R&D on GAGG single crystals for fast timing detectors in high-rate and radiation environments

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Gadolinium Aluminium Gallium Garnet

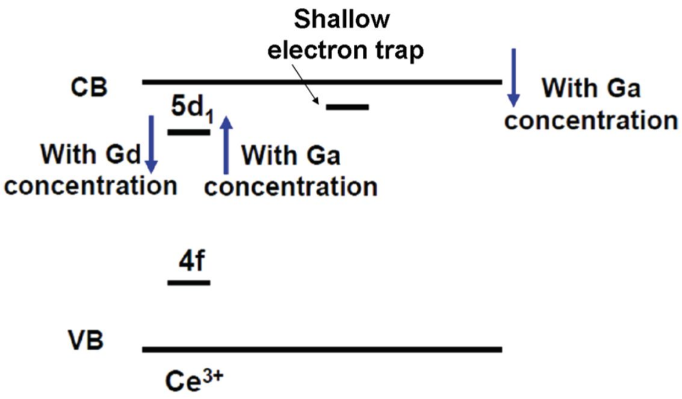
	light yield (photons/MeV)	first decay time (ns)	second decay time (ns)
$Gd_3Al_4Ga_1O_{12}$	15 895	316 (100%)	
$Gd_3Al_3Ga_2O_{12}$	**45 931**	**221 (100%)**	
$Gd_3Al_2Ga_3O_{12}$	**42 217**	**52.8 (73%)**	282 (27%)
$Gd_3Al_1Ga_4O_{12}$	17 912	42.2 (34%)	90.5 (66%)
$Gd_3Al_0Ga_5O_{12}$	0	*ND	*ND

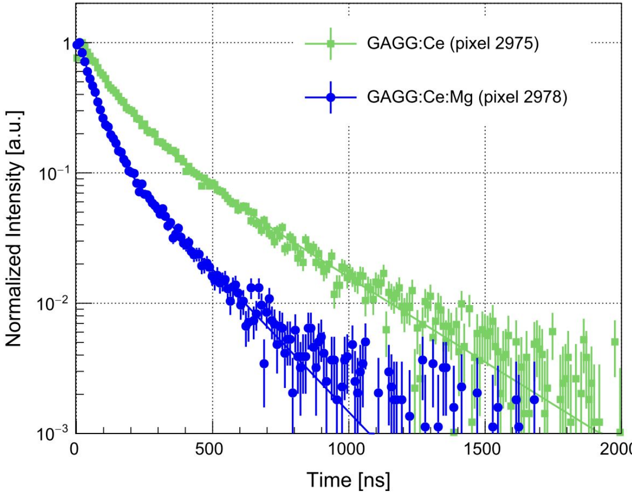
K. Kamada et al., Cryst. Growth Des. 2011, 11, 10, 4484–4490

Ce-doped multi-component garnets discovered in 2011. Amongst them is Gadolinium Gallium Aluminium Garnet $Gd_3Al_2Ga_3O_{12}$ (GAGG):

- **High light yield** and **fast scintillation**
- **Tunable composition**
  K. Kamada et al., Optical Materials 36 (2014) 1942–1945
- **Acceleration** of scintillation **via divalent ions codoping** (e.g. Magnesium)
  K. Kamada et al., Optical Materials 41 (2015) 63–66
  M. Lucchini et al., NIM A816 (2016) 176-183

Courtesy of K. Lebbou, ILM



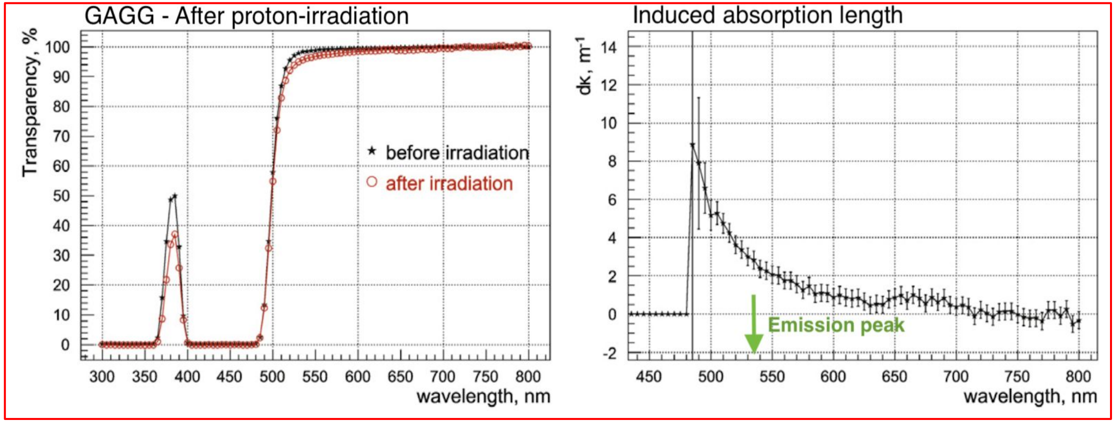


Radiation Hardness

Garnets are **radiation hard**

- **GAGG** irradiated w 3.1x10^{15} protons/cm^{-2} (24 GeV/c)
  - **Dose: 910 kGy**
  - Induced absorption below 4m^{-1} at the emission peak

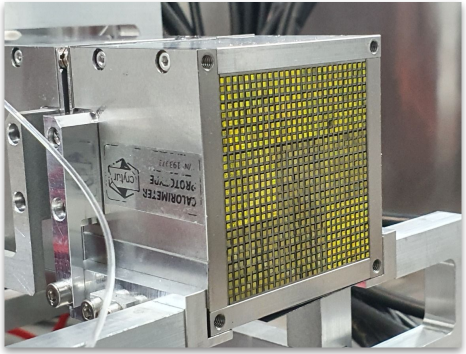
  See: *V. Alenkov et al., NIM A 816 (2016) 176*

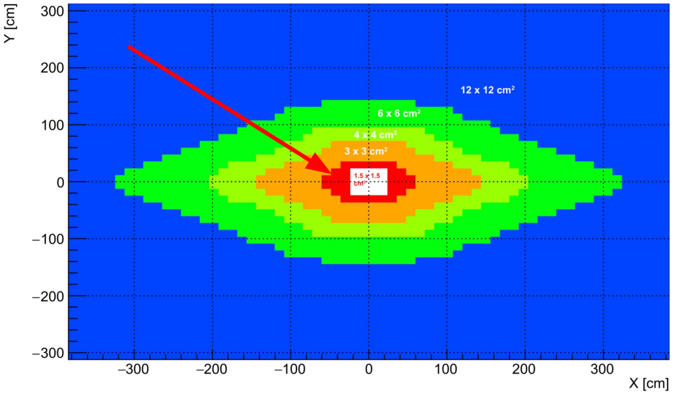


—> GAGG fibres are the baseline solution for the innermost region of the **LHCb PicoCal**

(see talk by E. Picatoste Tue 26)

