Lasing of Yb^{3+} in the non-centrosymmetric host $KGd(PO_3)_4$

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Abstract: Crystals of the self-doubling $KGd(PO_3)_4$ were grown with Yb-doping as high as 3.2×10^{20} at/cm^3. They were spectroscopically characterized and laser generation in the 1 μm range was achieved with a slope efficiency exceeding 55%.

OCIS codes: (140.5680) Rare earth and transition metal solid-state lasers; (160.5690) Rare earth doped materials

1. Introduction

Acentric crystal hosts, in which the laser effect and the nonlinear optical process, e.g. frequency doubling, take place simultaneously, are very promising for compact visible lasers because diode-pumped solid-state lasers operate mainly in the near-IR. Laser operation in self-frequency doubling hosts has been demonstrated mainly with Nd-doping, but more recently also several Yb-doped crystals, e.g. $Ca_4Y(BO_3)_3$ (YCOB), $Ca_4Gd(BO_3)_3$ (GdCOB), $YAl_3(BO_3)_4$ (YAB), and $LiNbO_3$ (LNB), were shown to be efficient laser materials. The Yb^{3+}-ion is an alternative to Nd^{3+} in the same wavelength range near 1 μm. However, it has no bands in the green region, so the inevitable reabsorption losses of Nd^{3+} in the wavelength range of the second harmonic can be avoided.

We propose here a new candidate for self-frequency doubling, Yb-doped $KGd(PO_3)_4$ (KGP). The host KGP is a monoclinic acentric crystal with the space group $P2_1$ (point group 2) which exhibits broad transmission window, extending from ≈180 nm to 4 μm, and covering both the fundamental and the second harmonic ranges [1]. Besides the large band-gap, KGP exhibits almost isotropic thermal expansion which is important for the crystal growth, processing of active elements, and during laser operation [2]. Furthermore, KGP is mechanically hard and chemically stable. We report here on spectroscopic results and laser operation near 1 μm of Yb:KGP in the continuous-wave (CW) regime, and show that KGP is phase-matchable for second-harmonic generation (SHG).

2. Crystal growth of Yb-doped KGP

We were able to grow inclusion free single crystals of Yb:KGP, by partial replacement of Gd by Yb, using the top-seeded solution growth (TSSG) slow-cooling technique. The maximum Yb-concentration in the solution was 15 at % which corresponds to ≈7.5 at % in the bulk crystal (≈3.2×10^{20} at/cm^3 measured by electron probe microanalysis). All spectroscopic and laser results presented here were obtained with a single crystal of composition $KYb_{0.024}Gd_{0.976}(PO_3)_4$ corresponding to an Yb-density of ≈1×10^{20} at/cm^3. To avoid impurities, this crystal was grown from its self-flux and the optimized solution composition was Yb_2O_3:Gd_2O_3:K_2O:P_2O_5=0.3:5.7:34:60 mol %. Because of the high viscosity of this solution, the seed holder was equipped with a platinum turbine rotating at 75 rpm. The axial temperature gradient was about 1.2 K/mm and the saturation temperature amounted to 958 K. The temperature was decreased to 12 K below the saturation temperature at a rate of 0.05 K/h. An ***a****-oriented parallelepipedic seed of undoped KGP was used. More details on the growth procedure can be found elsewhere [3].

3. Optical characterization of the KGP host

The orientation of the three principal optical axes of KGP, denoted as N_p, N_m, and N_g, according to the refractive indices $n_p<n_m<n_g$, was determined with respect to the crystallographic frame (***a***, ***b***, ***c***) at a wavelength of 632.8 nm. For a monoclinic crystal, one of the principal optical axes coincides with the ***b*** crystallographic axis; in the case of KGP this is N_p. N_g was found to lie at 37.3° clockwise from the ***c*** crystallographic axis with the positive direction of the ***b*** axis towards the observer. In order to establish if self-frequency doubling is possible in Yb:KGP we measured as a first step the dispersion of the three refractive indices of undoped KGP between 0.45 and 1.2 μm (Fig. 1). As can be seen, the three refractive indices are nearly equidistant. KGP is an optically positive biaxial crystal since the $2V_g$ angle between the two optic axes, lying in the N_p-N_g plane, is 61.3° at 632.8 nm. The experimental data points were fitted using one UV pole and an IR correction term and the Sellmeier equations obtained are included in Fig. 1.

