Continuous-wave Raman laser in H_2

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Recent developments in high-finesse cavities now make broadly tunable, continuous-wave Raman lasers possible. The design and preliminary characterization of what is to the authors' knowledge the first continuous-wave Raman laser in H_2 are presented. The threshold is currently at 2 mW of pump, making diode laser pumping possible. The maximum photon conversion efficiency observed was 35% at 7.6 mW of pump power.

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Raman scattering occurs when an incident photon interacts with a molecule (or an atom) and generates a red-shifted photon with the change in wavelength resulting from the conservation of energy when the excitation in the molecule occurs. The molecular excitation can be rotational, vibrational, or electronic, which accounts for the possibility of many different wavelengths. For example, Fig. 1 shows the levels involved for vibrational Raman scattering of a 532-nm laser in H_2 to 683 nm. At high laser intensities the Raman process can have gain and produce stimulated Raman scattering, and, because of the high intensities needed, stimulated Raman scattering is most often studied in the high-power, pulsed regime. Stimulated Raman scattering has also been utilized in the cw regime to make cw Raman lasers. However, because of the lower intensity of the cw pump lasers, these cw Raman lasers generally operate near a molecular (or atomic) resonance to increase the Raman gain and therefore are tunable only over narrow regions near the resonance. A few examples include a 67-μm cw Raman laser in NH_3,[1] a cw Na Raman laser near the *D* lines,[2] a two-photon-pumped cw Rb Raman laser near 776 nm,[3] and various cw Raman lasers near Ne resonances in a He–Ne laser discharge tube.[4–6] In addition, cw Raman lasing is also possible in optical fibers in which the long interaction length of the fiber and the small spot size in the fiber increase the gain so Raman lasing can occur. However, typically input pump powers of ~1 W are needed to pump these Raman fiber lasers.[7] In addition, the Raman shift in a fiber is only ~440 cm^{-1}, which is much smaller than the shift in H_2 of 4 155 cm^{-1} and is inconvenient if one is looking for substantial shifts of wavelength.

Recent developments in coating technology have led to the availability of new mirrors with reflectivities of 99.995% and higher. These low-loss mirrors have been used to build nonconfocal cavities with finesses of 50,000 and higher.[8,9] By use of the nonconfocal cavity, the spot size on the mirrors is of the order of 100 μm, so the degradation of the finesse that is due to wavefront distortion on the mirror is minimized and the high-finesse cavity is achieved.

With the advent of this new technology of high-finesse cavities it now becomes possible to consider using these cavities to do studies in nonlinear optics with cw lasers in the milliwatt range of power. Specifically, it is now possible to use these high-finesse cavities to build a widely tunable cw Raman laser that can be pumped with lasers at the milliwatt level. This low power requirement for pumping raises the possibility of driving these cw Raman lasers with compact and efficient diode lasers. With available room-temperature diode lasers, cw Raman lasers covering the spectrum from the visible to the near IR (~4 μm) appear possible.[10]

To predict the threshold for the cw Raman laser, we start with the single-pass gain G in a focused geometry[13]:

$$G = \frac{4\alpha}{\lambda_p + \lambda_s} \tan^{-1}(l/b) P_p \,, \tag{1}$$

where G is the Raman gain per pass, α is the Raman gain coefficient, λ_p (λ_s) is the wavelength of the

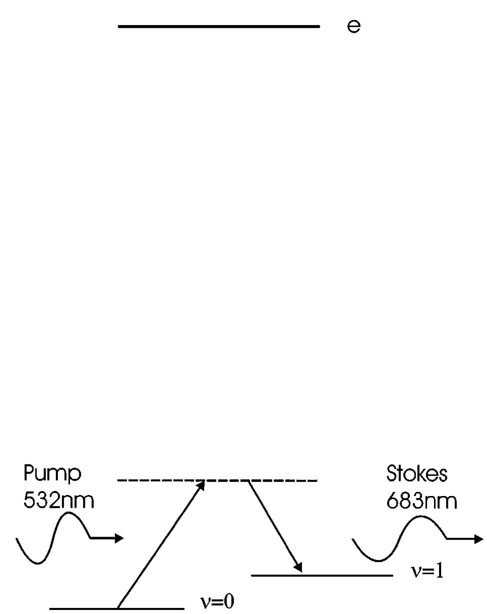

Fig. 1. Energy levels in H_2 (to scale) used for the cw Raman laser. Note that the virtual level (dashed line) is associated with the first electronic excited level (labeled e). Therefore this nonresonant Raman laser will be broadly tunable without large changes in the gain.

