

## Optical Parameters of $\text{Nd}^{3+}$ Ion in $\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$ Crystal

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**Abstract:** Spectral properties of  $\text{Nd}^{3+}$  ion in  $\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal were investigated, where the crystal was grown successfully by the Czochralski method. The absorption spectrum in the range of 200–1000 nm was measured. The fluorescence spectrum and lifetime were determined excited with 808nm wavelength, and the emission cross-section was calculated. Based on the Judd-Ofelt theory the spectral parameters were obtained, which were compared with others Nd-doped crystals.

**Key words:**  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal; optical properties; Judd-Ofelt theory

Due to a wide application of the diode pumped solid state lasers, research on more efficient new materials for diode pumped has gained much interest. The good laser materials should have the features of large absorption coefficient and linewidth, as well as large emission cross-section at the emission wavelength. Borate compounds possessing good chemical and physical properties are perfect types of laser gain media. For example,  $\text{YCOB}^{[1]}$ ,  $\text{GdCOB}^{[2]}$ ,  $\text{NGAB}^{[3]}$ , and  $\text{NYAB}^{[4]}$  are all excellent solid-state laser crystals. The rare earth calcium oxoborate  $\text{M}_3\text{Re}_2(\text{BO}_3)_4$  crystal (where  $\text{M}=\text{Ca}$ ,  $\text{Sr}$ ,  $\text{Re}=\text{Y}$ ,  $\text{Gd}$ ,  $\text{La}$ ) is a new host crystal<sup>[5-6]</sup>, which crystallizes in orthorhombic system with  $\text{Pc}2_1\text{n}$  space group. Since  $\text{Nd}^{3+}$ ,  $\text{Yb}^{3+}$  or other rare earth ions can be substituted for  $\text{Re}$  in  $\text{Ca}_3\text{Re}_2(\text{BO}_3)_4$  crystals, these  $\text{Ca}_3\text{Re}_2(\text{BO}_3)_4$  crystals are interesting as laser crystalline materials<sup>[7-14]</sup>. In this paper absorption spectrum, emission spectrum and fluorescence lifetime of  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal were measured. Based on the Judd-Ofelt theory, the investigation of the optical transition probability of  $\text{Nd}^{3+}$ -doped  $\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal was performed.

## 1 Experiment

$\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal was grown by the Czochralski method. The sample with dimensions  $2.1\text{mm}\times 3.5\text{mm}\times 3.0\text{mm}$  was polished for the spectroscopic experiments. The  $\text{Nd}^{3+}$  ion concentration in  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  is 5.12 at%(about  $4.04\times 10^{20}\text{ cm}^{-3}$ ), which was measured by ICP-AES (Ultima2, Jobin Yvon) method.

The absorption spectrum was measured using a Perkin Elmer UV-VIS-NIR (Lambda-35, PerkinElmer) spectrophotometer at room temperature. The fluorescence spectrum of the  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal was measured between 800 nm and 1500 nm using FLS920 spectrometer with a continuous Xe-flash lamp (FLS920, LifeSpec-Ps, Edinburgh) at room temperature.

## 2 Results and Discussion

Figure 1 shows the absorption spectrum in the wavelength from 300 nm to 900 nm at room temperature. The strong  $\text{Nd}^{3+}$  absorption occurs near 352, 524, 583, 749, 808 and 881 nm corresponding to  $4f^3-4f^3$  transition of  $\text{Nd}^{3+}$  ions. The absorption band at 808 nm has an FWHM of 14 nm. The wide absorption band of  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  means that it should be less sensitive to the diode temperature fluctuations in the laser performance. The absorption cross-section  $\sigma_a$  is determined using  $\sigma_a = \alpha/n_c$ , where  $\alpha$  is absorption coefficient and  $N_c$  is the concentration of  $\text{Nd}^{3+}$  ions in  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal which is  $4.04\times 10^{20}/\text{cm}^3$ . Then, the absorption cross-section  $\sigma_a$  is  $3.11\times 10^{-20}\text{ cm}^2$  at 808 nm.

