

A continuous wave SrMoO₄ Raman laser

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We demonstrate a cw, laser diode-pumped Nd:GdVO₄/SrMoO₄ crystalline Raman laser. First Stokes laser output at 1173.5 nm of 2.18 W was achieved with a diode-to-first Stokes efficiency of 8.7%. With intracavity frequency doubling in LiB₃O₅, 3.1 W of cw yellow emission at 586.8 nm was obtained with a 12.4% diode-to-yellow efficiency. The experimental results show that SrMoO₄ is an excellent stimulated Raman scattering gain material for high-power cw near-IR Stokes and yellow lasers. © 2011 Optical Society of America

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Stimulated Raman scattering (SRS) is a third-order, non-linear optical process that offers a simple and efficient means of frequency conversion [1–4]. In theory, this process can shift the wavelength of all kinds of lasers, and, when combined with frequency doubling, wavelengths spanning the UV to IR can be generated [5]. Such application of SRS and frequency doubling in crystalline materials has led to the generation of cw, solid-state laser sources with multiwatt outputs in the yellow spectral region [6], for which there are numerous applications in medicine, remote sensing, and detection.

Because SRS is an inelastic process, it generates heat within the Raman-active material; thus, the Raman-active material must have favorable thermal properties. In particular, crystalline Raman-active materials should have high thermal conductivity and a low thermo-optic coefficient (dn/dT) and thermal expansion, in addition to high Raman gain coefficients and low optical loss. Vanadates, tungstates, and molybdates, such as YVO₄, GdVO₄, BaWO₄, SrWO₄, KGd(WO₄)₂, KLu(WO₄)₂, CaWO₄, PbMoO₄, and CaMoO₄ have been identified as promising Raman crystals [6–11].

Strontium molybdate (SrMoO₄) has a tetragonal structure with a space group of $I41/a$, belonging to the sheelite family, and the growth of high-quality crystals using the Czochralski method has been well established [12,13]. SrMoO₄ has a Raman gain coefficient (5.7 cm/GW at 886 cm⁻¹ for polarization along the c axis) superior to that of a number of established Raman crystals (e.g., GdVO₄ 4.5 cm/GW) [14,15] and similar thermal expansion (5.9×10^{-6} °C⁻¹ along the a axis) [16], although the thermal conductivity and thermo-optic coefficients have not been reported to the best of our knowledge. Self-Raman laser operation of Nd:SrMoO₄ at the first Stokes wavelength 1163 nm has previously been demonstrated in the pulsed regime at low average powers [14].

Self-Raman laser configurations have advantages in terms of miniaturization and minimizing resonator losses; however, their wavelength options are restricted due to a fixed fundamental wavelength and Raman shift. Also, because of the combined effects of quantum defect and Raman heating, they suffer significant heat loading, which can limit their output power and beam quality. When separate laser and Raman crystals are used, a diversity of output wavelengths can be achieved, and, since

the thermal load is distributed across two crystals, thermal lensing is decreased and resonator stability is improved at higher incident pump powers, resulting in higher Raman laser output power.

In this Letter, we report for the first time a cw Raman laser based on crystalline SrMoO₄ in an intracavity configuration with a diode-pumped Nd:GdVO₄ laser generating the fundamental field for SRS. cw multiwatt output powers have been achieved for first Stokes Raman output and the second harmonic (586.8 nm) generated by intracavity second-harmonic generation (SHG) in lithium triborate (LBO).

The SrMoO₄ Raman crystal used in the experiments was grown at State Key Laboratory of Crystal Materials, Shandong University, using the Czochralski method. An a -axis SrMoO₄ crystal was used as the seed, and the resultant crystal was cut along the a axis (having lowest thermal expansion) with dimensions of 3 mm × 3 mm × 20.5 mm. The two end faces were polished and antireflection (AR) coated at 880, 1063, and 1173 nm. To the best of our knowledge, the thermo-optic coefficient for SrMoO₄ has not previously been reported, yet it is an important property predicting the thermal lensing that may occur in the Raman-active crystal. We used a He–Ne laser (633 nm) and Mach–Zehnder interferometer to observe small changes in optical path length as the crystal was heated from 25 °C to 70 °C and found that there was no net change in optical path length as the crystal temperature varied. Using the measured value for thermal expansion of SrMoO₄, 5.9×10^{-6} °C⁻¹ along the a axis reported in [16], we determined that the crystal had a corresponding thermo-optic coefficient of -11.24×10^{-6} °C⁻¹ ± 0.3 × 10⁻⁶ °C⁻¹ for 633 nm light polarized along the c axis.

