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NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A

Refractive index and absorption length of YAP : Ce scintillation crystal and reflectance of the coating used in YAP : Ce single-crystal matrix

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Abstract

Recent works show growing interest in optically isolated YAP : Ce single-crystal matrices as high spatial resolution detectors for imaging applications in Nuclear Medicine. Such matrices are manufactured by Preciosa Crytur (Czech Republic) by means of a special sticking procedure of YAP : Ce pillars about 10 mm in length and with cross section ranging between 0.6 mm × 0.6 mm and 3 mm × 3 mm. In this work, we carried out an intensive study on both the complex refractive index of YAP : Ce crystal and the reflectance, versus. the incidence angle, of the coating used as optical insulator in matrix pillar made by Preciosa Crytur. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

$YAlO_3$ (YAP) has a venerable history as laser material when doped with Nd [1]. The same crystal doped with Cerium has remarkable scintillation properties [2–6]. The main characteristics of YAP : Ce, well described in other works [4–6], can be summed up as follows: average Z 39, density 5.37 g/cm^3, average scintillation decay time 25 to 30 ns and emission peak at about 370 nm. Moreover, YAP : Ce has an intrinsic light yield of about 50% compared to NaI (Tl). Recent studies on medical imaging applications show that YAP : Ce crystal, when arranged in a multipillar matrix, is particularly suitable to detect the interaction position of gamma rays emitted by a Tc^{99m} source, the most common tracer used in SPET technique [7–9].

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Preciosa Crytur (Czech Republic) produces matrices by means of a special sticking procedure of YAP : Ce pillars 10 mm length and with square cross sections ranging between $0.6\times0.6\ \mathrm{mm}^2$ and $3\times3\ \mathrm{mm}^2$. Each pillar is optically isolated from the adjacent ones by a 5 μm thick reflective coating, therefore the total dead zone is only 10 μm thick. The light yield of the matrix composed by $1\times1\times10\ \mathrm{mm}^3$ pillars is about 24% with respect to one of the 1 cm thick planar YAP : Ce crystal [10] and the aim of the present paper is to study the cause of such a reduction. For this purpose, we measured the refractive index and absortion length of YAP : Ce crystal and the reflectance versus the incidence angle of the coating used as optical insulator between two adjacent pillars in the YAP : Ce matrix. These measurements allow some considerations about the light output of one single pillar.

2. Experimental

2.1. Refractive index and absorption length of YAP : CE crystal

Two YAP : Ce single crystals were grown by Preciosa Crytur from 0.1 mol% CeO_2-rich melt, the same melt is used for the matrix pillar production. The samples are 52 mm and 104 mm long with $10\times10\ \mathrm{mm}^2$ cross section; the $10\times10\ \mathrm{mm}^2$ surfaces are flat and optically polished, the remaining are ground. The YAP crystal is orthorhombic [11,12] therefore a biaxial optical anisotropy is expected [13]. Such an anisotropy was experimentally observed in undoped YAP crystal [14]. On the other hand unpolarised HeNe laser beam travels along the examined Ce-doped crystals without a macroscopic splitting and in accordance with the optics of isotropic medium. Moreover, reflectance at air‖YAP : Ce interfaces, at normal incidence, does not depend on the polarisation when a 0.05% sensitivity is available. For these reasons, in the present section the examined YAP : Ce crystals are considered optically isotropic.

As already reported elsewhere [15], the complex refractive index of a crystal can be determined from reflectance and transmittance measurements at normal incidence. In the present investigation both the measurements were performed with a commercial spectrophotometer (Lambda 9 Perkin Elmer) in the 200–850 nm wavelength range. The instrument reference values were performed without a sample for transmittance and with a first surface reference mirror (NSB SRM 2003) for reflectance.

In reflectance (R) measurement, because of the considerable sample lengths, only the beam reflected from the first interface air‖YAP was detected. Reflectance depends on the surface roughness that was measured with a Wyco TOPO 3D. Fig. 1 shows the reflectance measured on the surface having the lowest roughness (5.3 nm rms). Fig. 2 and Table 1 report the refractive index n deduced by the equation

$$R(\lambda) = \left|\frac{n+ik-1}{n+ik+1}\right|^2 \cong \left|\frac{n-1}{n+1}\right|^2, \quad (k \ll 1), \tag{1}$$

considering the roughness-corrected reflectance

$$R(\lambda) = R_{\mathrm{m}}(\lambda)\exp\left(\frac{4\pi\delta}{\lambda}\right), \tag{2}$$

