Side-pumped crystalline Raman laser

Richard P. Mildren

MQ Photonics Research Centre, Macquarie University, Sydney, NSW, 2109, Australia (rich.mildren@mq.edu.au)

Received September 29, 2010; revised November 19, 2010; accepted November 21, 2010; posted December 14, 2010 (Doc. ID 135870); published January 12, 2011

A crystalline Raman laser is pumped at 90° to the Raman laser axis by a single pass from a line-focused 532 nm pump laser of pulse duration 10 ns. The Raman laser threshold was 6.1 mJ, and at 12 mJ pump energy, a maximum output energy of 2.7 mJ was obtained with a slope efficiency of 46%. The threshold pump intensity is within a factor of 2 of the same device when end-pumped. The results highlight significant potential for coherent beam conversion and combination with enhanced degrees of flexibility and increased power. © 2011 Optical Society of America OCIS codes: 140.3550, 140.3580, 140.7300, 190.5890, 190.2640, 190.5650.

Optical pumping of lasers in directions noncollinear to the laser axis is an established method for increasing output power and design flexibility that has been widely applied to many lasers, such as rare-earth-doped crystal lasers and dye lasers. For these lasers that rely on a population inversion for laser gain, the absorption depth of pump light and the resonator laser mode dimensions must be carefully engineered to ensure adequate energy conversion into the output beam. In contrast, energy transfer in Raman lasers is principally determined by the overlap of pump intensity with the laser mode, thus providing fundamentally different design freedoms and opportunities.

Despite the demonstration [1] of a transverse-pumped Raman laser in nitrobenzene liquid soon after the discovery of stimulated Raman scattering (SRS), investigation of practical non-end-pumped Raman lasers has been very limited. Off-axis pumping of gaseous Raman lasers and amplifiers by excimer lasers has been studied in some detail (e.g., [2]) for small angles (<1°) between the pump and Stokes output beams. Raman crystals have the advantage of much higher gain in approximate proportion to the higher density of scattering centers, in addition to all-solid-state compatibility. Parasitic transverse SRS has been observed in a large-aperture, high-intensity laser system [3,4]. Multipass perpendicularly pumped Raman lasers, in which a Raman crystal is placed inside the cavity of a Q-switched Nd laser, have also been reported [5,6], but only very few details of these studies are available.

Further progress in side-pumped solid-state Raman lasers is needed to address several current technical challenges. Since the absorption depth of Raman material at the pump wavelength is generally very much longer than for inversion lasers, multiaxis pumping is straightforward for an arbitrary crystal shape and dimension, providing a promising approach to coherent beam combination [1,7]. Side pumping also provides an alternative that may enable efficient operation in cases where end-pumping is problematic owing to end-mirror coating constraints or the longer optical path through the medium. For example, diamond Raman lasers [8] are promising sources in the long-wave IR, yet they suffer from the problem of multiphonon absorption at the pump wavelength. A transverse configuration will reduce pump absorption by as much as the ratio of the beam diameter to the Raman material length. To date, laser gain and conversion efficiency of crystalline Raman lasers that involve noncollinear Raman scattering have not been investigated in detail and are not well understood. The present availability of high-gain and high-damage-threshold crystalline Raman materials such as the monoclinic tungstates, tetragonal tungstates, and synthetic diamond provides an interesting opportunity to explore these systems.

Here we investigate a KGd(WO₄)₂ Raman laser pumped by single pass perpendicular to the resonator axis. By comparing the performance with the same laser end-pumped, it is shown that the threshold pump intensities are within a factor of 2 and have similar slope efficiencies.

Although the theory underpinning gain and threshold for SRS was established during the 1960s, there are few detailed treatments of SRS in crystals that deal explicitly with scattering geometry and the associated dynamics of the vibrational wave. Shen and Bloembergen [9] specifically investigated SRS as a function of the optical phonon wavevector. The problem is adequately dealt with classically, since the interaction involves a large ensemble of photons. For near-threshold behavior, the depletion of pump field E_p can be neglected, and it is also assumed that most vibrational centers are in the ground state so that the anti-Stokes wave can be neglected. Coupled equations for the Stokes field E_s ∼ exp[i(k_s⋅r − ω_s t)] and vibrational waves Q_s ∼ exp[i(k_s⋅r − ω_s t)] were obtained using a Lagrangian method

\[
\beta^2 \nabla^2 Q_s + (\omega_0^2 - \omega_s^2 + 2i\omega_s \Gamma) \cdot \nabla \cdot (\nabla \times E_s) + (\epsilon_s \omega_0^2 / c^2) \cdot E_s = \left( N \cdot \frac{d\alpha}{dQ_s} \right) \frac{4\pi \omega_0^2 / c^2}{\beta^2} Q_s \cdot E_p \cdot E_s,
\]

where Q_s is the relative displacement of nuclear positions normalized by \( \sqrt{2}\rho \), \( \rho \) is the reduced mass density, \( \epsilon_s \) is the permittivity of free space of the Stokes wave, and \( c \) is the speed of light. The \( \beta^2 \) term allows for the propagation of crystal momentum where \( \beta \ll \omega_0 / k_s \) is equal to the acoustic phonon speed in the Raman medium. The equations describe a damped harmonic vibration with the driving term E_p⋅E_s and Maxwell’s equation for the Stokes field with the driving

