RMD
A Dynasil Company

Photonic Crystal Enhances Scintillation Light Extraction: Can It Improve Timing Resolution?

B. Singh, M. S. J. Marshall, and V. V. Nagarkar, RMD, Inc. (Watertown, MA)
M. Salomoni, R. Pots, and P. Lecoq, CERN (Geneva, Switzerland)
K. Munechika and C. Pina-Hernandez, aBeam (Hayward, CA)

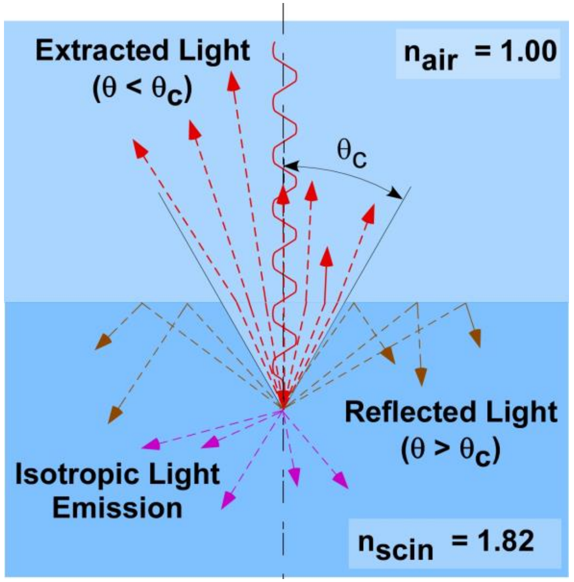
June 2nd, 2017

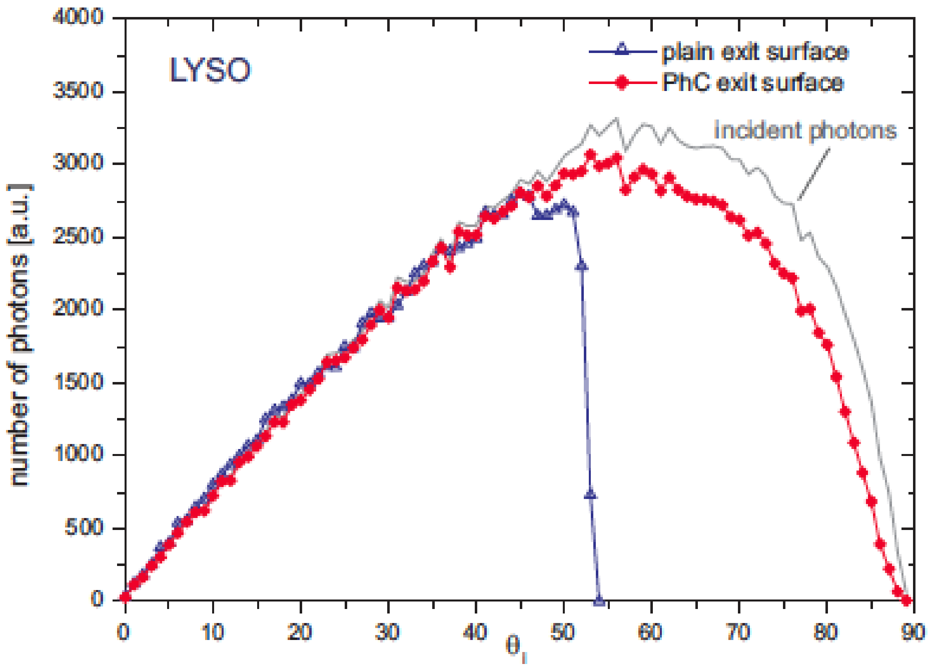
MEDAMI 2017 – V Mediterranean Workshop on Advanced Molecular Imaging, Orosei

This work was supported by the US Department of Homeland Security, Domestic Nuclear Detection Office, under competitively awarded contract(s) HSHQDC-13-C-B0040. This support does not constitute an express or implied endorsement on the part of the Government.

The Problem Definition

<u>Issue</u>: Overcoming refractive index mismatch at dielectric interface





A factor of two increase in photon flux output expected

IEEE Tran. Nucl. Sci., **57**(5), Part: 1, (2010) pp. 2475 – 2482
Int. J. Mod. Phys. A, **29**, (2014) 1430070
IEEE Tran. Nucl. Sci., **60**(3), (2013) pp. 1653 – 1657

From Snell's law

$$\theta_c = \sin^{-1}\left(\frac{n_{air}}{n_{scin}}\right)$$

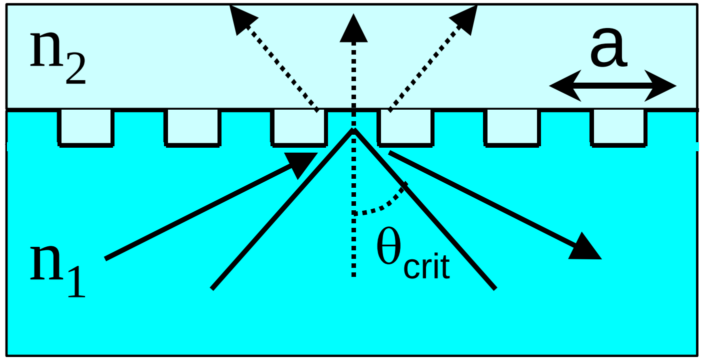
Larger RI mismatch → Smaller θ_c
Smaller RI mismatch → Larger θ_c

- Critical angle for scintillator with RI=1.82 coupled to RI=1.5: ~55°
- A large fraction of light hits the air/scintillator interface with angle > θ_c
- Due to the mismatch in refractive index between high-RI scintillator and low-RI window, light is lost due to total internal reflections.

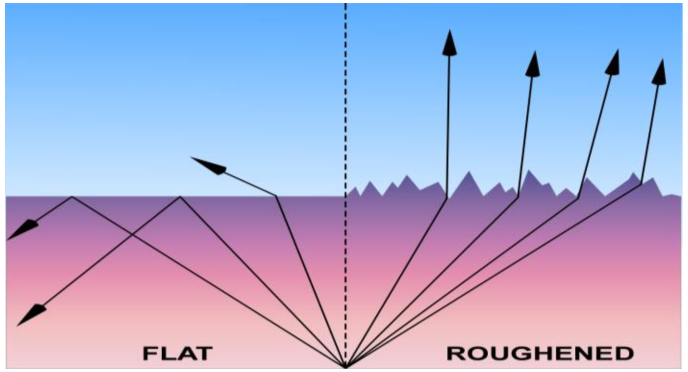
→ Light yield
Energy resolution
Timing resolution

What are the solutions?

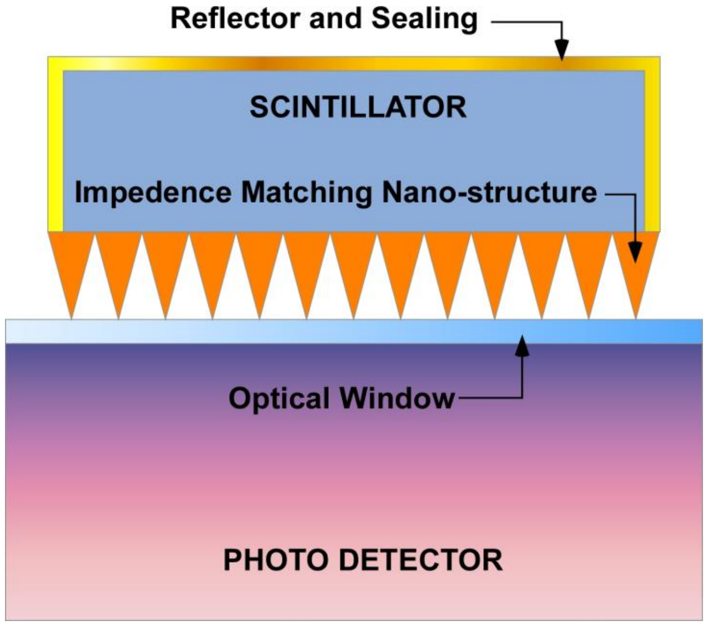
- To enable realization of the next-generation of high sensitivity, high performance detectors using *existing scintillation materials.*
- Grooves or gratings
- Surface roughness
- Photonic crystals (PhC) or gradient index nanostructures (GRIN)



Grooves or gratings



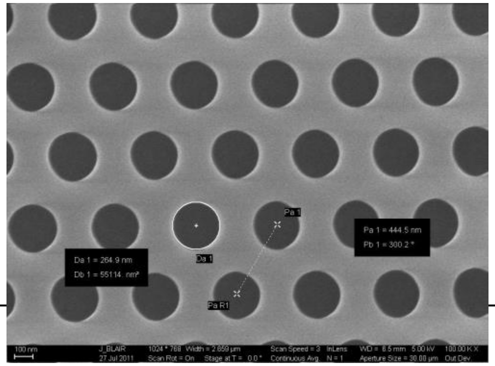
Surface roughening

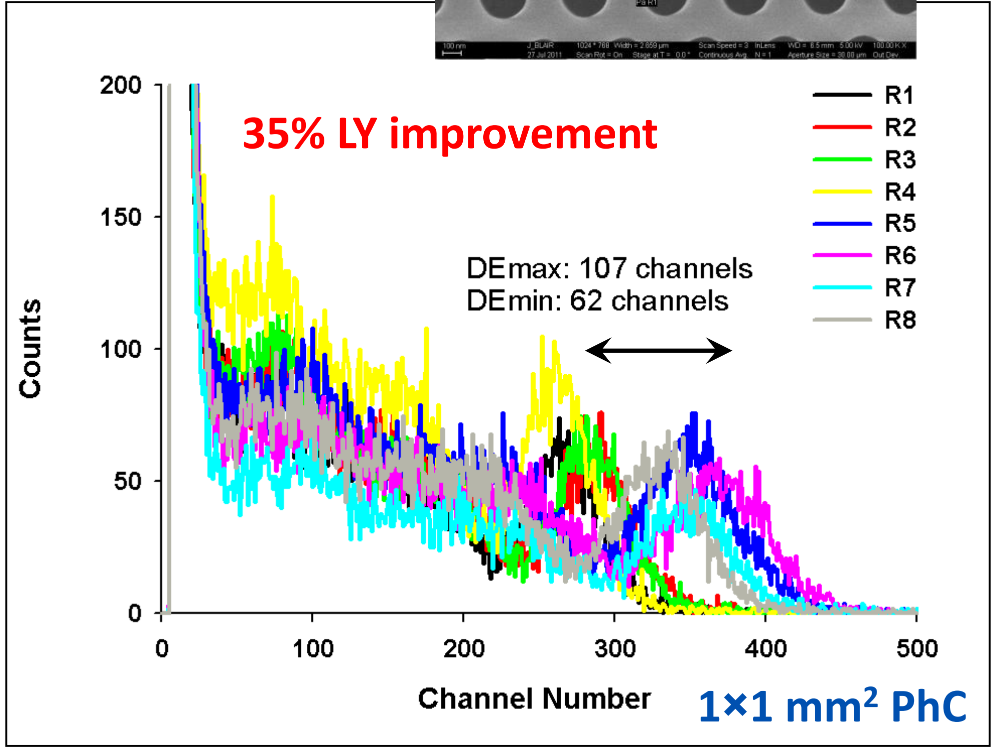


Photonic crystals or nanostructures

Photonic Crystals Fabrication Methods

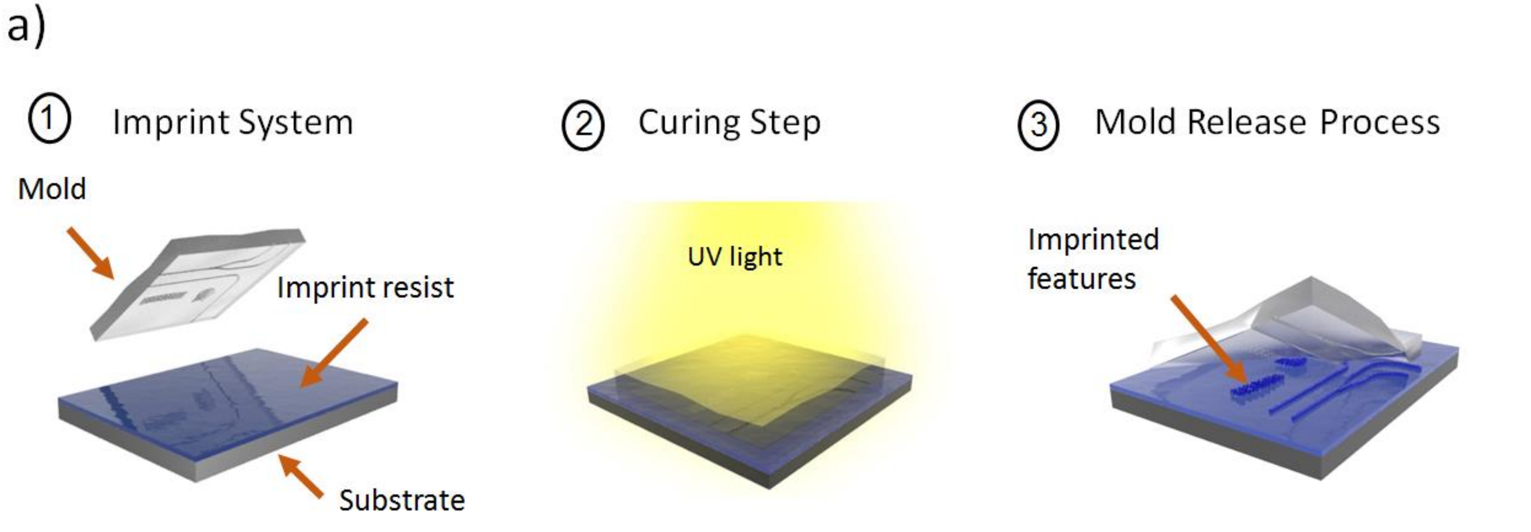
- Previously, we used e-beam lithography
  - Made PhCs (2mm×2mm) in high refractive index Si_3N_4
  - Demonstrated 35% improvement in light yield
  - E-beam lithography is expensive, slow, and can't be used for large areas
- Now, we use nanoimprinting
  - Low-cost technique
  - Flexible (mold material, polymer curing)
  - Repeatable
  - Scalable
  - Made large-area (1"×1" so far), low-cost PhC on scintillators
  - Demonstrated superior performance



