Harnessing the Purcell Effect for Faster Metascintillators

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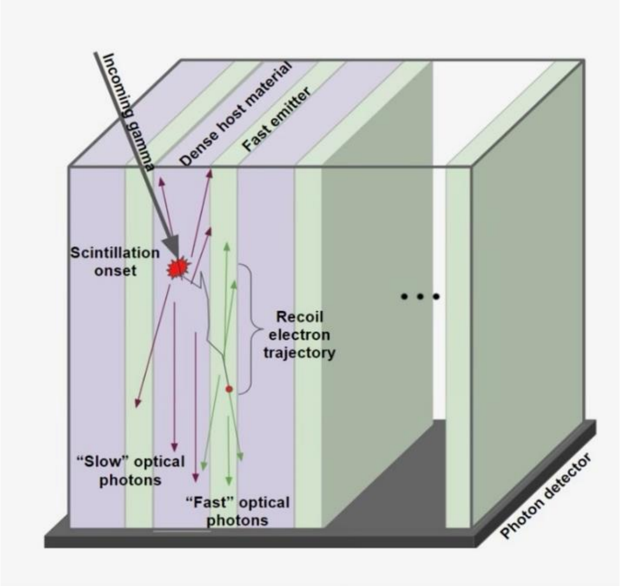
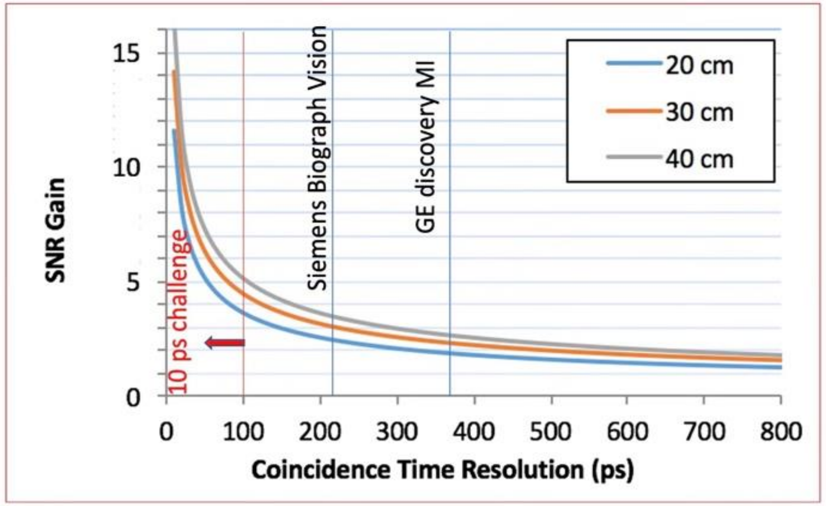


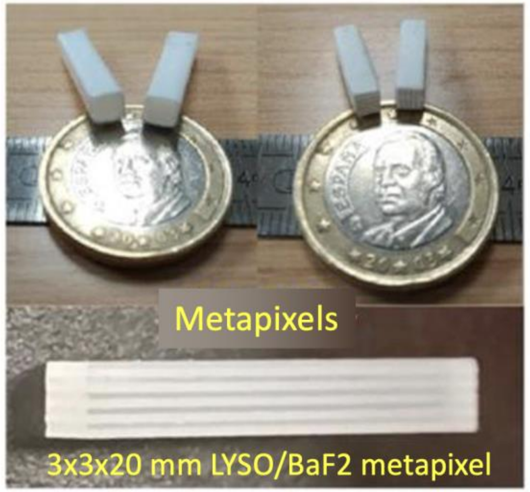
Shultzman et al., Towards a second generation of metascintillators using the Purcell effect, submitted TRPMS (2024)

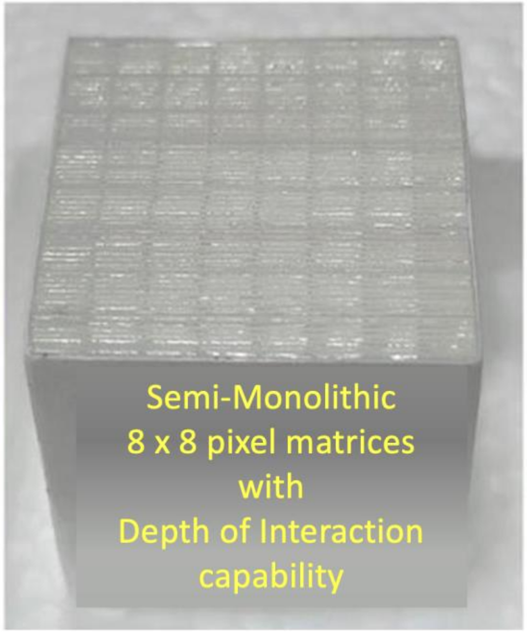
The 10 ps challenge

$$SNR_{TOF} = \sqrt{(2D/c\Delta t)} \cdot SNR_{conv}$$



- Requirements
  - Faster emission
  - Higher yield
  - Emission directionality





BGO/BaF2 & BGO/EJ232
200 ps CTR

LYSO/BaF2 & LYSO/EJ232
100 ps CTR

Scintillator engineering to boost the light emission yield and rate

- Crystal engineering limited so far to controling or compensating crystallographic defects to limit afterglow and improve radiation resistance
- Can we engineer the oscillator strength?
- Decay time is driven by the electric dipole moment between the excited and fundamental states

$$\Gamma_\nu = \frac{1}{\tau_\nu} \propto \frac{n}{\lambda^3}\left(\frac{n^2+2}{3}\right)^2 \sum_f \left| \langle f | \quad | i \rangle \right|^2$$

