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Multiphonon-assisted continuous-wave tunable vibronic laser in Yb:LuScO$_3$ crystal

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In this work, we demonstrate the phonon-assisted vibronic lasing of a Yb-doped sesquioxide Yb:LuScO$_3$ crystal. The electron–phonon coupling process was analyzed and the Huang-Rhys factor S was calculated to be 0.75 associated with the fluorescence spectrum at room temperature. By a rational cavity design to suppress lasing below 1100 nm, a continuously spectral tunability from 1121 to 1136 nm was realized in a Yb:LuScO$_3$ laser, which represents the longest achievable wavelength in the Yb-doped sesquioxide lasers. Moreover, the Raman spectrum indicated that the E$_g$ phonon mode with a frequency of 472 cm^{-1} was mainly devoted to the phonon-assisted transition process. This work broadens the achievable laser spectrum of Yb-doped sesquioxide, and suggests that the multiphonon–electron coupling strategy should be universal for other laser materials.

Keywords: multiphonon; vibronic laser; Yb-doped sesquioxide.

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1. Introduction

Laser wavelength is a crucial factor for determining particular applications in many fields, such as telecommunications, imaging, and laser processing. Therefore, extending the lasing wavelength with broadband tunability has been a long-time pursuit in laser physics and engineering[1,2]. Especially, lasers operating at the wavelength ranging from 1120 to 1160 nm are in urgent demand for some crucial applications[3,4]. For example, it is a pumping source to obtain a visible blue laser in thulium-doped upconversion fiber[5]. In addition, yellow-orange lasers at 560–580 nm can be generated by frequency doubling, which is a powerful tool for ophthalmic surgery[6,7]. However, direct tunable lasing at 1120–1160 nm is difficult to access in solid-state laser technology due to a few active materials in this range. For Ti:Sapphire and Cr^{3+}:LiSrAlF$_6$ lasers, the longest tuning wavelengths locate around 1110 nm[8]. For $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition of Nd:YAG, the $R_1 \rightarrow Y_6$ branch can generate lasing at 1123 nm, but with poor tunable capacity[9].

Ytterbium (Yb^{3+}) is a common laser gain media emitting at ~1 μm. At present, most of Yb-doped gain media support tunable laser below 1100 nm[10–13], such as Yb:YAG (992.5–1110.8 nm), Yb:YAP (985.6–1052.7 nm), Yb:KY(WO$_4$)$_2$ (1042–1075 nm), and Yb:CaGdAlO$_4$ (999–1089 nm). Moreover, it is possible to extend the lasing wavelengths of Yb-doped materials due to their strong electron–phonon coupling interaction. In recent years, our group has realized a vibronic lasing at 1140 nm in Yb:YCOB crystal[6]. Meanwhile, based on our proposed multiphonon coupling strategy, an unprecedented wavelength tunability (1110–1465 nm) beyond the fluorescence spectrum was also observed in Yb:YCOB crystal[1]. Therefore, there are great opportunities to extend the lasing wavelength beyond 1.1 μm in Yb^{3+}-doped materials.

Sesquioxide RE$_2$O$_3$ (RE = Sc, Y, and Lu) crystals doped with Yb^{3+} ions have high thermal conductivity, wide emission spectrum from large Stark splitting, and good mechanical stability[14]. Therefore, Yb:RE$_2$O$_3$ crystals were applied in high-power continuous-wave (CW) lasers[15] and ultrafast pulse laser generation[16]. In addition, the broadband tunable lasers beyond 1.1 μm were reported in some sesquioxide crystals[17,18], and the longest laser wavelength up to 1134.5 nm has been mentioned in Ref. [17] but without specific output power and efficiency. According to the spectral broadening principle, the structural disorders in mixed sesquioxide (RE = Sc + Lu, Sc + Y, or Lu + Y) can bring strong inhomogeneously spectral broadening and smooth wavelength tunability[19]. For example, the longest CW laser wavelength increased from 1100 nm in Yb:Y$_2$O$_3$ ceramic, to 1112 nm in mixed Yb:(Sc$_{0.51}$Y$_{0.49}$)$_2$O$_3$ ceramic[18]. Therefore, it is possible to obtain a smooth and wide lasing wavelength if we combine spectral broadening and multiphonon coupling strategy into mixed sesquioxide.