pump (Stokes) field, l (b) is the interaction length (confocal parameter) of the beam, and P_p is the incident pump power. The Raman gain coefficient, α, has the following functional form[14]:

$$\alpha = \frac{D(\nu_p - \nu_v)}{(\nu_i{}^2 - \nu_p{}^2)^2} = \frac{D\nu_s}{(\nu_i{}^2 - \nu_p{}^2)^2}\,, \tag{2}$$

where D is a constant that incorporates the pressure of the gas and the linewidth of the transition, ν_p (ν_s) is the pump (Stokes) frequency, and ν_i (ν_v) is the resonant electronic (vibrational) frequency of the transition. For diatomic hydrogen, $\nu_i = 8.48 \times 10^4\ \mathrm{cm}^{-1}$ and $\nu_v = 4155\ \mathrm{cm}^{-1}$. Note that ν_i corresponds to a resonance at 118 nm in the UV, so $\upsilon_i{}^2 - \upsilon_p{}^2$ will be a slowly varying function for ν_p in the visible and the IR parts of the spectrum. Thus the cw Raman laser in H_2 is expected to the broadly tunable. For a pump wavelength of 532 nm (18 800 cm^{-1}), α is 2.5×10^{-9} cm/W in the high-pressure limit.[14]

To increase the gain one can inject the pump laser into a high-finesse cavity filled with H_2, which is doubly resonant, at both the pump and the Stokes wavelengths. If the pump is then frequency locked to the cavity resonance, the circulating pump power inside the cavity will be substantially increased. The gain per pass in the cavity will then become[15]

$$G' = G\left(\frac{\sqrt{T_P}}{1 - \sqrt{R_p}}\right)^2, \tag{3}$$

where G' is the gain at the Stokes frequency with a pump resonant to the cavity, G is the gain from Eq. (1), and T_p (R_p) is the transmission (reflectivity) of the mirrors at the pump wavelength.

To couple the incident pump into the cavity efficiently requires not only that the incident laser be frequency locked to the cavity but also that it be spatially mode matched to the cavity. For a high-finesse cavity with two concave mirrors of radius of curvature C, the confocal parameter of a beam that is mode matched to the cavity is given by $b = [l(2C - l)]^{1/2}$, where l is the length of the cavity.[8]

If we assume that the cavity is resonant at both the pump and the Stokes transitions, we arrive at the following condition for the threshold of the cw Raman laser:

$$G' = -\ln(R_s)\,, \tag{4}$$

where G' is the gain from Eq. (3) and R_s is the reflectivity of the mirrors at the Stokes wavelength.

For mirrors coated at the pump and the Stokes wavelengths with reflectivities of 99.98% and transmission of the mirrors at 185 parts in 10^6, the threshold for the process is less than a milliwatt. This low threshold indicates the possibility of pumping the cw Raman laser with conventional diode lasers.

The apparatus used to make the first cw Raman laser in H_2 is shown in Fig. 2. For this first experiment we chose to use a frequency-doubled cw Nd:YAG laser with output powers up to 200 mW. The output of this Nd:YAG laser[16] at 532 nm was sent though a Faraday isolator to minimize feedback to the Nd:YAG laser. An electro-optic modulator (EOM) was used to place sidebands on the carrier frequency required for locking the pump to the cavity. The laser beam traveled though a two-element lens pair used to mode match the pump beam to the cavity. A polarizing beam splitter (PBS) cube in conjunction with a quarter-wave plate allowed for monitoring of the beam reflected from the cavity. A fast detector was then used to measure the error signal. A low-noise amplifier gave the signal 30 dB of amplification before the signal was mixed to dc. The error signal entered the servo,[17] and the slow corrections were sent to temperature tune the pump laser, while the fast corrections were sent to the pump laser piezoelectric transducer (PZT).

To ensure that the Stokes and pump cavity resonances fall within a Raman linewidth, either the pump laser or the Raman cavity must be scanned until a resonance occurs. In the present experiment this required a scan of seven free-spectral ranges (~20 GHz) to guarantee this overlap. However, the pump laser used in this experiment does not have enough tuning range to cover the needed seven free-spectral ranges. Therefore a PZT was placed inside the Raman cavity to adjust the cavity length. The Raman cavity was tuned though pump resonances until the Stokes output was seen. The second mirror was then adjusted mechanically by a New Focus picomotor until the Stokes output corresponded to a zero voltage on the cavity PZT. The picomotor and the cavity PZT were turned off, and the laser was locked to the doubly resonant cavity line.

