CVD-diamond external cavity Raman laser at 573 nm

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Abstract: Recent progress in diamond growth via chemical vapor deposition (CVD) has enabled the manufacture of single crystal samples of sufficient size and quality for realizing Raman laser devices. Here we report an external cavity CVD-diamond Raman laser pumped by a Q-switched 532 nm laser. In the investigated configuration, the dominant output coupling was by reflection loss at the diamond’s uncoated Brewster angle facets caused by the crystal’s inherent birefringence. Output pulses of wavelength 573 nm with a combined energy of 0.3 mJ were obtained with a slope efficiency of conversion of up to 22%.

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References and links

Solid-state Raman lasers are currently receiving much attention as important sources of pulsed and continuous output at particular wavelengths regions in IR, visible and ultraviolet (see eg. [1-6]). Diamond has the potential to radically alter Raman laser design and performance owing to its combination of broad optical transmission, high Raman gain, and high thermal conductivity [7]. Its extraordinary properties are also of intense interest for integrated monolithic diamond photonic devices and within the last decade promising first demonstrations of waveguiding [8], photonic crystal devices [9], radiation detection [10,11], and photon sources [12,13] have been made. Laser action in diamond has been discussed in several papers [14-16], however the difficulties in fabricating large, high-purity and quality, single crystals of consistent properties have restricted development until now. The recent progress in epitaxial growth of diamond by chemical vapour deposition (CVD) [17] has created an exciting opportunity to realize a range of advanced Raman laser devices. Diamond’s outstanding Raman properties result from its high-order lattice symmetry and strong covalent bonding. The first-order Raman spectrum consists of a single peak at 1332.5 cm⁻¹ of width as narrow as 2 cm⁻¹ [18], corresponding to a triply degenerate eigenmode of vibration between two interpenetrating face-centred cubic lattices [19].
Raman laser materials, as well as the largest shift. Current alternatives, of which barium nitrate, the metal tungstates (e.g., BaWO₄, KGd(WO₄)₂) and silicon are key examples, have main shifts in the range 520-1050 cm⁻¹ and the Raman gain coefficients are lower than diamond by at least 40%. Undoped diamond is transparent at wavelengths longer than 230 nm, apart from some absorption in the 3-6 μm range, making it suitable for direct generation in ultraviolet and far-infrared regions not well serviced by other materials. Thermal lensing, which is currently an important factor limiting Raman laser output power scaling beyond multi-watt levels, is strongly mitigated in diamond due to the very high thermal conductivity, an order of magnitude higher than silicon and two orders of magnitude higher than other current alternatives, as well as its low thermal expansion coefficient [22]. The well-known fracture strength and damage resistant nature of diamond also makes it an ideal optical material. These properties make diamond of significant practical interest for developing Raman lasers with enhanced output power, compactness, robustness and wavelength reach.

Despite the strong Raman line being first identified in 1930 [23] and stimulated Raman scattering observed in 1963 [24], there have been only a few studies reported since on stimulated Raman scattering in diamond [14, 25-27] and perhaps only one report of a genuine Raman laser [16]. McQuillan et al reported [14] stimulated Raman scattering of a Q-switched single-longitudinal mode ruby laser to multiple Stokes and anti-Stokes orders in a natural diamond plate (the parallel diamond facets provided some weak feedback that was demonstrated to slightly influence the spatial and spectral output characteristics). More recently, stimulated Raman scattering into multiple Stokes orders has been reported for CVD diamond using a picosecond pulsed Nd:YAG pump laser and its second harmonic [25-27]. Though single or double-pass stimulated Raman scattering arrangements may generally lead to high conversion efficiencies (e.g., [28]), these are rarely practical devices as the output beam quality is often very poor and output is often spread over multiple unwanted Stokes and anti-Stokes orders. On the other hand, a resonant cavity placed around a Raman gain medium reduces the threshold for Raman generation and enables the output spectrum and spatial beam properties to be constrained by the cavity optics. Demidovich et al reported [16] first-Stokes generation for a diamond microchip placed intracavity to a passive Q-switched diode-pumped solid-state laser, but very few details of this experiment are available.

A convenient architecture for demonstrating Raman laser performance is the so-called external cavity Raman laser in which the Raman crystal is placed inside a cavity resonant at the Stokes wavelengths and pumped by a separate laser source. The conversion efficiency for such systems based on crystals such as barium nitrate and potassium gadolinium tungstate several centimeters long can be as high as 60% [29,30].

