Optical Properties of YAG:Ce and GGG:Ce Scintillation Crystals Irradiated with a High Fluence Proton Beam

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Abstract—In this paper, we report on the study of the optical properties of YAG:Ce and GGG:Ce garnet crystals after irradiation in a 660 MeV proton beam with a fluence up to $8.9 \times 10^{14}$ protons/cm$^2$. We found that the transparency of both crystals fell by no more than 7% in the region of their own luminescence. The light yield of the YAG:Ce sample, measured one year after the irradiation, dropped by about 35%.

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INTRODUCTION

Particle physics moves toward higher energies and beam intensities. This will require the search for new materials and the development of detectors that provide high reliability and long-term stability in harsh radiation conditions.

Inorganic garnet crystals have recently been increasingly used in tomography, medical applications, in particle physics and nuclear physics. This is due to their properties. The structure of garnet crystal allows changing scintillation properties by doping the crystals with different ions [1]. Yttrium aluminum garnet crystals YAG:Ce ($Y_3Al_5O_{12} \cdot Ce$) doped with Ce are candidates for use in future experiments in particle physics [2]. Gadolinium gallium garnet crystals GGG:Ce ($Gd_3Ga_5O_{12} \cdot Ce$) are widely used in tomography, electronics [3], biology and medicine [4].

In this paper, we report on the results of our study of the optical properties of YAG:Ce and GGG:Ce garnet crystals before and after irradiation with a 660 MeV proton beam.

CRYSTALS

Samples of the yttrium aluminum garnet crystal YAG:Ce and the gadolinium-gallium garnet crystal GGG:Ce doped with Ce were studied. The Ce content is 0.2 at % in YAG:Ce, and 0.3 at % in GGG:Ce, that is, the proportion of cerium atoms in the composition of the crystal samples is 0.2 and 0.3%, respectively. The GGG:Ce crystal has a high density of 7.08 g/cm$^3$. The YAG:Ce crystal has a density of 4.6 g/cm$^3$. Both crystals have luminescence in the 510–650 nm region with a peak of about 540 nm. Crystal samples were cut from ingots grown by the Czochralski method at the Institute for Physical Research [5], Ashtarak, Armenia. Crystal samples of both types had a size of $9 \times 9 \times 1$ mm$^3$. All surfaces were optically polished. The optical parameters of the crystals were studied before and after irradiation with a proton beam.

PROTON BEAM

Crystal samples were irradiated in a 660 MeV proton beam at the phasotron of the Dzhelepov Laboratory of Nuclear Problems, JINR. The beam diameter was about 1.5 cm (FWHM). Samples were placed in the beam center. The total proton fluence through the samples was determined by measuring the induced activity in pure aluminum foil placed in the beam. The aluminum foil had a size of $30 \times 33$ mm$^2$ and was cut into 110 elements of $3 \times 3$ mm$^2$ each, forming a $10 \times 11$ matrix.

When protons pass through the aluminum foil, $^{24}$Na isotopes with a half-life of about 15 h are produced in it. From the known cross section of the interaction of protons with aluminum nuclei and the measured induced activity of the foil, the fluence of the protons passing through the foil is determined with a high accuracy. In our case, we used a different approach. First, the relative induced activity of each $3 \times 3$ mm$^2$ the aluminum piece was measured. Measurement of the activity of the aluminum pieces began about 4 h after the irradiation and lasted about three hours. The initial induced activity of each piece (immediately after irradiation) was calculated from the measured activity of the pieces with allowance for
the lifetime of the $^{24}$Na isotope and the individual delay of the measurement time. The beam profile was constructed using the calculated initial induced activity of each element and then was fitted with a three-dimensional Gaussian distribution. The absolute values for each $3 \times 3$ mm$^2$ aluminum piece were determined from the accelerator beam current during the irradiation of the samples. As a result of the fit, it was found that 31% of the total fluence of the proton beam passes through the central part $9 \times 9$ mm$^2$ where the samples were placed during the irradiation. The beam profile is shown in Fig. 1.

Two exposures of the samples were performed. During the first irradiation, $2.3 \times 10^{13}$ protons or $2.8 \times 10^{13}$ protons/cm$^2$ were passed through the samples. The second exposure added $6.97 \times 10^{13}$ protons, giving in total about $7.2 \times 10^{14}$ protons on sample or $8.9 \times 10^{14}$ protons/cm$^2$.

