WP8
Innovative calorimeters with optical readout
Task 8.3.1:
R&D on Crystal and nanomaterial scintillators

E. Auffray, CERN EP_CMX
on behalf of Sub task 8.3.1 team

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 101004761.

CERN FZU Fyzikální ústav Akademie věd České republiky · Research Institute for Nuclear Problems BSU · INFN LNF · INFN TORINO · INFN PERUGIA · crytur · GLASS to POWER

Task 8.3.1: Partner Institutes

- **Main Benificiaries:**
  - CERN: E. Auffray, N. Kratochwil with all CERN CCC team
  - FZU Prague: M. Nikl, V. Babin, J. Vitezslav,
  - INFN Frascati: M. Moulson, L. Bandiera,
  - INFN Perugia: C. Cecchi, G. De Nardo, E. Manoni, M. Merola
  - INFN Torino: N. Pastrone, I. Sarra
  - Vilnius: G. Tamulaitis, S. Nargelas, A. Vaitkevicius
- **Associated partners:**
  - CERN
    - *Minsk: M. Korzhik*
  - INFN
    - GlassToPower, Italy (company): S. Brovelli,
- **Other partner:**
    - Crytur, Czech republic (company): J. Houzvicka, S. Sykorova

Task8.3.1: Objectives

- Optimisation of crystal materials and processes for fast timing applications in radiation environments
- Industrialisation of the production process of fast and radiation-hard crystals

Task activities

- **Material investigation**
  - Optimisation of “standard” scintillating Materials to improve ti… performance:
    - Garnet, PWO, BGO
    - Study of crossluminescence materials
  - Study of scintillating nanomaterials
- **Development of instrumentation for timing char…sation**
  - Transient absorption
  - Time resolved spectroscopy
  - Time resolution

  => Milestone achieved in Mar… 2 ((see link)
- **Test beam study:**
  - Timing performan… mip and electron
  - Test of various prot…ypes for future calorimeter
  - Test Nanocal prototype (bluesky project)

Not too much news, since meeting January (see link)

Deliverable D8.2 submitted 28 feb2024

Milestones & Deliverables

Milestone:

MS32 (due to M12) **:** Test benches for testing detecting materials in picosecond and subpicosecond domains. Achieved in March 2022 (see link)

Deliverable

D8.2 (due to M35 end of February 2024)**:** Report on prototypes construction, performance and assessment of industrialization

New materials enabling high precision timing will be selected by means of test bench measurement and assembled into a prototype that will allow also the assessment of industrialisation for realistic detector systems

=> Report submitted 28 Feb2024

AIDA innova

Deliverable 8.2

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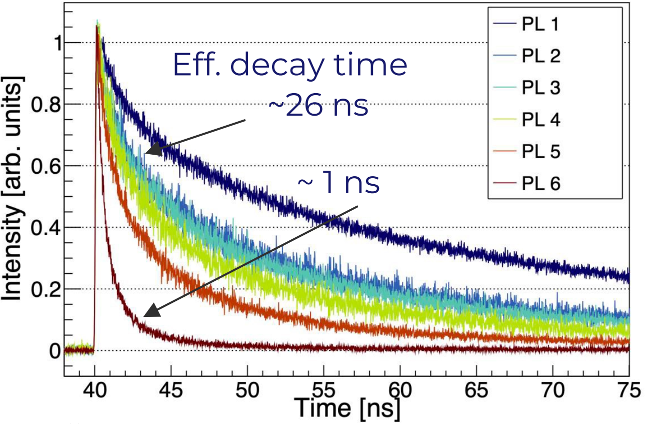
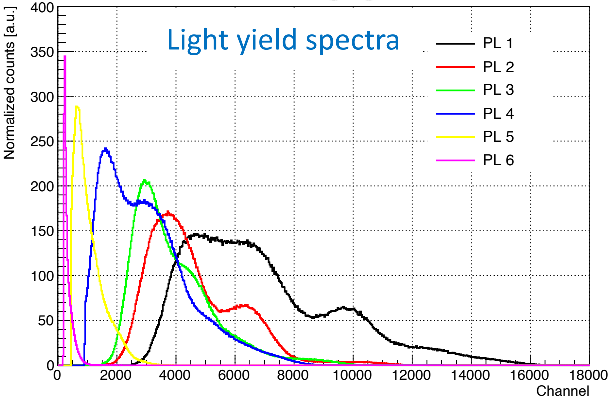
Task activities update

- **Material investigation**
  - Investigation of garnet materials:
    - ⇒ Strong reduction of decay time observed on GAGG with highly codoping Ce, Mg
      - ⇒ Crytur company starts to grow ingots
      - ⇒ CERN group has also collaboration with ILM, Lyon and ISMA, Kharkiv through a Twin project TWISMA (GA 101078960)
    - ⇒ Investigation of other garnets in Vilnius
  - Deep understanding of Ultra fast PWO: PWO-UF
    - ⇒ Vilnius and FZU
    - ⇒ Material produced by Crytur
    - ⇒ Test by INFN in the frame of R&D on CRILIN and INFN OREO (ORiEnted calOrimeter)
  - Nanocomposite scintillators based on nanomaterials
    - ⇒ Development by GlasstoPower & UNIMIB
    - ⇒ Characterisation at CERN and at Frascatti
      - ⇒ CERN, FZU , UNIMIB have a new pathfinder project UNICORN (GA 101098649) on development of nanocomposite
    - ⇒ Test in beam through Nanocal project (see today presentation M. Moulson in WP13 meeting)