In this paper, we report an external-cavity diamond Raman laser pumped by a commercial frequency-doubled Nd:YAG laser to generate output at the 573 nm first Stokes in the yellow spectral region. We observe conversion to 573 nm with slope efficiency 22%. We have deduced that the surface damage threshold of the diamond sample is currently an important factor limiting the output energy efficiency and approaches for improving performance are discussed.

2. Experiment

The Raman laser consisted of a 5 × 5 × 1.47 mm thick uncoated single crystal (Element Six, Ascot UK) placed at Brewster’s angle (67.5°) within a resonator consisting of an input coupler M1 and end-mirror M2 as shown in Fig. 1. The basic optical properties of the diamond sample, which was grown epitaxially in the [100] direction, including measurements of absorption and substantial depolarization at 1064nm, have been reported in ref [17] (refer Sample #6). Raman laser performance was investigated for the crystal oriented with each pair of facets normal to the beam axis and also the arrangement in Fig. 1. Though stimulated Raman emission was observed in each case, laser action was only observed in the latter arrangement in which the beam traverses the longest dimension of the crystal and losses are minimized for the vertical polarization.
The resonator mirrors were placed as close as practicable to the diamond crystal to form a total cavity length of 15 mm. The reflection values and curvatures of the resonator mirrors M1 and M2 are specified in Table 1. The Raman laser was pumped with 10 ns pulses of 532 nm output from a frequency-doubled Q-switched Nd:YAG laser (HyperYag, Lumonics) at the repetition rate of 10 Hz. The vertically polarized pump beam was compressed to approximately 0.35 mm beam diameter using a telescope and was reflected by M2 to make a double pass through the crystal.

The first and second Stokes output lines at 573 nm and 620 nm were identified using a calibrated fibre spectrometer (USB2000, Ocean Optics). Energy measurements of the output beam transmitted were performed using a joulemeter (ED100, Gentec) and the laser output pulse shapes were made using a fast-silicon photodetector (DET2-SI, Thorlabs) and oscilloscope (TDS3054, Tektronix) combination with 500 MHz detection bandwidth. In addition to the output beam transmitted by M2, we observed two output beams corresponding to the external reflection as indicated in Fig. 1; we could conveniently measure the output characteristics for the upper external reflection. The facet reflected beam carried a significant component of residual 532 nm pump light which was removed from the measurement by recording the difference between the total beam energy (pump + Stokes) and that transmitted through a short-pass filter (high transmission <540 nm).

### Table 1. Input coupler (M1) and end-mirror (M2) reflection characteristics at the pump and Stokes wavelengths

<table>
<thead>
<tr>
<th></th>
<th>R% 532 nm fundamental</th>
<th>R% 572 nm First Stokes</th>
<th>R% 620 nm Second Stokes</th>
<th>Radius of Curvature (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>M1</td>
<td>9</td>
<td>99.7</td>
<td>99.98</td>
<td>-1.0</td>
</tr>
<tr>
<td>M2</td>
<td>99.7</td>
<td>99.5</td>
<td>97.4</td>
<td>-2.0</td>
</tr>
</tbody>
</table>

Threshold for first Stokes generation was observed at the pump energy 1.1 mJ and the output performance was investigated for pump energies up to 2.5 mJ. Raman lasing was confirmed by misalignment of M2 and observing the decrease of laser output below measurement sensitivity. The Raman output pulse energy transmitted by mirror M2 was found to be much lower than that reflected from the upper Brewster facet and was not much above the measurement sensitivity of our pulse energy meter (5 μJ). As shown in Fig. 2, the output Stokes energy from the facet reflection increased linearly with pump energy up to a maximum of 0.05 mJ, or 2.1% of the pump energy. The onset of Stokes generation occurs just prior to the peak of the pump laser pulse and has duration 3.5 ns full width at half maximum as shown by the recorded laser pulse shapes on the inset plot of Fig. 2. The timing of the Stokes pulse is...
typical of that modeled and previously observed for first Stokes generation in external resonators (see eg., [31]).

![Graph showing output energy and efficiency characteristics of the diamond Raman laser.](image1)

**Fig. 2.** The 573 nm output energy and efficiency characteristics of the diamond Raman laser. The output energy is measured for a single beam reflection off the diamond facet. The efficiencies represent the total conversion efficiency of the pump to the 4 facet reflections combined. The inset shows the pulse durations of the incident pump laser and the first Stokes for 2.0mJ input energy.

