16-μm generation by CO₂-pumped rotational Raman scattering in H₂

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We have generated 50 mJ of 16.9-μm radiation by stimulated rotational Raman scattering in 3 atm of H₂ gas pumped by a CO₂ TEA-laser source. Threshold was reached by injection of a few microjoules of 16.9-μm radiation generated by four-wave mixing. We achieved 25% peak power, or 40% peak photon conversion efficiency.

In 1976 Byer proposed using stimulated rotational Raman scattering in hydrogen gas as an efficient method of frequency conversion in the infrared. In particular, it was noted that stimulated Raman scattering in para-H₂ gas with a CO₂ laser pump generates 16-μm radiation, which is of potential use for UF₆ isotope enrichment. It was subsequently learned that others have also suggested stimulated rotational Raman scattering in the infrared. Recently Frey et al. have generated 16-μm output at the 1-mJ level by vibrational Raman scattering in H₂ and N₂ pumped by a ruby-pumped dye laser.

We have successfully generated 50 mJ of 16.9-μm radiation by four-wave mixing-assisted stimulated rotational Raman scattering. We used a 2-J, 70-nsec CO₂ laser source to pump a 25-pass, 4-m-long cell filled with 3 atm of p-H₂ gas obtained from a liquid H₂ source. Based on theoretical calculations and rotational Raman threshold, scaled from careful measurements at 1.064 μm, we predicted a CO₂-pumped rotational Raman threshold in our multipass cell of 1.7-2.6 J in a 70-nsec gain-switched pulse. Our available CO₂ pump energy was not adequate to reach stimulated Raman threshold by amplification of spontaneous emission. However, when 16.9-μm radiation generated by four-wave mixing was injected into the multipass cell, the Raman gain was sufficient to lead to significant pump depletion and a peak photon conversion efficiency of 40% to the Stokes wave at 16.9 μm.

The four-wave mixing process was first described for interactions in crystals and later extended to gases as a means for generating widely tunable infrared radiation. Sorokin et al. have demonstrated four-wave mixing in H₂ gas using a CO₂ laser source as a method for 16-μm generation.

The equations governing both four-wave mixing and stimulated Raman scattering are given by

\[
\frac{\partial E_o}{\partial z} = -\frac{\omega_o}{2cn_o} \chi_{R''} (|E_o|^2 E_p + E_o*E_p e^{i\Delta k z}), \quad (1a)
\]

\[
\frac{\partial E_s}{\partial z} = +\frac{\omega_s}{2cn_s} \chi_{R''} (|E_s|^2 E_o + E_s*E_o e^{-i\Delta k z}), \quad (1b)
\]

\[
\frac{\partial E_i}{\partial z} = -\frac{\omega_i}{2cn_i} \chi_{R''} (|E_i|^2 E_p + E_i*E_p e^{-i\Delta k z}), \quad (1c)
\]

\[
\frac{\partial E_o}{\partial z} = +\frac{\omega_o}{2cn_o} \chi_{R''} (|E_i|^2 E_o + E_o*E_s e^{i\Delta k z}) \]

where \(\omega_o - \omega_s = \omega_i - \omega_p = \omega_R = \omega_p - \omega_R = \Delta k = -(k_p - k_s) + (k_1 - k_o)\) with \(\omega_R\) the characteristic Raman frequency of the medium with a peak Raman susceptibility \(\chi_{R''}\). In our case \(E_p\) is the Nd:YAG pump field at 1.064 μm, \(E_s\) is the generated Stokes field at 1.10 μm, \(E_i\) is the input field at 10.6 μm, and \(E_o\) is the generated output at 16.95 μm.

