How Photonic Crystals Can Improve the Timing Resolution of Scintillators

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***Abstract*—Photonic crystals (PhCs) and quantum optics phenomena open interesting perspectives to enhance the light extraction from scintillating media with high refractive indices as demonstrated by our previous work. By doing so, they also influence the timing resolution of scintillators by improving the photostatistics. The present contribution will demonstrate that they are actually doing much more. Indeed, photonic crystals, if properly designed, allow the extraction of fast light propagation modes in the crystal with higher efficiency, therefore contributing to increasing the density of photons in the early phase of the light pulse. This is of particular interest to tag events at future high-energy physics colliders, such as CLIC, with a bunch-crossing rate of 2 GHz, as well as for a new generation of time-of-flight positron emission tomographs (TOFPET) aiming at a coincidence timing resolution of 100 ps FWHM. At this level of precision, good control of the light propagation modes is crucial if we consider that in a $2 \times 2 \times 20\text{-mm}^3$ LSO crystal, the time spread (peak to peak) of extracted photons can be as large as 400 ps considering simple light bouncing only. This paper presents a detailed analysis of the light propagation and extraction modes in a LSO crystal combining the LITRANI light ray tracing and the CAMFR PhC simulation codes. Ongoing measurement results are shown with an attempt to unfold the contribution from the improved photostatistics that result from the total enhanced light output on one side and from the improved contribution of fast propagation mode extraction on the other side. Some results are also shown on a new and more industrial process to produce PhCs by the use of nano-imprint technology.**

***Index Terms*—Crystals, photonic crystal, positron emission tomography (PET), scintillation detectors, scintillation yield.**

I. INTRODUCTION

SEVERAL years ago, we identified photonic crystals (PhCs) as a tool with a high potential to significantly improve the performance of scintillator-based detectors [1], [2]. We have shown that well-designed PhCs can enhance the light output of dense scintillators with a high refractive index (between 1.8 and 2.2 for the majority of scintillators) by a large factor (>2 in air and by at least 50% in grease) [3], [4]. Indeed, they enable photons that are inside the crystal and impinging at angles greater than the Brewster angle (and thus usually undergo total internal reflection) to be transmitted, thus increasing the amount of scintillation light extracted. As a consequence, PhCs can offer attractive perspectives in the search for higher timing resolution in scintillator-based detectors [5].

Increasing the light output and, as a consequence, the rate of photoelectrons produced in the photodetector at the early stage of the signal generation has a direct impact on the timing resolution by virtue of the improved photostatistics as illustrated by the following formula derived from Hyman theory:

$$\Delta t \propto \frac{\sqrt{\tau}}{\sqrt{N_{\mathrm{phe}}/\mathrm{ENF}}} \qquad (1)$$

where τ is the scintillator decay time, $\mathrm{N_{phe}}$ is the number of photoelectrons and ENF is the excess noise factor of the photodetector.

Besides the gain in photostatistics, another factor, not apparent in the simplified formula (1), plays a determinant role for timing resolution. It is related to the scintillator rise time that affects the photoelectron density in the early stage of the signal, where the ultimate timing information is embedded [6]. As we have shown in a previous paper [7], several factors contribute to the rise time of the signal produced by a scintillator. Some of them are related to the light production mechanisms, and we have discussed them in detail in [7]. However, we have also shown in [8] that the light transport from the emission point in the crystal to the photodetector is playing a significant role if we were aiming at timing resolutions in the 100-ps FWHM range.

Section II describes the simulation tools used in this study as well as the procedure to produce the PhC samples.

Section III discusses how PhCs can be used to modify the balance of the number of photons distributed in the different propagation modes and what gain in timing resolution can be expected with a proper design.

Section IV presents measurement results on PhC samples. For practical reasons related to the equipment presently available for manufacturing the PhCs, the crystal samples are small and therefore not very sensitive to light transport time spread. Nevertheless, they show a significant improvement in timing resolution as compared to untreated crystals.

In Section V, we are describing our nano-imprint approach in view of developing a cost-effective mass production technology for nanostructuring the coupling face of the crystals to the photodetector.

Section VI is a discussion on the longer-term expectations and perspectives.

II. EXPERIMENTAL PROCEDURES

A. Simulations

The propagation of photons in the crystals was simulated using the light ray-tracing program LITRANI [9], whereas the LSO photo-absorption and Compton scattering coefficients of the crystals were taken from the NIST database [10].