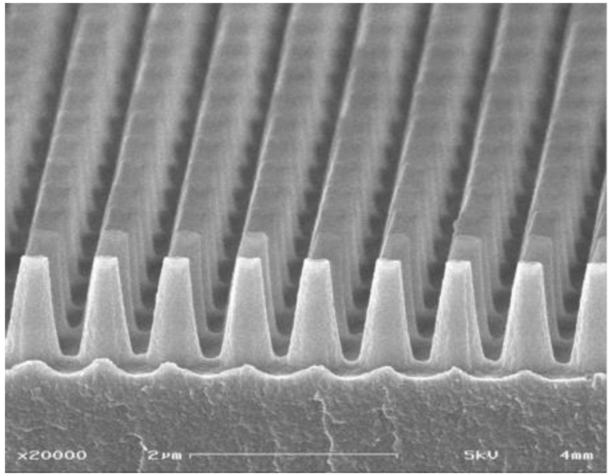


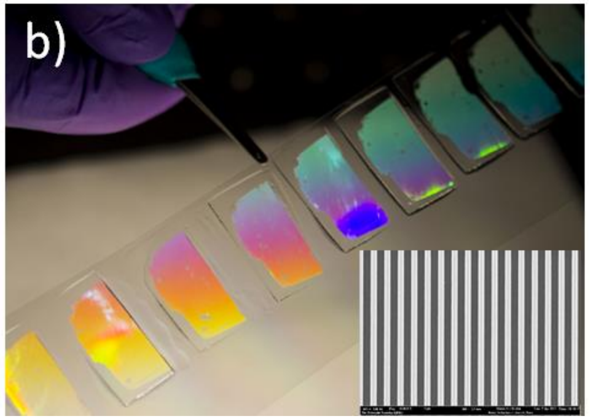
B. Singh, *et al.*, Proc. IEEE NSS/MIC, 2013

Nanoimprinting Process

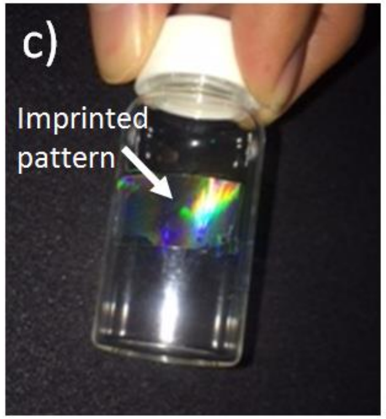


Conical PhC Master in Polymer

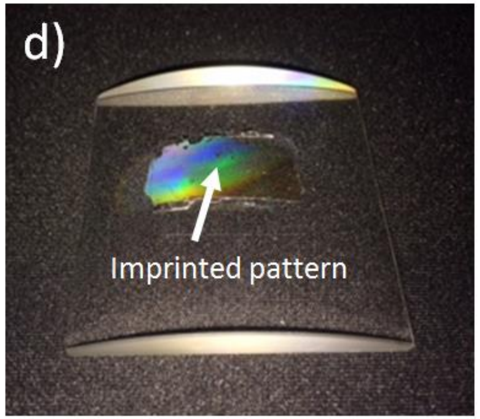




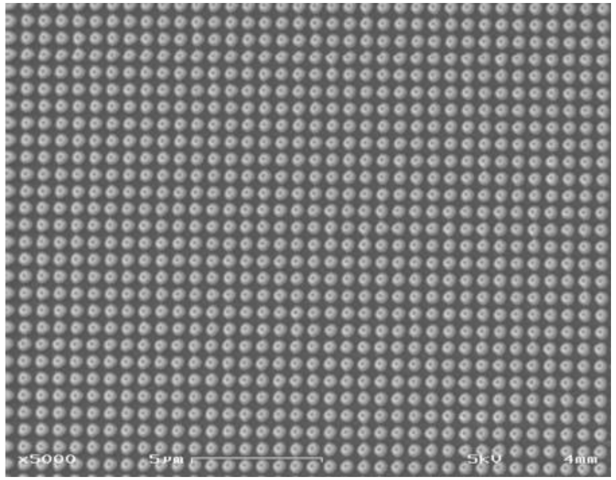
Optical grating (700 nm pitch)



