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# Optical spectroscopy of Ho<sup>3+</sup>, Pr<sup>3+</sup> co-doped YScO<sub>3</sub> crystal

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**Abstract:** YScO<sub>3</sub> single crystal co-doped with Ho<sup>3+</sup> and Pr<sup>3+</sup> ions has been successfully grown by the Czochralski method. The absorption spectrum, fluorescence spectrum and fluorescence decay curve were measured at room temperature. The absorption cross section of Ho,Pr:YScO<sub>3</sub> at 649 nm and 1130 nm was calculated to be  $0.93 \times 10^{-20}$  cm<sup>2</sup> and  $0.32 \times 10^{-20}$  cm<sup>2</sup>, respectively. Judd-Ofelt (J-O) theory was performed to evaluate the spontaneous emission probabilities and the radiative lifetimes. Ho,Pr:YScO<sub>3</sub> exhibited strong emission at 2040 nm and 2890 nm with a emission cross section of  $0.80 \times 10^{-21}$  cm<sup>2</sup> and  $1.22 \times 10^{-20}$  cm<sup>2</sup>, respectively. The fluorescence lifetime of the Ho<sup>3+</sup>:<sup>5</sup>I<sub>6</sub> and <sup>5</sup>I<sub>7</sub> levels were calculated to be 0.31 ms and 5.89 ms, respectively. All the results indicate that the Ho,Pr:YScO<sub>3</sub> crystal could be a potential laser material for 2.9 μm laser operation.

**Keywords:** Ho,Pr:YScO<sub>3</sub>; 2.9 μm; Spectroscopic property

## 1. Introduction

Laser sources operating in the 3  $\mu\text{m}$  spectral range have attracted much interest due to their potential applications in biomedical treatments, remote atmosphere sensing and military defense [1-3]. The reason is that this region is overlapped by strong absorption bands of both water and  $\text{CO}_2$  [4]. Moreover, laser around 3  $\mu\text{m}$  wavelength can be used as a pumping source for infrared optical parametric oscillators. Among various active ions ( $\text{Er}^{3+}$ ,  $\text{Dy}^{3+}$  ions, etc.) [5,6],  $\text{Ho}^{3+}$  ion is a promising candidate for 3  $\mu\text{m}$  laser owing to the  $^5\text{I}_6 \rightarrow ^5\text{I}_7$  transition. Unfortunately, the self-terminating transition ( $^5\text{I}_6 \rightarrow ^5\text{I}_7$ ) of  $\text{Ho}^{3+}$  ions, namely, the upper energy level  $^5\text{I}_6$  has a shorter lifetime than the lower energy level  $^5\text{I}_7$  [7], extinguished the probability of population inversion for lasing at around the 3  $\mu\text{m}$  wavelength. Co-doping with  $\text{Pr}^{3+}$  ions is an effective way to depopulate the  $^5\text{I}_7$  multiplet of  $\text{Ho}^{3+}$  for population inversion and practical laser operation [8-13]. Up to now, laser operations have been demonstrated in  $\text{Ho,Pr:LiLuF}_4$  [14,15],  $\text{Ho,Pr:LiYF}_4$  [16,17] and  $\text{Ho,Pr:CaF}_2$  [18].

Mixed sesquioxide  $\text{YScO}_3$  crystallizes in a cubic C (bixbyite) structure, and its disordered structure comes from the random distribution of  $\text{Sc}^{3+}$  and  $\text{Y}^{3+}$  [19,20]. When doped with rare earth ions, it will lead to an inhomogeneous broadening of emission bands which is beneficial for tunable and ultrashort laser operation. Large-size  $\text{YScO}_3$  crystals can be grown by the conventional Czochralski method using iridium crucibles due to the melting point below 2150  $^\circ\text{C}$  [21]. In 2021,  $\text{YScO}_3$  crystals doped with  $\text{Yb}^{3+}$  and  $\text{Er}^{3+}$  have been grown by the Czochralski method and the laser operation of  $\text{Yb:YScO}_3$  and  $\text{Er:YScO}_3$  was demonstrated [22]. In 2022, Suzuki et al. reported a continuous-wave  $\text{Tm:YScO}_3$  laser pumped by 780 nm laser diode, delivering as high as 0.5 W laser output at around 2100 nm with a slope efficiency of 45% [23].