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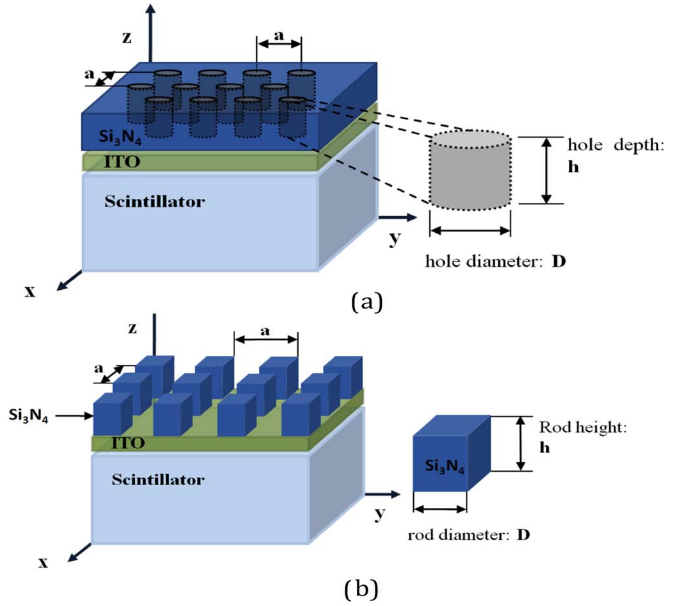

Fig. 1. PhC structures with (a) cylindrical holes and (b) cubic pillars.

The optical transmission of photonic crystals was simulated with a "rigorous coupled wave analysis" (RCWA), based on a full-vectorial Maxwell solver based on the eigenmode expansion method CAMFR [11].

The two simulations, LITRANI and CAMFR, were used sequentially to optimize the parameters of the photonic crystal structure (thickness, lattice constant, diameter of holes).

B. Sample Production

To evaluate the simulation results, several samples were manufactured using state-of-the-art E-beam lithography (EBL) and reactive ion etching (RIE) techniques. The fabrication process basically involves four steps:

1) sputter deposition of the pattern transfer material;
2) resist spinning;
3) electron beam writing;
4) reactive ion etching.

Since the scintillator material itself cannot be etched by standard nano-lithography, we deposited an auxiliary 300-nm silicon nitride layer on top of a 70-nm indium tin oxide (ITO) layer by sputter deposition. The conductive ITO layer is aiming at removing the charges during the E-beam lithography process and has no optical contribution. A resist layer of poly-methyl-methacrylate (PMMA) is then applied by spin coating to prepare for the third step that consists of writing the pattern with the electron beam. After the development of the pattern by chemically resolving the exposed areas of the PMMA resist, the pattern is etched into the silicon nitride by reactive ion etching.

We have designed and optimized six PhC structures (labeled P1 to P6) consisting of either cylindrical holes or of cubic pillars (Fig. 1). For each PhC design, the light diffraction properties at a fixed wavelength are governed by the following parameters:

- index of refraction of the bulk material;
- index of refraction of the filling material;
- lattice constant a;
- filling factor f.

TABLE I
PhC CHARACTERISTICS

Parameters	P4	P6
Period designed (nm)	640	640
Period measured (nm)	616	622
Gap designed (nm)	110	110
Gap measured (nm)	146	200
Light output gain/P7	1.33	1.56

For our prototype, we have structured several PhC patterns $1.2\times 2.6\ \text{mm}^2$ each on the 1-cm^2 face of a $10\times 10\times 5$-mm^3 block of LSO. We have then cut the block to produce six pixels (P1 to P6) of $1.2\times 2.6\times 5\ \text{mm}^3$ with a different PhC pattern on top of each of them. Another block of similar size but unpatterned has been cut from the same slab to be used as reference (P7). The choice of these dimensions was driven by technical constraints for the preparation of the PhC as well as by the need to decrease the time spread between the different light propagation modes due to multiple bouncing (see Section III). For the purpose of this work on timing, we have selected patterns P4 and P6, for which we have measured a light output gain of 1.33 and 1.56, respectively [4].

Their characteristics are listed in Table I.