Grating on a curved glass vial surface

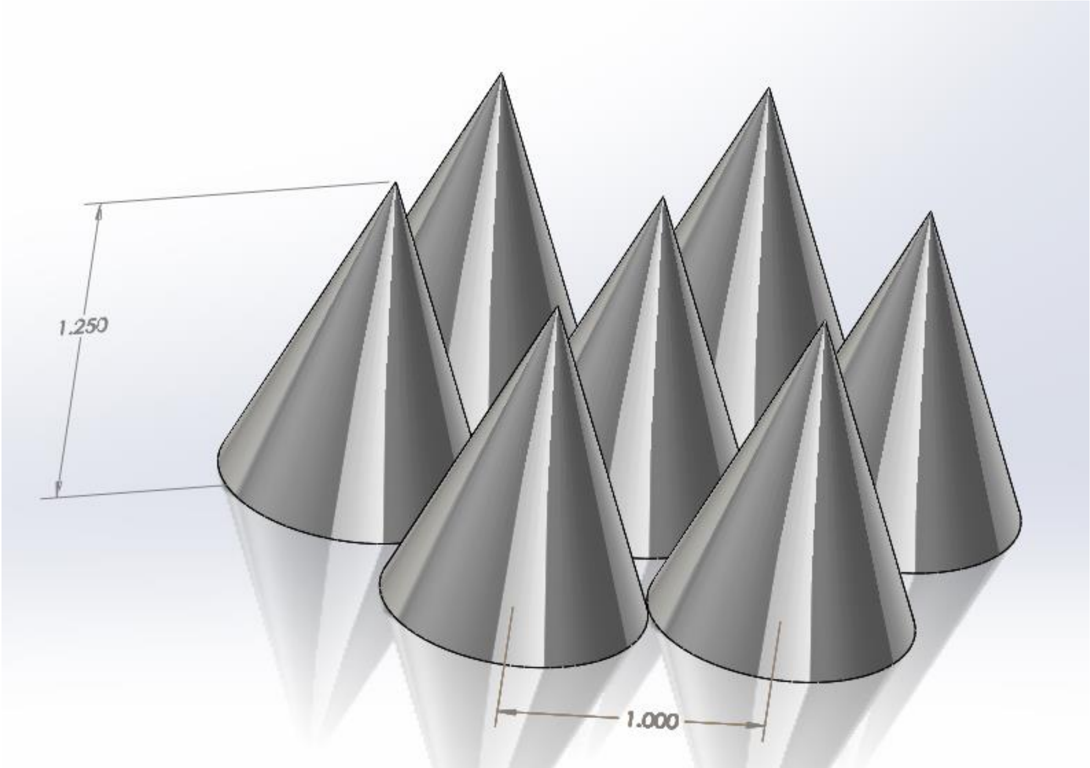


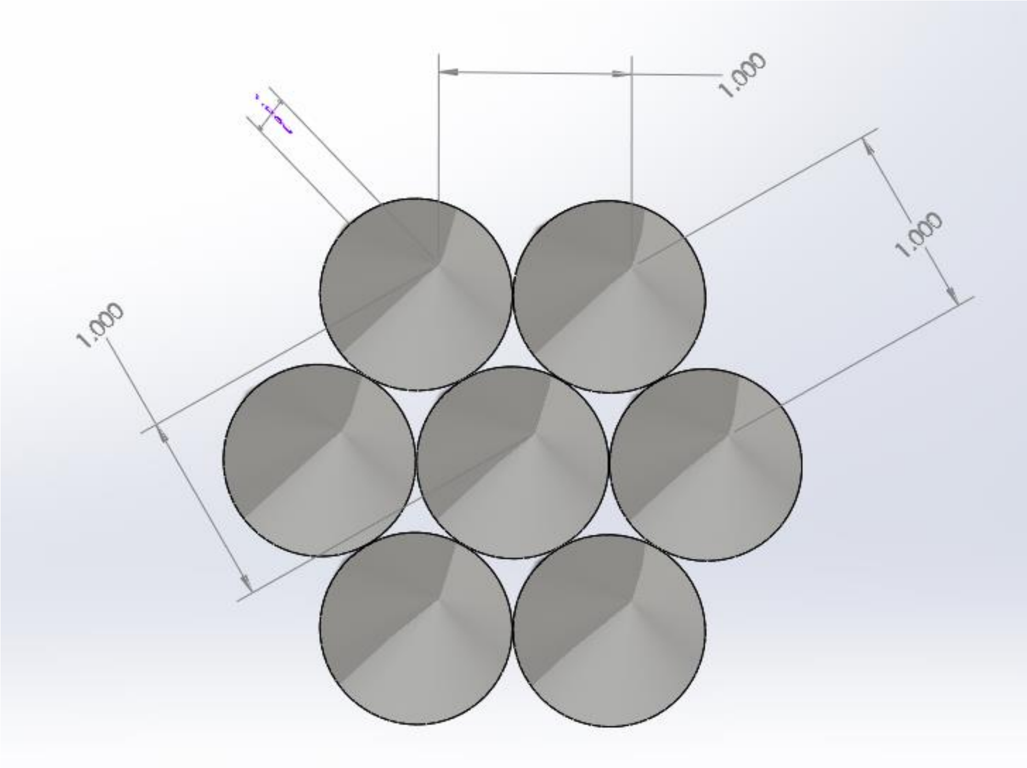
Grating on a cylindrical lens



Uniform, large-area coverage

Template Designed for Light Extraction Enhancements

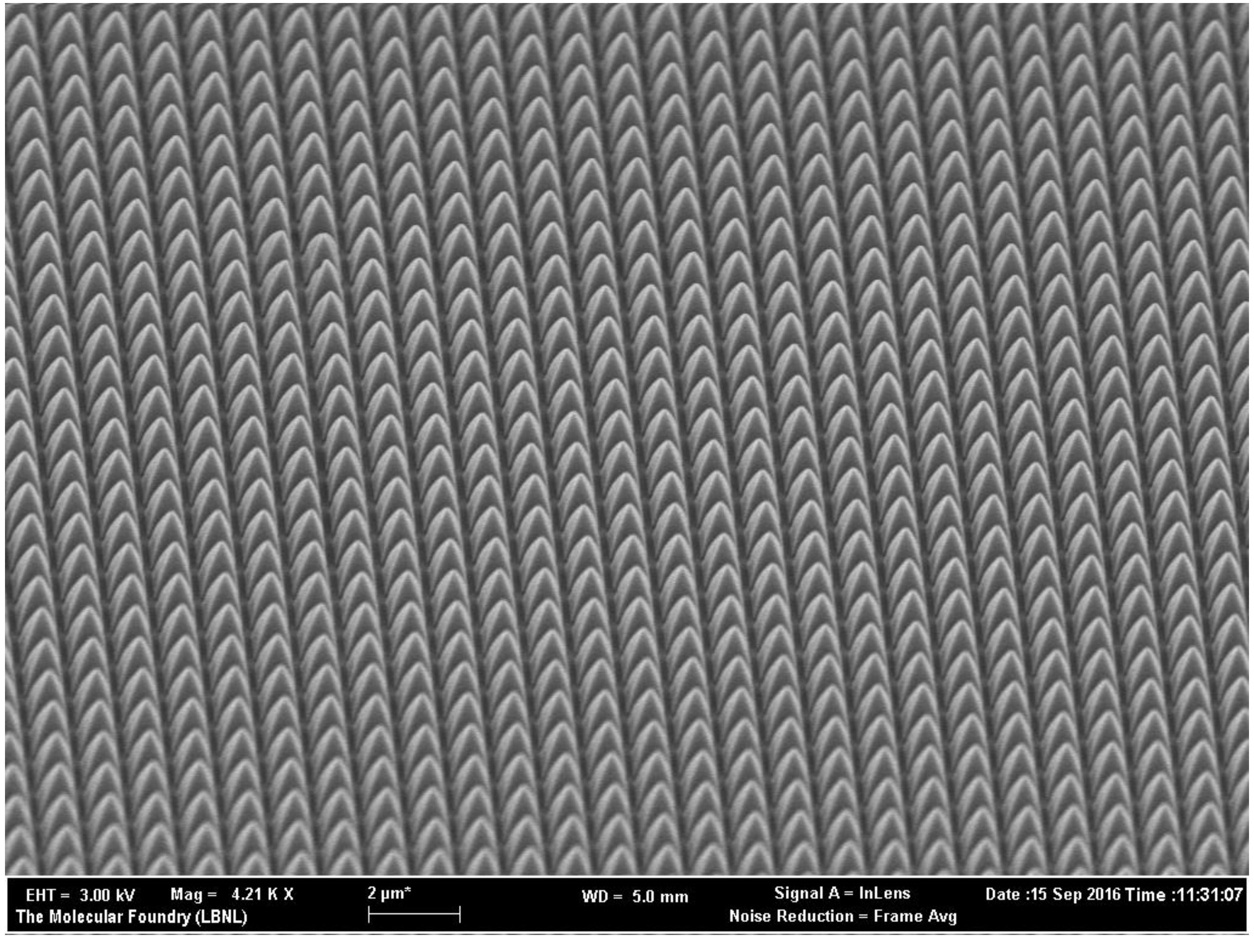




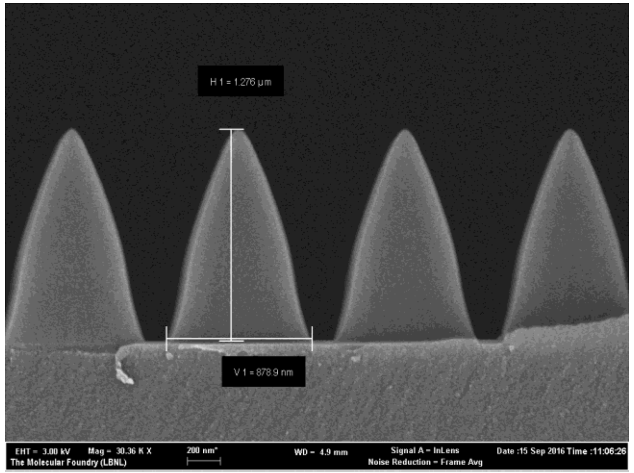
Optics Express, **23**(17), (2015) 22730

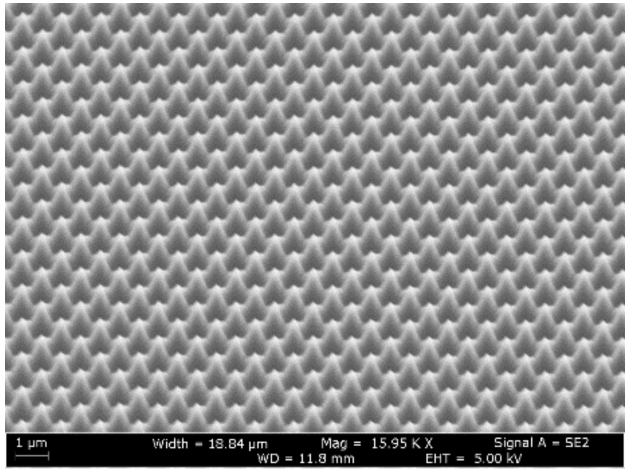
Hexagonally close-packed array of nanocones, with a pitch-to-height aspect ratio of approx. 1:1.25.
Optimum gain is obtained for aspect ratio of 1:1.25 to 1:1.5, substantially easing fabrication process.

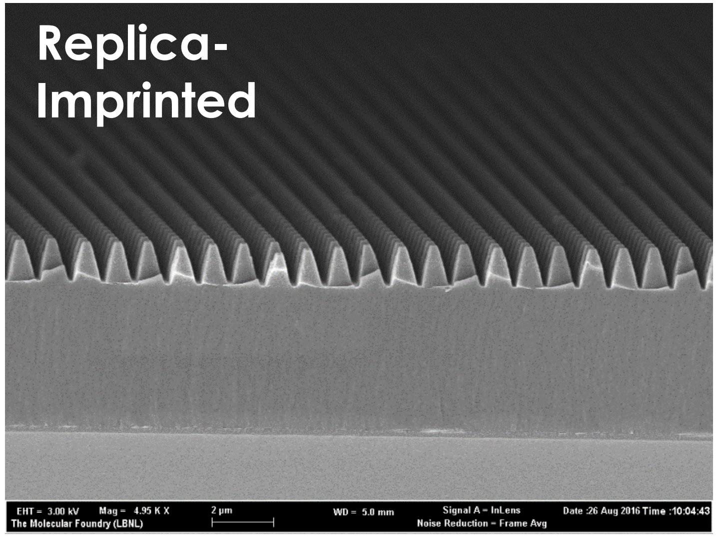
Nanoimprinting 1”×1” Master Mold



SEM image of the silicon template. The nanocones have a period of 800 nm, and a height of 1276 nm.

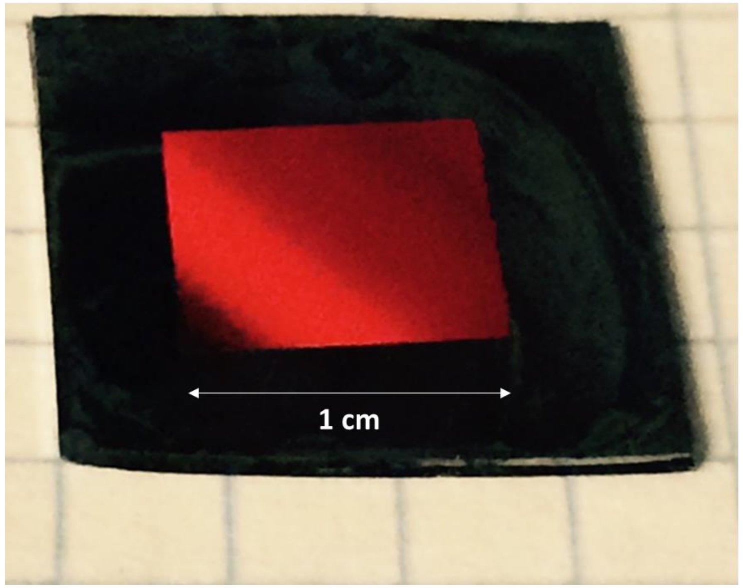




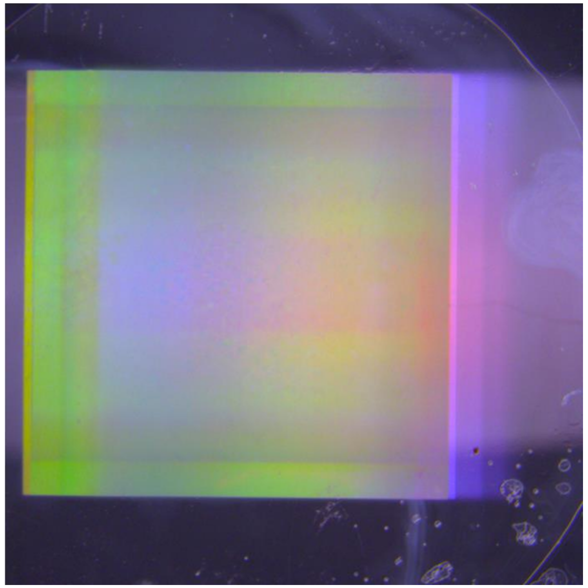


New New large molds – for 1 cm × 1 cm to 1”×1” GRINs

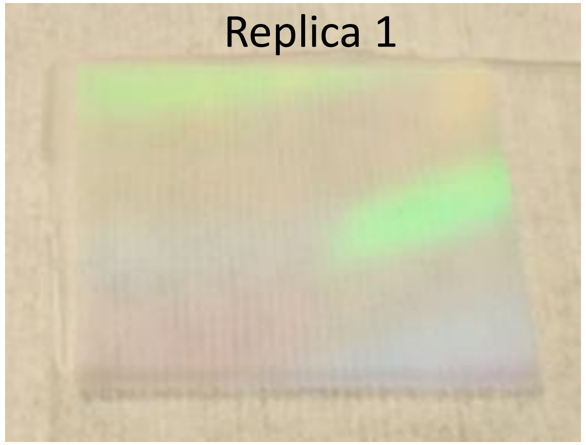
Master Mold

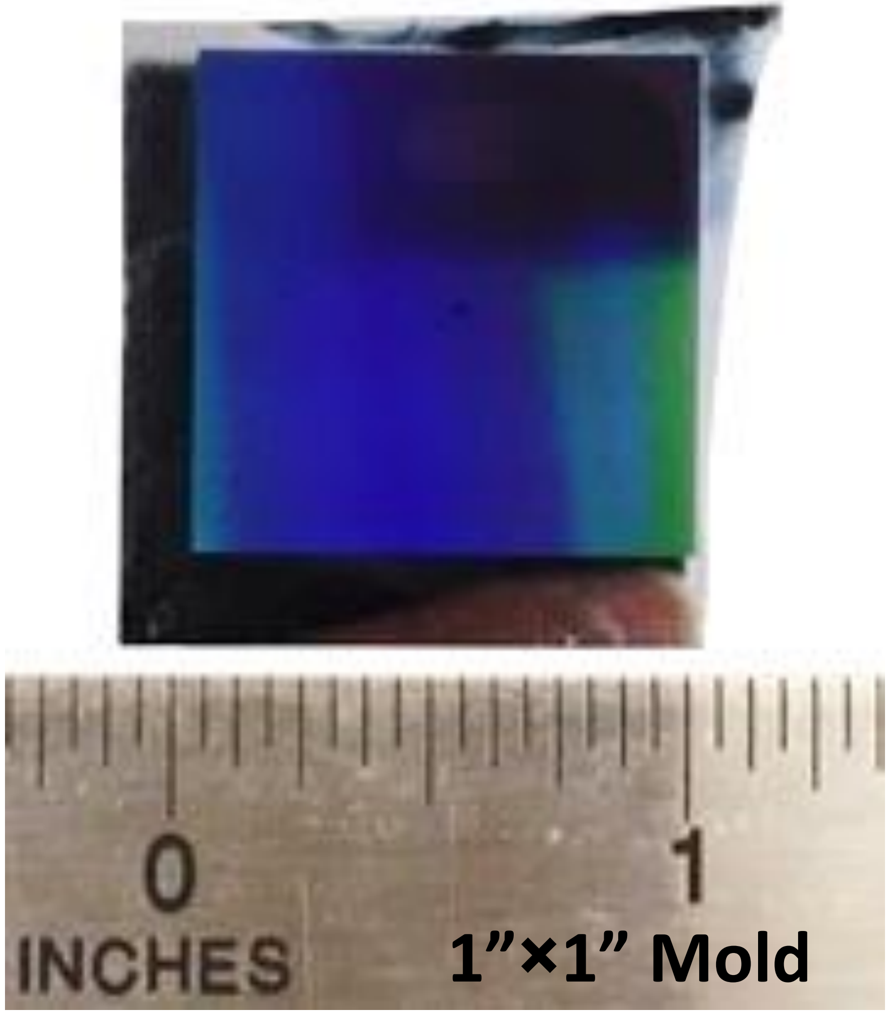


Imprinted Replica

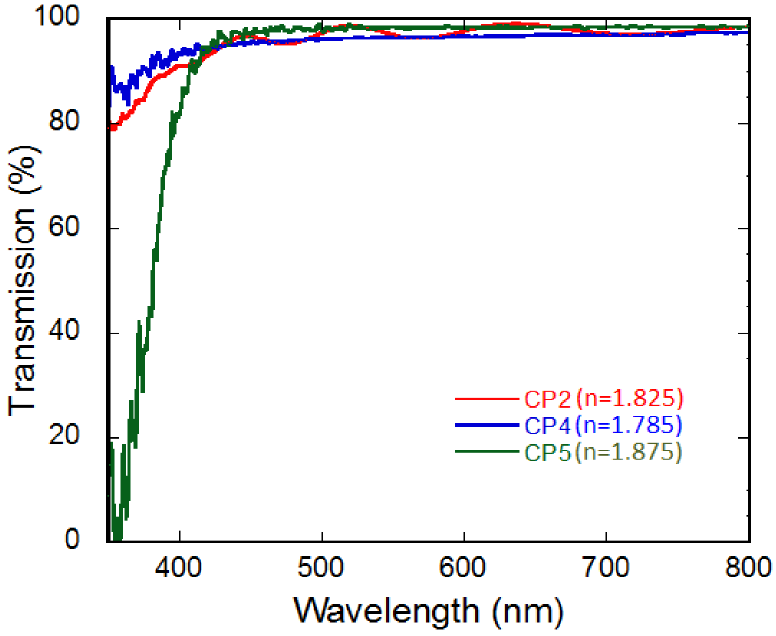


New large mold, for fabricating 1 cm – 1 inch GRINs.

