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Drastic scintillation yield enhancement of YAG:Ce with carbon doping

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Scintillation performances of carbon-doped YAG and YAG:Ce crystals obtained by the Czochralski method under novel conditions of Ar+CO reducing atmosphere are compared to their counterparts obtained by conventional technologies. While light yield and energy resolution in YAG crystals grown under different conditions are similar, a fast luminescence decay component of 4-6 ns was observed in YAG:C grown by the new procedure. Optimization of post-growth thermal annealing procedure of YAG:Ce,C scintillator provides the very high light yield of 28200 phot MeV⁻¹, and the energy resolution of 7.8 – 8.5 % at 662 KeV.

1. Introduction

New high-energy physics experiments at colliders (see, for ex., ^[1]), as well as future generation of medical tomography equipment ^[2] need bright scintillators with faster luminescence decay compared to currently widely used Ce³⁺-doped scintillators. Y₃Al₅O₁₂ (YAG) garnet crystals are considered as an efficient host for laser, phosphor, and scintillation materials. Scintillation properties of undoped YAG were barely explored in detail due to low light yield and slow luminescence decay. Meanwhile, Ce-doped garnets did not attract much attention until the development of extremely efficient Al/Ga-substituted multicomponent garnets with the light yields of up to 58000 phot MeV⁻¹.^[3,4] This value is near the theoretical limit considering the energy gap of these materials.^[5] The improvements in the light yield were attributed to the suppression of electron capture on shallow traps due to the electronic structure modification by cationic substitution.^[6]

In this sense, it is worth to explore whether it is possible to reduce carrier trapping and improve the scintillation performance in “simple” garnets, such as YAG:Ce, without considering solid solutions with other cations. This is important, because Gd₃Al_{5-x}Ga_xO₁₂:Ce (GAGG:Ce) and related Ga-containing crystals have to be grown in inert gases enriched with oxygen to reduce Ga evaporation. Only Ir crucibles are capable to withstand such atmosphere at temperatures near 2000 °C, if oxygen content does not exceed 1-2 vol.%. Nevertheless, Ga evaporation from the melt under these conditions is still rather strong. This, evidently, causes the tendency of crystals to cracking. As a result, the use of Ir crucibles and expensive thermal insulation material, as well as the high energy consumption during the crystal growth of rare-earth garnets, makes the crystals expensive for some applications. The economic factor is a problem also for other high-melting point complex oxide scintillators, such as Lu₂SiO₅:Ce (LSO:Ce) and Lu_{2-x}Y_xSiO₅:Ce (LYSO:Ce). All these factors promote the development of low-cost crystal growth technologies.

Procedures for aluminum garnets and perovskites growth in Mo and W crucibles have been developed since 1970-80s ^[7-9] until much more recently.^[10-12] Meanwhile, a new procedure of growth ^[13, 14] and post-growth heat-treatment ^[15] for garnet-based crystals was developed by some of authors of this work, and a series of undoped and Ce-doped YAG crystals were produced. Among the basic features of the novel procedure there is the use of cheap graphite heat insulation in crystallizer and reducing Ar+CO growth atmosphere. The latter is a cause of few hundreds wt. ppm (up to 1 at.%) carbon incorporation in crystals ^[15]. Since carbon impacts on YAG optical properties, ^[15] the crystals obtained in these conditions were denoted as carbon-doped crystals (YAG:C and YAG:Ce,C).

In this paper the scintillation parameters of YAG:C and YAG:Ce,C crystals grown by the developed procedure are compared with YAG and YAG:Ce grown by the conventional technologies.