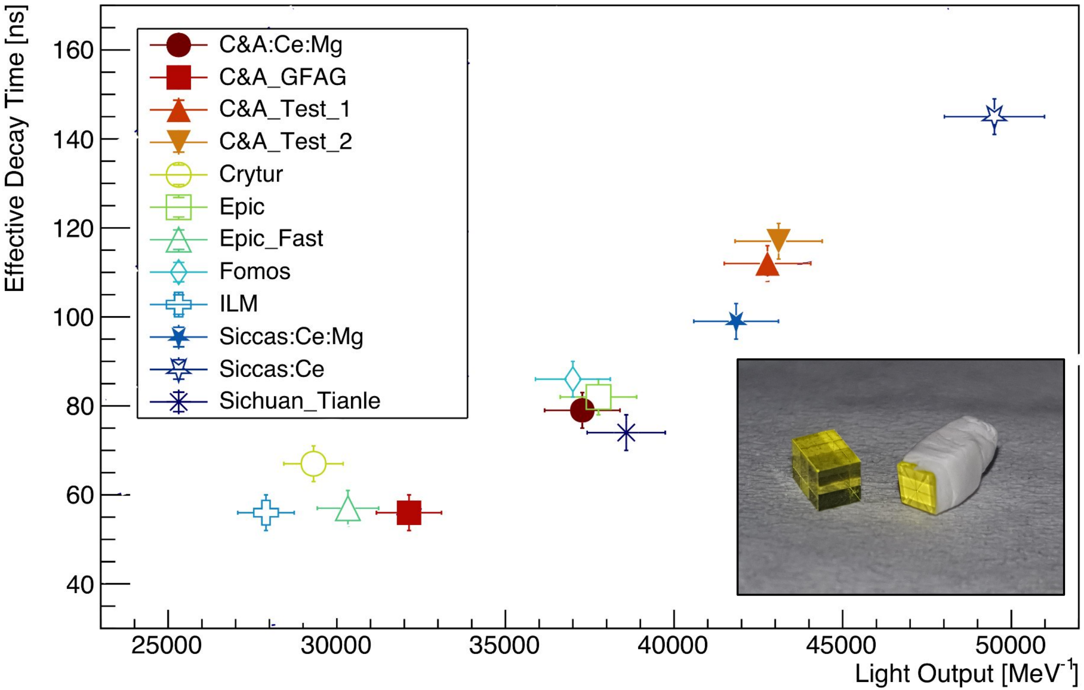
PicoCal





I. **Present GAGG**
II. Accelerating Scintillation

Light Output and Effective Decay Time

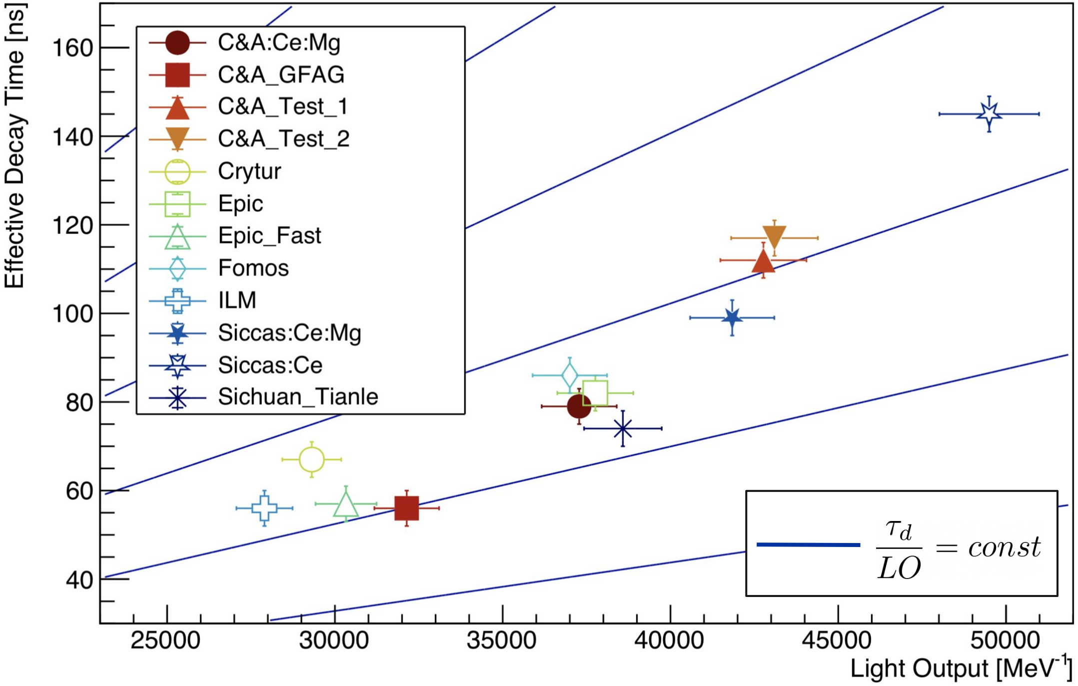


L. Martinazzoli, et al., NIM A, 2021, 165231

- Characterised GAGG of several producers
  - Light output
    from 30k to 50k photons/MeV
  - Decay times:
    from 50 to 150 ns
- Correlation between light output and decay time

High light yield → slow decay time

Light Output and Effective Decay Time



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$$\sigma_t \propto \sqrt{\frac{\tau_{d,eff}\ \tau_r}{LO}}$$

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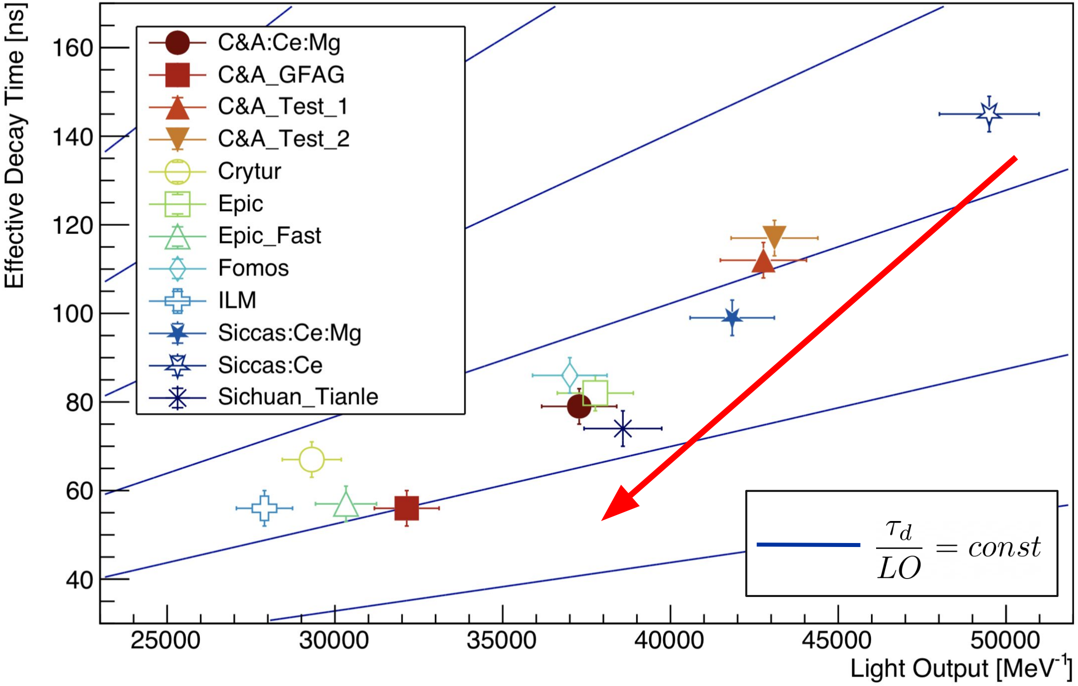