In this work, we select Yb:$LuScO_3$ crystal as a possible candidate. First, based on the fluorescence spectrum, we calculated its Huang–Rhys factor S and predicted its fluorescence emission line shape. In addition, by amplifying the three-phonon process and suppressing all few-ones, we realized a tunable vibronic laser from 1121 to 1136 nm by inserting a birefringent filter (BF) into a V-shaped resonant cavity, which is longer than previous laser wavelengths on Yb:$LuScO_3$, e.g., 1010–1100 nm[19]. Finally, the participated phonon modes in the multiphonon coupling process were analyzed and discussed.

2. Results and Discussion

Figure 1(a) displays the fluorescence spectrum of Yb:$LuScO_3$ pumped by an 880 nm laser diode at room temperature. There are four peaks at 976, 1015, 1038, and 1085 nm, corresponding to four sublevels of $^2F_{7/2}$ level, respectively. The zero-phonon line (ZPL) locates at 976 nm. The strongest fluorescence emission locates at 1038 nm, followed by an emission peak at the longest wavelength of 1085 nm. Despite there being no obvious emission peak beyond 1100 nm, there is a long fluorescence trailing due to phonon-assisted transitions. Therefore, the intensity of fluorescence emission extended to 1150 nm is not zero. To verify the electron–phonon coupling strength of Yb:$LuScO_3$ crystal, we calculated the Huang–Rhys factor S according to the following equation[20]:

$$e^{-S} = \frac{I_{ZPL}}{I}, \tag{1}$$

where I_{ZPL} is the fluorescence intensity of ZPL ($n = 0$) at 976 nm and I represents the sum of I_{ZPL} and other phonon-involved processes ($n = 1, 2, 3, \ldots$). The fluorescence intensity in Eq. (1) represents the integrated peak area. By fitting of peaks in the fluorescence spectrum, the S factor of Yb:$LuScO_3$ is calculated to be 0.75, which is smaller than that of the Yb:YCOB crystal ($S = 1.34$)[1]. According to the Huang–Rhys theory, the theoretical emission intensity for various phonon numbers can be predicted. As shown in Fig. 1(b), besides the ZPL at 976 nm, there are a series of fluorescence satellites originating from phonon-assisted electronic transitions. The intensities of phonon sidebands reach the maximum when the involved phonon numbers are equal to S and gradually decrease for larger phonon numbers. For a Yb:$LuScO_3$ crystal, the maximum satellite locates at $n = 1$. In addition, due to the large Stark splitting of Yb:$LuScO_3$, the vibronic emissions with phonon number $n = 1$ and $n = 2$ overlap with electronic transitions, thus leading to a broadband range for tunable lasers in Yb-doped sesquioxide[17,19]. Clearly, in order to obtain direct lasing at longer wavelength beyond 1.1 μm with more phonons ($n \geq 3$), these few-phonon ones and Stark lines below 1100 nm must be suppressed.

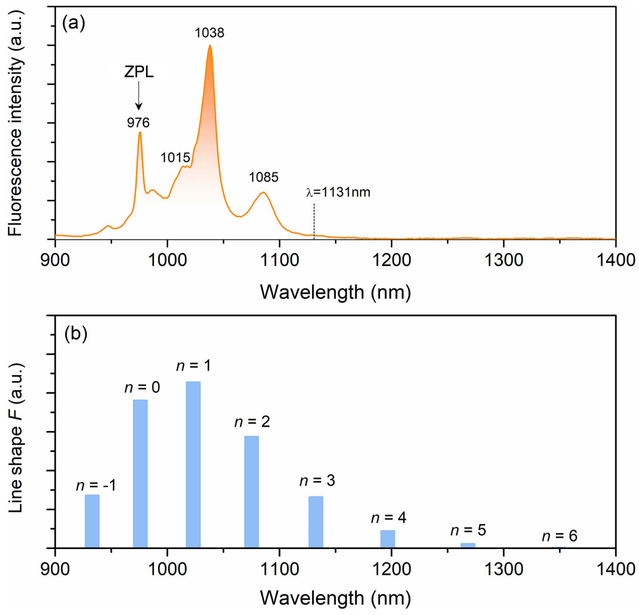

Fig. 1. (a) Fluorescence spectrum of a Yb:$LuScO_3$ crystal; crystal thickness is 1 mm. (b) The calculated fluorescence line shape F for multiphonon-assisted transitions; n represents the phonon number.