The laser resonator used to investigate laser performance at the first Stokes wavelength and intracavity frequency-doubled first Stokes (yellow at 586.5 nm) output is shown in Fig. 1. The pump source used in the experiment was a 25 W, fiber-coupled laser diode (200 μm core diameter, 0.22 NA) with a central emission wavelength of 880 nm. It was focused to a 430 μm spot diameter (63 mrad half-angle divergence) and was incident on a 4 mm × 4 mm × 10 mm, a -cut, 0.3 at.% Nd:GdVO₄ laser crystal, which was AR coated at 880, 1063, and 1173 nm.

The SrMoO₄ crystal was oriented such that the fundamental and first Stokes emissions were polarized along

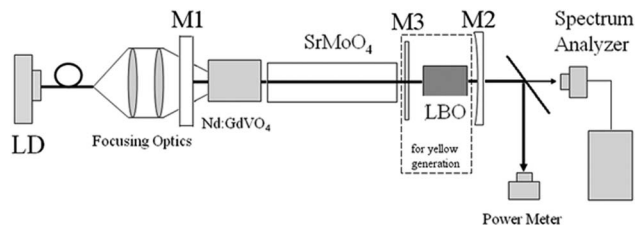


Fig. 1. Experimental configuration of the SrMoO₄ Raman laser.

the *c* axis. The crystal axes were determined by examining the spontaneous Raman spectrum, shown in Fig. 2 for the cases of laser excitation polarized along the *a* and *c* axes. The spectra were obtained using a Raman spectrometer (Enwave, Sense-I-250) with an excitation wavelength of 532 nm, spectral resolution of 6 cm⁻¹, and spectral coverage 250–4000 cm⁻¹.

The input mirror M1 was plano and high transmission (HT) coated at 880 nm and high reflecting (HR; $R > 99.994\%$) at 1063 and 1173 nm. To investigate the first Stokes Raman laser performance, the output coupler (OC) M2 was a concave mirror HR coated at 1063 and 1173 nm ($R = 99.99\%$ at 1063 nm and $R = 99.4\%$ at 1173 nm) with radius of curvature 250 mm. Mirror M3 and the LBO crystal were excluded. To investigate performance at the frequency-doubled first Stokes wavelength, M2 was replaced with a concave mirror, radius of curvature 100 mm, HR coated ($R > 99.994\%$) at 1063 and 1173 nm and HT coated at 586 nm. The mirror M3 was plano and AR coated ($R < 0.06\%$) at 1063 and 1173 nm on the side facing the Nd:GdVO₄ and AR coated at 1063 and 1173 nm and HR coated ($R = 98.7\%$) at 586 nm on the side facing the LBO crystal.

The LBO crystal with dimensions of 4 mm × 4 mm × 10 mm was AR coated for 586, 1063, and 1173 nm. The LBO was cut for noncritical phase matching ($\theta = 90^\circ$, $\varphi = 0^\circ$) and was maintained at a temperature of 44.8 °C for frequency doubling of the first Stokes wavelength. The laser and Raman crystals were wrapped with indium foil, mounted in water-cooled copper blocks, and maintained at a constant temperature of 20 °C.

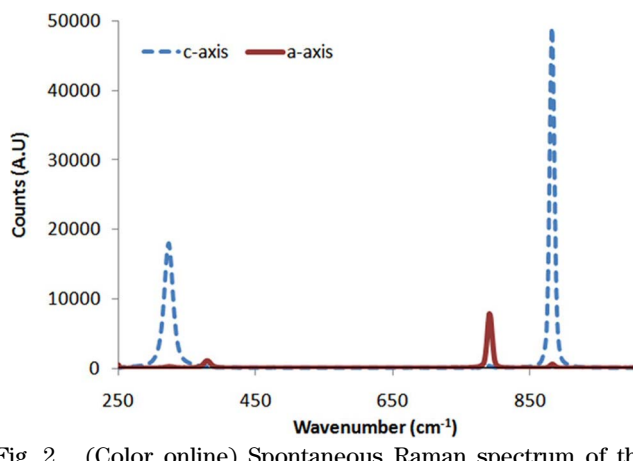


Fig. 2. (Color online) Spontaneous Raman spectrum of the SrMoO₄ crystal for laser excitation at 532 nm, polarized along the *a* and *c* axes.

Raman laser operation at the first Stokes wavelength at 1173.5 nm (corresponding to 886 cm⁻¹ Stokes shift for polarization parallel to the *c* axis of the SrMoO₄ crystal) was achieved with a threshold of 2.4 W. As shown by the data in Fig. 3, a maximum first Stokes power of 2.18 W was obtained for an incident pump power of 25 W, corresponding to an optical (diode to first Stokes) conversion efficiency of 8.8%. The first Stokes power was observed to saturate for incident pump powers greater than 20 W, which also correlated with an observed increase (of 12%) in the far-field diameter of the fundamental mode emitted from the resonator, indicative of strong thermal lensing within the laser and Raman crystals at high incident pump powers [17].