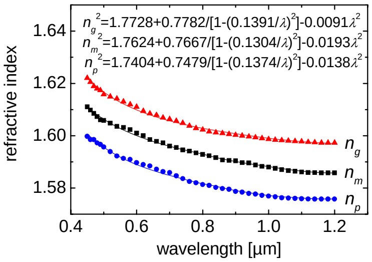

Fig.1: Dispersion of the principal refractive indices of KGP at room temperature: experimental points (symbols) and fits (curves).

Estimations based on these Sellmeier equations indicate that KGP is phase-matchable for type-I SHG near 1 µm both in the ***N$_p$-N$_m$*** (oo-e type interaction) and in the ***N$_m$-N$_g$*** (ee-o type interaction) principal planes. It should be outlined that biaxial crystals have in general greater potential for self-frequency doubling because they offer greater variety of phase-matching configurations. As can be seen in the case of KGP, both oo-e and ee-o phase-matching are possible and their effective nonlinearity is nonvanishing for point group 2 [4]. Thus one has greater freedom to select the polarization of the fundamental in such a way that the laser gain is also maximized.

4. Spectroscopic characterization of Yb:KGP

All further results reported here were based on a single $KYb_{0.024}Gd_{0.976}(PO_3)_4$ sample with dimensions of 2.34, 2.68, and 2.47 mm along the ***N$_p$***, ***N$_m$***, and ***N$_g$*** principal optical axes, respectively. The polarized absorption was measured in the temperature range from 6 to 300 K. The absorption cross-section is maximized for ***E***//***N$_m$***: at 977 nm (zero line transition) it amounts to 1.17×10^{-20} cm^2. The maximum absorption cross sections for ***E***//***N$_g$*** and ***N$_p$***, at the same wavelength, are 0.72×10^{-20} and 0.80×10^{-20} cm^2, respectively. From the low temperature spectra we determined the energies of the three Stark sublevels of the exited state multiplet $^2F_{5/2}$: 10245, 10309, and 10559 cm^{-1}.

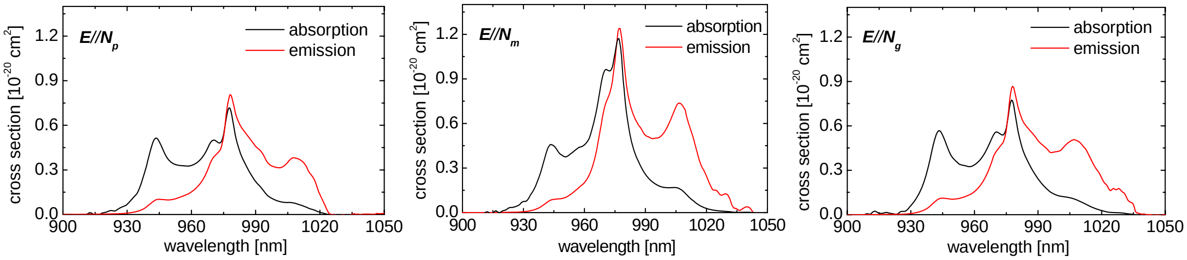

Fig.2: Measured absorption and calculated emission cross-sections of Yb:KGP at room temperature for the three polarizations.

The fluorescence was excited at 940 nm and recorded, both at 300 and 10 K, in a 90º geometry. From the 10 K spectra we determined the four Stark sublevels of the ground state multiplet $^2F_{7/2}$: 0, 125, 298, and 382 cm^{-1}. The reciprocity method was used to compute the emission cross sections σ_e from the absorption cross sections σ_a at room temperature and the level energies (Fig. 2). By averaging the calculated $\sigma_e(\nu)$ over the three polarizations, a radiative lifetime of 1.57 ms was obtained using the Füchtbauer-Ladenburg equation. The fluorescence decay time, measured by the pinhole method, was 1.25 (±0.10) ms. This leads to an intrinsic quantum efficiency of 80%. It can be seen that the oscillation wavelength of Yb:KGP would be rather short: From Fig. 2, lasing can be expected on the $^2F_{5/2}(0')\rightarrow{}^2F_{7/2}(2)$ transition, near 1010 nm. This was confirmed in the following laser experiments.