In this work,  $\text{Ho}^{3+}$ ,  $\text{Pr}^{3+}$  co-doped  $\text{YScO}_3$  crystal was grown by the Czochralski method for the first time. Room temperature spectroscopic properties including absorption, emission spectra, Judd–Ofelt analysis and fluorescence lifetime were discussed in detail.

## 2. Experimental

### 2.1. Crystal growth

$\text{Ho,Pr:YScO}_3$  crystal was grown by the Czochralski method. Raw materials of  $\text{Ho}_2\text{O}_3$ ,  $\text{Pr}_6\text{O}_{11}$ ,  $\text{Sc}_2\text{O}_3$ ,  $\text{Y}_2\text{O}_3$ ,  $\text{Al}_2\text{O}_3$  with purity of 99.99% were weighed according to the formula  $(\text{Ho}_{0.005}\text{Pr}_{0.0005}\text{Y}_{0.4945}\text{Sc}_{0.4945})_2\text{O}_3$  and then mixed together for 6 h. The mixture was cold pressed into the form of the block under 200 MPa, and then placed into an alumina crucible, heated up to 1200  $^\circ\text{C}$  and kept at this temperature for 20 h. The charge was then loaded into an iridium crucible for crystal growth. A  $\text{YScO}_3$  crystal was used as the seed. The pulling rate was 1 mm/h, the rotation rate was 10-30 rpm and the growth atmosphere was nitrogen. Finally, the  $\text{Ho,Pr:YScO}_3$  crystal was obtained. Fig. 1 shows the polished samples for spectroscopic and laser experiments. The crystals were greenish and no inclusions and bubbles were observed in the samples.

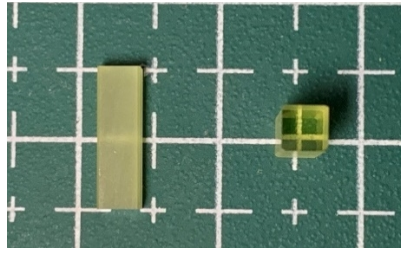


Fig. 1. Photograph of the polished Ho,Pr:YScO<sub>3</sub> crystal.

## 2.2. Spectral measurements

The Ho,Pr:YScO<sub>3</sub> crystal was cut and polished to the dimensions of  $10 \times 10 \times 1$  mm<sup>3</sup> for spectral measurements. The absorption spectrum was carried out on the spectrophotometer (Lambda 950, PerkinElmer UV-VIS-NIR) from 350-2200 nm with 1 nm as a footstep. The fluorescence spectrum and fluorescence decay curve were tested by Edinburgh Fluorescence Spectrophotometer (FLS1000) in the range of 1800-3200 nm under the excitation of 640 nm. All the measurements were performed at room temperature.

