DEFECTS IN GADOLINIUM GALLIUM GARNET SINGLE CRYSTALS IRRADIATED BY NEUTRONS

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Gadolinium gallium garnet ($Gd_3Ga_5O_{12}$) single crystals are widely used as substrate materials for epitaxial films and microwave devices. Presence of point defects and their associates in structure influences a number of physical properties and hampers use of these crystals in devices. During the last few years a considerable work has been carried out on study of optical properties of irradiated $Gd_3Ga_5O_{12}$ [1-5]. However, models explaining changes of optical properties offered by various authors are inconsistent. Results of investigation of the effects of irradiation with fast and thermal neutrons and thermal treatment on absorption spectra are presented for $Gd_3Ga_5O_{12}$ single crystals.

EXPERIMENTAL

The pure $Gd_3Ga_5O_{12}$ crystals and those doped with Ca^{2+}ions used in this investigation were grown by the Czochralski method in Russia and Ukraine. The samples were prepared in the form of a flat wafer 0.4 to 0.8 mm thick oriented in the (111) plane with polished surfaces.

The neutron irradiation was performed at Latvian 5 MW water-water research reactor. The fluence of fast neutrons with energy > 0.1 MeV was in the range of $10^{14} - 6.8 \cdot 10^{18}$ cm^{-2}. The irradiation temperature did not exceed 350 K. A cadmium filter was used for absorption of thermal neutrons. The thermal neutrons fluence did not exceed $6.8 \cdot 10^{18}$ cm^{-2}. The thermal evolution of absorption bands was observed at isothermal annealing and at isochronal annealing in thermal region 300-1100 K in electric field 1 kV/cm and without field. The lattice parameter has been measured for irradiated and annealed samples by X-ray difractometer DRON UM-2 (Russia).

The optical absorption spectra were measured using "Specord M-40" (Karl Zeiss Jena) double-beam spectrophotometer operating in the wavelength region of 50000-11000 cm^{-1} (200-900 nm). Optical measurements before and after irradiation were carried out at room temperature.

RESULTS AND DISCUSSION

The absorption spectra of pure $Gd_3Ga_5O_{12}$ shown the sharp lines in UV region connected with Gd^{3+} electron transition from ground state $^8S_{7/2}$ to excited states $^6P_{7/2,5/2,3/2}$, $^6I_{17/2,11/2,9/2.7/2}$ and $^6D_{9/2,7/2,3/2,1/2}$. The single crystal of $Gd_3Ga_5O_{12}$ containing Ca impurity has the additional band ~29000 cm^{-1} (350 nm). After neutron irradiation the additional bands 33500 cm^{-1} (298.5 nm) and 23000 cm^{-1} (435 nm) may be distinguished in optical absorption spectra. These band positions are in good agreement with those obtained by Matkovskii [4]. At fluence increasing up to 10^{16} cm^{-2} we observed one more absorption band at ~36000 cm^{-1} (280 nm). With fluence increase its intensity arises and bands overlap each other.

The evolution of the ~36000 cm^{-1} absorption band during isochronal annealing in pure and doped crystals is shown in Fig. 1. Activation energies of the thermal dissociation of defects connected with this band are determined using isothermal annealing data. This value for the additional absorption band ~36000 cm^{-1} annealed in the electric field is $E_a \approx 0.96$ eV and annealed without electric field – $E_a \approx 1.38$ eV. The activation energy depends on separation distance in the vacancy-interstitial ion pair and increases with growth of this distance. The distortion around this close pair reduces the displacement activation energy of interstitial ions [6]. The obtained results allow suggest that the thermal destruction of bands is related to the migration of interstitial ions.

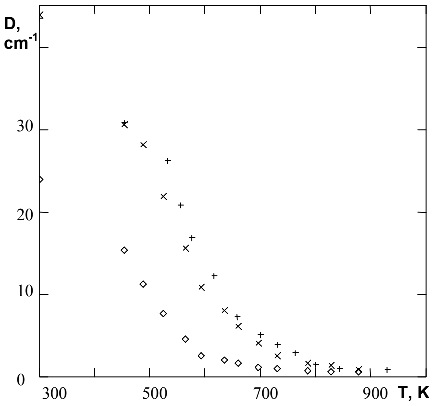

Fig. 1 Intensity of the ~36000 cm^{-1} absorption band as function of annealing temperature for $Gd_3Ga_5O_{12}$ irradiated with fast neutrons: ✕ – annealed in electric field; + – annealed without field; and $Gd_3Ga_5O_{12}$:Ca: ◇ – annealed without field.