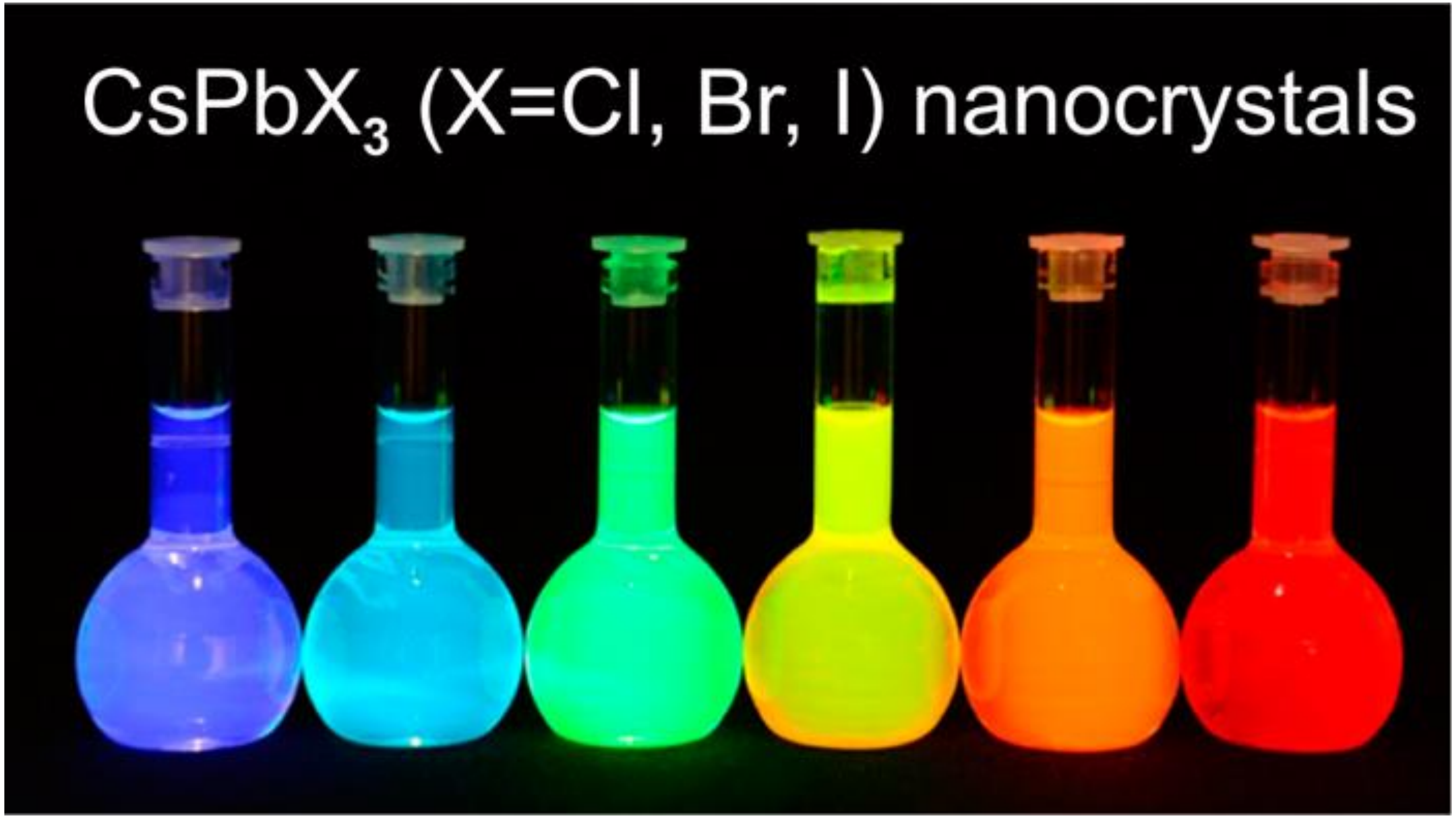
Local polarization field
Related to ion coordination
and local symetry level

Electric dipole moment operator

- Exciton confinement in quantum wells allows scintillation wavelength engineering aand leads to efficient and sub-ns luminescence
- Properly designed quantum dot-based heterostructures produce directional coherent phasing of dipoles over many unit cells and achieve Giant Oscillator Strength (GOS)

