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Growth and spectroscopic properties of Nd^{3+}-doped $Na_2Gd_4(MoO_4)_7$ crystal

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ABSTRACT

This paper reports the growth and spectral properties of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystals. An Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal with dimensions of Ø20 × 25 mm^3 has been grown by the Czochralski method. The spectroscopic properties of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal were investigated. The Judd–Ofelt theory was applied to calculate the spectral parameters. The polarized absorption cross-sections of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal are 4.25×10^{-20} cm^2 with full width at half maximum (FWHM) of 14.6 nm for the π-polarization and 2.87×10^{-20} cm^2 with FWHM of 16.2 nm for the σ-polarization, respectively. The emission cross-sections are 10.0×10^{-20} cm^2 at 1060 nm for π-polarization and 13.6×10^{-20} cm^2 at 1067 nm for σ-polarization, respectively. The fluorescence quantum efficiency has been estimated to be 90.0%. Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal may be considered as a potential laser gain medium for the diode laser pumping.

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1. Introduction

Laser diode pumped solid-state laser plays an important role in a wide variety of applications. Recently, with the rapid development of the solid-state laser with diode laser pumping, research on more efficient new laser materials has gained much interest [1–11]. Double molybdate with general format $MT(MoO_4)_2$ (M = alkali metal; T = rare earth ion) were investigated as new laser host materials [12–14]. A molybdate with general format $M_2T_4(MoO_4)_7$ (M = alkali metal; T = rare earth ion) is another molybdate family [15,16]. $Na_2Gd_4(MoO_4)_7$ crystal is a member of $M_2T_4(MoO_4)_7$ molybdate family. In order to explore new laser materials, this paper reports the synthesis, growth and spectroscopic properties of $Na_2Gd_4(MoO_4)_7$ crystal.

2. Synthesis and crystal growth

In order to obtain the melting point of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal, Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ compound was firstly synthesized by solid state chemical reaction according to the following chemical reaction equation:

$$Na_2CO_3 + \left(2 - \frac{x}{2}\right) Gd_2O_3 + \frac{x}{2}Nd_2O_3 + 7MoO_3 = Na_2Gd_{4-x}Nd_x(MoO_4)_7 + CO_2 \uparrow \quad (1)$$

where x is the concentration of Nd^{3+} ions in Nd^{3+}:$Na_2Gd_4(MoO_4)_7$. The initial substances used were Gd_2O_3 and Nd_2O_3 with purity of 99.99%, Na_2CO_3 and MoO_3 with analytical reagent. The raw materials of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ were accurately weighted. After grinding and extruding to form chunks, the samples were placed in the crucible and heated up to 750 °C, kept at this temperature for 24 h. The process was repeated once again and held at 850 °C for 24 h to assure adequate reaction. The synthesized Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ was checked by powder X-ray diffraction (XRD) using a *D*/max-rA type diffractometer and CuKα radiation (λ = 1.54056 Å) at room temperature in a range of 2θ = 10–65°. The powder X-ray diffraction pattern of the synthesized Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ is shown in Fig. 1(a). The melting point of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ compound was determined using a NETZSCH STA 449C Simultaneous Thermal Analyzer. The result of DSC analysis shows that Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ compound melts congruently at 1194 °C, as shown in Fig. 2.

Since Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal melts congruently at 1194 °C, it was grown by the Czochralski method. Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal was grown in a 2 kHz frequency fur-

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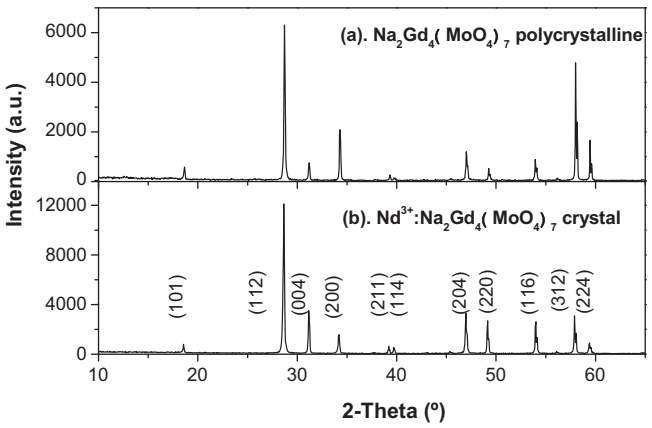

Fig. 1. Powder XRD patterns of (a) $Na_2Gd_4(MoO_4)_7$ polycrystalline and (b) as-grown Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal.

nace in air. The synthesized raw materials of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ with 3 at.% Nd^{3+} were molted in a platinum crucible with dimensions of Ø45 × 40 mm^3, where 1 wt.% excess amount of MoO_3 was added to the raw materials to compensate for the evaporation of MoO_3 during crystal growth. The fully charged crucible was heated up to 1300 °C. After adjusting the growing temperature, the crystal was grown at a pulling rate of 0.5–2.0 mm/h and rotating rate of 10–30 r/min. When the growth process ended, the grown crystal was cooled down to the room temperature at an annealing rate of 10–30 °C/h. Finally, A transparent Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal with dimensions of Ø20 × 25 mm^3 and free crack was obtained, as shown in Fig. 3.

