

Development of fine-sampling calorimeters with nanocomposite scintillating materials

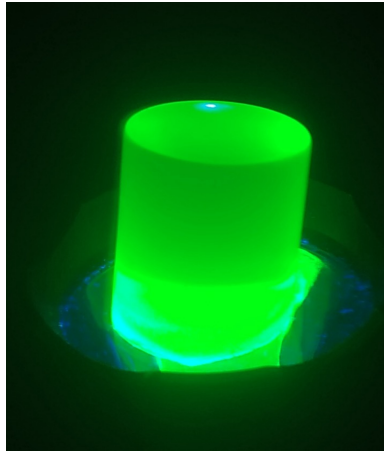
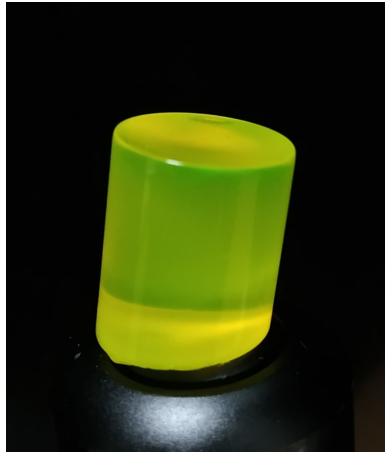
NanoCal Status Report

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For the NanoCal Project

Prospective and Technology-Driven Detector R&D
AIDAinova 3rd Annual Meeting, 19 March 2024



Nanomaterial composites (NCs)



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

- Perovskite (ABX_3) or chalcogenide (oxide, sulfide) nanocrystals
- Cast with polymer or glass matrix
- Decay times down to $O(100 \text{ ps})$
- Radiation hard to $O(1 \text{ MGy})$

Nanocrystals and composite can be **engineered** to obtain performance requirements

- **Nanocrystal:** emission wavelength, decay time, etc.
- **Composite:** concentration of nanocrystals and/or additional fluors, e.g. very high concentration of nanocrystals to obtain shorter radiation length

Can realize thin nanocrystal films to realize **fast timing layers**

Nanocrystal composites could make **very fast WLS devices** to efficiently couple light from fast scintillators to SiPMs

NCs have high potential for innovative, high-performance calorimetry

From test bench to detector

Nanocomposite scintillators have received much attention in the materials-science community

- Many studies of photoluminescence for $E_\gamma < 10 \text{ eV}$
- However, **almost no studies** have been done on the response of NC scintillators to **high-energy particles**

So far, **applications in HEP have received little attention**

No attempt yet to build a **real calorimeter with NC scintillator** and **test it with high-energy beams**

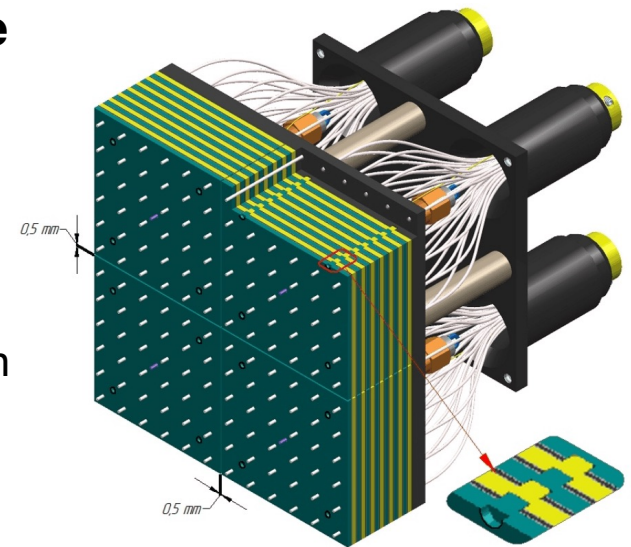
NanoCal project: Construct a calorimeter prototype with NC scintillator and test with high-energy beams

Shashlyk design naturally ideal as a test platform:

- Easy to construct with very fine sampling
- Primary scintillator and WLS materials required: both can be optimized using NC technology

Additionally exploring:

- **New dyes** for optimized conventional scintillators
- Fast, bright **green scintillators** for additional radiation hardness

