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Diode-pumped, actively Q-switched Nd,La:CaNb₂O₆ self-Raman laser at 1,174 nm

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Diode end-pumped Nd,La:CaNb₂O₆ self-Raman laser with acousto-optic Q-switching was successfully demonstrated for the first Stokes wave generation at 1,174 nm. A 1.0 at.% Nd³⁺ and 1.0 at.% La³⁺-doped CaNb₂O₆ crystal in dimensions $3 \times 3 \times 14.3$ mm³ was used as the self-Raman laser crystal. Doping 1 at.% La³⁺ ions into this crystal could subdue the fluorescence quenching caused by cross-relaxation between Nd³⁺ ions and finally improve the laser output performance. Under the incident pump power of 9.9 W, the first Stokes wave at 1,174 nm with a maximum output power up to 928 mW was obtained, with the diode to Stokes conversion efficiency of about 9.4%. The results show that the Nd,La:CaNb₂O₆ is also a promising self-Raman crystal for efficient fundamental and Raman laser operation.

KEYWORDS

self-Raman, solid-state laser, acousto-optic Q-switch, Nd,La:CaNb2O6 crystal, diode pumping

Introduction

As an efficient wavelength conversion technology, stimulated Raman scattering has received extensive attention in recent years. Many solid-state materials have been found as promising Raman gain media for all-solid-state Raman laser design [1–8]. A class of Raman crystals that can be used as excellent laser crystals by doping active ions have been widely concerned, such as vanadate [9–11] and tungstate crystals [12–15]. In addition to these commonly used Raman crystals, calcium niobate (CaNb₂O₆) is one of the excellent Raman crystals with strong Raman gain and can also be used as an excellent laser crystal by doping rare earth ions.

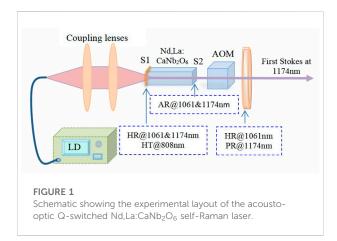
As early as 1963, Ballman et al. grew $CaNb_2O_6$ crystal by the Czochralski technique [16] and observed the laser phenomenon for the first time in $CaNb_2O_6$ doped with Nd^{3+} , Ho^{3+} , Pr^{3+} , Er^{3+} , and Tm^{3+} . In 1969, Kaminskii et al. reported the absorption and luminescence spectra of Nd: $CaNb_2O_6$ [17]. As the rare earth ion-doped $CaNb_2O_6$ crystal can be used as a laser crystal, many research studies on its fundamental laser have been reported [18–22]. In 1977, Husson et al. found that the $CaNb_2O_6$ crystal was also an excellent Raman crystal with a primary Raman shift of 904 cm⁻¹, and the polarized Raman spectra in different directions were measured [23]. In 2004, Silva et al. studied the

growth and characterization of CaNb2O6 single crystal fiber and measured its Raman spectra [24]. In 2009, Cheng et al. measured the thermal and mechanical properties of Yb3+-doped CaNb2O6 crystal, and the results show that it has larger thermal conductivity and specific heat than those of other materials such as Yb:LuVO₄ and Yb:KLu(WO₄)₂ [25]. In the same year, Kaminskii et al. reported the high-order stimulated Raman scattering phenomenon of pure and Nd3+-doped CaNb2O6 crystals [26] and realized Nd:CaNb2O6 self-Raman laser output. The average output power of 85 mW at 1,174 nm first Stokes wave with a 904 $\rm cm^{-1}$ Raman shift was obtained [27]. The abovementioned reports confirmed that the rare earth ion-doped CaNb₂O₆ crystal can be used as a self-Raman crystal and applied to the generation of the new near-infrared laser. However, both the output power and conversion efficiency of the Nd:CaNb₂O₆ crystal self-Raman laser were very low. In 2019, the laser performances of Nd:CaNb2O6 and Nd,La:CaNb2O6 were compared. It was found that Nd3+ ion-doped CaNb2O6 crystal would lead to fluorescence quenching caused by cross-relaxation between Nd³⁺ ions and resulted in low laser efficiency [28]. By doping 1.0 at.% La3+ ions into this crystal could subdue the fluorescence quenching and finally improve the laser output performance. Using this modified crystal, 2.6 W fundamental laser and 310 mW first Stokes laser have been obtained, and both results were much higher than those using Nd:CaNb₂O₆ crystal, respectively. Therefore, the Nd,La:CaNb2O6 crystal has achieved much better performance and higher conversion efficiency. In summary, the excellent Raman characteristics, thermal effect, and physical properties of CaNb₂O₆ crystal make it possible to obtain efficient self-Raman operation by co-doped with Nd³⁺ and La³⁺ ions.

