



# Spectroscopic properties and orthogonally polarized dual-wavelength laser of $\text{Yb}^{3+}:\text{NaY}(\text{WO}_4)_2$ crystals with high $\text{Yb}^{3+}$ concentrations



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## ABSTRACT

The optical properties and dual-polarization laser operation of  $\text{Yb}^{3+}:\text{NaY}(\text{WO}_4)_2$  doped with different concentrations of  $\text{Yb}^{3+}$  ions have been detailedly studied in this paper. Single crystals of  $\text{NaY}(\text{WO}_4)_2$  doped with 10 at%, 15 at%, 20 at%, 30 at% and 45 at%  $\text{Yb}^{3+}$  have been grown by the Czochralski method respectively. By comparing their room temperature polarized absorption and emission cross sections, we found that the 10 at%  $\text{Yb}^{3+}:\text{NaY}(\text{WO}_4)_2$  was more appropriate for laser operation. Finally the dual-polarization and dual-wavelength laser properties of 10 at%  $\text{Yb}^{3+}:\text{NaY}(\text{WO}_4)_2$  were measured with the output coupler of  $T = 3\%$ ,  $R_c = 75\%$ . An output of 2.33 W of was achieved, which was calculated by 1000 Hz and 0.3 duty ratio pulse, and its slope efficiency is 41.3%. An output of 1.75 W of orthogonally polarized dual-wavelength laser at 1050 nm and 1055 nm was also obtained.

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## 1. Introduction

$\text{Yb}^{3+}$  ions have the advantageous of simple electronic structure (only two manifolds,  $^2F_{7/2}$  and  $^2F_{5/2}$ ), preventing the parasitic effects as cross relaxation, excited-state absorption up conversion processes, cross, and concentration quenching.  $\text{Yb}^{3+}$  ion doping give the crystals broadband absorption at 0.9  $\mu\text{m}$  that applies to the InGaAs diode direct pumping, and large emission band around 1.0  $\mu\text{m}$  which can support tunable laser and femtosecond pulse generation [1]. These excellent properties attracted considerable attention [2,3], and  $\text{Yb}^{3+}$  has been used for the mode-locking and dual-wavelength laser [4–7], so it has promising prospect for THz wave by frequency difference technique.

Tungstate is an excellent host material for luminescence and solid-state lasers as the result its stable mechanical properties [8–12].  $\text{NaY}(\text{WO}_4)_2$  (NYW) crystal belongs to the tetragonal structure with space group  $I4_1/a$  (no. 88) and its essential structure belongs to the scheelite structure ( $\text{CaWO}_4$  has eight symmetry elements and a body-centered tetragonal primitive cell that includes two formula units) [13],  $\text{Na}^+$  and trivalent  $\text{Ln}^{3+}$  ions occupy the sites of  $\text{Ca}^{2+}$  ions [14,15], making its crystalline structure irregular and beneficial for optical properties. This crystal is congruent melt

(1200 °C), and thus can be grown by Czochralski method [16]. In addition, this tungstate is still stable under high press [17], which provides favorable surroundings to study the properties of activated ions.

The continuous wave and mode-locking laser operations of  $\text{Yb}^{3+}:\text{NYW}$  have been actively investigated [18–21]. These reports are all based on low concentration (<10 at%) of  $\text{Yb}^{3+}:\text{NYW}$ . In our previous study, we found the chemical properties of NYW crystal are very stable that can support high concentration of rare earth ions. But there are seldom reports about the spectroscopic properties with the change in concentration of  $\text{Yb}^{3+}$ -doped and about the orthogonally polarized dual-wavelength laser properties.

In this work, the large single crystals of NYW with different  $\text{Yb}^{3+}$ -doped concentrations have been grown by the Czochralski method. Then the optimal concentration of  $\text{Yb}^{3+}$ -doped in NYW for the laser operation was analyzed by their spectral characteristics. Finally, the 10 at%  $\text{Yb}^{3+}:\text{NYW}$  crystal was chose to test the laser properties, and a dual-polarization laser output at two wavelengths was simultaneously and successfully realized.

