Organic high-Z scintillators for a flexible and fast total body nuclear imaging

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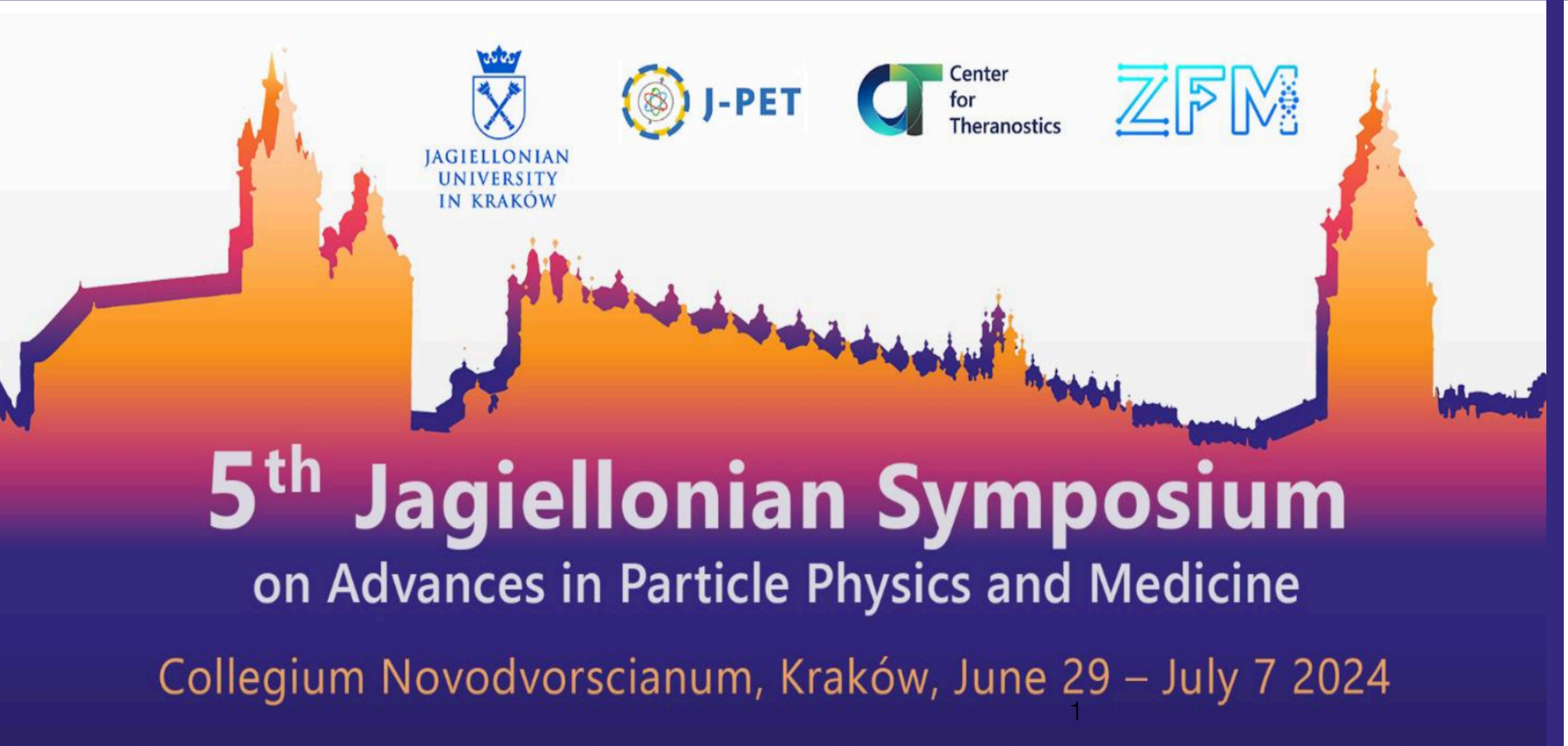

