Continuous-wave rotational Raman laser in H_2

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Received November 20, 2001

A diode-pumped, far-off-resonance cw Raman laser in H_2 with rotational Stokes emission is reported for the first time to our knowledge. The Raman laser can produce single-wavelength emission at either 830 nm (rotational Stokes) or 1180 nm (vibrational Stokes) depending on the frequency tuning of the pump laser. The mirrors for the rotational cw Raman laser are easier to produce; the laser also exhibits a wider continuous tuning range and is less sensitive to thermal effects than the previously studied vibrational Raman laser [Opt. Lett. **26,** 426 (2001) and references therein]. © 2002 Optical Society of America

OCIS codes: 140.3550, 140.3480, 140.3410, 140.3600, 290.5910.

The far-off-resonance cw Raman laser utilizing high-finesse-cavity (HFC) enhancement is a recent development for optical frequency downconversion.[1] Tunability[2,3] and a narrow linewidth[4] make this technique applicable in such areas as high-resolution laser spectroscopy, atomic physics, and remote sensing. To our knowledge, all such lasers reported so far have been based on a pure vibrational Raman transition in H_2 [i.e., the $Q_{01}(1)$ (Ref. 5) transition with a shift of 4155 cm^{-1}]. In this Letter we report for what is believed to be the first time a cw Raman laser with a pure rotational Stokes emission [the $S_{00}(1)$ (Ref. 5) transition with a shift of 586.9 cm^{-1}].[6]

The experimental setup used to generate the rotational Stokes is shown in Fig. 1 and is basically identical to the one reported in Ref. 2: Both the pump laser and the HFC are the same, except that the HFC mirrors in the current setup are degraded, with smaller reflectivity and larger absorption, because of aging and handling. In the current system the finesse of the HFC is estimated to be $\sim 4 \times 10^4$ at all three wavelengths (pump, rotational Stokes, and vibrational Stokes), whereas in Ref. 2 the finesse was $\sim 6 \times 10^4$ at all three wavelengths. An external-cavity diode laser with 30-mW maximum output power and a 792-nm center wavelength is used as the pump source. The standard Pound–Drever–Hall laser-locking technique[7] is used to stabilize the laser frequency to a resonance of the HFC. The H_2 pressure in the HFC is approximately 10 atm. With this setup, pure vibrational Raman shifting from 792 to 1180 nm has been reported.[2] In this Letter we report on the additional discovery of pure rotational Raman shifting from 792 to 830 nm when the pump frequency is appropriately tuned. We measure the powers of both the vibrational and the rotational Stokes emissions as functions of pump frequency tuning and pump power, as shown in Figs. 2 and 3.

In Fig. 2 we show that as the pump frequency is tuned the system exhibits sharp switching between the vibrational and rotational Stokes emissions. Each of the two Stokes modes operates in a single longitudinal and spatial cavity mode. These behaviors indicate the homogeneous nature of the Raman gain medium. Figure 3 shows how the rotational Stokes power grows and the transmitted pump power clamps after the Raman threshold is reached, the same dependence as with the vibrational cw Raman lasers previously studied.[2]

The incident pump light is circularly polarized. The emitted vibrational Stokes has the same circular polarization, whereas the rotational Stokes is counter-circularly polarized.[6] We choose the circularly polarized pump because (1) the rotational Raman gain is 1.5 times higher than with the linearly polarized pump[6] and (2) it allows us to avoid possible competition from rotational anti-Stokes generation.[6] It is known that when the rotational Stokes is pumped by linearly polarized light, the gain can be greatly suppressed by parametric Stokes–anti-Stokes coupling.[8] We are currently investigating the cavity-enhanced Stokes–anti-Stokes coupling and parametric gain suppression both theoretically and experimentally.

Using the theory given in Ref. 3, we model the tuning characteristics of this dual-output laser system. If a cavity mode of the Stokes is initially detuned by $\Delta(0)$ from the Raman gain line center, after a pump frequency tuning of $\delta\nu_p$, the detuning from the gain line center will become[3]

$$\Delta(\delta\nu_p) = \Delta(0) + \left(\frac{\lambda_p}{\lambda_s} - 1\right)\delta\nu_p ,$$

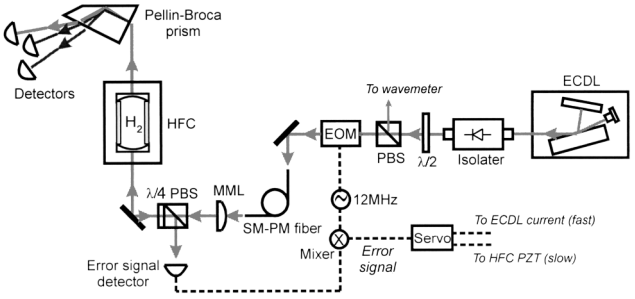

Fig. 1. Experimental setup used to study the rotational cw Raman laser. ECDL, external-cavity diode laser; $\lambda/2$, half-wave plate; $\lambda/4$, quarter-wave plate; PBS; polarization beam splitter; EOM, electro-optic modulator; SM-PM fiber, single-mode polarization-maintaining fiber; MML, mode-matching lens; HFC, high-finesse cavity; PZT, piezoelectric transducer. The standard Pound–Drever–Hall technique is used to lock the laser's frequency to a resonance of the HFC.