In this article, watt level first Stokes light at 1,174 nm of a diode end-pumped acousto-optic Q-switched Nd,La:CaNb₂O₆ self-Raman laser was successfully demonstrated. The fluorescence quenching caused by cross-relaxation between Nd³⁺ ions was subdued by doping La³⁺ ions into the Nd: CaNb₂O₆ crystal. An average output power of 928 mW was obtained under the incident pump power of 9.9 W, with the diode-to-Stokes conversion efficiency up to 9.4%.

Crystal characteristics of Nd,La: CaNb₂O₆ crystal

CaNb₂O₆ crystal has an orthorhombic symmetry structure with the P_{bcn} space group [16]. A lot of literature has proved that the crystal has excellent optical properties, thermal performance, and a large Raman gain coefficient. Some of the crystals were doped with a certain amount (0.58–2 at. %) of Nd³⁺ to realize laser operation, but Nd³⁺ as an isovalent ion doped into CaNb₂O₆ will form defects such as Nd_{Ca} with a positive charge and V_{Ca}" with a negative charge, leading to the formation of Nd³⁺ clusters, thus cross-relaxation between neodymium ions will take place

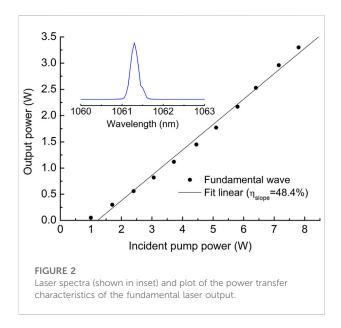


[28]. This phenomenon leads to fluorescence quenching related to the transition from ${}^{4}F_{3/2}$ multiple peaks. Therefore, the improvement of laser power and efficiency is limited. In order to solve this problem, we proposed that La³⁺ ions were doped into Nd:CaNb₂O₆ to form Nd³⁺-La³⁺ buffer clusters, so that more efficient lasers were successfully obtained in co-doped Nd,La: CaNb₂O₆ crystal. Therefore, in order to obtain stable high-power fundamental light and first Stokes light, we used 1.0 at.% Nd³⁺ and 1.0 at.% La³⁺ co-doped CaNb₂O₆ crystal as self-Raman laser crystal in this experiment.

The Raman properties of $CaNb_2O_6$ crystals are similar to those of conventional Raman crystals such as vanadates (YVO₄ and LuVO₄) and tungstates (KLu(WO₄)₂ and KGd(WO₄)₂) [29]. The primary Raman shift of the CaNb₂O₆ crystal is at 904 cm⁻¹ which is similar to that of KGd(WO₄)₂ at 901 cm⁻¹. The fundamental wavelength at 1,061 nm for the Nd:CaNb₂O₆ crystal could be shifted to the first Stokes wavelength at 1,174 nm based on the primary Raman shift of 904 cm⁻¹. The Raman gain at 1,064 nm of KGd(WO₄)₂ is 3.5 cm/GW, whereas it is 2.8 cm/GW for this crystal [27]. However, the thermal conductivity of CaNb₂O₆ crystal is much higher than that of vanadates and tungstates, which is approx. two times that of KGd(WO₄)₂ [25, 29]. The good thermal properties will help in achieving higher laser powers.