Wavelength engineering through quantum confinement

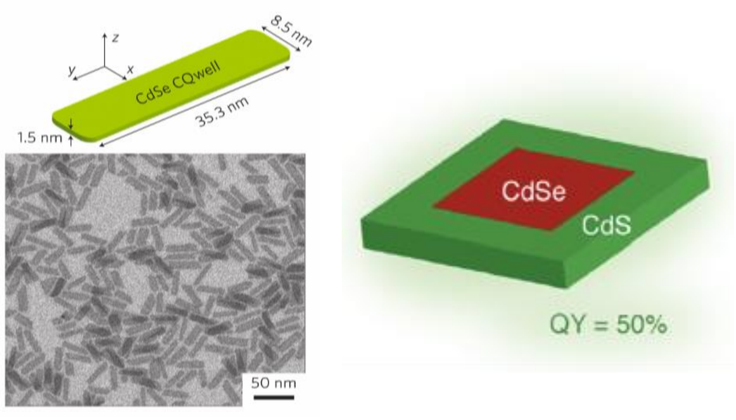
Confinement level



Protesescu et al., 2015

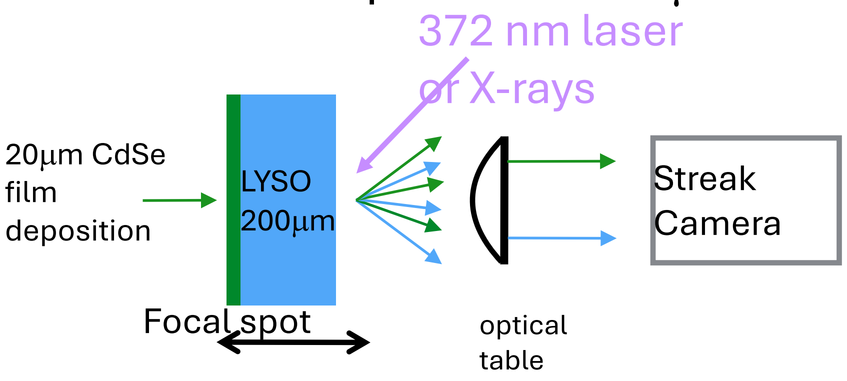
CdSe nanoplatelets on LYSO plates

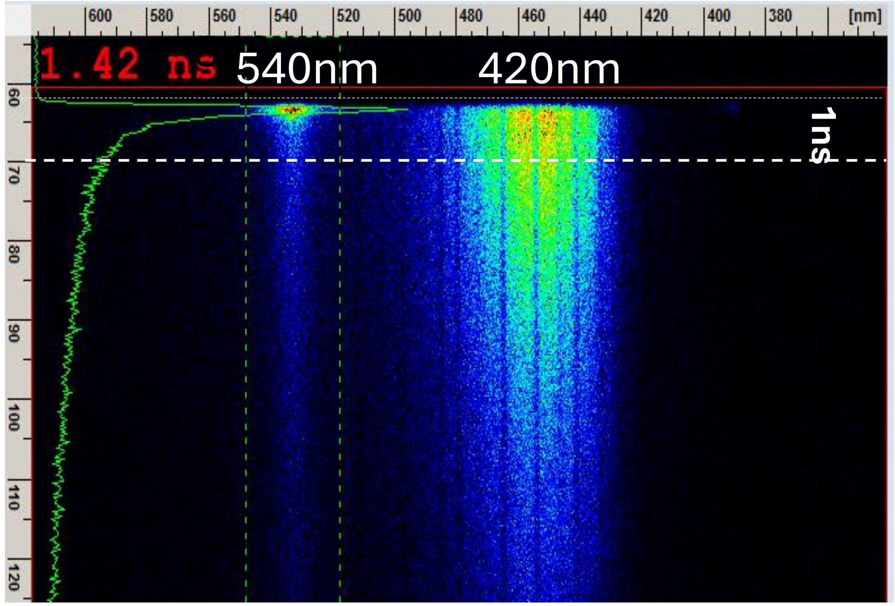
J.Grim, I. Moreels
ITT, Italy



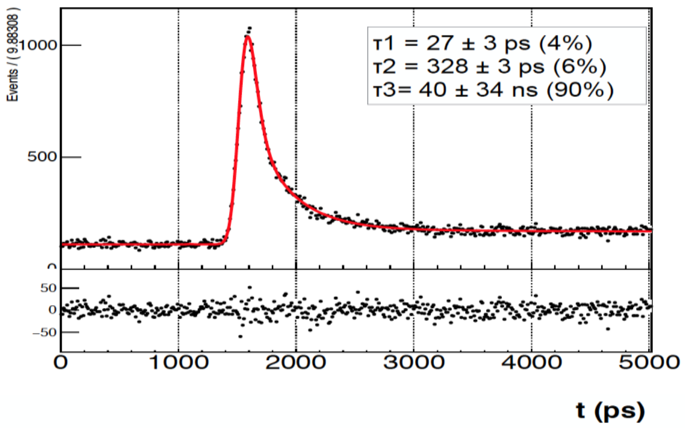
LYSO plate 200μm thick

+ CdSe/CdS nanoplate film 20μm thick



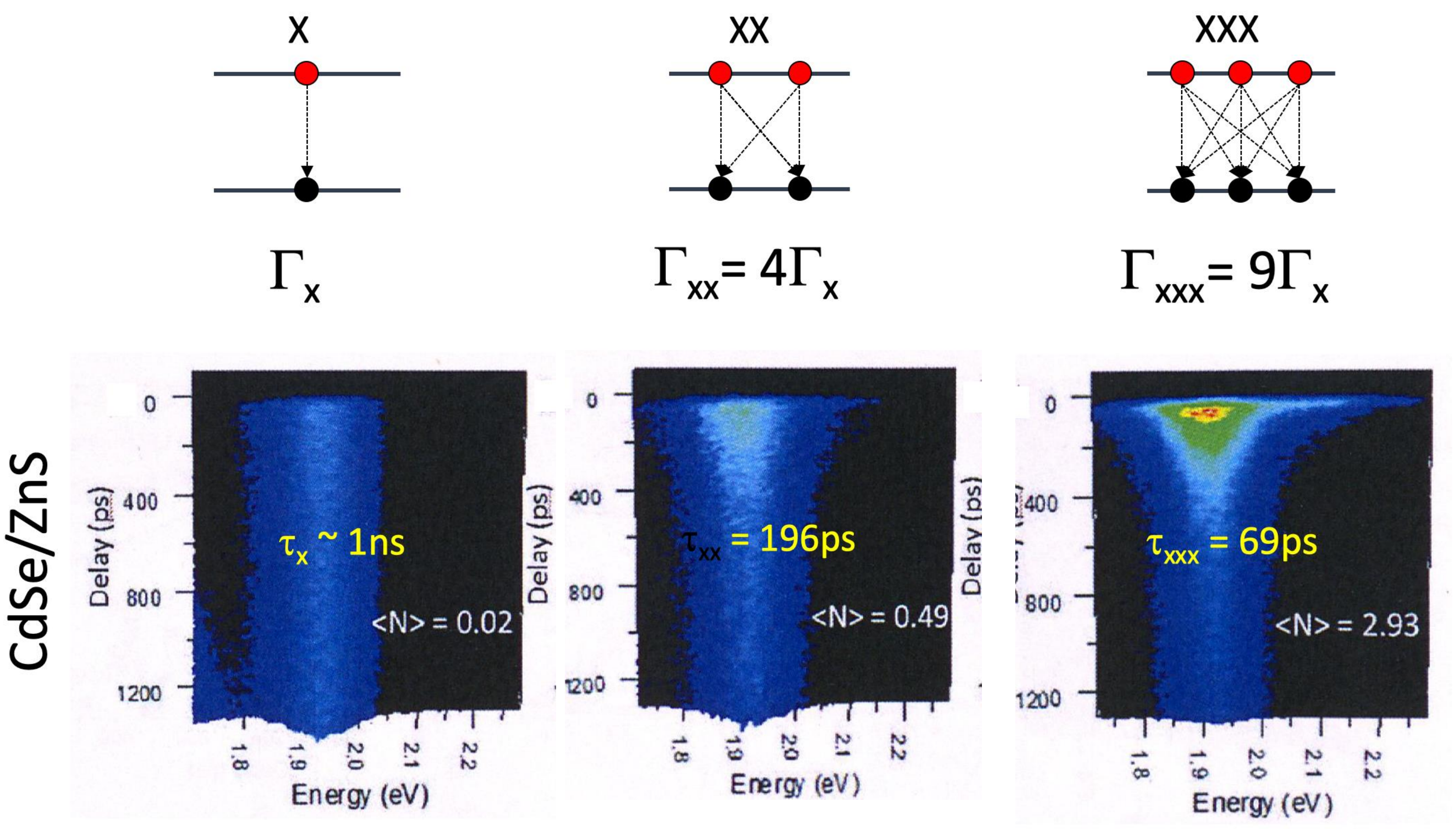


RL decay profile @530nm



R. Martinez Turtos et al., JINST_068P_06

Multi-exciton quantum confinement



ZnO:Ga QDs

Metal halide Perovkites

CdSe/CdS Nanoplatelets

InGaN/GaN Quantum wells

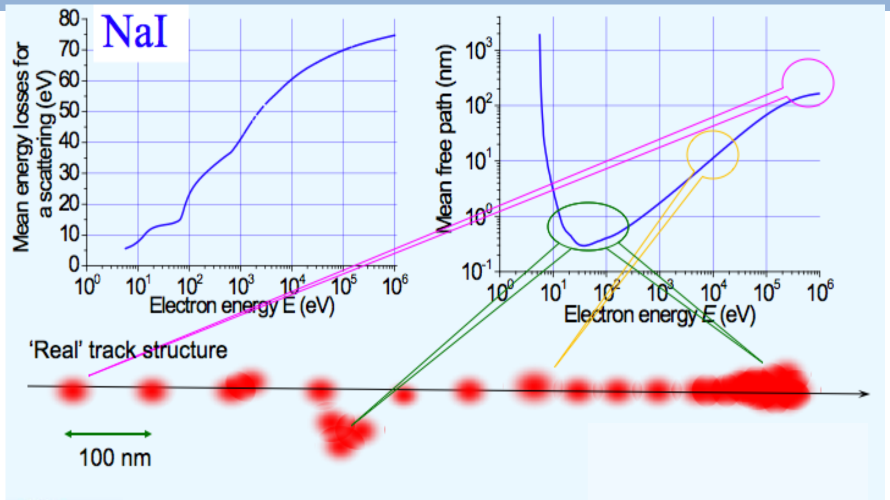
Padilha et.al., Nano Lett. 2013, 925-932

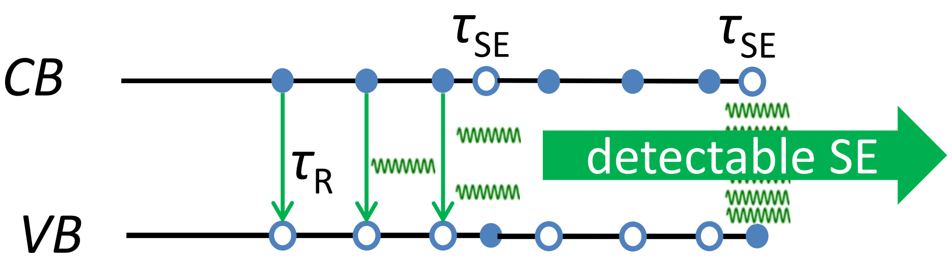
Towards a self-triggered lasing γ-ray detector

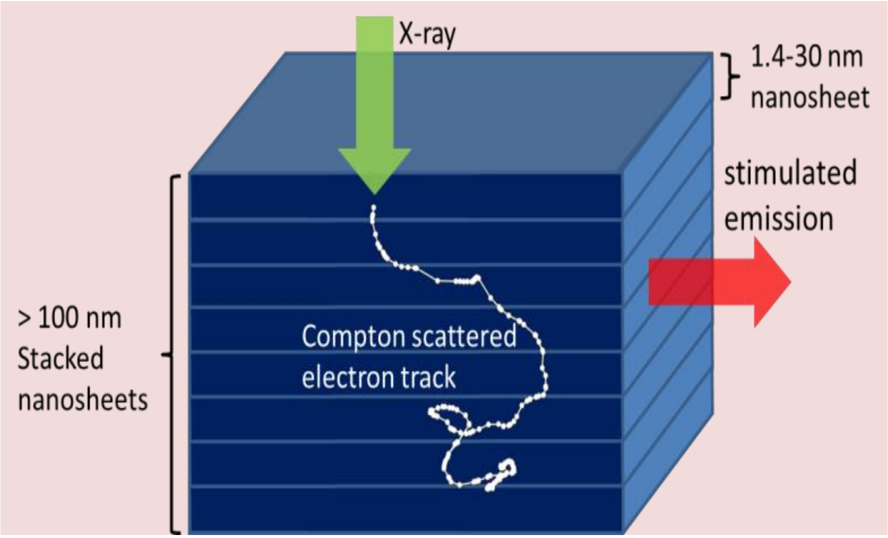
- Ionization density per dE/dx event can reach