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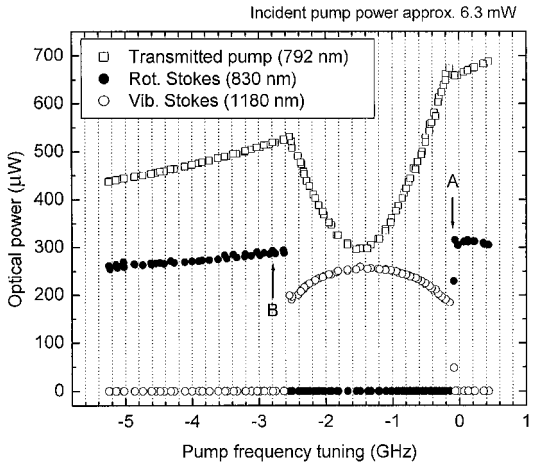

Fig. 2. Continuous tuning curve measured for the cw Raman laser. The pump frequency tuning is shown relative to 37 8440.00 GHz, as measured by a Burleigh wavemeter with 10-MHz resolution. Note that at point A, there is some coexistence of the rotational (Rot.) and vibrational (Vib.) Stokes. At point B, we measure the power dependence, as shown in Fig. 3, below.

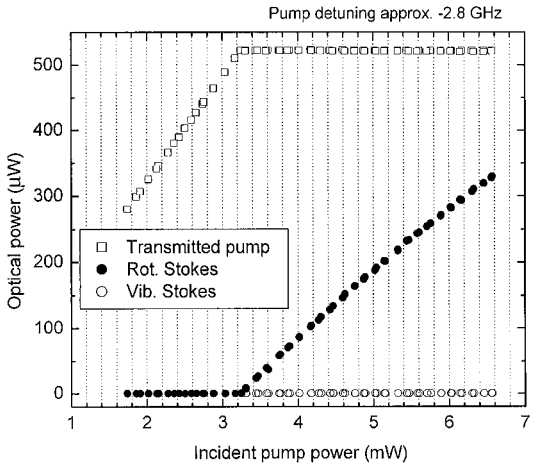

Fig. 3. Plot of the rotational Stokes power and transmitted pump power as a function of the incident pump power. The growth of the Stokes power and clamping of the pump power that can be seen are similar to what was observed for the vibrational cw Raman laser.[2]

where λ_p and λ_s are the wavelengths of the pump and Stokes, respectively. The Raman gain, pressure broadened at a high pressure of 10 atm, exhibits a homogeneous Lorentzian line shape for both the vibrational and the rotational transitions. The Raman plane-wave gain coefficient α associated with a cavity mode of the Stokes can then be modified as

$$\alpha(\delta\nu_p) = \alpha_0 \frac{(\Gamma/2)^2}{\Delta(\delta\nu_p)^2 + (\Gamma/2)^2},$$

where α_0 is the Raman plane-wave gain coefficient at the gain line center and Γ is the FWHM Raman gain linewidth. At a temperature of 300 K, a H_2 pressure of 10 atm, and a pump wavelength of 792 nm, we calculate that, for vibrational Raman transition $Q_{01}(1)$, $\alpha_{0\nu} = 1.5 \times 10^{-11}$ m/W (Ref. 9) and $\Gamma_\nu = 0.5$ GHz,[10] while for rotational Raman transition $S_{00}(1)$, $\alpha_{0r} = 0.54 \times 10^{-11}$ m/W (Refs. 11–13) $\Gamma_r = 1.02$ GHz.[14] Using these parameters, in Fig. 4(a) we plot theoretically the Raman plane-wave gain coefficients associated with the vibrational and rotational Stokes cavity modes as functions of the pump frequency tuning.

The tuning curve observed in Fig. 2 can now be qualitatively explained. Because of the homogeneous nature of the Raman transitions, the Stokes mode with the largest net gain will oscillate. Based on this rule, we calculate the powers of the transmitted pump, the vibrational Stokes, and the rotational Stokes as functions of the pump frequency tuning, using the theory given in Ref. 4, and plot them in Fig. 4(b). One can see a qualitative agreement between measurement and theory, except that only an ~6-GHz range of the pump

