# Fast Crystal Scintillators for GHz Hard X-Ray Imaging


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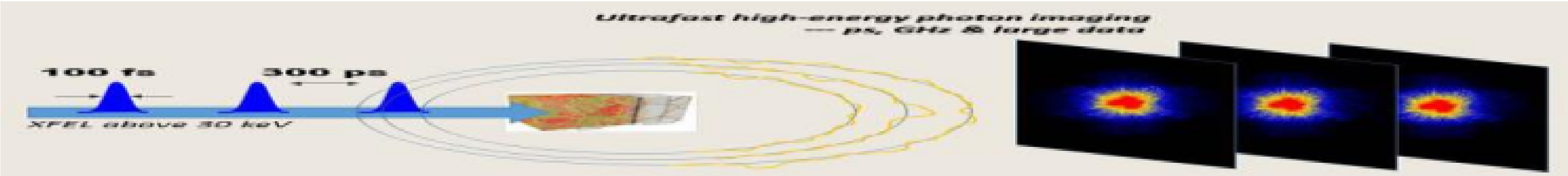
# GHz Hard X-Ray Imaging




**High-Energy and Ultrafast X-Ray Imaging Technologies and Applications**

*Organizers: Peter Denes, Sol Gruner, Michael Stevens & Zhehui (Jeff) Wang*[1]

*(**Location/Time**: Santa Fe, NM, USA /Aug 2-3, 2016)*

The goals of this workshop are to gather the leading experts in the related fields, to prioritize tasks for ultrafast hard X-ray imaging detector technology development and applications in the next 5 to 10 years, see Table 1, and to establish the foundations for near-term R&D collaborations.

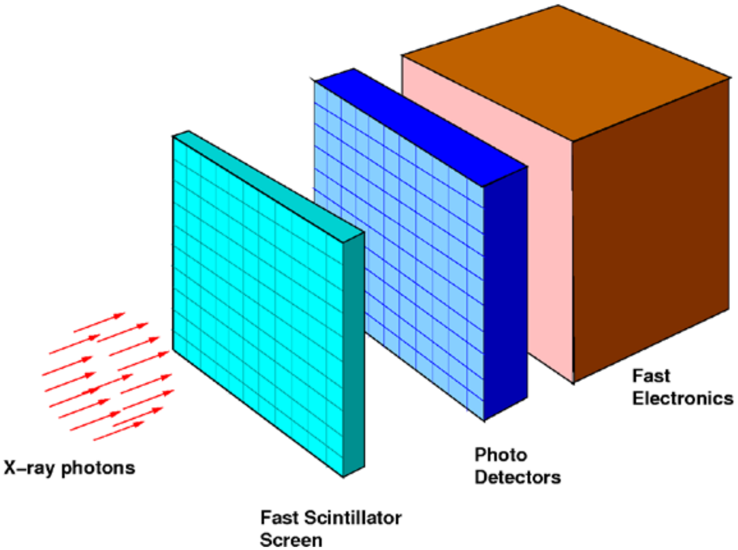
Table I. High-energy photon imagers for MaRIE XFEL

| Performance | Type I imager | Type II imager |
|---|---|---|
| X-ray energy | 30 keV | 42-126 keV |
| Frame-rate/inter-frame time | 0.5 GHz/2 ns | 3 GHz / 300 ps |
| Number of frames | 10 | 10 - 30 |
| X-ray detection efficiency | above 50% | above 80% |
| Pixel size/pitch | ≤ 300 μm | < 300 μm |
| Dynamic range | $10^3$ X-ray photons | ≥ $10^4$ X-ray photons |
| Pixel format | 64 x 64 (scalable to 1 Mpix) | 1 Mpix |

**2 ns and 300 ps inter-frame time requires very fast sensor**

# Why Crystal Scintillator?

- Detection efficiency for hard X-ray requires bulk detector.
- Scintillation light provides fast signal.
- Pixelized crystal detector is a standard for medical industry.
- A detector concept:
  - Pixelized fast scintillator screen;
  - Pixelized fast photodetector;
  - Fast electronics readout.

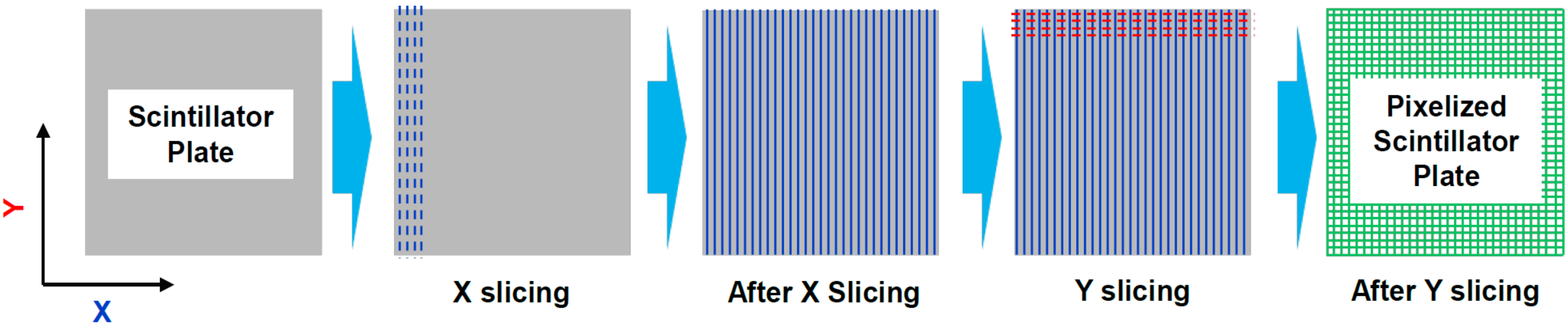



- Challenges:

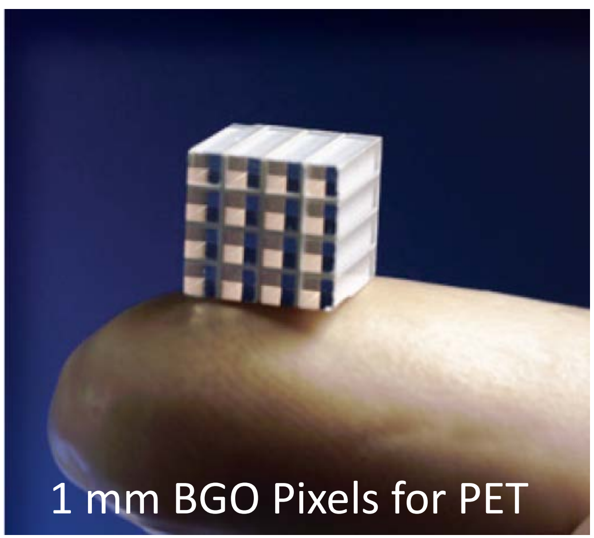
  Ultra-fast crystals, photodetectors and readout.

# Pixelized Crystal Detectors

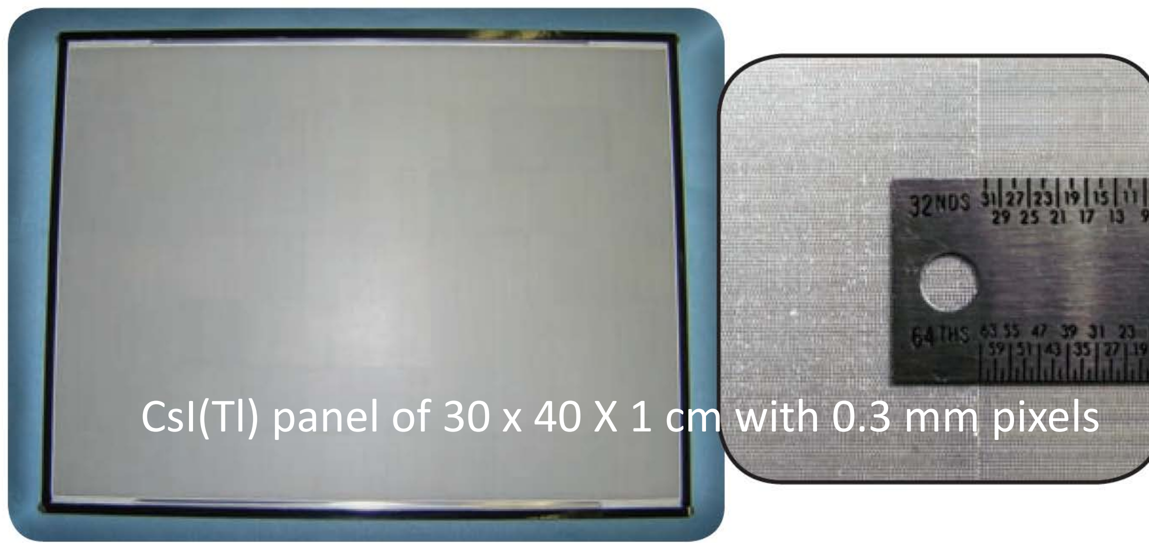
**Crystal panels of 300 µ pitch may be fabricated by classical mechanical processing**




**A Schematic showing pixelized scintillator plate processing**



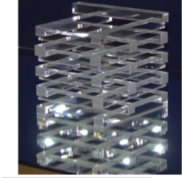
1 mm BGO Pixels for PET



CsI(Tl) panel of 30 x 40 X 1 cm with 0.3 mm pixels

**Laser slicing, micropore or not pixelized provide better coverage**

# Candidate Scintillators for Marie



| | LYSO (:Ce) | YSO:Ce | ZnO:Ga | $BaF_2$ | $BaF_2$:Y | YAP:Ce | YAP:Yb | YAG:Yb | LuAG:Ce | $LaBr_3$ (:Ce) |
|---|---|---|---|---|---|---|---|---|---|---|
| Density (g/cm³) | 7.4 | 4.44 | 5.67 | 4.89 | 4.89 | 5.35 | 5.35 | 4.56 | 6.76 | 5.29 |
| Melting points (°C) | 2050 | 2070 | 1975 | 1280 | 1280 | 1870 | 1870 | 1940 | 2060 | 783 |
| $X_0$ (cm) | 1.14 | 3.10 | 2.51 | 2.03 | 2.03 | 2.77 | 2.77 | 3.53 | 1.45 | 1.88 |
| $R_M$ (cm) | 2.07 | 2.93 | 2.28 | 3.1 | 3.1 | 2.4 | 2.4 | 2.76 | 2.15 | 2.85 |
| $\lambda_I$ (cm) | 20.9 | 27.8 | 22.2 | 30.7 | 30.7 | 22.4 | 22.4 | 25.2 | 20.6 | 30.4 |
| $Z_{eff}$ | 64.8 | 33.3 | 27.7 | 51.6 | 51.6 | 31.9 | 31.9 | 30 | 60.3 | 45.6 |
| dE/dX (MeV/cm) | 9.55 | 6.57 | 8.42 | 6.52 | 6.52 | 8.05 | 8.05 | 7.01 | 9.22 | 6.90 |
| $\lambda_{peak}$ [a] (nm) | 420 | 420 | 389 | 300<br>220 | 300<br>220 | 370 | 350 | 350 | 520 | 360 |
| Refractive Index[b] | 1.82 | 1.78 | 2.1 | 1.50 | 1.50 | 1.96 | 1.96 | 1.87 | 1.84 | 1.9 |
| Normalized Light Yield[a,c] | 100 | 80 | 6.6[e] | 42<br>4.8 | 1.7<br>4.8 | 9<br>32 | 0.19[e] | 0.36[e] | 35[f]<br>48[f] | 153 |
| Total Light yield (ph/MeV) | 30,000 | 24,000 | 2,000[e] | 13,000 | 2,000 | 12,000 | 57[e] | 110[e] | 25,000[f] | 46,000 |
| Decay time[a] (ns) | 40 | 75 | <1 | 600<br>0.6 | 600<br>0.6 | 191<br>25 | 1.5 | 4 | 981[f]<br>64[f] | 20 |
| Light Yield in 1st ns (photons/MeV) | 740 | 318 | 610[e] | 1200 | 1200 | 391 | 28[e] | 24[e] | 240 | 2,200 |
| 40 keV Att. Length (1/e, mm) | 0.185 | 0.334 | 0.407 | 0.106 | 0.106 | 0.314 | 0.314 | 0.439 | 0.251 | 0.131 |