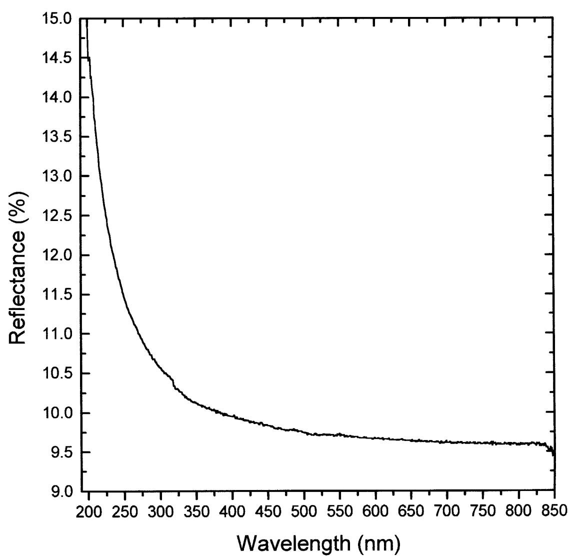

Fig. 1. Reflectance at near-normal incidence (about 7°) of an air‖YAP interface. The experimental error is ± 0.1%.The surface roughness, measured with a Wyko TOPO 3D, is 5.3 nm rms.

where $R_m(\lambda)$ is the measured reflectance, λ is the wavelength and δ is the rms of the surface roughness assumed to have a normal distribution [16]. Refractive index of undoped YAP crystal along the crystallographic axes [14] is also reported in Fig. 2. The doping obtained growing YAP crystal from 0.1 mol% CeO_2 rich melt reduces: (i) the refractive index of about 0.04: (ii) the difference among the main refractive indices (about 0.025 for the undoped) to less than 0.003 that is the experimental error due to the spectrophotometer sensitivity (0.05%). It should be noticed that the error on n reported in Table 1 (about 0.01) depends also on the reference mirror and on spectrophotometer reference drift occurring during the wavelength scanning.

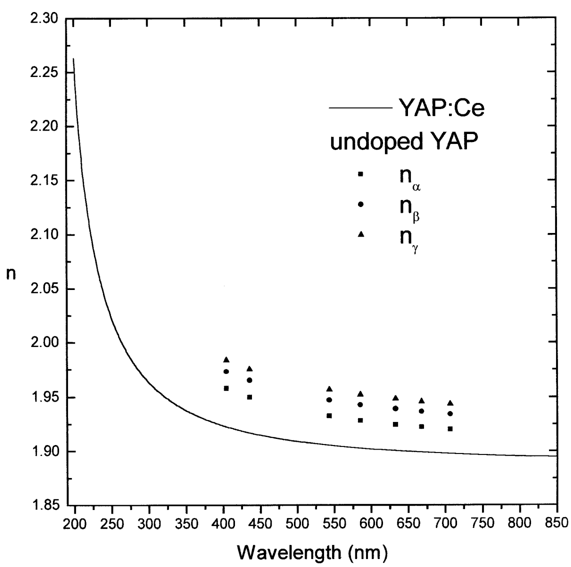

Fig. 2. Real part (n) of the complex refractive index of YAP : Ce crystal (line); the error is reported in Table 1. Refractive index of undoped YAP crystal along the crystallographic axes [14] (symbols).

Table 1
YAP refractive index at several wavelengths

λ(nm)	$n \pm \Delta n$
200	2.26 ± 0.01
225	2.091 ± 0.009
250	2.023 ± 0.009
275	1.986 ± 0.009
300	1.963 ± 0.009
325	1.948 ± 0.008
350	1.938 ± 0.008
375	1.930 ± 0.008
400	1.923 ± 0.008
450	1.915 ± 0.008
500	1.909 ± 0.008
550	1.905 ± 0.008
650	1.900 ± 0.008
750	1.897 ± 0.008
850	1.894 ± 0.008

Fig. 3 shows the transmittance (T) of the directly transmitted beam after the correction of the systematic error caused by the slight wedging of the polished surfaces. As a matter of fact the beam directly transmitted, even if a bit displaced, was detected. On the contrary the secondary contributions, reflected from the outgoing and incoming surfaces and crossing the crystal more than one time, were not detected. The displacement of the directly transmitted beam caused the transmittance depending on the crystal orientation around the longitudinal axis. In order to evaluate the systematic error related to a given orientation, the transmittance of each sample was also measured using a HeNe laser beam ($\lambda = 632.8$ nm) and