The laser emission spectrum was examined (using a high-resolution Ocean Optics fiber-coupled spectrometer) for any evidence of self-Raman generation in the Nd:GdVO₄ crystal, which would occur at the nearby wavelength of 1173.0 nm and would be in competition with the SrMoO₄ Stokes line. No self-Raman shifting by the Nd:GdVO₄ was observed: this is because of the higher Raman gain in the SrMoO₄ crystal resulting from a higher Raman gain coefficient, longer crystal length, and smaller mode size in the Raman crystal than in the laser crystal. We note that blue emission was observed from the SrMoO₄ crystal when the system was operated above the threshold for first Stokes emission. This emission peaked at 475 nm with a bandwidth of ~10 nm (FWHM); similar blue emission has been previously reported with Raman shifting in BaWO₄ [7], Nd:GdVO₄ [18], and KGW [19] crystals.

The dependence of yellow (586.8 nm) output power generated by intracavity SHG on incident diode pump power is also shown in Fig. 3. The threshold pump power for yellow output was 2 W, lower than for first Stokes operation due to the very high *Q* (at the first Stokes wavelength) of the resonator used for the yellow laser. A maximum yellow output power of 3.1 W was achieved for maximum incident pump power of 25 W, representing an overall diode-to-yellow optical conversion efficiency of 12.4%. We note that the yellow power scaling curve does not rollover as in the case of the first Stokes resonator, and there was no observable increase in the

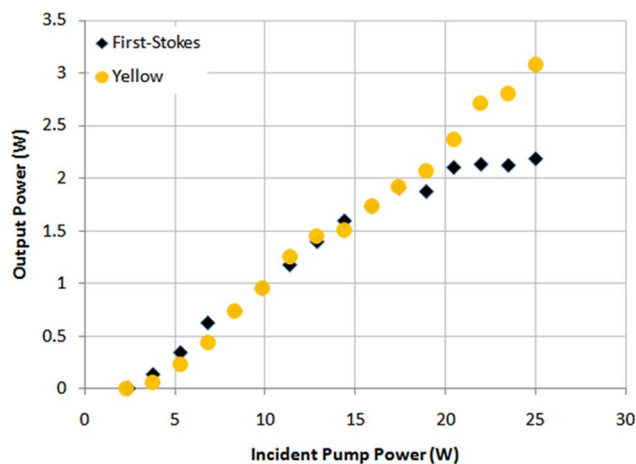


Fig. 3. (Color online) Output power versus incident pump power at the first Stokes and yellow wavelengths.

diameter of the fundamental mode through the OC. This is a result of increased resonator stability offered by the shorter radius of curvature OC. Additionally, as the thermal lens increased, the effective mode diameter in the LBO crystal decreased, which would further improve the SHG conversion efficiency.

It is valuable to compare the diode pump power thresholds and overall conversion efficiencies achieved for the first Stokes and yellow outputs with SrMoO₄ to those results reported for a BaWO₄ Raman laser using near-identical pump and resonator configurations. The SrMoO₄ Raman laser exhibited both a lower first Stokes (2.4 W cf. 2.5 W) and lower yellow threshold (2.0 W cf. 2.5 W) and higher overall optical conversion efficiencies, 8.8% cf. 7.8% at the first Stokes and 12.4% cf. 11% in the yellow, respectively. This was achieved using a substantially shorter Raman crystal length for SrMoO₄ (20.5 mm) than for BaWO₄ (48 mm). These comparisons suggest that the optical losses in the SrMoO₄ crystal are substantially lower than for the BaWO₄ crystal used in the experiments. In the near future, we plan to make *in situ* measurements of resonator loss and Raman gain using the technique described in [20]. There was no evidence of optical damage to the SrMoO₄ crystal within the range of pump powers employed in the current experiments. This, along with the lack of rollover in the yellow output power at maximum pump power, suggests that crystalline SrMoO₄ is well-suited for medium- to high-power (3–5 W) cw yellow generation.

In conclusion, we have reported the first (to our knowledge) cw SrMoO₄ Raman laser operating at the first Stokes and frequency-doubled first Stokes wavelengths of 1173.5 and 586.8 nm, respectively. A maximum first Stokes laser power of 2.18 W was achieved at an 8.7% optical conversion efficiency from the diode pump, and a maximum yellow output power of 3.1 W, limited by the maximum available diode pump power, was obtained at a diode-to-yellow conversion efficiency of 12.4%.

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