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Spectral properties of Yb^{3+} ions in $Lu_3ScAl_4O_{12}$ single crystal

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ABSTRACT

In this paper, a Yb^{3+} doped $Lu_3ScAl_4O_{12}$ single crystal has been grown by the Czochralski method. The cell parameters were analyzed with X-ray diffraction (XRD). The absorption spectrum, emission spectrum and fluorescence lifetime of $Yb:Lu_3ScAl_4O_{12}$ crystal were measured at room temperature. The absorption bandwidth, emission bandwidth, absorption cross-section, emission cross-section and fluorescence lifetime have been estimated as 25 nm, 12.6 nm, 0.54×10^{-20} cm^2, 1.47×10^{-20} cm^2 and 1.43 ms. The spectral properties of $Yb:Lu_3ScAl_4O_{12}$ crystal were compared with those of $Yb:Lu_3Al_5O_{12}$ (Yb:LuAG) crystal. The results indicate that $Yb:Lu_3ScAl_4O_{12}$ crystal should be a potential candidate used for generating short pulses in mode-locking operation.

1. Introduction

In recent years, ytterbium doped laser crystals have attracted considerable attention [1–6]. Trivalent ytterbium has important advantages in comparison with the widely used Nd laser in 1 μm laser applications. For example, Yb^{3+} ions have low thermal loading, long radiative lifetime of the upper laser level, large absorption width, broad emission width, and no concentration quenching, no excited-state absorption and no visible reabsorption loss. They offer high quantum efficiency, low quantum defects, reduced thermal effects and a potentially broad gain bandwidth [7–11].

Yb^{3+} doped mixed crystals are quite promising laser-active materials for ultrashort lasers because of their disordered natures, which leads to inhomogeneous broadening of fluorescence lines, with expectations of improving the laser performance in Q-switched and mode-locked regimes [12,13]. There are several reports of the optical properties and laser performance of Yb^{3+}-doped mixed garnets crystals and ceramics [14–16], in which partially replacing Al^{3+} ions with Sc^{3+} ions to form distorted $Y_3Sc_xAl_{5-x}O_{12}$ (YSAG) compounds. When enlarging the distance between dodecahedral lattice sites by introducing large Sc^{3+}, broad absorption and emission spectra were obtained in Yb:YSAG crystals and ceramics. At the same time, thermal and mechanical properties of YSAG crystals and ceramics are still comparable to that of YAG crystal or ceramics. $Lu_3Al_5O_{12}$ (LuAG) belongs to the rare-earth garnet family (space group Oh10-*Ia*3*d*) with lattice spacing 11.906 å. As a host material, LuAG possesses many attractive characteristics such as high thermal conductivity, excellent physical and chemical properties [17–22]. Particularly, partial-substitution of Sc^{3+} for Al^{3+} in $Lu_3Al_5O_{12}$ crystals forming distorted $Lu_3Sc_xAl_{5-x}O_{12}$ structure provides $Lu_3Sc_xAl_{5-x}O_{12}$ compounds with preferable conditions for trivalent lanthanide lasant doping. Therefore Yb^{3+} in $Lu_3Sc_xAl_{5-x}O_{12}$ crystal is expected to exhibit a larger emission band due to disordered crystal-field in $Lu_3Sc_xAl_{5-x}O_{12}$ crystal.

In this paper, the spectroscopic properties of $Yb:Lu_3ScAl_4O_{12}$ crystal were reported for the first time to our knowledge, and the spectral properties were compared with those of Yb:LuAG crystal.

2. Experimental

To grow $Yb:Lu_3ScAl_4O_{12}$ crystal with 10 at.% doped concentration, the chemicals of Yb_2O_3, Lu_2O_3, Sc_2O_3 and Al_2O_3 with purity of 99.999% were used. The mixture was ground, extruded to form pieces with diameter close to the inner diameter of the crucible at high pressure, then sintered in an alumina crucible at 1200 °C for 30 h. The charge was then loaded into the iridium crucible for crystal growth. The pulling rate was 1–3 mm/h and the rotation rate of the seed was 15–30 rpm. High-purity nitrogen gas was used as a protective atmosphere. The temperature was controlled by a EUROTHERM 818 controller/programmer with a precision of ±0.1 °C. In order to prevent the crystal from cracking, the crystal was cooled to room temperature slowly after growth.