## 3. Results and discussion

### 3.1. Optical absorption spectrum

Fig.2 shows the room temperature absorption spectrum of Ho,Pr:YScO<sub>3</sub> crystal in the wavelength range of 350-2100 nm. Obviously there are six absorption bands centered at around 421 nm, 455 nm, 543 nm, 649 nm, 1130 nm and 1940 nm with the full width at half maximum (FWHM) of 9.9 nm, 3.6 nm, 7.6 nm, 16.7 nm, 16.4 nm and 13.2 nm, respectively. The absorption bands are ascribed to the transitions from the ground state  $^5I_8$  to the different excited states:  $^5G_5$ ,  $^5G_6+^5F_1$ ,  $^5S_2+^5F_4$ ,  $^5F_5$ ,  $^5I_6$ , and  $^5I_7$ . The absorption coefficient and absorption cross section at 649 nm was calculated to be  $0.63 \text{ cm}^{-1}$  and  $0.93 \times 10^{-20} \text{ cm}^2$ , respectively. The absorption peak at 1145 nm, corresponding to the  $^5I_8 \rightarrow ^5F_5$  transition of Ho<sup>3+</sup> ions, matches well with the emitting wavelength of well-developed 1150 nm Raman fiber laser. The absorption coefficient and absorption cross section was  $0.20 \text{ cm}^{-1}$  and  $0.27 \times 10^{-20} \text{ cm}^2$ , with FWHM of 14.1 nm. The wide absorption band is mainly caused by the multisite characteristic of YScO<sub>3</sub>, which ensures the steady absorption of the pumping light. Due to the low doping concentration of Pr<sup>3+</sup> ions, the absorption band at around 1500 nm corresponding to the  $^3H_4 \rightarrow ^3F_3+^3F_4$  transition of Pr<sup>3+</sup> ions, could not be observed. While, the  $^3H_4 \rightarrow ^3F_3+^3F_4$  transition usually has the most intense absorption in Pr<sup>3+</sup>-doped oxide crystals [24].

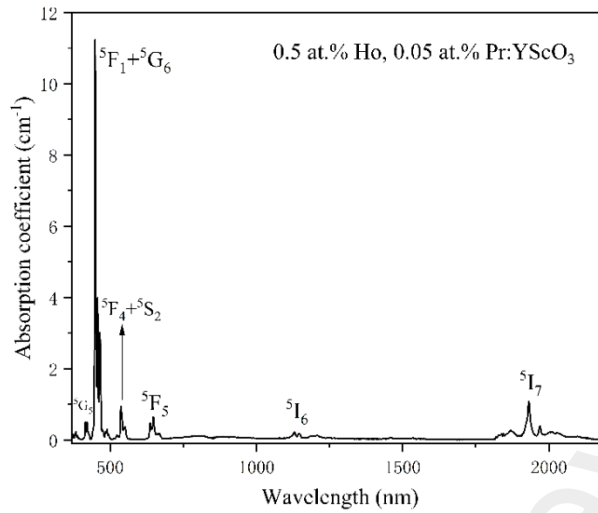


Fig. 2. The absorption spectrum of Ho,Pr:YScO<sub>3</sub> crystal

### 3.2. J-O analysis

The Judd-Ofelt (JO) theory [25,26] was used to analysis the absorption spectrum of Ho,Pr:YScO<sub>3</sub> crystal, and then the fluorescence branching ratios and radiative lifetimes of the excited multiplets can be evaluated. The reduced matrix elements used for absorption and emission transitions can be consulted in [27] and [28], respectively. The average wavelength of different transition, experimental and calculated line strength, and RMS deviation of Ho<sup>3+</sup> ion in YScO<sub>3</sub> crystal are listed in Table 1. The value of rmsΔS is 1.638×10<sup>-20</sup> cm<sup>2</sup>, which indicates a good agreement between the experimental and the calculated line strength.

Table 1. The average wavelength of different transition, experimental and calculated line strength, and RMS deviation of Ho<sup>3+</sup> in YScO<sub>3</sub> crystal.

Excited state (from <sup>5</sup> I <sub>8</sub> )	$\bar{\lambda}$ (nm)	S <sub>mea</sub> (10 <sup>-20</sup> cm <sup>2</sup> )	S <sub>cal</sub> (10 <sup>-20</sup> cm <sup>2</sup> )
( <sup>5</sup> G, <sup>3</sup> G) <sub>5</sub>	419	1.89	3.06
<sup>5</sup> G <sub>6</sub> + <sup>5</sup> F <sub>1</sub>	456	27.87	29.01
<sup>5</sup> F <sub>4</sub> + <sup>5</sup> S <sub>2</sub>	540	3.35	4.20
<sup>5</sup> F <sub>5</sub>	649	2.90	4.16
<sup>5</sup> I <sub>6</sub>	1107	1.22	2.43
<sup>5</sup> I <sub>7</sub>	1956	3.55	5.76
rmsΔS (10 <sup>-20</sup> cm <sup>2</sup> )		1.638	