L. Martinazzoli, et al., NIM A, 2021, 165231

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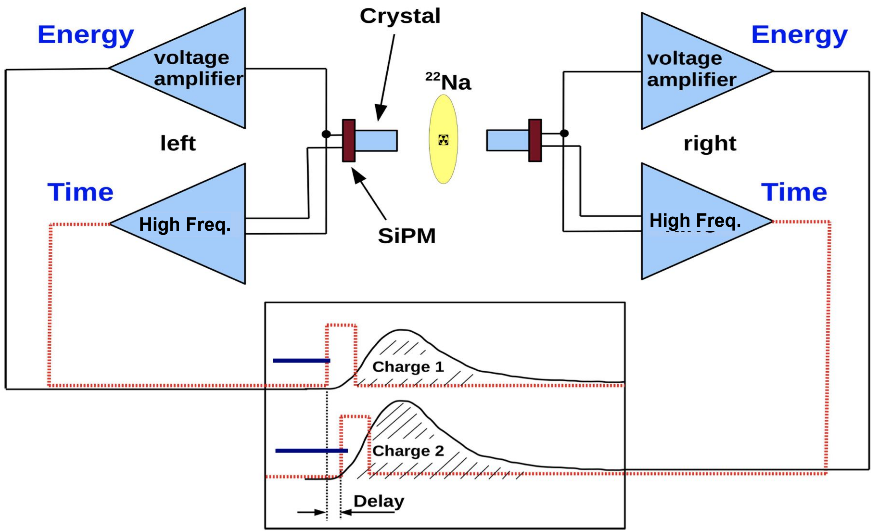
Faster samples have better timing

Coincidence Time Resolution



Adapted from S.Gundacker et al., 2013 JINST, 8, P07014

- Time resolution measured with two 511 keV photons in coincidence with ^{22}Na (CTR)
- High-Frequency amplifier (~1.5 GHz bandwidth) for timing

CTR of GAGG in line with LYSO:Ce

Crystal	CTR (HF) [ps] 2x2x3 mm^3
C&A GFAG	87 ± 2
ILM GAGG	90 ± 2
EPIC Fast GAGG	90 ± 2
LYSO:Ce	86 ± 2

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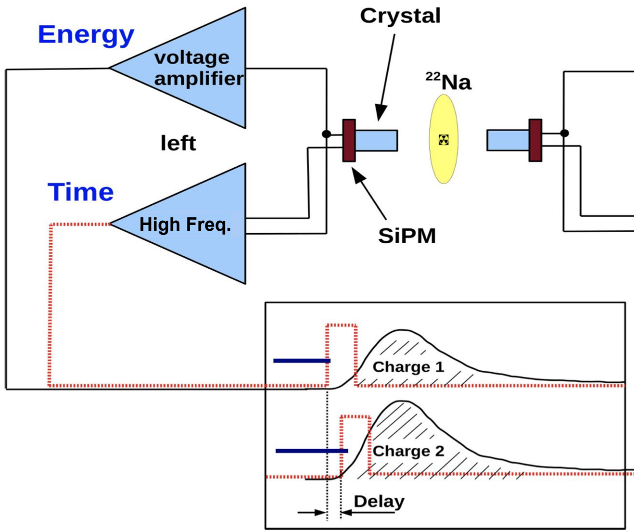
Coincidence Time Resolution



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However, decay time of ~ 50 ns (+ slow component)...

—> Pile-up at high rates!

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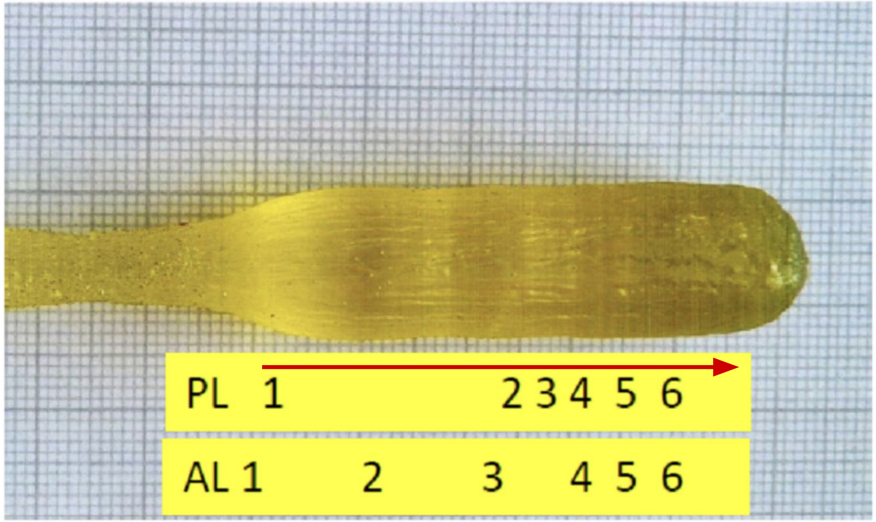
I. Present GAGG

II. Accelerating Scintillation

The Samples

L. Martinazzoli et al., Mater. Adv., 2022,3, 6842-6852



Samples produced from plates cut along the ingot

- AL to study composition
- PL to study luminescence

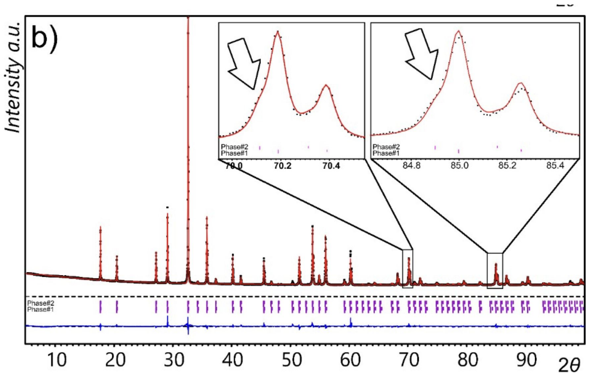
Along the ingot:

- Both Ce and Mg increasing
  - Up to 1 at%!
- Ga increasing

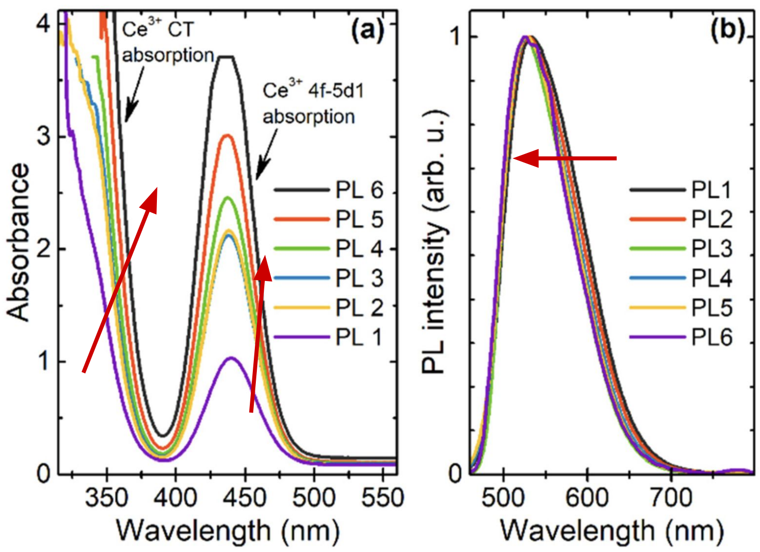
Rietveld analysis found 2 garnet phases (80%-20%)