[1] Spurrier, et al., *IEEE T. Nucl. Sci.* 2008,55 (3): 1178-1182.

a. Top line: slow component, bottom line: fast component;

b. At the wavelength of the emission maximum;

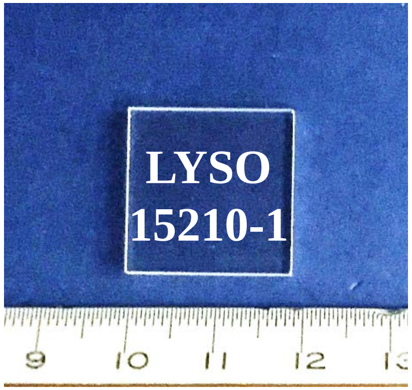
c. Excited by Gamma rays;

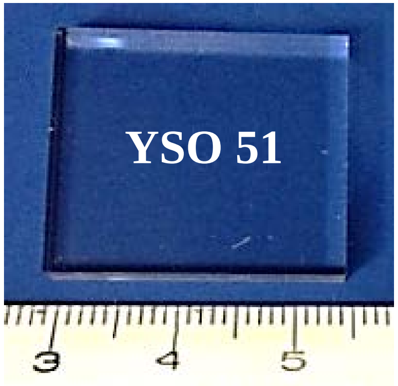
d. For 0.4 at% Ca co-doping;

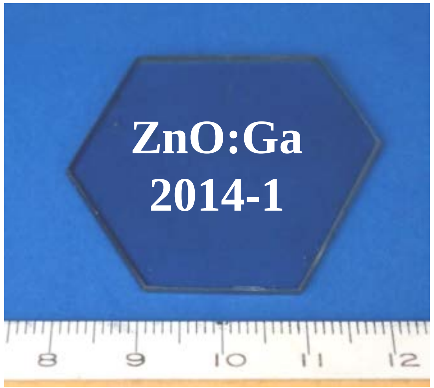
e. Excited by Alpha particles.

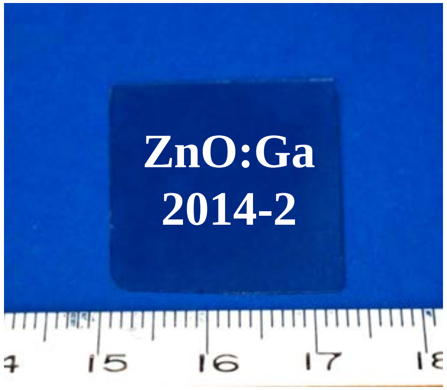
f. Ceramic with 0.3 Mg at% co-doping

# LYSO and ZnO:Ga Samples




| Crystal | Vendor | ID | Dimension ($mm^3$) |
|---|---|---|---|
| LYSO:Ce | SIC | 150210-1 | 19x19×2 |
| YSO:Ce | SIC | 51 | 25×25×5 |
| ZnO:Ga | FJIRSM | 2014-1 | 33×30×2 |
| ZnO:Ga | FJIRSM | 2014-2 | 22×22×0.3 |

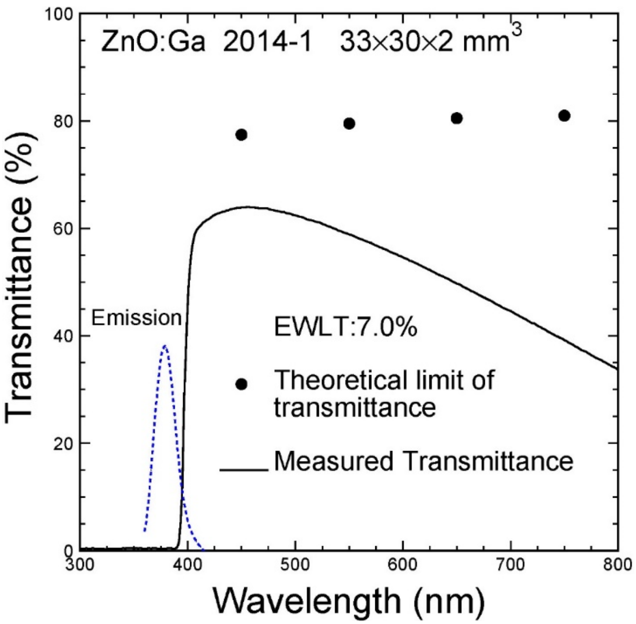
## Experiments

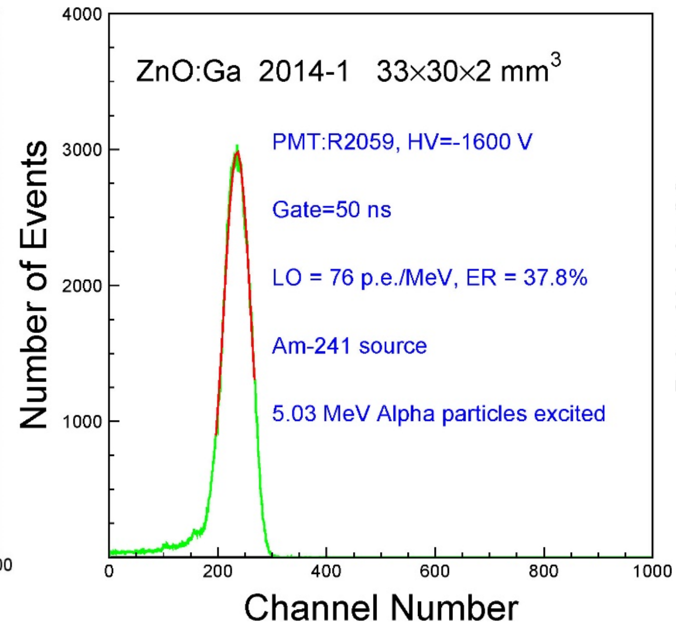
- Properties measured at room temperature : PL & Decay, Transmittance, PHS, LO & Decay kinetics

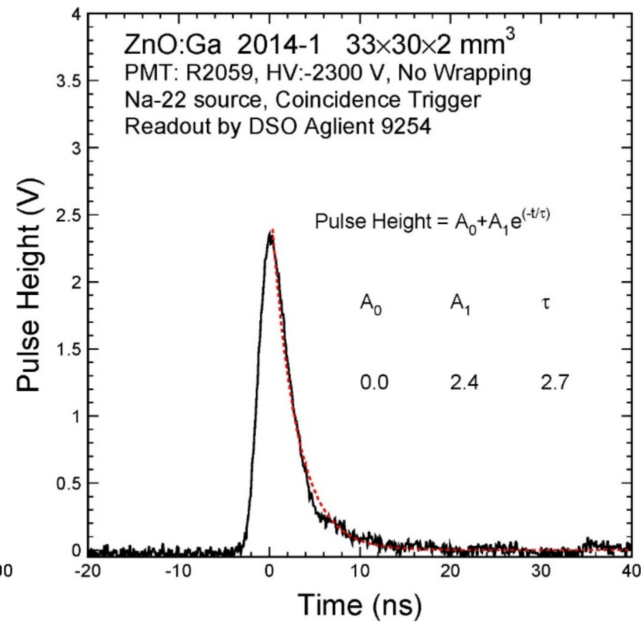
# FJIRSM ZnO:Ga-2014-1

✓ **Very short decay time**

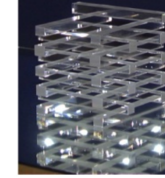
× **Low EWLT and LO due to severe self absorption**



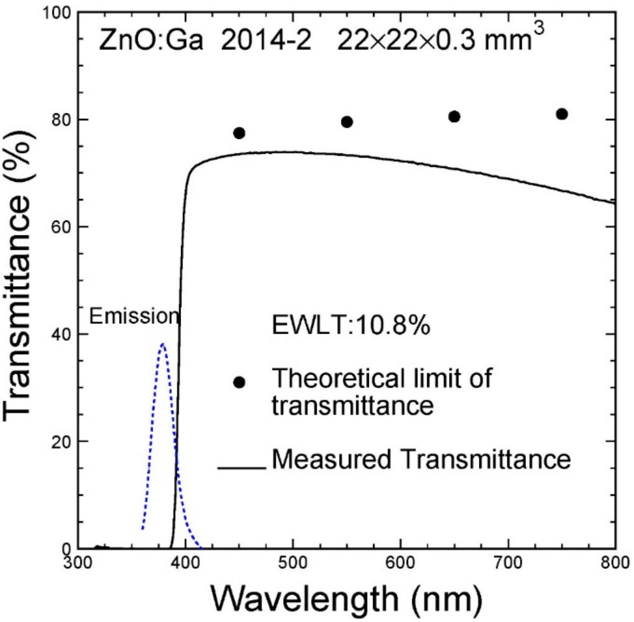



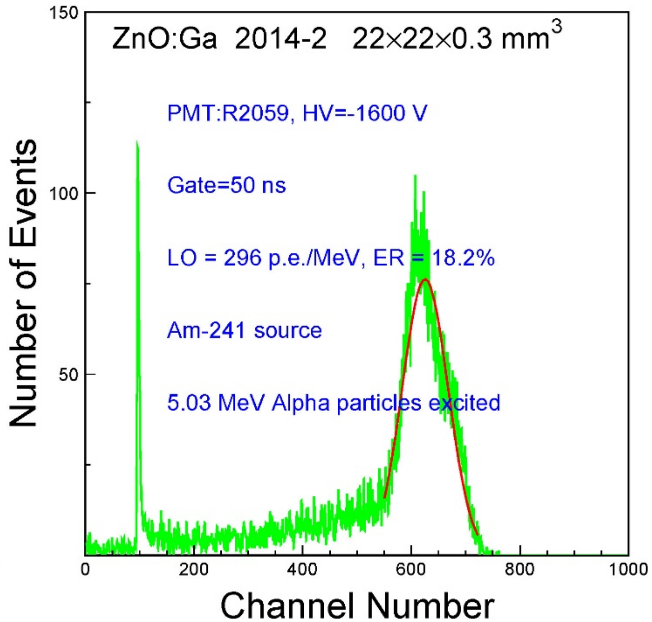

| ID | Dimension | EWLT (%) | ER (%) | 50 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| FJIRSM ZnO:Ga-2014-1 | 33×30×2 | 7.0 | 37.8 | 76 (α) | 2.7 |

