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Research Article

Optical characteristics of single crystal $Gd_3Al_2Ga_3O_{12}$: Ce

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Abstract

New emerging high-energy radiation detection techniques are based on the use of rare-earth ion doped materials. There is a great demand for new inorganic scintillators for medical applications, including X-ray and γ radiation detection. In these applications, the new scintillating materials must comply with the main requirements such as high optical quality, high light yield, short response time etc. Materials satisfying these requirements include $Gd_3Al_2Ga_3O_{12}$: Ce (GAGG : Ce) scintillating single crystals. By now the optical characteristics of GAGG : Ce have been studied insufficiently. We have therefore measured the spectral reflectance and transmittance characteristics of these crystals using optical spectroscopy in the 200–750 nm range. We have also measured the absorbance and refractive indices and the extinction coefficients, and assessed the optical band gap for GAGG : Ce crystals. For measuring the refractive indices, we have used two spectrophotometric methods, i.e. by the measured Brewster angle and by the reflectance for low incidence angles, i.e., close to the normal. Based on the results we have drawn up the dispersion functions of the refractive indices.

Keywords

$Gd_3Al_2Ga_3O_{12}$:Ce, scintillating single crystal, transmission spectrophotometry, absorption coefficient, optical band gap, reflection spectrophotometry, refractive index, extinction coefficient, dispersion

1. Introduction

$Gd_3Al_2Ga_3O_{12}$: Ce **(GAGG : Ce)** (gadolinium/aluminum/gallium garnet) scintillating single crystals are of a great research interest because of the novelty and unique properties of this material. For example, the number of publications dealing with this material grew up more than 10-fold in the 2011–2017 period. GAGG : Ce shows good promise for sensing probes in medical equipment, including positron emission tomographs [1–5]. The material was first synthesized and grown in 2011 [6]. GAGG : Ce has a cubic structure with the *Ia3d* space symmetry group [6–9] and the chemical formula $A_3B_2C_3O_{12}$ [6–9]. The chemical elements of the host lattice (Gd^{3+}, Ga^{3+} and Al^{3+}) are located at the structural positions formed by coordination polyhedrons with oxygen atoms at the vertices:

- Gd^{3+} is located at the dodecahedral positions *A*;
- Ga^{3+} and Al^{3+} may be located at the octahedral positions *B* and the tetrahedral positions *C*.

The ionic radii of the constituent elements of the single crystal are summarized in Table 1 [10, 11]. It can be seen from Table 1 that the Ce ions can only be located at the dodecahedral positions of the gadolinium atoms in the GAGG crystalline matrix without leading to significant lattice distortion [12–14]. However, Ce may have two different charge states in the lattice, i.e. 3+ or 4+ [15, 16], this having effect on scintillation.

Analysis of the literary data showed that the researchers pay the greatest attention to the study of the scintillation properties of GAGG : Ce [6, 8, 9, 17–19], whereas far less works dealt with the optical properties of this material. Currently there are known only two works [19, 20] in which the refractive indices of the material were assessed and the dispersion functions of *n* were obtained. It should be noted that the results of these two earlier works [9, 20] disagree.

The aim of this work is to determine the optical characteristics of GAGG:Ce crystals using optical spectrophotometry, including the values and dispersion dependences of the refractive indices.

2. Experimental

$Gd_3Al_2Ga_3O_{12}$: Ce single crystals were grown at JSC Fomos-Materials using the Czochralsky method in an argon + 1–2 % oxygen atmosphere from charge obtained by solid state synthesis from a stoichiometric mixture of high purity oxides of the constituent elements. The crystals were grown in iridium crucibles on Kristall-3M equipment by high-frequency heating.

The crystals were cut into specimens in the form of polished wafers ~0.05 cm in thickness and into complex shaped specimens having a ~0.7cm thickness in their plane-parallel portions.

The specimens were studied at the certified test laboratory "Single Crystals and Stock on their Base" of the National University of Science and Technology MISiS using certified spectrophotometric measurement methods and validated equipment, and the accuracy and repeatability of the results were controlled using references.

The spectral transmittance (*T*, %) and reflectance (*R*, %) curves were recorded on an Agilent Technologies Cary 5000 spectrophotometer with a UMA automatic universal measurement accessory.

The appearance of the Cary 5000 spectrophotometer with a UMA accessory and the experimental setup are presented in Fig. 1. The UMA accessory offers a wide range of unique measuring opportunities for the measurement of various optical characteristics, including measurements without specimen movement at the same probing point [21].