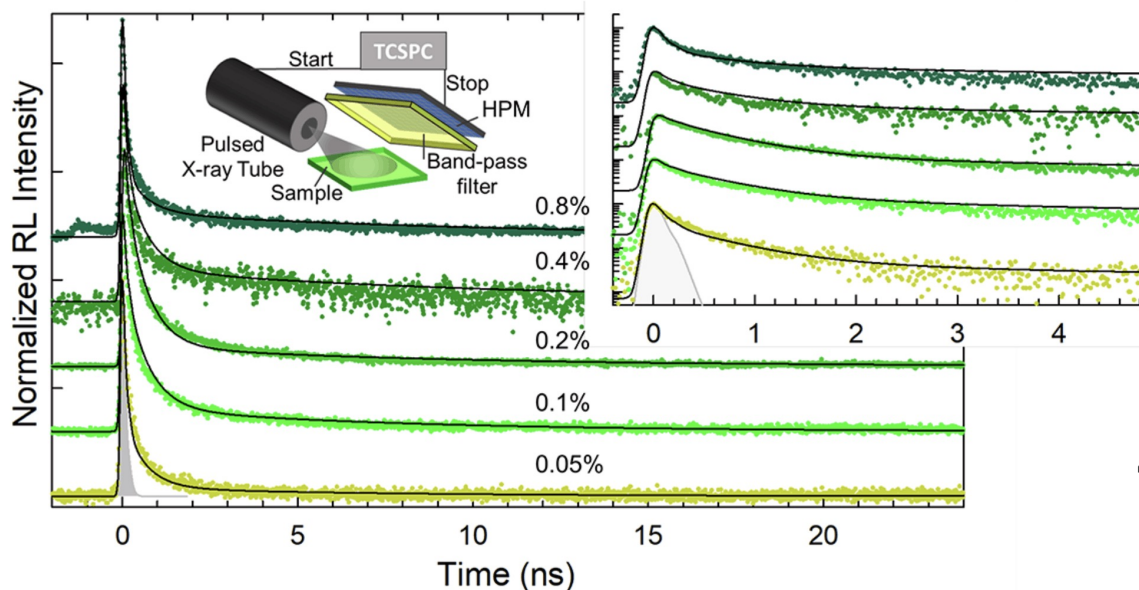


KOPIO/PANDA design
Fine-sampling shashlyk

The promise of lead halide perovskite NCs

Ultrafast and Radiation-Hard Lead Halide Perovskite Nanocomposite Scintillators

CsPbBr₃ nanocrystals (~10 nm)
0.05 to 0.8% w/w in UV-cured polyacrylate matrix, measured with **radio- and photoluminescence**

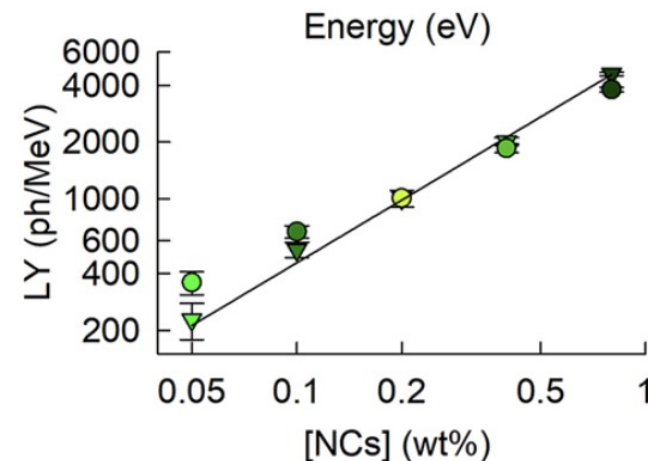


30% of light emitted in < 80 ps

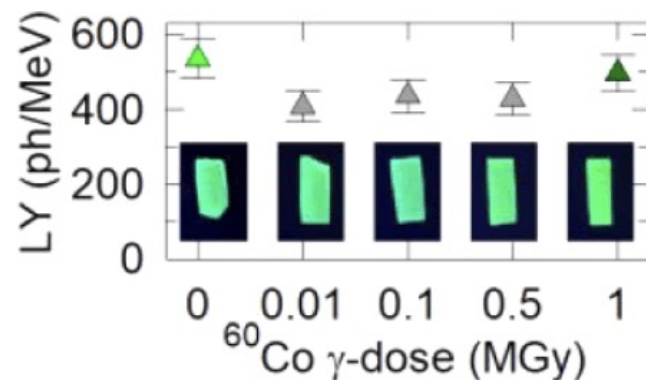
3-component fit to pulse shape for 0.2%

- 34% prompt (Gaussian, 160 ps IRF)
- 22% with $\tau \sim 600$ ps (R_1)
- 44% with $\tau \sim 10$ ns (R_2)

R_2 increases with concentration, prompt component maintained

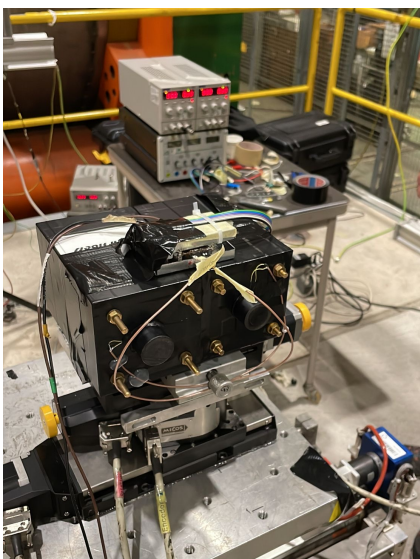
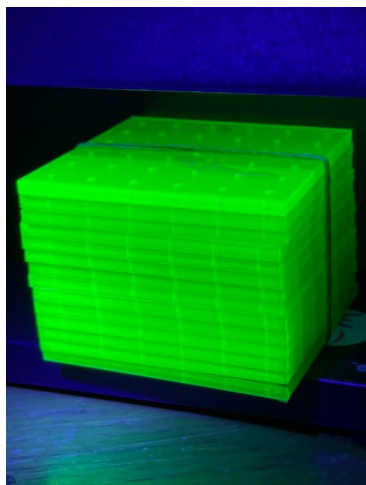
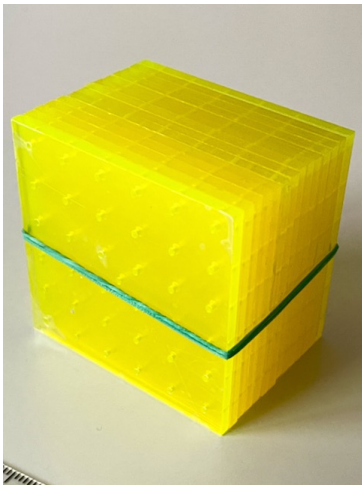


