Figures 2 and 3 show the results for 20- and 60-eV impact energies, respectively.

The integral cross sections are summarized in Table I. A more detailed description of the results as well as cross sections at other impact energies and for other electronic transitions will be published later.

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Observation of Stimulated Anti-Stokes Raman Scattering in Inverted Atomic Iodine*

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Nonresonant stimulated anti-Stokes Raman scattering from an inverted population in atomic iodine has been observed. The population inversion was achieved by flash photolysis of trifluoromethyliodide. A gain of e^7 was measured for a laser probe pulse at the anti-Stokes wavelength passing through the inverted iodine coincident with a laser pump pulse. This result is in good agreement with theoretical predictions.

In the presence of intense electromagnetic radiation of optical frequency ω_1, atoms in an excited metastable state of energy $\hbar\omega_0$ can decay through enhanced two-photon emission[1] and anti-Stokes Raman scattering.[2] In the first process, two photons are emitted with frequencies ω_1 and $\omega_2 = \omega_0 - \omega_1$. In the second process, the incident photon is absorbed and simultaneously a photon of energy $\omega_2 = \omega_0 + \omega_1$ is emitted. If the intensity at frequency ω_2 becomes sufficiently large, these emission processes will be stimulated. Resonant stimulated anti-Stokes Raman scattering has been observed by Sorokin *et al.*[3] in potassium vapor in which a population inversion between the two 4^2P excited states was produced through absorption of laser light by atoms in the $4^2S_{1/2}$ ground state. In this Letter we report the first observation of nonresonant stimulated anti-Stokes Raman scattering (ASRS) in a medium in which a population inversion was produced by an incoherent pumping source. An inversion between the $5^2P_{3/2}$ ground state and the metastable $5^2P_{1/2}$ excited state of atomic iodine was achieved by flash photolysis of trifluoromethyliodide (CF_3I). The ASRS was then observed by measuring the gain at the anti-Stokes wavelength experienced by a laser probe pulse as it passed through the inverted iodine coincident with a 1.06-μm laser pump pulse. Since the iodine metastable energy $\hbar\omega_0 = 7604$ cm^{-1} was less than the incident photon energy $\hbar\omega_1 = 9391$ cm^{-1}, only the anti-Stokes Raman decay process could occur. The possible use of ASRS in iodine to produce frequency up-conversion of high-power lasers was recently suggested by Vinogradov and Yukov.[4]

The experimental arrangement used is shown in Fig. 1. The neodymium-doped yttrium aluminum garnet oscillator[5] was electro-optically Q switched permitting reliable time synchronization between the laser pulse and the flashlamp discharge used to invert the iodine. The oscillator produced a train of pulses each of 0.75-nsec duration with a transform-limited bandwidth of 0.020 cm^{-1}. The pulse duration was as short as possible, consistent with the limit imposed by steady-state Raman scattering theory,

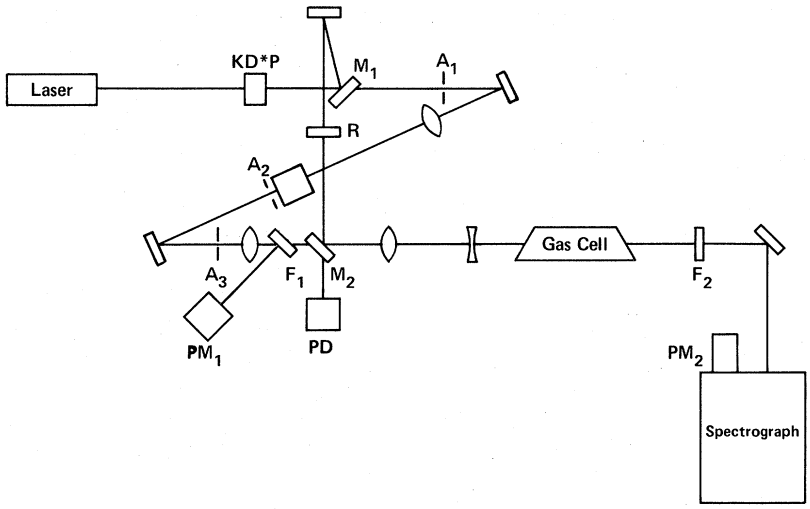

FIG. 1. Experimental arrangement used to measure anti-Stokes Raman gain in inverted iodine. A, aperture; F, filter; M, mirror; PD, photodiode; PM, photomultiplier; and R, half-wave rotator.

to minimize difficulties with optical breakdown and saturation of the Raman gain by depletion of the excited-state population. A single pulse from the train was selected, passed through a spatial filter, and then amplified by Nd:glass amplifiers to an energy of 0.25 J. After amplification, the beam intensity was 10^9 W/cm^2, and the divergence angle was 250 μrad.