Based on the Judd-Ofelt theory<sup>[15-16]</sup>, the date of absorption spectra can be used to predict the radiative lifetime of the  $^4F_{3/2}$  excited  $J$  manifold and the branching ratios of the fluorescence transition to the lower-lying  $^4I_J$  manifold. The experiment line oscillator strength  $S_{\text{mea}}$  of the transition between ground  $^4I_{9/2}$   $J$  manifold and the ex-

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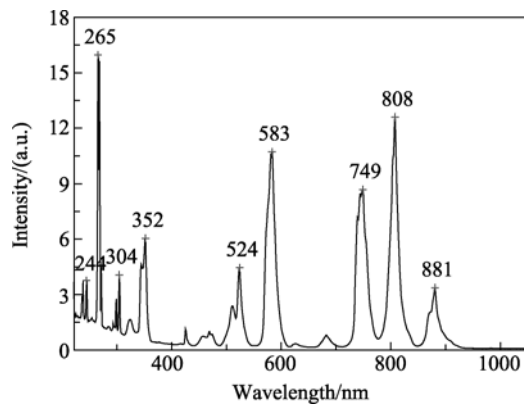


Fig. 1 Absorption spectrum of Nd<sup>3+</sup>:Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> crystal at room temperature

cited  $J$  manifold can be calculated using the following formula<sup>[17]</sup>:

$$\int \sigma(\lambda) d\lambda = \frac{8\pi^3 e^2 \bar{\lambda}}{3ch(2J+1)} \frac{(n^2+2)^2}{9n} S_{\text{mea}} \quad (1)$$

where  $\sigma(\lambda)$  is the absorption cross-section at wavelength  $\lambda$ ,  $e$  is the electron charge,  $\bar{\lambda}$  is the mean wavelength of the absorption band,  $J$  is the total angular momentum of the initial level ( $J=9/2$  for Nd<sup>3+</sup>),  $n$  is the refractive index which is 1.77.

According to the Judd-Ofelt theory, the experiment line

oscillator strength of an electric-dipole transition between the initial  $J$  manifold and the terminal manifold can be expressed as following:

$$S = \sum_{\lambda=2,4,6} \Omega_{\lambda} \left| \langle 4f^n(S, L) J \| U^{(\lambda)} \| 4f^n(S', L') J' \rangle \right|^2 \quad (2)$$

Where  $\|U^{(\lambda)}\|$  is the doubly reduced unit tensor operators calculated by Carnall for Nd<sup>3+</sup><sup>[18]</sup>. According to experiment line oscillator strength  $S_{\text{mea}}$  for different line bands, the intensity parameters  $\Omega_{\lambda}$  can be calculated by least square fitting of Eq. (2) and listed in Table 1. The oscillator strength  $S_{\text{cal}}$  of the ten absorption bands is calculated using these parameters, which are showed in Table 2. The root-mean-square deviation of the experiment and calculation line oscillator strength is defined by:

$$\text{rms}\Delta S = \sqrt{\frac{(S_{\text{mea}} - S_{\text{cal}})^2}{N_{\text{tr}} - N_{\text{par}}}} = 2.293 \times 10^{-21} \text{ cm}^2 \quad (3)$$

Where  $N_{\text{tr}}$  is the number of transitions and  $N_{\text{par}}$  is the number of parameters. A measurement of the relative error of the fit is given by  $\text{rms}\Delta S / \text{rms} = 8.48\%$ .

The radiative transition rate from initial  $J'$  manifold  $\left| (S', L') J' \right\rangle$  and the terminal manifold  $\left| (\bar{S}, \bar{L}) \bar{J} \right\rangle$  is expressed as following<sup>[15-16]</sup>:

**Table 1 Comparison of spectral parameters of Nd<sup>3+</sup>: Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> crystal with other Nd<sup>3+</sup>-doped crystal**

