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Enhanced light extraction of $Bi_3Ge_4O_{12}$ scintillator by graded-refractive-index antireflection coatings

Fei Tong, Bo Liu,[a] Hong Chen, Zhichao Zhu, and Mu Gu
Shanghai Key Laboratory of Special Artificial Microstructure Materials and Technology, School of Physics Science and Engineering, Tongji University, Shanghai 200092, People's Republic of China

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A three-layer graded-refractive-index antireflection coating is designed and prepared on the one surface of the $Bi_3Ge_4O_{12}$ scintillator by sol-gel technology. The emission intensity of the $Bi_3Ge_4O_{12}$ with a graded-refractive-index antireflection coating exhibits a broadband and omnidirectional enhancement of 15.9% compared with the reference sample without coating. This significant enhancement is attributed to the decrease of Fresnel reflection, which is consistent with the measurement of transmission spectra. Additionally, it is evident that the graded-refractive-index coating is superior to the conventional quarter-wave coating due to the omnidirectionality advantage. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4818821]

In the field of radiation detection based on scintillation luminescence, a high light output is vital to improve sensitivity, spatial resolution, and energy resolution of detection systems.[1] For instance, a high sensitivity can lead to a low radiation dose needed in the medical imaging, such as X-ray computerized tomography, which is possible to reduce the radiation damage for the patients. A variety of scintillators have high internal quantum efficiency. However, those inorganic scintillators with high refractive indices suffer from intense Fresnel reflection and total internal reflection, thus degrading the light extraction efficiency. Improvement of light extraction efficiency is important for practical applications. Recently, two-dimensional photonic crystals are designed on the surface of scintillators to decrease the total internal reflection by out-coupling the emission light with incident degree beyond the critical angle.[2,3] Fresnel reflection is a common phenomenon occurring when light propagates across a boundary between two media with different refractive indices.

$Bi_3Ge_4O_{12}$ (BGO) is a popular scintillator with extensive applications in nuclear medical imaging systems and high energy physics experiments.[4] It has a high refractive index of 2.15 at its central emission wavelength of 510 nm. Conventional quarter-wave (QW) antireflection (AR) coatings are widely used to eliminate Fresnel reflection by means of interference principle. However, QW coatings perform well only in the normal direction and with a limited bandwidth. In contrast, graded-refractive-index (GRIN) AR coatings are theoretically able to achieve broadband and omnidirectional AR charactersistic.[5] A lot of approaches such as oblique-angle deposition,[5–7] biomimetic templating technique,[8–10] self-organized nanodot arrays,[11] and custom tailored compositions[12,13] are proposed to achieve broadband and omnidirectional AR based on GRIN. Enhanced efficiencies by using GRIN AR coatings to eliminate Fresnel reflection have been well demonstrated in light-emitting diodes[7] and solar cells.[14] However, the application of GRIN AR coatings to scintillators has not been reported until now.

Sol-gel process is an important method to prepare materials with nanoscale structures, such as semiconductor nanocrystals,[15] oxide nanorods,[16] electron transport layer in light-emitting diodes.[17] Sol-gel derived nano-porous SiO_2 and TiO_2 films can achieve adjustable refractive index by controlling porosity of the coatings. Sol-gel technique requires inexpensive equipment and is easy to prepare samples with large area. In this letter, GRIN coatings using nano-porous SiO_2 and TiO_2 on BGO scintillators are designed and prepared for broadband omnidirectional AR. This investigation demonstrates that GRIN AR coatings can efficiently decrease Fresnel reflection and thus enhance the light output of scintillator.

A GRIN AR coating in the present study is designed according to the quintic graded-index profile which exhibits excellent performance.[18] Theoretically, when the refractive index continuously varies from the index of scintillation to the index of air, Fresnel reflection can be completely eliminated for omnidirectional incidence over a broad wavelength range. However, due to the technique limitation, we have to choose discrete layers with different refractive indices. Fig. 1 shows the thickness and the refractive index for each individual layer in a three-layer GRIN AR coating following the quintic graded-index profile. In Fig. 2, we present the calculated reflectivity with the average of transverse electric and transverse magnetic polarizations for BGO with GRIN coating, BGO with QW coating, and BGO without coating (as reference), assuming that the incident medium is BGO, and the emergence medium is air. Both the GRIN and QW coatings exhibit low reflectivity with respect to the reference. It is also found that the reflectivity for the GRIN coating is much lower than that of the QW coating in high degree and broadband.