4800 ph/MeV at 0.8% w/w



No decrease in LY up to 1 MGy

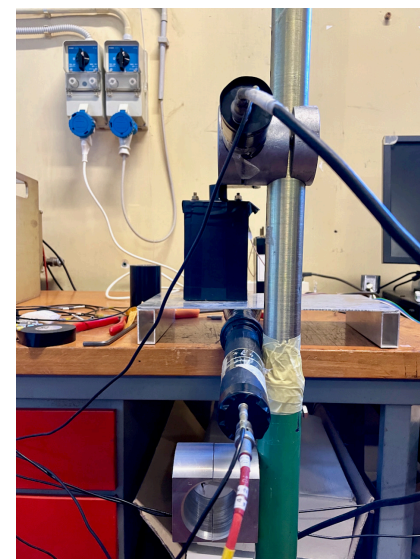
Tests of early prototypes



First test of NanoCal prototype in fall 2022, followed by laboratory studies with cosmic rays spring 2023

Comparison of light yield from conventional (Protvino) shashlyk with NanoCal ($\text{CsPbBr}_3/\text{PMMA}$) prototype

$\text{LY}_{\text{mip}}(\text{NanoCal}/\text{Protvino}) \sim 5\%$



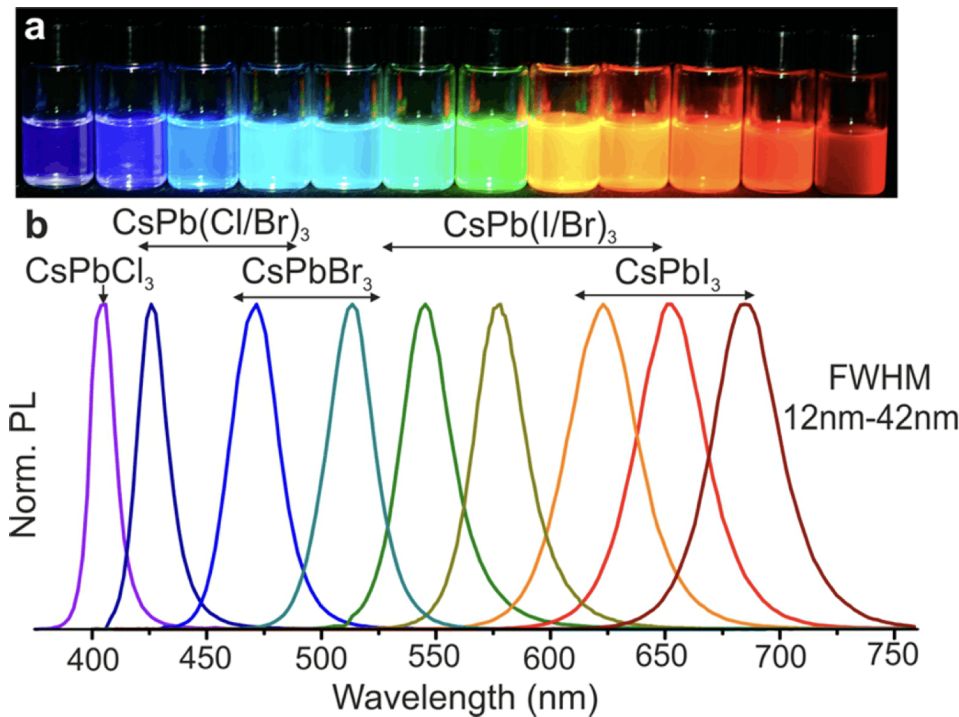
Possible issues:

1. Nanoparticles exhibit too much self-absorption?
2. Inefficient excitation of nanoparticles: concentration too low, no contribution from matrix?

NC scintillator optimization

Continue to optimize NC scintillator via additional tests with small modules before constructing full-scale prototypes

- **New ideas for NC scintillators with longer absorption length**



- Use co-dopant as internal WLS, like conventional fiber
 - Many fast (1-2 ns) candidates
- Use blue-emitting CsPbCl_3 to obtain primary emission at shorter wavelength
 - Tricky interface chemistry needed to stabilize CsPbCl_3 against non-emitting phases, but possible
 - Can tune emission by admixture of different halides

Example of nanocomposite optimization

CsPbBr₃ in PMMA (green emission) → **inconvenient to shift to red**

A promising idea:

1. Use CsPb(Br,Cl)₃: Substitution of some Br with Cl shifts emission to blue
2. Add coumarin-6 as a high-Stokes-shift WLS → shifts emission back to green

