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Induced Absorption in Gadolinium Gallium Garnet Irradiated by High Energy ^{235}U Ions

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The present work is devoted to investigation of optical absorption in pure $Gd_3Ga_5O_{12}$ (GGG) single crystals in the spectral range 0.2–1.1 μm induced under influence of the ^{235}U ions irradiation with energy 2640 MeV and a fluence 10^9–10^{11} cm^{-2}. The induced absorption for 10^9 cm^{-2} is caused by recharging of point defects, both growth ones and impurities. After irradiation by ^{235}U ions with fluences starting from 3×10^9 cm^{-2} the absorption rise is probably caused by contribution of the lattice destroying as a result of heavy ion bombardment as well as radiation displacement defects.

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1. Introduction

Gadolinium gallium garnet $Gd_3Ga_5O_{12}$ (GGG) single crystals doped with rare earth ions (Nd^{3+}, Er^{3+}, Tm^{3+}, Ho^{3+}, Pr^{3+}, Ce^{3+}) or with 3*d*-ions (Cr^{4+}, Cr^{3+}, Co^{2+}, V^{3+}) are the most perspective materials for laser engineering [1], specially for pulsed solid-state lasers of the 1.06–1.44 μm region (GGG:Nd) [2, 3].

Neodymium lasers are used in LIDAR optical systems and navigation systems that can operate outside of Earth's atmosphere [4]. Such laser systems operating on near-earth orbit are exposed with electrons, protons and high-energy cosmic radiation, whose absorbed doses during 5 years can reach 10^3 Gy [4]. For example, it was shown previously that stable colour centres (SCC) induced by gamma quanta irradiation of GGG:Nd crystals absorb the pumping light reducing the laser output efficiency [5].

There are many publications devoted to investigation of the colour centres (CC) created by various kinds of ionizing radiation (UV, gamma-quanta, electrons, neutrons) in pure and doped GGG crystals, see for example [6, 7]. However, the CC formation in the crystal under the ion irradiation was studied insufficiently. The present work is devoted to investigation of induced absorption in GGG single crystals after irradiation by high energy ($\approx$ 2.6 GeV) ^{235}U with 10^9-10^{11} cm^{-2} fluence at room temperature.

2. Experimental details

Investigated GGG crystal was grown by the Czochralski method at conditions described in [6]. Polished plates of the crystal with thickness of 1.87 mm were used for optical measurements. The crystals were irradiated by ^{235}U ions with energy per nucleon 11.23 MeV/u (the total particle energy was 2640 MeV) and fluences: 1×10^9 cm^{-2}, 3×10^9 cm^{-2}, 1×10^{10} cm^{-2}, 2×10^{10} cm^{-2}, 5×10^{10} cm^{-2} and 1×10^{11} cm^{-2}.

The irradiation was performed at room temperature, without control and without cooling. The samples irradiated by high-energy uranium ions were free of visible damages. The transmission spectra were recorded in the spectral range 0.2–1.1 μm using UNICAM UV 300 spectrophotometer.

The induced additional absorption (AA) was calculated as

$$\Delta K = \frac{1}{d}\ln\frac{T_1}{T_2},$$

where d is the sample thickness, T_1 and T_2 are the optical transmission values before and after irradiation, respectively.

3. Results

The optical absorption spectrum of as-grown GGG crystal in the region 45000–10000 cm^{-1} is shown in Fig. 1. This spectrum shows the narrow, high intensity lines and weak pronounced broad bands at $\approx$ 29000 cm^{-1}, and $\approx$ 23000 cm^{-1}. The origin of narrow lines are intra-center transitions of *f*-electrons in Gd^{3+} ions (transitions from

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ground $^8S_{7/2}$ state to splitted by crystal field multiplets ^6P, ^6I and ^6D terms), whereas colour centres connected with intrinsic point defects are responsible for broad absorption bands (F centres, hole O^- centres can be localized near V_{Ga}, Gd^{3+}_{octa} ions, $[Ca^{2+}F^+]$ and $[Ca^{2+}O^-]$ complex centres) [6, 8].

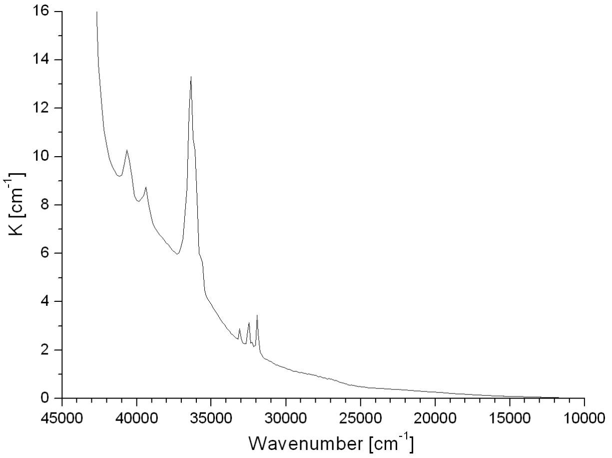

Fig. 1. Absorption of the as-grown GGG crystal.

Low fluency (10^9 cm^{-2}) irradiation of the GGG crystals with ^{235}U ions leads to the formation of slight coloration in UV–IR region with less or more appearing absorption maxima in 20000–25000 cm^{-1} and 26000–28000 cm^{-1} regions (Fig. 2). It is stable at room temperature and the absorption value does not exceed 0.07 cm^{-1}.

