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Improved light extraction efficiency of cerium-doped lutetium-yttrium oxyorthosilicate scintillator by monolayers of periodic arrays of polystyrene spheres

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In this Letter, monolayers of arrays of periodic polystyrene (PS) spheres are designed to couple onto the surface of cerium-doped lutetium-yttrium oxyorthosilicate scintillator to improve the light extraction efficiency. The enhancement of extraction efficiency up to 38% relative to the reference case without polystyrene spheres is achieved. Combining with the simulation for the transmission as well as its dispersion relation, detailed analysis of the effect of whispering gallery modes and diffraction on the extraction mechanism are given. As a result, the optimal diameter of 414 nm is obtained based on a trade-off between the transmission loss and the diffraction enhancement. [http://dx.doi.org/10.1063/1.4793303]

Cerium-doped lutetium-yttrium oxyorthosilicate (LYSO) with advantages of high light output (~30 000 ph/MeV), good energy resolution (~10%), and short decay time (~40 ns) has become an excellent scintillator for the applications in medical imaging and high energy physics experiments.[1] In spite of the high internal quantum efficiency of LYSO, the light extraction efficiency is rather low due to its high refractive index (1.83), which leads to a small critical angle (θ_c). The extraction efficiency from one side of the crystal-air interface is as low as 7.5% according to an approximate formula ($1/4n^2$).[2] In order to obtain a significant increase of light extraction, photonic crystals have been widely used in inorganic semiconductor light-emitting diodes (LEDs)[3–5] and organic electroluminescence devices.[6,7] Although the research on the enhancement light extraction efficiency in LEDs has attracted great attention, the application of photonic structures to the field of scintillators is scarcely reported.[8–10] For practical purposes in scintillation detection systems, large-area patterned photonic crystals fabricated in low cost are highly desirable. Therefore, two-dimensional photonic crystals consisting of a monolayer of self-assembled hexagonal-close-packed (hcp) dielectric spheres prepared in a very economical and effective way holds a great promise.[11]

An array of periodic wavelength-scale dielectric spheres exhibits special photonic characteristics because of both whispering gallery (WG) modes due to Mie resonance of individual spheres and Bragg diffraction arising to the periodic arrangement.[12] Based on the ability of controlling the redistribution of electromagnetic field in such systems, the arrays of periodic dielectric spheres obtain important applications in, such as solar cells,[13,14] biological and chemical sensors,[15] photocatalysis,[16] and field emission devices.[17] In this Letter, monolayers of periodic arrays of polystyrene (PS, n = 1.59) spheres are self-assembled onto the top surface of LYSO scintillator for the improvement of light extraction beyond the total internal reflection. The underlying physical mechanism for light extraction is discussed in detail.

Commercially available LYSO single crystal scintillator was cut and polished into 3 × 10 × 20 mm pieces. The monolayers of PS spheres with different diameters were coated onto the LYSO single crystal substrates using a modified self-assembly method.[18] SEM images of the samples show a monolayer of hcp PS spheres coating on the scintillator. The emission spectra were recorded by a fiber spectrometer under the excitation wavelength of 360 nm from an ultraviolet LED. The numerical simulations were performed based on a rigorous coupled wave analysis (RCWA) method.

The simulated transmission spectra at 415 nm (peak of LYSO emission band) shown in Fig. 1 exhibit a significant transmittance amplitude of light beyond the critical angle θ_c thus suggesting that the monolayers of PS spheres indeed enables to outcouple of light trapped in the scintillator. However, it should be noted that the transmission enhancement beyond the θ_c achieved by this structure is at the cost

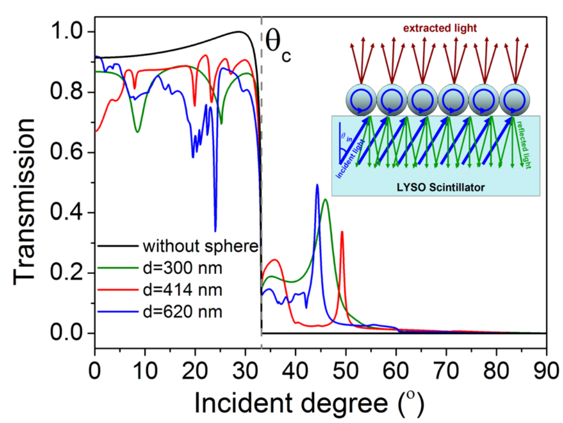

FIG. 1. Simulated transmission at 415 nm as a function of incident angle for the LYSO scintillator coated with the monolayers of PS spheres with different diameters.