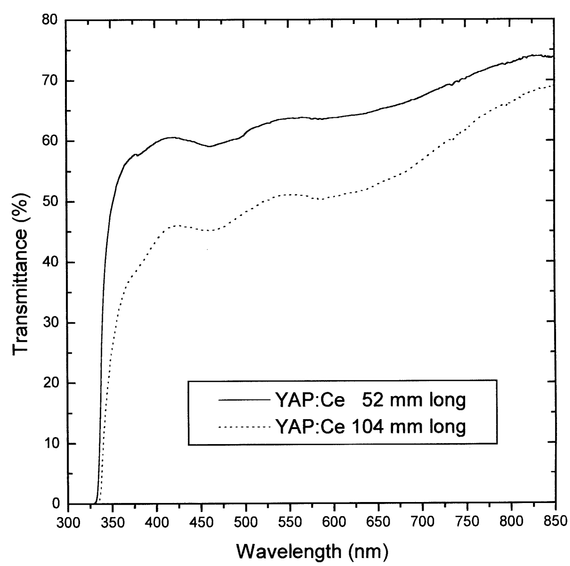

Fig. 3. Transmittance of the beam directly transmitted. The secondary contributions, reflected from the outgoing and incoming surfaces and crossing the crystal more than once were not detected. The typical experimental error is $\pm 0.15\%$.

a photomultiplier tube (mounted on a x–y micrometrical translator). Then the spectral transmittance curve was multiplied by the factor for which the measurement agrees with the value obtained with the HeNe and the photomultiplier tube.

Fig. 4 shows the absorption length $L = \lambda/4\pi k$ computed by the equation

$$T(\lambda) = (1 - R(\lambda))^2 \exp\left(-\frac{d}{L(\lambda)}\right), \tag{3}$$

where d is the crystal length. The absorption lengths of the two samples are very similar to each other. As an example at the luminescence peak, $\lambda = 370$ nm, the absorption length of the short (52 mm) and the long (104 mm) crystals are 14.74 ± 0.24 cm and 13.24 ± 0.11 cm, respectively.

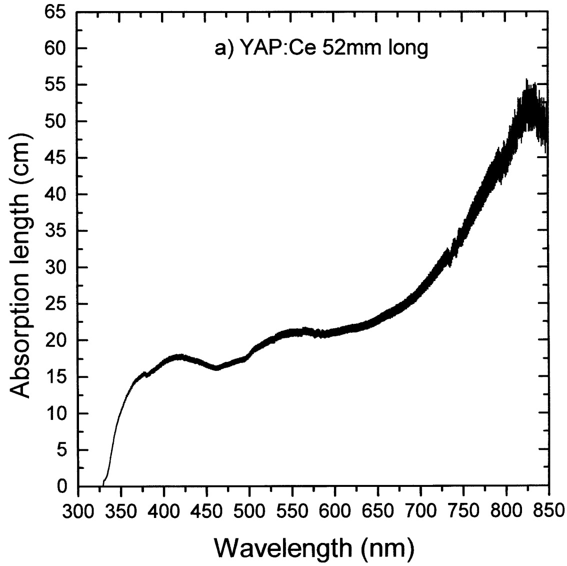

Fig. 4. Absorption length $L = \lambda/4\pi k$ of the 52 mm long (a) and 104 mm long (b) YAP : Ce samples.

2.2. Optical characterisation of the reflective coating

Each pillar of YAP : Ce matrix is optically isolated from the other by means of a coating having high reflectance and very low transmittance. This coating has a metallic appearance and probably it is a multilayer including a metallic film thick enough to reduce the light transmission. For such a coating, the reflectance of the system YAP : Ce ‖coating‖ air strongly depends on the incidence versus [17] and, on the other hand, the most interesting case is when the light is coming from YAP : Ce. The measurement of this reflectance, versus the incidence angle was performed on a hemispherical YAP : Ce sample, manufactured by Preciosa Crytur, whose radius is 10 mm and the flat surface is treated with the same coating used in YAP : Ce matrix. The adopted experimental set up is reported in Fig. 5. The beam of a UV enhanced Ar Laser (Coherent Innova 200) (L) is sent to the centre of the flat surface, through the hemispherical surface of the sample (S). The intensity of the reflected beam depends on the coating reflectance, but also on the crossing of the two air‖YAP : Ce interfaces and on the 10 mm path through the YAP : Ce. Both the air‖YAP : Ce transmittance and the 10 mm path attenuation can be evaluated on the basis of the YAP : Ce complex refractive index reported in Section 2.1. The 150 mm focal lens (F) reduces the spot size of the laser beam to about 0.1 mm in proximity of the sample. The rotation axis of the rotating platform (RP) is lying on the circular flat surface and passes through its centre. The incidence angle θ is measured from the orientation for which incidence and reflected beams coincide, with an error lower than 0.5°. The incident beam is p (electric vector parallel to the incidence