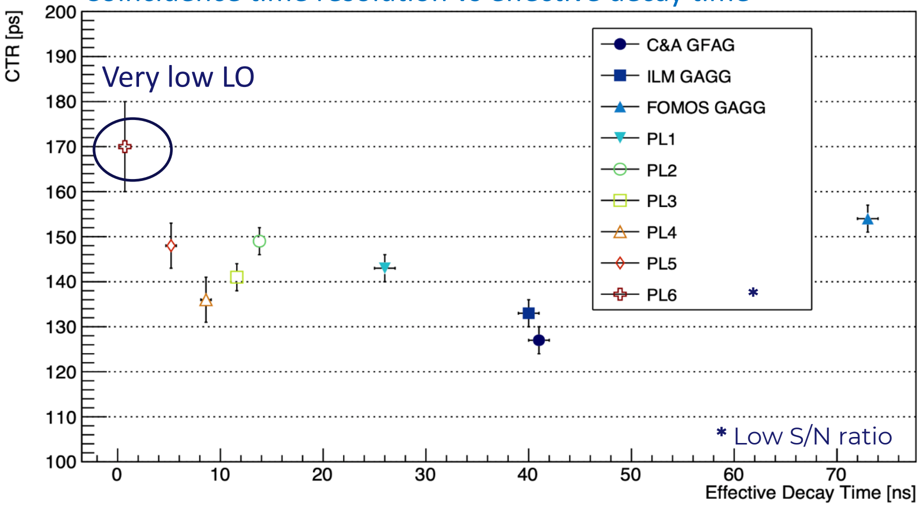
CERN, FZU; VILNIUS: GAGG:Ce Scintillation acceleration by heavy Ce/Mg doping

Decay time spectra





Coincidence time resolution vs effective decay time



No major loss of time resolution!
Decay time decrease compensated the Light output reduction
=> the same photon time-density

Joint paper CERN; FZU; Vilnius, L. Martinazzoli et al, Mater. Adv., 2022,3, 6842-6852 .

Technology transfer of heavily doped GAGG:Ce,Mg From FZU to CRYTUR

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Samples from the seed and end part of crystal ∅10x1mm

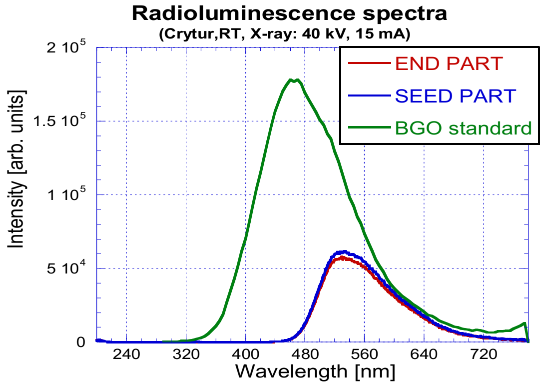
From about 20 growth experiments of GAGG:Ce,Mg in FZU the starting composition of the melt was defined with the goal to reach 1/e decay time below 10 ns

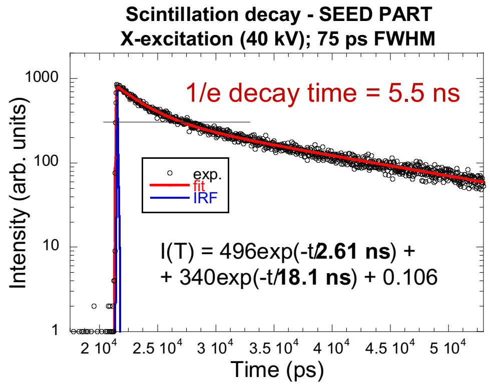
=> technology transfer

Industrial Czochralski growth in CRYTUR was adapted

=> able to grow first ingot: ∅ 25mm,

First task was to obtain **HOMOGENEOUS COMPOSITION AND CHARACTERISTICS** within all the crystal body



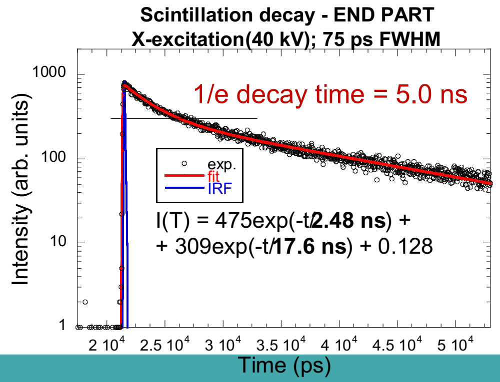


SUCCESS!!!

RL intensity, 1/e decay time and LY (~5000 ph/MeV] values are within 10% difference at the seed and end part of the crystal.

Fibers of ~50mm length can be cut!

=> First step to large production !