In order to realize vibronic lasing at long wavelengths, we designed a special resonant cavity with a judicious choice of mirrors (see Fig. 2). We used a three-mirror V-shaped resonator configuration. The input mirror is a plane mirror with high-reflectance (HR) coating at 1100–1200 nm ($R > 99.9\%$) and high-transmittance (HT) coating at 950–990 nm. In addition, another surface of input mirror is antireflection (AR) coating at 1000–1100 nm to suppress laser oscillations inside the fluorescence spectrum. Both M2 and M3 mirrors with the curvature of 50 mm are HT-coated at 1000–1100 nm and HR-coated at 1120–1390 nm. This design can simultaneously suppress conventional lasing and amplify the weak phonon sideband emissions beyond 1.1 μm. A MgF_2 BF with a thickness of 1 mm is inserted as a tuning element. The output power behind M2 is dominant. The output power behind M3 is very weak and can be ignored. Therefore, we collected the tunable laser power by one of the output beams behind the M2, and the output power in the following tuning experiment was measured by the beam, which was parallel to the pump light.

The 1% (atomic fraction) Yb^{3+}-doped $LuScO_3$ crystal used in this work was fabricated by the optical floating zone (OFZ) method[14]. The heating source was a quad-ellipsoidal reflector focused Xenon lamp with a maximum temperature of 3000°C. The feed rod with dimensions of $\Phi 4 \times 30\,\mathrm{mm}^3$ was heated at

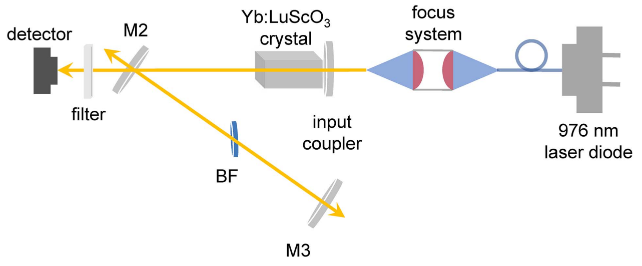

Fig. 2. Experimental setup of the tunable laser beyond 1.1 μm on Yb:$LuScO_3$ crystal.

2400°C. The growth atmosphere was oxygen atmosphere. The Yb:$LuScO_3$ sample for laser experiment had dimensions of 2 mm × 2 mm × 4 mm, and two end faces were precisely polished. To remove the heat produced by the quantum defect effect during the lasing process, Yb:$LuScO_3$ crystal was enclosed by indium foil, then housed in a copper heat sink. The laser crystal was maintained at a constant temperature of 15°C by water-cooling. A fiber-coupled 976 nm laser diode was used for pumping (maximum power was 10 W; the fiber core radius was 52.5 μm and the numerical aperture was 0.22). The pump light was re-imaged and then delivered onto the laser crystal by a focusing lens, whose beam compression ratio was 1:1. The power meter (Newport, Model 1916-R) was placed behind a filter to eliminate the influence of pump light while measuring. Meanwhile, the laser wavelength was measured by a spectrometer (YOKOGAWA, AQ6370C). In addition, Raman spectra of Yb:$LuScO_3$ (also Yb:Lu_2O_3 and Yb:Sc_2O_3) were obtained using a Horiba iHR550 Raman spectrometer with a scanning step size of 0.5 nm and a 633 nm laser excitation source.

Based on this experimental setup, a tunable CW laser generation beyond 1.1 μm was realized in Yb:$LuScO_3$ crystal. A BF was inserted along the Brewster's angle to minimize the loss of the laser oscillation. As shown in Fig. 3, by rotating the angle of the BF, a continuously tunable laser can be obtained from 1121 to 1136 nm, corresponding to an overall tuning range of 15 nm. These wavelengths are longer than the previous result of Yb:$LuScO_3$ (1010–1100 nm)[19], and also longer than other Yb-doped gain media[21–23], such as Yb:LYSO (1030–1111 nm), Yb:$LiYF_4$ (993–1110 nm), and Yb:YAB (1016–1090 nm). During the tuning range, the output power maintains above 5 mW in 1124–1134 nm. The highest output power is located around 1131 nm, with a maximum output power of 20 mW.