January 15, 2011 / Vol. 36, No. 2 / OPTICS LETTERS 235
term $\mathbf{Q}_p \cdot \mathbf{E}_p$, respectively. The damping constant for the vibrational wave is $\Gamma (= 1/T_2$ where $T_2$ is the phonon dephasing time). The strength of the Raman coupling is $N - d\alpha / dQ$, where $\alpha$ is the optical polarizability tensor and $N$ is the number density of scattering centers, and is related to the steady-state Raman gain coefficient by $g = 2\pi \omega_0^2(N - d\alpha / dQ)/c^2k_0\omega_0 \Gamma$. Conservation of energy requires $\omega_p = \omega + \omega_0$.

From Eqs. (1) and (2), it is seen that the phonon–photon coupling strength is independent of the propagation and depends only on pump and Stokes polarization and the properties of $\alpha$. The only direction dependence comes from the requirement for momentum conservation ($k_p = k_s + k_\gamma$). For 90° scattering, the magnitude of the phonon wavevector is between those seen for forward and backward scattering, as shown in Fig. 1(a). However, it is almost always generally assumed that the phonon wavevector is very small compared to the Brillouin zone boundary, which is a regime where phonon dispersion is low. For the example of Raman backscattering at visible wavelengths, $k_\gamma$ is the order of $10^5$ cm$^{-1}$, or approximately 1% of the zone boundary. As a result, the variation in the resonant frequency of the optical phonon $\omega_0^\gamma = (\omega_0^2 - \beta^2 - k_\gamma^2)^{0.5}$ (see, e.g., [10]) is negligible and phonons of frequency within the Raman linewidth can be generated in momentum-conserving interactions independent of the scattering direction. It has also been suggested that $\Gamma$ is dependent on the phonon wavevector magnitude [9], in which case Raman gain would be affected in the steady-state regime. The author is unaware of any evidence for significant $\Gamma$ dependence on $k_\gamma$, and the effect is assumed negligible. Thus it is concluded that the Raman gain is, to first order, independent of scattering geometry.

The apparatus for the transverse pump Raman laser is depicted in Fig. 1(b). A 1.0 cm high $\times$ 1.0 cm wide $\times$ 2.5 cm long rectangular Kd(WO$_4$)$_2$ prism was placed in a stable resonator of length 3.4 cm. The end facets were antireflection coated at the Stokes wavelengths, whereas the front and back pump faces were uncoated (the input energy data presented herein factor in the front face Fresnel loss). The crystal was aligned so that the $N_m$ crystallo-optic axis was approximately parallel to the pump beam polarization to provide maximum $d\alpha / dQ$ for the 901 cm$^{-1}$ Raman mode. The resonator consisted of a broadband high reflector 530–650 nm and an output coupler highly reflecting at the first Stokes (559 nm) and 70% transmitting at the 589 nm second Stokes output wavelength (both having radii of curvature 20 cm). The 532 nm pump beam was TEM$_{00}$ mode of pulse duration 8 ns and had a measured $M^2$ beam-quality factor of approximately 1.5. The 0.6 cm diameter output beam was expanded in the horizontal direction using a 10× cylindrical telescope. The wings of the pump stripe were clipped using an aperture so that only the central portion of the pump beam is used and that the pump beam illuminates only the central 90% of the crystal length. Clipping ensures that, for the range of pump energies used, the thresholds for crystal damage were not exceeded for the end corners of the prism and the bulk at the most intense region of the line focus. The line focus was formed in the crystal by focusing in the vertical direction using a 4.1 cm focal length cylindrical lens. The length of the pump stripe in the crystal was 2.2 cm. Based on the known beam parameters for the pump, the calculated vertical waist minor radius and Rayleigh range were 5 μm and 100 μm, respectively.

To contrast performance with an end-pumped configuration, the high reflector was exchanged with a dichroic input coupler 92% transmitting at 532 nm and highly reflective at the Stokes wavelengths. The pump beam was focused into the crystal with an f = 50 cm spherical lens [see Fig. 1(c)] to provide a waist radius and Rayleigh range of approximately 55 μm and 3.5 cm, respectively.