If we neglect four-wave mixing for the moment, Eqs. (1a)–(1d) separate into two pairs of equations that describe stimulated Raman scattering pumped at \(E_p\) and \(E_i\). In the absence of pump depletion, the Stokes power increases exponentially as \(P_s(l) = P_s(0) \exp(g_s l)\), where \(P_s(0)\) and \(P_s(l)\) are the input and output Stokes power, respectively, and \(g_s\) is the Raman power-gain coefficient given by

\[
g_s = \frac{\omega_s \chi_{R''} |E_p|^2}{n_s c} = \frac{4\pi \chi_{R''} I_p}{|\lambda_n n_p| c}. \quad (2)
\]

The input power to the Raman cell \(P_s(0)\) is usually provided by blackbody or spontaneous Raman noise. In our case the spontaneous noise is dominant so that the noise in a single polarization and single spatial mode within the Raman linewidth \(\Delta \nu_R\) is \(P_s(0) = h \nu_s \Delta \nu_R \approx 10^{-12} W\). Thus, for a CO₂ pump power of 10 MW, the net gain required to amplify the spontaneous power up to the order of the pump power is \(g_s l = \ln[P_s(l)/P_s(0)] = \ln[10^7 W/10^{-12} W] = 44\).

The 16.95-μm radiation generated by four-wave mixing can also be amplified by the stimulated Raman gain process. It has been previously shown and is evident from Eqs. (1a)–(1d) that the four-wave mixing conversion efficiency in the absence of significant Raman amplification and pump depletion is given by

\[
\frac{I_s}{I_p} = \left(\frac{\omega_o}{\omega_p}\right) \supl2 n_i n_p I_s \frac{n_o n_s}{I_p}, \quad (3)
\]

where \(I_i, I_o, I_s,\) and \(I_p\) are the intensities and \(n_i, n_o, n_s,\) and \(n_p\) are the refractive indices at the four fields. In the present case the Nd:YAG laser source at \(\omega_p\) converts 40% of its energy to the Stokes wave by stimulated Raman scattering on the first transit of the 25-pass cell. The incident CO₂ laser intensity at \(\omega_i\) then generates
output at $\omega_o$ by four-wave mixing. The generated output power at 16.95 $\mu$m is given by $P_o = 0.4(1.10 \mu$m/16.95 $\mu$m)$^2 P_i = 1.68 \times 10^{-3} P_i$. A 10-MW CO$_2$ laser produces 16.8 kW or 193 $\mu$J of energy in an 8-nsec pulse. In practice, less energy is produced because of the imperfect spatial overlap of the beams and the nonzero phase-mismatch factor $\Delta k$. The generated 16-$\mu$m energy is then amplified on the remaining passes by the CO$_2$ laser-pumped Raman gain.

The generated four-wave mixing power is 16 orders of magnitude greater than the spontaneous Raman power and thus significantly reduces the Raman gain required to reach threshold. The required net Raman gain with four-wave mixing injection is $g_i l = \ln(P_o(t)/P_o(0)) = \ln(10^{10} W/10^4 W) \approx 7$. The required Raman gain and thus input pump power is, therefore, reduced by a factor of 44/7 or 6.2 times compared to the amplified spontaneous-emission case.

Figure 1 shows a schematic of the experimental apparatus. The CO$_2$ laser source consists of a Lumonics 103 oscillator followed by a Lumonics 102 amplifier. A Q-switched unstable resonator Nd:YAG laser operating at 1.064 $\mu$m provided the pumping for generating the four-wave mixing signal. The Nd:YAG laser pulse energy was limited to 70 mJ, which was 35 times the measured 2-mJ threshold because of the onset of damage of the metal turning mirror. The Nd:YAG and CO$_2$ beams were combined on a ZnSe plate and were focused into the multipass cell by a 9-m-radius mode-matching reflector followed by a dielectric bandpass filter and is also monitored by a pyroelectric energy meter. The wavelength of the Raman signal measured with a grating monochromometer is 16.95 $\mu$m, as expected.

The multipass cell consists of a 3.77-m-long, 15.24-cm-diameter stainless steel tube, with steerable 12.70-cm-diameter, 2-m-radius copper mirrors mounted at each end. Light is coupled into and out of the cell through salt windows mounted on the sides of the tube. Light entering the cell is directed down the axis of the tube by a 1.27-cm-diameter steering mirror. The beam then reflects back and forth between the 12.70-cm-diameter mirrors refocusing on each transit and wandering in a circle around the rims of the mirrors. After 25 transits, the beam is coupled out of the multipass cell by a second 1.27-cm-diameter steering mirror and enters the screen room. The monochromator is 16.95 $\mu$m, as expected.

