

Infrared (1.2–1.6 μm) luminescence in $\text{Cr}^{4+}:\text{Yb}_3\text{Al}_5\text{O}_{12}$ single crystal with 940 nm diode pumping

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Abstract

Infrared (1.2–1.6 μm) luminescence in a ytterbium aluminium garnet (YbAG) crystal, doped with Cr (0.05 at.%) ions, was investigated under CW laser diode pumping ($\lambda = 940 \text{ nm}$). The Cr^{4+} emission band was observed with its peak at 1.34 μm and measured to be about 1.3 times with respect to Yb^{3+} IR luminescence ($\lambda = 1.03 \mu\text{m}$). We demonstrate that for the excitation wavelength of 940 nm Yb^{3+} ions act as sensitizers of the ${}^3\text{B}_2({}^3\text{T}_2) \rightarrow {}^3\text{B}_1({}^3\text{A}_2)$ emission of Cr^{4+} ions. This crystal is promising as a high-efficient system for tunable laser (1.2–1.6 μm) output.

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1. Introduction

Tetravalent chromium ion (Cr^{4+}) has been recognized as a popular and efficient ions for obtaining tunable laser operating in the near-infrared region (1.2–1.6 μm) [1–10]. Operating around 1.3 and 1.5 μm , the second and third telecommunication windows respectively, Cr:YAG lasers should find applications in spectroscopy, remote sensing, medicine and in the laboratory characterization of fibre communication systems [11]. The material has a broad absorption peak around 1 μm and is usually pumped by Nd:YAG lasers operating at 1.06 μm [12–13].

It is well-known that rare-earth and transition metal elements are used both as active centers and sensitizers. The ytterbium ions have been studied as sensitizers in yttrium aluminium garnet (YAG) crystals [14–16]. $\text{Yb}_3\text{Al}_5\text{O}_{12}$ and $\text{Y}_3\text{Al}_5\text{O}_{12}$ belong to the crystal family of the rare-earth alu-

minium garnets ($\text{RE}_3\text{Al}_5\text{O}_{12}$, Re=rare earth or yttrium). They are isostructural with only about a 1.5% difference in unit-cell size and a 200 °C separation in their melting points, and they have a cubic symmetry with space group $Ia3d$ [17]. In our recent letter [18] we showed that chromium can be easily doped into YbAG crystals to form tetrahedral Cr^{4+} ions for the purpose of obtaining a compact, efficient, highly stable, passive self-Q-switched thin chip solid-state lasers, and the Yb^{3+} ions act as active centres and Cr^{4+} ions act as excellent saturable absorbers. In this letter, we report on the experimental investigation of NIR luminescence of Cr^{4+} doped ytterbium aluminium garnet crystal under 940 nm excitation. The results demonstrate that it can act as a suitable material for Cr^{4+} tunable laser output.

2. Experiments

Raw materials of 5N pure Yb_2O_3 , Al_2O_3 , Cr_2O_3 and CaCO_3 were weighed, according to the stoichiometric composition. After the compounds were ground and thoroughly

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mixed, they were pressed into pieces. The pieces were heated to 1200 °C and kept at the temperature for 20 h. Then the mixture was charged into an Iridium crucible of 50 mm in diameter for crystal growth. The pulling and rotation rates were 1 mm h⁻¹ and 10–20 rpm, respectively, under the nitrogen or argon atmosphere, respectively. The initial growth boundary in solid-melt was convex towards the melt so that dislocation and impurities were reduced or eliminated from the crystal. After that, the growth boundary became flat. In order to prevent the crystal from cracking, the crystal was cooled to room temperature slowly after growth. The crystal was blue-green and free from crack, inclusions and precipitations. When the 20 mW He–Ne laser beam passed through the as-grown Cr⁴⁺:YbAG, the light beam was almost unseen, which indicated very few scattering particles in Cr⁴⁺:YbAG crystal. The crystal changed to brown after annealing in oxygen atmosphere at 1400 °C for 24 h.

Samples for spectroscopic measurements were cut out of the boules and surfaces perpendicular to the (111)-growth axis were polished. The thickness of the samples was 0.35 mm. A JASCO V-570 UV–vis/NIR Spectrophotometer was employed for acquisition of the absorption spectra at room temperature. The fluorescence spectra were acquired by a TRIAX 550 spectrophotometer with InGaAs LD as the pump source (excited at 940 nm). The decay time was measured by a computer controlled transient digitizer.