- Possibly due to Al/Ga subclusters
  See: Phys. Status Solidi A 2018, 215, 1701034
- No perovskite phase

	g	Gd	Ce	Mg	Ga	Al
SC		2.955	0.015	0.03	3	2
AL1	0.065	2.917	0.0065	0.0051	2.787	2.284
AL2	0.155	2.944	0.0063	0.0053	2.813	2.231
AL3	0.416	2.978	0.0096	0.0058	2.845	2.160
AL4	0.618	2.954	0.0131	0.0039	3.043	1.986
AL5	0.703	2.959	0.0164	0.0060	3.108	1.982
AL6	0.789	2.952	0.0279	0.0112	3.251	1.758



Photoluminescence



1. Cerium increases in both Ce^{3+} and Ce^{4+} valence state
2. Emission spectrum blue-shifted due to Ga increase

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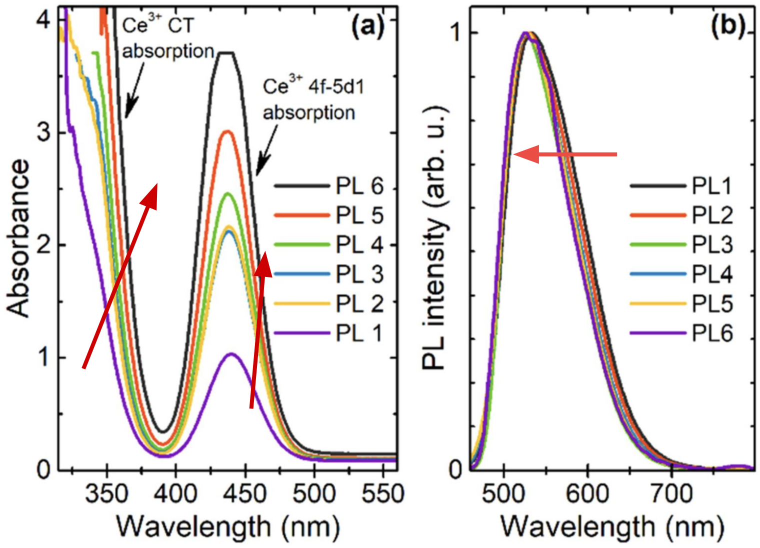
Photoluminescence



1. Cerium increases in both Ce^{3+} and Ce^{4+} valence state
2. Emission spectrum blue-shifted due to Ga increase
3. $5d^1$-4f photoluminescence accelerated by the Mg^{2+}

	g	Gd	Ce	Mg	Ga	Al
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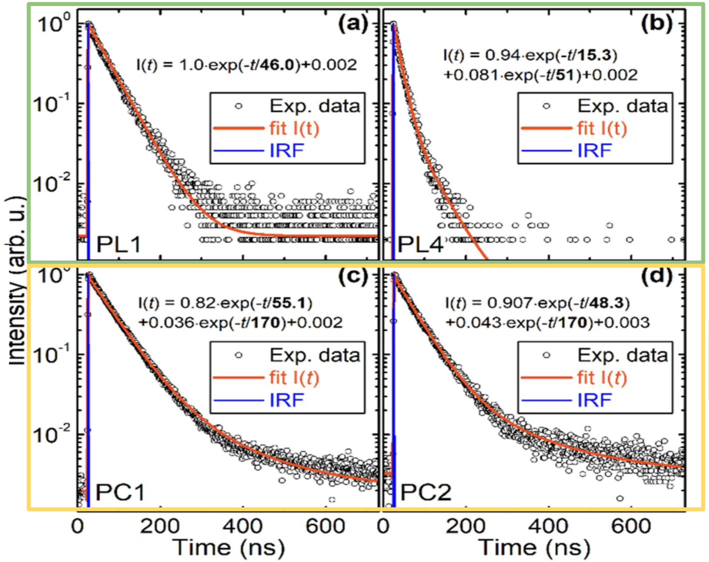
Increasing Ce quantity



with Mg^{2+}

without Mg^{2+}

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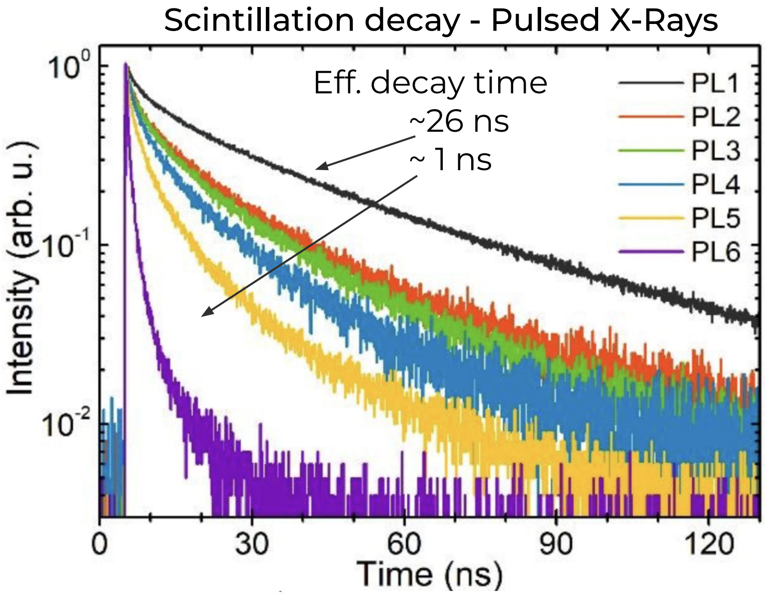
Scintillation



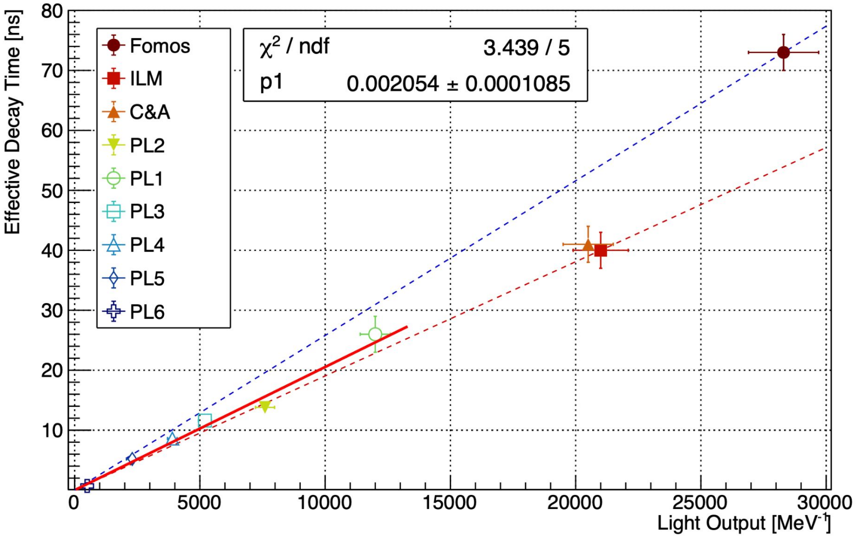
Samples of **1x1x5 mm³** used

Light quenching observed moving along the ingot:

- **Light output reduction** by > 10x
- **Decay time accelerated** by > 10x

Acceleration compensating the Light Yield reduction to maintain the same initial photon time-density

Sample	Light output (ph MeV^{-1})	Commercial sample	Light output (ph MeV^{-1})
PL1	12 000 ± 600	GFAG C&A	20 500 ± 1000
PL2	7600 ± 380	GAGG Fomos	28 300 ± 1400
PL3	5200 ± 260	GAGG ILM	21 000 ± 1100
PL4	3900 ± 190		
PL5	2300 ± 230		
PL6	500 ± 300		



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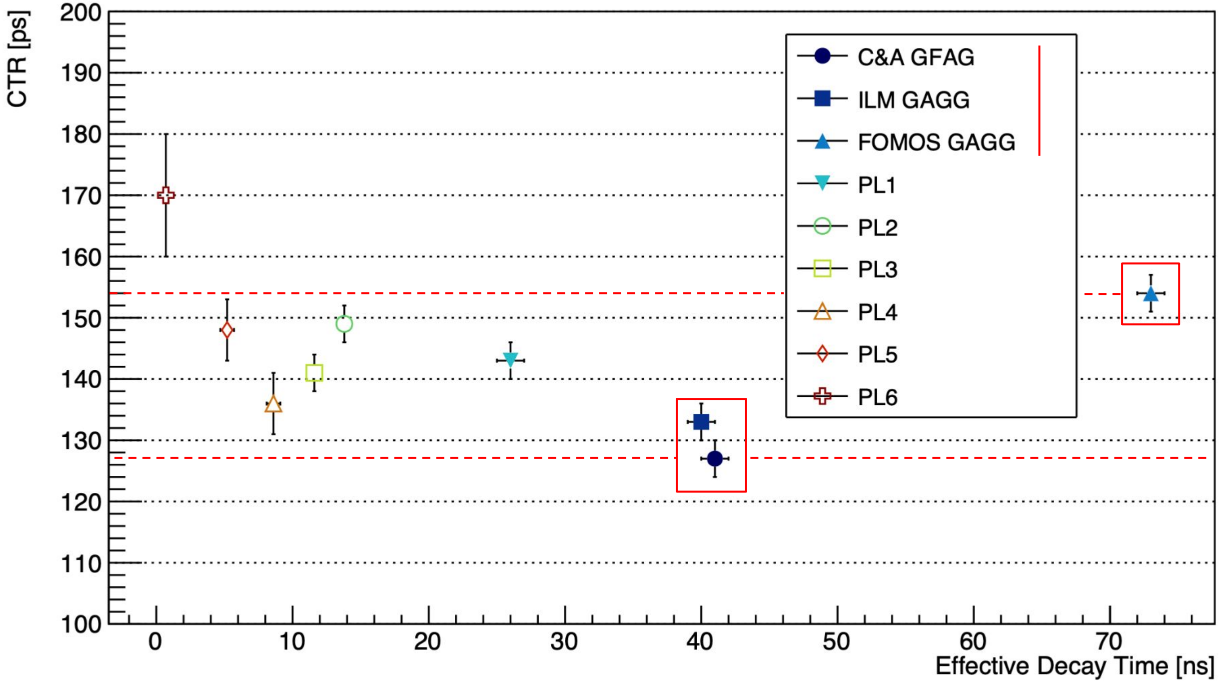
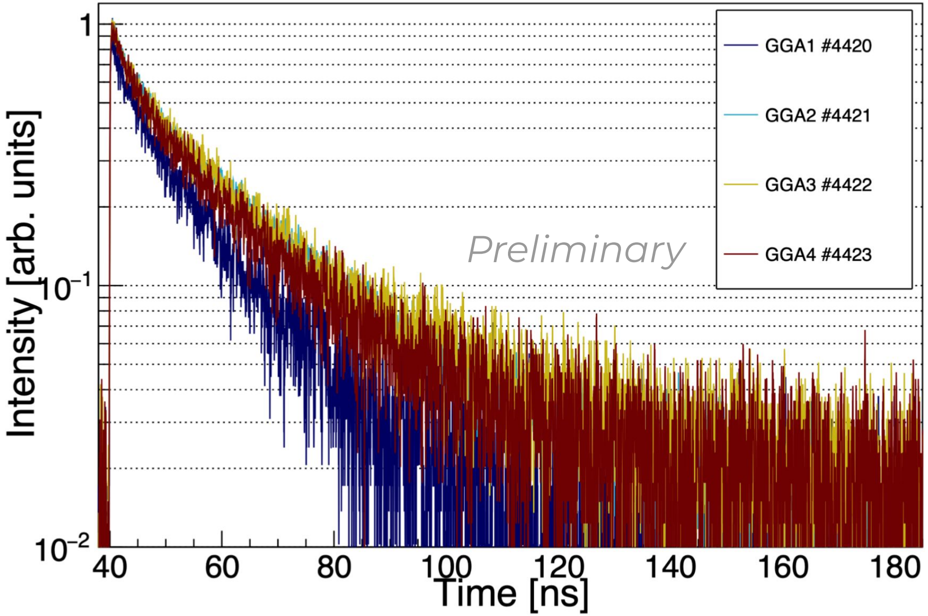
Timing