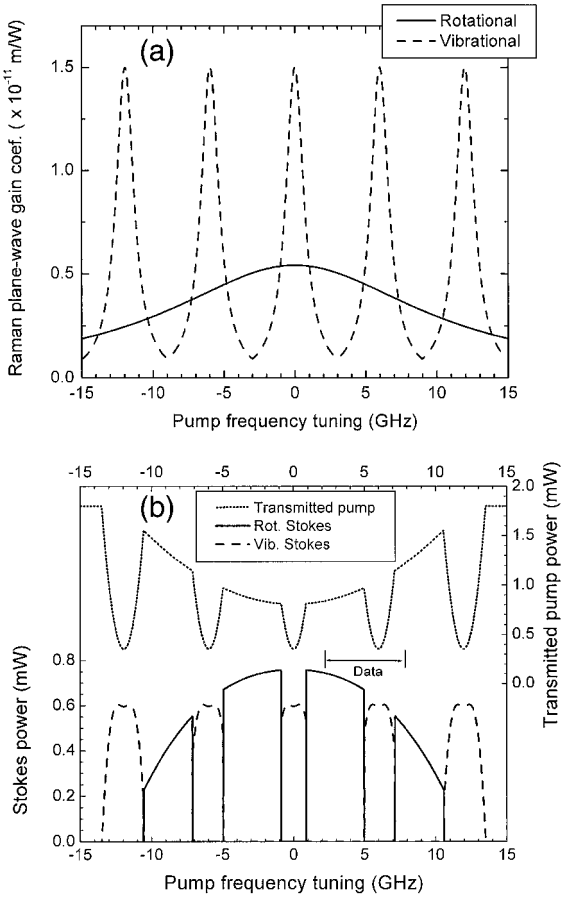

Fig. 4. (a) Theoretical plot of the Raman plane-wave gain coefficients of the rotational and vibrational transitions as functions of pump frequency tuning. The plot shows higher peak gain for the vibrational transition but wider tuning linewidth for the rotational transition in our double-resonance cavity. The cavity resonances of both Stokes are assumed to be at the gain line center when the relative pump frequency is zero [i.e., $\Delta(0) = 0$]. (b) Based on these gain profiles, the power of the transmitted pump, the rotational Stokes, and the vibrational Stokes are calculated as functions of pump tuning. The parameters used for the calculation are mirror reflectance, 0.9999; mirror absorptions, 40 parts in 10^6 at all wavelengths and both mirrors; and input pump power, 5 mW. The range shown is in qualitative agreement with the measured data presented in Fig. 2.

frequency can be tuned in the measurement because of the limited output range of the locking servo for the cavity piezoelectric transducer of 0–150V, which corresponds to a physical tuning range of ~1.2 μm of the piezoelectric transducer tube between the cavity mirrors.

Two phenomena in the laser's tuning behavior are noted. First, in spite of its smaller gain, the rotational Stokes can emit at higher power than the vibrational Stokes [see Fig. 2 or the middle range in Fig. 4(b)]. This counterintuitive effect is due to the smaller Raman shift of the rotational transition. In other words, more energy from each pump photon is transferred to the rotational Stokes photon than to the vibrational Stokes photon. Second, there is some coexistence of the two Stokes when the pump frequency is tuned to the switching point (see point A in Fig. 2). We believe that this coexistence is due to the slight inhomogeneity of the Raman gain (caused by Doppler broadening).

We did not quantitatively compare the theoretical calculation with the experimental data because we found that the interference effect between the two surfaces of the HFC mirrors caused significant modulation of the mirrors' effective reflectance as a function of the laser's frequency (for example, the results of our cavity ringdown measurements showed as large as 20% variation). This effect thus adds background modulation to the cavity-transmitted pump and Stokes powers as their frequencies are tuned and makes quantitative comparison difficult. The same problem is common in cavity ringdown spectroscopy (see, for example Ref. 15). To compare the theory with the data quantitatively, one should use mirrors with back surfaces that are wedged and (or) antireflection coated.

The fact that the rotational Raman shift is considerably smaller than the vibrational transition (586.9 versus 4155 cm^{-1} in H_2) can be advantageous. First, for the rotational Raman transition, the bandwidth of a single-wavelength mirror coating can be sufficient to cover both the pump and Stokes wavelengths, whereas the double-wavelength coating required for the vibrational cw Raman laser is far more expensive and difficult to manufacture. Second, the continuous tuning range of the Stokes emission near four times threshold is $\Gamma(\lambda_s/\lambda_p - 1)^{-1}$,[3] and thus a rotational Raman laser can have a much wider continuous tuning range (>21 GHz for the 792–830-nm transition if the cavity length change has no physical limitation) than a vibrational Raman laser (~1 GHz for the 792–1180-nm transition). Third, the thermo-optic effects caused by heat deposition in the Raman gas are less of a problem for the rotational Raman transition because of its smaller photon energy shift, whereas it has been observed that in the high-power vibrational Raman laser thermo-optic effects can lower the conversion efficiency and cause instabilities.[16,17]

In summary, we have reported a diode-pumped cw Raman laser with rotational Stokes emission. In many fields, such as laser spectroscopy and atomic physics, a small shift of an existing laser wavelength is often necessary to match the preferred wavelength exactly. The rotational cw Raman laser provides a new technique that is useful in such applications.

This material is based on work supported by the National Science Foundation under grant 0097222. J. Carlsten's e-mail address is carlsten@physics.montana.edu.

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