The Raman cavity used in this experiment has a design similar to the high-finesse cavity described in Ref. 9. The cavity mirrors had a radius of curvature of 50 cm and were spaced by 7.28 cm of ULE 7971 from Corning. The cavity was filled with 10 atm of H_2, resulting in a Raman linewidth (FWHM) of 510 MHz.

The output of the cw Raman laser was measured at the exit of the cavity, with a narrow-band filter at 683 nm placed in the beam at a slight angle to separate the pump and the Stokes beams. The output power of the pump laser was varied, and the pump laser was locked to the cavity. The average pump and Stokes powers were measured, and a photodiode was used to monitor the amplitude noise of the Stokes beam.

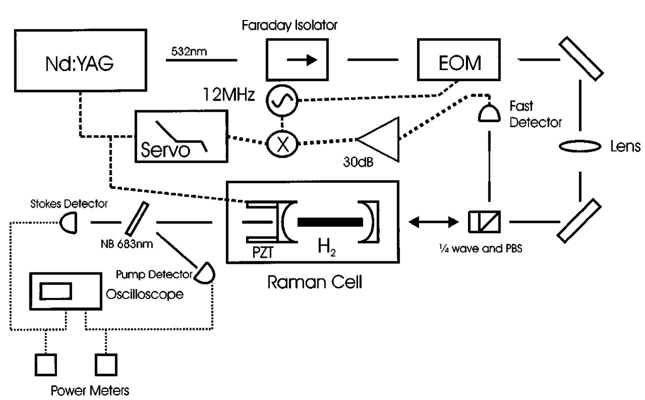

Fig. 2. Experimental diagram for the cw Raman laser in H_2: NB, narrow-band.

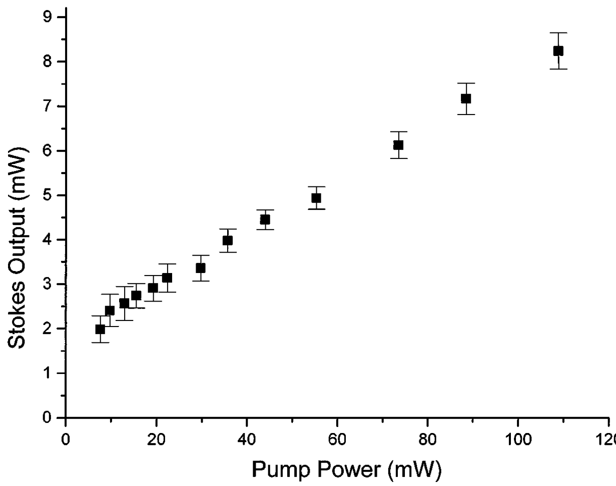

Fig. 3. Stokes output power of the cw Raman laser versus the pump input power. The threshold occurs near 2 mW, and the maximum photon conversion efficiency is 35% at an input pump power of 7.6 mW.

Figure 3 shows the power of the Stokes field at the exit of the cavity. The Raman laser has a threshold of ~2 mW. The maximum photon conversion efficiency is 35% at a pump power of 7.6 mW. The Raman laser has a maximum output of 8.2 mW.

The theory predicts that the threshold for the Raman laser should be of the order of 600 μW for mirror reflectivities (transmissions) of 0.99983 (92 parts in 10^6).[18] However, in the present setup we see a threshold of ~2 mW. We believe that this discrepancy can be attributed to the accuracy of the overlap of the double resonance of the cavity for the pump and the Stokes beams.

Currently, the intensity fluctuations on the pump and the Stokes beams at the exit of the cavity are less than 1% for high pump powers and increase to 5% for low pump powers. We believe that these intensity fluctuations, which occur in a frequency band of 30–300 kHz, are related to the interaction of the locking servo loop with the intensity buildup in the cavity, which occurs on a time scale of approximately 1–10 μs. The Raman laser is more efficient at lower pump powers, causing a larger initial oscillation in the locking signal, which is more difficult for the servo to suppress.

In conclusion, what is to our knowledge the first nonresonant cw Raman laser in H_2 was demonstrated with a maximum output of 8.2 mW and a maximum photon conversion efficiency of 35%. The threshold of the cw Raman laser was seen to be 2 mW, making diode laser pumping possible.

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