The experimental demonstration was performed on the BGO samples which were cut and polished into $20 \times 10 \times 3$ mm^3 pieces. The three-layer GRIN coatings were prepared on the one side of the BGO pieces using base/acid two-step catalysis according to the Refs. 19 and 20. In the first step, $Si(OC_2H_5)_4$ (TEOS), $NH_3 \cdot H_2O$ and ethanol were mixed with a molar ratio of 1:2:40 and then aged at room temperature for 1 week. The sol was under reflux at 80 °C for 10 h afterwards, and this is called the sol A which was prepared

[a]Author to whom correspondence should be addressed. Electronic mail: lbo@tongji.edu.cn

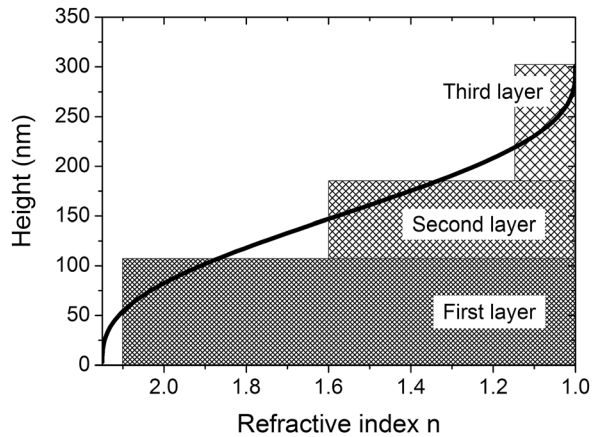

FIG. 1. Design of the three-layer GRIN AR coating on a BGO scintillator according to the quintic profile.

by only a base one-step catalysis. A mixture of TEOS, water, ethanol, and HCl with a molar ratio of 1:2:40, which is called the sol B formed by only an acid one-step catalysis, was added into the formed sols in the second step. TiO_2 sol was prepared by using tetrabutyl titanate, acetylacetone, acetic acid, ethanol, and deionized water with a molar ratio of 1:0.2:1:30:3 which was mixed at room temperature. Then the TiO_2 sol was added into the acid-catalyzed SiO_2 sol which was prepared by the same way which was described in the second step. After that, the mixed SiO_2-TiO_2 sol was formed and kept at room temperature for coating. The refractive index was controlled by the ratio of SiO_2 and TiO_2. The layers of coating were prepared with the dip method. Both QW and GRIN coatings were prepared on the one side of BGO pieces. The thickness was controlled by the dip-speed. The thickness and the refractive index of each layer were measured by ellipsometry. For the single-layer QW coating, the thickness is 87 nm and the refractive index is 1.46. For the three-layer GRIN coating, the thickness and the refractive index for each layer are listed in Table I.

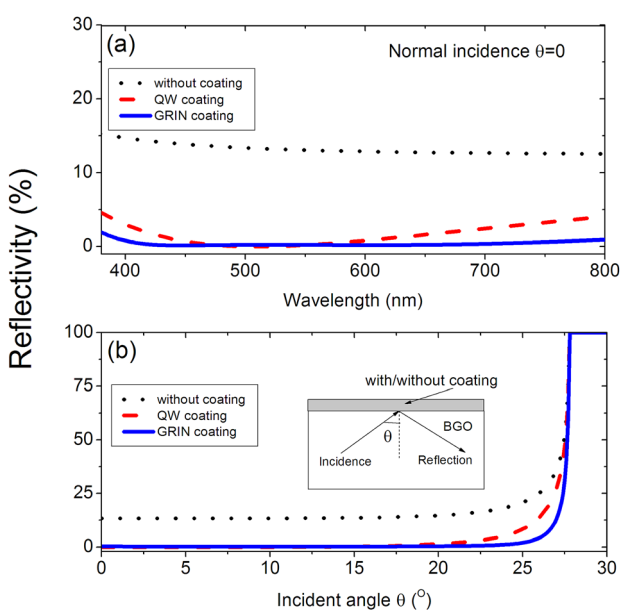

FIG. 2. Calculated reflection spectra in the normal incidence with a function of wavelength (a) and angle (b) for the BGO with GRIN coating, QW coating, and the reference sample without coating.

TABLE I. Component, refractive index, and thickness for each layer of the GRIN coating.

Layer	1	2	3
Component	SiO_2/TiO_2	SiO_2/TiO_2	SiO_2
Refractive index	2.10	1.60	1.16
Thickness (nm)	104	80	117

The experimental transmission spectra in the normal direction with an unpolarized light source are shown in Fig. 3(a) which exhibit good coincidence with the theoretical spectra shown in Fig. 3(b) calculated with the parameters listed in Table I. From the transmission spectrum of the reference sample (without coating), the intrinsic absorption in the range of emission wavelength is so weak that it can be neglected. The emission spectra recorded by a fiber spectrometer with a FFT-CCD (PG2000-Pro-Ex, Ideaoptics Co.) under the excitation wavelength of 266 nm from an ultraviolet laser are shown in Fig. 4. The emission spectra of BGO are ranged from 400 to 750 nm with a peak at 510 nm. The broadband of antireflection is benefit to the overall emission spectrum. As shown in Fig. 4(a), the emission spectra of BGO in the normal direction are increased by about 13% for the QW coating and the GRIN coating samples compared with the reference sample. The emission intensity for the BGO with GRIN coating is slightly higher than that of the BGO with QW coating for the wavelength longer than 650 nm. Therefore, compared with the QW coating, the GRIN coating is not obviously superior in the normal direction but exhibits more significant enhancement in high degree of emission. The angular-dependent emission intensities at 510 nm are shown in the inset of Fig. 4(a). For the reference sample, the angular-dependence exhibits an approximate Lambertian emission pattern which follows a cosine dependence on the emission angle.[21] For the sample with QW coating, the angular-dependence exhibits a faster decay with increasing degree than that of the reference sample. In contrast, the sample with GRIN coating exhibits a better angular characteristic, which can be attributed to the fact that the GRIN coating is able to significantly lower Fresnel reflection in omnidirection. Figs. 4(b)–4(d) present an overall performance of angular-dependent emission spectra for the