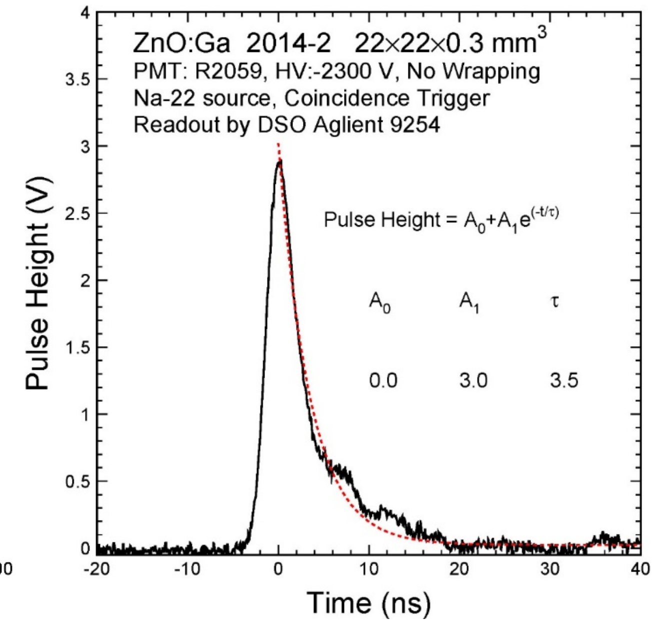


# FJIRSM ZnO:Ga-2014-2

- × Reduced self absorption due to 0.3 mm thickness
- × May pursue QD, NP or thin film based solution

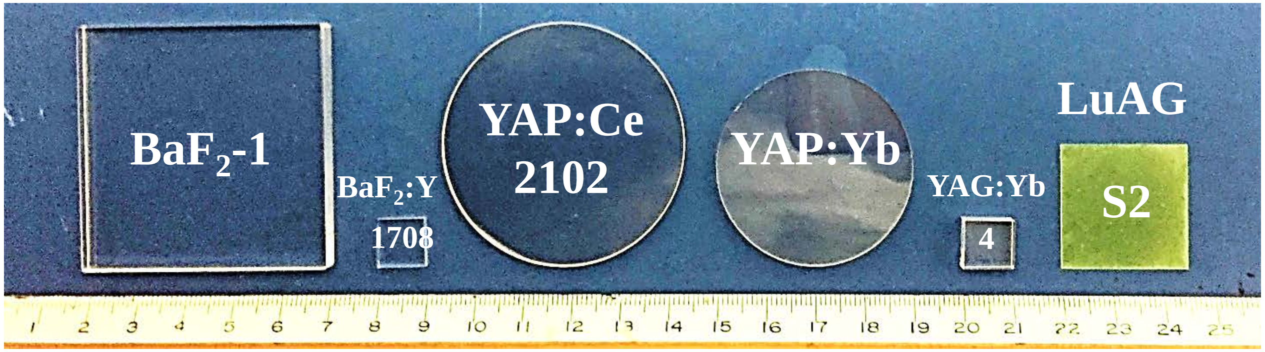



| ID | Dimension | EWLT (%) | ER (%) | 50 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| FJIRSM ZnO:Ga-2014-2 | 22×22×0.3 | 10.8 | 18.2 | 296 (α) | 3.5 |

# $BaF_2$ and Other Samples




| Crystal | Vendor | ID | Dimension ($mm^3$) |
|---|---|---|---|
| $BaF_2$ | SIC | 1 | 50×50×5 |
| $BaF_2$:Y | BGRI | 1708 | 10×10×2 |
| YAP:Ce | Dongjun | 2102 | Φ50×2 |
| YAP:Yb | Dongjun | 2-2 | Φ40×2 |
| YAG:Yb | Dongjun | 4 | 10×10×5 |
| LuAG:Ce | SIC | S2 | 25×25×0.4 |

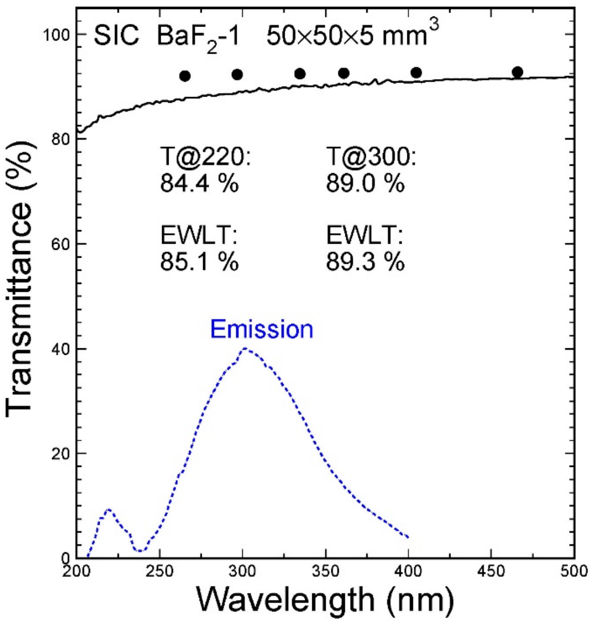
## Experiments

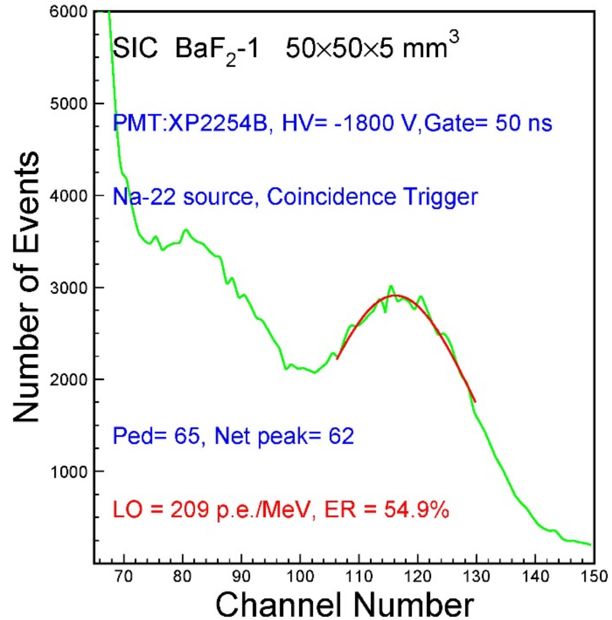
- Properties measured at room temperature : PL & Decay, Transmittance, PHS, LO & Decay kinetics