2. Methods

Crystal growth: YAG:C and YAG:Ce,C crystals were grown by the developed procedure in Mo and W crucibles using graphite heat insulation under Ar+CO reducing atmosphere. The growth procedure details are described elsewhere. ^[13, 14] The carbon concentration was up to 1 at.% in YAG:C crystals. The Ce concentration in YAG:Ce,C was around 0.1 at.%. Carbon concentration was not measured in YAG:Ce,C, but it is presumed to be the same as in YAG:C, because the crystal growth procedure was the same. Cerium was introduced in the form of CeAlO₃ single crystals, whose production technology was developed by us in. ^[16] As-grown YAG:C were greenish, and YAG:Ce,C were yellow-greenish as the result of interaction with the reducing atmosphere (**Figure 1**). The post-growth heat treatment of crystals at 1200 °C under air and at 1500 – 1850 °C under Ar+CO atmosphere provided the

bleaching of YAG:C and less deep yellow coloration of YAG:Ce,C. The reference YAG and YAG:Ce crystals were grown by the Czochralski method from Ir crucibles in Ar gas atmosphere. Samples with the dimensions of $10 \times 10 \times 2 \text{ mm}^3$ were fabricated for optical and scintillation measurements.

Figure 1. The view of as-grown YAG (a), YAG:C (b), and YAG:Ce,C (c) crystals, and air-annealed YAG:Ce,C scintillation element (d).

Measurement of spectral properties: The absorption spectra were measured using a Specord 40 spectrophotometer (Analytik Jena AG). X-ray luminescence (RL) spectra were measured using a home-made apparatus featuring a CCD detector (Jobin-Yvon Spectrum One 3000)

coupled to a monochromator (Jobin-Yvon Triax 180). RL irradiation was performed by a Philips 2274 X-ray tube.

Determination of light yield and energy resolution: Light yield (LY) and energy resolution (R) were measured under irradiation with a 662 keV ^{137}Cs gamma source. A R1307 Hamamatsu PMT ran at 800 V and HV with linear dynode voltage divider were used for registration of YAG:Ce,C parameters. Meanwhile, a R2059 Hamamatsu PMT run at 1400 V with better sensitivity in UV-range was used for measurements with YAG:C. PMT output was connected to the charge-sensitive preamplifier BUS 2-94 and BUI-3K shaping amplifier. Signal from preamplifier was shaped by a shaping amplifier with 2 μs (for YAG:C) and 8 μs (for Ce-doped YAG) shaping times. In order to collect the whole scintillation light the samples together with open part of PMT photocathode were covered with Teflon reflector. No optical contact between samples and PMT window was provided. For absolute light yield determination the yields were compared to $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ (BGO) produced at ISMA, and Lu_2SiO_5 (LSO:Ce) sample obtained from Proteus Inc., USA. The quantum efficiencies were 0.1867 (BGO), 0.1158 (YAG:Ce,C), 0.2578 (LSO:Ce) for measurements with R1307 PMT, and 0.1472 (BGO) and 0.2108 (YAG:C) for measurements with R2059 PMT based on the luminescence spectra of the crystals and the PMT quantum efficiency. Energy resolutions of ^{137}Cs 662 keV peaks are determined by approximation of the obtained pulse-height spectra with Gaussian function.

Determination of decay times: Scintillation decay times were determined at X-ray excitation (50 kHz excitation repetition rate, FLE 400 filter $\lambda > 400\text{nm}$). Pulsed X-ray decay measurements were performed using a Hamamatsu N5084 light-excited X-ray tube set at 30 kV as irradiation source. The optical excitation of the tube was performed with a Hamamatsu PLP-10 picosecond light pulser. The scintillation decay was registered by a PMA 165-C PMT from PicoQuant operating in single photon counting regime. According to the data sheet, the

transit time is below 180 ps and the rise and fall time are 750 ps providing a time resolution of 180ps.

3. Experimental results and discussion

3.1. YAG:C

The as-grown crystals are colored, while the annealed crystals are highly transparent both in UV- and in the visible. In annealed YAG:C the color centers are not re-formed under any subsequent reducing or oxidizing annealing (see ^[15] for details). The irreversible bleaching was observed for YAG:C crystals grown in W, Mo, and Ir crucibles (if raw material was prepared under Ar+CO atmosphere). This means that the possible introduction of crucible material atoms in the crystals is not responsible for this phenomenon. The lack of coloration after subsequent annealing was attributed ^[15] to the existence of competing centers of electron capture linked to complex C-related defects. Thus, a high transparency of annealed YAG:C crystals and their scintillation parameters sustain under any further annealing.