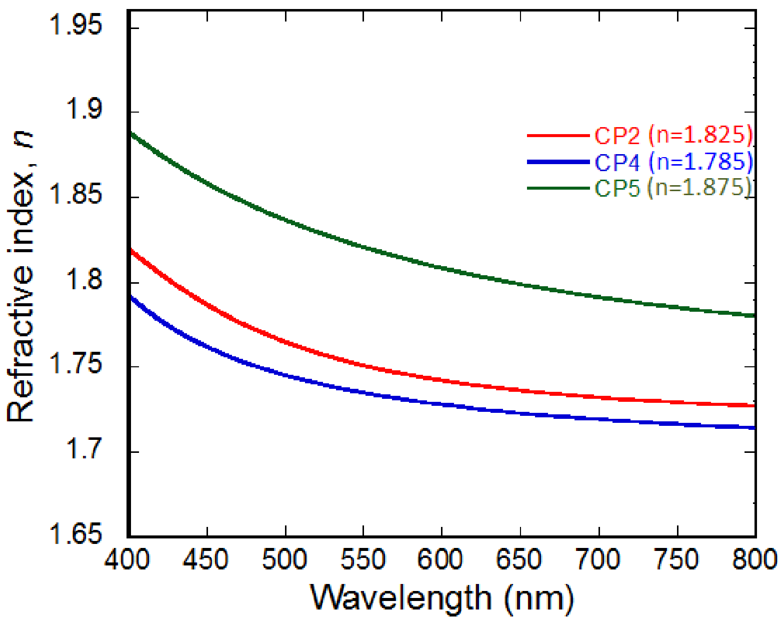




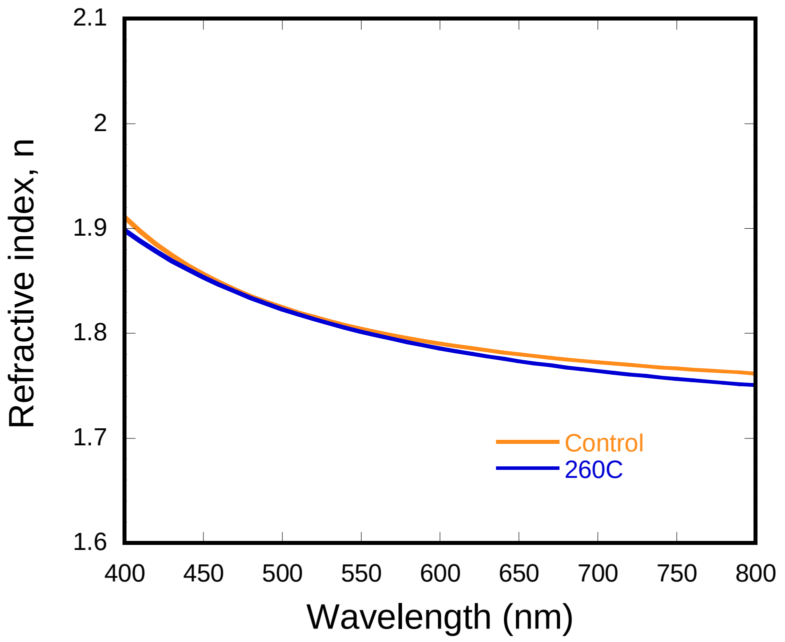
High Refractive Index Polymer



Transmission measured with UV-VIS spectrometer.



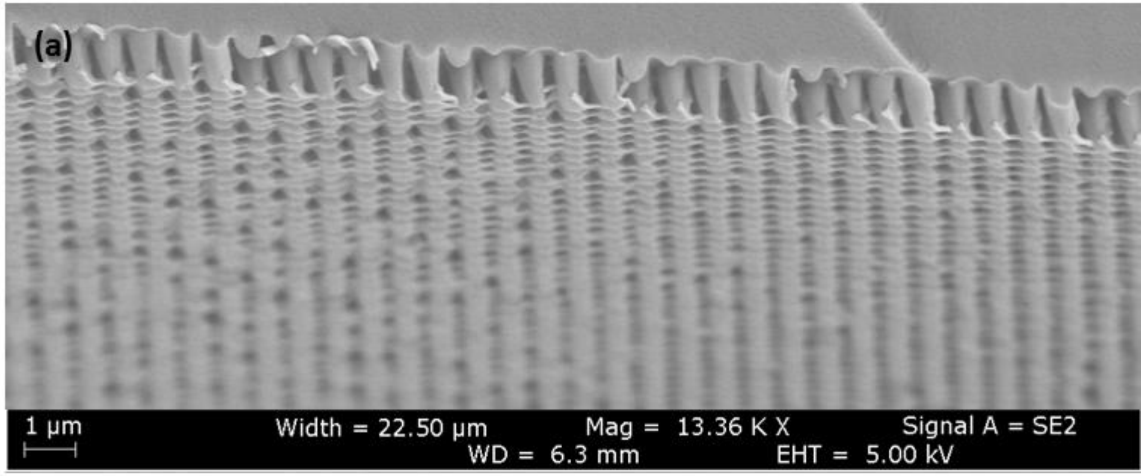
Refractive index of CP2, CP4, and CP5 polymers measured with an ellipsometer.

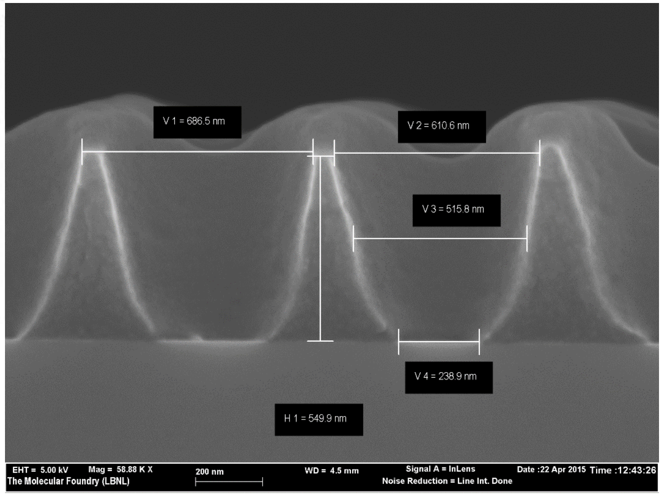


Refractive index is stable up to 260°C.

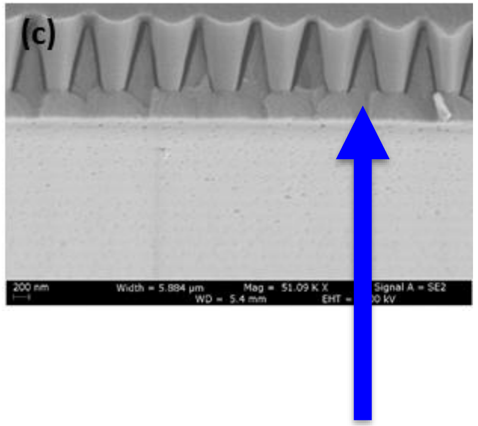
Polymer	Refractive Index	Wavelength	Curing method
CP2	1.825	400 nm	UV curing
CP4	1.785	400 nm	UV curing
CP5	1.875	400 nm	Thermal curing
CP5a	1.930	400 nm	Thermal curing

Characterization of Nanoimprinted Layers



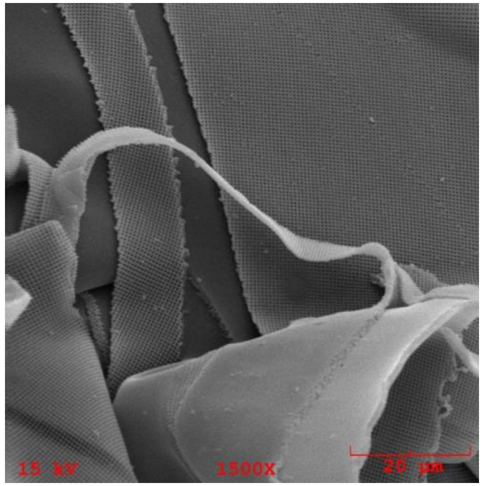


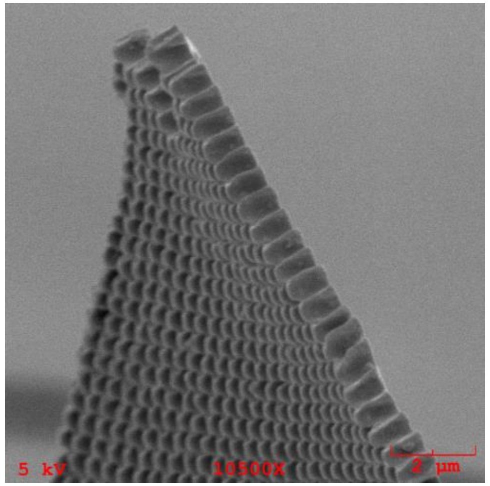
Zero residual layer



Residual layer of several hundred nanometers.

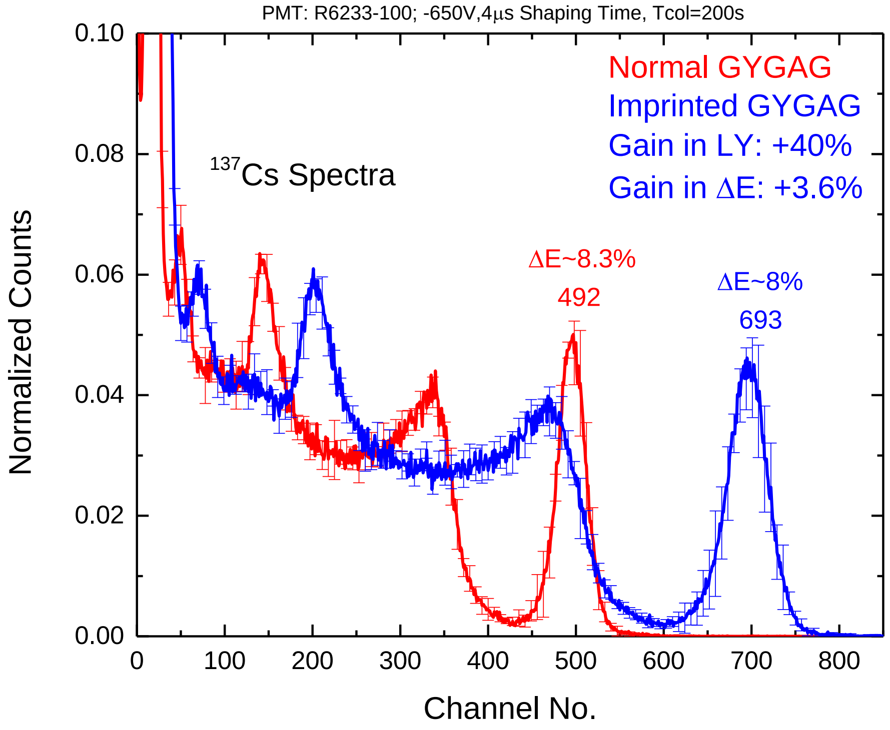
- SEM images of nanoimprinted PhC after removal from scintillator.
- Any layer of polymer between scintillator and PhC increases light absorption (residual polymer layer), and decreases light output.
- **Key**: Develop a process to decrease residual layer.
- Nanoimprinted PhC are quite flexible, and show excellent uniformity (and strength!).