III. EFFECT OF PhCs ON LIGHT PROPAGATION MODES IN A SCINTILLATOR

We have already shown in [8] that for an emission point in the center of a $2\times 2\times 20$-mm^3 LSO crystal ($n_{\text{LSO}} = 1.82$, $L = 20$ mm) coupled to a photodetector with silicon grease of $n_{\text{Si grease}} = 1.41$, the maximum time spread between the shortest path-length photon, emitted forward along the crystal axis, and the longest path-length photon, emitted backwards at the total reflection angle on the lateral faces, is given by

$$\Delta t_{\text{max photons}} = \frac{2L}{c} \cdot \frac{n_{\text{LSO}}^3}{n_{\text{Si grease}}^2} = 404\ \text{ps} \tag{2}$$

not considering multiple reflections on the extraction face. Simulation shows that without a PhC, nearly 50% of the photons are reflected at the first incidence and reappear for several times (if not absorbed) with a delay ranging from 243 and 384 ps times the number of recurrences n_{rec} of the light rays on the coupling face to the photodetector.

With a PhC grating, more photons are being extracted the first time they hit the "out-coupling" face of the crystal. This effectively increases the number of fast photons and decreases the number of back-reflected photons reappearing later, as shown in the light ray-tracing simulation plots (Litrani) in Fig. 2.

On the other hand, there is also a chance that the PhC structure extracts a large fraction of photons, delayed from multiple reflections on the crystal sidewalls, which normally would not have been extracted because of too large an incidence angle with the extraction face. These photons would of course not contribute to the improvement of timing resolution if they were detected too late w.r.t. the first detected photon.

In this work, we wanted to get a better understanding of the propagation modes in a scintillator in order to optimize the PhC for best timing resolution. Simulation results are shown

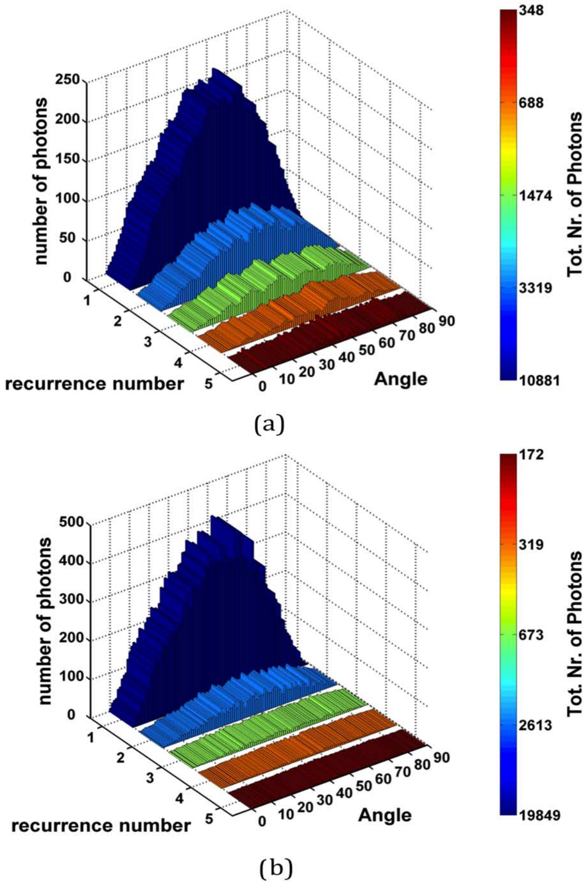

Fig. 2. Angular distribution of photons at each recurrence on the extraction face (a) without PhC and (b) with PhC.

in Figs. 3 and 4 for the actual PhC (pattern 6) and for the "as designed" PhC, respectively, deposited on a $1.2 \times 2.6 \times 5$-mm^3 LSO crystal. We have chosen this crystal and this PhC pattern because they have been fully characterized in terms of light output improvement. The deviations between the design pattern and the actually produced PhC have been precisely measured. For both cases, the simulation results of an unwrapped LSO crystal are shown since wrapping does not necessarily improve the timing performance (wrapping mainly improves the collection of photons delayed by multiple reflections).

It clearly appears that for this crystal geometry, almost no gain is to be expected in the 0–50 ps range. This is not surprising as these photons correspond to the fastest propagation mode (forward emission along the crystal axis) and would be extracted anyway. However, a significant gain is visible in the 50–100 ps range, corresponding to photons emitted at larger angles and being reflected a few times on the lateral crystal walls before reaching the coupling face at an angle for which extraction is only allowed if the PhC is present. The influence of the deviations in the geometry of the produced pattern as compared to the designed one is also visible.

IV. RESULTS

The timing measurements were made in a coincidence setup optimized for precision time of flight (TOFPET) studies.