Technology transfer of heavily doped GAGG:Ce,Mg From FZU to CRYTUR

FZU Fyzikální ústav Akademie věd České republiky

crytur

CRYSTAL CLEAR

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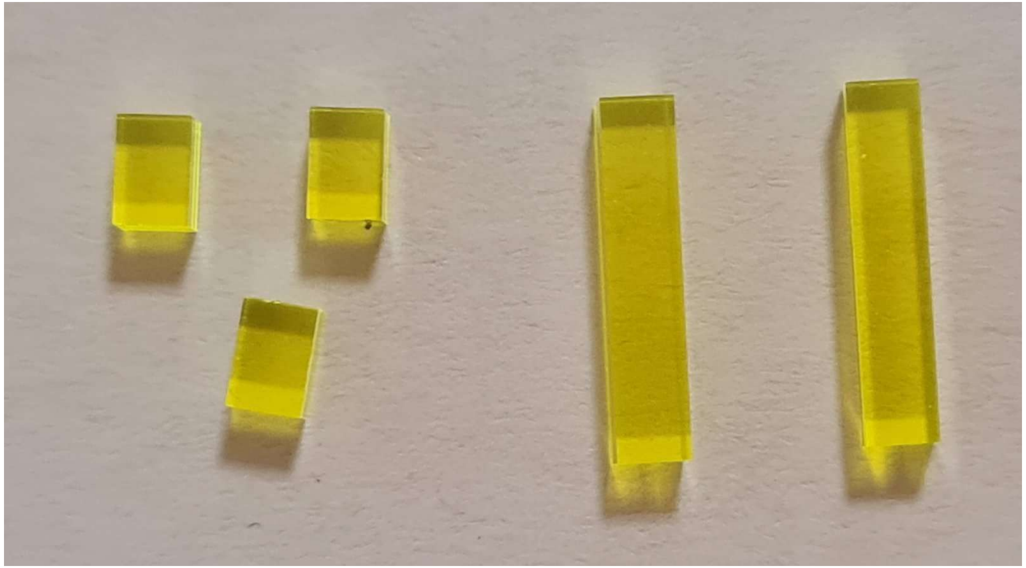
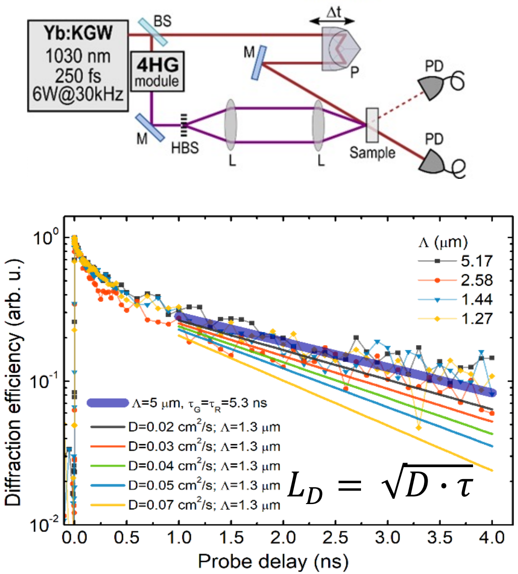
GAGG Samples produced by CRYTUR

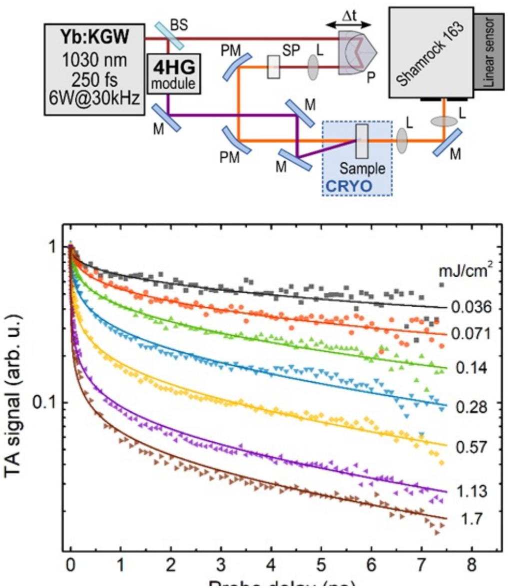


Self-activated scintillators. PWO



The estimated upper limit of the diffusion length is 100 nm.

- The emission is caused by excitons self-trapped at oxyanions WO_4^{2-};
- at high La, Y doping, the luminescence decay is nonexponential due to inequivalent surrounding of WO_4^{2-};
- Auger-type nonradiative recombination becomes important at high excitation intensities;
- the time constant of scintillation decay can be decreased down to, at least, of 640 ps, however, at a light yield down to 5 phe/MeV