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of the reduction of transmission below the θ_c. There are many dips with different number and width dependent on the diameters of spheres shown in the transmission spectra. It is evident that the dips increase in numbers and become wider with the increasing diameters of the spheres. The simulated and experimental transmission spectra in the normal direction are shown in Fig. 2. In order to obtain insight into the nature of the dips, the simulated transmission spectra include a monolayer of PS spheres (d = 414 nm) coated on the scintillator and a free standing monolayer of PS spheres. The transmission spectra are presented in terms of both wavelength and reduced frequencies $\omega = \sqrt{3} \cdot d/(2 \cdot \lambda)$. For the free standing monolayer (Fig. 2(a)), the four sharp transmission dips at ω = 0.874, 0.854,0.717, and 0.711 correspond to the leaky modes, which can strongly confine the electromagnetic field within or in the surroundings of the spheres depending on the Mie resonances (WGM) of individual spheres and the coupling among the spheres.[19] When putting the monolayer of PS spheres onto the scintillator substrate (Fig. 2(b)), these transmission dips show a slight frequency shift and the width are significantly broaden, which suggests that additional openings of dissipation channels through the substrate due to the converting of evanescent waves into propagating waves.[20] This also implies that the confinement of electric field by the spheres is broken, and as a result, the field is expanded into the substrate. Besides these dips, there is a broad Fabry-Perot-type oscillation with small amplitude, covering the whole range of wavelength, which is due to the contrast of effective refractive index of spheres and the substrate. With the diameters of sphere increases, these dips below θ_c in Fig. 1 become sharp and numerous because that the high-order WG modes (large *l* value) could be excited, which exhibit sharp profiles due to strong localization and Q factor. The experimental transmission spectrum (Fig. 2(c)) shows the characteristic dips at the same wavelengths compared with the simulated spectrum. These dips exhibit a little broadening, which is due to the size distribution of PS spheres in the experiment. The evident discrepancy is an overall low transmittance in the experimental spectrum compared with the simulated spectrum since the experiment involves the Fresnel reflection at the scintillator-air interface in the whole spectrum and the intrinsic absorption of LYSO for the wavelength below 410 nm.

The achievement of outcoupling to air is attributed to the diffraction effect. The diffraction could take place when the frequency ω is higher than $1/n_{sub}$ (0.546) for the normal incidence. While the incident light meets the periodic structure of dielectric constant, the wave vector will be split into different harmonics. The harmonics satisfying the following condition could be radiated into air:

$$|k_{//} + nG_0| < \frac{2\pi}{\lambda_0}, \tag{1}$$

where $k_{//}$ is the in-plane wave vector, λ_0 is the wavelength in vacuum, n is an integer, and $G_0 = 2\pi/d$ is the reciprocal lattice vectors with *d* the lattice constant. The light into air has an emergence angle as

$$\theta_{air} = \arcsin((\lambda_0/2\pi)|k_{//} + nG_0|). \tag{2}$$

The simulated angle dependence of the transmission spectra for *p* polarized incidence in the Γ-M direction of reciprocal space shown in Fig. 3 exhibits the WG modes (black solid lines), which is consistent with the dispersion relation described in Ref. 12. The emission spectra in the normal direction under excitation of an ultraviolet LED are shown in Fig. 4, which exhibit broad band emissions of Ce^{3+} peaked at 415 nm. All the samples coated with monolayers of PS spheres give rise to a significantly enhanced emission and the spheres with diameter of 414 nm perform the best. The enhancement ratios in the normal direction for different diameters of spheres with a function of wavelength are shown in the inset of Fig. 4. In this simulation, the multi-reflection by the lower interface between scintillator and air, the multi-diffraction by the monolayer spheres, and the absorption in the bulk of scintillator are taken into account. The simulation is consistent with the experimental result, which exhibits an evident wavelength-dependence. The large enhancement ratio for 414 nm spheres is in the range of 400–480 nm, which covers the main emission band of LYSO, leading to a significant extraction enhancement. Although the enhancement for 500 nm spheres is also observed, the

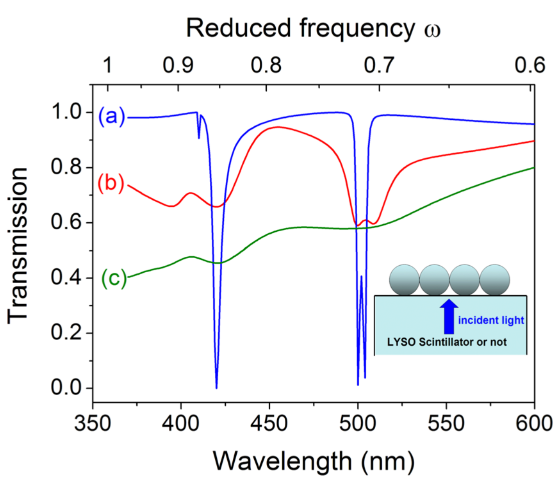

FIG. 2. Simulated transmission spectra in the normal direction for a free standing monolayer (a), and for a monolayer of PS spheres (d = 414 nm) coated on the scintillator (b). Experimental transmission spectrum in the normal direction for a monolayer of PS spheres (d = 414 nm) coated on the scintillator (c).