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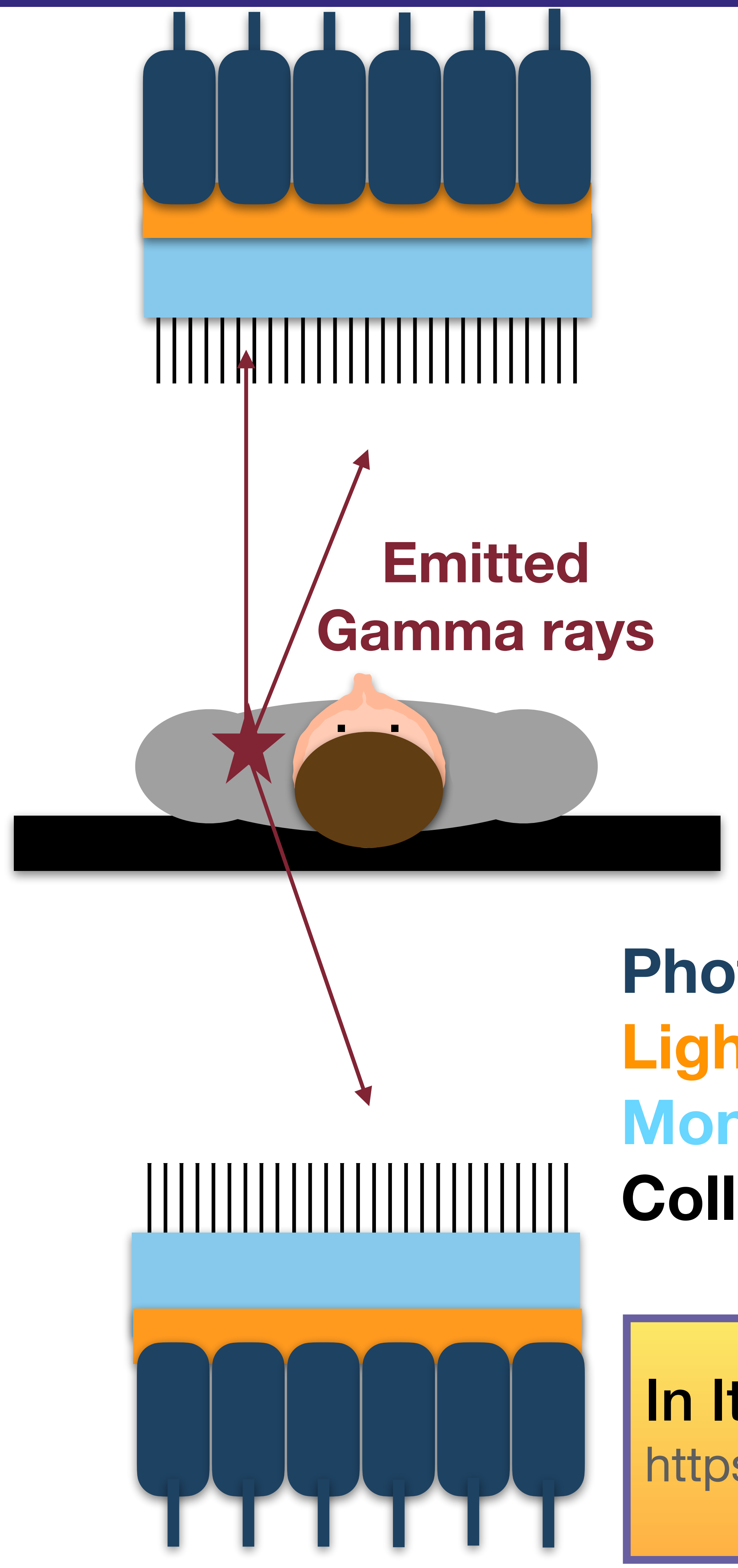
PRIN 2022 - Protocollo n. 2022Z72Y3K, CUP F53D23001500006

Single-Photon Emission Computed Tomography (SPECT)



- **Nuclear imaging technique** that allows to investigate the patient's physiological processes.
- Useful tool for **cancer detection**, cardiac and neurological diagnoses.
- Conventionally, the detection of the gamma rays is carried out by a device called **Anger Camera**.