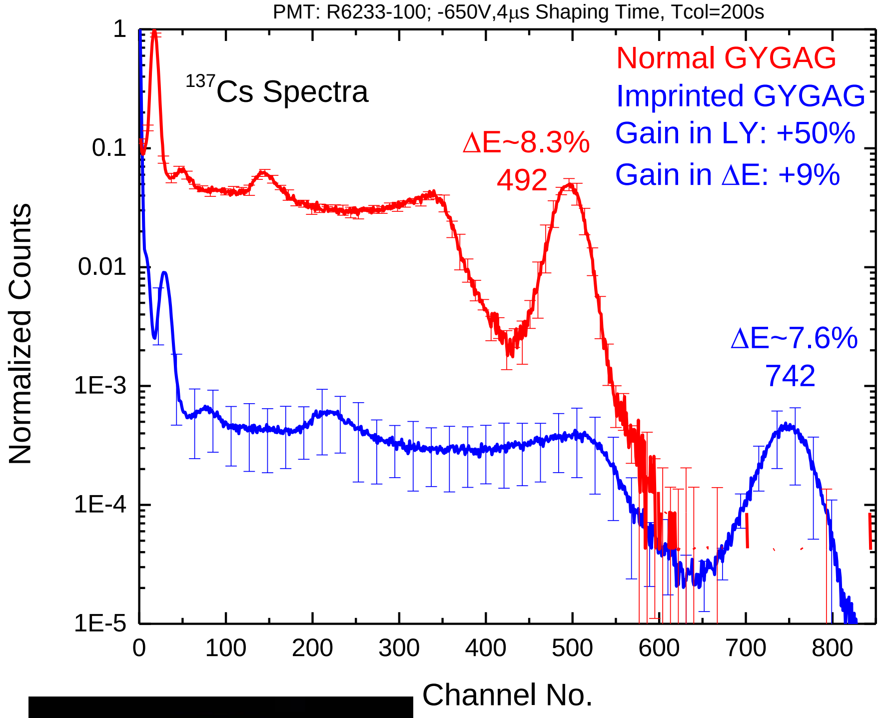


Nanoimprinted GYGAG

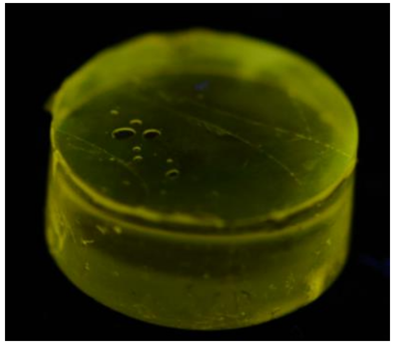
CP2 polymer, RI=1.825



CP5 polymer, RI=1.875

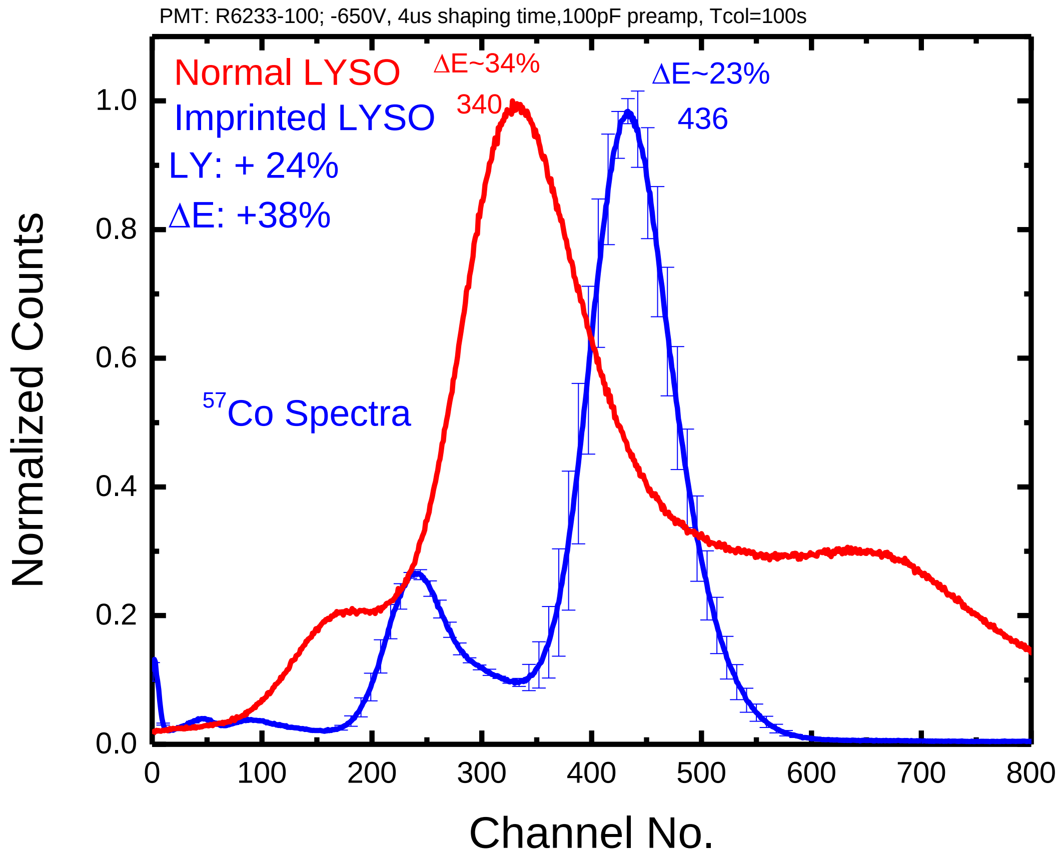


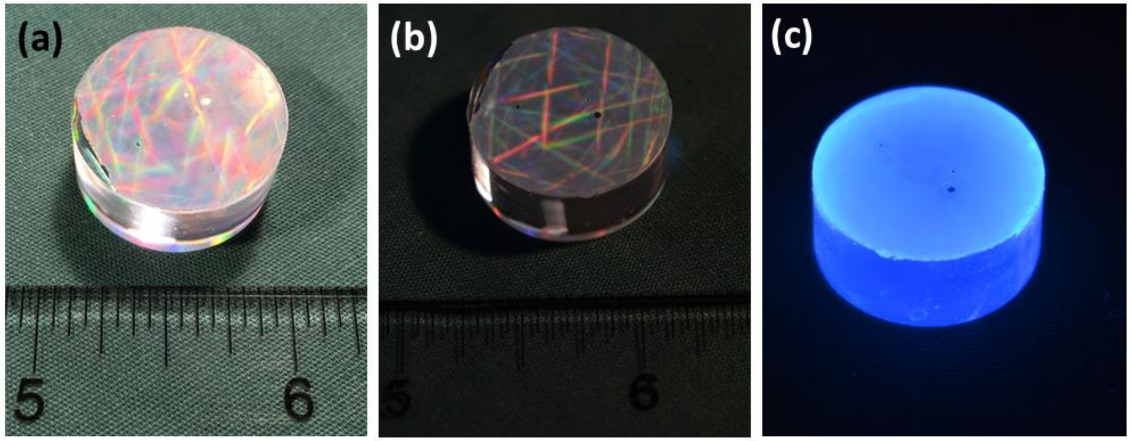
Custom Polymer	RI	Gain in LY	Gain in ER
CP5	1.875	50%	9%
CP2	1.825	40%	3.6%



Non-uniformity during imprinting. This is a process engineering issue, may be leading to degradation of energy resolution.

Nanoimprinting on LYSO

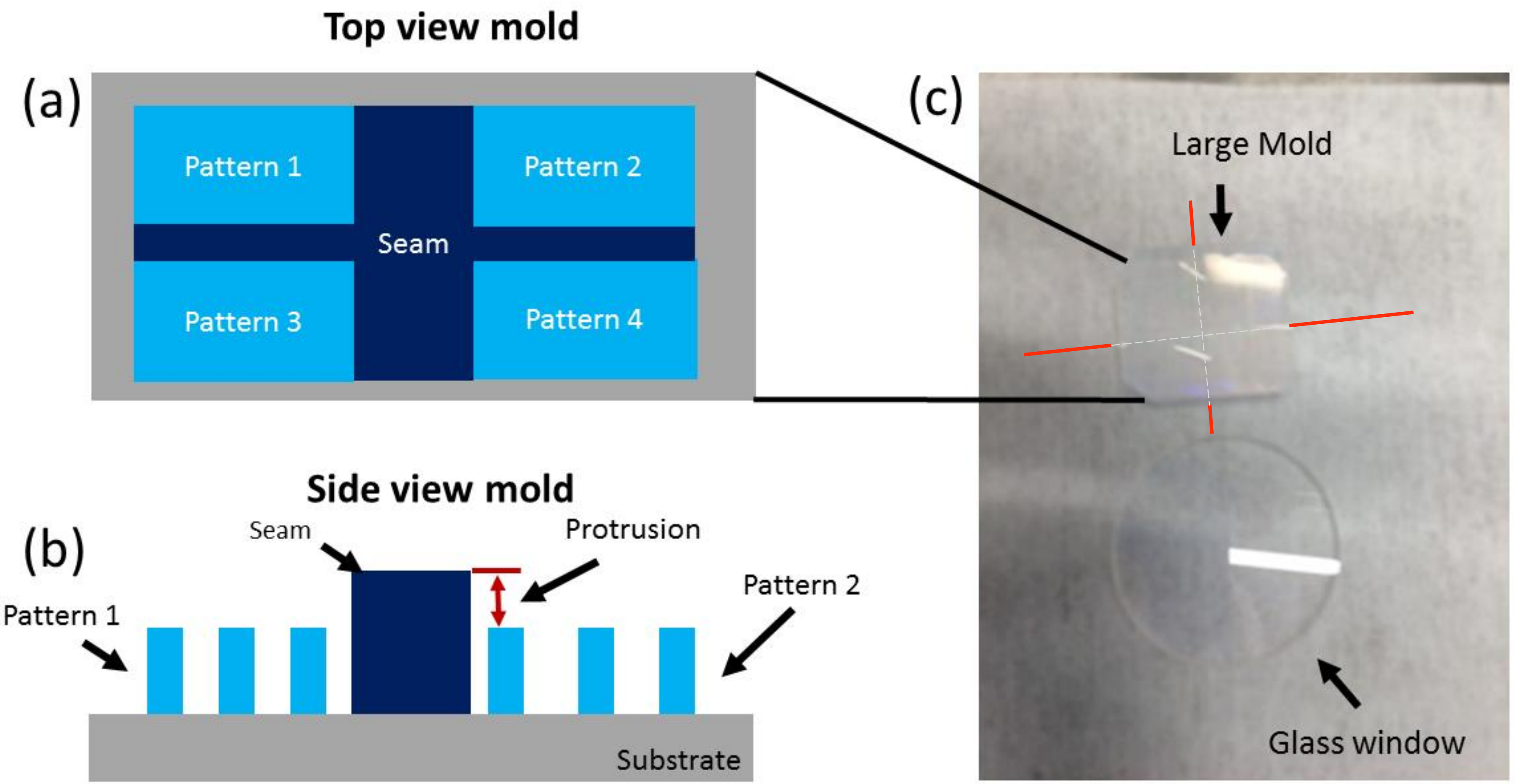




22 mm diameter, 10 mm tall LYSO imprinted with the 1”×1” mold into CP4 (RI=1.78).

- Imprinting in CP4 polymer (RI=1.78).
- 24% gain in light output.
- 38% improvement in ER.

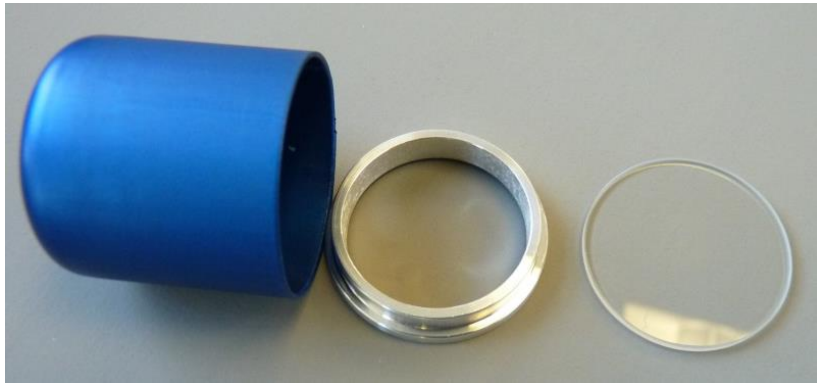
Stamp and Repeat



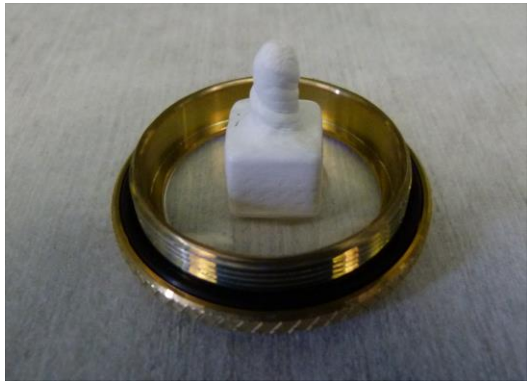
Stamp and repeat is challenging, but doable. Need **larger molds.**

Imprinting large areas, hygroscopic scintillators

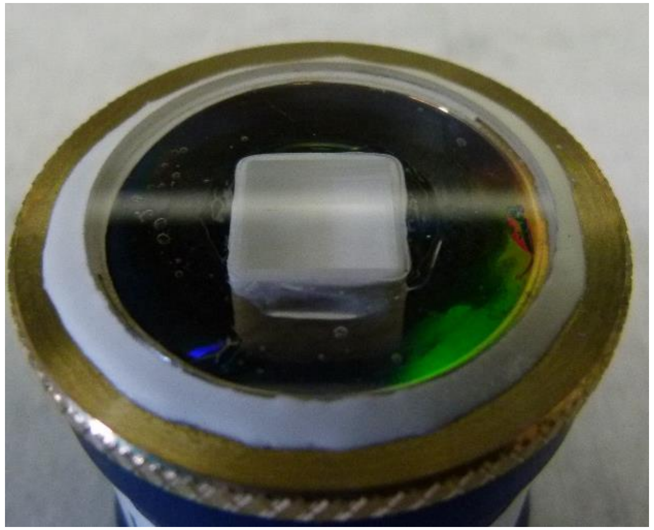
Imprinting on a window facilitates the PhC approach even on difficult to handle ***hygroscopic scintillators***.