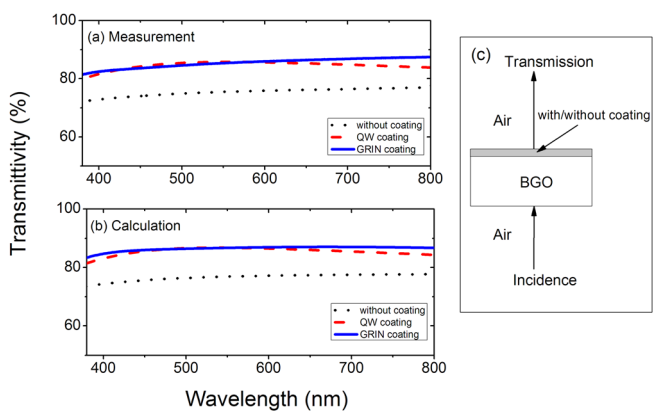

FIG. 3. Measured (a) and calculated (b) transmission spectra in the normal direction for the BGO with GRIN coating, QW coating, and the reference sample. The sketch of layout is shown in (c).

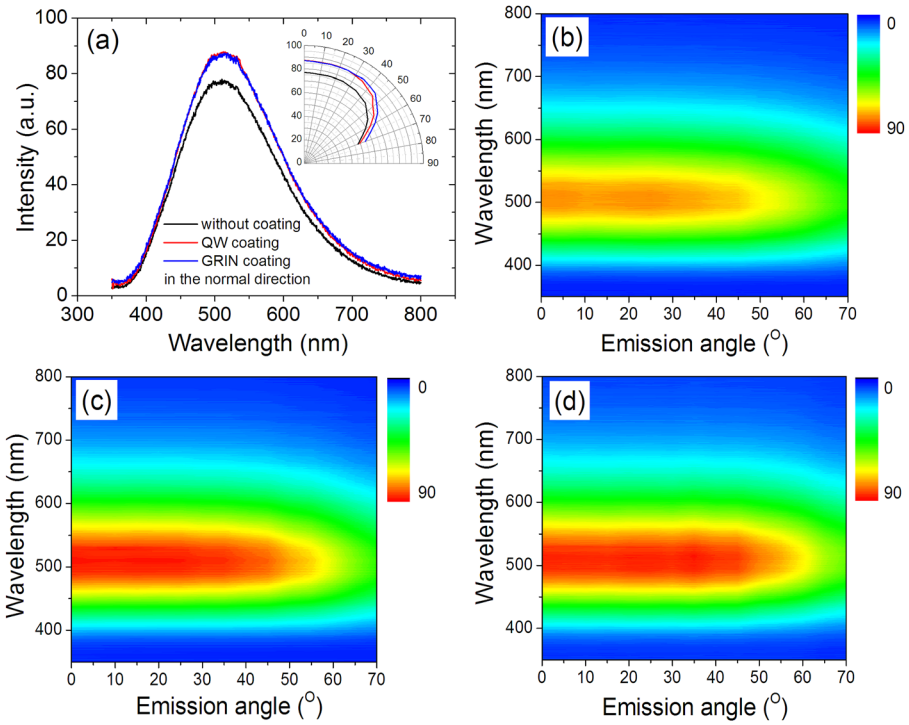

FIG. 4. Emission spectra under the excitation of 266 nm for BGO with GRIN coating, QW coating, and reference sample (a). The angular profile at 510 nm emission is shown in the inset of (a). Angular-dependent emission spectra for reference sample (b), BGO with QW coating (c), and BGO with GRIN coating (d).

samples with GRIN and QW coatings and the reference sample. Compared with the reference sample, the spectral and angular integration intensities increase by 10.7% and 15.9% for the sample with QW coating and the sample with GRIN coating, respectively. As a result, it is well demonstrated that the performance of the GRIN coating is evidently superior to the QW coating in both bandwidth and directionality.

In summary, a three-layer GRIN AR coating on BGO scintillator prepared by sol-gel method can significantly enhance the scintillation light output in broadband and omnidirection due to the significant reduction of Fresnel reflection. The GRIN AR coating is superior to the conventional QW AR coating. The concept of graded-refractive-index would be very beneficial to the application in the scintillation detection systems.

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