**Synthesis of
CsPbBr₃**



**Substitution of
50% of Br with Cl**



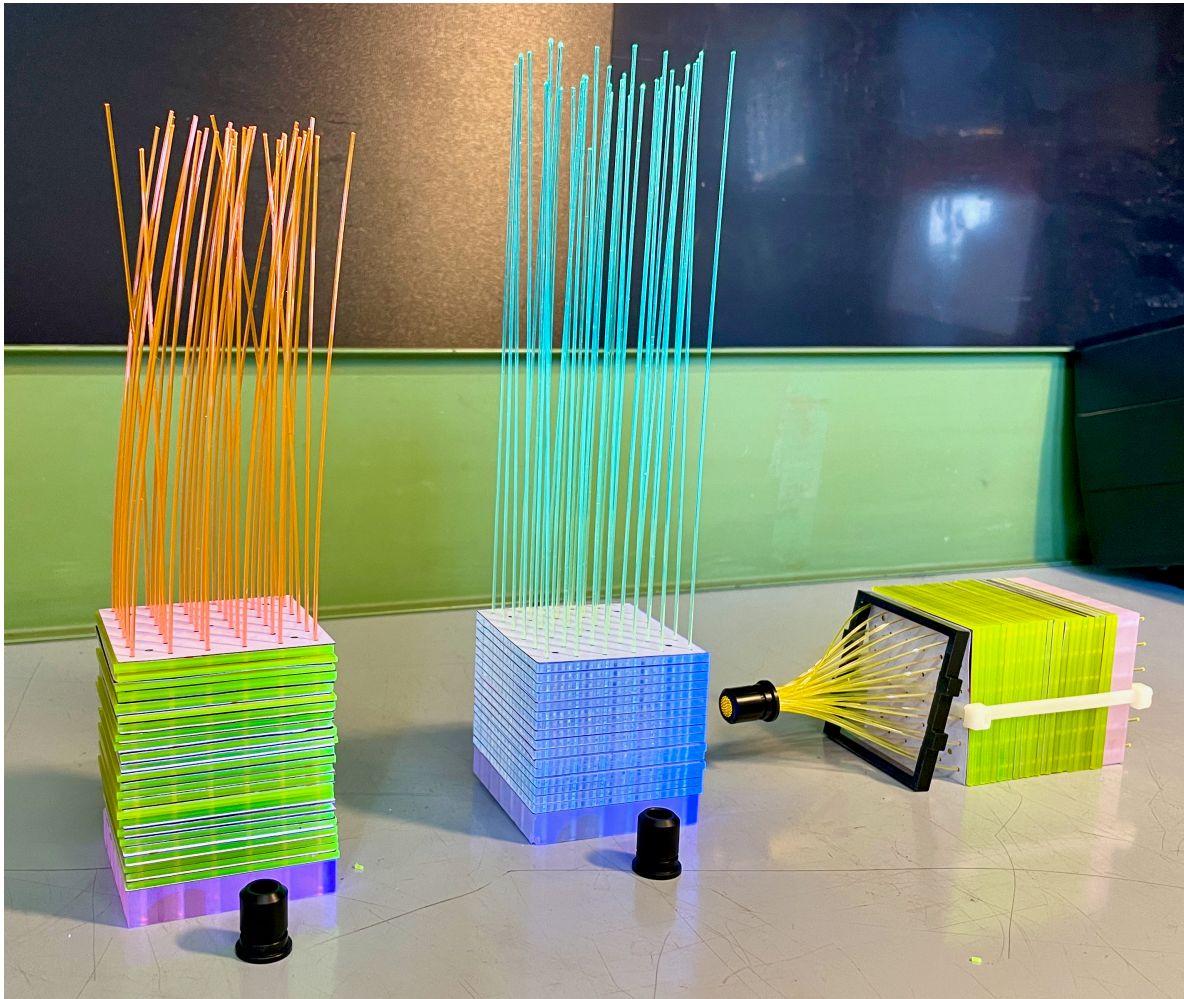
**Addition of
coumarin-6**



However, surface passivation of nanocrystals destroyed during substitution reaction, leading to aggregation (milky appearance, poor transparency)

First prototype tested anyway with custom (NCA-1) fibers at June 2023 test beam

New prototypes for summer 2023 tests



Prototypes tested in PS T9, June 2023:

- Conventional scintillator (Protvino), Y-11 fibers
- Conventional scintillator (PVT + BTP 0.02%), Y-11 fibers
- PMMA + CsPbBr₃ 0.2%, O-2 fibers
- PMMA + CsPbBr₃ 0.2%, custom NCA-1 fibers
- PMMA + CsPb(Br,Cl)₃ + coumarin-6, NCA-1 fibers

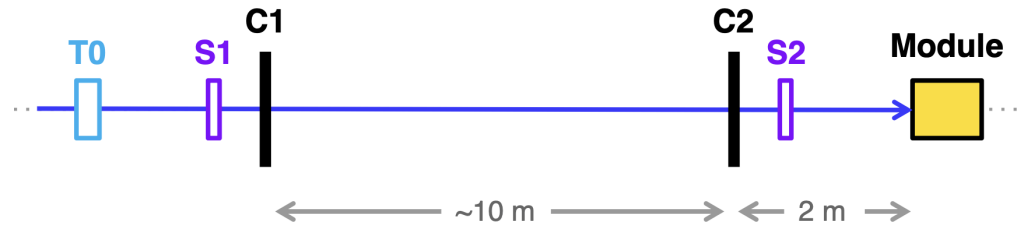
NanoCal setup and T9 test results

Electron beam, 1, 2, 4 GeV
MIP beam (μ^- or π^-), 10 GeV
Cerenkov ID for $e/\mu/\pi/p$

For each prototype:

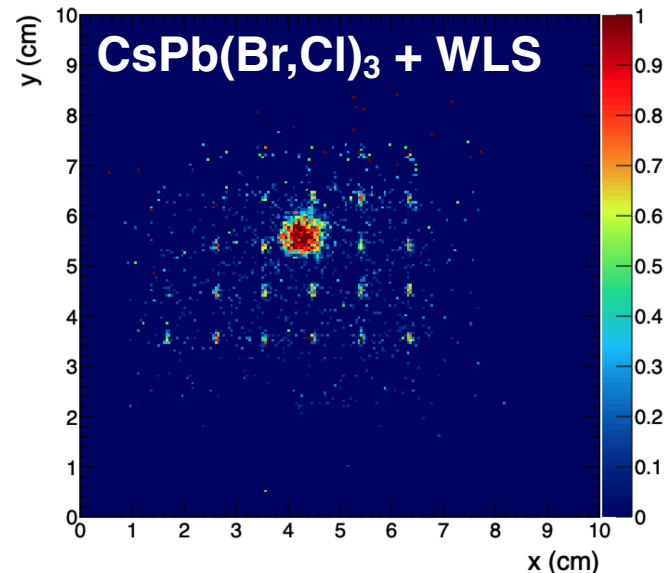
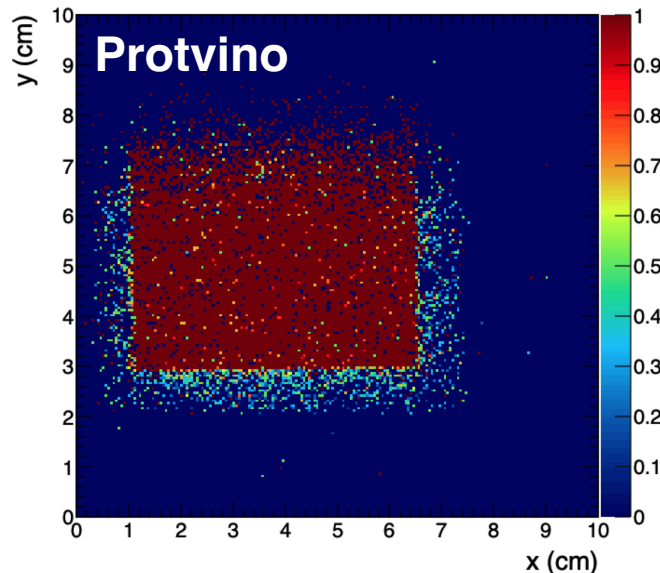
- MIP response, efficiency
- e^- response
- Time resolution