The values of Ω<sub>2,4,6</sub> were calculated to be 15.63×10<sup>-20</sup> cm<sup>2</sup>, 5.74×10<sup>-20</sup> cm<sup>2</sup>, 3.02×10<sup>-20</sup> cm<sup>2</sup>, respectively. Table 2 shows the J-O parameters of different Ho<sup>3+</sup>,Pr<sup>3+</sup> co-doped crystals. Ω<sub>2</sub> is related to covalency in rare earth doped material. The value Ω<sub>2</sub> of Ho,Pr:YScO<sub>3</sub> is higher than that of Ho,Pr:LiLuF<sub>4</sub>, Ho,Pr:LiYF<sub>4</sub>, Ho,Pr:Sc<sub>2</sub>O<sub>3</sub>, Ho,Pr:Lu<sub>2</sub>O<sub>3</sub> and Ho,Pr:CaGdAlO<sub>4</sub>, indicating that the covalency of Ho,Pr:YScO<sub>3</sub> is larger than that of Ho,Pr:LiLuF<sub>4</sub>, Ho,Pr:LiYF<sub>4</sub>, Ho,Pr:Sc<sub>2</sub>O<sub>3</sub>, Ho,Pr:Lu<sub>2</sub>O<sub>3</sub> and Ho,Pr:CaGdAlO<sub>4</sub>. Ω<sub>4</sub> and Ω<sub>6</sub> are dependent on the host environment, and the ratio of Ω<sub>4</sub>/Ω<sub>6</sub> is used to judge the spectroscopic quality. The value Ω<sub>4</sub>/Ω<sub>6</sub> of Ho,Pr:YScO<sub>3</sub> is 1.90, suggesting that Ho,Pr:YScO<sub>3</sub> crystal is a promising medium for efficient laser operation around 2.9 μm.

Table 2. The J-O intensity parameters of different Ho<sup>3+</sup>, Pr<sup>3+</sup> co-doped crystals.

Crystals	$\Omega_2$ (10 <sup>-20</sup> cm <sup>2</sup> )	$\Omega_4$ (10 <sup>-20</sup> cm <sup>2</sup> )	$\Omega_6$ (10 <sup>-20</sup> cm <sup>2</sup> )	$\Omega_4/\Omega_6$	Ref.
LiLuF <sub>4</sub>	3.25	4.45	3.81	1.17	[8]
LiYF <sub>4</sub>	1.52	1.96	1.05	1.85	[9]
Sc <sub>2</sub> O <sub>3</sub>	6.56	1.67	0.52	3.21	[10]
Lu <sub>2</sub> O <sub>3</sub>	7.10	2.67	0.33	8.09	[11]
CaGdAlO <sub>4</sub>	4.82	6.75	4.91	1.13	[12]
YScO <sub>3</sub>	15.63	5.74	3.02	1.90	This work

Through the calculated intensity parameters, the calculated radiative transition rate, fluorescence branching ratio and radiative lifetimes of Ho<sup>3+</sup> ions in YScO<sub>3</sub> crystal. were given in Table 3. We can see that the branching ratio of the <sup>5</sup>I<sub>6</sub> → <sup>5</sup>I<sub>7</sub> transition is about 13.89%. The radiative lifetime of <sup>5</sup>I<sub>6</sub> energy level was calculated to be 1.70 ms, which is lower than that of Ho:LaF<sub>3</sub> (18.99 ms [29]), Ho,Pr:Sc<sub>2</sub>O<sub>3</sub> (6.20 ms [10]) and Ho,Pr:Lu<sub>2</sub>O<sub>3</sub> (7.97 ms [11]).

Table 3. The calculated radiative transition rates, luminescence branching ratios, and radiative lifetimes of Ho<sup>3+</sup> ions in YScO<sub>3</sub> crystal.