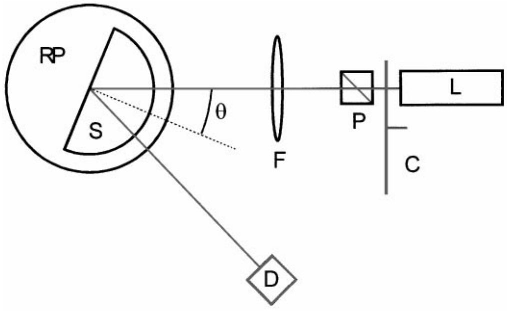

Fig. 5. Experimental set-up used for the coating reflectance measurement versus the incidence angle. L = UV enhanced argon laser; C = chopper; P = polarizer; F = 150 mm focal lens; S = hemispherical shaped YAP : Ce sample (the coating is on the flat surface); RP = rotating platform; D = detector.

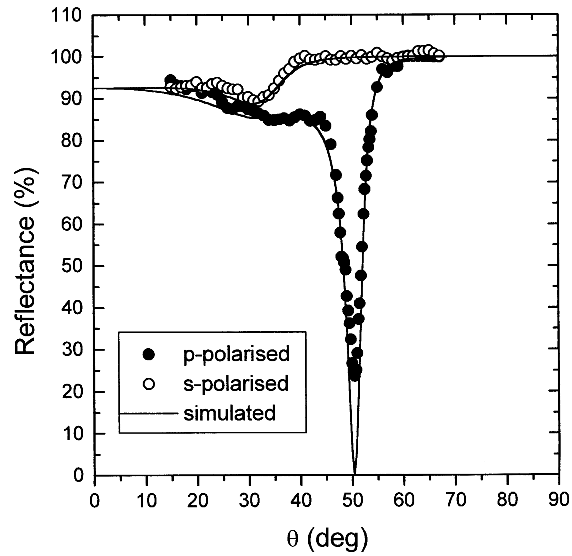

Fig. 6. Experimental reflectance (YAP : Ce side), versus the incidence angle, of the system YAP : Ce‖coating‖air for p and s polarisations. The continuous lines are the reflectances computed according to the model YAP : Ce‖MgF_2(170 nm)‖Al(thicker than 50 nm)‖air and with the refractive indexes reported in literature [18,19].

plane) or s linearly polarised according to the orientation of the polariser (P). The laser power is distributed over 8 lines located between 333.6 and 385.8 nm with 355 nm power weighted mean. The beam intensity is modulated with the mechanical chopper (C) and the intensity of the reflected beam, hitting the UV enhanced silicon detector (D), is measured with a lock-in amplifier.

In this experimental situation YAP : Ce crystal shows its anisotropy: the polarisation state of the reflected beam almost perfectly coincides with one of the incident beam (p or s), only for four sample orientations (90° spaced) around the hemisphere radius orthogonal to the flat surface. This behaviour, let us suppose that one of the three YAP : Ce principal axis nearly lie along the sample flat surface, so that the incident beam polarisation is conserved only when this principal axis is parallel or (almost) orthogonal to the incidence plane. This phenomenon and the considerations discussed in Section 2.1 indicate that the difference between the principal refractive indices is lower than 0.003 and greater than or nearly equal to $0.25\lambda/d \sim 10^{-5}$ (that is the minimum difference required to observe a macroscopic disagreement between the polarisations of the incidence and the reflected beams [13]) with λ e.m. wavelength and d crossed YAP : Ce thickness. In conclusion, the influence of the YAP : Ce anisotropy on the magnitude of the coating reflectance is very low compared to the sensitivity of our apparatus (0.5%); on the contrary, the sample must be carefully oriented around the hemisphere radius orthogonal to the flat surface in order to minimise the modification of the probe beam polarisation.