No major loss of time resolution wrt commercial samples

Note: different (worse) surface state for the new samples

New Samples

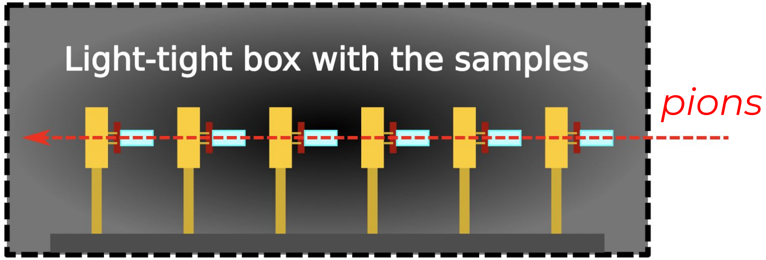


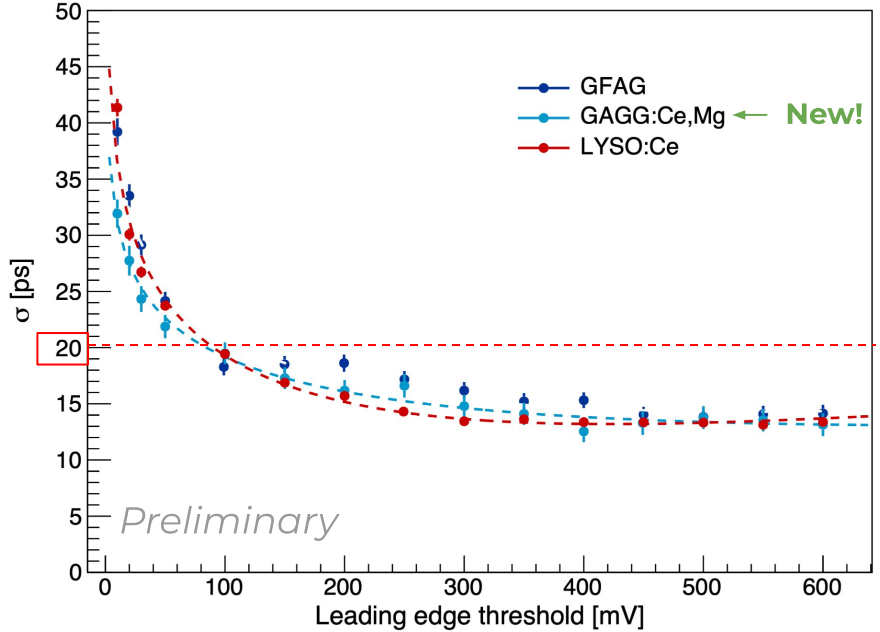
New and larger samples of **$2x2x10\ mm^3$** produced by the FZU institute, Prague.

- 4 samples with similar composition (and scintillation)
- Main decay time of 15-20 ns
- Slow component under study

Crystal	Photons Output [MeV^{-1}]	$\tau_{d,1}$ [ns]	%	$\tau_{d,2}$ [ns]	%	$\tau_{d,3}$ [ns]	%	$\tau_{d,eff}$ [ns]
Sample 1	6'200	3	10	15	66	99	24	12
Sample 2	7'400	3	8	19	67	77	25	16
Sample 3	7'400	3	7	18	66	82	27	15
Sample 4	6'200	3	10	17	67	80	23	14

Timing with MIPs





Tested the timing performance with 150 GeV/c pions at CERN SPS.

.

- MIPs travelling through 10 mm
- Crystals wrapped in Teflon and glued to a HPK S13360-3050PE SiPM

Time resolution compatible with:

- Fast Commercial GAGG (C&A GFAG)
- LYSO:Ce

see presentation of R. Calà on Friday 29.

Conclusions

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Cerium-doped **GAGG** is:

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- **fast and with high light output**
- **commercially available today**

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Decay time can be accelerated with heavy (co)-doping:

- **Heavy doping** achieved **up to ~1 at%**
  - Thanks to the cubic structure and the melting temperature of GAGG
  - No perovskite phase observed
- **Acceleration** of the scintillation **by >10x** paired by **reduction in light output**
- **Time resolution remains competitive**

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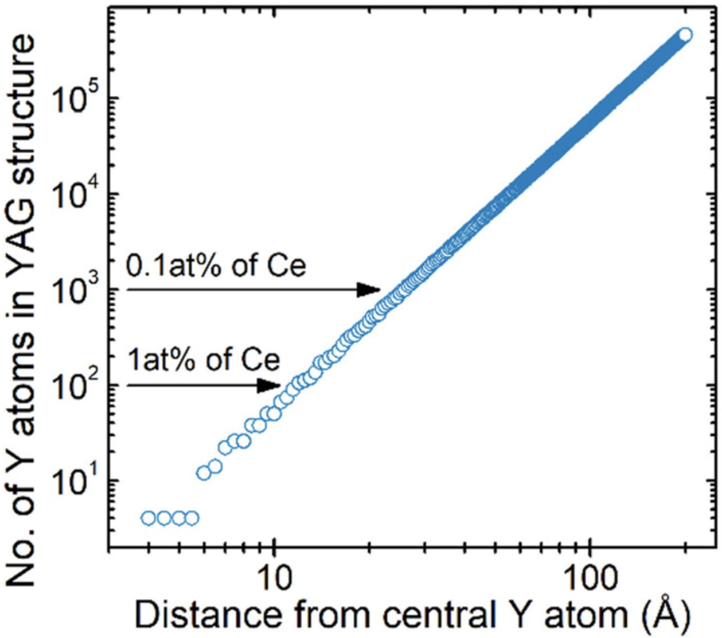
Ongoing R&D to develop large-size ingots with accelerated scintillation in:

- **Crystal Clear Collaboration** and the **TWISMA project**
  TWIN European project between ISMA, CERN, ILM Lyon (Grant agreement 101078960)
- Institutes taking part in the **LHCb PicoCal**

Back-up

Ce-Mg pairs



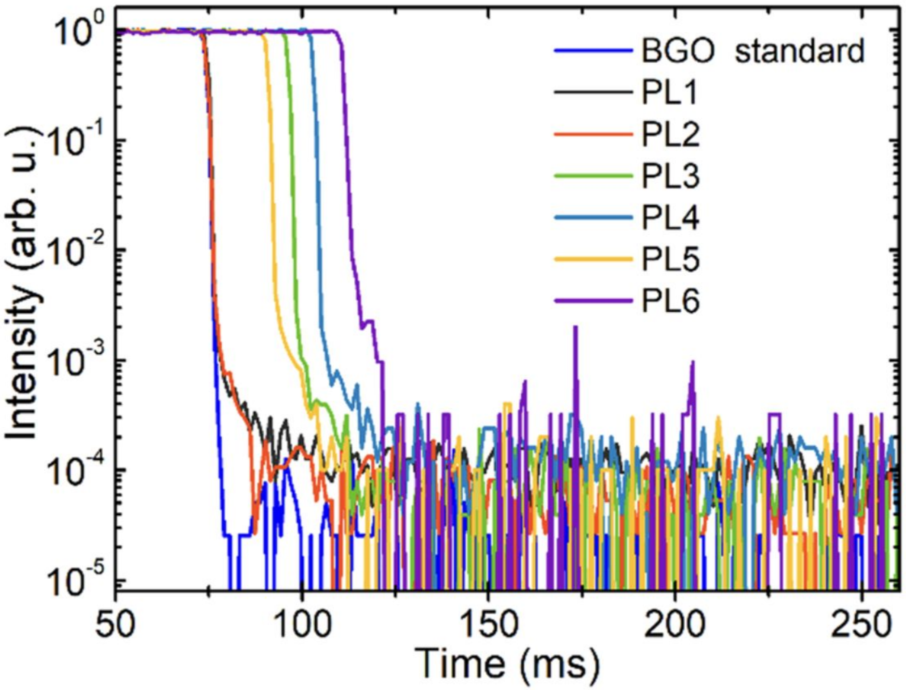
- Recent literature suggests that PL quenching in co-doped garnets is due to {Ce-Mg} pairs
  see:
  *Babin et al., Optical Materials **83** (2018) 290*
  *Babin et al., J. Lumin. **215** (2019) 116608*

- Doping of 1 at% reduces the distance of dopants at the level of the lattice parameter

- Different components explained by discrete available distances between dopants.