J→	J'	$\lambda$ (nm)	$A_{ED}$ (s <sup>-1</sup> )	$A_{MD}$ (s <sup>-1</sup> )	$A_{(J-J')}$ (s <sup>-1</sup> )	$\beta$ (%)	$\tau$ (ms)
<sup>5</sup> I <sub>7</sub> →	<sup>5</sup> I <sub>8</sub>	2044	211.719	36.0959	247.815	100	4.03
<sup>5</sup> I <sub>6</sub> →	<sup>5</sup> I <sub>8</sub>	1205	505.092	0	505.092	86.110	1.70
	<sup>5</sup> I <sub>7</sub>	2913	59.757	21.7487	81.506	13.890	
<sup>5</sup> I <sub>5</sub> →	<sup>5</sup> I <sub>8</sub>	908	194.185	0	194.185	39.090	2.01
	<sup>5</sup> I <sub>7</sub>	1660	267.465	0	267.465	53.840	
	<sup>5</sup> I <sub>6</sub>	3901	24.472	10.6363	35.108	7.070	
<sup>5</sup> I <sub>4</sub> →	<sup>5</sup> I <sub>8</sub>	752	28.804	0	28.804	9.221	3.20
	<sup>5</sup> I <sub>7</sub>	1223	141.071	0	141.071	45.165	
	<sup>5</sup> I <sub>6</sub>	2153	115.536	0	115.536	36.989	
	<sup>5</sup> I <sub>5</sub>	4578	21.528	5.4059	26.934	8.623	
<sup>5</sup> F <sub>5</sub> →	<sup>5</sup> I <sub>8</sub>	648	6561.315	0	6561.315	75.347	0.11
	<sup>5</sup> I <sub>7</sub>	947	1754.322	0	1754.322	20.145	
	<sup>5</sup> I <sub>6</sub>	1401	368.280	0	368.280	4.229	
	<sup>5</sup> I <sub>5</sub>	2387	23.927	0	23.927	0.274	
	<sup>5</sup> I <sub>4</sub>	4658	0.203	0	0.203	0.002	
<sup>5</sup> S <sub>2</sub> →	<sup>5</sup> I <sub>8</sub>	537	4173.168	0	4173.168	52.037	0.12
	<sup>5</sup> I <sub>7</sub>	727	3034.947	0	3034.947	37.844	
	<sup>5</sup> I <sub>6</sub>	1001	548.799	0	548.799	6.843	
	<sup>5</sup> I <sub>5</sub>	1354	131.288	0	131.288	1.637	
	<sup>5</sup> I <sub>4</sub>	1950	129.063	0	129.063	1.609	
	<sup>5</sup> F <sub>5</sub>	3134	2.270	0	2.270	0.028	

### 3.3. Fluorescence spectrum

Under 640 nm excitation, the fluorescence spectrum of Ho,Pr:YScO<sub>3</sub> was recorded. The stimulated emission cross section can be calculated from the measured fluorescence spectrum via the Füchtbauer Ladenburg (F-L) formula:

$$\sigma_{em}(\lambda)_{JJ'} = \frac{\lambda^5 A(J'' \rightarrow J')}{8\pi c n^2} \frac{I(\lambda)}{\int \lambda I(\lambda) d\lambda}$$