RESULTS AND DISCUSSION

Transmission of the samples was measured using a Shimadzu SolidSpec-3700 DUV spectrophotometer [6]. Light transmissions of the YAG:Ce and GGG:Ce samples before and after irradiations are shown in Fig. 2. The measurement results demonstrated that after the second irradiation, the transparency of both crystal samples fell by no more than 7% in the region of their own luminescence (approximately 510–650 nm). However, there are differences in the transparency behavior of the two crystals after the first and second exposures. The transparency of the GGG:Ce sample fell mainly after the second exposure, while a significant decrease in the transparency of the YAG:Ce sample occurred immediately after the first exposure.

The luminescence spectra of the samples before and after irradiation were measured using a monochromator of an SF-46 spectrophotometer (manufactured by the “LOMO” company, Leningrad/S. Petersburg, discontinued). Both samples have a luminescence peak at around 540 nm. As an example, the normalized luminescence spectra of a YAG:Ce crystal before and after the second irradiation are shown in Fig. 3. The luminescence of the irradiated sample was measured one year after irradiation. One can see that the shape of the luminescence spectrum of the crystal did not change when $7.2 \times 10^{14}$ protons pass through the sample.
The light output of the samples before and after the second irradiation was estimated by the response of the crystals to the beta source of $^{90}\text{Sr}$. The decay electrons passed through the crystal under study and hit the trigger counter made of a larger YAG:Ce crystal. The registration of energy deposited in the trigger counter allowed us to select electrons intersecting the crystals under study with an energy of about 1.5–2.28 MeV (the beta decay spectrum continues up to 2.28 MeV). The change in the light output of the investigated crystals was determined by the peak position in the spectrum of the electrons from the $^{90}\text{Sr}$ beta decay intersecting crystals.

Protons crossing the crystal samples produce radioisotopes in the crystal due to nuclear reactions [7]. The high residual activity of the crystals after irradiation greatly complicates the measurement of the spectrum of beta decay electrons. First measurements of the irradiated samples were made 9 weeks after the exposure.

Earlier, we developed and tested a method for extracting the contribution from an external source against the background. When subtracting two spectra, from intrinsic radioactivity and from external source plus intrinsic radioactivity, taking into account the fraction of the external source and the intrinsic radioactivity, one can reliably distinguish the contribution from the external source even with its relatively small fraction in the total spectrum [8].

Figure 4 shows the spectra from the non-irradiated YAG:Ce crystal (line 1) and after the second irradiation (lines 2 and 3) due to $^{90}\text{Sr}$ beta source. The beta source contribution to the spectrum of the irradiated sample was obtained by subtracting two spectra: the residual activity spectrum was subtracted from the total spectrum due to residual activity plus external irradiation by the $^{90}\text{Sr}$ beta source.

After the irradiation, both samples were found to show an increase in light output when irradiated with the $^{90}\text{Sr}$ beta source. The first measurement of the YAG:Ce sample taken 9 weeks after the irradiation shows signal increase of almost 30% (line 2) compared with the non-irradiated samples. After 18 weeks, the output signals from the irradiated sample (line 3) were still higher than that from the non-irradiated samples, as seen in Fig. 4 for the YAG:Ce crystal. The increase in light output from the GGG:Ce crystal after irradiation was much smaller than for the YAG:Ce sample, but is still visible. Spectra from the GGG:Ce crystal are presented in Fig. 5. The irradiated sample was

![Graph showing YAG:Ce luminescence spectra before and after irradiation.](image)

![Graph showing spectra from non-irradiated and irradiated samples of the YAG:Ce crystal.](image)

![Graph showing spectra from non-irradiated and irradiated samples of the GGG:Ce crystal.](image)
measured 9 and 18 weeks after the exposure as well. We observed such an effect earlier on some crystals. Apparently, this is due to radioisotopes and excited states arising in the samples as a result of irradiation with protons.

Data taken from both the YAG:Ce and the GGG:Ce samples several months after the irradiation show that eventually light output from the irradiated samples became lower compared to non-irradiated samples. Figure 6 shows the response of the non-irradiated and irradiated YAG:Ce samples to the $^{22}$Na gamma source. The spectrum from the irradiated sample was taken about a year after the irradiation. The full absorption peak shows that the irradiated YAG:Ce sample lost about 30% of the light output compared with the non-irradiated sample.

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