Experimental setup

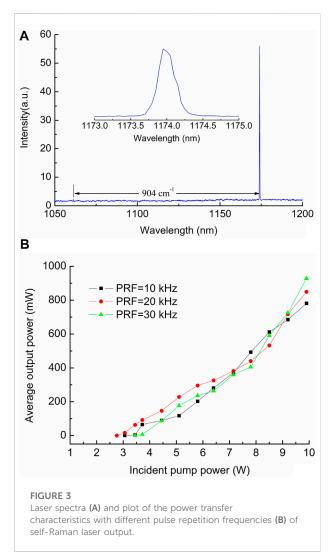
The experimental setup for the acousto-optic Q-switched Nd,La:CaNb₂O₆ self-Raman laser is shown in Figure 1. A fibercoupled laser diode at 808 nm (fiber core diameter of 200 µm and numerical aperture of 0.22) is re-imaged into the laser crystal with a spot diameter of about 320 µm. A c-cut 1.0 at.% Nd³⁺ and 1.0 at.% La³⁺-doped CaNb₂O₆ crystal with the size of $3 \times 3 \times$ 14.3 mm³ was used as the self-Raman laser crystal. The crystal was wrapped in indium foil and mounted on TEC-cooled copper blocks, where the surface temperature was kept at about 20°C,



eliminating the thermal generated in the crystal. In order to make the structure more compact, the input side of the self-Raman crystal was worked as a high-reflective cavity mirror with high reflectivity (HR, R>99.9%) for fundamental wavelength at 1,061 nm and first Stokes wavelength at 1,174 nm, and high transmittance (HT, T>95%) coated for pump wavelength at 808 nm. The other side was anti-reflectivity (AR) coated at both 1,061 nm and 1,174 nm. The output mirror was HR (R> 99.9%) coated at 1,061 nm and a partial reflectivity (PR) coated at 1,174 nm (T = 4.1%). The cavity formed by the input side of the self-Raman crystal and the output mirror was shared for both fundamental light and first Stokes light oscillation. The total cavity length was about 6 cm. An acousto-optic Q-switch modulator (AOM, model QS041-10G-GHI2, Gooch and Housego Co.) was installed between the self-Raman laser crystal and the output mirror to achieve pulsed fundamental laser operation.

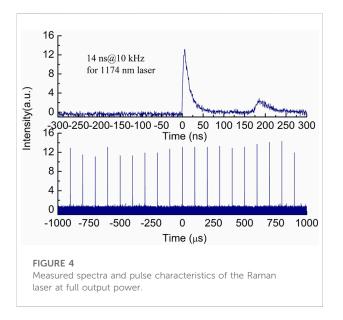
Results and discussion

First, the continuous-wave fundamental laser operation at 1,061 nm was investigated with the use of a compact two-mirror cavity. An efficient 1,061 nm laser was obtained using an output mirror with a transmission of 15%. The output power versus the incident pump power and laser spectra of the fundamental laser measured with the maximum incident pump power fixed at 7.8 W are displayed in Figure 2. The center wavelength of fundamental light was 1,061.3 nm, which was measured by the grating monochromator (model Omni λ -500) with a resolution of 0.05 nm. The threshold of the fundamental light output is around 1 W with respect to incident pump power, and



the output power increases with the increase of the pump power from the laser threshold. The overall linear relationship is well, and the slope efficiency reaches 48.4%. The maximum continuous-wave output power of 3.3 W is finally achieved at the incident pump power of 7.8 W, and the conversion efficiency reaches 42.3%.