Where  $A(J'' \rightarrow J')$  stands for the radiative transition rates,  $I(\lambda)$  is the emission intensity at the wavelength  $\lambda$ ,  $c$  is the speed of light,  $n$  stands for the refractive index. The spectra of the stimulated emission cross section for the  $^5I_7 \rightarrow ^5I_8$  and  $^5I_6 \rightarrow ^5I_7$  transitions are presented in Fig. 4. The peak emission cross sections at 2040 nm and 2890 nm were calculated to be  $0.80 \times 10^{-20} \text{ cm}^2$  and  $1.22 \times 10^{-20} \text{ cm}^2$ , with a FWHM of 187.0 nm and 197.4 nm, respectively. The value of emission cross section at 2890 nm is lower than that of Ho,Pr:LiLuF<sub>4</sub> ( $1.91 \times 10^{-20} \text{ cm}^2$  at 2944 nm [8]) and Ho,Pr:CaGdAlO<sub>4</sub> ( $2.38 \times 10^{-20} \text{ cm}^2$  at 2860 nm [12]), but higher than that of Ho,Pr:LiLiF<sub>4</sub> ( $0.68 \times 10^{-20} \text{ cm}^2$  at 2950 nm [9]), Ho,Pr:Sc<sub>2</sub>O<sub>3</sub> ( $0.54 \times 10^{-20} \text{ cm}^2$  at 2864 nm [10]), Ho,Pr:Lu<sub>2</sub>O<sub>3</sub> ( $0.42 \times 10^{-20} \text{ cm}^2$  at 2893 nm [11]) and Ho,Pr:CaF<sub>2</sub> ( $1.04 \times 10^{-20} \text{ cm}^2$  at 2851 nm [18]). The FWHMs are larger than those of Ho,Pr:Sc<sub>2</sub>O<sub>3</sub> (159.0 nm at 2110 nm, 144.6 nm at 2864 nm [10]), which displays the Ho,Pr:YScO<sub>3</sub> crystal may be used as a gain medium for tunable and ultrashort pulse lasers.

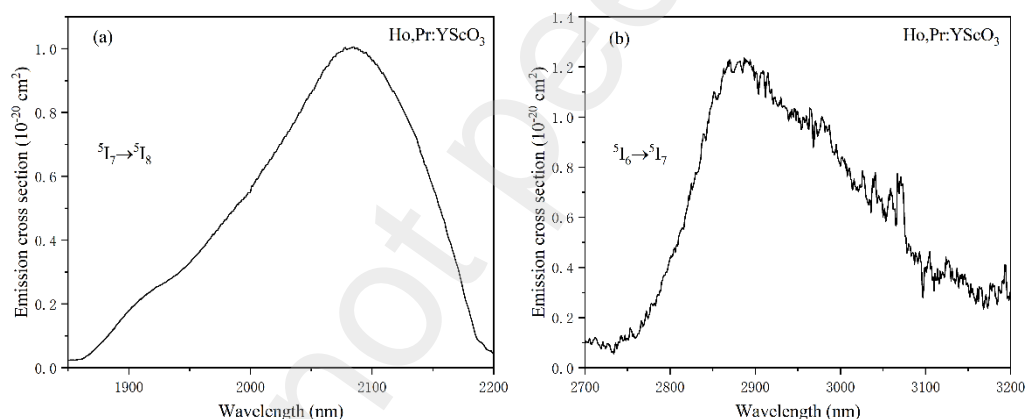


Fig. 4. Emission cross section around (a) 2  $\mu\text{m}$  and (b) 3  $\mu\text{m}$  of the Ho,Pr:YScO<sub>3</sub> crystal.

### 3.4. Fluorescence lifetime

Fig. 5 depicts the fluorescence decay curves of the  $^5I_6$  and  $^5I_7$  multiplets of Ho,Pr:YScO<sub>3</sub> crystal under excitation at 640 nm. The measured decay curves show a single exponential decaying behavior. The fluorescence lifetime of the  $^5I_6$  and  $^5I_7$  levels was fitted to be 0.31 ms and 5.89 ms, leading to a fluorescence quantum efficiency of 7.6% and 54.2%, respectively. The fluorescence of  $^5I_6$  multiplet is much higher than that of Ho,Pr:CaGdAlO<sub>4</sub> (0.194 ms [12]), but lower than that of Ho,Pr:LiLuF<sub>4</sub> (1.47 ms [8]), Ho,Pr:Sc<sub>2</sub>O<sub>3</sub> (1.64 ms [10]), Ho,Pr:Lu<sub>2</sub>O<sub>3</sub> (0.55 ms [11]) and Ho,Pr:CaF<sub>2</sub> (2.73 ms [18]). It is noticed that the fluorescence lifetime of the  $^5I_6$  state is much shorter than that of  $^5I_7$  state in Ho,Pr:YScO<sub>3</sub> crystal, which can cause the 2.9  $\mu\text{m}$  laser transition to self-terminate. Unfortunately, we have not obtained the laser output around 2.9  $\mu\text{m}$  from Ho,Pr:YScO<sub>3</sub> crystal. Pr<sup>3+</sup> ion has been proved to be an effective deactivation ion for  $^5I_7$  level of Ho<sup>3+</sup> ion, population inversion and practical laser