Fig. 6 shows the measured reflectances of the coating for p (R_p) and s (R_s) polarisations of the incident (and reflected) beam. In both cases at large incidence angle ($\theta \geq 60°$) the reflectances are 100%, when the total reflection occurs [13]. Moreover for $40 \leq \theta \leq 60°$, $R_s \sim 100\%$ and $R_p \sim 20\%$ at $\theta \sim 52°$, when there is photon tunnelling in the external film of a dual layer at θ higher than the critical angle [18]. These observations allow us to assume that the coating is composed of two layers: an internal low refractive index film and an external metallic layer. As a validation of this hypothesis, the reflectance of the system YAP : Ce ‖MgF_2(170 nm)‖Al(thicker than 50 nm)‖air, reported in Fig. 6 as a continuous line and computed using the refractive index data reported in literature [19,20], is in good agreement with the experimental data for both the polarisations. The disagreement in the minimum of R_p is probably due to both the imperfect conservation of the polarisation (caused by the anisotropy) and the imperfect monochromaticity of the used laser beam.

It must be emphasised that the fall in the reflectivity, for the p-polarised wave, at 52° has no influence on: (i) the overall optical insulation between one pillar and the adjacent ones, because no reflected photons are absorbed in the metallic layer; (ii) the coating performs as light guide, because the R_p minimum is very narrow and does not occur for the s polarisation.

3. Discussion

Usually the YAP : Ce matrix is optically coupled with the PMT window by some optical grease. The refractive index of both the PMT window and the optical grease ($n \sim 1.5$) is lower than that of YAP : Ce. Therefore, total reflection occurs when the photons generated inside the YAP : Ce pillar impinge the YAP : Ce‖PMT interface with an incidence angle greater than the critical angle [13]

$$\theta_C = \arcsin(n/n_{\mathrm{YAP:Ce}}) \sim 51^\circ.$$

When volume and surface scattering is negligible, the angle of the photon trajectory with the pillar longitudinal axis is not affected by reflections on the pillar surfaces (because the surfaces are parallel or orthogonal to the longitudinal axis) and the maximum value of the ratio *detected to emitted photons* (D/E) for an isotropic source located inside the pillar is

$$D/E_{MAX} = 2\ 2\pi(1 - \cos\theta_C)/4\pi \sim 37\%.$$

This represents the best light output from pillars with optically polished surfaces and it is effectively obtainable in the following conditions: (i) total reflection ($R = 1$) on the surfaces of the pillar not coupled with the PMT; (ii) infinite absorption length of YAP : Ce. In the real case none of the above conditions are satisfied and D/E is lower.

As a matter of fact taking into consideration the 1 cm long matrix composed by $1 \times 1 \times 10\ \mathrm{mm}^3$ pillars, the results [21] of a Monte Carlo simulation show that the ratio D/E is equal to 23%. The major cause of the scintillation light loss is to be ascribed to the absorption into the coating. The results of the Monte Carlo simulation show, in fact, that 70% of the emitted light is absorbed into the coating while only 7% is absorbed into the YAP : Ce. Each single pillar behaves as an optical fibre guiding and focusing the scintillation light (by means of multiple internal reflections) to the output matrix surface. The resulting light output can be made, therefore, very narrow at the expense of the number of collected photons.

It is worth noting that the Monte Carlo results do not change by considering the coating reflectance linearly growing from 85% (0°) to 100% (90°), then the fall of the reflectance around 50° for the p polarisation does not dramatically affect, as already stated above, the pillar light output.

4. Conclusions

The optical anisotropy of YAP : Ce single crystal, grown by Preciosa Crytur from 0.1 mol% CeO2-rich melt, is lower than the one of undoped YAP crystal: the maximum difference among the principal refractive indices is lower than 0.003 and greater or about equal to 10^{-5} for YAP : Ce and about 0.025 for undoped YAP crystal. In short, the photon trajectory in the pillar can be treated according to the geometrical optics, but generally the photon polarisation is not conserved along the path through YAP : Ce bulk. As experimental results, the refractive index and the absorption length of YAP : Ce at 370 nm are 1.93 and 14 cm, respectively.

In Preciosa Crytur matrices, the surfaces of YAP : Ce pillars are optically polished and treated with a coating. The thickness of the external metallic layer is thick enough to ensure the optical insulation. A more internal layer, having a refractive index lower than that of YAP : Ce, causes the total reflection for incidence angle greater than 40°, although there is tunnelling in the metallic layer of p polarised photons when the incidence angle is about 52°. The coating reflectance averaged on the incidence angle is about 90%.

The number of absorbed photons into the crystal is very small compared to the ones absorbed into the coating because of the high measured YAP : Ce absorption length at 370 nm (14 cm). As a matter of fact, the scintillation light is guided by means of multiple internal reflections in the matrix output light surface, and the very high number of hits against the coating suffered by each single emitted photon makes its absorption into the coating highly probable.

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