Gamma-ray induced color centers in Yb:YAG crystals grown by Czochralski method

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Abstract

20 at.% Yb:YAG single crystals have been grown by the CZ method and gamma-ray irradiation induced color centers and valence change of Fe^{3+} and Yb^{3+} ions in Yb:YAG have been studied. One significant 255 nm absorption band was observed in as-grown crystals and was attributed to Fe^{3+} ions. Two additional absorption (AA) bands located at 255 nm and 345 nm, respectively, were produced after gamma irradiation. The changes in the AA spectra after gamma irradiation and air annealing are mainly related to the charge exchange of the Fe^{3+}, Fe^{2+}, oxygen vacancies and F-type color centers. Analysis shows that the broad AA band is associated with Fe^{2+} ions and F-type color centers. The transition $Yb^{3+} \rightarrow Yb^{2+}$ takes place as an effect of recharging of one of the Yb^{3+} ions from a pair in the process of gamma irradiation.
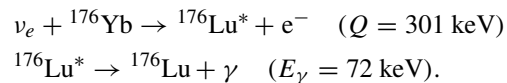© 2006 Elsevier Ltd. All rights reserved.

1. Introduction

Owing to favorable properties, especially the high thermal conductivity and excellent physical and chemical properties of the host crystals, Yb^{3+} doped crystals are attractive diode pumped solid-state laser (DPSSL) systems. Especially, recent advances in a high-performance InGaAs laser diode with a wavelength between 900 and 1100 have stimulated interest in developing LD pumped Yb^{3+}-doped lasers [2,1]. Due to a broad absorption band suited for diode-pumping, relatively larger laser transition cross section, the lowest heat generation of any diode-pumped solid-state laser medium, and high quality and highly doped crystals which can be easily grown, Yb:YAG crystals have become an important components in diode pumped high power laser systems [3]. Yb:YAG could also be used for solar neutrino detection [4]. It was demonstrated that the interaction of a neutrino with ^{176}Yb can be described by the following formulas:

$$\nu_e + {}^{176}\text{Yb} \rightarrow {}^{176}\text{Lu}^* + e^- \quad (Q = 301 \text{ keV})$$
$${}^{176}\text{Lu}^* \rightarrow {}^{176}\text{Lu} + \gamma \quad (E_\gamma = 72 \text{ keV}).$$

A prompt electron plus a delayed gamma-signal is the signature of a neutrino event. A delayed coincidence within 50 ns is required to have good discrimination power against background noise [5,6].

Solid-state lasers are sometimes used in the same environment (e.g. instruments in orbital space missions, space-based light detection and ranging systems) [7,8]. Scintillators are designed to work in the strong external fields of ionizing radiation (e.g. gamma-ray, neutron). Therefore, It is important to study the ionizing irradiation effects on Yb:YAG crystals. However, there seem to be few reports on gamma irradiation effects on Yb:YAG crystals. It is the purpose of the present investigation to explore impurity defects and gamma-ray induced color centers in Yb:YAG crystals grown by the CZ method.

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2. Experiments

Yb:YAG crystals were grown by the Czochralski method [9]. The starting materials were Y_2O_3 (5 N), Al_2O_3 (5 N) and Yb_2O_3 (5 N), weighed according to a specific molar ration. After the compounds were ground and thoroughly mixed, they were pressed into the form of blocks. These pieces were sintered at 1350 °C for 24 h in air and then loaded into an iridium crucible for crystal growth. The growth atmosphere was highly pure nitrogen gas. The pulling rate was 1 mm/h and the rotation rate was about 20 rpm.

Samples were cut from the as-grown Yb:YAG crystals perpendicularly to the growth axis and polished on both sides. We performed gamma-ray irradiation by means of a ^{60}Co gamma source (average gamma energy 1.25 MeV) on the sample up to an absorbed dose of 2×10^6 Gy with the dose rate of about 250 Gy/min at room temperature. The absorption spectra were recorded by means of a V-570 UV/VIS/NIR spectrophotometer. The light sources were a deuterium lamp (190–350 nm) and a halogen lamp (340–1200 nm), and the spectral resolution was 1 nm. To make accurate comparison, gamma irradiation was performed on the same piece of sample. Values of induced additional absorption (AA) due to the irradiation or thermal processing were calculated from the formula:

$$\Delta k = \frac{1}{d} \ln \frac{T_1}{T_2} \tag{1}$$

where k is the absorption, d is the sample thickness and T_1 and T_2 are the transmissions of the sample obtained before and after gamma-irradiation or thermal treatment, respectively.

