Optical transmission damage of undoped and Ce doped $Y_3Al_5O_{12}$ scintillation crystals under 24 GeV protons high fluence

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Abstract— This report presents results on the optical transmission damage of undoped and Ce doped $Y_3Al_5O_{12}$ scintillation crystals under high fluence of 24 GeV protons. We observed that, similarly to other middle heavy scintillators, it possesses the unique radiation hardness at fluence values as high as $5*10^{14}$ p/cm^2 and it is thus promising for the application in the detectors at High Luminosity LHC. The crystalline structure of the garnet scintillator allows to control and further optimize its scintillation parameters, such as scintillation decay time and emission wavelength, and shows a limited set of the radioisotopes after the irradiation with protons.

Index Terms— Experiments at colliders, inorganic scintillation material, radiation damage, γ-quanta irradiation, proton irradiation

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1.1 Introduction

Future physical programs at the LHC will require a significant increase of the accelerator instantaneous luminosity up to $L = 3 * 10^{34}$ cm^{-2} s^{-1}, and an integrated luminosity of 3000 fb^{-1} to be collected throughout the High Luminosity phase of LHC beyond 2025 [1]. At such beam luminosities, charged hadrons with fluences higher than 10^{14} p/cm^2 per year in the largest pseudo-rapidity regions of the detectors will have a non-negligible influence on the radiation damage of the materials. Moreover, with the increasing activation of the experimental equipment, it will become more difficult to periodically replace and maintain the detector components. Therefore, the selection of materials for new detectors in such high radiation environments requires more reliable assessment of the risks of detector failures due to severe radiation damage. During last few years we performed systematic studies of the damage effects in inorganic scintillation materials [2-9]. We concluded that mid-heavy, Ce-doped materials are very stable under γ-quanta and the most resistant in terms of the damage effects from high-energy protons. Hadron fluence values of the order of $5*10^{13}p/cm^2$ do not appear to set limits to the use of the crystals such as orthosilicates and garnets in the future calorimeter designs. Ce doped garnets, in comparison with orthosilicates, have following advantages: their luminescence is shifted to the green-yellow spectral range which well matches the spectral sensitivity of Silicon Photo Multipliers (SiPMs), garnet structure allows a variety of technological possibilities to engineer the scintillation properties, particularly the scintillation kinetics and the wavelength of the scintillation maximum. The shift of the scintillation light towards the green-yellow spectral range also targets a further reduction of the induced absorption due to the fact that most of the spectral changes in a wide band gap oxide materials under irradiation with high energy protons occur in the UV and blue spectral ranges. For instance, among several colour centers which appear in Lu_2SiO_5:Ce (LSO) after irradiation, two of them, having maxima near 390 nm (3.18 eV) and 470 nm (2.66 eV) respectively, overlap the scintillation band [4]. Since the discovery of scintillation properties of $Y_3Al_5O_{12}$:Ce (YAG:Ce) crystals 40 years ago [10], these crystals have found a wide application in imaging techniques. The technology of YAG crystals was very well developed in the 90's [11]. It was shown that the presence of Ce^{3+} in the crystal prevents the appearance of colour centers in the visible range. More recently, a good radiation hardness of YAG:Ce crystals to γ-irradiation and 150 MeV protons also was confirmed [12,13]. Here we

investigated the change of the crystal optical transmission after the irradiation with 24 GeV protons and a fluence $5*10^{14}$ p/cm^2. The set of the radioisotopes after the irradiation with protons is also described.

1.2 Samples and experimental setup

Studied crystals were produced by CRYTUR (Turnov, Czech Republic) [14] by the Czochralski method. The samples were cut from the middle part of the ingots and had dimensions 10x10x10mm^3 with two opposite sides polished. The first crystal was undoped (#2962), the second one (#2961, slightly doped) has trace of Ce^{3+} at the level less than 0.01 atomic % and the third one (#2960) was a sample from a Ce doped ingot used for commercial production. An additional YAG Ce doped sample of dimensions 10x10x10mm^3 was used to measure the set of radio-isotopes in the crystal after irradiation with 24GeV protons to a fluence of $3*10^{13}$ p/cm^2. This sample become available for the radio-isotope set measurements with germanium detector at CERN Radiation Protection Service in one month after irradiation.