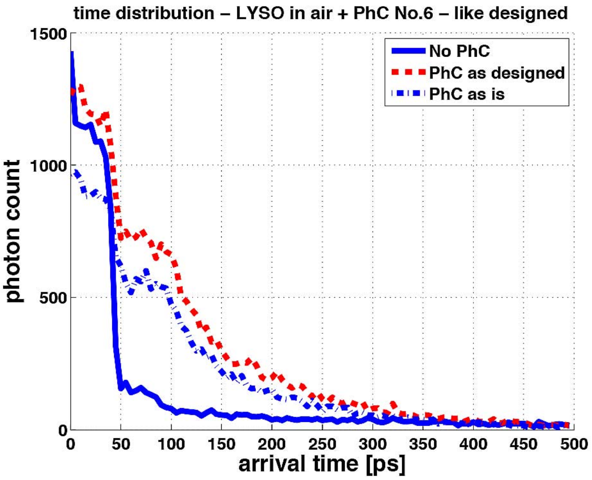

Fig. 3. Time distribution of extracted photons for the "as designed" and "as produced" PhC compared to an untreated LSO crystal.

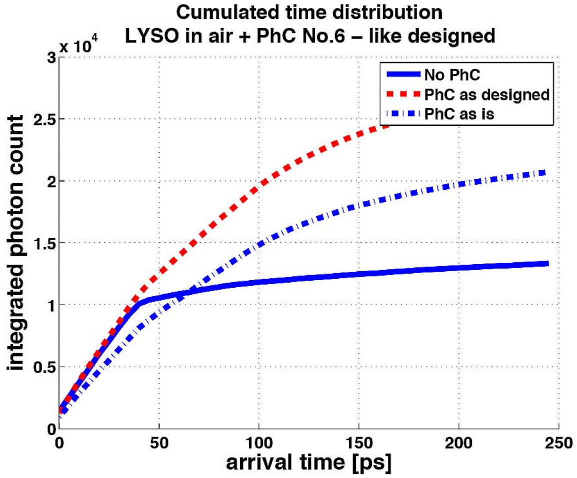

Fig. 4. Integrated photon yield over time for the "as designed" and "as produced" PhC compared with an untreated LSO crystal.

The crystal under study was installed in a black PVC holder without wrapping and in dry contact with a Hamamatsu MPPC (10931-050p) with 3600 50-μm single photon avalanche diodes (SPADs). On the opposite detector arm, a reference LYSO-crystal of $2 \times 2 \times 10$ mm^3 was mounted and fully wrapped with Teflon and coupled with Rhodorsil optical glue to a similar MPPC. The front-end electronics are based on the time-over-threshold method using the fast and low noise NINO discriminator as described in [12].

Each measurement was repeated several times, limiting the error to $\pm 3\%$. As expected from the improved light output, samples P4 and P6 exhibit an improvement in coincidence time resolution (CTR) with the reference LSO crystal as compared to the unpatterned crystal. Normalized to the unpatterned crystals, the measured relative CTR has visibly improved and is 87% and 74% for samples P4 and P6, respectively, normalized to 100% for the reference P7. It is interesting to notice that the expected value resulting from the improved photostatistics should be 88% for P4 and 80% for P6 (Table II). The observed additional gain

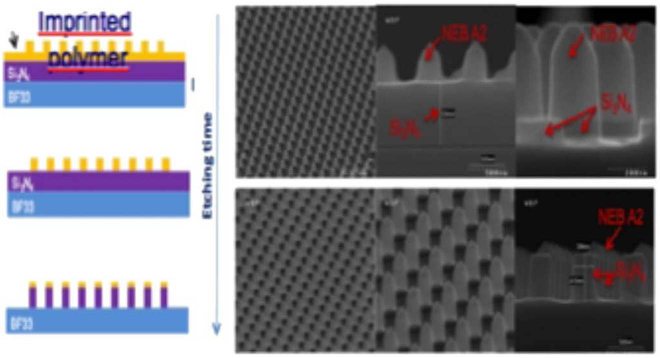

Fig. 5. Soft nano-imprinting technology applied to a Si_3N_4 layer deposited on a BF33 glass sample. *(left)* Etching phases as a function of time from top to bottom. *(right)* Photographs taken in the beginning (top) and at the end (bottom) of the etching process. The polymer used for this test was NEB A2.