5. Laser operation of Yb:KGP

A standard astigmatically compensated Z-shaped cavity was used as a laser set-up (Fig. 3). The cavity length was 129 cm. The pumping was in a single pass, by a Ti:sapphire laser (977.1 nm, FWHM<1 nm, max. 2 W). The estimated pump waist in the focus of the f=6.28 cm lens L was ≈30 µm. The resonator contained two curved folding mirrors (M1 and M2) with RC=-10 cm, and two plane reflectors (rear mirror M3 and output coupler M4). The uncoated Yb:KGP sample was attached to a Cu-holder without active cooling and positioned under Brewster angle.

CW laser operation was obtained at room temperature for pumping with ***E***//***N$_m$*** and ***E***//***N$_p$*** (propagation in both cases along ***N$_g$***). The laser always had the same polarization as the pump. No generation was possible for ***E***//***N$_g$***. The input-output characteristics are shown in Fig. 4 against the absorbed pump power P_{abs}. With a maximum incident pump power of ≈2 W (corresponding to P_{abs}=206 mW at 977.1 nm), the maximum output power for $\boldsymbol{E}//\boldsymbol{N}_m$ was 93 mW (T_{oc}=1%). The corresponding slope efficiency was η=53.2%. Pumping with $\boldsymbol{E}//\boldsymbol{N}_p$, the maximum output power reached 72 mW for P_{abs}=294 mW, also with T_{oc}=1%. In this case the slope efficiency was lower, η=31.3%.

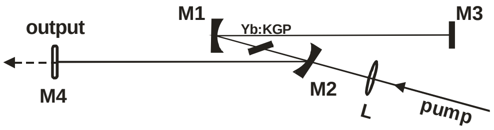

Fig.3: Schematic set-up of the Yb:KGP laser.

The laser thresholds for $\boldsymbol{E}//\boldsymbol{N}_m$ and $\boldsymbol{E}//\boldsymbol{N}_p$ were 51 and 74 mW, respectively, both for T_{oc}=1%. The lower η and higher threshold for $\boldsymbol{E}//\boldsymbol{N}_p$ can be explained by the lower gain. For $\boldsymbol{E}//\boldsymbol{N}_m$ and T_{oc}=3% we obtained η=55.6% while for T_{oc}=5% the output power was only 12 mW and reliable estimation of η was not possible. In the latter case the oscillation wavelength was as short as 1012.1 nm. The shorter λ_L at higher T_{oc} is typical for Yb-lasers and is related to the maximum of the gain curve. In this case we have obviously oscillation on the $^2F_{5/2}(0')\rightarrow{}^2F_{7/2}(2)$ transition.

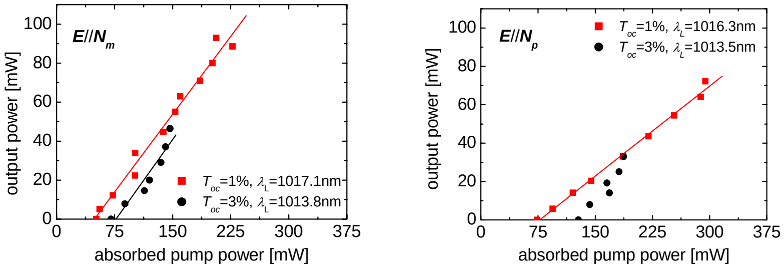

Fig.4: Room temperature CW laser performance of Yb:KGP. Solid lines are fits to the experimental points for estimation of the slope efficiency.

Under lasing conditions the crystal absorption was quite low (≤15%) but almost constant. This is a consequence of the low Yb-density and the relatively low absorption cross-sections. Under non-lasing conditions the absorption was completely bleached at the maximum incident power. However, in the lasing state, the intracavity power in the three-level Yb-system increases the saturation intensity for the pump, balancing the bleaching effect.

6. Conclusion

We successfully grew inclusion free single crystals of Yb:KGP by the TSSG technique. The maximum Yb-density achieved until now is $\approx 3.2\times10^{20}$ cm^3. However, there are indications that it can be increased up to $\approx 1\times10^{21}$ cm^3 [3]. We determined the Stark splitting of the two electronic states of Yb^{3+} from absorption and emission measurements, both at room and low temperatures, and calculated the emission cross sections at room temperature. Lasing has been demonstrated for the first time with $KYb_{0.024}Gd_{0.976}(PO_3)_4$. Although the maximum output power achieved (93 mW) was limited by the available size and doping level, the more than 55% slope efficiency obtained with this first sample is rather promising for the future. Finally, by measuring the dispersion of the refractive indices of KGP, we confirmed that this host possesses phase-matching properties for self-frequency doubling of the Yb-laser.

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