Photomultiplier tubes
Light guide
Monolithic Inorganic Scintillator → **Typically Sodium Iodide activated with Thallium NaI(Tl)**
Collimator

In Italy, SPECT scans are more widespread than PET scans!
https://www.dati.salute.gov.it/dati/dettaglioDataset.jsp?menu=dati&idPag=80

1. **Radiopharmaceutical production**
2. **Administration of the radiopharmaceutical**
3. **Data acquisition (gamma rays detection)**
4. **Tomographic image reconstruction**
5. **Image analysis (diagnosis)**

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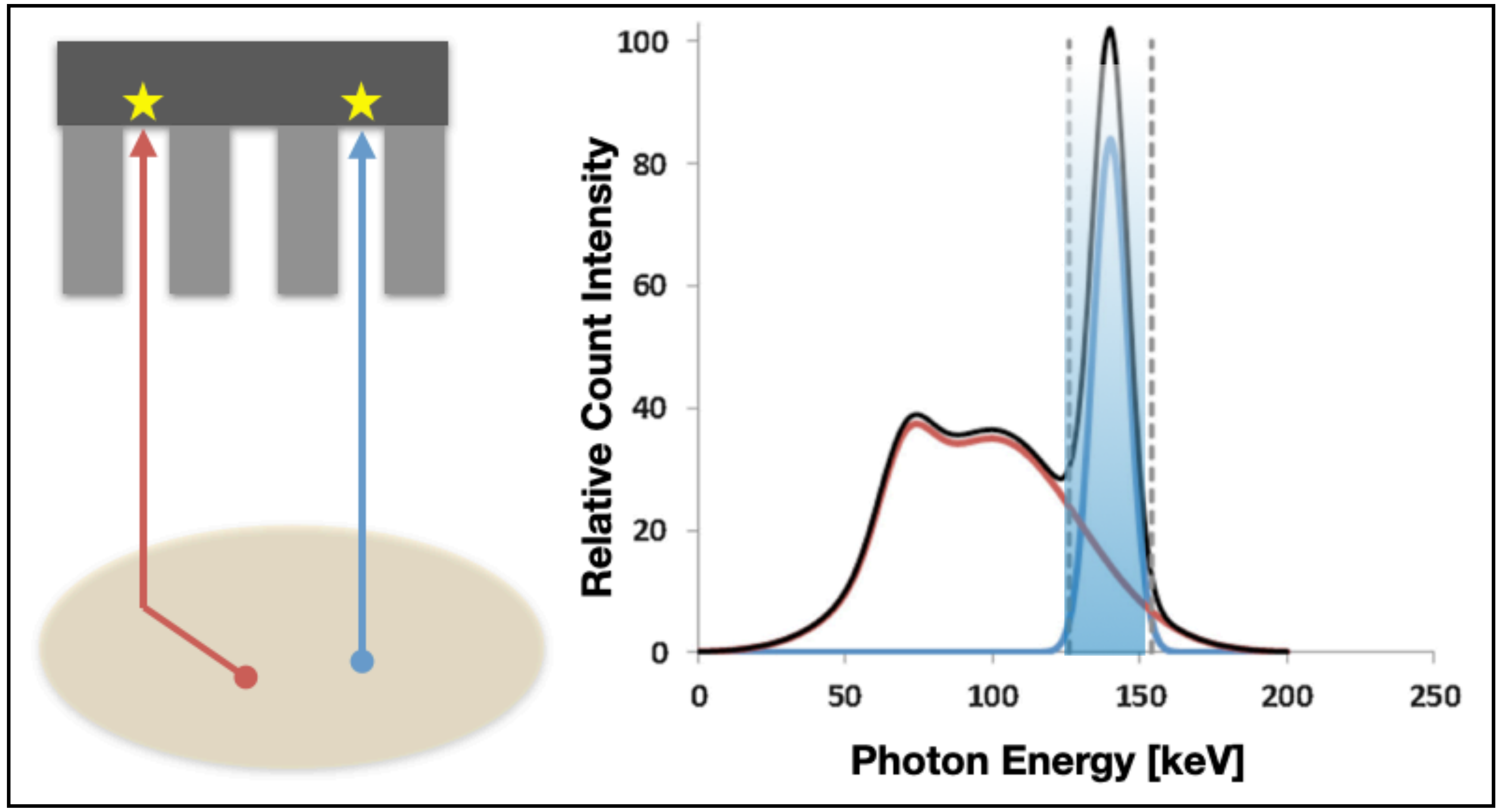
Scintillators for imaging

The most used radionuclide for SPECT exams is **Technetium-99 metastable**, that emits **140 keV γ-rays ($\tau_{1/2}$ ~ 6 h)**.