T0	Time reference detector
S1, S2	Trigger scintillator paddles
C1, C2	Si strip tracking chambers, $10 \times 10 \text{ cm}^2$
Module	Module to be tested



Efficiency maps with 10 GeV μ , threshold = $5\sigma_{\text{noise}}$

Disappointing result from new nanocomposite: only light is from readout fibers!

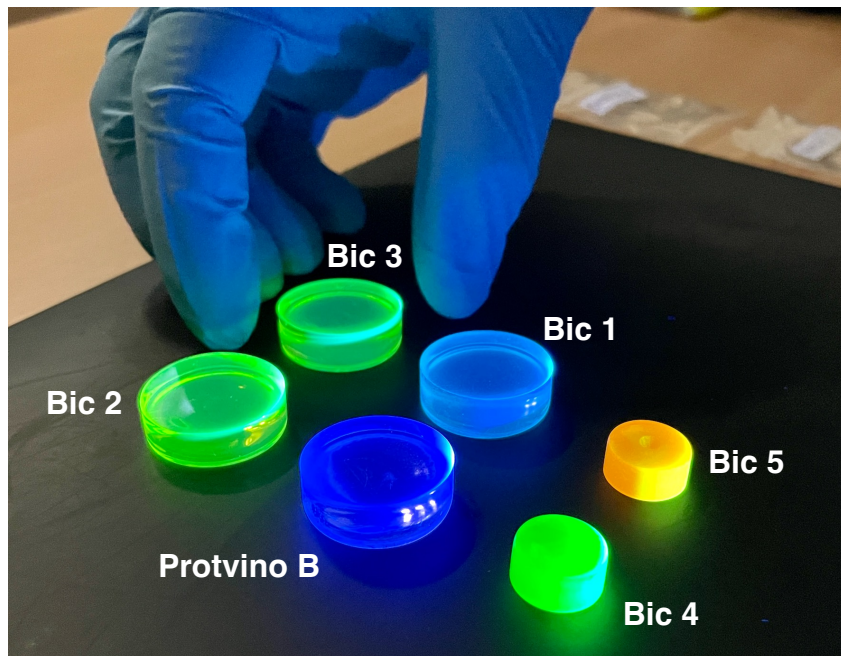


New samples for fall 2023 tests

Still have many of good ideas for next steps

- Direct synthesis of $\text{CsPb}(\text{Br},\text{Cl})_3$ to preserve surface passivation
- Use of an aromatic matrix material, e.g., PVT as in conventional scintillator
 - First formulations use PMMA: gives no primary scintillation contribution
 - Now have new protocol to use perovskites with thermally polymerized matrix, with or without additional WLS

New samples synthesized and tested in fall 2023:



All samples 90:10 PVT/DBV matrix
with 1.5% PTP as primary dye

Protvino B	0.04% POPOP
Bicocca 1	0.04% benzothiophene (BTP)
Bicocca 2	0.04% coumarin-6
Bicocca 3	0.04% BTP + 0.04% coumarin-6
Bicocca 4	1% $\text{CsPbBr}_3:\text{Yb}$
Bicocca 5	1% $\text{CsPbBr}_3:\text{Yb}$ + perylene dyad