According to Fig. 1(b), the spectral range beyond 1.1 μm is attributed to phonon number $n = 3$. To analyze the participating vibrational modes in this multiphonon-assisted lasing, we performed the measurement of the Raman spectrum of Yb:$LuScO_3$ crystal. Meanwhile, the Raman spectra of Yb:Lu_2O_3 and Yb:Sc_2O_3 crystals were also measured for comparison. $LuScO_3$ belongs to the cubic $Ia3$ space group. The irreducible representations for its optical branches are $\Gamma_{op} = 4A_g + 4E_g + 14F_g + 5A_{2u} + 5E_u + 16F_u$[24], where A_g, E_g, and F_g modes are Raman-active and F_u modes are infrared-active. As depicted in Fig. 4, the strongest Raman peak of Yb:$LuScO_3$ has the frequency of 405 cm^{-1} (A_g mode), followed by a relatively weak Raman peak located at 472 cm^{-1} (E_g mode). Compared to the Raman spectrum of Yb:Lu_2O_3 crystal, it can be seen that the Raman peaks of Yb:$LuScO_3$ have a certain degree of high-frequency shift as the increasing component of Sc^{3+} ions, because the atomic mass of Sc^{3+} ions is smaller than that of Lu^{3+} ions, thus leading to the increased vibrational frequency of corresponding phonons. As stated above, the ZPL of Yb:$LuScO_3$ locates at 976 nm. Therefore, the frequency shift of 1131 nm lasing is about 1404 cm^{-1}, which can be assigned to a three-phonon coupling process of 472 cm^{-1}.

In addition, we can see that the Raman peak width of Yb:$LuScO_3$ is much wider than those of Yb:Lu_2O_3 and Yb:Sc_2O_3 crystals owing to the structural disorders in mixed crystal. This inherent phonon broadening would be also favorable for laser tunability beyond 1.1 μm because the multiphonon-assisted lasing is a synergistic effect combining electronic broadening and phonon dispersion concurrently[1]. In our experiments, the output mirrors were coated to suppress laser oscillation at 1086 nm; their transmittance at 1100–1120 nm may not be low enough. Therefore, the shortest tunable wavelength is limited at 1121 nm. If we can improve coating technology, a wider tunable laser range for $n = 3$ can be expected.

Moreover, we used a compact plano–concave cavity to optimize the output power of the CW laser at 1131 nm [see Fig. 5(a)].

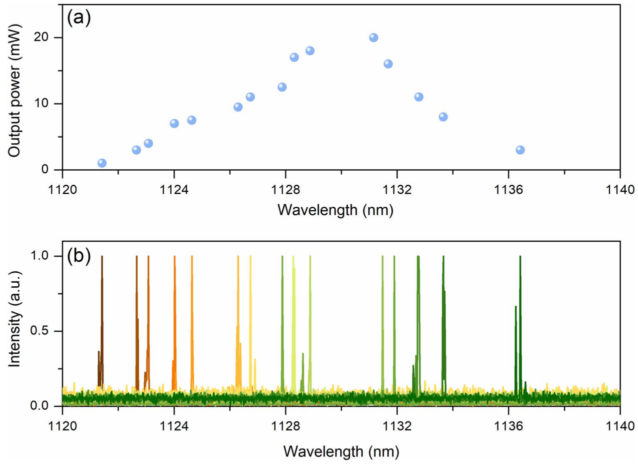

Fig. 3. (a) Wavelength tuning curve for the Yb:$LuScO_3$ crystal at the incident pump power of 5.1 W. The tunable spectrum is discontinuous. (b) Tunable laser spectra in the range of 1121–1136 nm. The bandwidth is narrow below 0.1 nm, which is consistent with the CW laser operation.

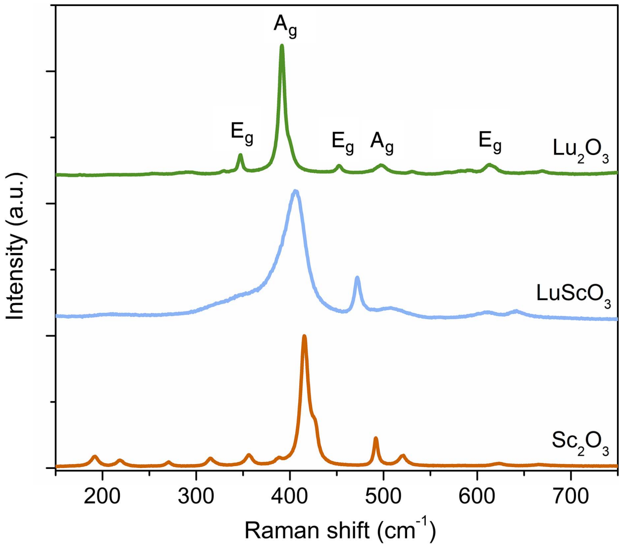

Fig. 4. Raman spectra of Yb^{3+}-doped Lu_2O_3, $LuScO_3$, and Sc_2O_3 crystals. Two main Raman peaks of Yb:$LuScO_3$ crystal are 405 cm^{-1} (A_g mode) and 472 cm^{-1} (E_g mode).