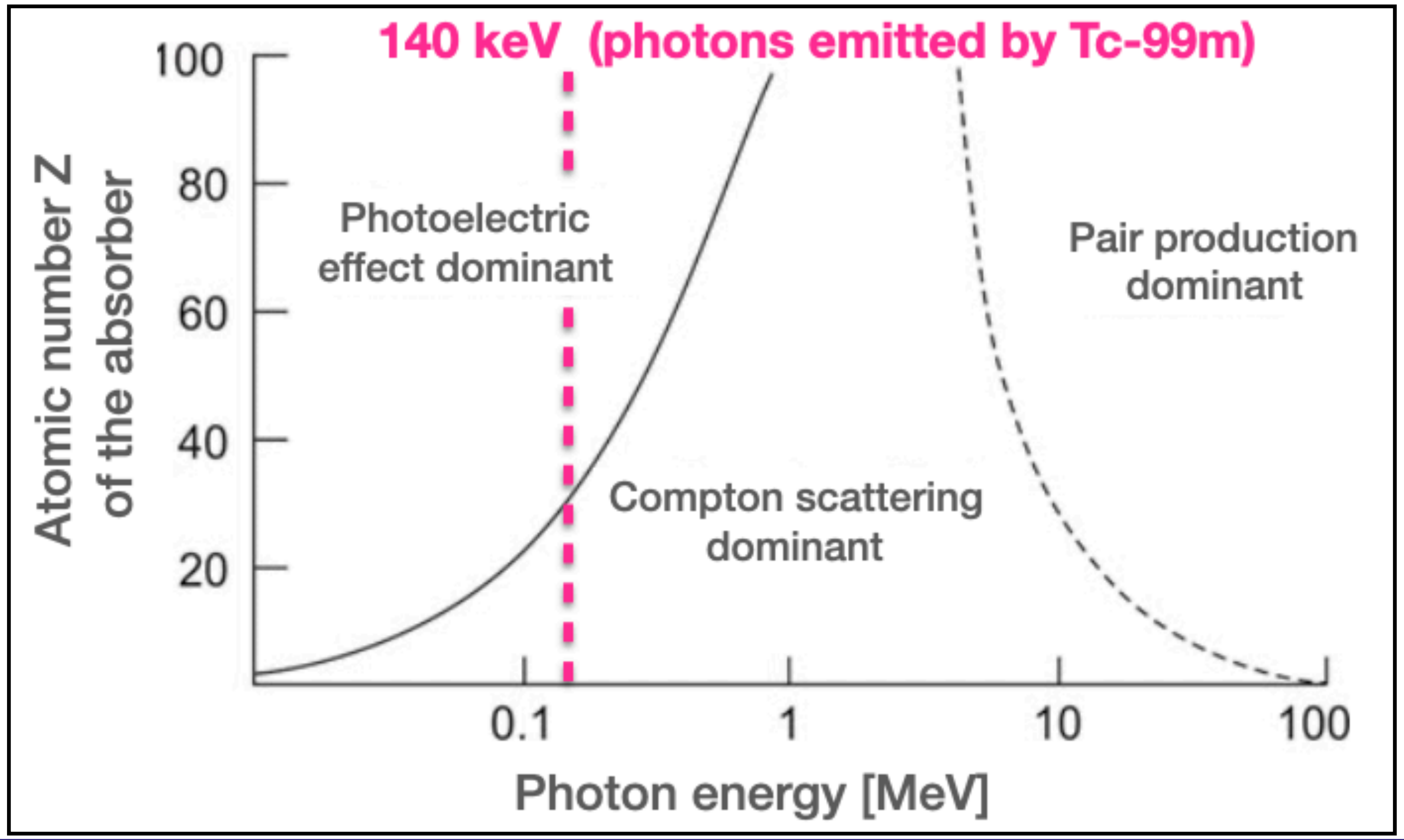


Tc-99m photopeak: these events carry the correct information about the radiopharmaceutical position.