3. Results and discussion

Fig. 1 shows the absorption spectrum of as-grown 20 at.% Yb:YAG crystals at room temperature. The broad absorption band in the range of 190–250 nm, which is not observed in the spectra of the undoped sample, is assigned to the transitions from the $^2F_{7/2}$ ground state of Yb^{3+} to the charge transfer state (CTS) [10]. The four strong absorption bands at 914, 940, 969 and 1028 nm are attributed to the $^2F_{7/2} \rightarrow \ ^2F_{5/2} Yb^{3+}$ transition. It can be seen that one prominent absorption band at 255 nm occurred in Yb:YAG grown by the CZ method. It has been shown from optical absorption and the electron paramagnetic resonance (EPR) technique that the absorption band at 255 nm in YAG was due to an Fe^{3+} charge-transfer band, which was made up of contributions from substitutional Fe^{3+} ions in octahedral and tetrahedral sites [11]. Work by Jiang et al. [12] also suggested the absorption peak at 255 nm in YAG crystals grown by CZ is associated with Fe^{3+} ions. In order to further determine if Fe^{3+} ions exist in our samples, we measured the electron paramagnetic resonance (EPR) of the as-grown Yb:YAG and compared the result with that of Fe^{3+}:YAG reported in previous work [11]. The ESR result was shown in the inset of Fig. 1. As we can see, the positions of ESR peaks are in good agreement with that of the intentionally Fe^{3+} doped sample. Therefore, it is believed that the Fe^{3+} ions
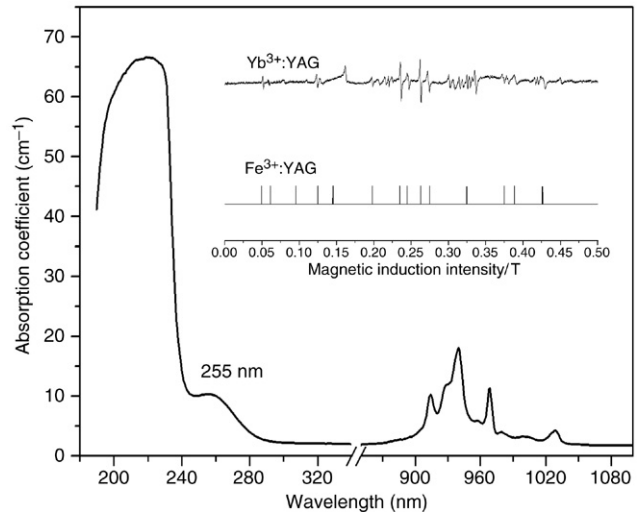

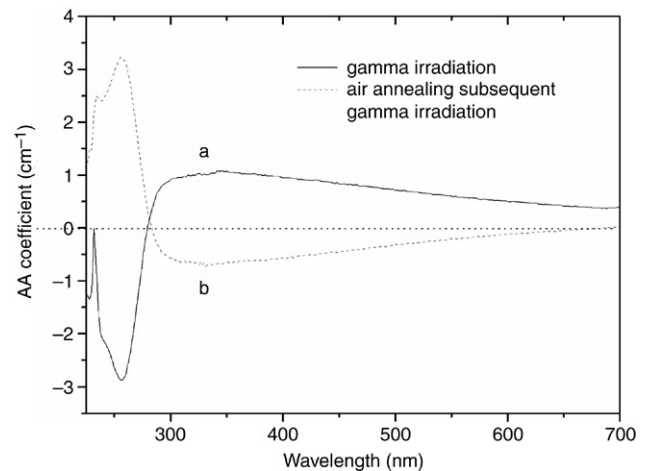Fig. 1. Absorption and EPR spectrum of the as-grown Yb:YAG crystals.

Fig. 2. AA spectrum of 20 at.% Yb:YAG after gamma irradiation (a) and annealing in air at 800 °C for 12 h subsequent gamma-ray irradiation (b).

are responsible for the absorption band at 255 nm in Yb:YAG crystals. However, our further experiments indicate that the 255 nm band was not observed in 5 and 10 at.% Yb:YAG. Therefore, the presence of the uncontrolled impurity Fe^{3+} ions may strongly depend on growth conditions.

Fig. 2 curve (a) shows the additional absorption spectrum of 20 at.% Yb:YAG crystals after γ-irradiation at room temperature. One negative additional absorption band and one broad positive AA band, located at 255 nm and 345 nm, respectively, were produced. It has been shown that the 255 nm band is correlated to Fe^{3+} impurity ions. The negative AA value indicates that the concentration of the Fe^{3+} ions decreases after gamma irradiation. During the gamma-ray irradiation process, high-energy photons (1.25 MeV) produced a large number of free electrons in crystals (mainly involving the Compton Effect, Photoelectric Effect and Pair Production processes). These free electrons could be trapped by Fe^{3+} ions and this leads to recombination of the Fe^{3+} to Fe^{2+} ions. This process could be expressed as: $Fe^{3+} + e \rightarrow Fe^{2+}$.