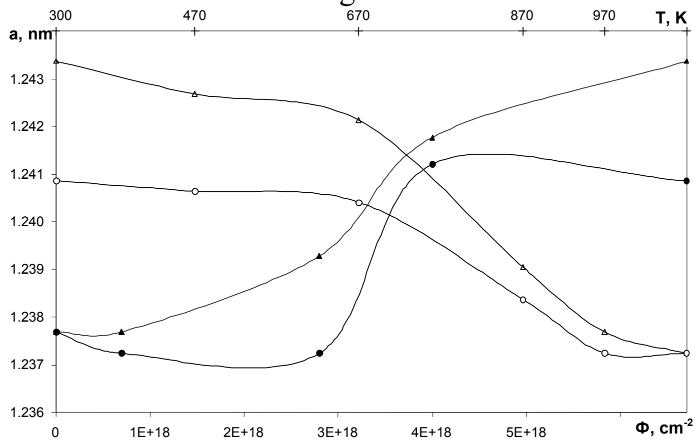

Fig. 2. Lattice parameter of $Gd_3Ga_5O_{12}$ as a function of neutron fluence: ● – fast neutrons, ▲ – thermal neutrons. Lattice parameter of $Gd_3Ga_5O_{12}$ as a function of annealing temperature: **O** – fast neutrons, △ – thermal neutrons.

The change of lattice parameter after irradiation with fast and thermal neutrons and after isochronal annealing is shown in Fig. 2. One can see that the fluence increase produces the considerable growth of the lattice parameter. The isochronal annealing up to 1100 K restores the lattice parameter and absorption spectra revert to the original shape.

The magnitude of gadolinium entering in octahedral sublattice may be determined from the lattice parameter change. The magnitude of gadolinium entering in octahedral sublattice was estimated using the lattice parameter change and the equation from Allibert at al. [7]. This value is 0.011 for nonirradiated crystal, 0.345 in the case of irradiation with thermal neutrons (maximum fluence $6.7 \cdot 10^{18}$ cm^{-2}) and 0.218 in the case of irradiation with fast neutrons. The $Gd_3Ga_5O_{12}$ lattice parameter depends not only on gadolinium and gallium correlation in a crystal [8] and presence of oxygen vacancies [9] but also on presence of vacancies in gallium sublattice [10], which is in agreement with conditions of the $Gd_3Ga_5O_{12}$ single crystal growth [11].

During irradiation of the samples, the radiation energy is mainly spent on excitation of the electron subsystem of the crystal, i.e. on ionizing. A part of radiation energy is lost during elastic collisions of radiation particles or secondary ones with crystal atoms. There are different ways in which atoms can be displaced by elastic collision processes, but it is evident that in any case a certain minimum amount of energy has to be used for displacement of a lattice ion [12]. If oxygen vacancies are present in the crystal, they should form electron traps and produce F^+ centers under exposure to ionizing radiation. The energy levels of F^+ center vary widely in oxide hosts. In yttrium aluminium garnet (YAG), three broad optical absorption bands at 800, 450 and 350 nm have been identified as transition of the F^+ center [13, 14]. However, the F^+ center in YAG is unstable at room temperature. If the F^+ center exists in $Gd_3Ga_5O_{12}$, it also may be unstable at room temperature [15]. The displacement defect formation in gadolinium gallium garnet single crystals under electron and neutron irradiation was investigated in details in [2, 4, 16]. They suppose that in the irradiated $Gd_3Ga_5O_{12}$ crystals, as the capture of electrons generated during irradiation on oxygen vacancies created in $Gd_3Ga_5O_{12}$ due to atom displacement is quite probable, F^+ centers with the absorption band in the 33500 cm^{-1} range are formed.

Since the additional band ~36000 cm^{-1} arises only in the neutron irradiated samples at fluence about ~10^{16} cm^{-2} we assume that this band is connected with “vacancy – interstitial” pairs. The lattice parameter changes after neutron irradiation confirm this assumption. Transition of a gadolinium ion into octahedral sublattice causes the displacement of a gallium ion into interstitial site. The annealing restores the initial equilibrium state. The restoration process is more intensive in electric field. Distinction of thermal destruction energies obtained for annealing in and without electric field allows believe that presence of electric field increases probability of recombination of a vacancy – interstitial pair.

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