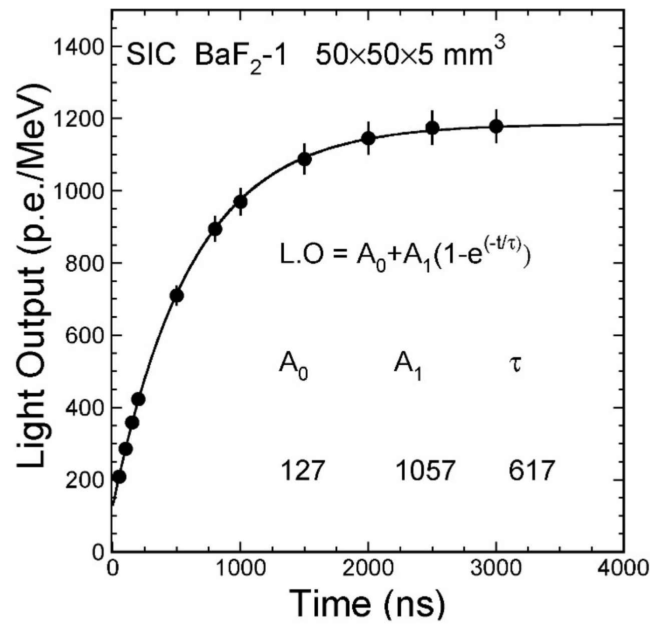
# SIC $BaF_2$-1

✓ The highest LY in 1st ns among all non-hygroscopic scintillators

× ~600 ns slow component may be suppressed by Y doping



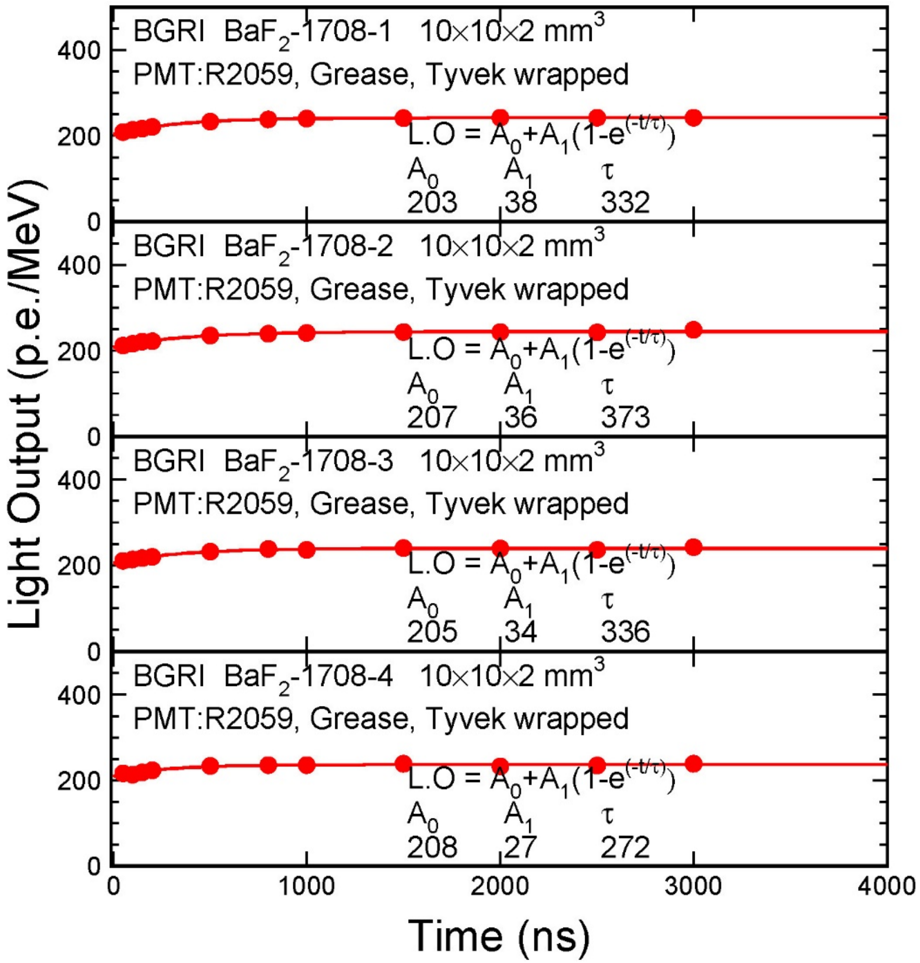



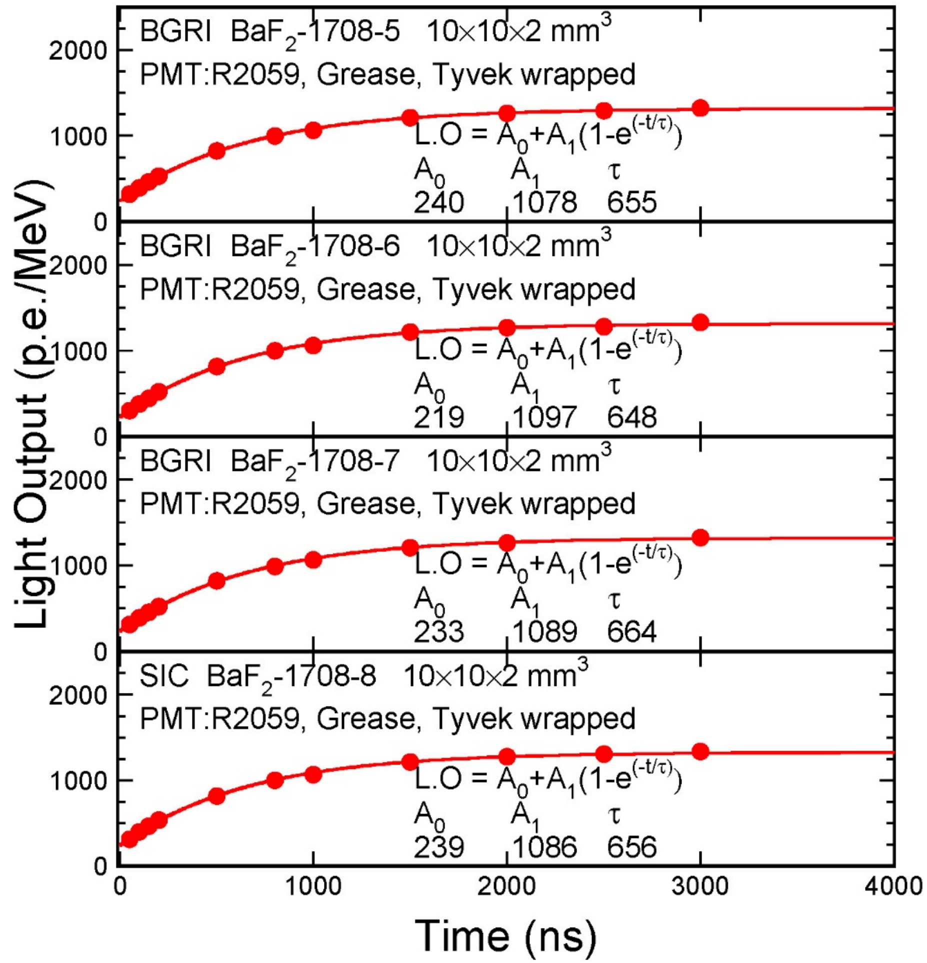




| ID | Dimension | EWLT (%) | ER (%) | 50 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| SIC $BaF_2$-1 | 50×50×5 | 85.1 | 54.9 | 209 | 0.6 |

# BGRI Y Doped/Undoped $BaF_2$

F/S ratio increased from 0.21 to 6 .2




Will be discussed in details in NSS2017 at Atlanta

# Use Thin Layer Scintillators

Proc. of SPIE Vol. 9504 95040N

A multilayer high QE photocathode coated thin fast scintillators concept was proposed for GHz hard X-ray imaging:

- Spatial resolution determined layer thickness,
- Overall efficiency defined layer number,
- Maximized conversion of scintillation photon to p.e.,
- Magnetic field extraction of p.e. and image preserving,
- Off-beam p.e. multiplication,
- On-board charge storages.

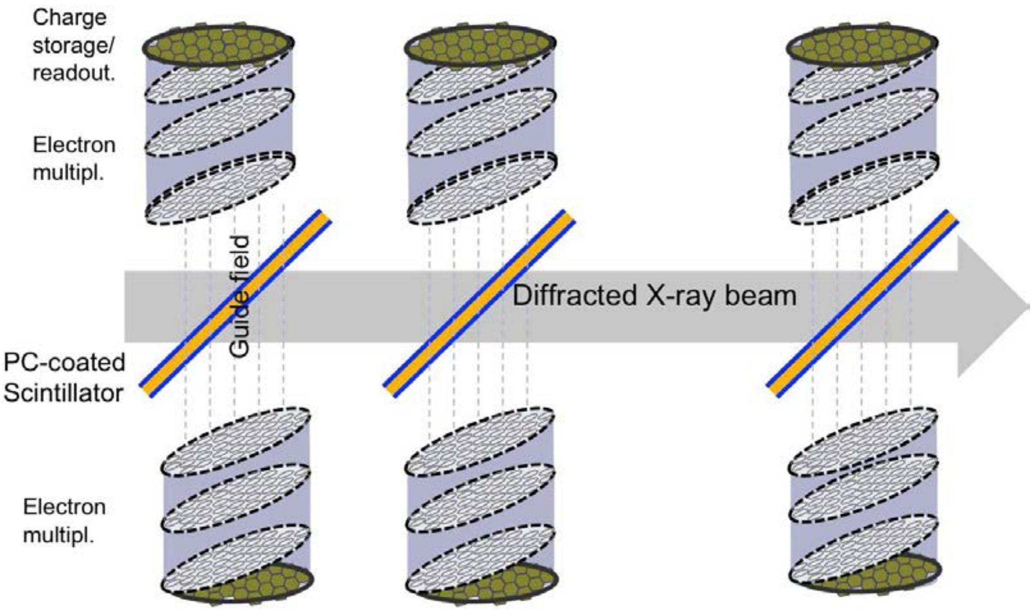



**Figure 6.** A multi-layer detector architecture for efficient and fast imaging of diffracted X rays. A guide magnetic field perpendicular to the X-ray direction guide the photoelectrons to amplification and storage. The magnetic field also preserves the image contrast due to X-ray absorption at the scintillator location.

# Ag/Au-ZnO Core-Shell Nano Particles

Nature Scientific **Reports** | 5:14004 | DOI: 10.1038/srep14004

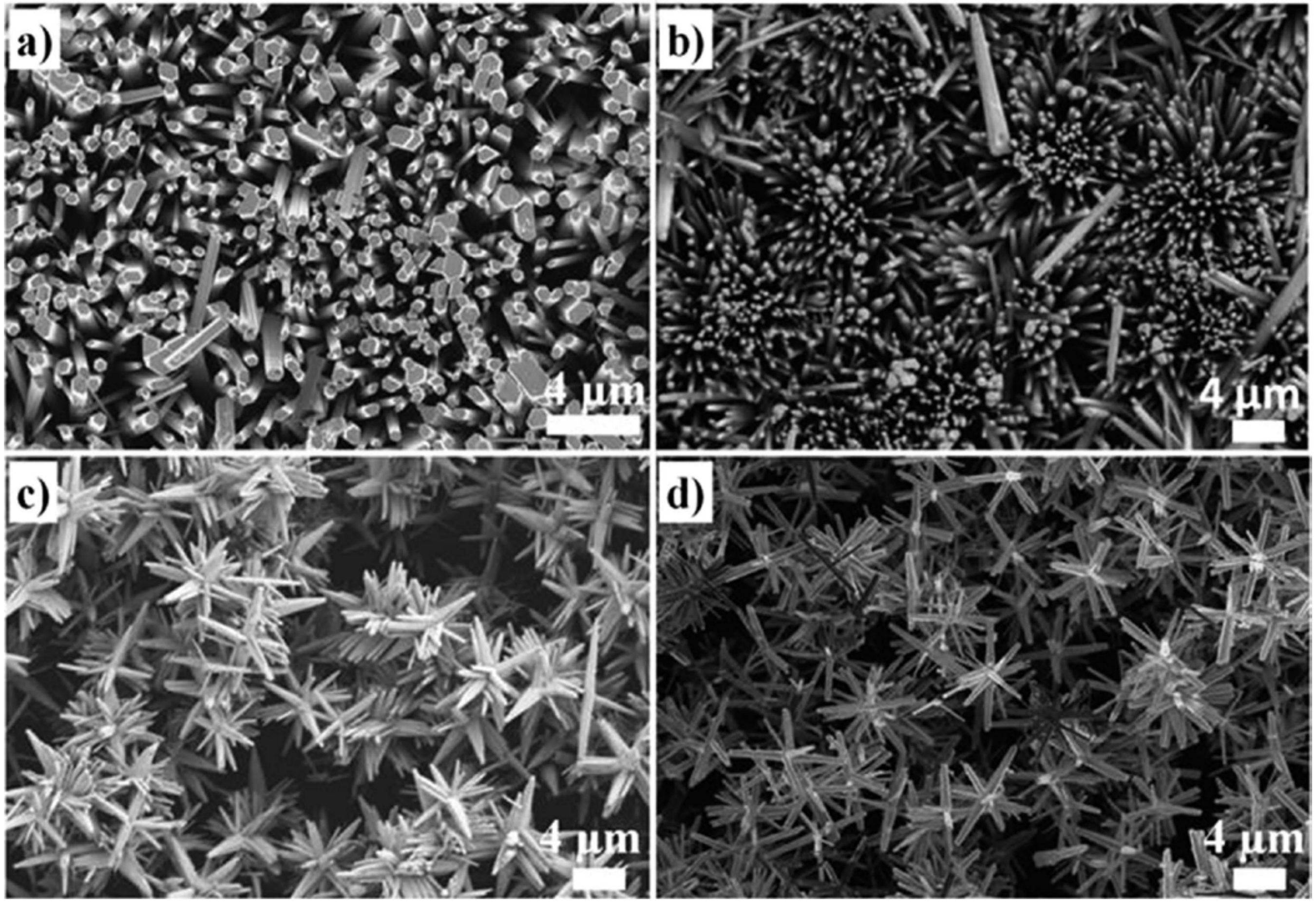
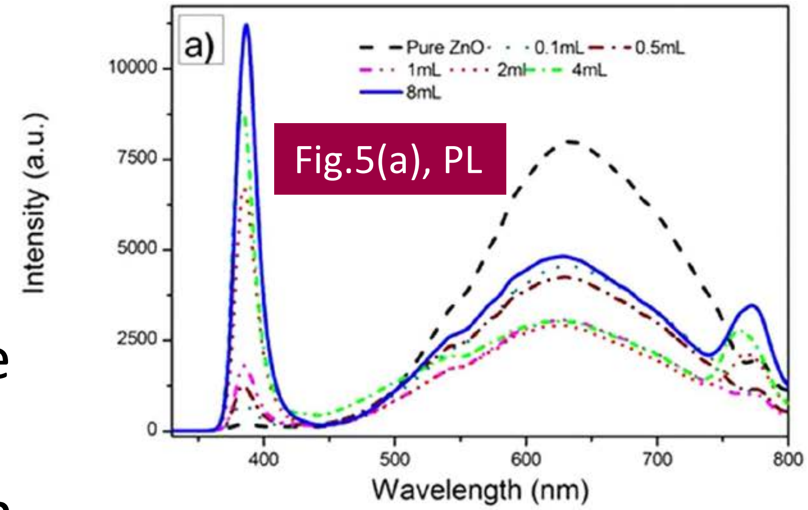


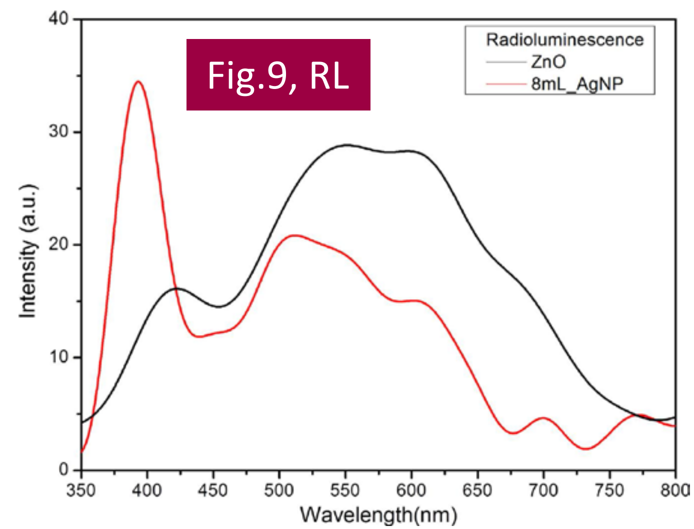

**Figure 2.** SEM images for ZnO samples without nanoparticles (**a**), with 2 mL_AgNP (**b**), 8 mL_AgNP (**c**), and 8 mL_AuNP (**d**) illustrating the change in shape of the particles. The particles in the lower two micrographs are referred to as "star" or "thistle" shaped in the text.