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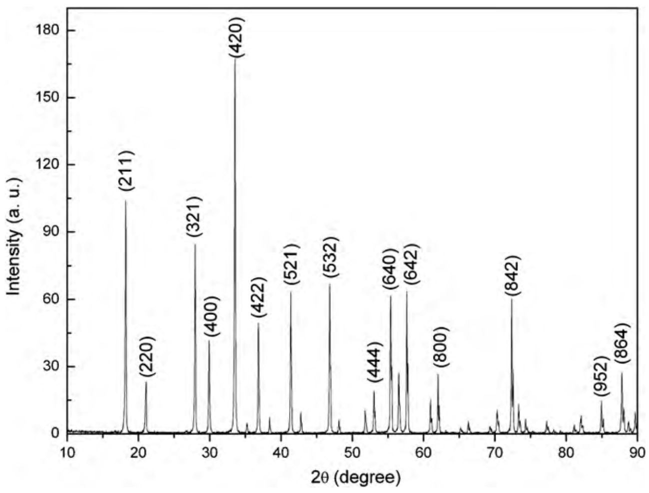

Fig. 1. Powder XRD patterns of Yb:$Lu_3ScAl_4O_{12}$ crystal.

The crystal structure of the as-grown Yb:$Lu_3ScAl_4O_{12}$ single crystal was analyzed by X-ray diffraction (XRD) using Cu Kα radiation (Ultima IV diffractometer, Rigaku, Japan) at a scan width of 0.02° within 2θ = 10–90°. Fine ground powder of the as-grown Yb:$Lu_3ScAl_4O_{12}$ single crystal was used as the sample. Samples for spectroscopic measurements with surfaces perpendicular to the <1 1 1>-growth axis were polished. The thickness of the sample is 1.08 mm. The absorption spectrum was measured in the wavelength range from 850 nm to 1100 nm using a Lambda 900 spectrophotometer (Perkin-Elmer Company). The luminescence spectrum of the sample was recorded by a spectrofluorometer (Fluorolog-3, Jobin Yvon, Edision, USA) equipped with a Hamamatsu R928 photomultiplier tube. A 940 nm continuous wave diode-laser was used as the excitation source. The decay time was measured by a computer controlled transient digitizer. All measurements were performed at room temperature.

3. Results and discussions

The structure of Yb:$Lu_3ScAl_4O_{12}$ crystal has been determined by X-ray diffraction analysis, as shown in Fig. 1. The result reveals the Yb:$Lu_3ScAl_4O_{12}$ crystal crystallizes in cubic with space group *Ia*3*d* and has the cell parameters: a = 1.1941 nm, V = 1.7026 nm^3.

The room temperature absorption spectra of Yb:$Lu_3ScAl_4O_{12}$ and LuAG crystals after annealing in the range of 850–1100 nm are shown in Fig. 2. The absorption characteristics of Yb:$Lu_3ScAl_4O_{12}$ crystal are similar to those of Yb:LuAG crystal. There are four absorption peaks centered at 916, 939, 968 and 1029 nm, respectively. 916, 939 and 968 nm are suitable for laser-diode pumping. The absorption coefficient at 939 nm is 7.81 cm^{-1}, which is smaller than that of Yb:LuAG crystal (~8.44 cm^{-1}). The absorption cross-section is 0.54×10^{-20} cm^2 at 939 nm. The absorption bandwidth (FWHM) of Yb:$Lu_3ScAl_4O_{12}$ crystal centered at 939 nm is 25 nm, which is 3 nm wider than that of Yb:LuAG crystal. Therefore, the Yb:$Lu_3ScAl_4O_{12}$ crystal is more suitable for InGaAs diode-laser pumping owing to the broader band absorption features. A wide absorption bandwidth means that the laser crystal is less sensitive to diode wavelength specification and the output power of the laser remains stable. Therefore, the FWHM of absorption band at pump wavelength is one of the important parameters for laser crystal.

