of the uncoated silicon sample. The small spatial extent (~ 0.03 cm) of our focused interaction region and the fact that no attempt was made to polish the ends of our sample perfectly parallel suggest that misalignment losses may have considerably raised the Stokes oscillation threshold. The effect of feedback below the oscillation threshold is to increase the apparent gain of the Raman amplifier. Our observed gain, however, is smaller than that predicted on the basis of spontaneous measurements. It seems, therefore, that Stokes feedback was not a dominant influence in our experiment. Inhomogeneous spatial distribution of the incident pump beam was also neglected. An anomalously large value of γ obtained in calcite[17] has been attributed to inhomogeneity effects in the laser far-field pattern. As stated above, however, our result on γ is opposite.

Stimulated Brillouin scattering (SBS) in silicon at 1.06 μm[4] may be an important additional nonlinear loss mechanism in our experiment. For intensities up to about 20 MW/cm^2, no nonlinear loss at the laser frequency is attributable to backward SBS.[15] However, power densities as high as 600 MW/cm^2 were used in our measurements, and significant depletion of the pump radiation by backward SBS may have occurred. Such an additional nonlinear loss mechanism would still further reduce the observed Raman gain from that expected solely on the basis of spontaneous measurements and the two-photon absorption correction made above.

IV. CONCLUSIONS

We have measured the spontaneous-Raman-scattering efficiency of silicon at 1.064 μm relative to that of CS_2, which has been given in absolute units by Skinner and Nilsen.[5] Using our absolute Raman-scattering efficiency of Si and the measured linewidth, we have calculated the stimulated gain coefficient.

Stimulated Raman scattering has been observed in silicon using a Q-switched YAG : Nd laser. We believe this to be the first reported stimulated Raman effect in an opaque medium. The uncertainty of the gain coefficient obtained from our data stems mainly from the estimate of the effective focal volume and neglect of multimode effects. We have considered the effect of two-photon absorption, but neglected that of SBS. The gain coefficient calculated from the spontaneous-scattering efficiency is in satisfactory agreement with that estimated from the stimulated data.

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†Work supported in part by the Office of Naval Research.

*Alfred P. Sloan Foundation Fellow.

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