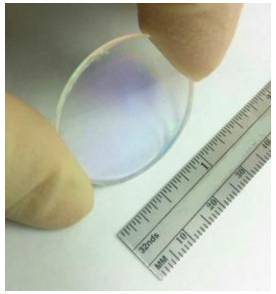
Packaging can for hygroscopic scintillators.



Teflon wrapped SrI_2 scintillator.

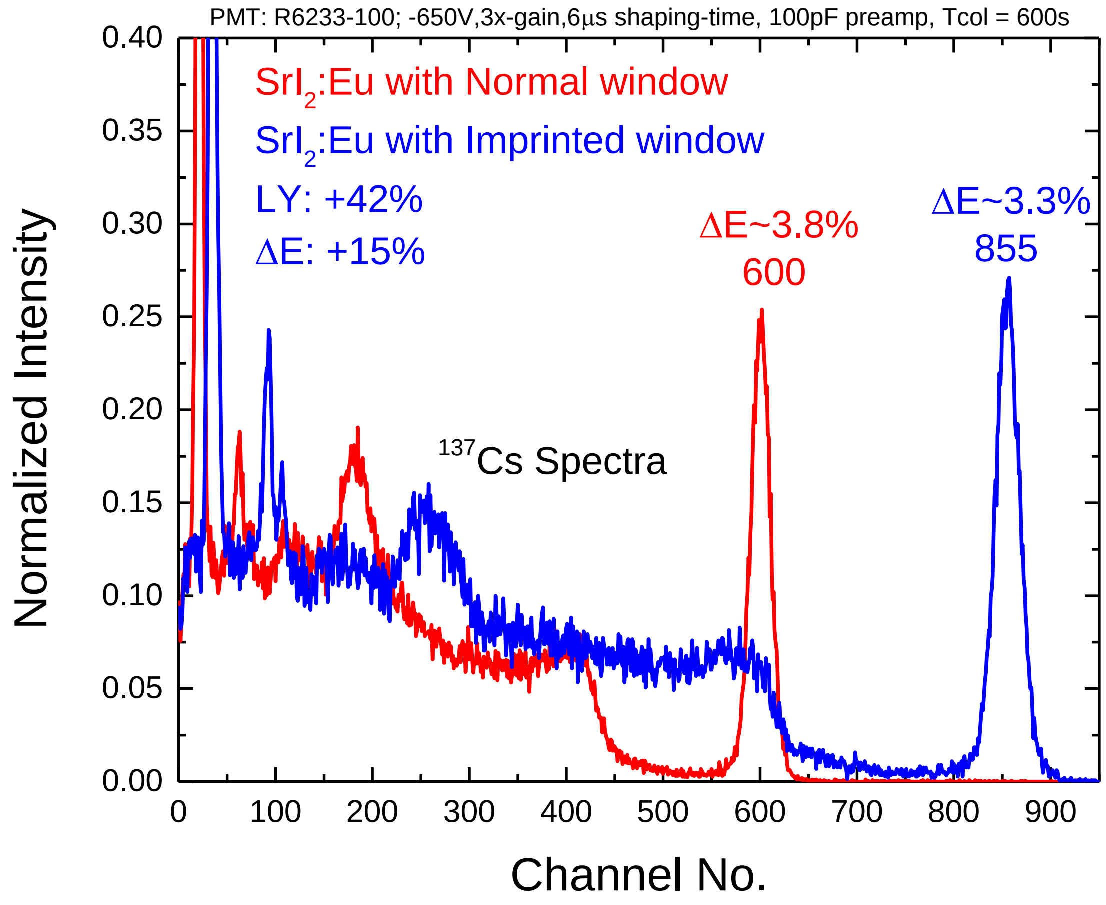


Packaged, Teflon wrapped SrI_2 scintillator with nanoimprinted window.



Nanoimprinted 30 mm diameter quartz window.

Hygroscopic Scintillators: SrI_2 : Eu

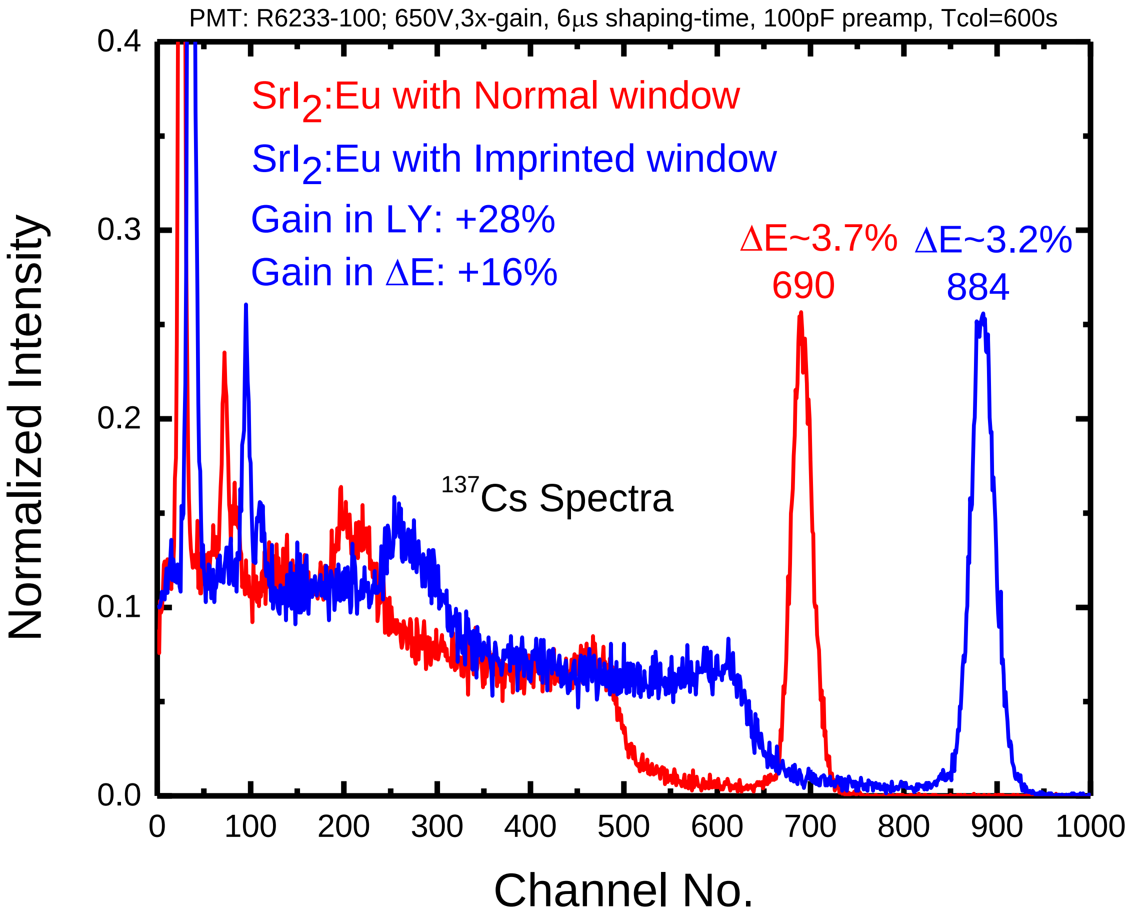


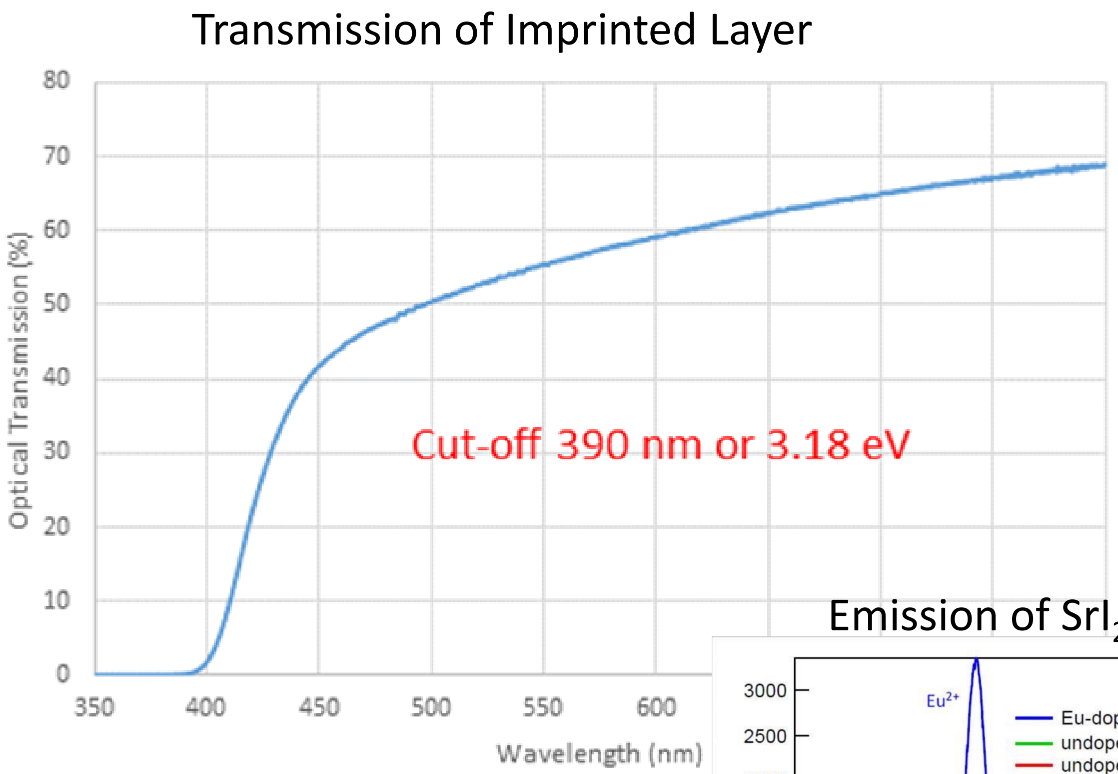
SrI_2:Eu light output & energy resolution measured through:

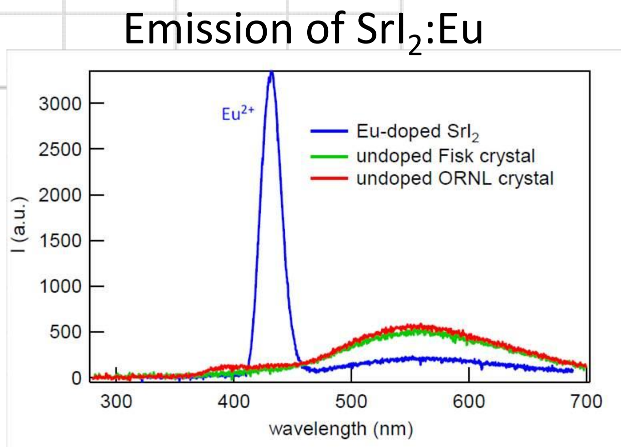
- a regular window;
- a window with PhC imprinted in CP5 polymer (RI=1.88)