  $> 10^{20}$ eV/ cm^3 $\approx$ 10J/cm^3

- Ultrafast X-ray or γ-ray self-triggered stimulated emission is within reach





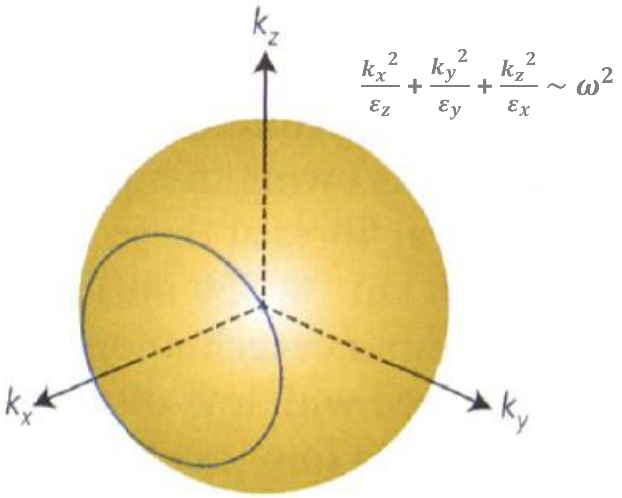


Courtesy J.Grim, I.Moreels, ITT, Italy

Increasing the Photonic Density of States with Hyperbolic Metamaterials

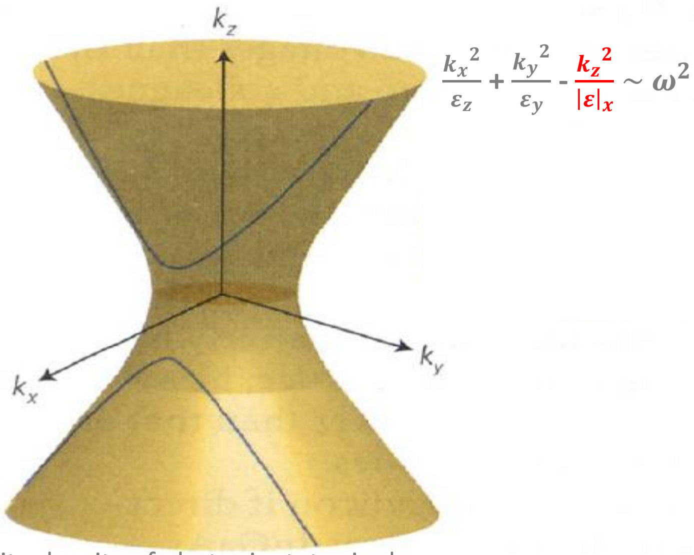
Isofrequency contour from dispersion formula: $k = \frac{2\pi}{\lambda} \sim \omega\frac{n}{c} \sim \omega\sqrt{\epsilon}$

for isotropic dielectric material

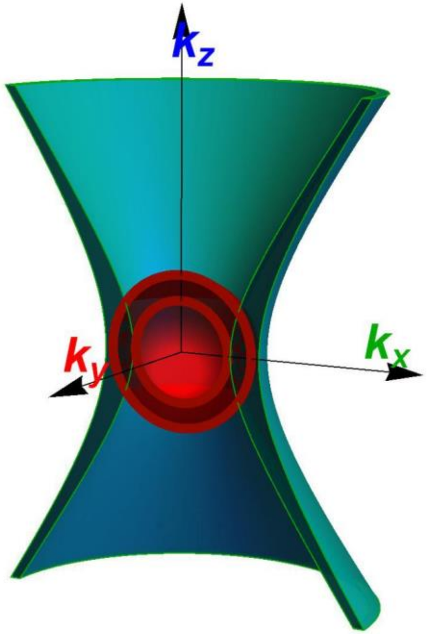


Finite density of photonic states in dω

for anisotropic hyperbolic material



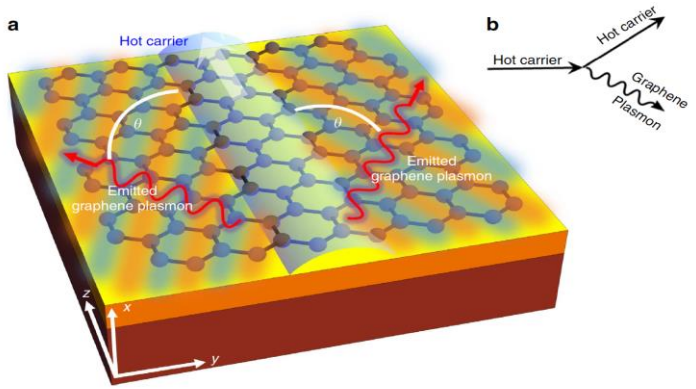
Infinite density of photonic states in dω



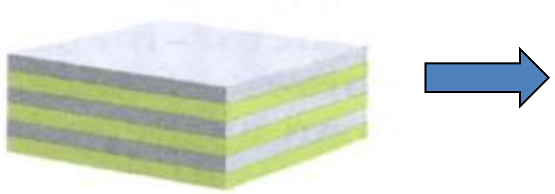
An intuitive counting procedure in k-space consists of calculating the volume between the isofrequency contours at ω(k) and ω(k) + Δω.

Lowering Čerenkov threshold ?

- The Čerenkov effect offers the fastest energy conversion scheme from charge particles to photons
- Čerenkov threshold (101 keV in LSO) can be strongly lowered (↘100’s eV) in specifically designed nanostructured metamaterials
- Produce transition radiation constructive interference through resonant plasmonic states in photonic crystals
- Related more formally to the Smith Purcell effect
  - A kind of Cerenkov or transition radiation emission → free electron lasers
  - $\lambda = \frac{a}{m}\left(\frac{1}{\beta} - cos\theta\right)$!! No velocity threshold !!