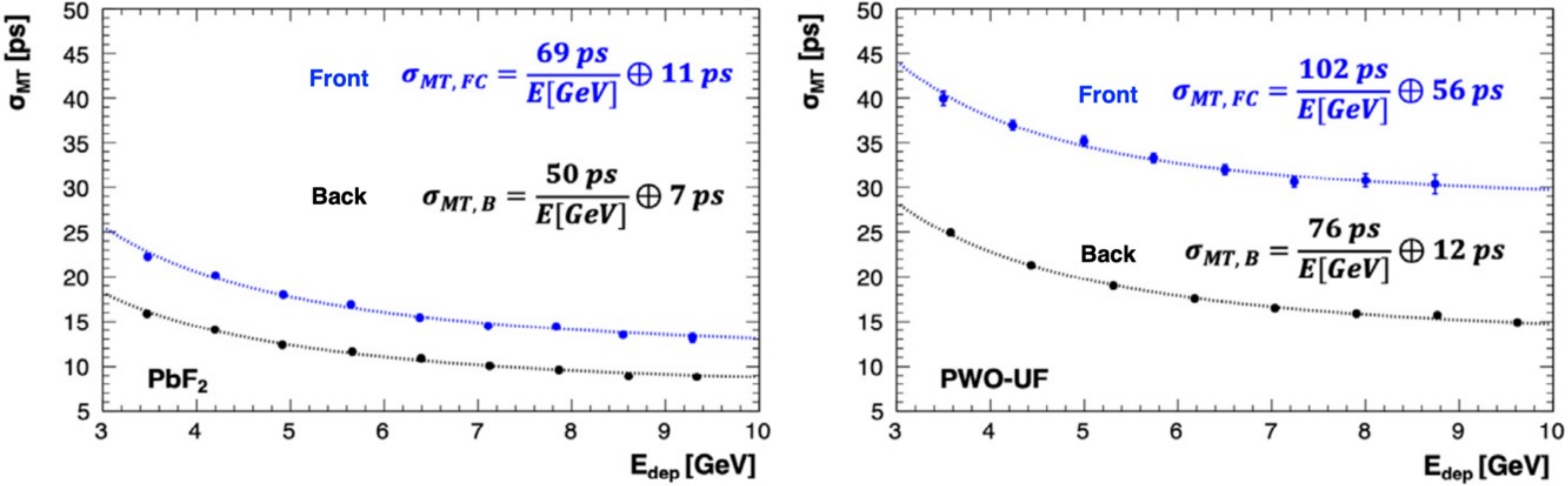


Lines are calculated by dipol-dipol interaction taken into account.

⇒ The dipole-dipole interaction might be an important mechanism causing the fast initial decay of the population of radiative centers in PWO observed experimentally .

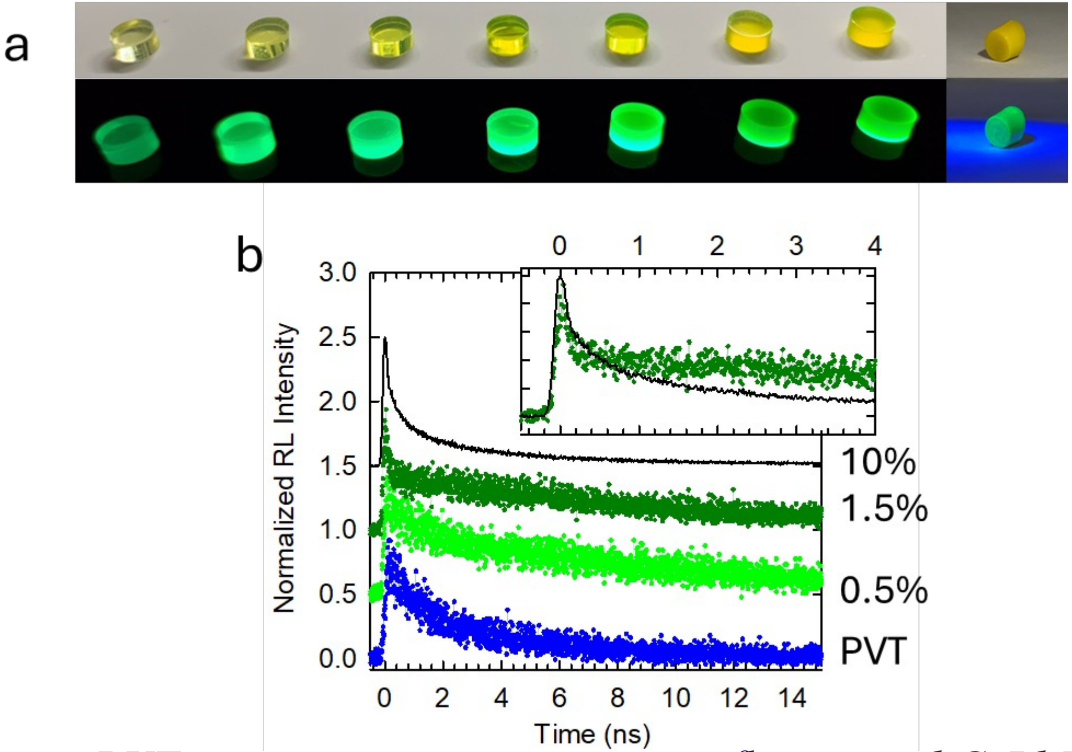
G. Tamulaitis, et al., J. Materials Chemistry C, 10, 9521 (2022)

Test of PWO-UF in beam test



Time resolution obtained with single 10 × 10 × 40 mm² crystals of PbF_2 (left) and PWO-F (right) as a function of deposited energy, from beam tests with high energy electrons. The blue (black) curves are for data taken in "front" ("back") configurations.

Nanomaterial development: Challenge



PVT nanocomposites containing fluorinated $CsPbBr_3$ NCs (concentration 0.01-10% left to right)

GLASS to POWER

Pushing the concentration of NCs in composites

Increasing the Light Yield (especially for gamma) requires higher densities.

→ HOW CAN WE BOOST THE NC LOADING IN POLYMERS?
→ HOW CAN WE TAKE ADVANT

Good news is,
we actually have $CsPbBr_3$ NCs that survive T>100K

Effective embedding in plastics

Two problems:

1. Conventional LHP NCs do not withstand thermal polymerization due to thermal damage

→ incompatible with polystirene derivatives

2. Photopolymerization of acrylates is self limited in [NC] due to competitive absorption of UV light (→ cheese toast effect)

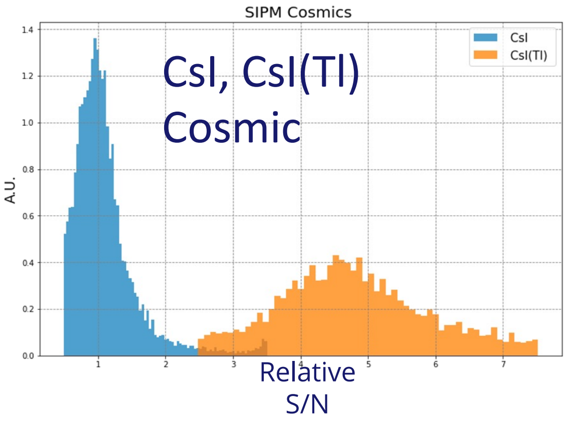
→ Impossible to go above 1% with conventional free radical routes.