TABLE II
RELATIVE CTR IMPROVEMENT FOR PhC SAMPLES P4 AND P6 COMPARISON WITH EXPECTED GAIN FROM IMPROVED LIGHT OUTPUT

Relative parameter value/P7 (un-patterned)	P4	P6
Light output	1.33	1.56
Expected relative CTR from photostatistics	0.88	0.80
Measured relative CTR	0.87	0.74

on timing is compatible with the modification of the time distribution of the detected photons induced by the PhC with a significant increase of their number in the 50–100 ps range as shown in Fig. 3. Although small, because of the small size of the crystal that reduces the influence of photon travel time on the timing resolution, the observed effect is significant and larger than the measurement error, at least for the P6 sample. Other tests are being prepared with crystal samples of different length ranging from 5 to 30 mm.

V. PHOTONIC CRYSTALS NANO-IMPRINT PRODUCTION

Considering the good results already obtained in terms of light output gain and the very encouraging ones on the timing resolution, we have started a study to mass-produce PhCs in a cost-effective way. Indeed, the technology used for the preparation of our samples is very flexible, allowing us to test different patterns. However, the electron beam patterning we have used for preparing our samples is time-consuming and expensive.

We are therefore developing a new method based on the nano-imprint technology that will allow patterning of the whole surface of a slab cut from a crystal ingot before cutting the slab into pixels. The principle is to produce a nanostructured Si stamp that will be used to imprint the resin (PMMA) deposited by standard spin-coating techniques on the slab.

For this purpose, CEA-LETI in Grenoble, France, has produced a stamp with a nanostructure design validated by our simulation programs. This stamp was made on an 8-in silicon wafer by UV lithography and dry etching. A first attempt to stamp the PMMA layer deposited by spin coating on a $1 \times 1\text{-cm}^2$ BGO crystal resulted in an inhomogeneous structure with imperfections correlated to surface defects on the BGO crystal. One possibility to alleviate this problem would be to optically polish the crystal face first to a flatness of $\lambda/2$ (about 250 nm). As this is likely to become cost-ineffective, we have developed another method. The hard silicon stamp is first used to imprint an intermediate polymer stamp, which is then used to produce a replicate of the initial pattern on a soft poly-dimethyl-siloxane film. This material is soft enough to compensate the flatness imperfections of the crystal surface, but hard enough to correctly imprint the spin-coated PMMA layer on this surface. First attempts on a 300-nm-thick Si_3N_4 layer deposited on a BF33 glass sample that was not specifically treated for good flatness show excellent results as exemplified in Fig. 5.

VI. DISCUSSION

The promising results presented in this paper should take into consideration the following remarks.

- The PhC pattern chosen for these tests has been constrained by practical limitations related to the equipment we had access to for the preparation of the samples. They have been modeled by simulation programs to quantify the expected gain in light output and timing resolution. Although the expected gain with this PhC configuration is significant (and confirmed by experimental results), it cannot be judged as being optimized for LSO. Further simulation studies are likely to design patterns with even better performance. We expect in particular a similar gain in light output and timing performance over untreated crystal optically glued to the photodetector by using a substrate with a higher index of refraction than Si_3N_4.
- The overexposure during the lithography process has led to a pattern slightly different from the designed one with reduced performance.
- The crystal thickness of 5 mm only strongly reduces the time spread between the different propagation modes. The sensitivity of the redistribution of the light propagation modes to the timing resolution is therefore limited with such small crystals. New tests on crystal samples of different lengths (5–30 mm) are in preparation to better assess and quantify the influence of this parameter.

An additional feature of the PhC is to collimate the light in some preferred directions, in particular in the forward direction. As the quantum efficiency of most of the photodetectors is angular-dependent, with a maximum at normal incidence, we intend to study PhC structures optimized for forward collimation and the additional gain that can be expected from the number of photoelectrons produced and in timing resolution.

So far, the PhC is structured on a few hundreds of nanometers of Si_3N_4 layer deposited on the crystal. The reason is the easy deposition of this layer by sputtering techniques and the well-understood etching procedures on this material. However, this additional step in the preparation of the crystals is not mandatory and adds cost to the process. We are therefore investigating methods to directly nanostructure the crystal itself.

VII. CONCLUSION

We have demonstrated that a PhC made of a nano-structured Si_3N_4 layer, deposited on the coupling face of a LSO crystal by standard lithography techniques, can significantly enhance the light output of the crystal and, therefore, the timing resolution as a result of improved photo statistics. Moreover, the PhC, if properly designed, allows a redistribution of the scintillation photons to the fast propagation modes in the crystal, further improving the timing resolution.

Nano-imprint technologies offer attractive perspectives to implement PhC structures on scintillator pixels in a cost-effective way.

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