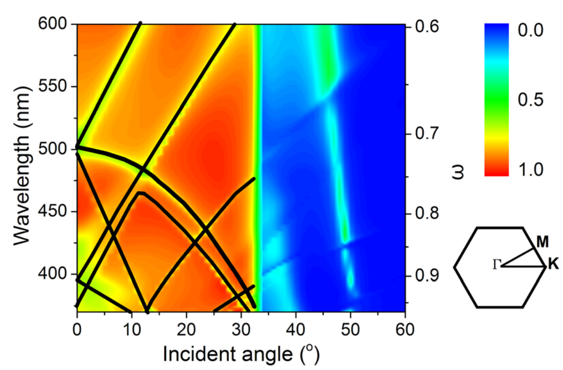

FIG. 3. Simulated transmission spectra of monolayer of PS spheres (d = 414 nm) coated on the scintillator as a function of incident angle for *p* polarized incident in the Γ-M direction of reciprocal space. The black solid lines indicate the WG modes.

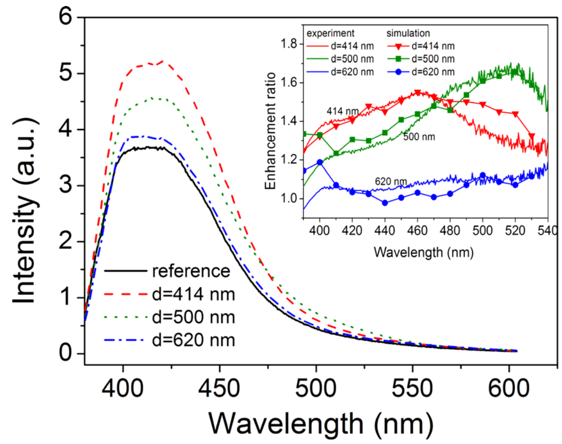

FIG. 4. Emission spectra excited by an ultraviolet LED for the structures with different diameters in the normal direction. The inset represents the enhancement ratio with respect to the reference sample, which shows a good consistence between experiment and calculation.

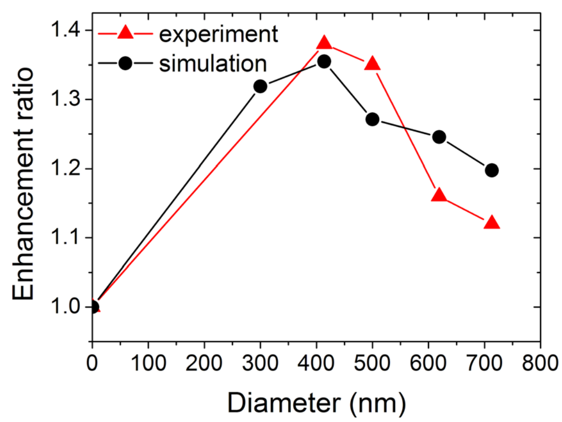

FIG. 5. Experimental and simulated enhancement ratio of light extraction at 415 nm emission with various diameters of spheres.

maximum locates at 520 nm, which is out of the main emission of LYSO.

The enhancement ratio of angle-integrating emission intensity at 415 nm by the monolayers of PS spheres relative to the reference sample is shown in Fig. 5. The experimental extraction efficiency increases by up to 38% for an optimal diameter with 414 nm. Although the experimental results are in agreement with the simulation in general, it is worthwhile to note that the size distribution of PS spheres in the experiment can cause some discrepancy with simulation. In the present experiment, the size distribution of the spheres has a standard deviation below 10%. The effect of size distribution would lead to broadened dips in transmission for the individual spheres and form average lattice constants for the arrays, which can be accurately dealt with using a method of the weighted average of size distribution.[14]

In order to ensure a high extraction efficiency, we must consider the trade-off between the transmission increase beyond the θ_c and the transmission decrease below θ_c. Looking back into the Fig. 1, when the diameter increases, the number of transmission dips significantly increases as a consequence of the excitation of higher order WG modes, which gives rise to the detriment to the light extraction. Therefore, in order to obtain an optimal result, it is required that the diameter should be small enough for ensuring that only low order WG modes can be excited, and at the same time, it should be large enough for ensuring that the diffraction can take place effectively. As a result, experimental and simulated results suggest that the optimal diameter is close to the wavelength involved.