# Enhanced UV Emission in Ag/Au-ZnO

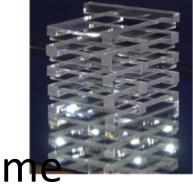
Nature Scientific **Reports** | 5:14004 | DOI: 10.1038/srep14004

- Enhancement of ZnO near-band-edge (UV) emission centered at 385 nm was reported in PL and RL of Ag/Au-ZnO core shell nanoparticles.
- The enhanced luminescence and the decreased free exciton lifetime suggest a plasmon-coupled-emission mechanism.
- This suggests that plasmon-coupled luminescence can be employed for the development of improved scintillators.



# Purcell effect for enhancing ZnO luminescence (Theoretical framework)



Total field :

$$\langle \hat{\boldsymbol{E}}_m(\boldsymbol{r})\rangle = \boldsymbol{E}_m^L(\boldsymbol{r}) + \frac{\omega^2}{\epsilon_0 c^2}\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}_0;\omega)\cdot\boldsymbol{\mu}\langle S\rangle$$

(a) (b) (c)

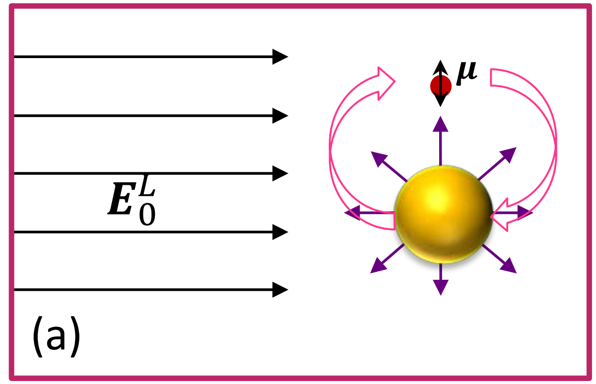
Dipole moment: $\langle \hat{S}\rangle = \frac{-\Omega[2\Delta - i\gamma_m]}{4\Delta^2+2|\Omega|^2+\gamma_m^2}$

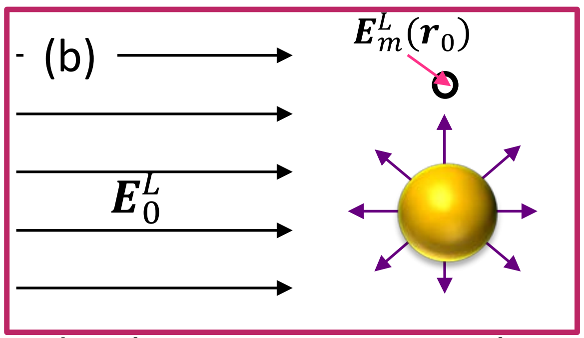
Rabi frequency: $\Omega = 2\boldsymbol{\mu}\cdot\boldsymbol{E}_m^L(\boldsymbol{r}_0)$

$\Delta$ is detuning

$\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}_0;\omega)$ is Dyadic Green's function

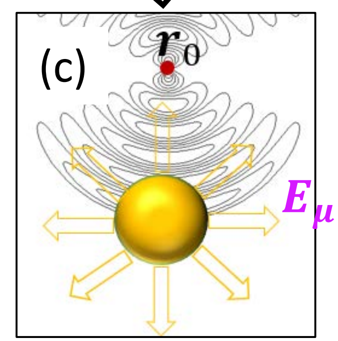
Hybrid system as experimental frame




No dipole: Mie scattering theory

NOTE: dipole is considered as a point in theory. In experiment, ZnO is the dipole.



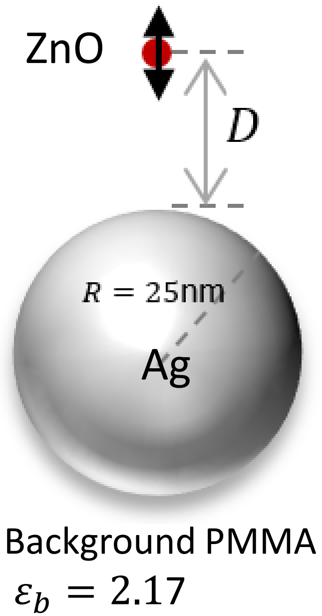

Dipole vs. Nanostructure: dyadic Green's function

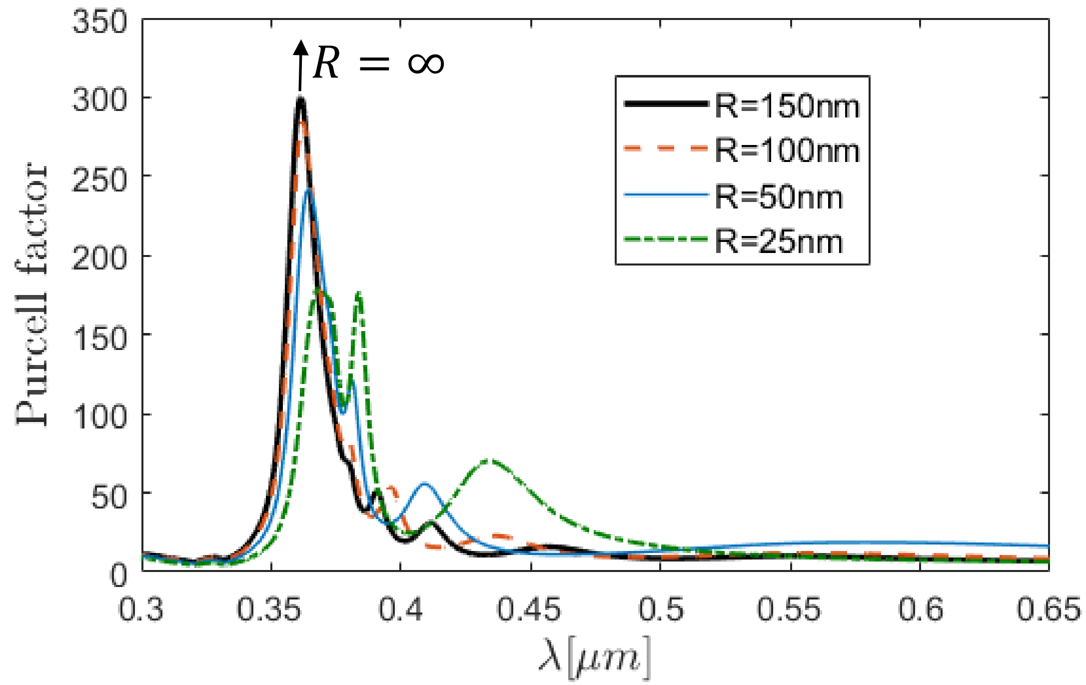
In numerical calculation, the dyadic Green's function is the kernel.

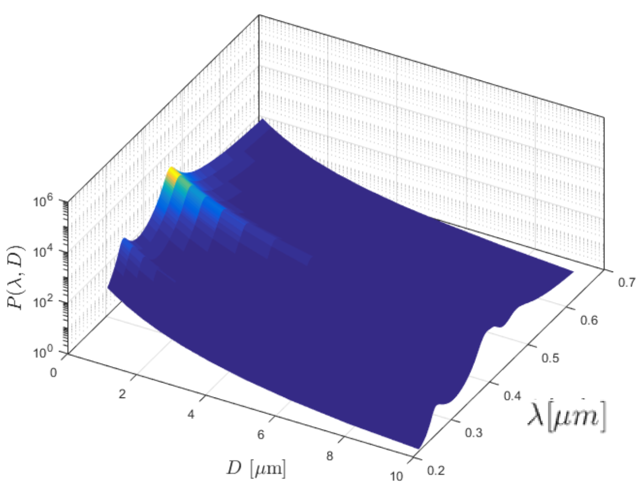
$$\gamma_m = 2\mathrm{Im}[\boldsymbol{\mu}\cdot\boldsymbol{E}_\mu(\boldsymbol{r}_0)] = 2Im[\boldsymbol{\mu}\cdot\overleftrightarrow{\boldsymbol{G}}(\boldsymbol{r},\boldsymbol{r}_0;\omega)\cdot\boldsymbol{\mu}]$$

