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MODE-STRUCTURE INDEPENDENCE OF STIMULATED RAMAN-SCATTERING CONVERSION EFFICIENCIES*

F. J. McClung, W. G. Wagner, and D. Weiner†

Hughes Research Laboratories, Malibu, California

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The power density needed to convert a given amount of laser energy to stimulated Raman-scattered radiation has been found to be about an order of magnitude less in nitrobenzene than theoretically expected for the case of a Raman cell external to the laser cavity.[1] It has been suggested[1,2] that the multimode character of the laser pump might be responsible for this disagreement. Bloembergen and Shen[2] have given a semiquantitative estimate of the enhancement of the Raman gain. It indicates that with a typical multimode laser a Raman gain ~4 to 8 times greater than that for a single-mode laser should be produced. It is the purpose of this communication to show that the Raman gain in nitrobenzene for a single-mode laser pump is precisely that for a multimode laser pump, and to suggest other reasons for the above-mentioned disagreement.

In this experiment, the collimated beam from our giant pulse laser was directed onto a 1.45-mm aperture 70 cm from the laser, and the portion of the beam transmitted by the aperture passed through a 10-cm cell of nitrobenzene onto a MgO diffuse reflector placed 50 cm after the cell. The power of the laser and the first Stokes line was monitored by suitably filtered fast photodetectors that sampled the diffuse reflection from the MgO. The cell was tilted at ~3° with respect to the laser beam. This arrangement is like that described in more detail by Weiner, Schwarz, and McClung,[1] except that the aperture and cell are now further from the laser and closer to the MgO. Also, another beam splitter to allow a measurement of the far-field pattern has been added.

Our laser has been mode-selected to give single-transverse and longitudinal mode behavior. The details of the mode-selection techniques will be given elsewhere.[3] The transverse mode structure was determined with the aid of a 1-m focal-length camera. The longitudinal mode structure was determined with the aid of a 2-cm spaced Fabry-Perot etalon with $\lambda/80$ flat plates of ~1% transmission. When completely mode-selected, the laser produced 2 MW of power in a beam whose divergence equaled the diffraction limit corresponding to the laser-beam diameter. When not mode-selected, the laser output was ~10 MW, the beam divergence $\sim 1.5\times 10^{-3}$ rad, and the spectral width $\sim\frac{1}{2}$ cm^{-1}. The pulse length was ~30 nsec for both cases. The results of the conversion-efficiency measurements for mode-selected and non-mode-selected lasers are shown in Table I. The experimental arrangement is the same for both cases. The relative error for the power measurements is ~5%. Measurements at other power densities for our mode-selected laser gave a conversion-efficiency curve which agreed very well with our previous curve for a non-mode-selected case.[1] The data of Table I indicate strongly that the anomalously high gain is not caused by the laser mode structure.

The theoretical gain in nitrobenzene at 20 MW/cm is 0.028 cm^{-1}. This gain is computed using a formula of Hellwarth[4] and a recently measured peak Raman-scattering cross section of 1.3 ± 0.4 cm^{-2}. This cross section agrees within the expected error with our earlier[5] and less accurate measurement of 2.3 ± 1.2 cm^{-1}. It also agrees with the recent measurement of Damen, Leite, and Porto[6] in the fol-

Table I. Conversion efficiency for mode-selected and non-mode-selected pumping of nitrobenzene.

Description of mode structure	Incident-peak power density (MW/cm^2)	Power converted to first Stokes radiation (%)
Two TEM_{00} modes of ~4:1 intensity ratio separated by ~800 Mc/sec	19	0.6
~100 transverse modes ~10-20 longitudinal modes	19.5	0.6

lowing sense. If we scale our data for benzene cross sections by the ratio of the nitrobenzene cross sections (1.3/2.3) and then compute the cross section/molecule at 6328 Å, we find a value of $0.7 \pm 0.2 \times 10^{-28}$ cm^2. Here we assume a λ^4 wavelength dependence and use the observed angular dependence for Raman scattering. This value agrees (within the stated error) with their[6] value of $0.56 \pm 0.1 \times 10^{-28}$ cm^2.

To estimate the Raman gain at 20 MW/cm^2 from our conversion-efficiency data, we assume noise is amplified during a double pass (through the cell to the laser reflectors and back through the cell) to the observed level of 10^5 W/cm^2. The dominant contribution to the noise which initiates the generation of the Raman-shifted radiation comes from the quantum-mechanical zero-point vibrations of the electromagnetic field. In a spectral interval of $d\lambda$, and within a cone of opening angle θ, centered on the axis of the laser beam, there is a zero-point power flux[7] given by $(\pi\theta^2\hbar\omega/n\lambda^3)(d\lambda/\lambda)$. Our measurements indicate that the bulk of the Stokes radiation emerges within an angle, θ, which is not greater than 0.017 rad, and that the fractional wavelength spread, $d\lambda/\lambda$, is of order 3×10^{-4}, and thus the starting power flux is estimated to be $\leq 1.2 \times 10^{-2}$ W/cm^2. Then 10^5 $W/cm^2 = 1.2 \times 10^{-2}$ $W/cm^2 \times \exp(g \times 20$ cm), so that the gain per cm $g = 0.80$ cm^{-1}. The gain is only slightly lower if calculated for multiple double passes where the feedback is supplied by diffuse reflection from the cell window and the MgO reflector, since the solid angle of acceptance imposed by our geometry is so small. Thus the observed gain is at least 25 times greater than that calculated from the cross sections.

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†Present address: Department of Electrical Engineering, University of California, Berkeley, California.

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