The samples were irradiated with 24GeV protons of the PS accelerator at CERN with the 10^9 p/cm^{2}s flux in a plastic container. Fluence was measured by activation of Al foil. The samples #2962, 2961, 2960 were irradiated to a fluence of $5*10^{14}$ p/cm^2. Manipulation with irradiated samples was performed by CERN Radiation Protection Service. When crystals become available for measurements in three moth after irradiation, their optical transmission was measured by Varian spectrophotometer at CERN authorized facilities allowing operations with radioactive samples. All measurements were performed at room temperature. The details of the irradiation procedure, the experimental methods and the spectra deconvolution analysis are described elsewhere [2-4].

1.3 Experimental results and discussion

Figures 1-3 show the optical transmission spectra of the samples measured before the irradiation and 3 months after the irradiation. The transmission curve of the doped crystal (#2960) does not show significant changes in the spectral range of scintillation whereas the undoped (#2962) and the slightly doped (#2961) samples demonstrate a more complex behavior over the whole spectrum. Table 1 summarizes the colour centers observed in the studied YAG samples.

Figures 4-6 show the induced absorption spectra of the YAG samples after the proton irradiation. To make an approximation by Gaussians, an energy scale in eV was used. At the deconvolution of spectra we used prior information, such as peak position and band width, of several bands of color centres which were observed in the YAG:Ce after γ-irradiation [15,16] and Ce^{3+} ions as well. The Pearson's χ^2 chi-square criteria was used to obtain best approximation of experimental curve with set of Gaussians.

The induced absorption spectrum of the undoped YAG crystal #2962 contains several bands with maxima located in the vicinity of 747 nm (1.66 eV, C1), 480 nm (2.58 eV, C2), 409 nm (3.03 eV, C3), 348 nm (3.56 eV, C4), 277 nm (4.47 eV, C5). The center C1 was also detected in the undoped as-grown crystals [11] and most probably appears due to crystal growth defects. The colour center similar to C2 was previously observed in the γ-irradiated crystal [15]. The drop of the induced absorption below zero in the vicinity of 277 nm is caused by the conversion of the colour centers located in UV spectral range and having band maximum near 277 nm. Due to their conversion a strong induced absorption band with maximum around 348 nm appears in the induced absorption spectrum. It is worth to note that only the C1 and C2 colour center absorption bands marginally overlap with the Ce^{3+} luminescence band in the crystal.

The crystal #2961 contains two weak Ce^{3+} bands with maxima near 460 and 345 nm and a sharp band near 370 nm in the initial spectrum before irradiation. The last band is due to the F^+ colour center as described in article [16]. The effect of the conversion of the 277 nm band in the induced absorption spectrum was not observed. It seems that the set of the colour centers which appear in the crystal after the crystal growth is sensitive to the presence of Ce, even if in a small amount (<0.01 atomic %). Moreover, the transmission spectrum in the range above 500 nm is occupied by the color centers due to the defects of the crystal growth. We did not observe the F^+ centers causing 230 nm absorption band in the initial spectrum because of the large optical density in that spectral region, however its presence became visible through its conversion after the crystal was irradiated.

A strong peak with the maximum near 248 nm, which appears due to its conversion into the F center, was detected. A conversion of the F+ center having maximum 370 nm into the F center is also observed similar to the irradiation with γ-quanta [16]. Due to the presence of the F^+ centers and traces of Ce^{3+}, the deconvolution of the induced absorption spectrum becomes

more complicated. Nevertheless, similar to the undoped crystal, both C1 and C2 can be identified in the slightly doped sample #2961.