## 2. Experimental procedures

### 2.1. Crystal growth

NYW is stable under its melt temperature with tetragonal phase [22], and can be grown by the CZ method. The initial chemicals used were A.R. grade  $\text{Na}_2\text{CO}_3$ ,  $\text{WO}_3$  and 4N  $\text{Yb}_2\text{O}_3$ , 5N  $\text{Y}_2\text{O}_3$ . Firstly we synthesized the  $\text{NaYb}_x\text{Y}_{1-x}(\text{WO}_4)_2$  ( $x = 0.1, 0.15, 0.2, 0.3$  and  $0.45$ ) polycrystals compounds. The initial materials were

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thoroughly mixed and pressed to be tablets. These tablets were put into a platinum (Pt) crucible and slowly heated up to 900 °C for 3 days. Then they were ground, mixed, pressed and heated again to make sure that the chemicals reacted thoroughly.

The polycrystals compounds were placed in the DJL-400 furnace and heated up to a temperature 50 °C higher than the crystallization temperature for about 1 h so as to melt completely and homogeneously. Transparent single crystals with dimensions of  $\Phi 20$  mm  $\times$  30 mm were obtained by using the NYW seed (with the *c*-axis) attached to a Pt rod. During crystal growth, the Pt rod rotated at a rate of 12–15 rpm and the pulling rate was 0.8–1.2 mm h<sup>-1</sup>. When the growth process was over, the crystals was drawn out of the molten and cooled down to room temperature at a rate of 9–45 °C h<sup>-1</sup>. We found that it is hardly to grow single crystal by CZ method when the concentration of Yb<sup>3+</sup> exceeded 30 at%.

Fig. 1a shows the single crystal, we picked out the transparent parts with few defect to process and do test. Fig. 1b shows the 45 at% Yb<sup>3+</sup>:NaY(WO<sub>4</sub>), which is not transparent and without any usable part of crystal. Fig. 1c presents the sample for spectroscopic measurement was cut along a axis with the thickness of 1 mm from the above crystals and the cross section of 10.0  $\times$  10.0 mm<sup>2</sup>. The *a*-cut one is suit for polarization fluorescence-emission and dual-polarization laser operation. Fig. 1d shows the crystal samples used for laser operation with a dimension of 5  $\times$  3  $\times$  3 mm<sup>3</sup>, both end faces of the crystal has polished. Yb<sup>3+</sup> segregation coefficient in NYW crystal is 94.6% [23], therefore the Yb<sup>3+</sup> concentrations in crystals of this paper need to multiply segregation coefficient.

## 2.2. XRD characterization

The XRD pattern of Yb<sup>3+</sup>:NYW crystals recorded by Mini FlexII Desktop X-ray Diffractometer using Cu K $\alpha$  radiation ( $\lambda = 1.54059$  Å). Data were collected in the range of  $2\theta = 15$ –75° with a scan step of 0.02°. The data analyses were carried out with the aid of the JADE 5.0 software package.

## 2.3. Spectroscopic properties

The room temperature polarized absorption spectra of the crystal in this work have been recorded by a Perkin–Elmer UV–VIS–NIR Spectrometer (Lambda-900). The absorption in the wavelength range from 870 nm to 1070 nm. We calculate the absorption cross sections by the following formula [24]:

$$\sigma_{ab} = \alpha / N_c, \quad \alpha = A / L \log e \quad (1)$$

where *A* is absorption intensity, *L* is crystal thickness, *N<sub>c</sub>* is ion concentration.

The room temperature polarized emission spectra of Yb<sup>3+</sup>:NYW single crystals were measured by an Edinburgh Instruments FLS920 spectrophotometer. The corresponding emissions across sections were calculated by fuchtbauer–Ladenburg (F–L) method as the following formula [25–26]:

$$\sigma_{em}(\lambda) = \frac{\lambda^5}{8\pi c n^2} \frac{1}{\tau_r} \frac{I(\lambda)}{\int \lambda I(\lambda) d\lambda} \quad (2)$$

where *n* is refractive index,  $\tau_r$  is radiative lifetime which is 0.480 ms [14],  $\lambda$  is wavelength, *c* is velocity of light.