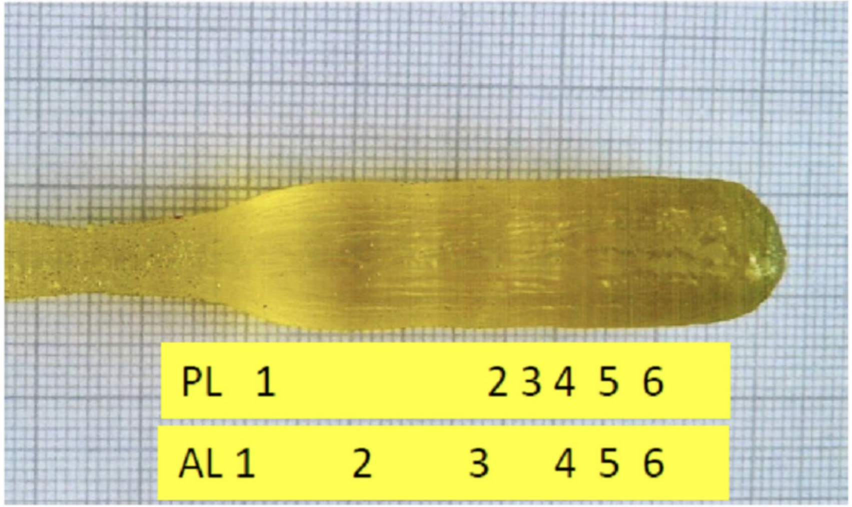
- Similar behaviour observed in CeF_3
  see: *M. Nikl, et al., J.Phys.Cond.Mat. 7, 6355-6364 (1995)*

Afterglow



BGO standard
PL1
PL2
PL3
PL4
PL5
PL6
Intensity (arb. u.)
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
50
100
150
200
250
Time (ms)

Ingot



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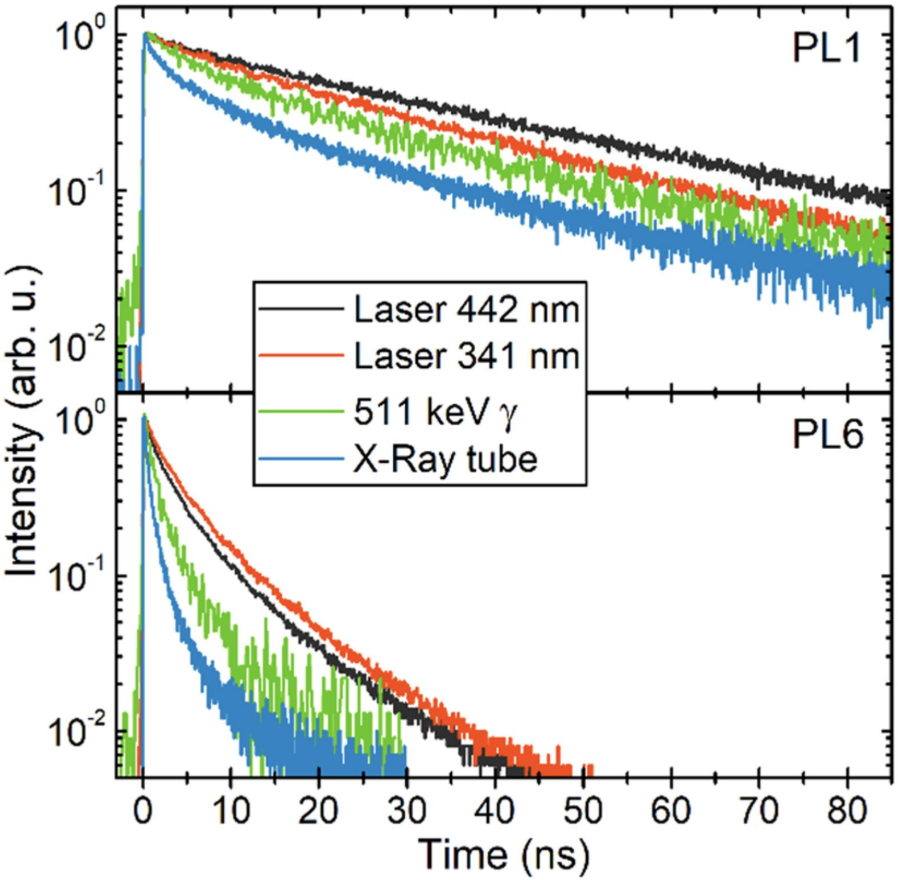
- Diameter: 12.5 mm
- Czochralski - Iridium crucible
- Atmosphere: N + 2% O
- Cyberstar Mini-oxypuller machine
- Elementary cell parameters, 12.28279(5) Å and 12.29436(13) Å, with their volume ratio of 0.8 : 0.2.