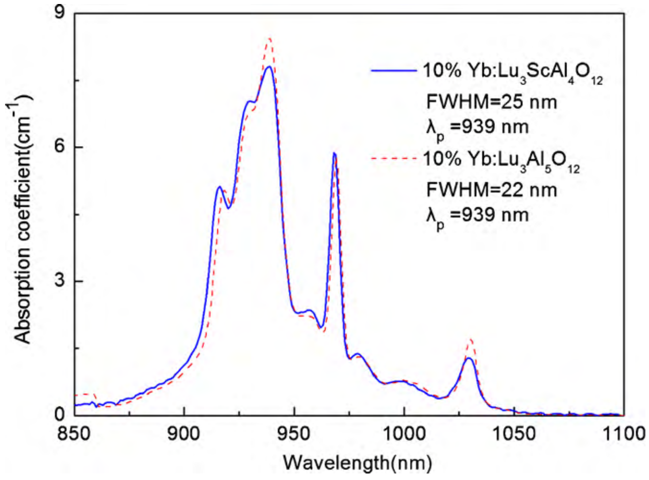

Fig. 2. Absorption spectra of Yb:$Lu_3ScAl_4O_{12}$ and Yb:LuAG crystal with the same doping level.

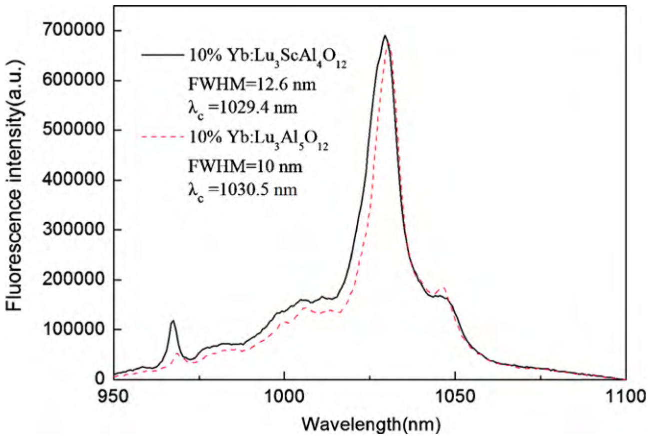

Fig. 3. Fluorescence spectra of the Yb:$Lu_3ScAl_4O_{12}$ and Yb:LuAG crystal.

Fig. 3 shows the fluorescence spectra of the Yb:$Lu_3ScAl_4O_{12}$ and Yb:LuAG crystals. The emission spectrum of Yb:$Lu_3ScAl_4O_{12}$ crystal is similar to that of Yb:LuAG crystal. There is a strong emission peak located at wavelength 1029 nm, and the emission shifts to shorter wavelength. The emission bandwidth of Yb:$Lu_3ScAl_4O_{12}$ crystal (~12.6 nm) is 1.26 times larger than that of Yb:LuAG single crystal (~10.0 nm). The value is similar to that of Yb:$Y_3ScAl_4O_{12}$ ceramics (~12.5 nm) [15], but smaller than that of Yb:$Y_3Sc_2Al_3O_{12}$ crystal (~14 nm) [14]. The wide emission wavelength range is useful for generating short pulses in mode-locking operation. Fluorescence decay curve is shown in Fig. 4. The fluorescence lifetime was measured to be 1.43 ms, which is longer than that of Yb:LuAG crystal with the same Yb^{3+} concentration (~1.32 ms) [19].