- Light Yield gain of +42%
- Energy resolution improvement of +15%

Hygroscopic Scintillator: SrI_2: Eu

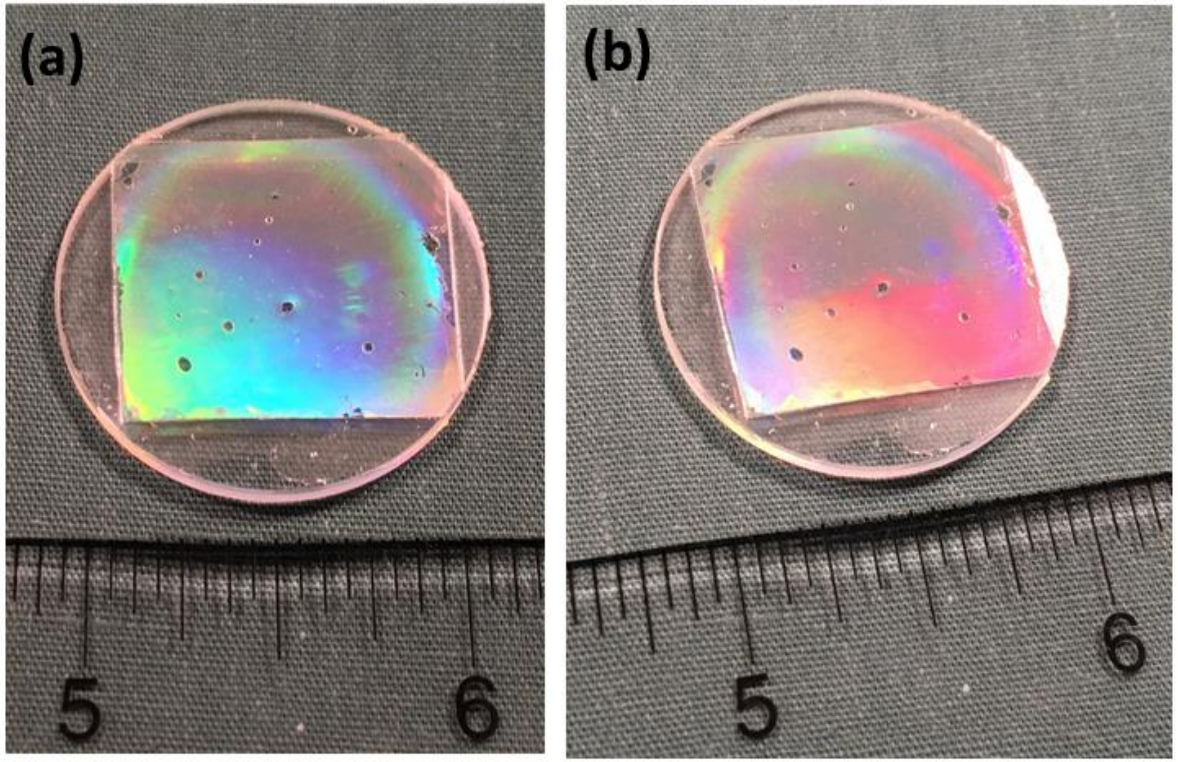




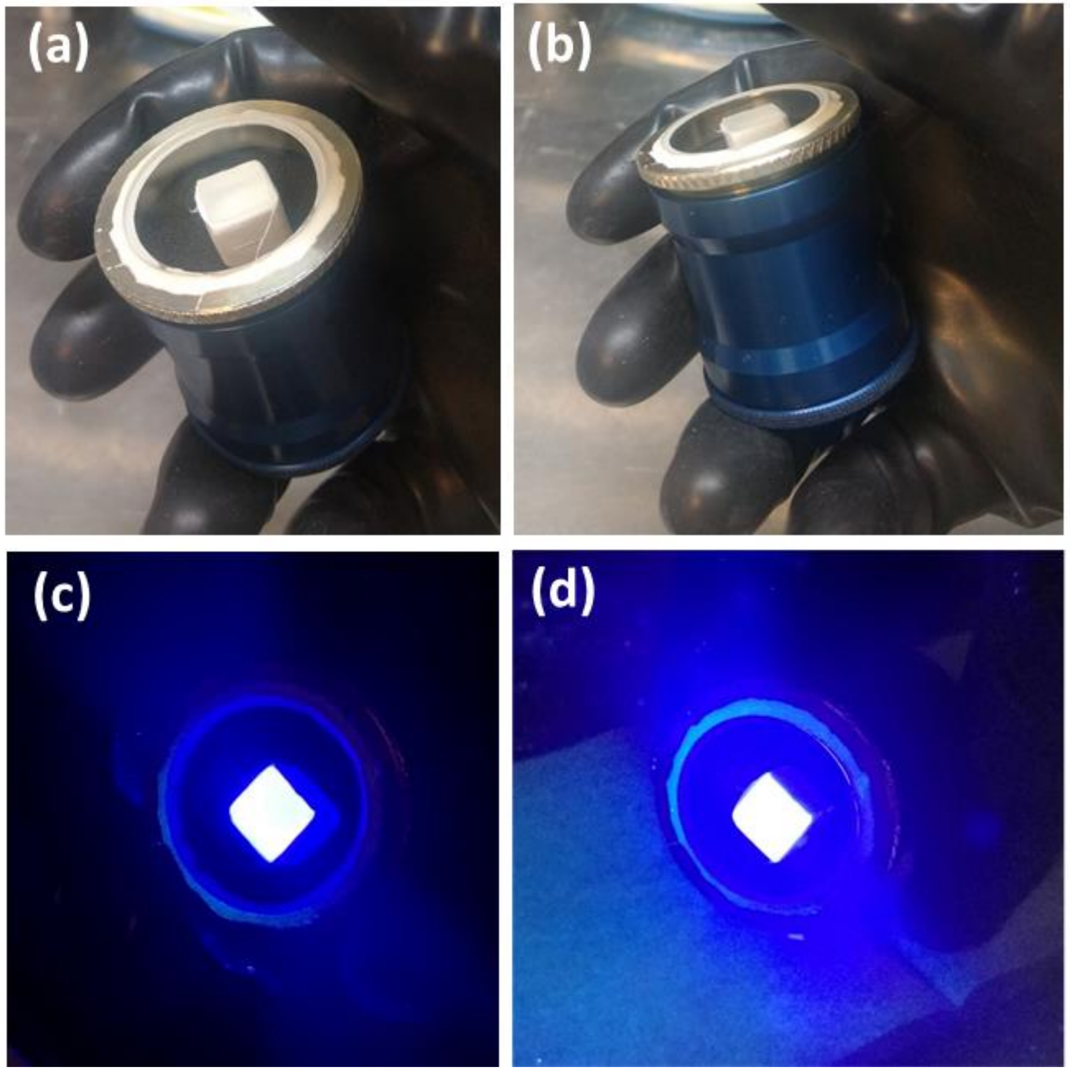


- Light Yield gain of +28%
- Energy resolution improvement of +16%

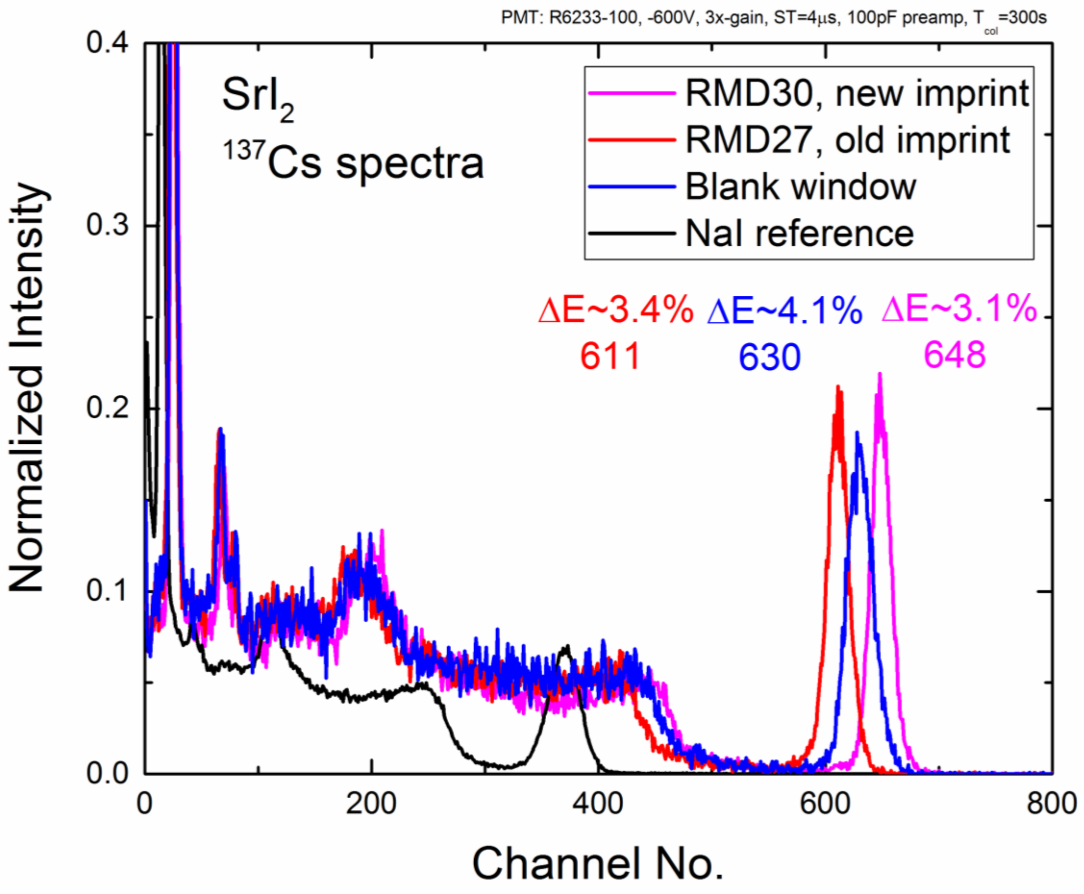
Imprinting using the new 1”×1” Mold



30 mm diameter fused silica optical window imprinted using the new 1 inch mold into CP4 polymer