The sample #2960 does not contain the colour centers related to the defects of the crystal growth like C1 and C2. Obviously, the presence of the Ce activator improves the quality of the crystal structure and minimizes the amount of oxygen defects in the crystal. Cerium ions can be stabilized in a 3+ and 4+ valence states in oxides. When Ce^{3+} substitutes yttrium Y^{3+} in the matrix host, no charge compensation is required. However, the stabilization of Ce^{4+} in the Y^{3+} position introduces a non-compensated negative charge. This reduces the amount of the oxygen vacancies, which introduce non-compensated positive charge in the crystal. In fact, two C^{4+} ions compensate one oxygen vacancy. This assumption is proven by the absence of F^{+}, F- centers absorption bands in the spectra before and after the irradiation of YAG:Ce. The spectral regions 2.5-2.8 and 3.5-3.8 eV are blocked by strong f→d Ce^{3+} electronic transitions, therefore precise deconvolution of the induced absorption spectrum of the sample #2960 was difficult. We observed induced absorption bands dedicated only to the C3, C4, C9, C0 centres. Moreover, as seen from the deconvolution analysis, the peak maxima of the colour centers in the YAG:Ce crystal are two times smaller than in the undoped samples. It indicates that these colour centers are related to the defects of the crystal growth and their content is decreased due to the higher concentration of Ce dopant. The only color center which can be attributed to the proton irradiation damage is the C8 center, however its observation is masked by the strong absorption band related to the interconfiguration f →d transition in the YAG:Ce sample.

We did not observe in the Ce doped crystal other new defects due to the irradiation with protons. This indicates that the clusters and interstitials atoms produce the color centers whose absorption bands are likely to be located in the spectral range above 5 eV. Given the presence of Ce^{3+} ions, their observation by the standard optical spectroscopic methods is difficult. In addition, a shift of the fundamental absorption cut-off due to the strong absorption of the C11 center in this spectral region was not observed.

1.4 Irradiation-induced radio-isotopes

Hadrons induce the nuclear reactions and produce the radioisotopes [8,9]. The radioisotopes emit α-, β-, and γ-radiation, initiating scintillation process in the crystals and thus producing a background light in the crystal. The higher the scintillator light yield is, the higher the light background in the crystal from the activity of the radioisotopes occurs.

Table 2 reports the set of the radio-isotopes measured in YAG:Ce scintillation crystal one month after the irradiation with protons. Besides the isotopes created by the nuclear reactions (p, Y), the relatively long living ^{22}Na isotope due to (p,Al) reaction is also detected. ^{22}Na isotope, is a 511 keV γ-quanta emitter. Considering the typical YAG:Ce light yield of about 20000 ph/MeV, this isotope could increase the flux of the parasitic photons due to radio-luminescence. A comparison with Y_2SiO_5:Ce crystal can be found in earlier study [17]. However, the density and the effective charge Z_{eff} of YAG are relatively small in comparison with Lu_2SiO_5 or $Lu_3Al_5O_{12}$ crystals [18], therefore a significant increase of the parasitic photons in the YAG:Ce detecting elements with thickness of a few mm is not expected. We also note that the undoped YAG crystal is an intrinsic scintillator with the emission wavelength located in the UV spectral range due to the radiating recombination of self trapped excitons and self trapped holes [19]. These scintillation processes will be subject to a strong negative influence of the colour centers C3-C9. Thereby, the undoped YAG is not an optimal candidate for application in a high dose radiation environment.

1.5 Conclusions

The above analysis shows that the YAG:Ce scintillating crystals can be used in the detectors of the ionizing radiation and can sustain heavy radiation loads without significant loss of performance. Even the hadronic component of the ionizing radiation is unlikely to be a limiting factor. Moreover, high-energy protons do not appear to create a large amount of new colour centers in the crystal. In particular, the induced absorption coefficient in the spectral range of scintillation (540 nm) remains below $1m^{-1}$ after irradiation with 24 GeV protons to a fluence of $5*10^{14}$ p/cm^2. Thus, material is bright and radiation tolerant scintillator to construct calorimetric modules and segmented detector arrangements operating in harsh radiation environments at the future high luminosity collider experiments, particularly at the High Luminosity LHC.