Crystals	$\Omega_2/(\times 10^{-20}, \text{cm}^2)$	$\Omega_4/(\times 10^{-20}, \text{cm}^2)$	$\Omega_6/(\times 10^{-20}, \text{cm}^2)$	$\tau_{\text{rad}}/\mu\text{s}$	$\eta_c/\%$	Ref
5.3 at% Nd: $\beta$ -NLSB	2.85	3.69	4.73	226	49.4	[20]
3 at% Nd: GAB	1.89	4.41	4.14	293	18.7	[3]
3 at% Nd: BYB	0.62	0.90	1.06	1014	6.9	[21]
Nd: KLa(WO <sub>4</sub> ) <sub>2</sub>	3.09	1.04	1.04	685	30.7	[22]
8.2 at% Nd: $\alpha$ -NLSB	3.92	4.41	4.14	225	49.8	[23]
Sr <sub>6</sub> NdSc(BO <sub>3</sub> ) <sub>6</sub>	1.11	2.88	3.09	385	12.5	[24]
1.5% Nd <sup>3+</sup> : YAG	2.71	2.68	5.22	294	91	[19]
1.5% Nd <sup>3+</sup> : YAP	0.69	3.69	4.56	205	88.0	[25]
1.5% Nd <sup>3+</sup> : GSGG	0.35	2.35	3.23	284	88.0	[26]
Nd <sup>3+</sup> : Sr <sub>3</sub> Gd <sub>2</sub> (BO <sub>3</sub> ) <sub>4</sub>	2.436	4.142	5.99	226	23	This work

**Table 2 The measured and calculated line oscillator strength of Nd<sup>3+</sup> ion in Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> crystal**

$J$ Manifold	$\int \sigma(\lambda) d\lambda (\times 10^{-20})$	$\lambda / \text{nm}$	$S_{\text{mea}}/(\times 10^{-20}, \text{cm}^2)$	$S_{\text{cal}}/(\times 10^{-20}, \text{cm}^2)$
$^4F_{3/2}$	1.510	883	1.081	1.279
$^4F_{5/2} + ^2H_{9/2}$	5.439	796	4.317	4.109
$^4S_{3/2} + ^4F_{7/2}$	4.663	748	3.941	4.14
$^4F_{9/2}$	0.1785	684.5	0.1648	0.29
$^4G_{5/2} + ^2G_{7/2}$	4.820	585	5.206	5.236
$^2G_{9/2} + ^2D_{3/2} + ^4G_{7/2} + ^2K_{13/2}$	1.742	516	2.133	1.826
$^4G_{11/2} + ^2G_{9/2} + ^2K_{15/2}$	0.3537	467	0.4786	0.403
$^2P_{1/2} + ^2D_{5/2}$	0.0910	426.5	0.1349	0.152
$^4D_{1/2} + ^4D_{3/2} + ^4D_{5/2}$	1.339	356	2.377	2.477

$$A[{}^4F_{3/2}, (\bar{S}, \bar{L}, \bar{J})] = \frac{64\pi^2 e^2}{3h(2J'+1)\bar{\lambda}^3} \frac{n(n^2+2)^2}{9} \times \quad (4)$$

$$\sum_{\lambda=2,4,6} \left| \left\langle {}^4F_{3/2} \left\| U^{(\lambda)} \right\| (\bar{S}, \bar{L}, \bar{J}) \right\rangle \right|^2$$

Where  $\left| \left\langle {}^4F_{3/2} \left\| U^{(\lambda)} \right\| (\bar{S}, \bar{L}, \bar{J}) \right\rangle \right|^2$  are given in Ref[19]

for the  $\text{Nd}^{3+}$  ion.

The fluorescence branching transition ratios are defined by

$$\beta\{(\bar{S}', \bar{L}')J', (\bar{S}, \bar{L})\bar{J}\} = \frac{A[(\bar{S}', \bar{L}')J', (\bar{S}, \bar{L})\bar{J}]}{\sum_{\bar{S}, \bar{L}, \bar{J}} A[(\bar{S}', \bar{L}')J', (\bar{S}, \bar{L})\bar{J}]} \quad (5)$$

Where the sum is over all possible terminal manifolds  $|(\bar{S}, \bar{L})\bar{J}\rangle$ . The sum represents the total transition probability for radiative decay from the initial manifold. Thus, the fluorescence branching ratios  $\beta_c$  are obtained, which are listed in Table 3. The radiative lifetime is calculated using relation:

$$\tau_{\text{rad}} = \frac{1}{\sum_{\bar{S}, \bar{L}, \bar{J}} A[(\bar{S}', \bar{L}')J', (\bar{S}, \bar{L})\bar{J}]} \quad (6)$$