Scattered photons: these events must be discharged to exclude the photons that changed direction inside the patient.

In order to reduce the discharge of "good" events, **the probability of photoelectric absorption inside the scintillator must be maximized**.

For 140 keV gamma rays, photoelectric absorption becomes dominant starting from Z~30



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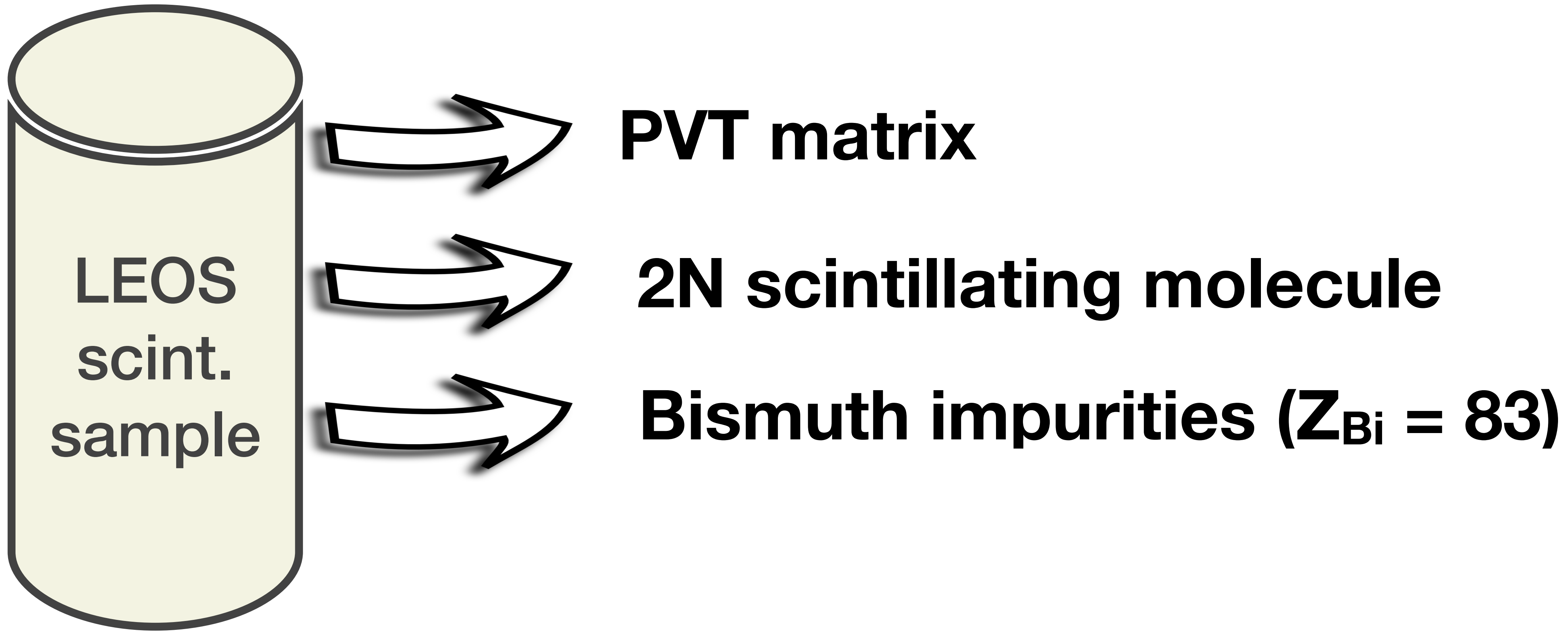
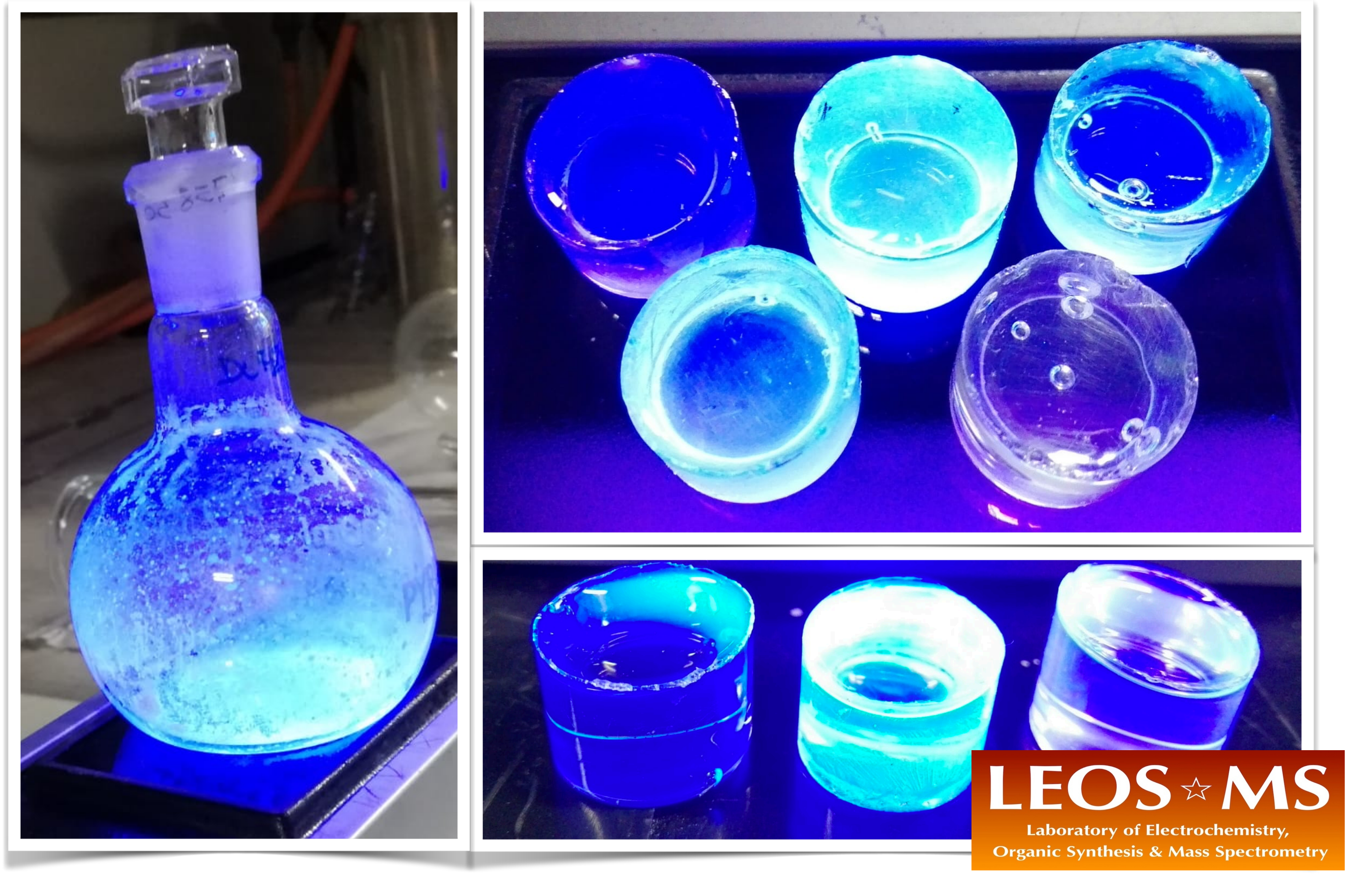
The reSPECT project: plastic scintillators

Use plastic scintillators since they are extremely **fast, light, inexpensive and easy to shape**. But the low atomic number of their constituents strongly affects the photoelectric absorption probability: to increase it, the samples of plastic scintillator must be **enriched with high-Z impurities**.

The **Laboratory of Electrochemistry and Organic Synthesis (LEOS)** have synthesized several organic scintillating molecules with promising performances in terms of light output and timing properties [1]. The **2N** has been selected to produce Bismuth-enriched samples.