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Table 1.Colour centers observed in YAG crystals after crystal growth, irradiation 24 GeV protons.

Sample	#2960	#2961	#2962	Description of the defect
C1, nm (eV) / peak amplitude, m^{-1}		736 (1,69) / 3,7	747 (1.66) / 2,4	Crystal growth defect
C2, nm (eV) / peak amplitude, m^{-1}		480 (2.58) / 1,2	480 (2.58) / 6,3	Crystal growth defect
C3, nm (eV) / peak amplitude, m^{-1}	405 (3,1) / 4		409 (3.03) / 8,4	Crystal growth defect
C4, nm (eV) / peak amplitude, m^{-1}	367 (3,4) / 8		348 (3.56) / 23,6	F-center due to conversion of C5
C5, nm (eV) / peak amplitude, m^{-1}			277 (4.47) / -27.7	F+ center on a base of crystal growth defect
C6, nm (eV) / peak amplitude, m^{-1}		386 (3,22) / 42		F-center due to conversion of C5
*C7, nm (eV) / peak amplitude, m^{-1}		370 (3,35) / -68		F+ center on a base of crystal growth defect
C8, nm (eV) / peak		335 (3,7) / 25		

amplitude, m^{-1}				
C9, nm (eV) / peak amplitude, m^{-1}	307 (4,15) / 45	307 (4,04) / 90		Crystal growth defect
C10, nm (eV) / peak amplitude, m^{-1}	248 (5) / 80	248 (5,0) / 210		F-center due to conversion of C10
*C11, nm (eV) / peak amplitude, m^{-1}		230 (5,4)		F+ center on a base of crystal growth defect

* was observed in the crystal with the low Ce doping concentration [16]

Table 2. The set of the radio-isotopes and its activity measured in the YAG:Ce scintillation crystals 1 month after the irradiation with 24GeV protons with a fluence $3*10^{13}$ p/cm^2. The volume of the sample was 1cm^3. An error of the activity measurements with Germanium Detector was at the level of 20 Bq. The half-life time data are listed in the table to show that no long living radioisotopes have been created after proton irradiation.

$Y_3Al_5O_{12}$:Ce		
Nuclide	Half-life, days	Activity, Bq
Be-7	5,31E+01	6,8E+02
Na-22	926	2,1E+03
Se-75	1,20E+02	2,7E+02
Rb-83	8,62E+01	1,06E+03
Rb-84	3,28E+01	5,6E+02
Y-88	1,07E+02	3,74E+03
Zr-88	8,34E+01	1,8E+02

Figure captions

Fig. 1. Optical transmission spectrum of crystal #2960 measured at room temperature before irradiation and 3 months after irradiation with 24 GeV protons to a fluence of $5*10^{14}$ p/cm^2.

Fig. 2. Optical transmission spectrum of crystal #2961 measured at room temperature before irradiation and 3 months after irradiation with 24 GeV protons to a fluence of $5*10^{14}$ p/cm^2.

Fig. 3. Optical transmission spectrum of crystal #2962 measured at room temperature before irradiation and 3 months after irradiation with 24GeV protons to a fluence of $5*10^{14}$ p/cm^2.

Fig. 4. Proton-irradiation-induced absorption spectrum of YAG sample #2962 and its approximation by a set of Gaussian type bands, O-experimental points. Pearson's chi-square test value $\chi^2 = 2 \cdot 10^{-5}$.

Fig. 5. Proton-irradiation-induced absorption spectrum of YAG:Ce sample #2961 and its approximation by a set of Gaussian type bands, O-experimental points. Pearson's chi-square test value $\chi^2 = 2 \cdot 10^{-5}$.

Fig. 6. Proton-irradiation-induced absorption spectrum of YAG:Ce sample #2960, O-experimental points. The inset shows the change of the induced absorption in the scintillation spectral range.

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