Output at the second-Stokes at 620 nm was observed visually at the high pump energies, but the output was not stable and the energy consisted of less than 10% of the total output under all pump conditions. The output spectrum transmitted by M2, which shows the fundamental pump wavelength, the first Stokes at 573 nm and the second Stokes at 620 nm, is shown in Fig. 3.

![Graph showing diamond laser output spectrum for operation near maximum energy.](image2)

**Fig. 3.** The diamond laser output spectrum for operation near maximum energy.

The energy reflected from the facet was approximately 5 times that transmitted by M2, thus it is deduced from the measured M2 transmission (0.5% for first Stokes, refer to Table 1) that the output coupling for the facet reflection was in the range 2-3%. The large reflection at
the Brewster-angled facet is attributed to depolarization of the intracavity beam on each transit of the crystal, brought about by residual stress-induced birefringence created during the growth process. For the sample placed between crossed polarizers, the transmission was found to be between 5 and 17.5% (depending upon rotation of the crystal with respect to the analyzer axis) for 1064 nm light traversing the smallest dimension of the crystal [17]. Consequently, the intracavity beams are substantially depolarized on each pass of the crystal and the main loss from the resonator occurs by reflection at the Brewster facets. The observed polarizations of the output beams are consistent with this hypothesis; the polarization of the beam transmitted by M2 is collinear with the pump beam (i.e., p-polarized), whereas the facet reflections are predominantly orthogonally polarized. Note that the rejection of the s-polarized component at Brewster’s angle is very high in diamond ($R_s = 50\%$) owing to its high refractive index ($n = 2.42$).

In order to calculate more precisely the potential efficiency and scaling properties of the diamond Raman laser, a more detailed analysis of the energy output is required. Given that the main mechanism for output coupling is a result of the diamond’s birefringence as described above, the total energy generated in the Stokes beams can be calculated. For Stokes radiation incident on the internal diamond-air facet, the s-polarized component of intensity $I_s$ will be attenuated by $R_s$ with the reflected beam exiting the cavity. The remaining fraction $(1-R_s)$ propagates to a cavity mirror and back to the same diamond facet without changing its state of polarization where a further fraction $R_s$ will be lost from the cavity by reflection of intensity $(1-R_s)$. Thus the ratio of the energy in the external and internal facet reflected beams is

$$\frac{(1-R_s)}{R_s} = 1.2$$

and the total energy outputted by the two internal and two external facet reflections is 6 times the energy measured by the upper external reflection. Note that the $R_s$ value is sensitive to small changes in the actual angle of the diamond so that the uncertainty in $R_s$ is approximately $\pm 10\%$. The conversion efficiency of the pump to the sum of the facet reflections is also plotted in Fig. 2. The total conversion efficiency is 13% and the slope efficiency increases to 22% at the maximum pump energy.

In comparison, external cavity Raman lasers based on much longer crystals (5 cm long) of other material (e.g., barium nitrate and potassium gadolinium tungstate) have a conversion efficiency of 50-60% and slope efficiencies up to 70% [29,30]. The lower efficiency in the present case is primarily attributed to the additional losses expected when operating near threshold. Previous observations show an increase in slope efficiency as a function of input power at pump energies less than twice the threshold [30,32]. This is attributed to, for example, an increasing fraction of the pump beam cross-section reaching threshold for Raman laser action. We therefore expect much higher slope efficiency at higher pump energies. In the present case, the onset of optical surface damage prevented us from investigating peak pump irradiances higher than $\sim 300$ MW/cm$^2$ (note that the actual damage threshold value is likely to be higher due to the superposition of the Stokes field, which often contains fast transients of high peak power). However, much higher efficiencies can be expected by reducing the threshold through either using lower loss resonators, using diamond samples of reduced birefringence or by using longer crystals likely to be soon available. Anti-reflection coated or low-birefringence crystals will also enable standard single-output beams configurations to be realized.

4. Conclusion

In conclusion, we observe conversion of 532 nm pump laser in an external cavity Raman laser using a 5 mm CVD grown diamond crystal, with total conversion efficiency to the first-Stokes of 13%. The results foreshadow development of compact diamond based laser devices at wavelengths and power densities not available in other materials. This represents a significant step in the global effort toward integrated diamond devices, with capabilities not available in other materials and wholly underpinned by the recent progress in diamond synthesis and processing strategies.