operation could be realized by optimizing the concentration of  $\text{Ho}^{3+}$  and  $\text{Pr}^{3+}$  ions in  $\text{YScO}_3$  crystal.

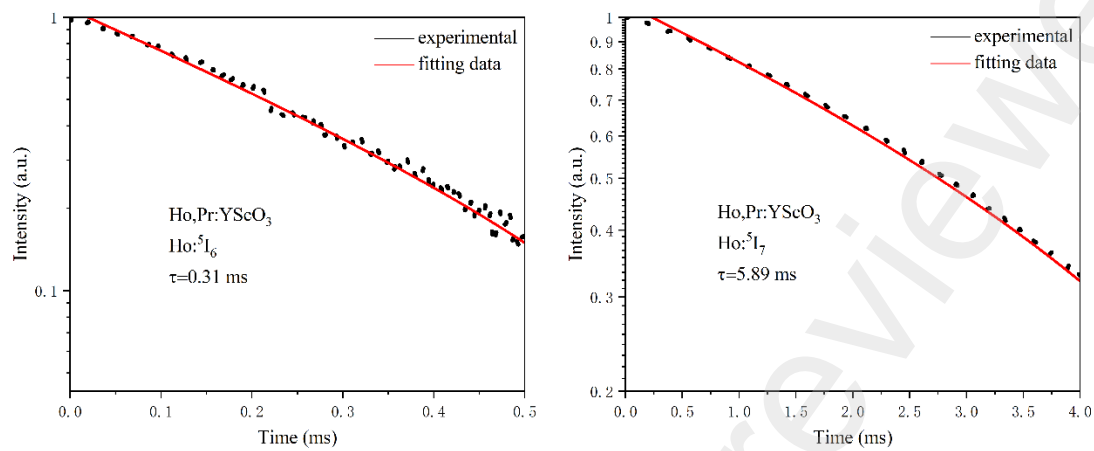


Fig. 5. Fluorescence decay curves of the  $^5\text{I}_6$  and  $^5\text{I}_7$  multiplets of  $\text{Ho,Pr:YScO}_3$  crystal.

#### 4. Conclusion

In summary,  $\text{Ho,Pr:YScO}_3$  crystal has been successfully grown by the Czochralski method. Room temperature absorption spectrum, fluorescence spectrum and fluorescence decay curve were measured and analyzed. The peak absorption cross section of  $\text{Ho,Pr:YScO}_3$  at 649 nm and 1130 nm was calculated to be  $0.93 \times 10^{-20} \text{ cm}^2$  and  $0.32 \times 10^{-20} \text{ cm}^2$ , with a FWHM of 16.7 nm and 16.4 nm, respectively. The intensity parameters  $\Omega_2$ ,  $\Omega_4$  and  $\Omega_6$  were obtained to be  $15.63 \times 10^{-20} \text{ cm}^2$ ,  $5.74 \times 10^{-20} \text{ cm}^2$ ,  $3.02 \times 10^{-20} \text{ cm}^2$ , respectively. The calculated emission cross sections is  $0.80 \times 10^{-20} \text{ cm}^2$  at 2040 nm and  $1.22 \times 10^{-20} \text{ cm}^2$  at 2890 nm, with a FWHM of 187.0 nm and 197.4 nm, respectively. The fluorescence lifetimes of the  $^5\text{I}_6$  and  $^5\text{I}_7$  levels were measured to be 0.31 ms and 5.89 ms. All the results indicate that  $\text{Ho,Pr:YScO}_3$  crystal could be a potential laser material for mid-infrared lasers.

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