This pulse passed through a deuterated potassium dihydrogen phosphate crystal in which approximately 10% of the energy was converted to the second-harmonic wavelength of 5324 Å. Mirror M_1 reflected the pulse at the fundamental wavelength and transmitted the second harmonic. The second-harmonic pulse was then used to pump a traveling-wave rhodamine B dye laser which provided the ASRS probe pulse. The dye laser consisted of a 1-cm quartz cell containing a $10^{-4}M$ solution of the dye in water. Its emission occurred in a 100-cm^{-1} bandwidth with the peak at 5850 Å. The dye cell windows were tilted to Brewster's angle with respect to the laser pulse direction so interference effects were avoided and a smooth spectral intensity distribution was produced. The apertures A_1, A_2, and A_3 were used to restrict the diameter and angular divergence of the dye laser probe pulse to equal, as nearly as possible, the diameter and divergence of the pump pulse. This restriction was necessary since only the portion of the probe pulse which overlapped with the pump pulse in the iodine could experience gain and any additional probe light reaching the detection apparatus would have reduced the signal-to-noise ratio. To avoid saturation of the Raman gain, the power of the probe pulse was reduced to about 100 W by filter F_1. The probe-pulse energy E_1 was measured by the photomultiplier PM_1. A half-wave plate was used to make the polarization of the pump pulse parallel to that of the probe pulse. The photodiode PD measured the pump-pulse intensity and the time at which the photodiode signal occurred was compared with the flashlamp discharge current to verify proper time synchronization. The probe and pump pulses were recombined at mirror M_2. Considerable attention was given to maintaining exact spatial and temporal overlap of the two beams over the length of the iodine cell. An inverted Gallilean telescope reduced the beam diameters to 2 mm. Entering the iodine cell, the pump-pulse intensity was approximately 10^{10} W/cm^2. The maximum laser pump intensity in the collimated beam was limited by the damage threshold of the quartz cell window. A collimated-beam, rather than a focused-beam, geometry was chosen again to avoid optical breakdown and gain saturation, and also to minimize the effects of line broadening due to the optical Stark effect. As the two pulses passed through the inverted iodine, the portion of the dye emission spectrum at the anti-Stokes Raman wavelength was amplified as indicated in Fig. 2. After passing through the iodine cell, the pump pulse was blocked by the filter F_2, and the probe pulse entered the 1-m Spex spectrograph which was set to transmit a bandwidth of 0.3 cm^{-1} at the anti-

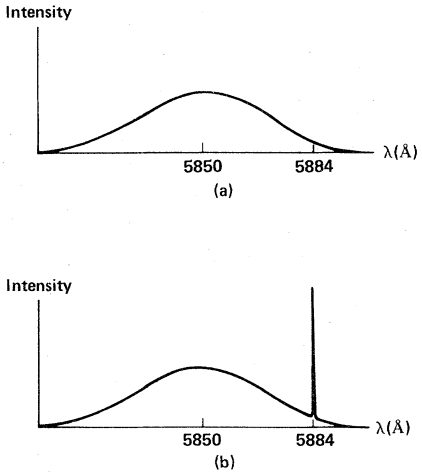

FIG. 2. Schematic representation of the laser probe pulse spectrum (a) before and (b) after passing through the inverted iodine coincident with the laser pump pulse.

Stokes wavelength. The amplitude of the signal from the photomultiplier PM_2 was proportional to the transmitted probe pulse energy E_2. The ASRS gain was obtained from the ratio E_2/E_1 which was measured to be 7.4 ± 0.5. No gain was observed when the iodine was not inverted, when the laser intensity was reduced by a factor of 3, or when the spectrograph was tuned 0.5 cm^{-1} away from the anti-Stokes wavelength.

Atomic iodine was chosen for these investigations for several reasons. A number density of inverted atoms sufficient to produce a measurable electronic Raman gain could be generated by flash photolysis of CF_3I under reasonable flashlamp pumping conditions. Since the transition $^2P_{1/2} \rightarrow {}^2P_{3/2}$ is the 1.315-μm atomic iodine laser transition, the ways in which to produce efficiently and measure quantitatively the population inversion had been well studied,[6] and information concerning the relevant kinetics was available.[7] At the laser intensities used in this experiment, stimulated vibrational Raman scattering did not occur in CF_3I or the dissociation products.

An iodine laser head designed to assure uniform pumping of the CF_3I was used for these experiments. The CF_3I was contained at a pressure of 100 Torr in a tube 60 cm long with 12 mm diam. Pumping was accomplished by four linear flashlamps with 7-mm bore and 50-cm arc length. The flashlamp discharge lasted 20 μsec with equal 10-μsec rise and decay. With the addition of an external mirror to provide feedback at 1.315 μm, laser action in the iodine cell occurred. The iodine transition is magnetic-dipole allowed with a single-photon emission cross section $\sigma = 2\times10^{18}$ cm^2 for 100 Torr CF_3I. Measurements of the output energy at laser threshold allowed an estimate of the peak inversion density of $N = (2-5)\times10^{16}$ atoms/cm^3. Significantly higher inversion densities were not possible because of the superfluorescence decay of the excited state. Since for CF_3I pressure of 100 Torr, the excited-state lifetime of ~50 μsec is long compared to the flashlamp discharge, the peak inversion occurred at the end of the discharge. It was a major concern that shock waves would be generated in the CF_3I by flashlamp heating before a significant inversion existed and the resulting light scattering would prevent the observation of gain. To study this effect, a He-Ne laser beam of various diameters was passed through the iodine cell followed by a lens-aperture spatial filter. The transmitted intensity, measured by a photomultiplier, decreased abruptly 25 μsec after initiation of the flashlamp discharge. Thus, the laser pulses had to pass through the iodine cell within 5 μsec of the end of the discharge. Since the excited-state lifetime is sensitive to small concentrations of impurity molecules, notably I_2 and O_2,[8] the gas cell was pumped and refilled after each shot.