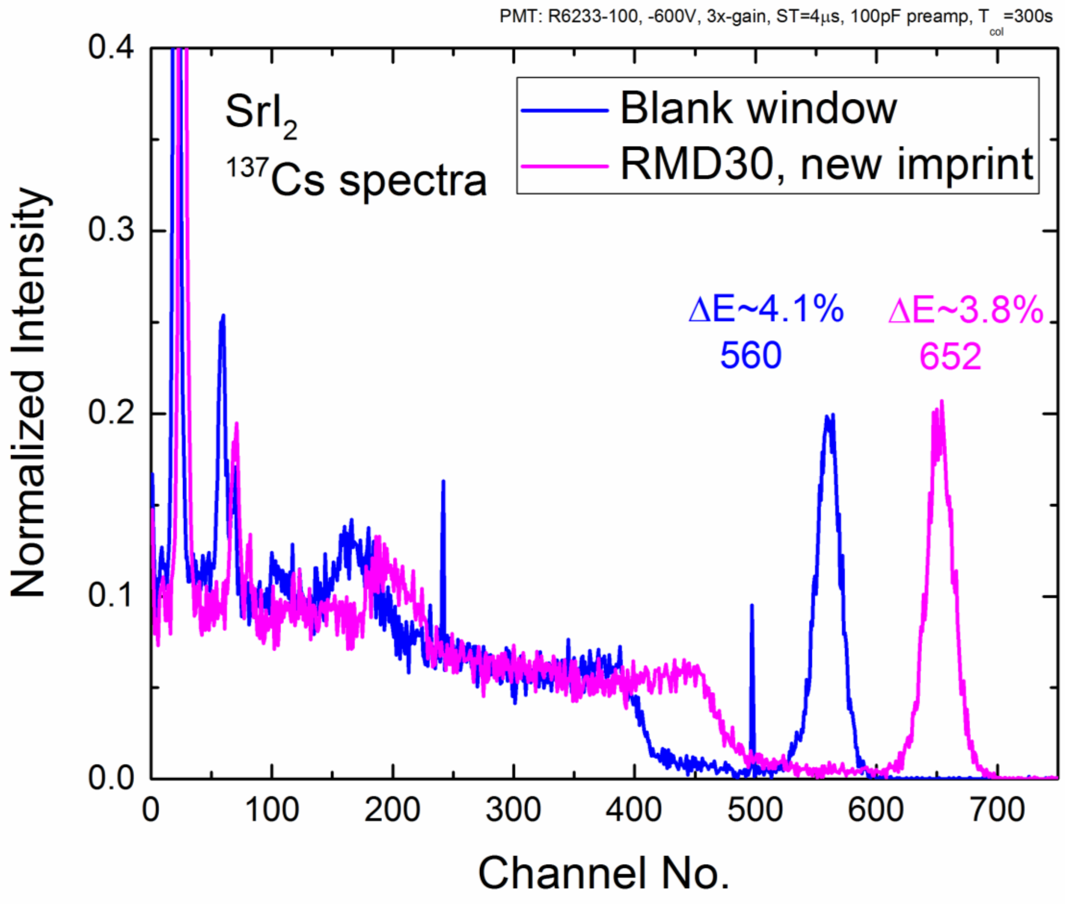


SrI_2 latest results using new mold



ER: ~28% improvement

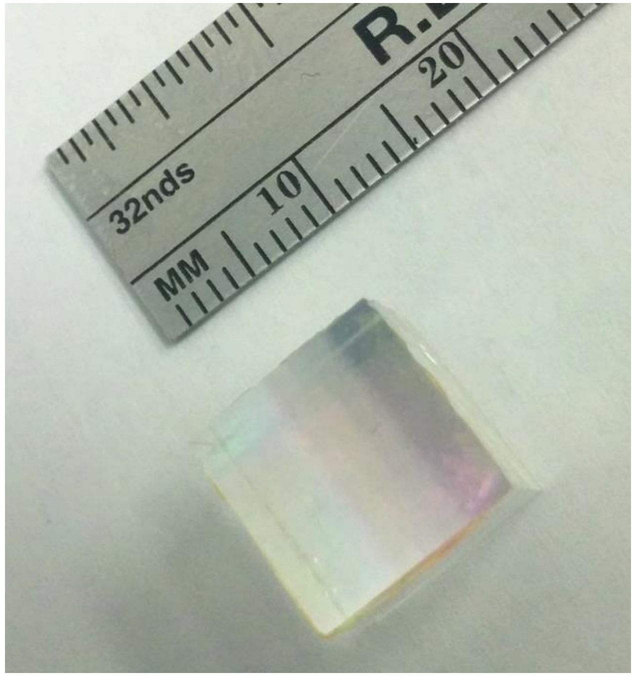
ER: ~19% improvement



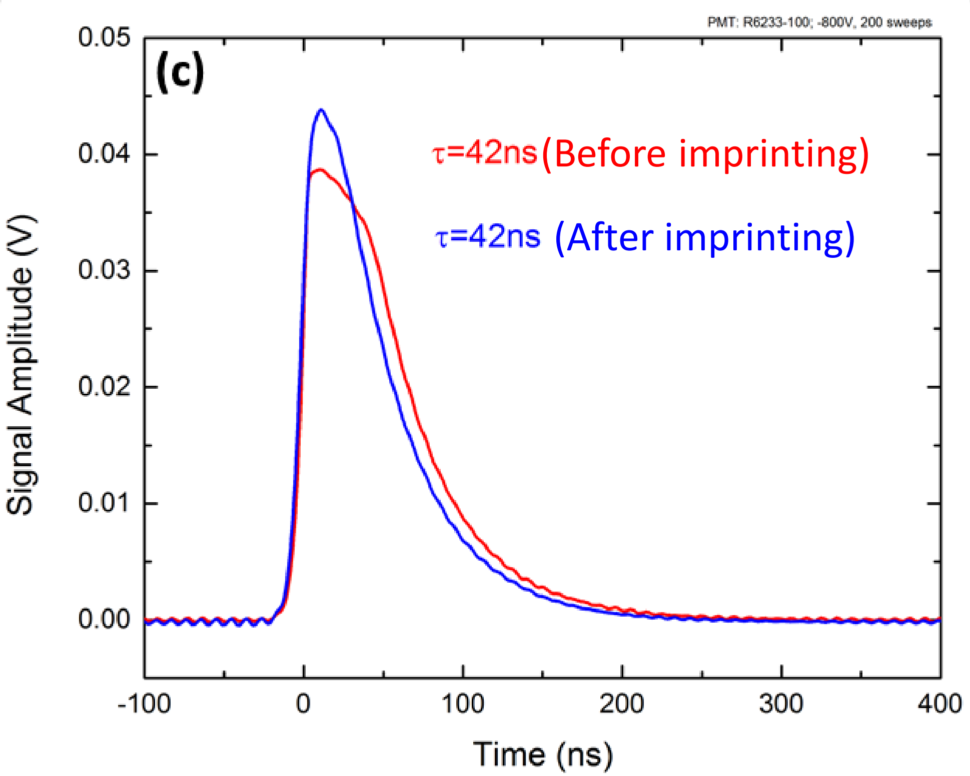
ER: ~8% improvement

LY: ~15% improvement

Timing Resolution Measurement



1 × 1 × 1 cm^3 LYSO scintillator, with the imprinted nanostructured surface displaying optical interference effects.



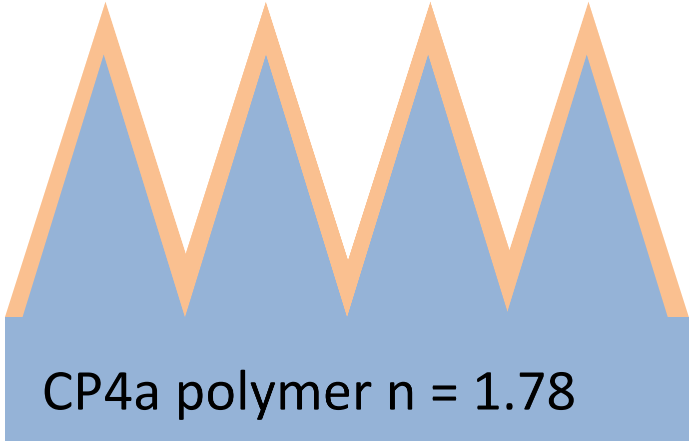
Preliminary timing tests do not show any change in performance. Needs further experimentation.

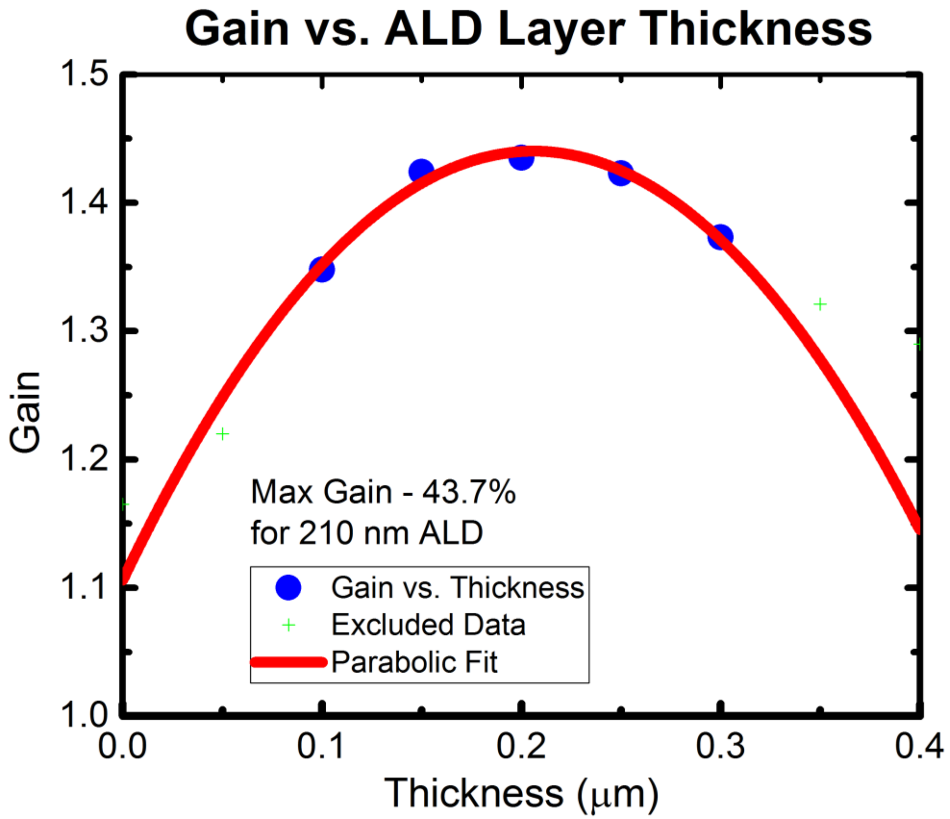
Coincidence timing resolution (*measurements done at U. Penn, courtesy Joel Karp*):

- *Before imprinting*: 333 ps
- *After imprinting*: 335 ps

Increasing the refractive index

Name	Composition	RI (n)
Alumina	Al_2O_3	1.7
Hafnia	HfO_2	2.2
Titania	TiO_2	2.6

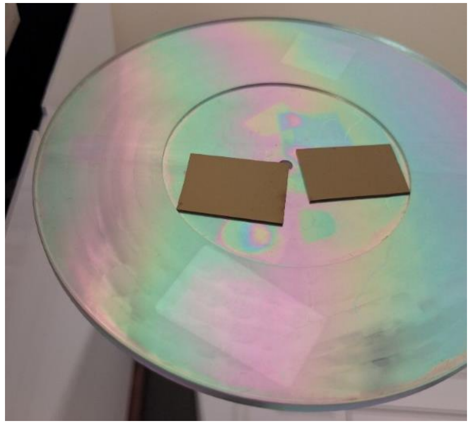




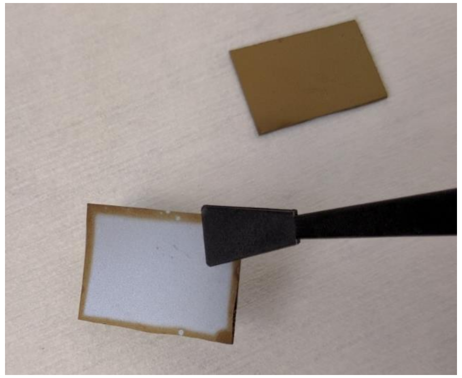
- Simulations show the gain as a function of layer thickness for ALD-grown thin films of high-RI TiO_2.
- Potential gains of 44% for a 210 nm thick film

ALD Thin Films

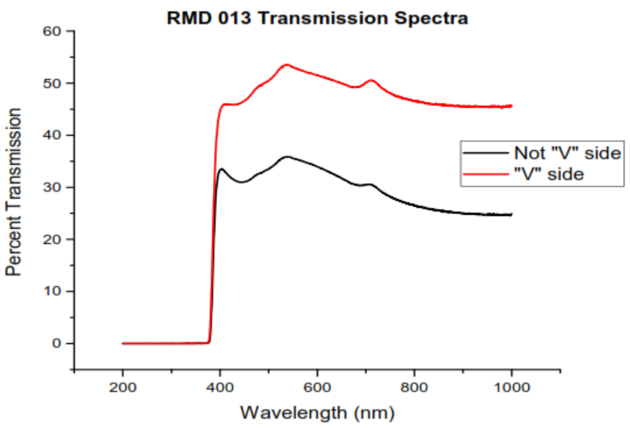
TiO_2 RI=2.46, film thickness 34 nm



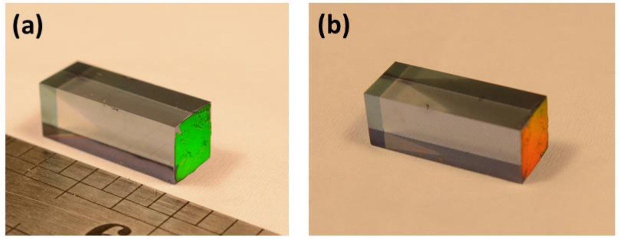
TiO_2 on the platter of the growth chamber

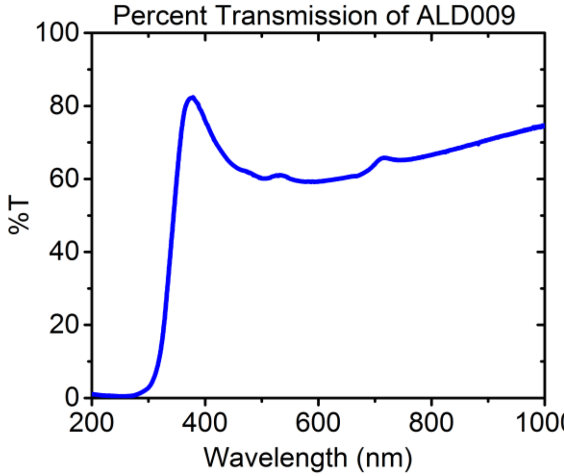


Silicon sample after removing from chamber

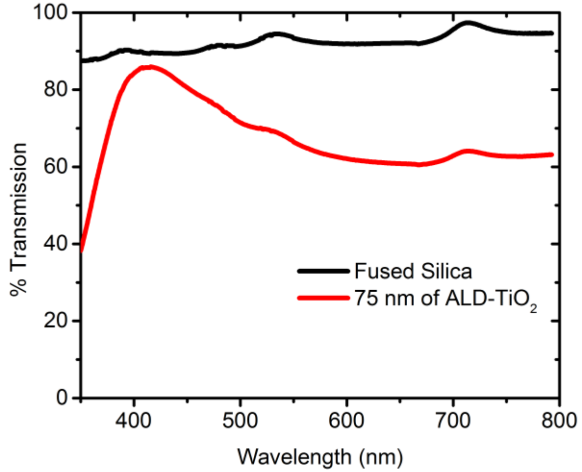


50 nm thick ALD TiO_2 on LYSO





50 nm thick ALD TiO_2 on fused silica



75 nm thick ALD TiO_2 on fused silica

RMD
A Dynasil Company

Summary

- Demonstrated large-area nanoimprinting of photonic crystals.
  - Overcome cost and size limitations of e-beam fabrication.
- PhCs demonstrated 40%-50% improvements in light yield and energy resolution even in highly hygroscopic crystals like SrI_2.
- PhCs enable current scintillators to perform in the realm of new materials → cost savings!

What is planned?

- Demonstrate improvements in timing resolution.
- *Polymer development*: Lower cost, higher refractive index, lower cut-off wavelength.
- *Roll-to-roll process*: Fabricate master template on a roller.
- Implement in commercial instruments for customer evaluations.