In summary, a monolayer of periodic arrays of PS spheres is an effective structure to enhance the light extraction of LYSO scintillator. The experimental results show the maximum enhancement of light extraction of up to 38% can be obtained for the PS spheres with the diameter of 414 nm. Balancing the loss and the attribution to the light extraction, it is concluded that the spheres with diameter close to the emission wavelength could be an optimal choice.

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[1]W. Chewpraditkul, M. L. Swiderski, F. M. Moszynski, M. T. Szczesniak, M. A. Syntfeld-Kazuch, C. Wanarak, and P. Limsuwan, IEEE Trans. Nucl. Sci. **56**, 3800 (2009).
[2]K. McGroddy, A. David, E. Matioli, M. Iza, S. Nakamura, S. DenBaars, J. S. Speck, C. Weisbuch, and E. L. Hu, Appl. Phys. Lett. **93**, 103502 (2008).
[3]J. Jewell, D. Simeonov, S.-C. Huang, Y.-L. Hu, S. Nakamura, J. Speck, and C. Weisbuch, Appl. Phys. Lett. **100**, 171105 (2012).
[4]J. J. Wierer, Jr., A. David, and M. M. Megens, Nature Photon. **3**, 163 (2009).
[5]E. Matioli, E. Rangel, M. Iza, B. Fleury, N. Pfaff, J. Speck, E. Hu, and C. Weisbuch, Appl. Phys. Lett. **96**, 031108 (2010).
[6]K. Ishihara, M. Fujita, I. Matsubara, T. Asano, S. Noda, H. Ohata, A. Hirasawa, H. Nakada, and N. Shimoji, Appl. Phys. Lett. **90**, 111114 (2007).
[7]C. Liu, V. Kamaev, and Z. V. Vardeny, Appl. Phys. Lett. **86**, 143501 (2005).
[8]A. Knapitsch, E. Auffray, C. W. Fabjan, J.-L. Leclercq, P. Lecoq, X. Letartre, and C. Seassal, Nucl. Instrum. Methods Phys. Res. A **628**, 385 (2011).
[9]A. Knapitsch, C. W. Fabjan, J.-L. Leclercq, X. Letartre, R. Mazurczyk, and P. Lecoq, 2011 IEEE Nuclear Science Symposium Conference Record N18-7 (2011), p. 994.
[10]P. Pignalosa, B. Liu, H. Chen, H. Smith, and Y. Yi, Opt. Lett. **37**, 2808 (2012).
[11]J. F. Galisteo-López, M. Ibisate, R. Sapienza, L. S. Froufe-Pérez, Á. Blanco, and C. López, Adv. Mater. **23**, 30 (2011).
[12]H. T. Miyazaki, H. Miyazaki, K. Ohtaka, and T. Sato, J. Appl. Phys. **87**, 7152 (2000).
[13]J. Grandidier, D. M. Callahan, J. N. Munday, and H. A. Atwater, Adv. Mater. **23**, 1272 (2011).
[14]J. Grandidier, R. A. Weitekamp, M. G. Deceglie, D. M. Callahan, C. Battaglia, C. R. Bukowsky, C. Ballif, R. H. Grubbs, and H. A. Atwater, Phys. Status Solidi A **210**, 255 (2013).
[15]C. R. Yonzon, E. Jeoungf, S. L. Zou, G. C. Schatz, M. Mrksich, and R. P. V. Duyne, J. Am. Chem. Soc. **126**, 12669 (2004).
[16]Y. Li, T. Sasaki, Y. Shimizu, and N. Koshizaki, J. Am. Chem. Soc. **130**, 14755 (2008).
[17]Y. Li, X. S. Fang, N. Koshizaki, T. Sasaki, L. Li, S. Y. Gao, Y. Shimizu, Y. Bando, and D. Golberg, Adv. Funct. Mater. **19**, 2467 (2009).
[18]X. Ye and L. Qi, Nano Today **6**, 608 (2011).
[19]M. López-García, J. F. Galisteo-López, C. López, and Á. García-Martín, Phys. Rev. B **85**, 235145 (2012).
[20]Y. Kurokawa, H. Miyazaki, and Y. Jimba, Phys. Rev. B **65**, 201102 (2002).