From the absorption and fluorescence spectra we determined the Stark energy levels. The resulting Stark energy-level diagram of Yb^{3+} in the $Lu_3ScAl_4O_{12}$ crystal-field at room temperature is

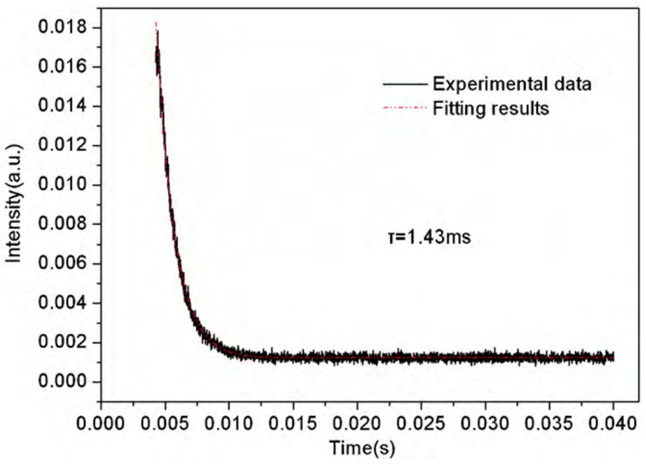

Fig. 4. Decay curve of the $^2F_{5/2}$ manifold of Yb:$Lu_3ScAl_4O_{12}$ crystal.

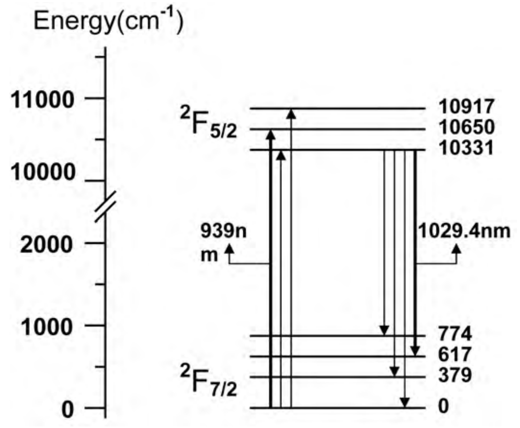

Fig. 5. Stark energy-level diagram of the $^2F_{5/2}$ and $^2F_{7/2}$ manifold of Yb^{3+} in $Lu_3ScAl_4O_{12}$ crystal.

shown in Fig. 5. The ground-state energy-level splitting, giving by $\triangle E = E_{zl} - E_{ext}$, was calculated to be 774 cm^{-1} and the zero-line energy E_{zl} is equal to 10331 cm^{-1}.

The emission cross-section of Yb^{3+}: $^2F_{5/2} \to {}^2F_{7/2}$ transition can be calculated from the ground-state absorption cross-section using the reciprocity method (RM) [11].

$$\sigma_{em}(\lambda) = \sigma_{abs}(\lambda)\frac{Z_l}{Z_u} \exp\left[\frac{E_{ZL} - (hc/\lambda)}{kT}\right] \qquad (1)$$

where $\sigma_{abs}(\lambda)$ is the absorption cross-section, Z_l and Z_u are partition functions of lower and upper manifolds, respectively. E_{ZL} is the zero-line energy, and is defined as the energy gap difference between the lowest Stark level of the upper manifold and the lowest Stark level of the lower manifold. For $Yb:Lu_3ScAl_4O_{12}$ crystal, the zero-line λ_{ZL} is equal to 968 nm and the value of Z_l/Z_u is 0.85. The emission cross-section is 1.47×10^{-20} cm^2 at 1031 nm.

4. Conclusion

$Yb:Lu_3ScAl_4O_{12}$ single crystal has been grown successfully by the Czochralski technique. The absorption spectrum, emission spectrum and fluorescence lifetime of $Yb:Lu_3ScAl_4O_{12}$ crystal were measured at room temperature. The absorption bandwidth of $Yb:Lu_3ScAl_4O_{12}$ crystal centered at 939 nm is 25 nm and the emission bandwidth of $Yb:Lu_3ScAl_4O_{12}$ centered at 1029 nm is 12.6 nm. The absorption, emission cross-section and fluorescence lifetime have been estimated as 0.54×10^{-20} cm^2, 1.47×10^{-20} cm^2 and 1.43 ms. The spectroscopic parameters of $Yb:Lu_3ScAl_4O_{12}$ crystal were compared with those of Yb:LuAG crystal with the same doping level. The results indicate that $Yb:Lu_3ScAl_4O_{12}$ crystals are potential candidates used for generating short pulses in mode-locking operation.

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