I. Kaminer et al., Nature Comm., 13 June 2016



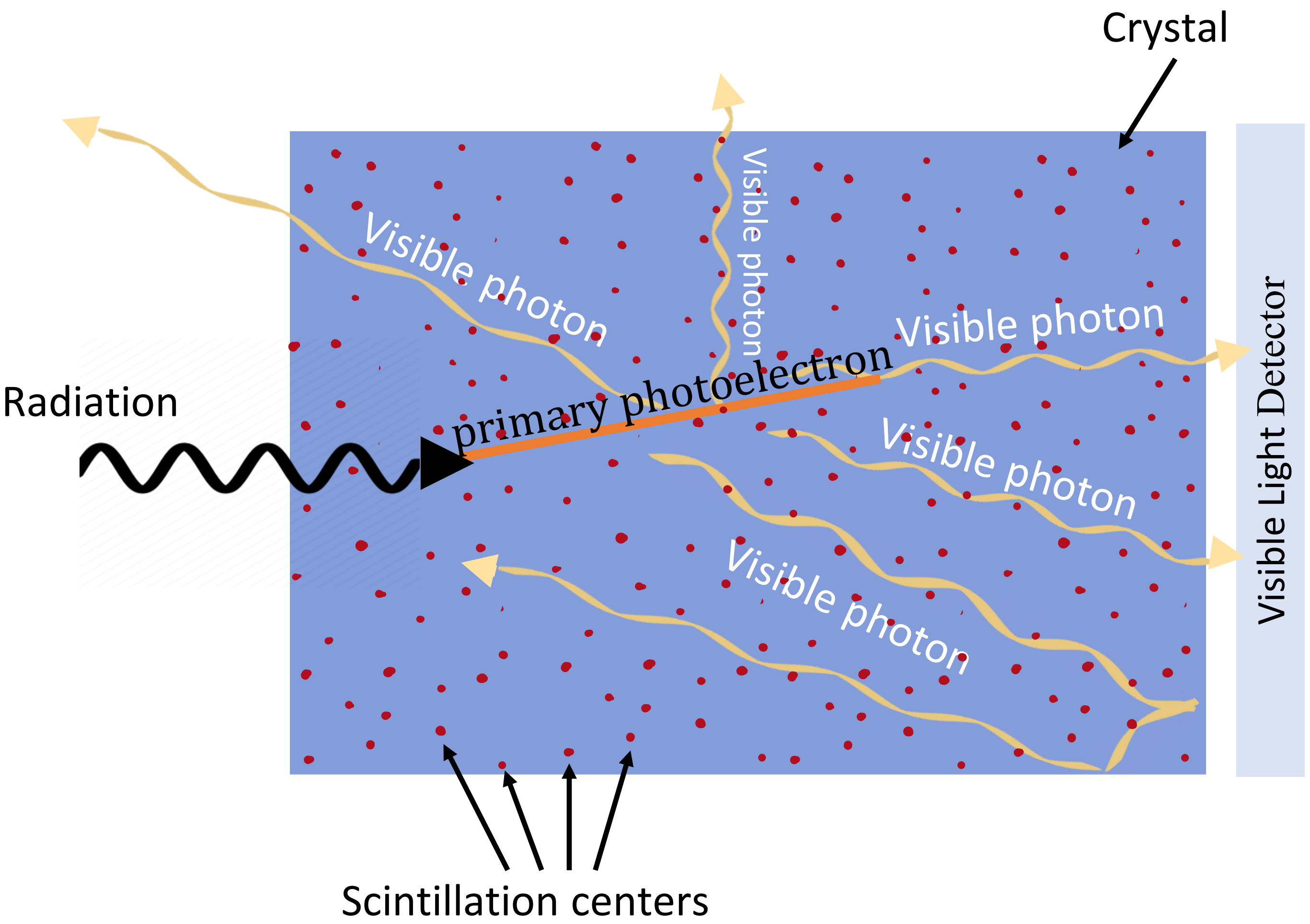
D. Lu et al., Nat. Photon. 11, 2017

Stack of Gold + SiO_2

Cerenkov threshold ↘ 250eV

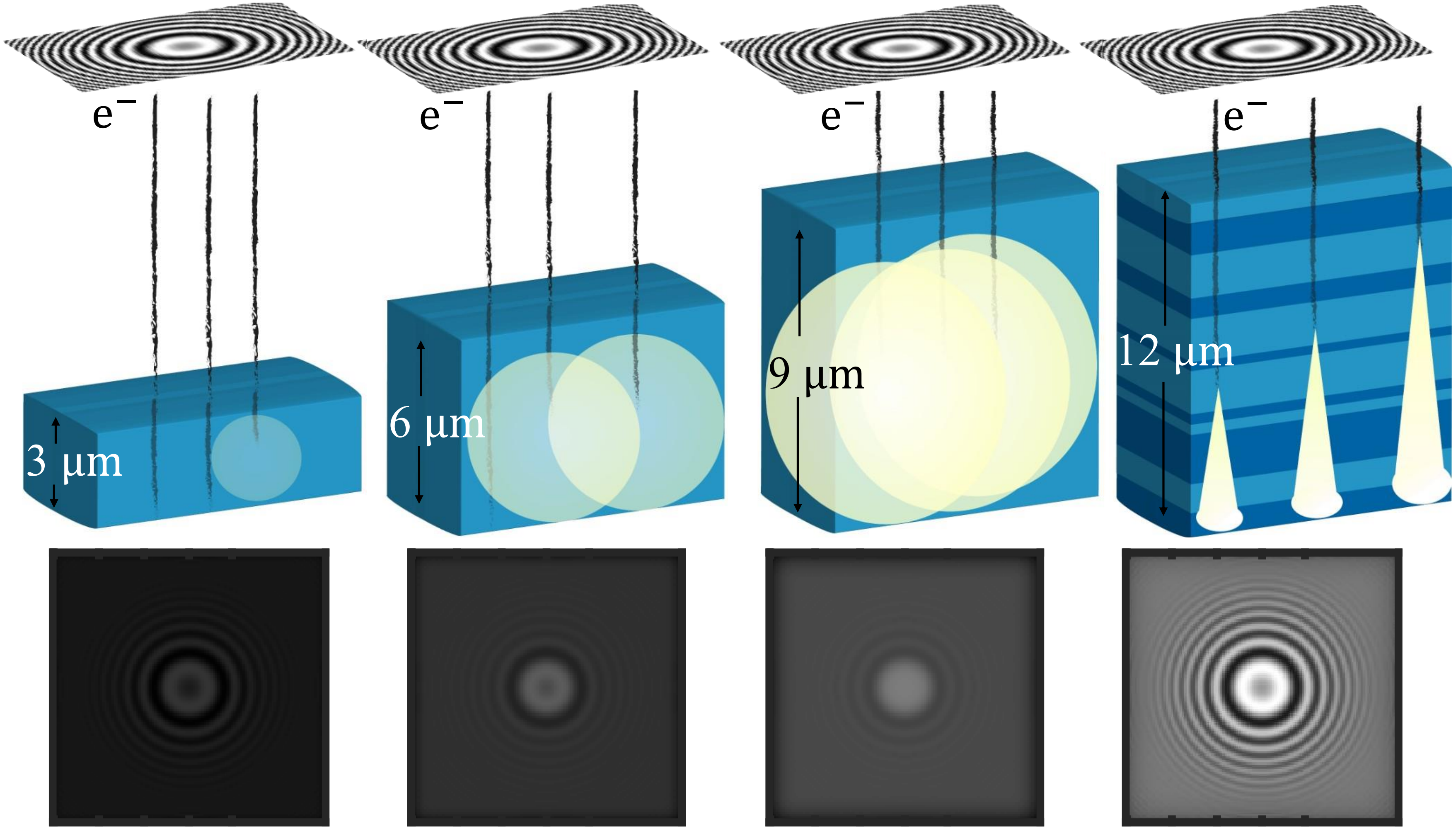
Cerenkov yield ↗ > 100

Mechanism of Scintillation based Detection



Efficiency vs Spatial resolution challenge

Optimization of a Multilayer Nanostructure
to Enhance Imaging Characteristics (PSF)



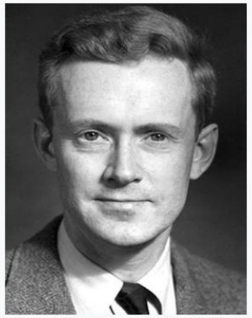
Nanophotonic Purcell scintillators

- Purcell effect: Spontaneous emission rate depends on its EM surrounding

$$\mathrm{F_p} = \Gamma/\Gamma_0 \cong \rho/\rho_0 \propto \left|\vec{E}\right|^2$$

- For a dipole in a cavity - $F_\mathrm{P} \propto \frac{Q}{V}$

- Exploit in an untraditional way



Edward Mills Purcell (1912–1997)