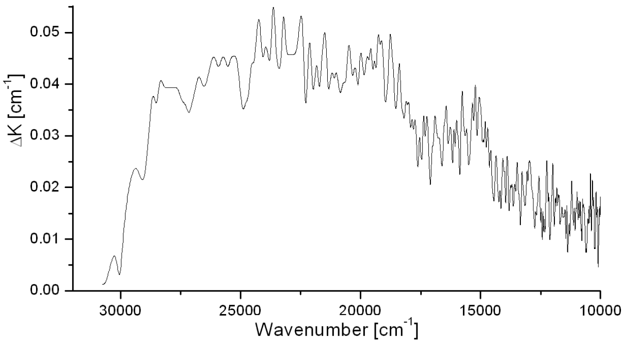

Fig. 2. AA of the GGG crystals irradiated by ^{235}U with the fluence of 10^9 cm^{-2}.

The energy losses of incident ions with specific energies above 1 MeV/u are defined by electronic interactions and lead to the primary ionisation of lattice atoms followed by creation of secondary free electrons and holes [9]. Those electrons can be trapped by different point defects or impurities, or can recombine with holes. In the first case, ionisation processes may lead to recharging of genetic defects and impurities.

The induced absorption is observed in the same where it appears in GGG after γ-irradiation that allows to attribute it to ionization recharging of growth defects and impurities present in the crystal namely to formation of F-centres (absorption in the region of 24000–21000 cm^{-1}) and O^--centres (30000–27000 cm^{-1}) [6]. A concentration of the irradiation induced displacement defects, produced in crystal volume under ion bombardment with the ion fluence 1×10^9 cm^{-2}, is very low in comparison with concentration of genetic defects, and they cannot influence the crystal absorption.

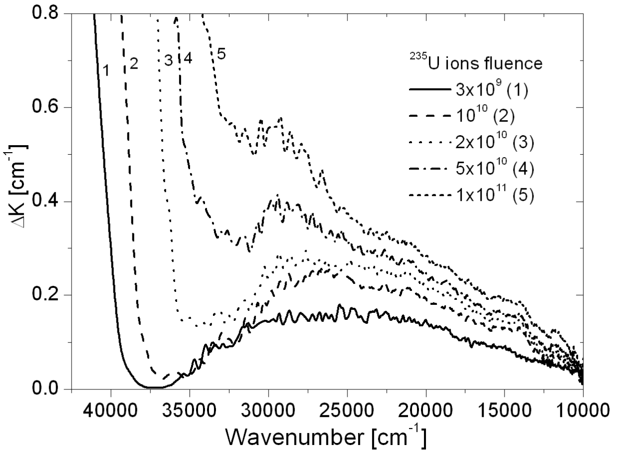

Fig. 3. Induced absorption spectra in GGG irradiated by ^{235}U ions with fluences: 3×10^9 cm^{-2}, 1×10^{10} cm^{-2}, 2×10^{10} cm^{-2}, 5×10^{10} cm^{-2} and 1×10^{11} cm^{-2}.

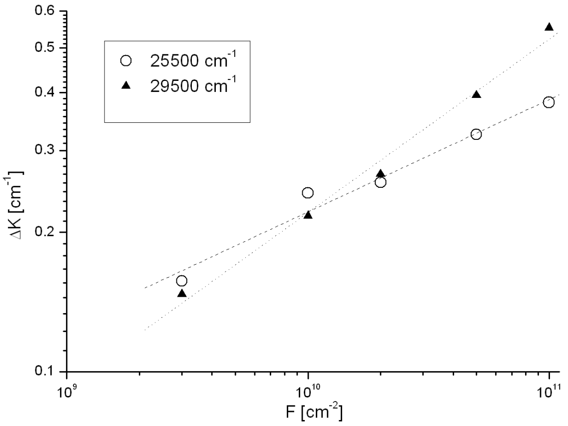

Fig. 4. Induced optical absorption at 29500 cm^{-1} and 25500 cm^{-1} versus the fluence of ^{235}U ions.

After irradiation by ^{235}U ions with the fluence above 1×10^{11} cm^{-2}, a much stronger absorption rise was observed (Fig. 3). The irradiation by ^{235}U ions results in formation of coloration in the 26000–34000 cm^{-1} region with not well resolved band and some shift of fundamental absorption edge (Fig. 3). The induced absorption spectra are stable at room temperature.

The dose dependence for 29500 cm^{-1} and 25500 cm^{-1} shows a linear growth in double-logarithmic scale

(Fig. 4). It is considered that the absorption rise is probably caused by the lattice destroying as a result of heavy ion bombardment as well as formation of CC in disturbed lattice. It is known that heavy ion irradiation can produce particle tracks in the crystal. In part, it was shown for fluoride crystals that heavy ions irradiation produce not only single defects in tracks but aggregates of defects as well [9]. That is why observed strong absorption is considered as contribution of the lattice destroying to optical losses as a result of heavy ion bombardment. The lattice disordering induced in oxide crystals irradiated with ^{238}U ions was also observed for example in sapphire irradiated with 0.48–3.40 MeV/u energies and fluences extending from 1.2×10^{12} to 2.5×10^{12} ions cm^{-2} [10].

4. Conclusions

Irradiation of pure gadolinium gallium garnet by ^{238}U ions with the energy $\approx$ 11 MeV/u and the fluence of 1×10^9 cm^{-2} does lead to creation of the absorption changes of the crystals as the results of ionisation of lattice atoms. The strong absorption rise during irradiation of crystals with the fluence above 3×10^9 cm^{-2} is due to contribution of the lattice destroying induced by heavy ion bombardment.

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