Depending on the spectroscopic parameters mentioned above, the laser properties of the Yb<sup>3+</sup>-doped materials can be assessed by the gain cross sections according to the following function [27]:

$$\sigma_{ga} = \beta \sigma_{em}(\lambda) - (1 - \beta) \sigma_{ab}(\lambda) \quad (3)$$

where  $\beta$  is the population inversion rate of Yb<sup>3+</sup> ions in crystals,  $\sigma_{ab}$  is the absorption cross sections,  $\sigma_{em}$  is the emission cross sections. The gain cross sections were calculated with several values of  $\beta$  ( $\beta = 0.2, 0.4, 0.6$  and  $0.8$ ).

## 3. Results and discussion

### 3.1. XRD pattern

Fig. 2 shows the obtained XRD pattern of Yb<sup>3+</sup>:NYW crystals. No unused peak was observed. And all diffraction peaks can be indexed as a tetragonal structure with the space group of I4<sub>1</sub>/a. From the XRD pattern it can be concluded that no disintegration and phase change occurred in the crystals.

### 3.2. Absorption spectra

Fig. 3 presents the polarized absorption spectra of Yb<sup>3+</sup>:NYW at room temperature. The absorption band of crystals is in range from 900 nm to 1050 nm, which is the regular absorption of Yb<sup>3+</sup> ions. The main absorption peaks for *E*⊥*c* ( $\sigma$ ) in the spectrum are at 962 nm, 974 nm and 996 nm, while that for *E*||*c* ( $\pi$ ) spectrum are at 935 nm and 974 nm. From Fig. 5 we can see the absorption coefficient for  $\sigma$  polarization is higher than that of  $\pi$  case. The absorption coefficient of the 30 at% Yb<sup>3+</sup>-doped is highest in the four doping concentrations. Then the half maximum broad bandwidth (FWHM) is 45 nm for *E*⊥*c* ( $\sigma$ ), and 55 nm for *E*||*c* ( $\pi$ ).

In Yb<sup>3+</sup>:NYW, the 974 nm peak has the largest absorption cross section calculated by the formula (1), the results are shown in Table 1. Take into account of the Yb<sup>3+</sup> concentration, the absorption cross section of 10 at% Yb<sup>3+</sup> is  $1.83 \times 10^{-20}$  cm<sup>2</sup> and is higher than it of Yb<sup>3+</sup>:YAG, which is  $0.8 \times 10^{-20}$  cm<sup>2</sup> [28]. We also compared it with other laser crystals in Table 2, and found Yb<sup>3+</sup>:NYW has a great advantage.

### 3.3. Emission spectra

The room temperature polarized emission spectra of Yb<sup>3+</sup>:NYW excited by 976 nm are shown in Fig. 4. The emission band ranges from 950 nm to 1050 nm, and the main emission peak is centered at about 1026 nm. The emission intensity increases with the increase of Yb<sup>3+</sup>-doped concentration. We can find the wavelength of strongest peak between *E*||*c* ( $\pi$ ) and *E*⊥*c* ( $\sigma$ ) is about 5 nm interval, as for the peak of laser spectrum is much narrower, so we

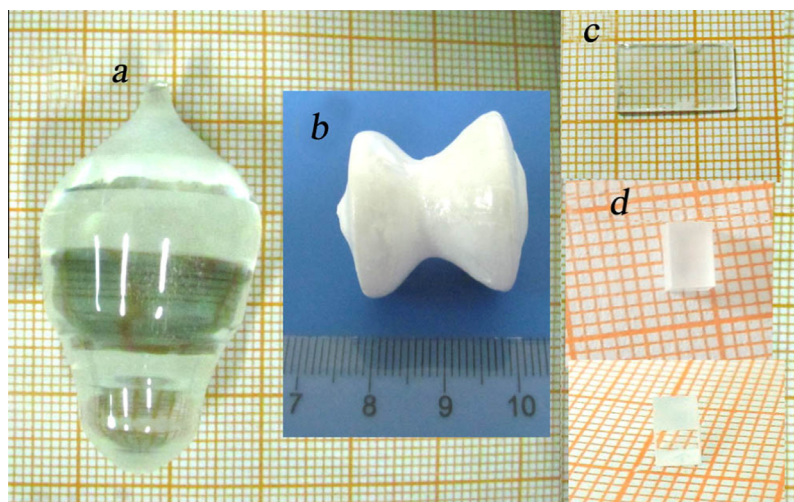


Fig. 1. Single crystal (a), the 45 at% Yb<sup>3+</sup>:NYW (b), crystal sample for optical properties test (c) and sample for laser operation (d).

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