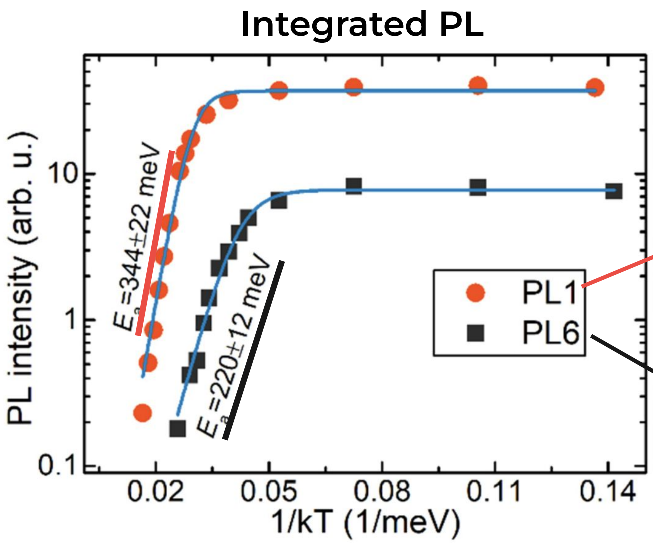
Kinetics Comparison

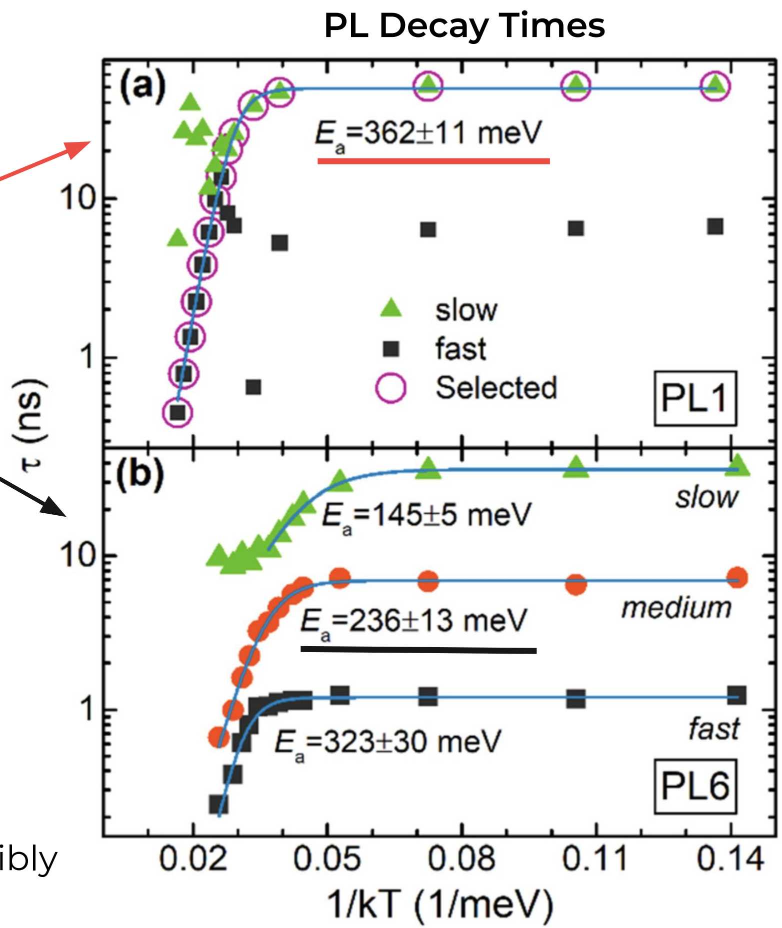


X-Rays Kinetics

Sample	τ_r (ps)	τ_{d1} (ns)	R_1 (%)	τ_{d2} (ns)	R_2%	τ_{d3} (ns)	R_3 (%)	$\tau_{d,eff}$ (ns)
C&A GFAG	32	6.0	4.6	44.5	69.2	222	26.3	41
ILM GAGG	37	4.0	3.2	40.4	56.4	138	40.4	40
Fomos GAGG	30	2.2	0.5	53.1	41.7	166	57.8	73
PL1	13	2.5	3.3	25.4	48.0	79.2	48.8	26
PL2	8	2.1	7.2	16.6	54.6	66.2	38.2	13.8
PL3	5	1.6	6.2	12.6	47.5	46.0	46.3	11.6
PL4	5	1.5	9.2	11.3	53.9	45.4	36.9	8.6
PL5	5	1.0	11.0	7.1	51.8	40.8	37.2	5.2
PL6	5	0.2	19.5	1.5	53.0	14.9	27.5	0.7

Temperature dependence of Photoluminescence

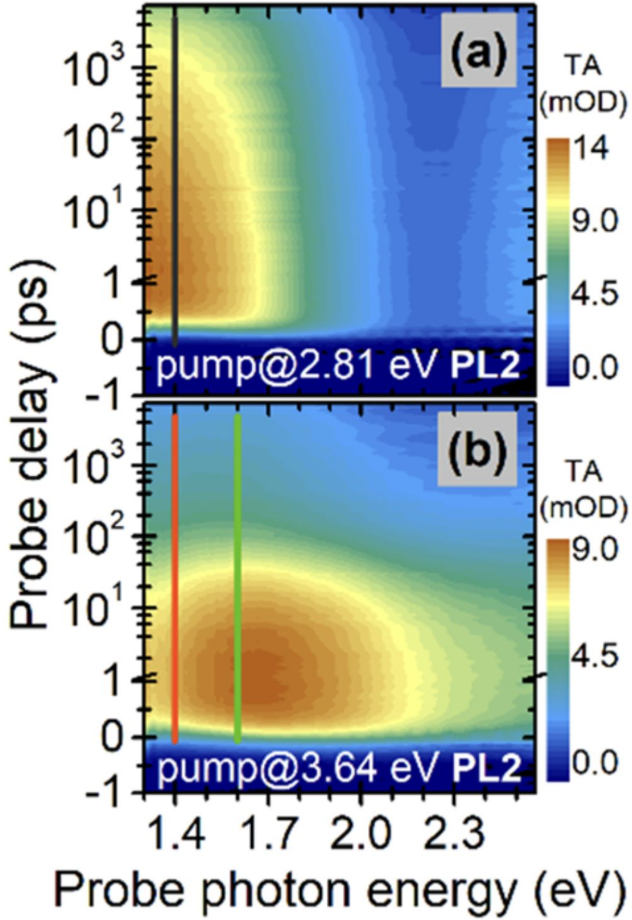




- Integrated PL shows **decreasing energy barrier** for light quenching
  - Same energy barrier for the PL decay time of the main component

- Up to **3 components** visible at the highest doping
  - Different energies for different components, possibly due to discrete distances of the Ce-Mg pairs

Transient Absorption



Studied non-equilibrium absorption bands:

- Pump 4f-5d[1] - 2.8 eV (a) :
  - absorption band in line with literature for GAGG

    see: *G. Tamulaitis et al., J. Appl. Phys. 124, 215907 (2018)*
    and Keynote presentation on Wednesday by G. Tamulaitis
- Pump 4f-5d[2] - 3.6 eV (b):
  - new **absorption band peaking at 1.7 eV**
  - new band extending towards higher eV increasing dopants

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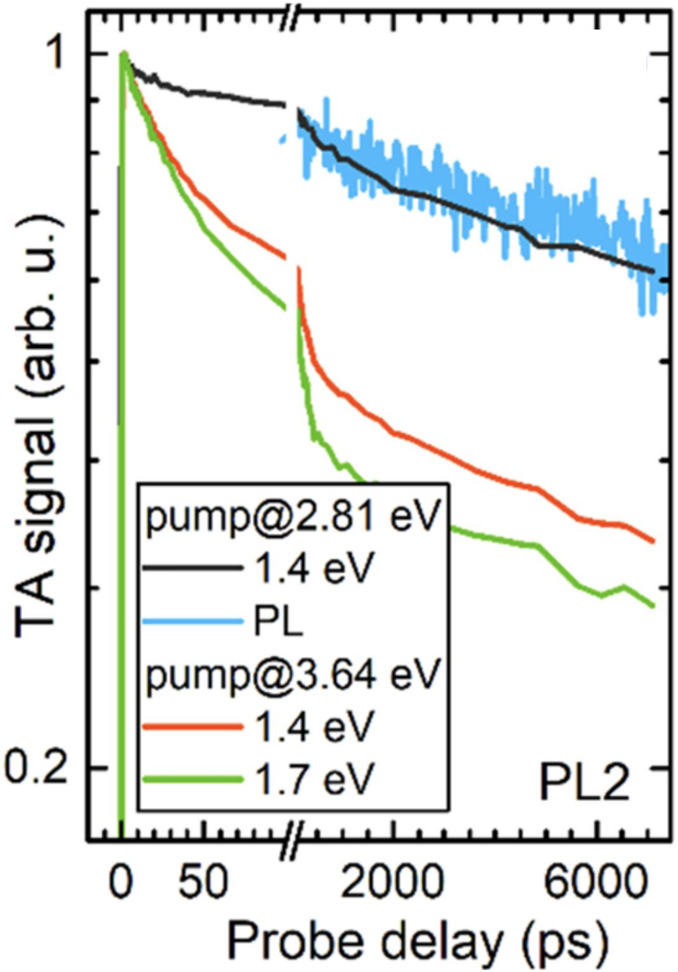

Studied non-equilibrium absorption bands:

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- Pump 4f-5d^2 - 3.6 eV (b):
  - new **absorption band peaking at 1.7 eV**
  - new band extending towards higher eV increasing dopants

- Non-exponential decay of electrons in 5d^1 and 5d^2 confirms **non-radiative recombination channels**

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