Light yield (10000 – 14000 phot MeV⁻¹), energy resolution (17 % at 662 keV) and scintillation decay ($\tau = 400$ ns + slow component) of annealed crystals are similar to those reported in,^[11, 16, 17] while light yield and energy resolution of as-grown YAG:C are significantly worse due to the formation of color centers (Table 1).

Table 1. Scintillation parameters of YAG:C and YAG crystals.

	Colored(as grown)YAG:C (this work)	Colored YAG	Transparent (annealed) YAG:C (this work)	Transparent YAG
Light yield, phot/MeV	<10000	50000/5.5 MeV ^[11]	up to 14000	14300 ^[16] 60000/5.5MeV ^[11]
Luminescence decay times, ns	~5 + slow	3-4 + slow ^[17] 35, 179, 974 ^[11]	~ 400 + slow	460 [16] 3-4, 750-1000 ^[17] 51, 373 ^[11]

Energy resolution at 662 keV, %	Not identified	16-18 ^[17]	17	11-14 ^[17]
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Meanwhile, colored YAG:C, apart of few hundred ns component ascribed to the self-trapped exciton (STE) emission, and long microsecond components, possesses a very fast luminescence decay component with $\tau = 4\text{-}6$ ns under X-ray excitation. The decay curves are well fit by the three-exponential decay. The contribution W_1 of the fast component with $\tau_1 = 4.8$ ns at fitting (**Figure 2**) to the light sum within the 30 microsecond gate is $\sim 2\%$, according to the equation 1.

$$W_1 = \frac{\int \tau_1 A_1}{\int \tau_1 A_1 + \int \tau_2 A_2 + \int \tau_3 A_3} * 100 \quad (1)$$

The fast nanosecond component under selective UV-excitation and synchrotron excitation was already reported before in the literature. Probably, the most detailed analysis of emission centers in undoped garnets was presented in ^[18] where the 2-3 ns decay components with emission band peaked around 400 nm (3.095 eV) and excitation at 3.33; 5.37; 6.56; 7.3 eV were attributed to F^+ -centers localized near Y_{Al} antisite defects. The same luminescence band peaked at 397 nm and excited at 370 and 236 nm was registered in our YAG:C crystals^[19] (in that work they are denoted as “Type III” crystals). Similar data on fast (<10 ns) luminescence decay in garnets by γ -ray excitation were reported previously in, ^[17] but it is impossible to evaluate its contribution based on the there presented data.

Figure 2. Scintillation decay after X-ray irradiation of colored YAG:C crystal fit by the function $y = 0.003 + 0.888 \cdot \exp(-t/4.8) + 0.064 \cdot \exp(-t/296.4) + 0.058 \cdot \exp(-t/3137.9)$; inset: the same data within the time range of first 60 ns.

X-ray excited luminescence spectra are shown in **Figure 3**. While YAG spectrum demonstrates just one dominant band peaked at 310 nm, which is evidently a superposition of emission of excitons trapped near antisite defects ^[20] and Gd³⁺ impurity emission, the spectrum of transparent YAG:C crystals contains much wider luminescence band in the same range with the main peak near 300 nm of AD-related exciton emission, and shoulder at ~245 nm attributed to self-trapped exciton emission. The wide 300 nm peaked band is a superposition of peaks of exciton emission related to different types of AD-related defects.

Unlike transparent crystals, in colored YAG:C the complex luminescence band arises at >395 nm, which is evidently related to the registered fast F⁺-center emission. Similar luminescence band was observed in YAG grown under Ar+H₂ atmosphere ^[11], and the persistent blue luminescence was revealed in carbon-doped YAG ^[21, 22], which was proposed for dosimetry applications. However, we have not noticed a clear correlation between the contribution of the fast luminescence decay component and intensity of the >370 nm luminescence bands on X-ray luminescence spectra.