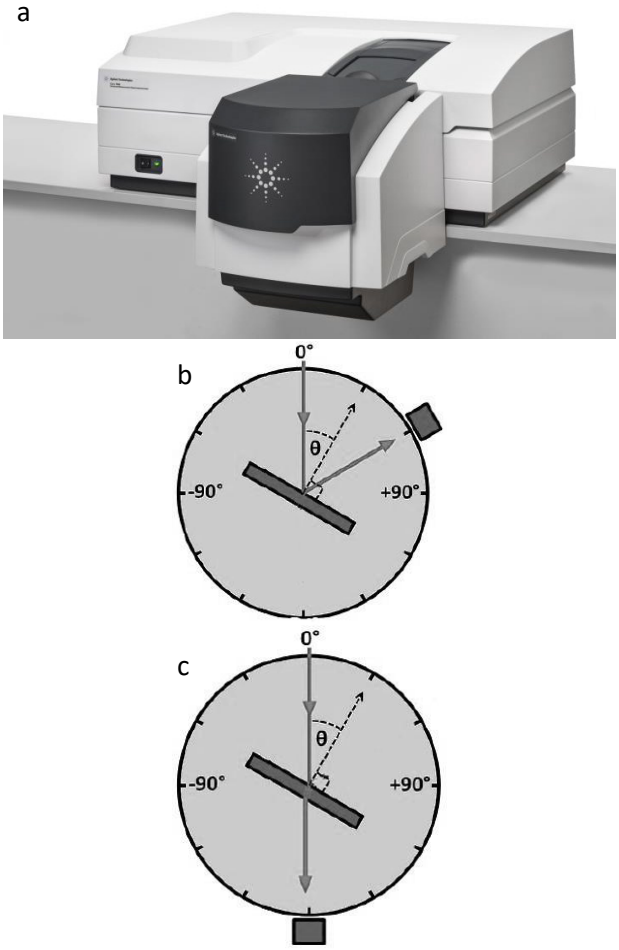

Figure 1. (a) Appearance of the Cary 5000 spectrophotometer with a UMA accessory, (b) specular reflectance measurement experimental setup and (c) transmittance measurement experimental setup.

Table 1. Ionic radii of elements with different charge and coordination states [10, 11]

Element	Degree	Ionic Radius, nm			
		Effective [10]	Coordination number [11]		
			4	6	8
Gd	3+	0.94	—	0.94	1.06
Al	3+	0.57	0.39	0.53	—
Ga	3+	0.62	0.47	0.62	—
O	2–	1.36	1.38	1.40	1.42
Ce	3+	1.02	—	1.01	1.14
	4+	0.88	—	0.87	0.97

The absorption coefficient (α, cm^{-1}) was determined from the curve of spectral transmittance T taken from a small thickness wafer (d, cm) using the Buger-Lambert law [9]:

$$\alpha = -\frac{\ln(T)}{d}. \quad (1)$$

The refractive indices were determined from the spectral angular reflectance curves taken in *p*- and *s*-polarized light. It is well-known [22] that if the incidence angles of the *p*- and *s*-polarized beams differ, there are two regions which allow measuring the refractive indices using spectrophotometric methods (Fig. 2).

In Region 1 (Fig. 2) the refractive index was assessed using the Brewster method in accordance with the following equation [9, 23]:

$$\text{tg}\,\varphi_B = n. \quad (2)$$

For this method, the reflectance spectra of *p*-polarized light are measured at different incidence angles, and the Brewster angles are calculated for every wavelength used, as described in detail elsewhere [10]. The Brewster method provides discrete values of the refractiuve indices. The dispersion functions of n were obtained using special approximation equations [24].

The accuracy of the refractive index obtained using the Brewster method was assessed using a lithium niobate reference and proved to be $\Delta = \pm 0.01$ with the confidence probability $P = 0.95$.

In Region 2 (Fig. 2) the refractive index was assessed using the low incidence angle reflection method in accordance with the following equation [22, 25].

$$R = \frac{(n-1)^2 + \kappa^2}{(n+1)^2 + \kappa^2}, \quad (3)$$

where R is the reflectance for one face of the specimen, rel. units, and κ is the extinction coefficient.

Along with the refractive index, Eq. (3) contains the extinction coefficient κ. The spectral function of the extinction coefficient can be assessed using the following expression:

$$\kappa = \frac{\alpha\lambda}{4\pi}, \quad (4)$$

where α is the absorption coefficient [25].

If κ is negligible compared with the first terms in Eq. (3), then one can assess n using the converted formula [22]:

$$n = \frac{1+\sqrt{R}}{1-\sqrt{R}}. \quad (5)$$

For low incidence angles (the R_0 method) the reflection method can provide a continuous experimental dispersion function of the refractive index.