$\gamma_m/\gamma_0$ is Purcell enhancement factor

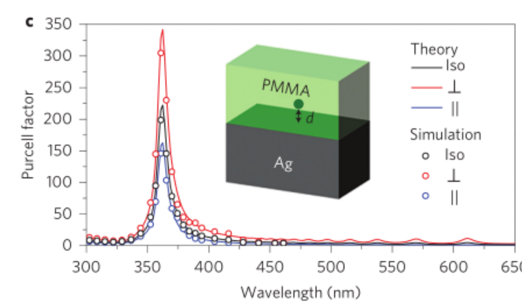
# Purcell Factor for Ag Particles



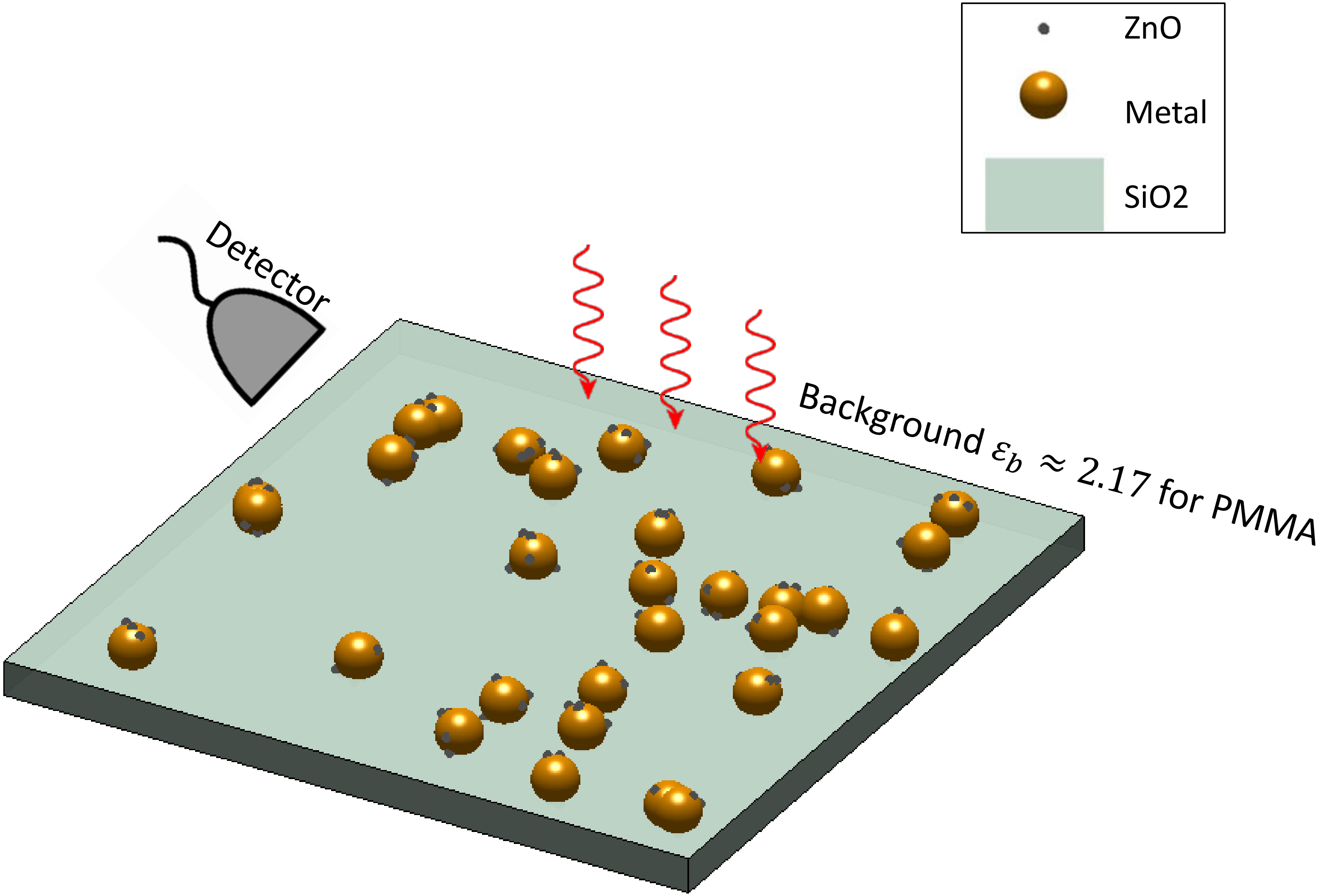

$R = \infty$ is equivalent to a infinite layer. Our simulation is greatly agree with experiments (red circles) as right figure. Agreement includes the peak value, wavelength and bandwidth.





-Lu, Dylan, et al. "Enhancing spontaneous emission rates of molecules using Nano patterned multilayer hyperbolic metamaterials." *Nature nanotechnology*9.1 (2014): 48.

# Experimental Proposal



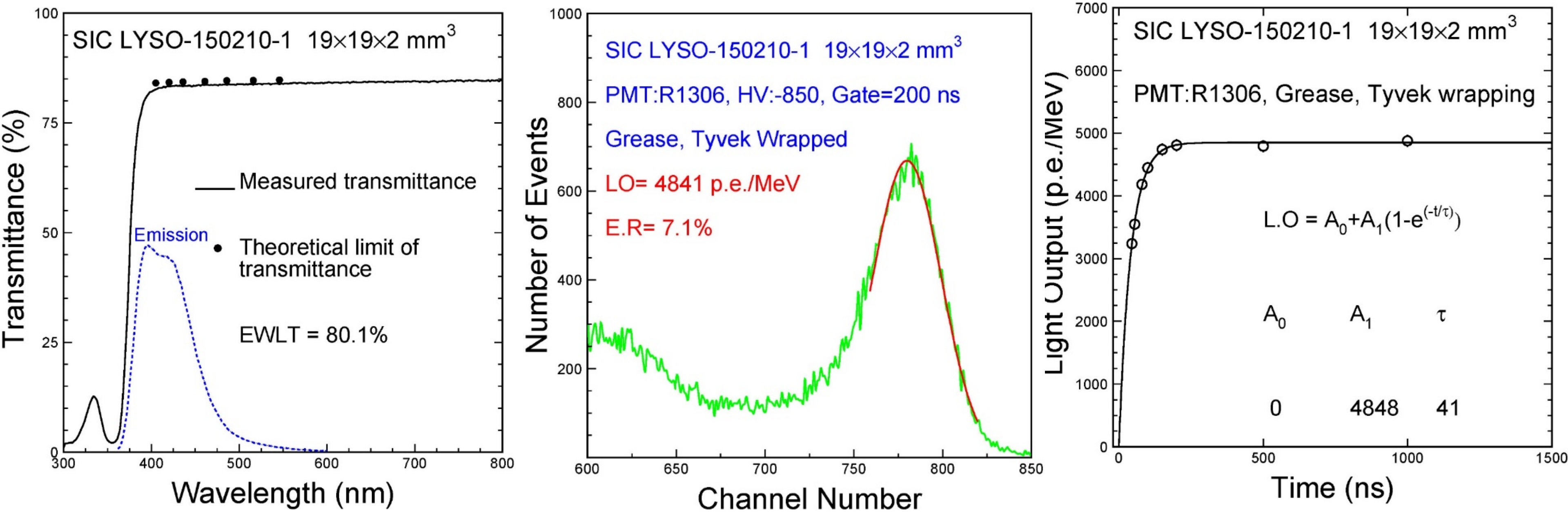
# Summary

- GHz hard X-ray imaging for the proposed Marie project presents an unprecedented challenge to the speed and radiation hardness of inorganic scintillators.
- $BaF_2$ crystals provide sufficient fast light with sub-ns decay time and excellent radiation hardness beyond 100 Mrad and 1 x $10^{15}$ h/cm$^2$. With its slow component effectively suppressed by yttrium doping Y:$BaF_2$ promises a fast and robust front imager.
- Bulk ZnO:Ga crystals suffer from serious self-absorption. Enhanced UV emission observed in Ag/Au ZnO core-shell nano particles hints a thin film based approach.
- Our plan is to investigate along both lines: Y:$BaF_2$ crystals, and ZnO QD/NP based thin film for the Marie project with a close collaboration between the NP, HEP and material science community.

# SIC LYSO:Ce-150210-1 (in LANL)

✓ **High LO, good transmittance and ER, short decay time**

× **Decay time too long for X-ray frame rate of a few ns**

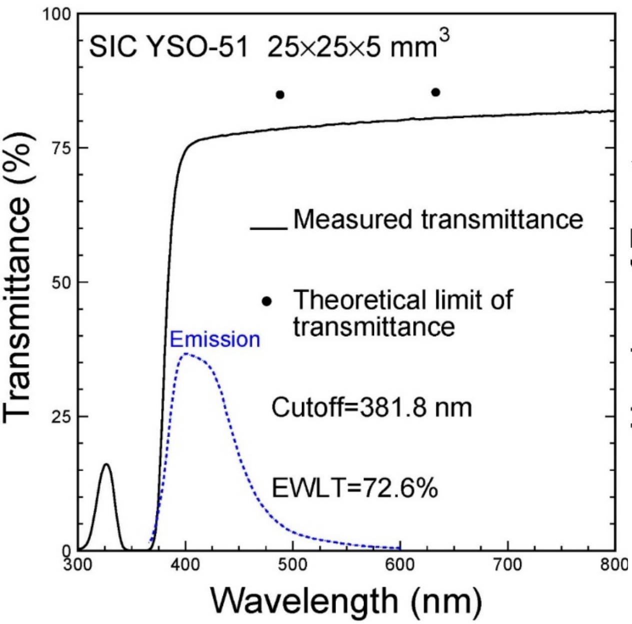


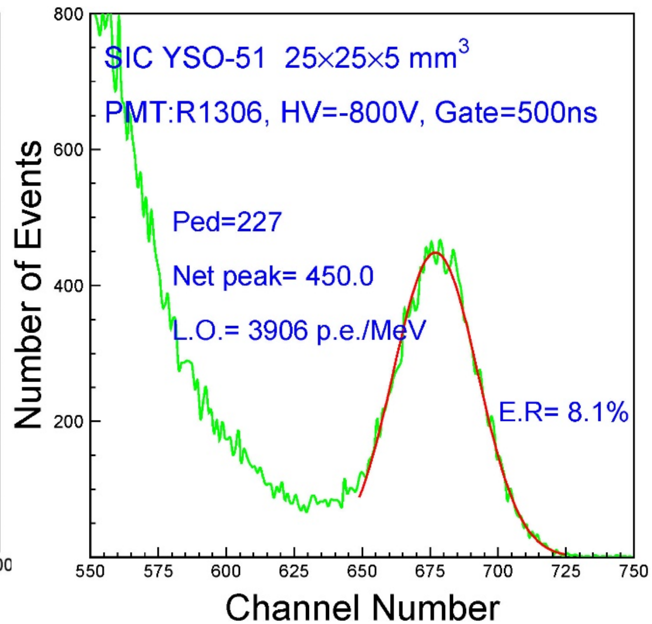

| ID | Dimension | EWLT (%) | ER (%) | 200 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| SIC LYSO-150210-1 | 19×19×2 | 80.1 | 7.1 | 4841 | 41 |

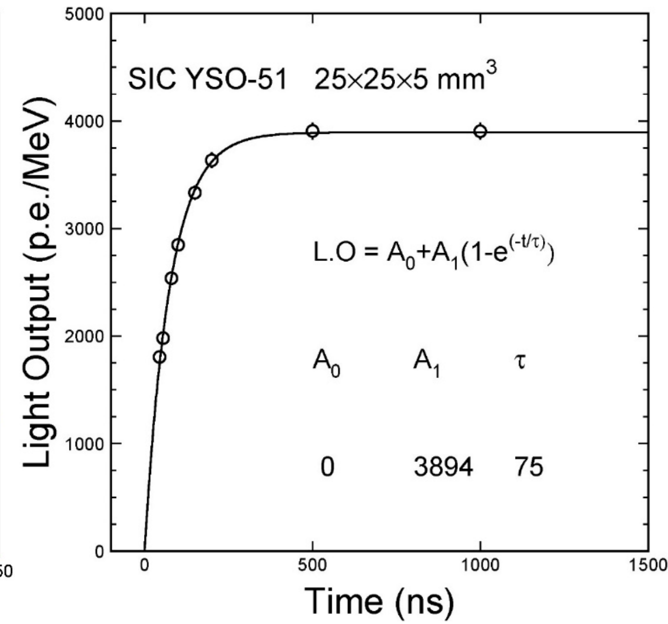
# SIC YSO:Ce-51 (in LANL)