Figure 3. Typical X-ray luminescence spectra of YAG and YAG:C crystals

3.2. YAG:Ce,C

While the light yield of as-grown YAG:Ce,C is similar to the light yields of YAG:Ce reported in most of the papers,^[23-25] the light yield of YAG:Ce,C after thermal annealing under air or Ar+CO atmosphere increases by 1.5 - 2.0 times compared to the as-grown crystal (**Figure 4**, **Table 2**). The highest light yield registered after annealing in air is 330 % relatively to BGO taking into consideration the quantum efficiency of these scintillators with a R1307 PMT. Assuming the averaged light yield of BGO reference sample of 8600 phot MeV⁻¹ according to 8000 – 9000 phot/MeV values prevailing in literature,^[23, 26-28] the light yield of air-annealed YAG:Ce,C is 28200 phot MeV⁻¹ (**Table 2**). The LSO:Ce reference light yield of 28000 phot/MeV determined by our procedure lies within the standard values for this scintillator, see for example, ^[29] and confirms the correctness of our data. **To the best of our knowledge, just one paper reports the similar light yield around 30000 phot/MeV in YAG:Ce, though without the pulse height spectrum ^[30] (**Table 2**).** The energy resolution in air-annealed sample presented in **Figure 4** is 8.1 %, though it ranges from 7.8 to 8.5 % in different samples.

Figure 4. Pulse height spectra of YAG:Ce,C crystals compared to the reference BGO and LSO:Ce samples.

Table 2. Scintillation parameters of YAG:Ce,C crystals compared to their counterparts obtained by the conventional technologies.

Crystal	Growth method	Crucible material	Light yield [phot MeV ⁻¹]	Energy resolution [%]	Scintillation decay time [ns]	Background level [%] after 3 μ s
YAG:C,Ce	Cz ^{a)} , as-grown	Mo/W	14200	9.7	73 (30%), 254 (70%)	2.94
YAG:C,Ce	Cz ^{a)} , CO-anneal.	Mo/W	21700	12.9	100 (77%), 331 (23%)	1.77
YAG:C,Ce	Cz ^{a)} , Air-anneal.	Mo/W	28200	7.8 -8.5	98 (79%), 349 (21%)	1.30
YAG:Ce [10]	HDC ^{b)}	Mo	15000-18000	8-10	No data	No data
YAG:Ce [23]	Cz ^{a)}	No data	16700	No data	No data	No data
YAG:Ce [24]	Cz ^{a)}	No data	No data	No data	119+slow	No data
YAG:Ce [30]	Cz ^{a)} , Air-anneal.	Mo	30260	No data	No data	No data

a) Czochralski method; b) Horizontal Directional Freezing method

The growth and post-growth annealing under the reducing conditions should promote the reduction of cerium into the optically active trivalent state. We presume that in as-grown YAG:Ce,C a high Ce³⁺ concentration does not lead to light yield increase because of high concentration of defects in crystals. The light yield improvement after the reducing annealing coincides with the suppression of the absorption band peaked at 371 nm and related to F⁺ centers, ^[18, 19] as shown in **Figure 5**. As one might expect the inverse effect of increase of trap

and color center concentrations at reducing annealing, we attribute the absorption decrease, by analogy to YAG:C, to the presence of carbon, which creates carrier capture centers competing for electrons with F and F^+ centers. Meanwhile, the energy resolution in YAG:Ce,C deteriorates after the reducing annealing.

Figure 5. Absorption spectra of YAG:Ce,C crystals; on the inset – difference spectra between air- and Ar+CO-annealed samples.

The air annealing significantly improves both light yield and energy resolution. This is evidently attributed to the elimination of oxygen vacancies and related defects. According to the intensity of Ce^{3+} absorption bands at 458 and 340 nm, and absorption band around 300 nm attributed to $Ce^{4+} - O^{2-}$ charge transfer complexes,^[31] the weakening of Ce^{3+} bands and reinforcement of $Ce^{4+} - O^{2-}$ band in the air-annealed sample compared to CO-annealed sample (see the difference spectrum at the inset in **Figure 5**) certify a partial oxidation of Ce^{3+} into Ce^{4+} . According to^[32], Ce^{4+} competes for electron capture with shallow electron traps in garnets, and the coexistence of Ce^{3+} and Ce^{4+} reduces electron trapping and promotes fast and bright scintillation.