The powder XRD pattern of the grown crystal is shown in Fig. 1(b), which agrees with that of synthesized Nd^{3+}:$Na_2Gd_4(MoO_4)_7$. The lattice parameters of the grown crystal were determined to be: $a = b = 5.2365$ Å, $c = 11.4573$ Å and $\alpha = \beta = \gamma = 90°$ by the Rigaku AFC7R diffractometer. The single XRD shows that Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal is of isomorphism of $Li_2Gd_4(MoO_4)_7$ crystal with tetragonal system and lattice parameters: $a = b = 5.192$ Å, $c = 11.30$ Å [15]. According to the lattice parameters of the grown crystal, all XRD lines can be indexed (see Fig. 1). The XRD results confirm that the grown crystal belong to Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal.

3. Spectroscopic properties

Nd^{3+} concentrations in Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal were measured to be 1.96 at.%, i.e. 1.43×10^{20} cm^{-3} by the inductively

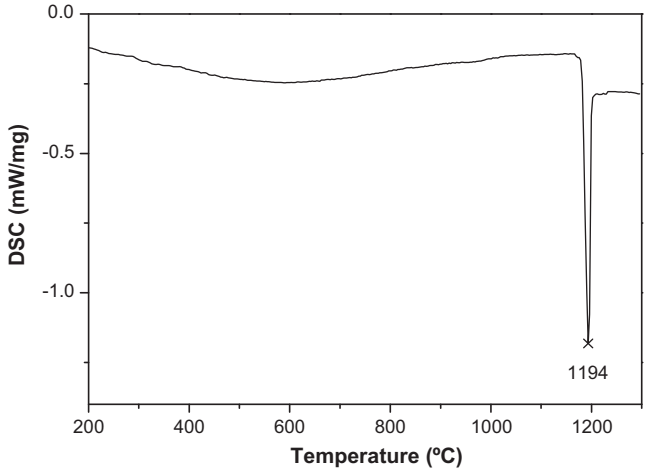

Fig. 2. DSC curve of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ compound.

Fig. 3. Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal grown by the Czochralski method (where white color is reflecting light).

coupled plasma and atomic emission spectrometry (ICP-AES). A plate sample with dimensions of 9 × 5 × 1 mm^3 cut from the as-grown crystal along the *c*-axis was used for the spectral measurement. The σ- and π-polarization were defined in terms of the *E*-vector being perpendicular or parallel to the *c*-axis, respectively. The polarized absorption spectrum of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal was measured using a Perkin-Elmer UV–VIS–NIR Spectrometer (Lambda-900) in a range of 350–950 nm at room temperature. The polarized fluorescence spectrum and fluorescence lifetime were determined using an Edinburgh Analytical Instruments FLS92 Fluorescence Spectrophotometer with a continuous Xe-flash lamp.

The polarized absorption spectrum of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal at room temperature is shown in Fig. 4. All the absorption bands are attributed to the transitions of Nd^{3+} ions from the ground state $^4I_{9/2}$ to the excited state J' manifolds, which are marked in Fig. 4. The absorption band at 807 nm has an absorption cross-section of 2.87×10^{-20} cm^2 with full width at half maximum (FWHM) of 16.2 nm for the σ-polarization, and has an absorption cross-section of 4.25×10^{-20} cm^2 with FWHM of 14.6 nm for the π-polarization, which is closed to the laser output wavelength of AlGaAs diode laser (λ ≈ 808 nm). Since the emission wavelength of the diode laser is increased at 0.2–0.3 nm/°C with the operating temperature of the laser device, the temperature stability of the output wavelength of the diode laser needs to be crucially controlled. Thus, such broad absorption band is suitable for diode laser

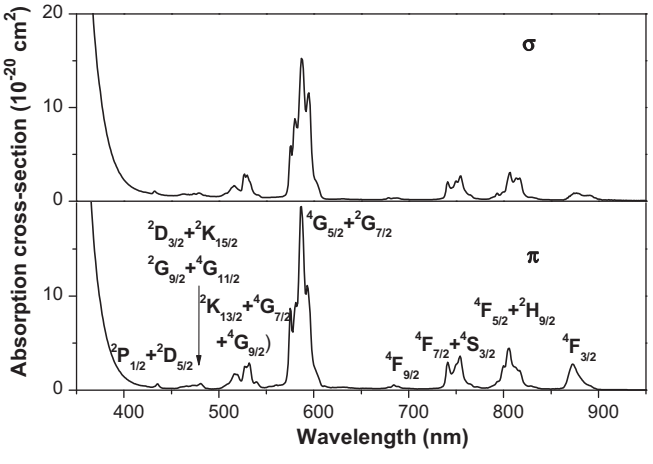

Fig. 4. Polarized absorption spectra of Nd^{3+}:$Na_2Gd_4(MoO_4)_7$ crystal at room temperature.

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