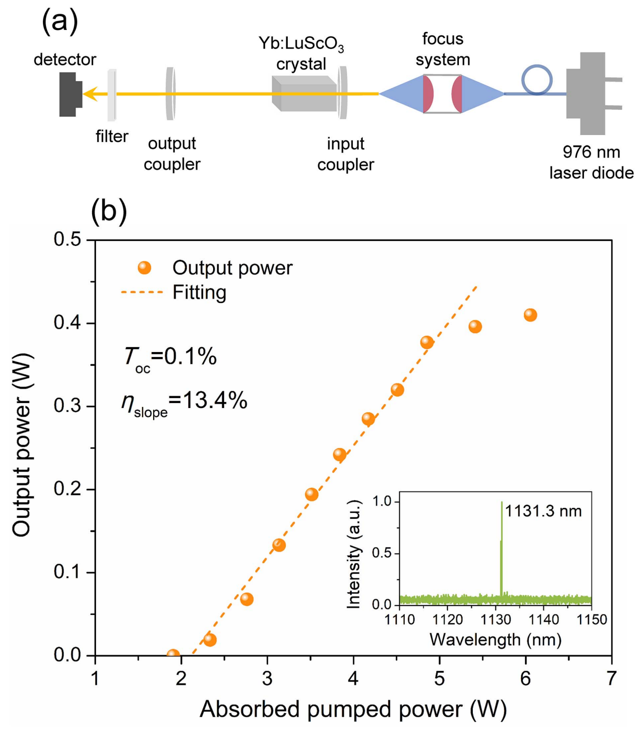

Fig. 5. (a) Experimental setup of the CW laser at 1131 nm; (b) characterizations of laser in the Yb:LuScO$_3$ crystal; output power as a function of absorbed pump power; inset, CW laser spectra of Yb:LuScO$_3$ with T_{oc} = 0.1%.

A concave mirror with HT coating at 1000–1100 nm and partial transmittance at 1120–1200 nm ($T_{oc} = 0.1\%$) is applied as an output coupler. By adjusting the cavity length to be ~51 mm, the pumped spot diameter is 105 μm, and the laser mode in the crystal is calculated to be 92 μm. As depicted in Fig. 5(b), the lasing threshold is 1.9 W. The output power is linearly dependent on the absorbed pump power with a slope efficiency of 13.4% under relatively low absorbed pump power. When the pump power is more than 5 W, the slope efficiency starts to decrease owing to possible thermally induced losses. The maximum output power of laser can reach 410 mW when the absorbed pump power is 6.1 W. In addition, we also tried some output couplers with higher transmittance ($T_{oc} = 1\%$ at 1120–1200 nm) in the laser experiment. However, we were unable to obtain lasing at 1131 nm, but a conventional lasing at 1038 nm. Therefore, in order to obtain high-power lasing beyond 1.1 μm in the Yb:LuScO$_3$ crystal, the lasing process below 1.1 μm must be totally suppressed by a precisely designed cavity.

3. Conclusion

In summary, based on the multiphonon coupling mechanism, we realized a CW laser operating at the fluorescence sidebands in a Yb:LuScO$_3$ crystal. The central wavelength of this vibronic laser was 1131 nm with a maximum output power of 410 mW. In addition, by inserting a BF plate, a continuous tuning from 1121 to 1136 nm was obtained. The participated phonon modes were analyzed and discussed. In this work, we obtained a vibronic lasing with phonon number $n = 3$ by suppressing $n = 1$ and $n = 2$ (as well as Stark lines) in the resonant cavity. In the future, the tunable range and output power are expected to improve with an optimized experimental setup. This work not only represents the longest achievable laser wavelength with Yb-doped sesquioxide but also inspires the study of multiphonon coupling for the broadening of laser wavelengths in other active materials.

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