B10. Spontaneous Emission Probabilities at Radio Frequencies. E. M. PURCELL, *Harvard University*.—For nuclear magnetic moment transitions at radio frequencies the probability of spontaneous emission, computed from

$$A_\nu = (8\pi\nu^2/c^3)h\nu(8\pi^3\mu^2/3h^2)\ \mathrm{sec.}^{-1},$$

is so small that this process is not effective in bringing a spin system into thermal equilibrium with its surroundings. At 300°K, for $\nu = 10^7$ sec.$^{-1}$, $\mu = 1$ nuclear magneton, the corresponding relaxation time would be 5×10^{21} seconds! However, for a system coupled to a resonant electrical circuit, the factor $8\pi\nu^2/c^3$ no longer gives correctly the number of radiation oscillators per unit volume, in unit frequency range, there being now *one* oscillator in the frequency range ν/Q associated with the circuit. The spontaneous emission probability is thereby increased, and the relaxation time reduced, by a factor $f = 3Q\lambda^3/4\pi^2V$, where V is the volume of the resonator. If a is a dimension characteristic of the circuit so that $V\sim a^3$, and if δ is the skin-depth at frequency ν, $f\sim\lambda^3/a^2\delta$. For a non-resonant circuit $f\sim\lambda^3/a^3$, and for $a<\delta$ it can be shown that $f\sim\lambda^3/a\delta^2$. If small metallic particles, of diameter 10^{-3} cm are mixed with a nuclear-magnetic medium at room temperature, spontaneous emission should establish thermal equilibrium in a time of the order of minutes, for $\nu = 10^7$ sec.$^{-1}$.

E.M. Purcell, Phys. Rev. (1946)

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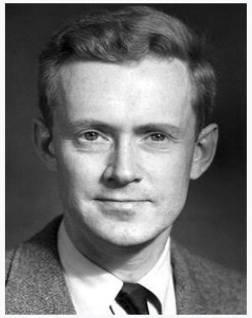

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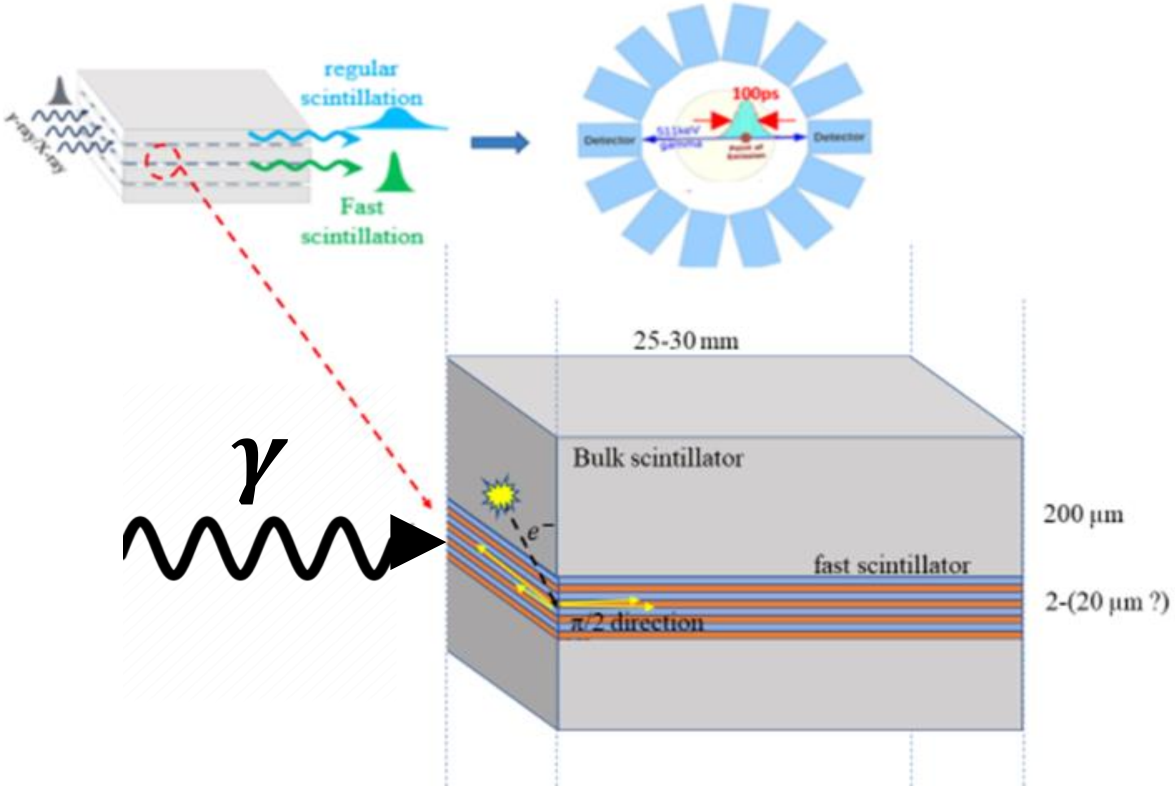
Surpassing the 100 ps CTR barrier

- Metascintillator + Nanophotonics
- Replacing the fast layers with nanophotonic Purcell scintillators
  - CsPbBr3 perovskite nanocrystals
  - Silica
- Enhance the emission parallel to the layers



Surpassing the 100 ps CTR barrier

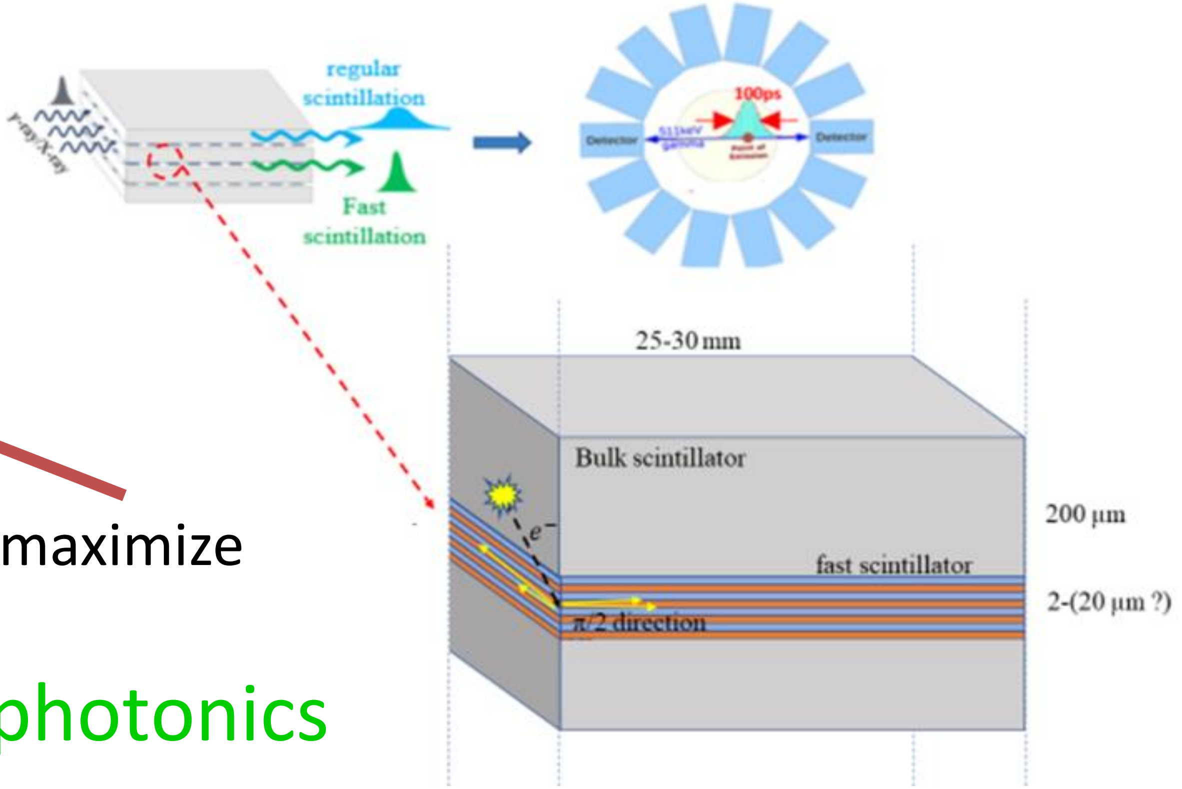
- Metascintillator + Nanophotonics

- CTR enhancement

$$\underline{CTR} \propto \sqrt{\frac{\tau_r \tau_d}{\eta}} = \sqrt{\frac{\tau_r}{\eta\Gamma}}$$