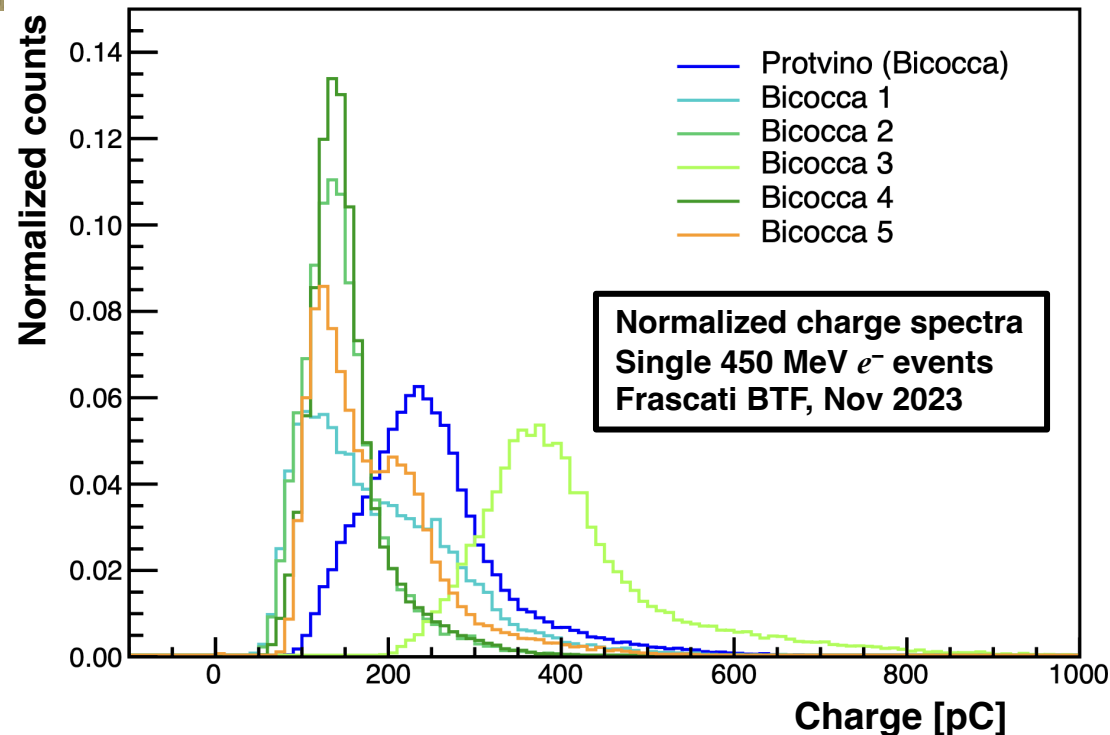
Light yield tests with new samples



Tests with mip and e^- beams:

CERN T9	6, 10 GeV electrons
Oct 2023	10, 15 GeV hadrons/mips
Frascati BTF	450 MeV electrons
Nov 2023	Similar to mips for small samples

- Reference sample:
1.5% PTP + 0.04% POPOP in PVT (“**Protvino**”)
- Bicocca **4**, **5**: CsPbBr₃:Yb in PVT
~50% ILY of ref. sample
Our first nanocomposites with good mip response!
- Bicocca **3**: Coumarin-6 in PVT with PTP + BTP
~160% LY of ref. sample!



New scintillators and future directions

New samples with 1-2.5% CsPbBr₃:F in PVT with/without additional dyes to be tested at BTF in Apr 2024!

- Surface passivation with fluorine allows use of thermally polymerized matrix (PVT) and higher nanocrystal concentration
- Complete set of control samples to isolate contributions from components