✓ Good LO, transmittance, ER, and short decay time

× All these performance are inferior to LYSO:Ce



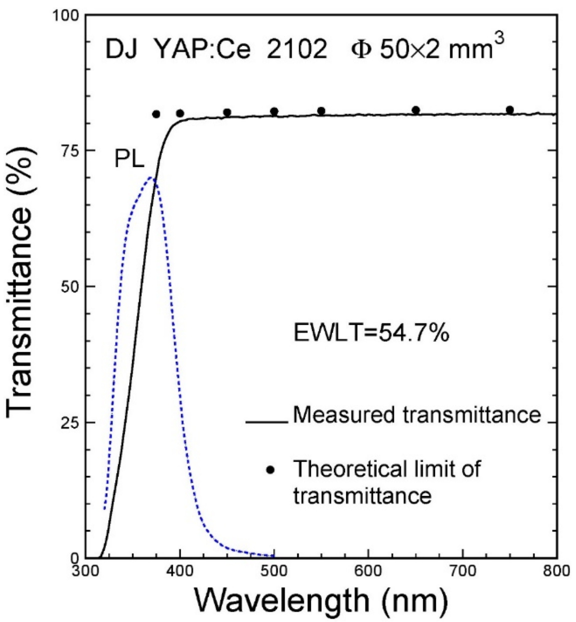



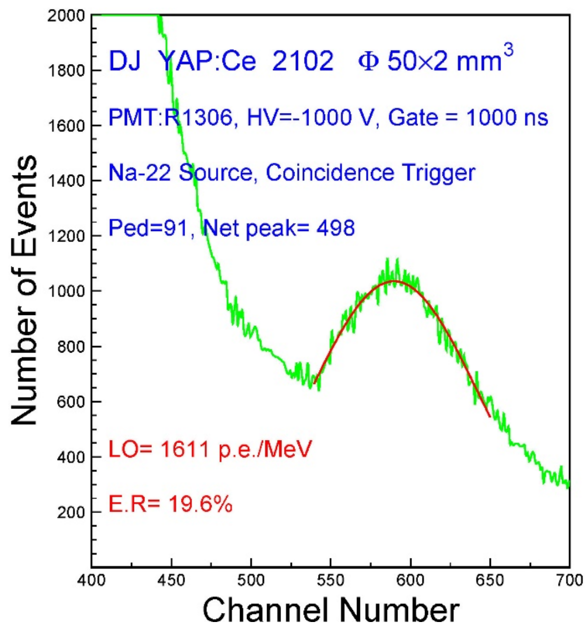

| ID | Dimension | EWLT (%) | ER (%) | 500 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| SIC YSO-51 | 25×25×5 | 72.6 | 8.1 | 3906 | 75 |

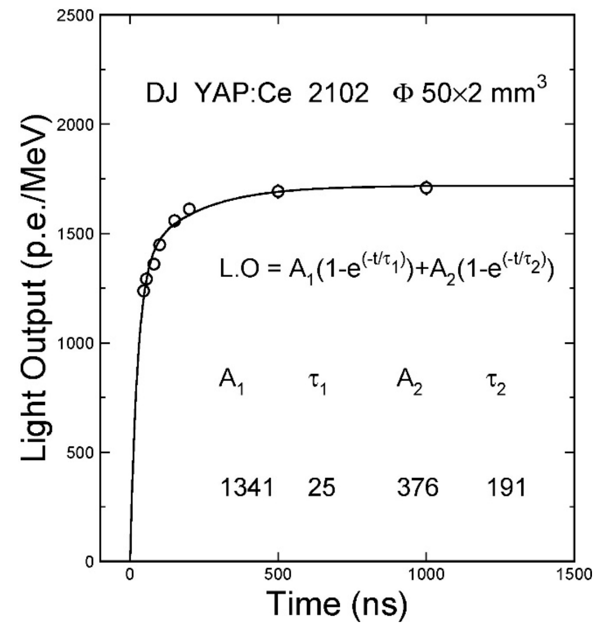
# DJ YAP:Ce-2102

✓ **Adequate LO and ER**

× **Self absorption and slow component**



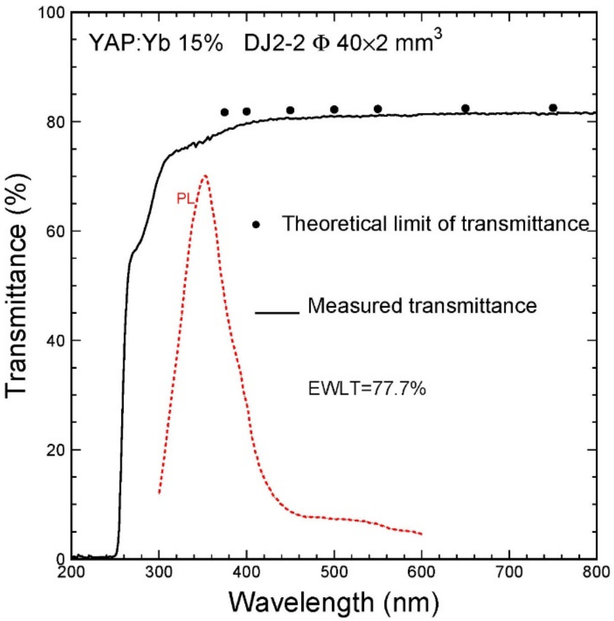



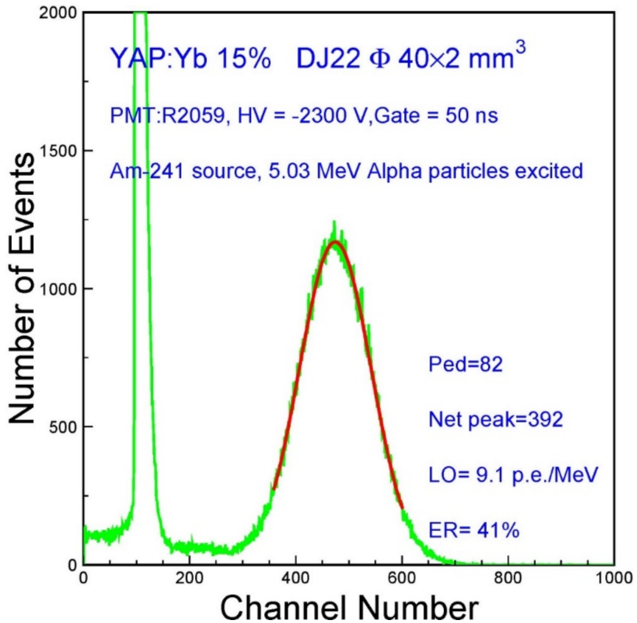

| ID | Dimension | EWLT (%) | ER (%) | 200 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| DJ YAP:Ce-2102 | Φ50×2 | 54.7 | 19.6 | 1611 | 25 |

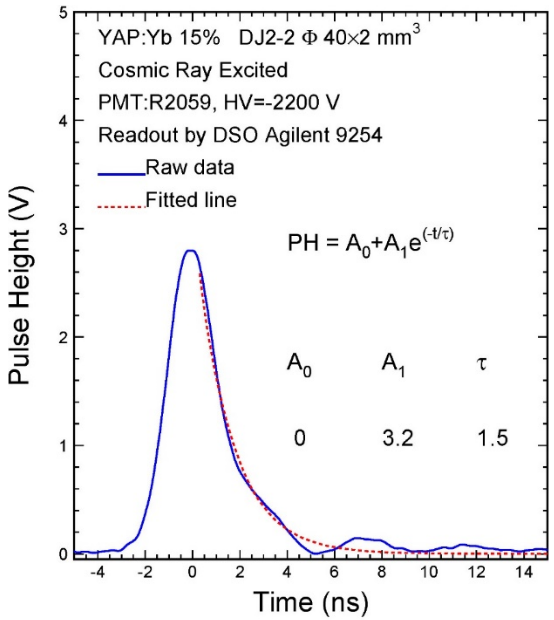
# DJ YAP:Yb-2-2

✓ Very short decay time

× Low LO due to thermal quenching



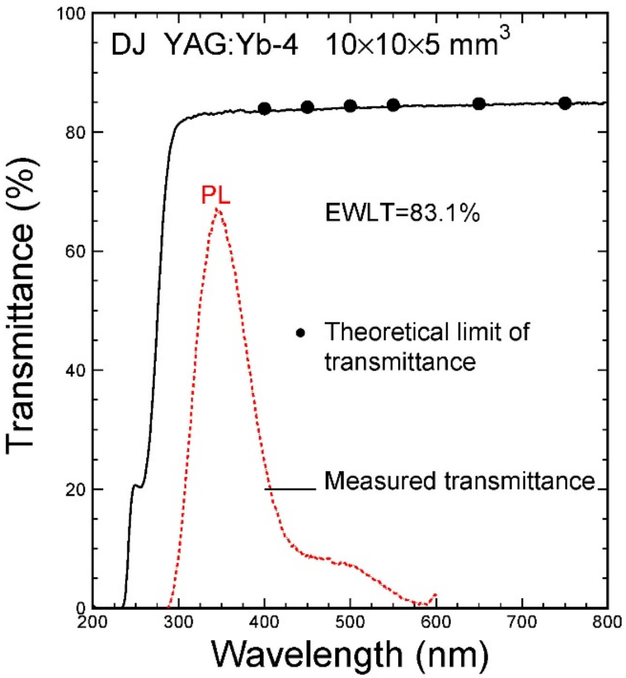



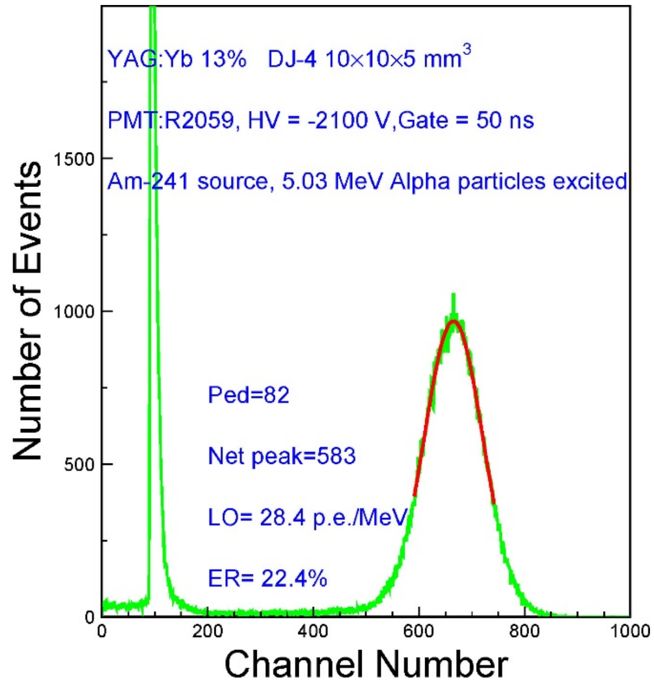

| ID | Dimension | EWLT (%) | ER (%) | 50 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| DJ YAP:Yb-2-2 | Φ40×2 | 77.7 | 41 | 9.1 (α) | 1.5 |