The side-pumped Raman laser was investigated for pump energies up to 12 mJ. The Raman resonator was aligned by using the amplified spontaneous Raman scattering observed in the plane of the pump beam when pumping at high pulse energies. First, the high reflector axis was aligned with the pump stripe by maximizing the observed double pass first Stokes SRS signal. The output coupler was then put in place and aligned to maximize second Stokes laser output. Energy conversion of the aligned Raman laser is shown in Fig. 2. The pump threshold for output was 6.2 mJ as defined by the linear fit for pump energies > 6.5 mJ. In comparison, the energy threshold for end pumping is 0.16 mJ, or 39 times lower than side-pumped.

The pump intensities at threshold allow the Raman gain coefficients for end and side pumping to be compared. The growth in the Stokes intensity near threshold is given by $dI = I_s(q \cdot I_p(z) - L)dz$ in each case where the round-trip loss coefficient $L$ is assumed to be fixed. Thus at the measured threshold, the deduced gain coefficient is inversely proportional to the integral.

![Fig. 1](image-url)

Fig. 1. (a) Phase-matching diagrams showing the range of $k_s$ vector magnitudes and directions for forward, backward, and 90° scattering. Schematic of the (b) side-pumped and (c) end-pumped Raman lasers.
of $I_p(z)$ per pass through the crystal. The $\int I_p(z) \cdot dz$ values at threshold are similar to each other within a factor of 2, as shown in Table 1 along with the parameters used to calculate them. The departure from parity expected from theory may be attributed to the invalidity of assumed mode overlap between pump and resonator mode volumes and effects arising from the presence of multiple longitudinal modes in the pump laser. For the side-pumped laser, it should also be noted that the far-field output beam profile is highly asymmetric ($M^2_x/M^2_y \sim 750$, with $M^2_y = 1.8$ where $x$ is the pump direction), which suggests that the seeding of Stokes modes is substantially different to the end-pumped case (for which $M^2_{x,y} < 1.5$).

The transverse Raman laser output energy scales linearly with pump energy with slope 46%, slightly lower than for the end-pumped system (53%). At maximum pump energy the conversion efficiency is 22%, which is more than 100 times more efficient than the previous single-pass side-pumped demonstration for nitrobenzene [1]. Future work at higher pump energies and with improved mode control of the pump beam is expected to enable much higher conversion efficiencies and approach the maximum values seen from end-pumped lasers, i.e., (>50%, e.g., [11,8]). Operation using unstable resonators, or use as power amplifiers, are also interesting directions of further study to achieve high brightness output. The results foreshadow development of a range of efficient coherent beam combiners and novel Raman laser systems.

The author is grateful to H. Rhee and H.-J. Eichler of the Technical University of Berlin for interesting discussions on this topic and acknowledges the support of an Australian Research Council Future Fellowship (FT0990622).

Table 1. Comparison of Threshold Parameters for Side- and End-Pumped Systems

<table>
<thead>
<tr>
<th></th>
<th>Side-Pumped</th>
<th>End-Pumped</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pulse energy [mJ]</td>
<td>6.2</td>
<td>0.12</td>
</tr>
<tr>
<td>Waist dimensions [mm]</td>
<td>0.01 (w) × 22(l)</td>
<td>0.055 radius</td>
</tr>
<tr>
<td>Power density [GW/cm²]</td>
<td>0.36</td>
<td>0.16</td>
</tr>
<tr>
<td>$\int I_p \cdot dz$ [GW/cm]</td>
<td>0.76</td>
<td>0.39</td>
</tr>
</tbody>
</table>

Fig. 2. Output energy as a function of the pump energy transmitted by the pump face of the crystal for the side-pumped and end-pumped (inset) configurations.

References

# Optics Letters

**Table of Contents:**

- ISSN: 0146-9592
- Title: Optics Letters
- Publishing Body: Optical Society of America
- Country: United States
- Status: Active
- Start Year: 1977
- Frequency: Semi-monthly
- Volume Ends: Jan - Dec
- Document Type: Journal; Academic/Scholarly
- Refereed: Yes
- Abstracted/Indexed: Yes
- Media: Print
- Alternate Edition ISSN: 1539-4794
- Size: Standard
- Language: Text in English
- Price: USD 2,650 combined subscription per year domestic to institutions (Print & Online Eds.)
- Subject: PHYSICS - OPTICS
- Dewey #: 535
- LC#: QC350
- CODEN: OPLEDP
- Special Features: Illustrations
- Article Index: Index Available
- Composition: Web, offset
- Editor(s): Alan E Willner (Editor-in-Chief)
- URL: [http://www.opticsinfobase.org/ol/journal/ol/about.cfm](http://www.opticsinfobase.org/ol/journal/ol/about.cfm)

**Description:**
Covers the latest research in optical science, including atmospheric optics, quantum electronics, fourier optics, integrated optics, and fiber optics.

## Additional Title Information

- **Alternate Title:** Medline Abbreviated title: Opt Lett; Abbreviated title: O L

---

Copyright © 2010 ProQuest LLC | Privacy Policy | Terms of Use | Contact Us