The accuracy of refractive index obtained using the reflection method was assessed using a molten quartz reference and proved to be $\Delta = \pm 0.001$ with the confidence probability $P = 0.95$ [21].

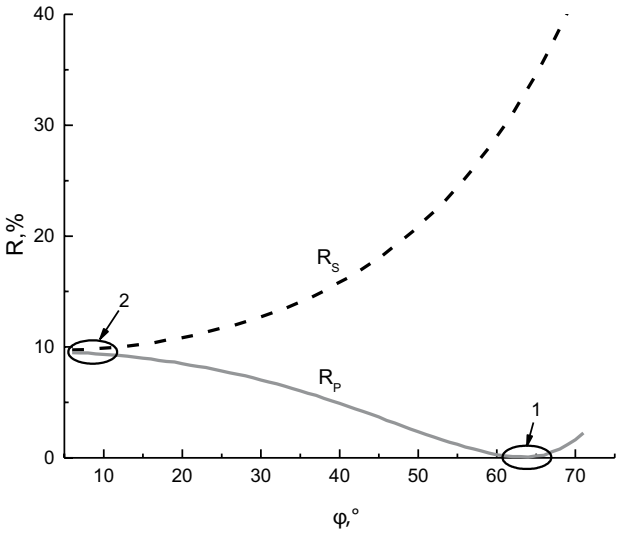

Figure 2. Reflection angular spectra for *p*- and s-polarized light: Region 1 for Brewster measurement of n; Region 2 for measurement at low light incidence angle close to the normal

The most significant limitation of these spectrophotometric methods is the shape of the specimens. The R_0 method requires the specimen shape and/or surface finished to avoid multiple reflection. The Brewster method allows studying various shape specimens, including two-side polished wafers.

The metrological tests made for the references proved that the accuracies of both the refractive index measurement methods, i.e. the Brewster angle and the low incidence angle reflection (R_0), are comparable and provide guaranteed accuracy to the third decimal digit.

3. Results and Discussion

To obtain the spectral absorbance characteristics of $Gd_3Al_2Ga_3O_{12}$:Ce we measured the spectral transmittance characteristics for normal light incidence. Earlier [9] the spectral absorbance characteristic of the material was obtained for the 200–750 nm range, and absorption bands were detected at 420–460, 330–350 and 270 nm. To determine the fundamental absorbance edge and the optical band gap of the crystals we recorded the absorbance index curves for small thickness specimens with $d \approx 0.5$ mm in the 200–750 nm range.

Based on the experimental absorbance index curves α(λ) we obtained the spectral function of the extinction coefficient κ (Fig. 3) in accordance with Eq. (4). The results are presented in Fig. 3 and suggest that the α(λ) and κ(λ) spectral functions are nonmonotonic and have expressed absorption bands peaking at 440, 340, 300–310, 270 and 230 nm. Earlier [8, 9, 26–29] the absorption bands were accounted for as follows:

- the 340 and 440 nm bands were attributed to the $4f - 5d^2$ and $4f - 5d^1$ transitions in Ce^{3+}, respectively;

- the 300–310, 270 and 230 nm bands were attributed to the $^8S - {}^6P_j$, $^8S - {}^6I_j$ and $^8S - {}^6D_j$ transitions in Gd^{3+}, respectively.

The optical band gap was determined in accordance with the Tauc law [30]:

$$\alpha = \alpha_0 \frac{\left(h\nu - E_g\right)^{r/2}}{h\nu}, \quad (6)$$

where E_g is the band gap, α_0 is the material's constant and r is the exponent which is 1 for direct band gap materials and 4 for indirect band gap ones.

For direct band gap materials which include $Gd_3Al_2Ga_3O_{12}$: Ce [31, 32], the Tauc equation can be transformed as follows:

$$(\alpha h\nu)^2 = \alpha_0(h\nu - E_g). \quad (7)$$

The optical band gap for these materials is determined graphically as described elsewhere [33–36].

The optical band gap of single crystal GAGG : Ce as determined graphically at room temperature using the Tauc law (Fig. 4) proved to be $E_g \approx 5.88 \pm 0.05$ eV.