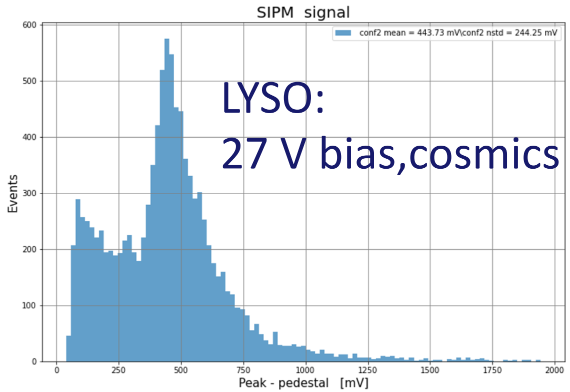
Good news is,
Free radical polymerization is not the only option available…

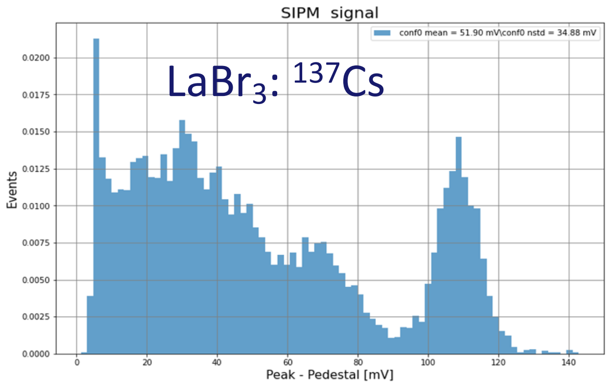
GLASS to POWER

UNIVERSITÀ DEGLI STUDI DI MILANO BICOCCA

Belle II upgrade: various materials readout with SiPM







See presentation C. Cecchi today

Test beam activities

Task Beam activities

- **Many Test beams in 2023:**

  - Timing performance with mip and electron by CERN group in June 23

  => Test many scintillators,

  - Test of various prototypes for future calorimeter CRILIN; Klever, HIKE June, August, October 23

  => PbF2, Ultra fast PWO, oriented PWO (INFN groups)

  - Test Nanocal prototypes (bluesky project) June 23, Nov23

  ⇒Different types of of material tested (standard plastic scintillator and nanocomposites based on $CsPbBr_3$ and $CsPb(Br,Cl)_3$

CERN: Test beam in sps: Assesment of timing performance under MIP

August 2021:

Readout performed with NINO ASIC electronics

September 2022:

Custom high frequency SiPMs readout

6 crystals measured in a row

150 GeV charged pion beam

Pulses were recorded for offline analysis

June 2023:

Same readout chain of 2022 test beam

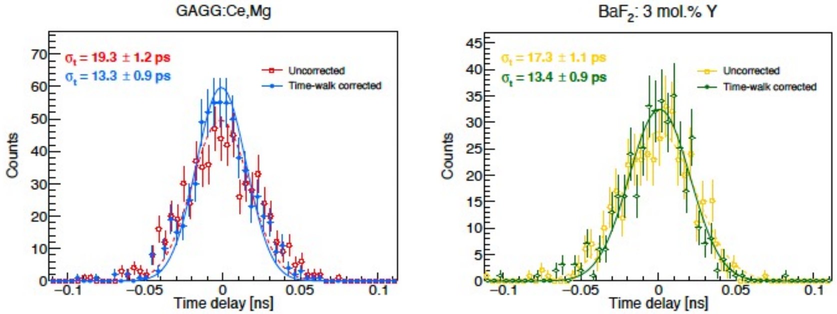
Up to 5 crystals measured in a row

150 GeV charged pion beam

Temperature stabilization system implemented

Crystal	Time resolution σ_t (ps) Sept. 2022 TB	Time resolution σ_t (ps) June 2023 TB	Energy deposited (MeV)
GFAG	14.3 ± 0.6	14.1 ± 0.8	9.5
GAGG:Ce,Mg - 1	16.4 ± 0.9	-	9.5
GAGG:Ce,Mg - 2	-	13.3 ± 0.9	9.5
BGO	36.4 ± 1.5	37.9 ± 0.9	9.9
BGSO	31.1 ± 0.5	32.9 ± 1.7	9.9
BSO	-	35.7 ± 1.3	9.9
BaF_2 *	15.8 ± 0.6	14.3 ± 0.6	6.7
BaF_2:Y *	17.0 ± 0.4	13.4 ± 0.9	6.7

* Sept. 2022 TB: $3 \times 3 \times 10\,mm^3$ samples measured with $6 \times 6\,mm^2$ VUV-SiPMs.
June 2023 TB: $2 \times 2 \times 10\,mm^3$ samples measured with $3 \times 3\,mm^2$ VUV-SiPMs.