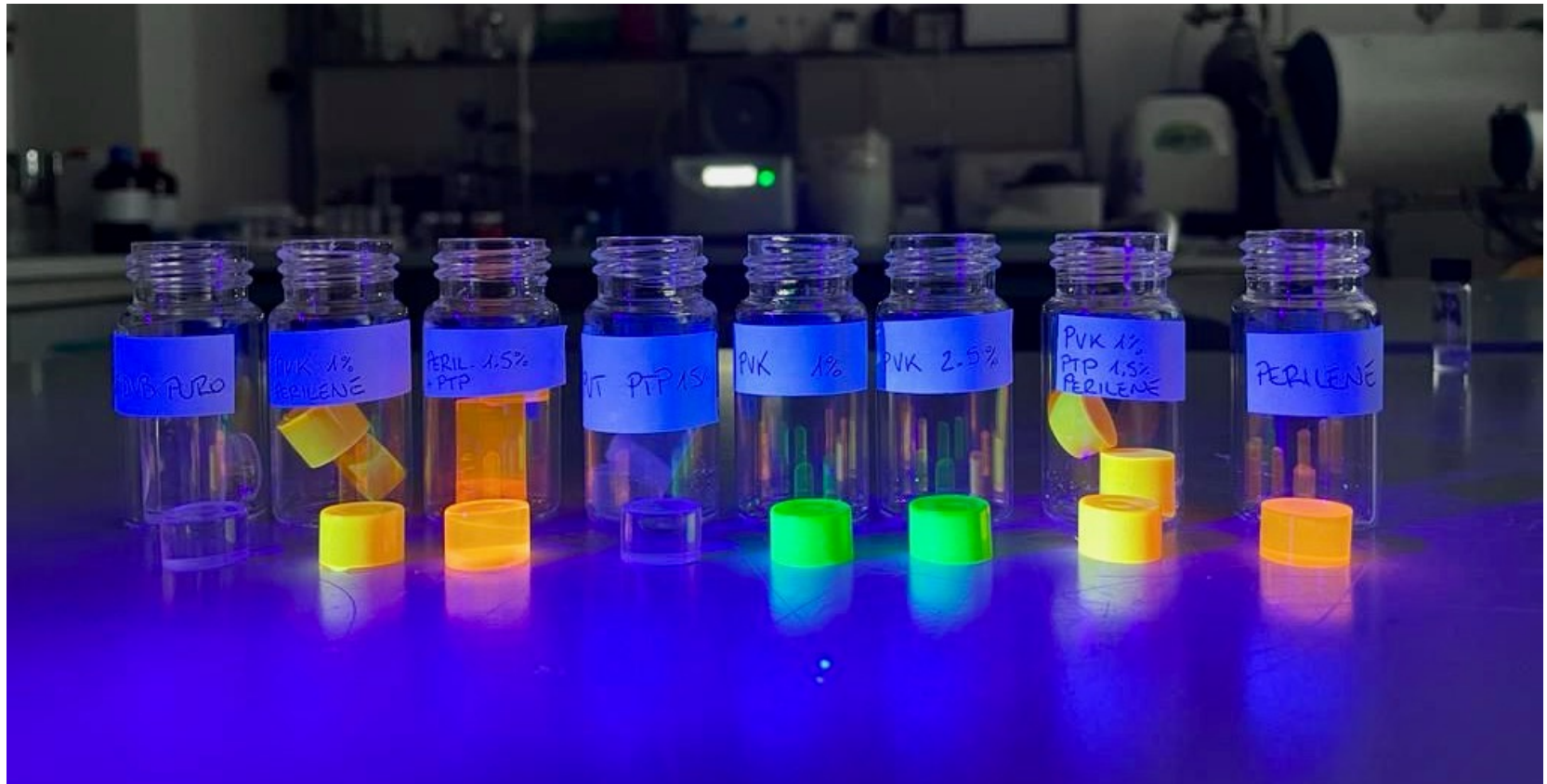
All samples 90:10 PVT/DBV matrix

Blank_0	Only matrix (control for NC24_0, 1)
Blank_1	1.5% PTP (control for NC24_2)
Blank_2	Perylene dyad (control for NC24_3)
Blank_3	1.5% PTP + perylene dyad (control for NC24_4)
NC24_0	1% CsPbBr ₃ :F
NC24_1	2.5% CsPbBr ₃ :F
NC24_2	1.5% PTP + 1% CsPbBr ₃ :F
NC24_3	1% CsPbBr ₃ :F + perylene
NC24_4	1.5% PTP + 1% CsPbBr ₃ :F + perylene

New scintillators and future directions

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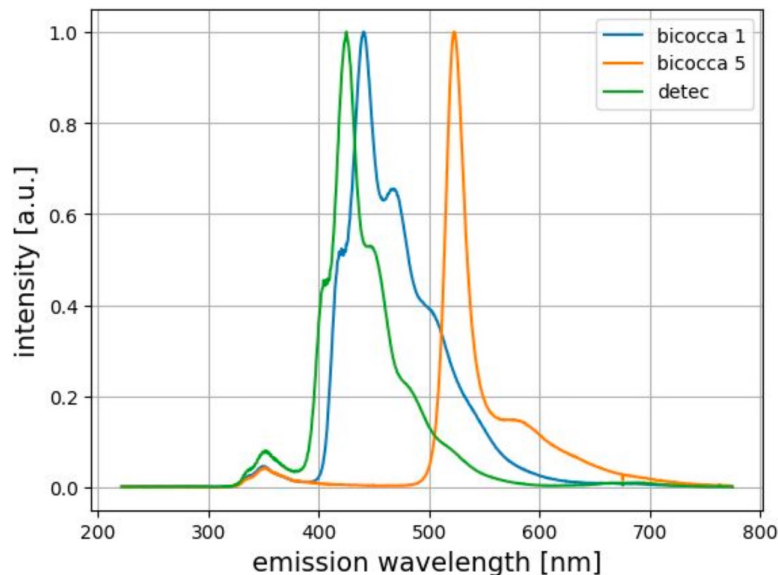
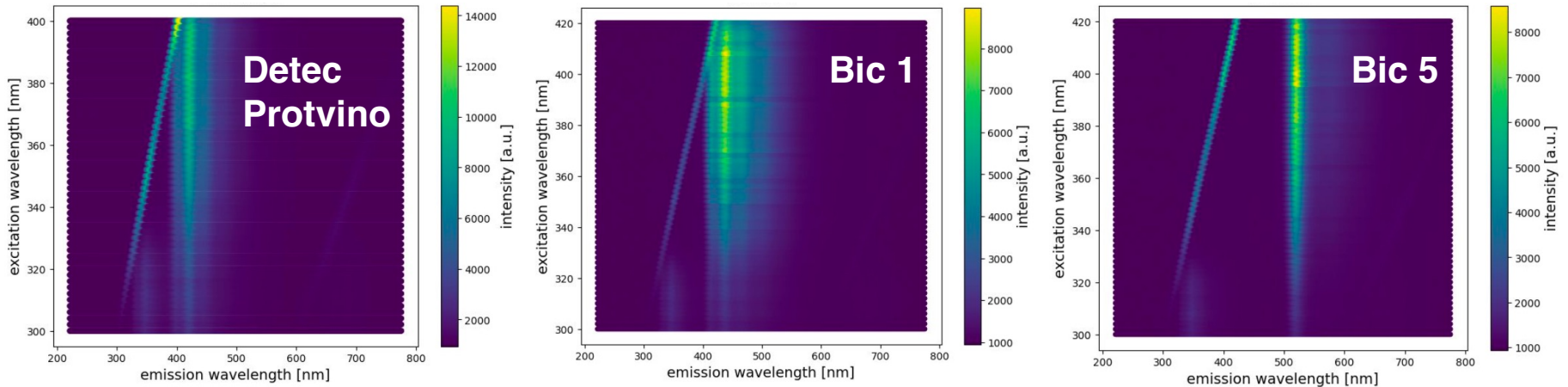
- Surface passivation with fluorine allows use of thermally polymerized matrix (PVT) and higher nanocrystal concentration
- Complete set of control samples to isolate contributions from components



Characterization of new samples

Samples are ready and are being characterized in laboratory and with cosmic rays in preparation for beam test