3. Results and discussion

The room temperature absorption spectra of YbAG and Cr⁴⁺:YbAG crystals are displayed in Fig. 1. As it is seen from the spectra, the main absorption bands in YbAG and Cr⁴⁺:YbAG crystals lie between 900 and 1050 nm (Yb³⁺, transition ²F_{7/2}–²F_{5/2}), which is characteristic for all Yb³⁺-activated materials. Each absorption peak of Cr⁴⁺:YbAG in-

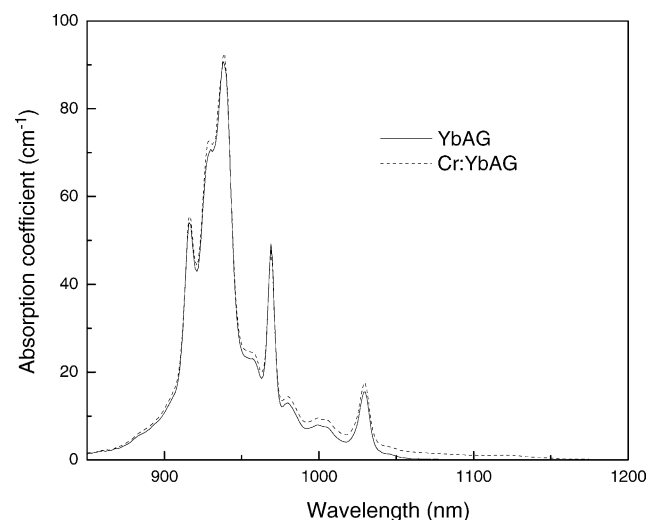


Fig. 1. The absorption spectra of YbAG and Cr⁴⁺:YbAG crystals.

creases in intensity compared with YbAG. The absorption coefficient at 1030 nm increases from 15.6 to 17.5 cm⁻¹, and the increase of the absorption is due to the increase of the concentration of Cr⁴⁺ ions. The band centered at 1030 nm is believed to be caused by the ³A₂ → ³T₁ transition of Cr⁴⁺ ions. The absorption spectrum of Cr⁴⁺, as shown in Fig. 2, can be calculated by subtracting the absorption spectrum of YbAG from the absorption spectrum of Cr⁴⁺:YbAG. It can be seen that Cr⁴⁺ ions have broad absorption band, and the absorption is similar to that of Cr⁴⁺:YAG crystal [4].

Fig. 3 shows the fluorescence spectra of Cr⁴⁺:YbAG crystal under 940 nm diode pumping. There is a emission peak located at wavelength 1032 nm, which is the characteristic emission of Yb³⁺. The fluorescence band peaks at ~1.34 μm and covers a wide range from 1.2 to 1.6 μm. The spectrum is similar to that of Cr⁴⁺:YAG crystal previously reported [2]. In our experimental conditions, the intensity of the luminescence centered at 1.34 μm was notably high and was estimated to be 1.3 times of the emission at 1032 nm. This fact supports the prospect of achieving lasing in Cr⁴⁺:YbAG under 940 nm diode pumping.

In Cr⁴⁺:YbAG crystal, which is closely analogous to Cr⁴⁺:YAG, the Cr⁴⁺ ions occupy trivalent cation sites with tetragonally distorted tetrahedral coordination, and are charge compensated by substitutional Ca²⁺ ions. Accordingly, the relevant Cr⁴⁺ energy levels, all of which are derived from the ³F ground term of the free ion, are labeled by the irreducible representations of tetragonal group D_{2d} followed in parentheses by the corresponding irreducible representations of tetrahedral group T_d. Three such levels are shown in Fig. 4. According to the Tanabe-Sugano diagram for tetrahedrally coordinated Cr⁴⁺:YAG [6], the ³B₁(³A₂) → ³A₂(³T₁) transition is the only fully electric-dipole allowed transition, to the extent that the tetrahedral crystal field relaxes the parity selection rule, and is therefore a target for energy transfer since it is in resonance with the emission of Yb³⁺. The

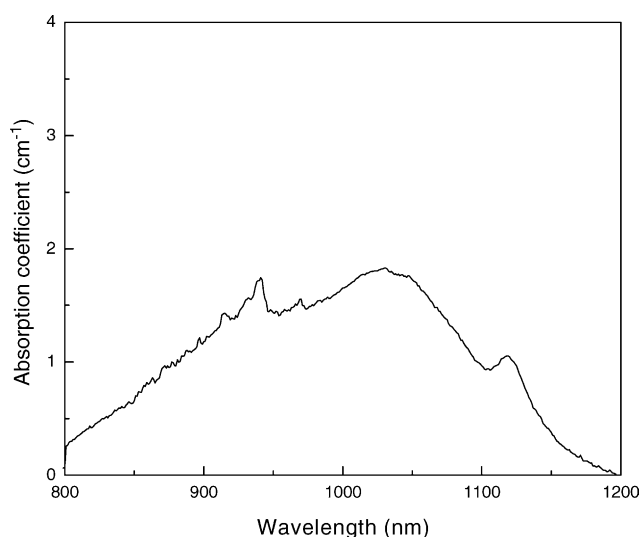


Fig. 2. The absorption of Cr⁴⁺ in Cr⁴⁺:YbAG crystal.

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