From R. Cala' Sienna Conference Sept23

INFN Frascatti, Torino, Napoli: Test beam activities for CRILIN & Klever

R&D for future experiments to develop new concepts for crystal calorimetry

KLEVER small-angle calorimeter (SAC)

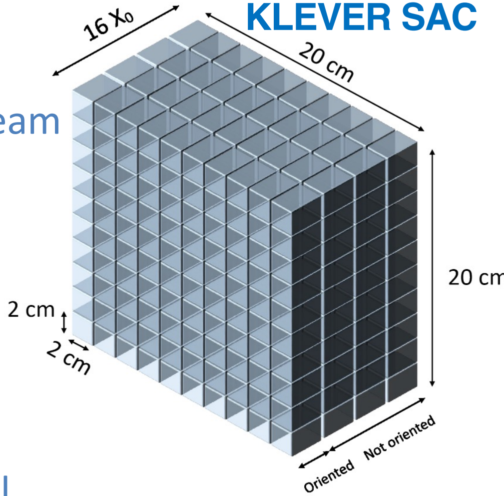
- Operates inside neutral beam, rejects γs from KL → π0π0 escaping through beam hole
- Low-sensitivity for > 400 MHz of beam neutrons
- Possibilities for γ/n discrimination: multilayer structure/longitudinal segmentation

ECAL for Muon Collider (CRILIN design)

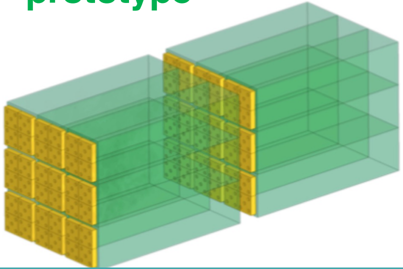
- Resolves jet substructure: fine segmentation, both transverse and longitudinal
- Rejects of beam-induced background from low-energy shower particles
- Good sensitivity for low-energy-release topologies (e.g. signal muons)
- Excellent time resolution to allow rejection of background hits at cell level

Crystals under consideration fast PbF2 or Ultra Fast (UF) PWO

Concept for KLEVER SAC



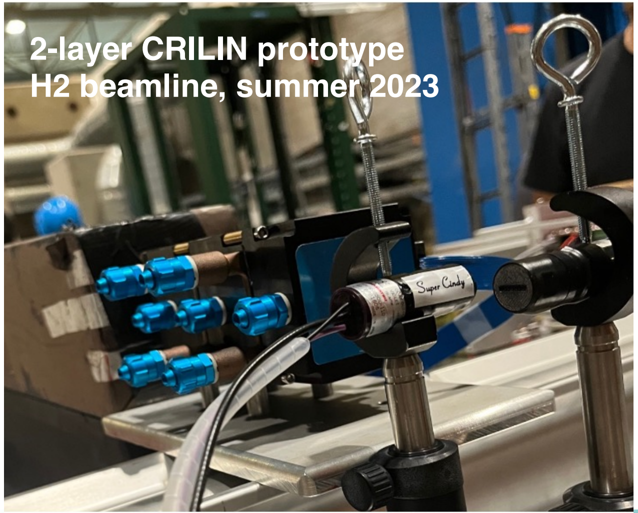
Concept for CRILIN prototype

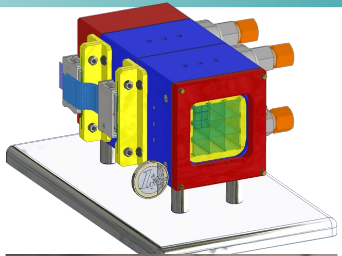


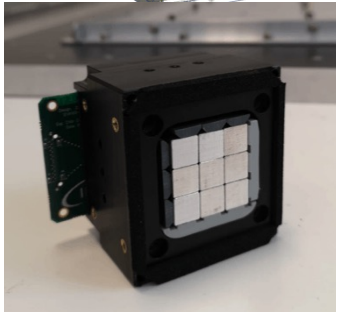
INFN Frascatti, Torino, Napoli: Test beam activities for CRILIN & Klever

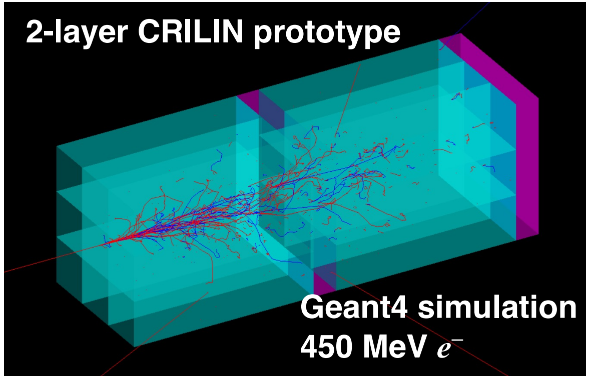
Collaboration with CRILIN to study:

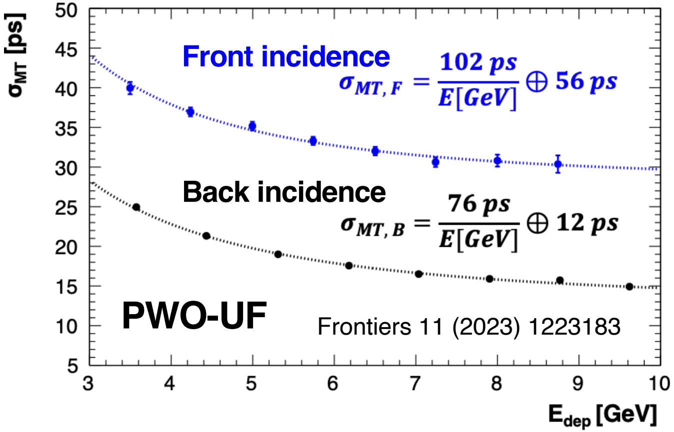
- Materials: PbF_2 vs PWO-UF
- Radiation resistance of crystals
- Photosensors: SiPMs, front-end
- Light collection in small crystals
- Longitudinal segmentation
- Mechanics, cooling, integration











For single crystals of PWO-UF: $\sigma_t < 20$ **ps** for $E_{dep} > 5$ GeV!