Spectroscopic characterization: Emission vs excitation spectra



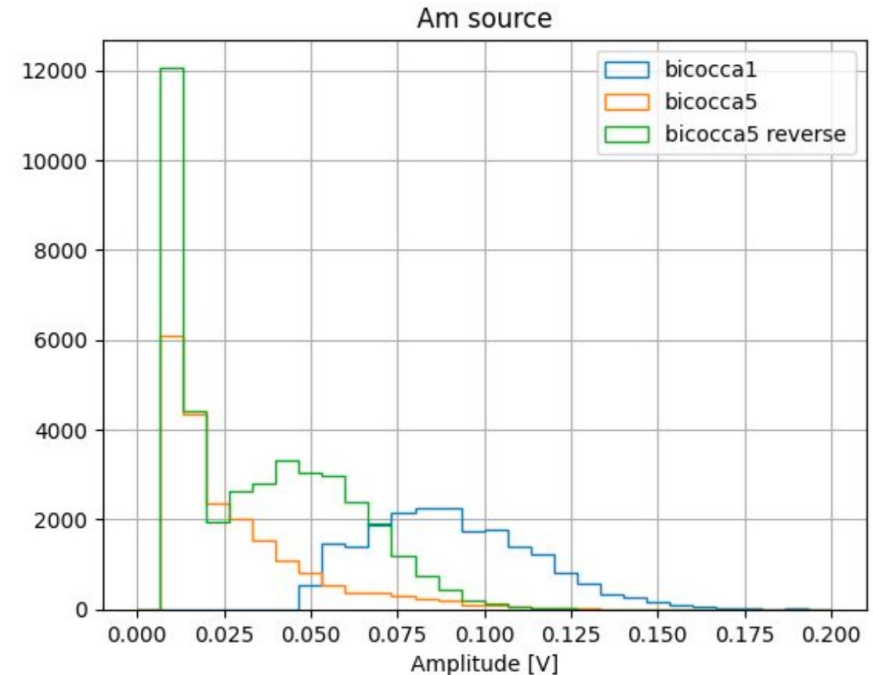
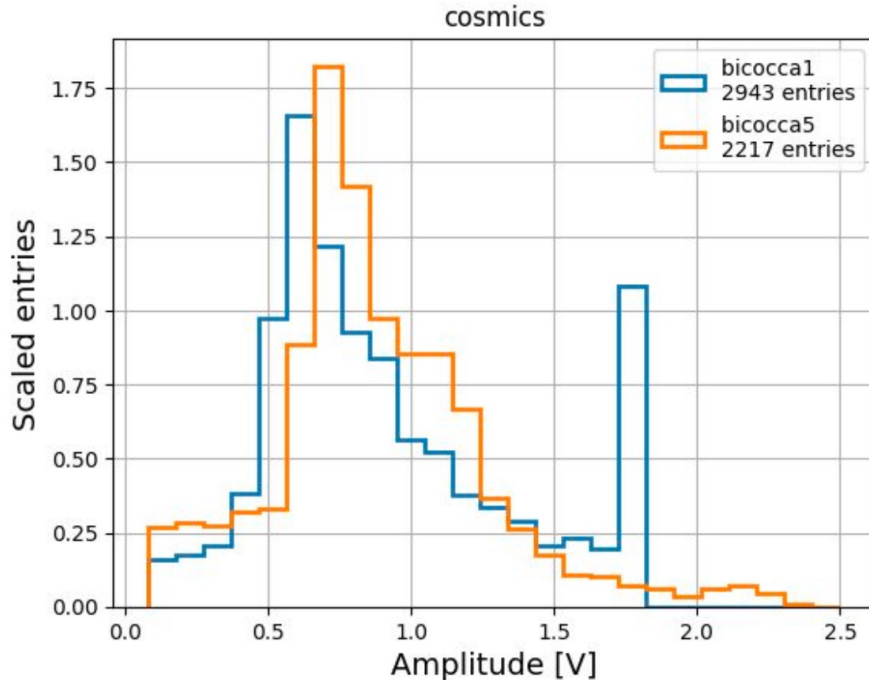
Emission for excitation at [320, 400] nm

- Detec/ISMA (similar to Protvino)
- Bicocca 1 (PVT/BTP)
- Bicocca 5 (CsPbBr₃:Yb/PTP/perylene)
- Emission from PTP, highly attenuated, short absorption length; efficient WLS from perylene
- Together with analysis of blanks, can understand contributions from components

Characterization of new samples

Samples are ready and are being characterized in laboratory and with cosmic rays in preparation for beam test

Performance and quality control: Cosmic-ray/alpha spectra

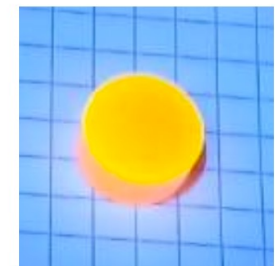
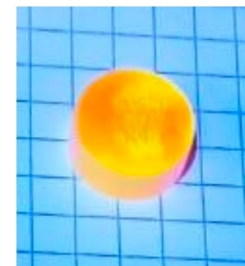


Cosmic rays: Bicocca 5 (CsPbBr_3 /perylene) has slightly higher output than Bicocca 1 (conventional)

Alpha source: Bicocca 5 has lower output

- Depends on side illuminated by alpha source — could depend on quality control considerations

A side on SiPM **B side on SiPM**



Outlook for measurements

By summer, we should have complete results from spectroscopy, cosmic rays, sources, and BTF electrons for *all* samples

Better setup for measurements with cosmic rays and beams:

- New laboratories in Frascati and Napoli
- New sample holders with better optical coupling, easier sample handling
- Electronics with reduced noise
- New DAQ system for digitizers
- Addition of Medipix-2 pixel detector to BTF setup for multiplicity counting

Survey will allow us to identify the best candidate(s) for a small prototype to be tested with mips and electrons in CERN PS T9 in September 2024:

- Isolation of contributions from nanocrystals and dyes and better understanding of how NC scintillators work

Final observations

Original polyacrylate/CsPbBr₃ formula originally very promising in radioluminescence and photoluminescence studies, but light output is very small for single mip

Project focus shifted from prototyping to sample characterization to explore ideas for better NC scintillators for HEP:

- NCs with WLS shifting dyes
- Thermally polymerized NCs with aromatic matrix materials
- NC scintillators with higher concentrations of nanocrystals

Survey now in progress with cosmic rays, sources, and mip and electron beams, with improved measurement setup, to obtain:

- **Identification of most promising candidates for prototyping**
- **Better understanding of how NC scintillators work**