minimize

maximize



- Rise time: τ_r
- Decay time : $\tau_d = 1/\Gamma$ nanophotonics
- Efficiency: η nanophotonics

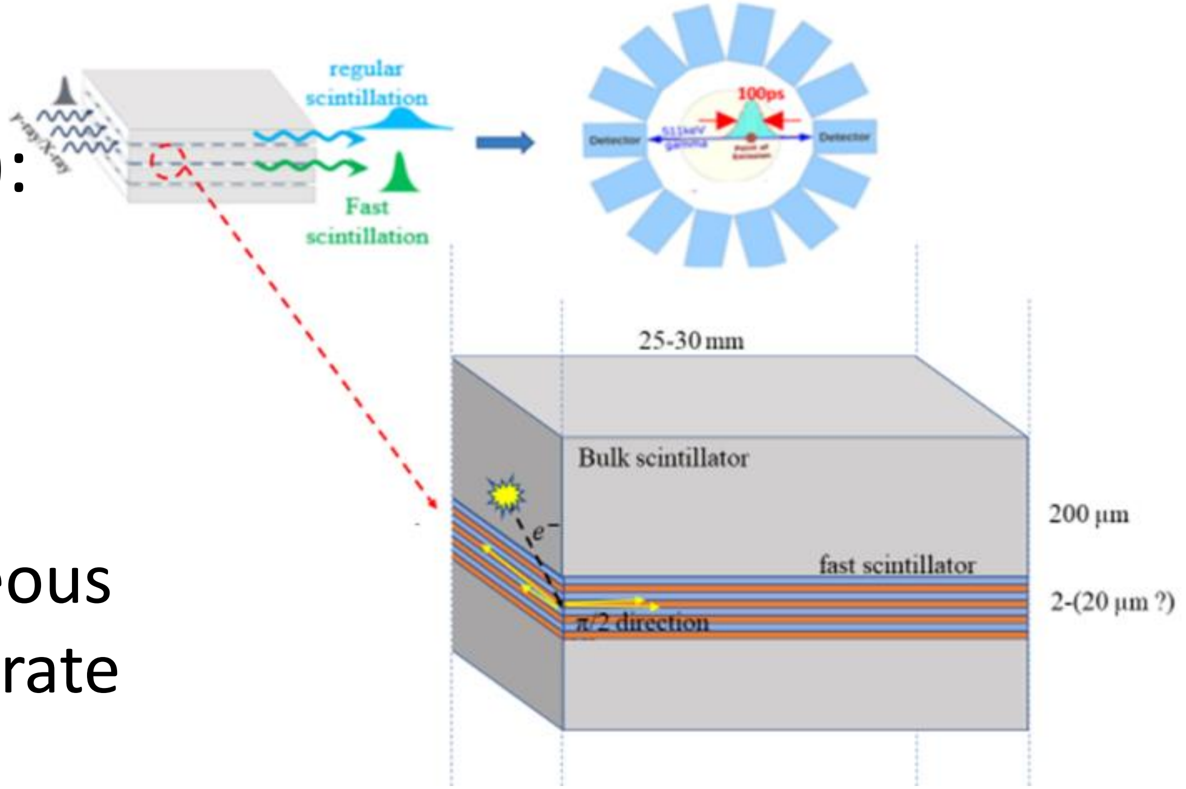
Surpassing the 100 ps CTR barrier

- Metascintillator + Nanophotonics

- Design strategy (minimize the CTR):

$$\max_{\mathbf{d}} \int_{\Delta\theta} d\theta\, \eta(\mathbf{d})\Gamma(\theta;\mathbf{d})/\Gamma_0$$