The radiative quantum efficiency of the  $|(\bar{S}', \bar{L}')J'\rangle$  manifold is defined as

$$\eta_c = \tau_f / \tau_{\text{rad}} \quad (7)$$

Where  $\tau_f$  is the fluorescence lifetime. The measured fluorescence decay curve of the  ${}^4F_{3/2}$  multiplet at room temperature is shown in Fig. 2 in semilog scale. The linear relationship in the figure displays the single exponential behavior of the fluorescence decay. The fluorescence lifetime can be obtained from the slope of the fitting line  $k$ , i.e.,  $\tau_f = -(1/2.303k)$ . By linear fitting, the fluorescence lifetime of the multiplet is about 52  $\mu\text{s}$ . Thus, the radiative lifetime  $\tau_{\text{rad}}$  is 226  $\mu\text{s}$  and the radiative quantum efficiency  $\eta_c$  is 23%. The rate of non-radiative transition in crystals is mainly caused by the effect of concentration quenching and multi-phonon relaxation. In Nd-doped double borate the fluorescence quenching is weak<sup>[7]</sup>. Compared to other crystal matrixes, the phonon energy of borate crystal is relatively higher. Taking the narrow energy gap between  ${}^4F_{3/2}$  and  ${}^4I_{15/2}$  manifold into account (about 5000  $\text{cm}^{-1}$ ), the rate of non-radiative transition of  ${}^4F_{3/2}$  manifold in the crystal becomes higher than that in other crystal hosts. So the multi-phonon relaxation rate is relatively high. As a result, the fluorescence quantum efficiency of  ${}^4F_{3/2}$  manifold in  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal is low.

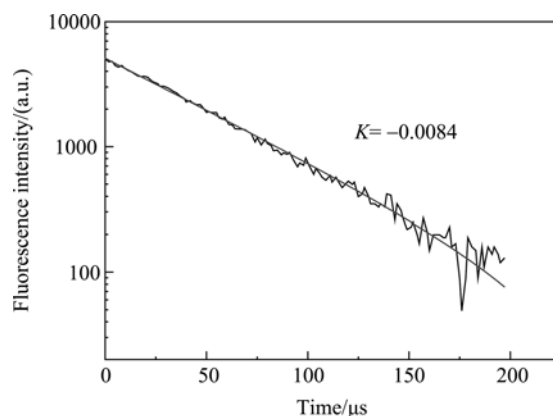


Fig. 2 Room temperature fluorescence decay curve of  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal

Table 3 fluorescence branching ratio  $\beta$  and transition probabilities of  ${}^4F_{3/2} \rightarrow {}^4I_j$

${}^4F_{3/2} \rightarrow (\bar{S}', \bar{L}')J'$	$\lambda/\mu\text{m}$	$A/\text{s}^{-1}$	$\beta_{\text{cal}}$
${}^4F_{3/2} \rightarrow {}^4I_{9/2}$	0.88	1752	0.396
${}^4F_{3/2} \rightarrow {}^4I_{11/2}$	1.06	2137	0.483
${}^4F_{3/2} \rightarrow {}^4I_{13/2}$	1.35	525	0.119
${}^4F_{3/2} \rightarrow {}^4I_{15/2}$	1.88	13.5	0.003

Figure 3 shows the emission spectrum of  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal excited with 808 nm laser radiation. Emission bands corresponding to  ${}^4F_{3/2} \rightarrow {}^4I_j$  transition are observed at 852–945, 1026–1133, 1296–1446 nm regions with peaks at 909, 1064 and 1338 nm, respectively. The emission band FWHM at 1064 nm is about 30 nm, which is much larger than those of  $\text{Nd}^{3+}:\text{YAG}$  (0.8 nm) and  $\text{Nd}^{3+}:\text{YVO}_4$  (3.5 nm) crystals<sup>[27-28]</sup>. The broad FWHM is beneficial to the generation of tunable and ultra-short pulse lasers and high peak power when operating in the Q-switched regime. The emission cross-section of  ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$  was derived using the fluorescence spectrum. For a Lorentz line, the emission cross-section  $\sigma_p$  is related