The Nd:YAG laser power results in an increase of 3 orders of magnitude greater than the spontaneous Raman signal. As expected. Figure 2 shows the rotational Raman threshold dependence on p-H$_2$ pressure at 1.06 $\mu$m in the multipass cell. As the pressure increases, the threshold rapidly drops to about 2 atm, where it levels off at about 2 mJ. In a long cell, pressure-induced absorption in H$_2$ gas at 10.6 $\mu$m is significant and limits the maximum operating pressure. The second curve in Fig. 2 is a theoretical curve showing the fraction of CO$_2$ energy absorbed versus H$_2$ pressure. Measurements of CO$_2$ transmittance versus pressure were in agreement with this curve. Taking both pressure-induced absorption and Nd:YAG threshold data into account, it is apparent that there is an optimum operating pressure of between 1 and 4 atm.

The Nd:YAG laser linewidth was also varied from single axial mode to about 0.5 cm$^{-1}$ by inserting etalons into the Nd:YAG resonator. The Raman threshold was found to be independent of laser linewidth, in agreement with theory. Our CO$_2$ laser source was thus operated without linewidth control to obtain the maximum output pulse energy. Based on the Nd:YAG threshold measurements, we predicted a Raman threshold for the CO$_2$ laser of between 1.7 and 2.6 J in a 70-nsec pulse for 3 atm of p-H$_2$ in the multipass cell. The gas-breakdown limit in our present cell is 4.5-J input CO$_2$ energy.

With both the YAG and CO$_2$ lasers properly aligned and synchronized in the H$_2$ cell, significant 16-$\mu$m energies were generated. Figure 3 shows the 16-$\mu$m output energy versus CO$_2$ laser peak input power. The ±50-nsec timing jitter between the YAG and CO$_2$ lasers resulted in large pulse-to-pulse variations in 16-$\mu$m energy. What is displayed are clusters of points that represent the best results at each CO$_2$ laser-power setting. The dots represent data taken at 1.5 atm, and the X's represent data taken at 3-atm H$_2$ pressure in the cell. An important point to notice is that the 16-$\mu$m energy is displayed on a log scale. An increase of 3 in CO$_2$ laser power results in an increase of 3 orders of magnitude.
magnitude in 16-μm energy. In a four-wave mixing process, the 16-μm energy should be limited to the 100-μJ level and increase linearly with CO₂ power. The exponential increase in 16-μm energy is due to CO₂-pumped Raman gain. The slopes of the lines give the Raman exponential gain coefficient for the multipass cell, which is 0.28/MW. This gain coefficient is a factor of 3 less than the plane-wave gain value predicted from Nd:YAG laser threshold scaling and is accounted for by the spatial mode overlap factor of the 10.6- and 16.9-μm beams proportional to \((\lambda_s + \lambda_p)/\lambda_p\).

The maximum observed 16-μm pulse energies were 50 mJ. The 8-nsec-long 16-μm pulse generated by four-wave mixing was significantly shorter than the 70-nsec CO₂-laser pulse, and, therefore, depleted a small portion of the energy in the CO₂ pulse. The 50-mJ pulse energy corresponded to a 27% peak power depletion or to a 40% peak photon conversion from 10.6 to 16.9 μm.

We have successfully used four-wave mixing injection to reduce the CO₂ power required to reach stimulated rotational Raman threshold on the \(S(0)\) transition in \(p-H₂\) gas. This technique should permit wider application of stimulated Raman scattering processes in the infrared as an efficient means of frequency conversion. The use of a tunable high-pressure CO₂-laser source should permit continuous tuning over wide regions of the infrared extending from 8 μm to beyond 20 μm by the generation of Stokes and anti-Stokes output from various rotational levels of hydrogen and its isotopes.

The high conversion efficiency and direct scalability to high peak and average powers of CO₂-pumped stimulated rotational Raman scattering should permit this approach to meet the large-scale isotope enrichment source requirements.

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