Comparison of X-ray luminescence spectra (**Figure 6**) indicates that both position and shape of Ce^{3+} luminescence bands at 530 nm are the same in YAG:Ce and YAG:Ce,C, and they are

not affected by the thermal treatments of YAG:Ce,C. Note that the most intense X-ray excited Ce^{3+} luminescence is registered in CO-annealed samples, while air-annealed YAG:Ce,C possesses the highest light yield. The 310 nm complex band in YAG:Ce, by the analogy to YAG, is related to Y_{Al} antisite luminescence and Gd^{3+} emission. The excitation peak at 273 nm and emission peak at 312 nm clearly related to Gd^{3+} luminescence were detected in YAG:Ce spectra (not shown in this paper). Meanwhile, the weakening of the band peaked 310 nm suggests the decrease of concentration of antisites in Ce,C-codoped crystals. The absence of Gd^{3+} -related peak in carbon-codoped samples can be attributed to the enhanced energy transfer from Gd^{3+} to Ce^{3+} . Therefore, the improved energy transfer to Ce^{3+} luminescence centers is an additional reason of the light yield improvement in YAG:Ce,C scintillator.

Figure 6. X-ray luminescence spectra of YAG:Ce,C.

Pulsed X-ray luminescence decay curves of YAG:Ce,C within the 2 μ s range were fit by the double exponential function $y = y_0 + A_1 \cdot \exp(-t/\tau_1) + A_2 \cdot \exp(-t/\tau_2)$. The scintillation decay constants of as-grown YAG:Ce,C (**Table 2**) are similar to those published in literature (see, for ex., ^[24]) with the fast ($\tau_1 = 73$ ns) and slow ($\tau_2 = 254$ ns) components. Although the calculated decay constants are somewhat longer in the case of the annealed samples (98 ns, and 100 ns), the overall decay is faster, as one may see in **Figure 7**, due to the decrease the slow component contribution down to 21 – 23 %. The luminescence decay almost does not depend on the annealing atmosphere (air or Ar+CO) suggesting that the annealing temperature is the main factor calling the reduction of carrier trapping responsible for the delayed luminescence. The phosphorescence level (background) after 3 μ s decreases by more than twice after the thermal annealing (see Table 2). Despite the very high measured light yield, we suppose that there is still room for further improvement of scintillation parameters by the optimization of Ce concentration and other parameters of the preparation process.

Figure 7. Luminescence decay curves after X-ray irradiation of YAG:Ce,C crystals (blue scatters – as-grown YAG:Ce, red scatters – CO-annealed YAG, black scatters – air-annealed YAG).

4. Conclusions

Scintillation performance of carbon-doped YAG and YAG:Ce crystals is explored. The YAG:C parameters are similar to those in YAG obtained by other methods. Meanwhile the fast luminescence with $\tau \sim 5$ ns attributed to F^+ -centers under extrinsic excitation was revealed in as-grown YAG:C.

High-quality YAG:Ce,C crystals were obtained by the proposed Czochralski process using Mo/W crucibles and optimized post-growth annealing with a very high light yield of up to 28200 phot MeV⁻¹ and a good energy resolution of 8 % at 662 keV. The contribution of fast ~ 100 ns luminescence decay component in air-annealed YAG:Ce,C reaches 79 %.

The obtained scintillation parameters are the result of complex interactions between electrons and holes with cerium in different valence states, carbon, as well as various types of intrinsic defects in the garnet structure. Thus, scintillation mechanisms in YAG:Ce,C still need for detailed study.

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Light yield of YAG:Ce scintillator was drastically enhanced by carbon-doping in

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Keywords: Scintillators, YAG:Ce, light yield

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Drastic scintillation yield enhancement of YAG:Ce with carbon doping

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Pulse height spectra of air-annealed YAG:Ce,C crystal.