efficiency — spontaneous emission rate

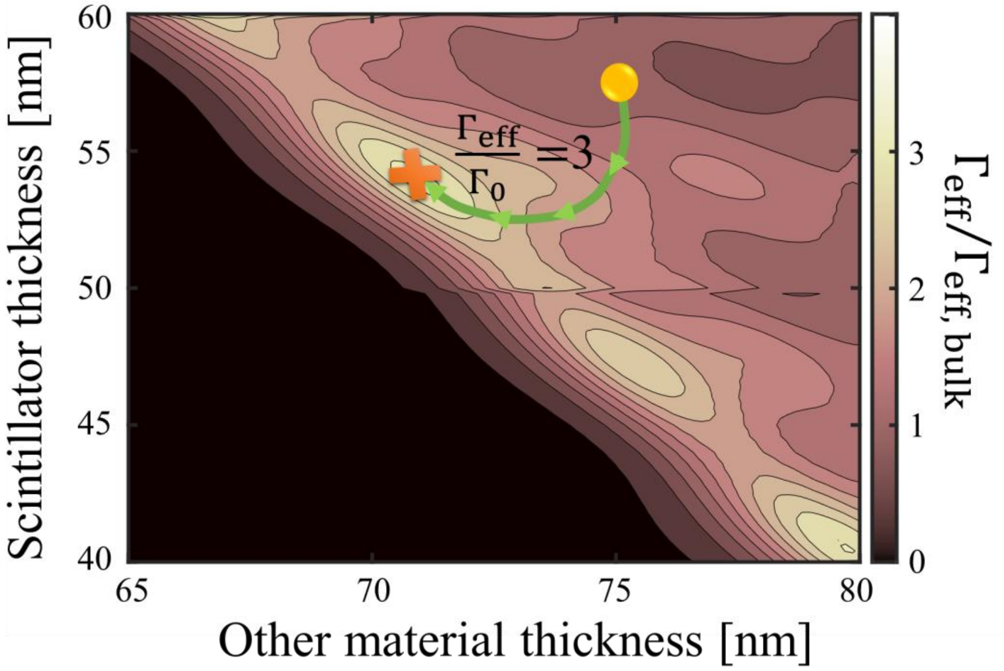


- Optimize over the thicknesses - $\mathbf{d}$

Design Tool – Scint City

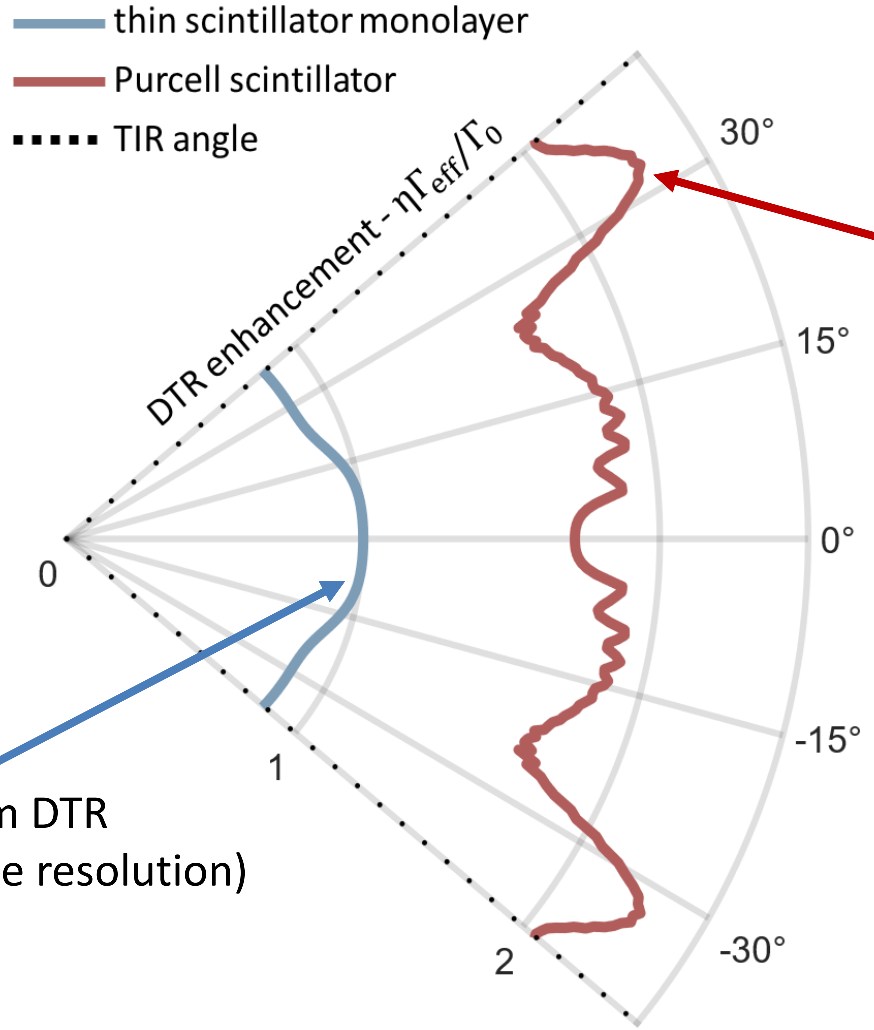
- Calculate the effective emission rate
- Optimize the effective emission rate
- Optimize for the CTR
- Optimize for the point-spread-function (PSF)

Example of the design of a nanophotonic scintillator



A. Shultzman et al., Enhanced Imaging Using Inverse Design of Nanophotonic Scintillators, Advanced Optical Material (2023)

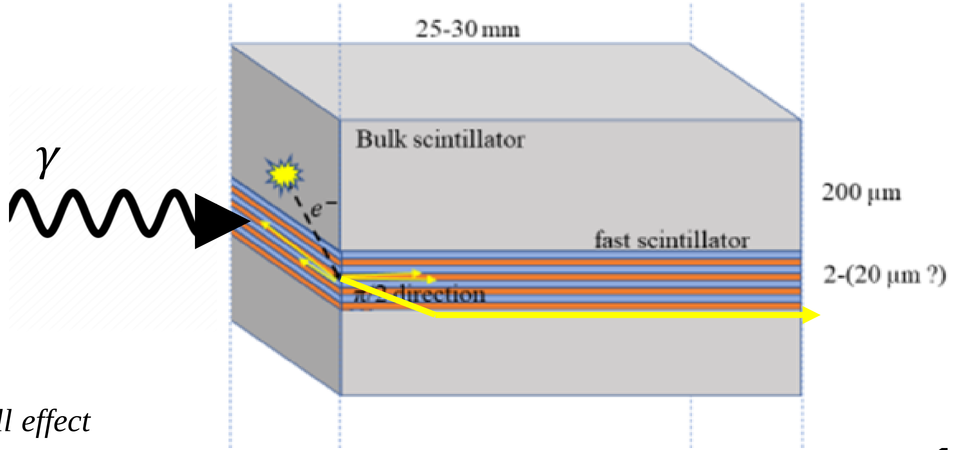
Surpassing the 100 ps CTR barrier



DTR angular Enhancement by **1.65**

Travel in parallel in the BGO

Uniform DTR
(Detection time resolution)

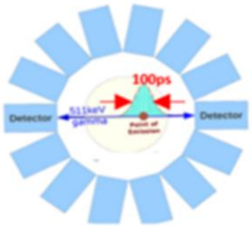


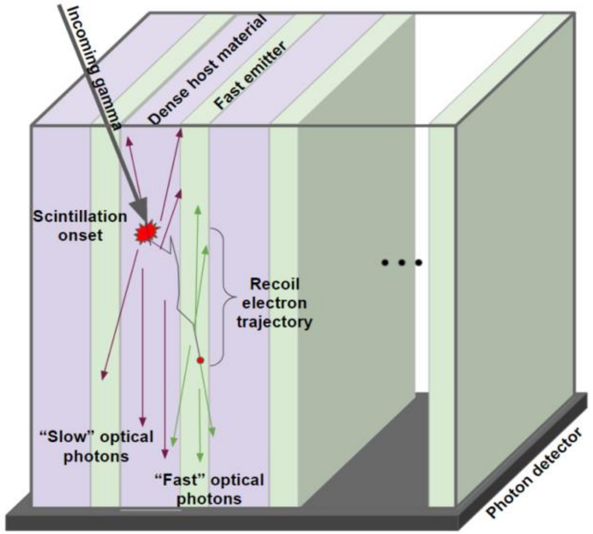
A. Shultzman et al., Towards a second generation of metascintillators using the Purcell effect
Submitted to IEEE TRPMS, Feb 2024

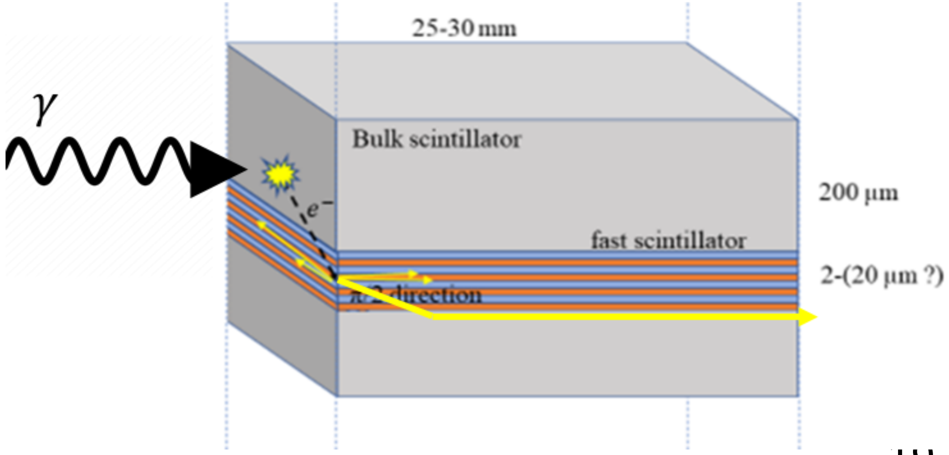
Conclusions and Future Directions

Conclusions

- Benefits of meta-nanophotonic scintillators
- Shaping the emission to be parallel
- Proposal to surpass the 100 ps CTR barrier and approach the 10ps target
  - BGO-based ~ 100ps
  - LYSO-based ~ 50 ps







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The best time to plant a tree was 20 years ago
The second best time is now