An improvement of the pulse shape discrimination properties of Gd₃Ga₃Al₂O₁₂:Ce single crystal scintillator

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Introduction

Single crystals of Gadolinium gallium aluminum garnet (Gd₃Ga₃Al₂O₁₂, also called GGAG) doped with Cerium (Ce) has shown the best combination of the scintillation characteristics exhibiting high density (6.7 g/cm³), effective atomic number (55), light yield [LY] of 57,000 ph/MeV and fast decay time of 55 ns [1]. The fast emission at 550 nm makes this scintillator very useful in the fabrication of compact detectors based on silicon photo-sensors. However the mentioned scintillation properties in GGAG:Ce crystal have shown a strong correlation with various co-doping. Tyagi et al., have reported the change in light output and timing properties of GGAG:Ce single crystals with co-doping of boron, barium and calcium. It is due to the change in defect centers inside GGAG:Ce crystal structure by co-doping [2]. GGAG:Ce has shown a great potential in pulse shape discrimination (PSD) of charged particles and gammas with a high figure of merit (FOM) of 3.46. However, the role of co-doping on the PSD properties is yet to be understood. Therefore, a detailed study of different codoped and annealed samples of GGAG:Ce would help us to understand its decay time and PSD mechanism.

In this communication, we have carried out experiments to study the effect of codoping and post annealing temperatures on the scintillation kinetics and PSD properties of GGAG:Ce scintillators. Thermo-luminescence (TL) and green light stimulated luminescence (GSL) measurements were carried out to understand the role of defect centers altered by the co-dopant and the relation with PSD properties.

Experimental details

GGAG single crystals doped with Ce and having different co-dopants were grown via Czochralski technique. The samples were cut from the grown crystals and polished optically. The samples were annealed after the growth at about 500 °C and 1000 °C for 10 h to alter the oxygen related defect centers. The processed samples were coupled to a 1" Hamamatsu PMT using optical grease. The scintillation measurements were carried out with Am-Pu alpha and 60Co gamma sources. Decay curves of alpha particles and gamma rays for all the samples were obtained in a fast Tektronics oscilloscope. The PSD of alpha and gamma excitations in GGAG:Ce, GGAG:Ce,B and GGAG:Ce,Ca samples was carried out employing charge integration method in a CAEN V1730 16 channels digitizer. In order to study the effect of co-doping on PSD properties of GGAG:Ce samples, the TL and OSL studies was performed on Lexsyg research imaging TL-OSL-RF system. The TL measurements incorporate preheating upto 350 $^{\circ}$ C at 5 A $^{\circ}$ C/s and cooling is done upto 50 A $^{\circ}$ C. Then samples were irradiated by beta (Sr-90) irradiation unit which provides a homogeneous irradiation field at a dose rate of about 0.1 Gy/s for 20 sec. Subsequently, TL was performed and the samples were heated upto 260 °C at a rate of 5 A °C/s. After TL, preheating with instructions mentioned earlier was carried out to get rid of filled trapped centers for GSL measurement. A green light stimulation (525 nm) performed by LEDs is used for OSL excitation passing through a filter of 365 nm.

Results and discussion

Fig 1. shows the scintillation decay time curves of GGAG:Ce, GGAG:Ce,Ca and GGAG:Ce,B samples excited with alpha gamma sources.

Sample	Channel Number	FWHM	Alpha			Gamma				
			τ ₁ (ns)	τ ₂ (ns)	$\begin{array}{c} \tau_{lpha_avg} \ (ns) \end{array}$	τ ₁ (ns)	$ au_2 ext{(ns)}$	$\begin{array}{c} \tau_{\gamma_avg} \\ (ns) \end{array}$	$\frac{\tau_{\alpha_avg}}{\tau_{\gamma_avg}}$	α/γ Ratio
GGAG:Ce	1226	134	261	47	106	354	53	93	1.1	0.11
GGAG:Ce,Ca	918	101	119	35	54	83	36	48	1.1	0.10
GGAG:Ce,B	976	100	497	175	342	462	69	164	2.1	0.14
$GGAG:Ce,B + annealed at 500 \ ^{\circ}C$	1413	129	143	630	295	58	569	90	3.3	0.10
GGAG:Ce,B + annealed at 1000 °C	1466	148	104	501	284	61	488	108	2.6	0.17

Table.1 Decay components of alpha and gamma excitations in GGAG:Ce, GGAG:Ce,Ca and GGAG:Ce,B annealed at 500°C and 1000°C.

The decay curves have been fitted exponentially having two components. The dependence of scintillation decay times for alpha and gamma excitations on co-doping of boron and calcium in GGAG:Ce crystals have been presented in table 1. The decay time measurements of boron co-doped samples showed the highest difference amongst them which would lead to best PSD properties. The results in the table also demonstrate that the oxygen vacancies were effectively affected by the post-growth annealing which consequently affects the PSD properties. The ratio of average decay times of alpha and gamma gives an insight on the crystal's PSD ability. The alpha and gamma average time difference and α/γ ratio is also maximum in GGAG:Ce,B crystal with post growth annealing at 1000 °C, thus indicating highest potential for PSD.

Therefore, the defect centers in GGAG:Ce have a significant role on the dependence of scintillation decay time pulse shapes with the kind of incident radiation. The TL was, therefore, carried out as it is a tool to extract information about the defect centers of co-doped GGAG:Ce samples. However, the TL of different GGAG:Ce co-doped samples had only shown change in the presence of number of shallow defect centers. We have observed that calcium codoping drastically reduces trap centers upto 250 °C. While the effect of boron co-doping lies in between of GGAG:Ce and of GGAG:Ce,Ca. But when stimulated with green light, OSL data signifies that Ca co-doped crystals have deep trap centers, while no emission was observed in OSL of GGAG:Ce and GGAG:Ce,B samples. Despite showing maximum change in timing properties due to reduction in trap centers, calcium codoped sample showed no PSD ability owing to its deep trap centers confirmed by GSL. The results of TL and GSL would be presented later in details.



Fig. 1 Alpha and gamma scintillation decay curves of boron and calcium co-doped GGAG:Ce samples.

Summary

The pulse shape discrimination capabilities were observed to be dependent on the presence of co-doping and after growth annealing treatment temperatures. The TL and OSL results further indicated the role of defect centers on charged particle and gamma PSD characteristics of GGAG:Ce scintillators. Thus in GGAG:Ce single crystal, structure of defect centers along with the charge density quenching plays an important role in the PSD capability of these single crystals.

References

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