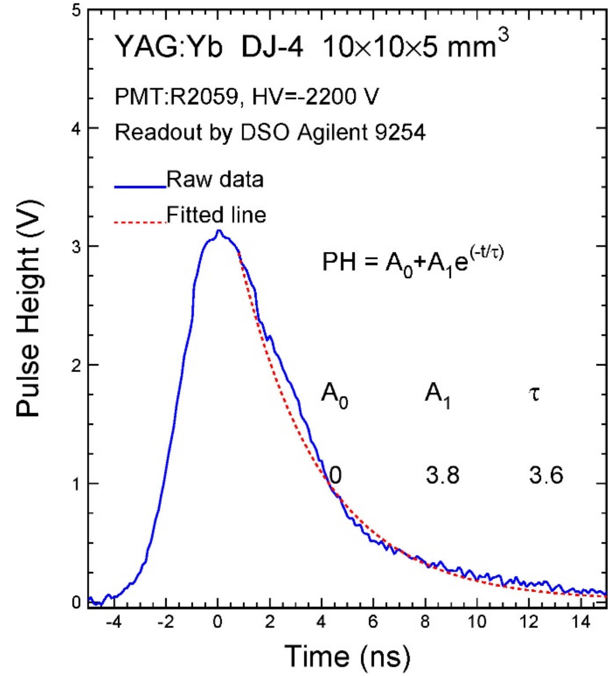
# DJ YAG:Yb-4

✓ **Very short decay time and good transmittance**

× **Low LO due to thermal quenching**



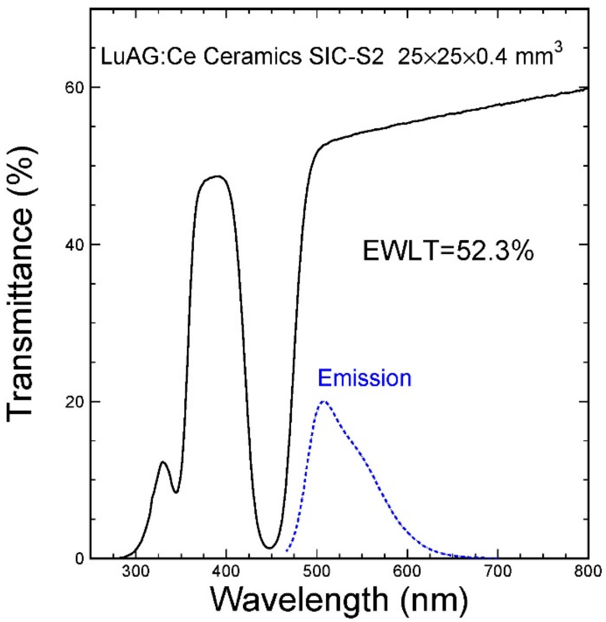



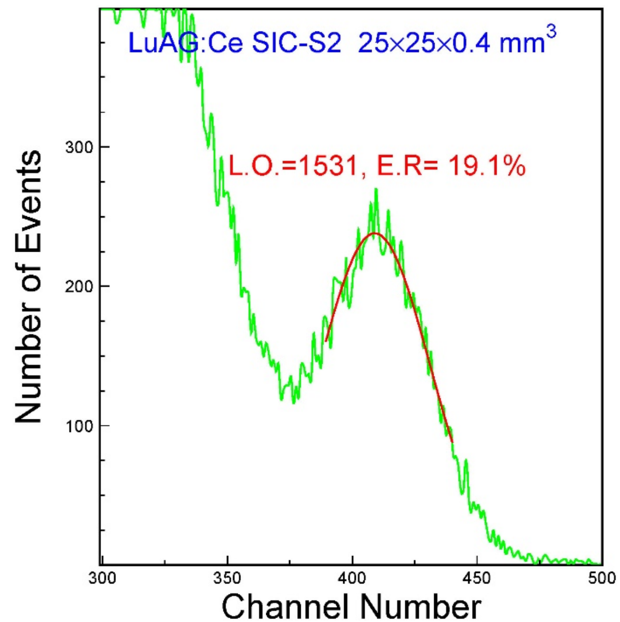

| ID | Dimension | EWLT (%) | ER (%) | 50 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| DJ YAG:Yb-4 | 10×10×5 | 83.1 | 22.4 | 28.4 (α) | 3.6 |

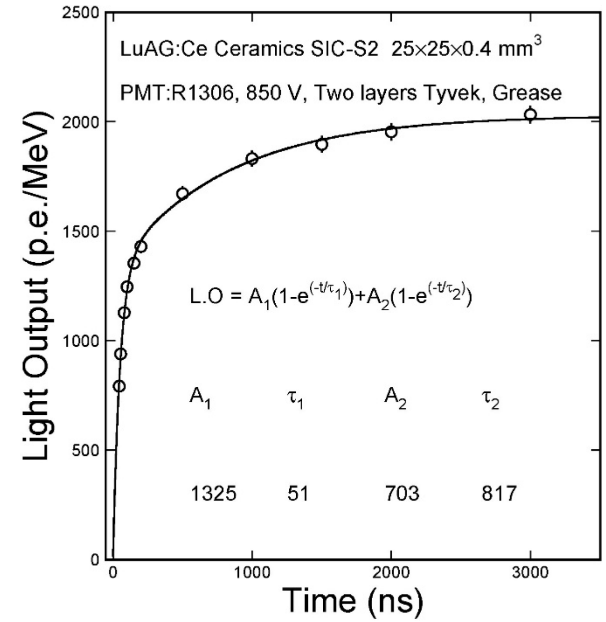
# SIC LuAG-S2 Ceramics

✓ **Good LO and ER, and short decay time**

× **~ 1 μs slow component**

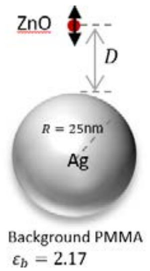


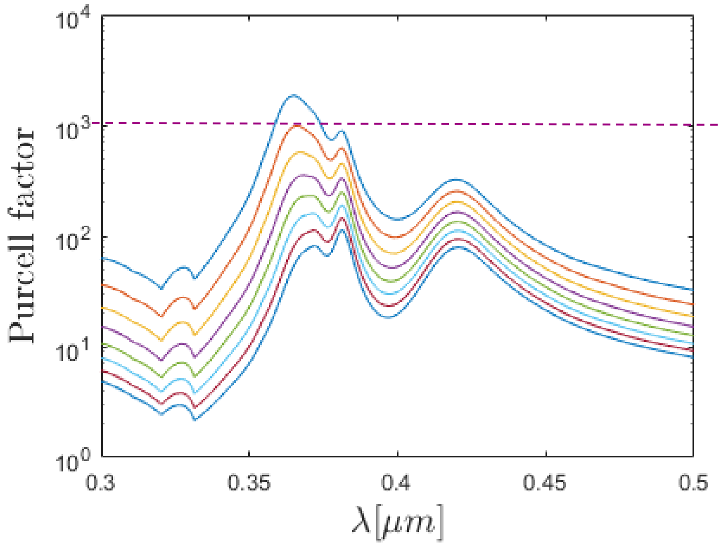




| ID | Dimension | EWLT (%) | ER (%) | 200 ns LO (p.e./MeV) | Primary Decay Time (ns) |
|---|---|---|---|---|---|
| SIC LuAG-S2 | 25×25×0.4 | 52.3 | 19.1 | 1531 | 51 |

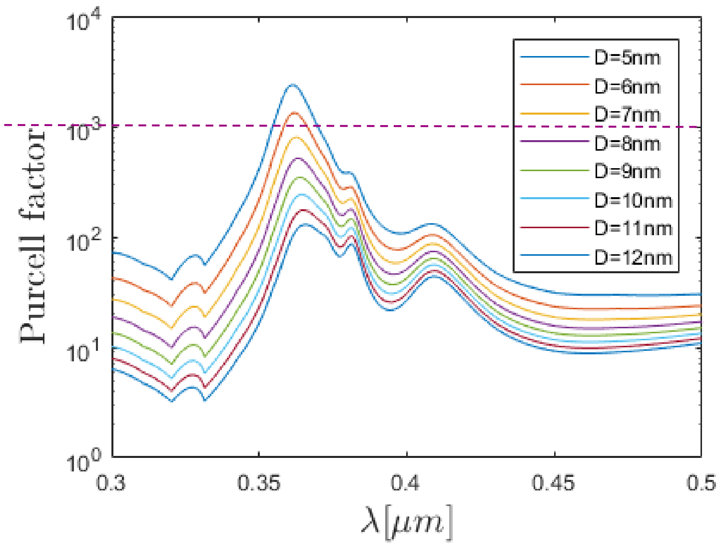
# Purcell Factor for Ag Particles




Particle size: R=20nm



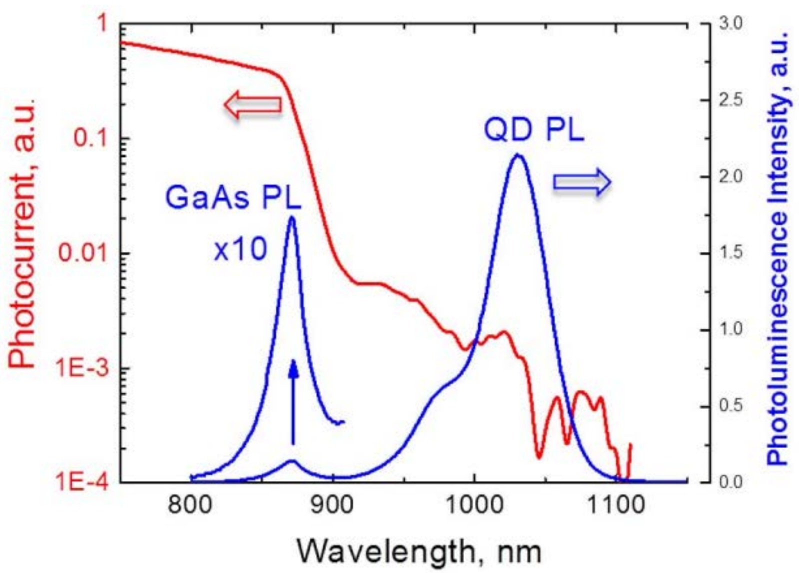

Particle size: R=50nm




- The Purcell factor is sensitive to the distance between ZnO and metallic nanoparticle.
- The bandwidth of Purcell factor is stable for variety of distance D.

# InAs/GaAs QD based Crystal

**In recent study[1], *S. Oktyabrsky, et al. report* an ultrafast, no self-absorption, high-efficient room-temperature semiconductor scintillator based on InAs QDs embedded in a GaAs matrix.**




**Room temperature photocurrent spectra overlapped with PL spectra of the same QD structure with reduced wetting layer placed in a p-n junction.**

**Comparison of Some Fast Inorganic Scintillators (source: Scintillator.lbl.gov) With Projected Performance of InAs/GaAs Qd Scintillator**

| Parameter | $BaF_2$ | LYSO | GaAs/InAs QDs |
|---|---|---|---|
| Density ($g/cm^3$) | 4.89 | 7.1 | 5.32 |
| Radiation length, cm | 2.03 | 1.1 | 2.3 |
| Decay constant, ns | 0.8 ns | 40 | 1 |
| Peak emission, nm | 195; 220 | 428 | 1050 |
| Photon Yield (photons/MeV) | 1,400 | 34,000 | 240,000 |
| Time between first photons, for 1MeV | 0.57ps | 1.2 ps | 2 fs |
| Poisson-limited energy resolution at 1MeV (keV) * | 62 | 13 | 4.8 |
| Radiation hardness, Gy | $10^4$-$10^5$ | $10^4$-$10^5$ | >$10^4$ |
| Coupling efficiency | <50% | <50% | ~100% |

*Assuming collection efficiency = 1

- **Due to inhomogeneous broadening of very narrow ground-level luminescence peaks, the self-absorption in such a medium is very low, <1 $cm^{-1}$ for $10^{15}$ QDs/$cm^3$ [1]**

*Ref.: [1] S. Oktyabrsky, et al., IEEE Trans. Nucl. Sci. 63, 656 (2016).*