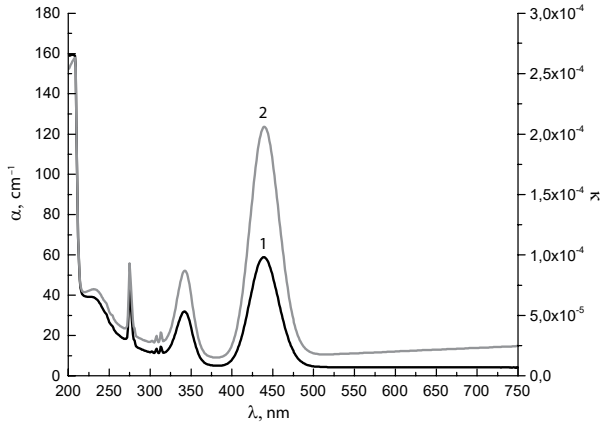

Figure 3. Spectral functions of absorption coefficient and extinction coefficient: (*1*) absorption coefficient and (*2*) extinction coefficient

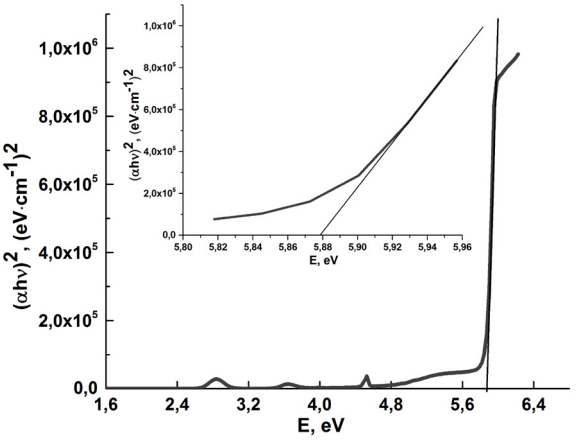

Figure 4. $Gd_3Al_2Ga_3O_{12}$: Ce optical band gap determination using Tauc method

Band gap estimates for GAGG : Ce were also published earlier [9, 16]. These results suggest that the band gap of GAGG : Ce is (5.8–6.8) eV. Our optical band gap estimate falls within this range.

Single crystal GAGG : Ce is a cubic material and has one refractive index n. To estimate this index we measured the p-polarized light reflectance spectra at different incidence angles (the Brewster method) and non-polarized light reflectance spectra at a low incidence angle (6 arc deg) close to the normal (the R_0 method). To obtain authentic data for the refractive indices we performed all the measurements at the same point of the non-plane-parallel portion of the complex shaped specimen.

The calculated extinction coefficient of $Gd_3Al_2Ga_3O_{12}$: Ce for the 200–750 nm range is $\kappa \sim 10^{-6} \div 10^{-4}$ (Fig. 3). Since the extinction coefficient is negligible compared with the first terms of Eq. (3), it is safe to assess the refractive index using the R_0 method in accordance with Eq. (5) [37].

Figure 5 shows the dispersion functions of the refractive index for the material obtained using the Brewster method and the low incidence angle reflection method (R_0). The results obtained using the two methods are in a good agreement.

4. Summary

The transmittance spectra of the $Gd_3Al_2Ga_3O_{12}$: Ce scintillating single crystals in the 200—750 nm range were obtained. The absorbance and extinction spectral functions were calculated based on the experimental spectral curves. The extinction coefficient of $Gd_3Al_2Ga_3O_{12}$: Ce is within the 10^{-6}–10^{-4} range.

The optical band gap of the $Gd_3Al_2Ga_3O_{12}$: Ce single crystals was estimated by means the Tauc method is 5.88 ± 0.05 eV.

The refractive indices were calculated from the spectral angle reflectance functions for p-polarized light using the Brewster method and from the reflectance spectra for a low incidence angle (6 arc deg) close to the normal (the

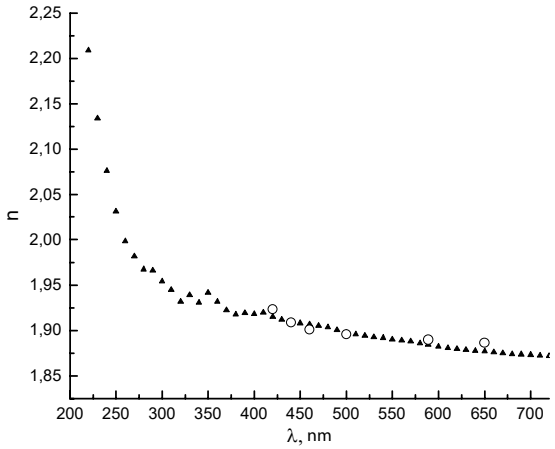

Figure 5. Dispersion functions of refractive index obtained using (○) Brewster method and (▲) reflection at low incidence angle close to normal

R_0 method). The results obtained using the two methods are in a good agreement.

Acknowledgments

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