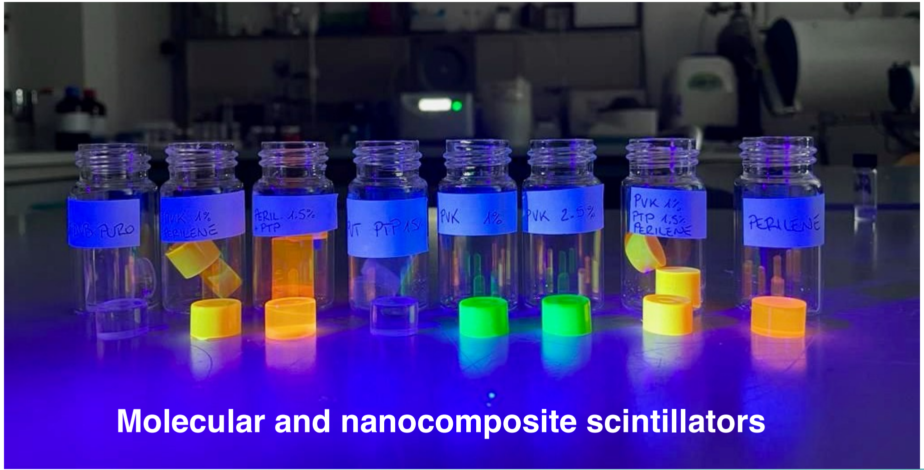
AIDA innova   GLASS to POWER   INFN Istituto Nazionale di Fisica Nucleare

TB with nanocomposite scintillators Nanocal

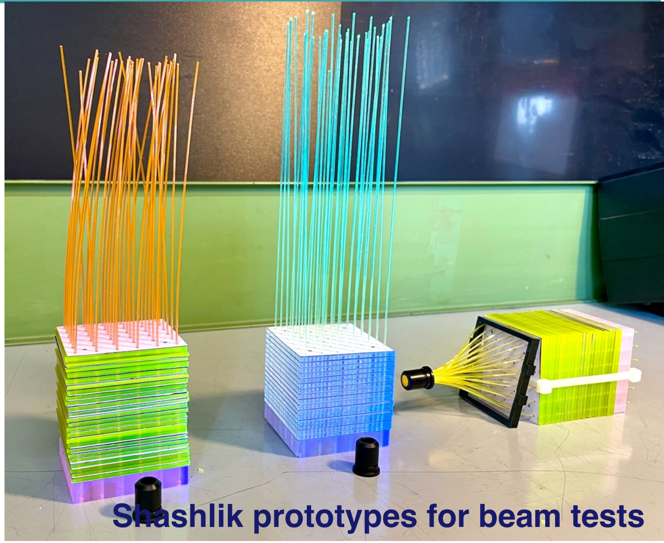
Use of nanocomposite scintillators under investigation in collaboration with AIDAinnova project NanoCal

Semiconductor nanostructures used as sensitizers/emitters for ultrafast, robust scintillators:

- Perovskite (typically $CsPbX_3$, X = Br, Cl…) nanocrystals cast into polymer matrix
- Decay components << 1 ns
- Radiation hard to O(1 MGy)



Molecular and nanocomposite scintillators


Shashlik prototypes for beam tests

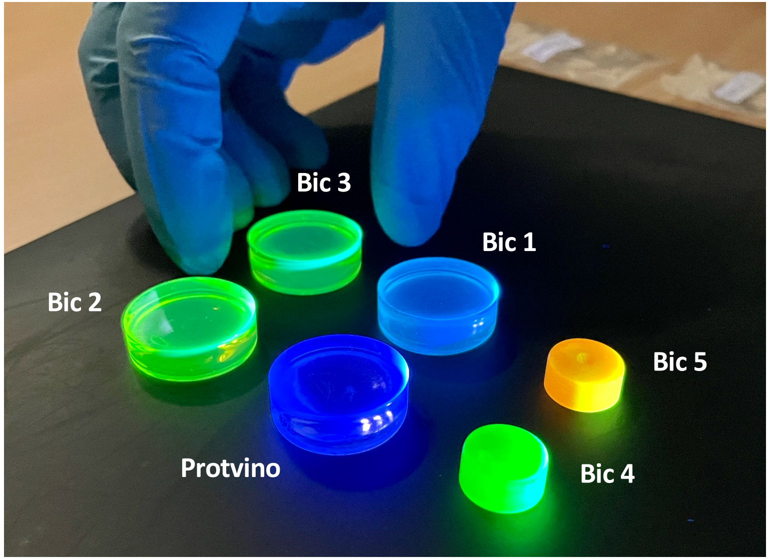
Additionally exploring:

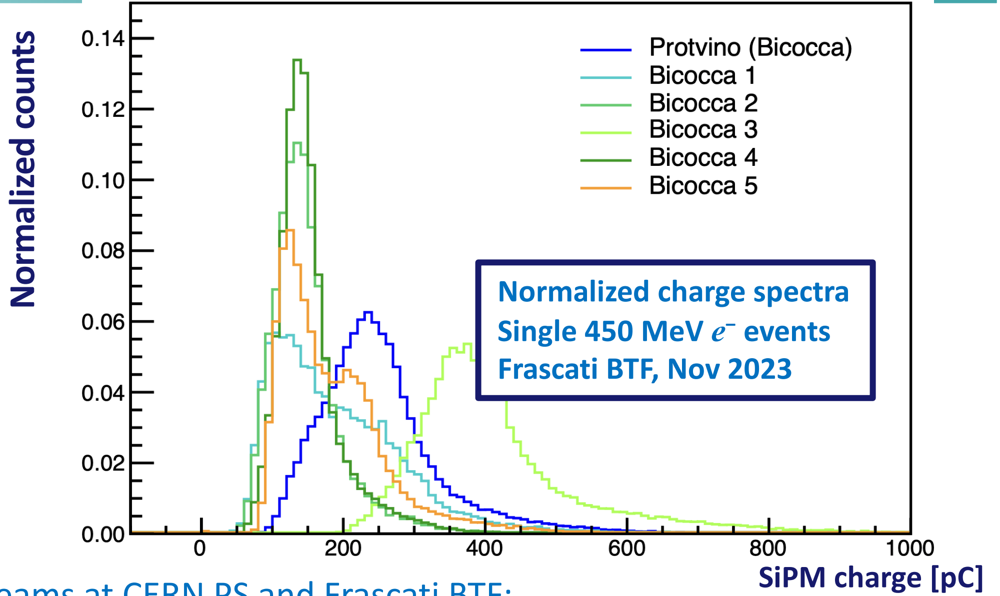
- **New dyes** for optimized molecular scintillators
- Fast, bright, radiation hard **green scintillators**

2022-23: Test scintillators/fibers/SiPMs with beams and cosmic rays

2024-25: Construct full-scale prototype if promising candidate found

TB with nanocomposite scintillators Nanocal





Tests with mip and e^- beams at CERN PS and Frascati BTF:

- Reference sample: 1.5% PTP + 0.04% POPOP in PVT (“Protvino”)
- Bicocca 4, 5: $CsPbBr_3$:Yb perovskites in PVT have ~50% light yield of ref. sample
  Our first nanocomposites with good mip response!
- Bicocca 3: Coumarin-6 (green) scintillator with ~160% light yield of ref. sample

Many new samples to be tested in Feb 2024 BTF run!

- # Publications:
  - L. Martinazolli et al., Compositional engineering of multicomponent garnet scintillators: Towards an ultra-accelerated scintillation response, , Mater. Adv., 2022,3, 6842-6852
  - G. Tamulaitis et al, Transient optical absorption as a powerful tool for engineering of lead tungstate scintillators towards faster response, J. Materials Chemistry C, 10, 9521 (2022), DOI: 10.1039/d2tc01450e.
  - F. Pagano et al., A new method to characterize low stopping power and ultra-fast scintillators using pulsed X-rays, Front. Phys. 10:1021787.doi: 10.3389/fphy.2022.1021787
  - G. Tamulaitis et al, Transient optical absorption technique to test timing properties of LYSO:Ce scintillators for the CMS Barrel Timing Layer, Radiation Physics and Chemistry 206, 110792 (2023).
  - S. Nargelas et al., Influence of heavy magnesium codoping on emission decay in Ce-doped multicomponent garnet scintillators, J. Mater. Chem C, 11, 12007 (2023), 11, 12007-12015 (2023)
  - Y. Talochka et al., Acceleration of emission decay in Ce-doped Gd-containing garnets by aliovalent codoping due to blocking excitation transfer via gadolinium subsystem, Radiation Physics and Chemistry, **218**, 111589 (2024).
  - A. Erroi, et al. ACS Energy Lett. 8, 3883-3894, (2023)
  - C. Cantone et al., Beam test, simulation, and performance evaluation of PbF2 and PWO-UF crystals with SiPM readout for a semi-homogeneous calorimeter prototype with longitudinal segmentation, Front. Phys. 11:1223183, doi:10.3389/fphy.2023.1223183
  - L. Bandiera et al., A highly-compact and ultra-fast homogeneous electromagnetic calorimeter based on oriented lead tungstate crystals, Front. Phys. 11:1254020, doi: 10.3389/fphy.2023.1254020
  - L. Bandiera et al., Investigation of radiation emitted by sub GeV electrons in oriented scintillator crystals, NIMA Vol 1060, March 2024, 169022

Presentations:

- At SCINT2022: 16th Int. Conference on Scintillating Materials & their Applications, September 19-23, 2022, Santa Fe, USA
  - G. Tamulaitis: Transient Optical Absorption Technique as a Tool for Routine and In-depth Characterization of Fast Scintillators, (keynote)
  - N. Kratochwil: Characterization of dense Cherenkov/Scintillation/Semiconductor materials for fast timing at future colliders
  - R. Cala' : Exploring BaF_2:Y Ultra-fast Emission for Future HEP Applications
  - L. Martinazolli: Acceleration of the scintillation response in garnet type multicomponent scintillators
  - S. Nargelas: Influence of matrix composition on excitation relaxation and emission spectrum of Ce ions in $(GdxY1\text{-}x)_3Al_2Ga_3O_{12}$:Ce scintillators
- R. Cala‘, Exploration of fast materials and light production mechanisms for high energy charged particles time detectors, Fast timing workshop, May 2023
- E. Auffray, Recent developments in the field of scintillators for fast radiation detectors, Fast timing workshop, May 2023
- E. Auffray, Recent developments in the field of scintillators for radiation detectors, TIPP2023
- R. Cala‘, Recent developments in the field of scintillators for fast radiation detectors, IPRD23 (16th Topical Seminar on Innovative Particle and Radiation Detectors)

Conclusion

- Different scintillating materials have been investigated
  - Many synergies between participants
  - Joint activities

- Fruitful test beam activities in 2023

- Deliverable D8.2: submitted on Feb 28