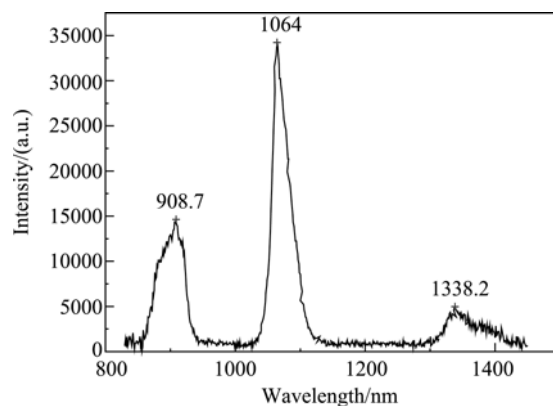


Fig. 3 Emission spectrum of  $\text{Nd}^{3+}:\text{Sr}_3\text{Gd}_2(\text{BO}_3)_4$  crystal at room temperature

to the radiative transition probability by:

$$\sigma_p = \frac{A\lambda_p^2}{4\pi^2 n^2 \Delta\nu} \quad (8)$$

Where  $\Delta\nu$  is the frequency full width at half-maximum,  $\lambda_p$  is the wavelength of the emission peak, and  $A$  is the radiative transition rate. Then, the obtained emission cross-section  $\sigma_p$  (1.06  $\mu\text{m}$ ) is  $1.08 \times 10^{-20} \text{ cm}^2$ . Although small emission cross-section is a drawback for a cw operation, it will favor Q-switched operation because of the enhanced energy storage capacity. The fluorescence lifetime is another crucial parameter affecting the Q-switched pulse energy. The lifetime of  $^4F_{3/2}$  energy of the 5.12 at% Nd<sup>3+</sup>:Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> crystal is 52  $\mu\text{s}$ , which will benefit the generation of pulse with high repetition rate for Q-switched laser.

In comparison with other well-known Nd-doped laser borate, such as NYAB<sup>[29]</sup>, NGAB<sup>[30]</sup> and NAB<sup>[31]</sup>, the value of radiative quantum efficiency is similar. FWHM for absorption at peak wavelength is wider but the emission cross-section is smaller than those of NYAB, NGAB and NAB. The moderate absorption cross-section and the large FWHM indicate that Nd<sup>3+</sup>:Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> crystal is preferable to be pumped by the GaAlAs laser diode(LD) as a novel solid state laser material.

### 3 Conclusion

Spectral parameters of Nd<sup>3+</sup> ions in Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> crystal have been investigated based on the Judd-Ofelt theory. The intensity parameters  $\Omega_\lambda$  are  $\Omega_2=2.436 \times 10^{-20} \text{ cm}^2$ ,  $\Omega_4=4.142 \times 10^{-20} \text{ cm}^2$ ,  $\Omega_6=5.99 \times 10^{-20} \text{ cm}^2$ . The radiative lifetime is 226  $\mu\text{s}$ , and the quantum efficiency is 23%. The fluorescence branch ratios are calculated:  $\beta_1=0.396$ ,  $\beta_2=0.483$ ,  $\beta_3=0.119$ ,  $\beta_4=0.003$ . As compared with other Nd<sup>3+</sup>-doped oxoborate Nd<sup>3+</sup>:YCa<sub>4</sub>O(BO<sub>3</sub>)<sub>3</sub>, Nd<sup>3+</sup>:GdCa<sub>4</sub>O(BO<sub>3</sub>)<sub>3</sub> and Nd<sup>3+</sup>:LaCa<sub>4</sub>O(BO<sub>3</sub>)<sub>3</sub> crystals, Nd<sup>3+</sup>:Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> crystal can be regarded as a novel laser material.

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## Nd<sup>3+</sup>: Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> 晶体的光谱特性研究

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**摘 要:** 用提拉法成功生长出 Nd: Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> 晶体, 并对其光谱性能进行了研究. 测量了晶体在 200~1000nm 波段的吸收谱. 用 808nm 的波长激发, 测量了晶体的荧光光谱和荧光寿命, 计算得到晶体的发射截面. 根据 J-O 理论计算了晶体的光谱参数, 与其它 Nd 掺杂的晶体的光谱参数做了比较和分析.

**关 键 词:** Nd<sup>3+</sup>: Sr<sub>3</sub>Gd<sub>2</sub>(BO<sub>3</sub>)<sub>4</sub> 晶体; 光谱特性; J-O 理论

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