Then, the first Stokes laser operation based on self-Raman conversion with the experimental setup as shown in Figure 1 was studied. With optimization of the focused pump beam spot position in the self-Raman crystal and the repetition rate of the acousto-optic Q-switch modulator, the higher average output power for the first Stokes laser at 1,174 nm was obtained under a higher incident pump power of 9.9 W. The laser spectrum of self-Raman output was also measured by the grating monochromator with the results shown in Figure 3A. The spectrum was scanned from 1,050 to 1,200 nm, and only the wavelength of the first Stokes laser was detected. The center wavelength was 1174 nm, and the line width was about 0.25 nm. As it was converted from



the fundamental wave at 1,061.3 nm, the calculated Raman shift was about 904 $\rm cm^{-1}$, which was well fitted with the measured Raman spectra.

Figure 3B shows the relationship between the measured average output power and the incident pump power under different pulse repetition frequencies of 10, 20, and 30 kHz. According to the plot of the power transfer curve, the threshold pump powers were from 2.7 to 3.5 W. As the incident pump power increases, the average output power corresponding to the three repetition frequencies also increases. Under the incident pump power of 9.9 W, a slightly higher average output power of 928 mW was achieved at a pulse repetition frequency of 30 kHz, and the diode-to-Stokes conversion efficiency was 9.4%. As we know, this is the highest first Stokes light output power and conversion efficiency in all studies using the CaNb₂O₆ crystal as a self-Raman laser crystal.

The laser pulse characteristics of the first Stokes output were detected by an InGaAs free-space photodetector and recorded on a 500 MHz oscilloscope (Model DPO3052B). Under the maximum incident pump power of 9.9 W, the narrowest pulse widths were 14, 17, and 19 ns for different pulse repetition frequencies of 10, 20, and 30 kHz, respectively. The pulse energy and peak power were calculated to be about $31\,\mu J$ and $1.63\,kW$ at a full output power of 928 mW, respectively. Though the average output power was only 781 mW for a pulse repetition frequency of 10 kHz, the pulse energy was calculated up to 78 μ J, which was much higher than those of higher pulse repetition frequencies. However, we also see the sub-pulse as the fundamental photons in the cavity re-accumulated and exceeded the Raman threshold again after the principal pulse disappeared at the low pulse repetition frequency. Figure 4

shows the temporal pulse profile and train for the first Stokes laser at the pulse repetition frequency of 10 kHz. The subpulse generated also caused instability in the pulse train. We calculated the amplitude fluctuation up to $\pm 9\%$ at 10 kHz pulse repetition frequency.

Conclusion

In conclusion, we have successfully generated a 1,174nm first Stokes wave with high output power and efficiency in an acousto-optic Q-switched Nd,La:CaNb₂O₆ self-Raman laser. A c-cut Nd,La:CaNb2O6 (1.0 at. % Nd3+ and 1.0 at. % La³⁺ doped) in dimensions $3 \times 3 \times 14.3 \text{ mm}^3$ was used as a self-Raman laser crystal. Nd:CaNb2O6 crystal laser performance was improved by doping La³⁺ ions into this crystal to subdue fluorescence quenching caused by crossrelaxation between Nd³⁺ ions. Under the incident pump power of 9.9 W, the first Stokes light at 1,174 nm with a maximum output power up to 928 mW and peak power of 1.63 kW was obtained at a pulse repetition frequency of 30 kHz, with the diode-to-Stokes conversion efficiency of about 9.4%. As a new self-Raman crystal, the higher Raman laser operation can be expected by further optimizing Nd³⁺ and La³⁺ ion concentration and crystal growth conditions. The results show that the Nd,La:CaNb2O6 is also a promising self-Raman crystal for efficient fundamental and Raman laser operation.

Data availability statement

The original contributions presented in the study are included in the article/Supplementary material; further inquiries can be directed to the corresponding author.

Author contributions

YZ was responsible for the investigation and experiment. WM was responsible for the experiment and writing. XG was responsible for crystal growth and performance analysis. XJ and ZL were responsible for data curation and editing. YH was responsible for the experiment, data curation, and editing. YD was responsible for conceptualization, methodology, writing, and funding acquisition. All authors contributed to the final manuscript.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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