According to the theory of stimulated Raman scattering,[9] exponential amplification $I_2 = I_1 \times \exp(gI_L L)$ occurs if the amplified ASRS intensity I_2 is small compared to the laser intensity I_L. I_1 is the incident ASRS intensity, L is the interaction length, and the steady-state gain coefficient g is given by[10]

$$g = \frac{2\lambda_{AS}^2 N(\partial\sigma/\partial\Omega)}{\hbar\omega_L} \frac{\Gamma}{\Gamma^2 + \Delta\omega^2}. \quad (1)$$

Here $\partial\sigma/\partial\Omega$ and Γ are the spontaneous Raman scattering differential cross section and linewidth (half width at half-maximum), respectively; λ_{AS} is the anti-Stokes wavelength; ω_L is the laser frequency; and $\Delta\omega = \omega_L - \omega_{AS} - \omega_0$ is the frequency offset from exact resonance. The Raman scattering cross section for atomic iodine can be estimated from general formulas.[11] Since the energy levels of interest are far from all other atomic levels and from the ionization continuum, a summation over virtual states can be carried out approximately by the use of sum rules. Such an estimate gives the value of $\partial\sigma/\partial\Omega \simeq 10^{-28}$ cm^2/sr for a pump wavelength of 1.06 μm. The collisionally broadened Raman linewidth is $\Gamma \simeq 0.05$ cm^{-1}.[12] The optical Stark shift of the iodine levels under

the influence of a laser intensity of 10^{10} W/cm^2 is approximately 0.03 cm^{-1}. From Eq. (1) the expected ASRS gain was estimated to be approximately e^7. Since the pump pulse duration t_p satisfies the relationship $t_p > gI_L L\Gamma^{-1}$, the gain may be calculated by use of the steady-state theory.[13]

The experimentally measured quantity is the energy ratio E_2/E_1 where E_1 and E_2 are, respectively, the incident and amplified pulse energies in the 0.3-cm^{-1} (full width at half-maximum) bandwidth transmitted by the spectrograph. Thus, to compare with the experiment, the ASRS intensity gain must be expressed in terms of this ratio. The pulse energies are obtained from the intensities by integration over time t, radius r, and frequency offset $\Delta\omega$:

$$E_{1,2} = \int I_{1,2}(r,t,\Delta\omega)2\pi r\,dr\,dt\,d\Delta\omega. \quad (2)$$

The incident probe and pump pulses are taken to be Gaussian with the same radius and duration:

$$I_{1,L} = I_{10,L0}\exp\{-[(r/r_0)^2 + (t/t_0)^2]\}. \quad (3)$$

In addition, we assume the pump pulse to be monochromatic, and the probe intensity to be constant as a function of frequency over the bandwidth of interest.

We introduce the parameters $R = r/r_0$, $T = t/t_0$, $\omega = \Delta\omega/\Gamma$, and $\gamma = g(0)I_{L0}L$ which is the gain coefficient on resonance at the maximum laser intensity. The integration to obtain E_1 can be done exactly. However, only the radial part of the E_2 integral can be done, so an approximation must be made. For $R^2, T^2, \omega^2 \ll 1$ and $\gamma \gg 1$, I_2 may be approximated by

$$I_2 \simeq I_{10}\exp[\gamma(1 - R^2 - T^2 - \omega^2)]. \quad (4)$$

The effects of "gain narrowing" are immediately apparent as the half-width in R, T, or ω of the amplified pulse is proportional to $(\ln 2/\gamma)^{1/2}$. Thus for sufficiently large values of γ, the amplified pulse energy E_2 is obtained to a high degree of accuracy by integration over the volume in the (R,T,ω) parameter space which does not violate the initial assumption of the approximation given by Eq. (4) to I_2. The calculated value of the energy ratio is then $E_2/E_1 = 0.3(e^\gamma/\gamma^2)$. From the experimental value of the ratio $E_2/E_1 = 7.4 \pm 0.5$, the peak ASRS gain e^γ occurring under the conditions of this experiment is found to be $e^{7.1\pm0.2}$. Using this value for the peak gain, the error in the approximation to E_2 was estimated by numerical integration and found to be less than 15%. In view of the uncertainties in experimental parameters, the observed gain is in good agreement with theoretical predictions.

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