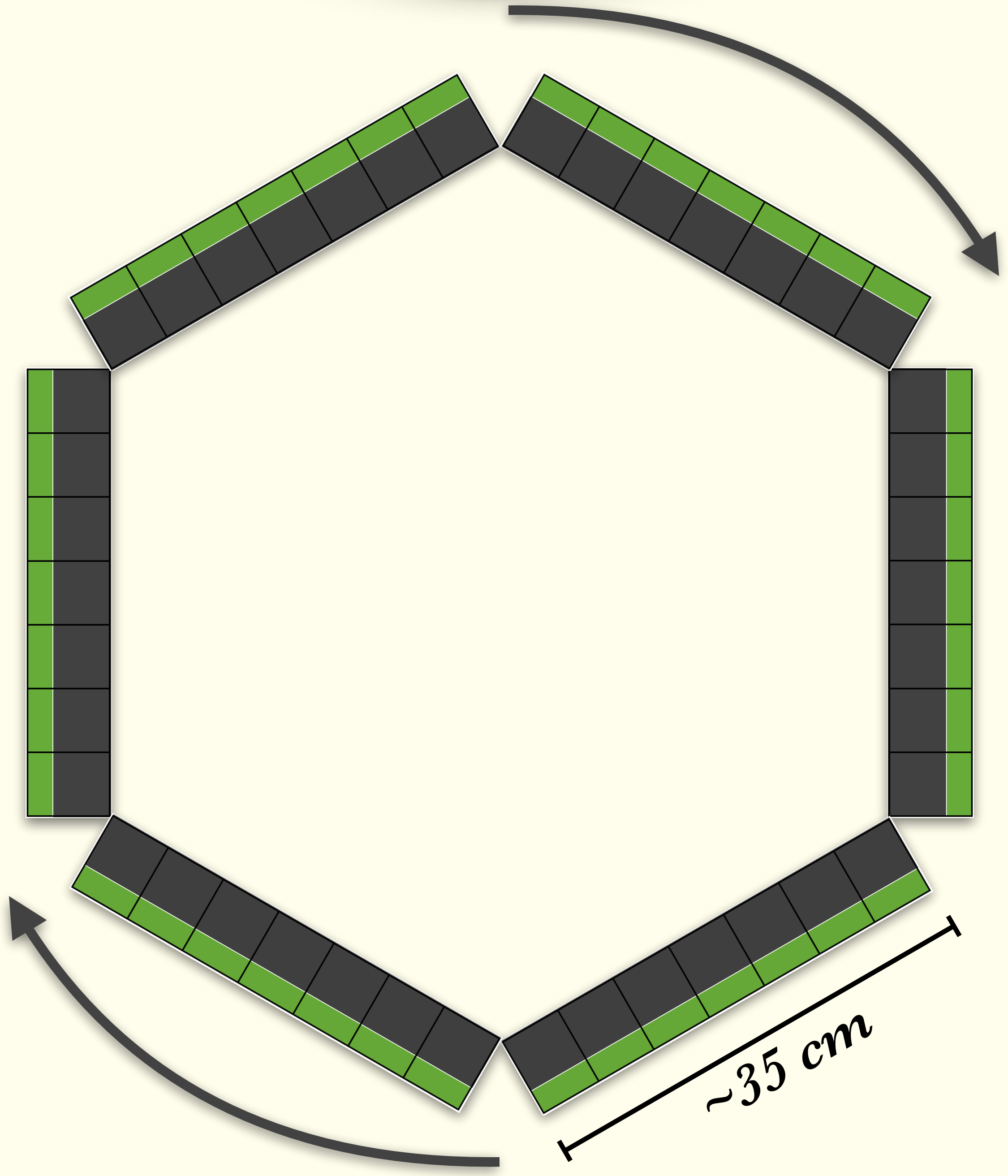




[1] Mattiello L;, Patera V.; Belardini A.; Rocco D.; Marafini M.; Organic Scintillator. *Patent* WO2023156957A1, 2023.

The reSPECT project: geometry and readout system

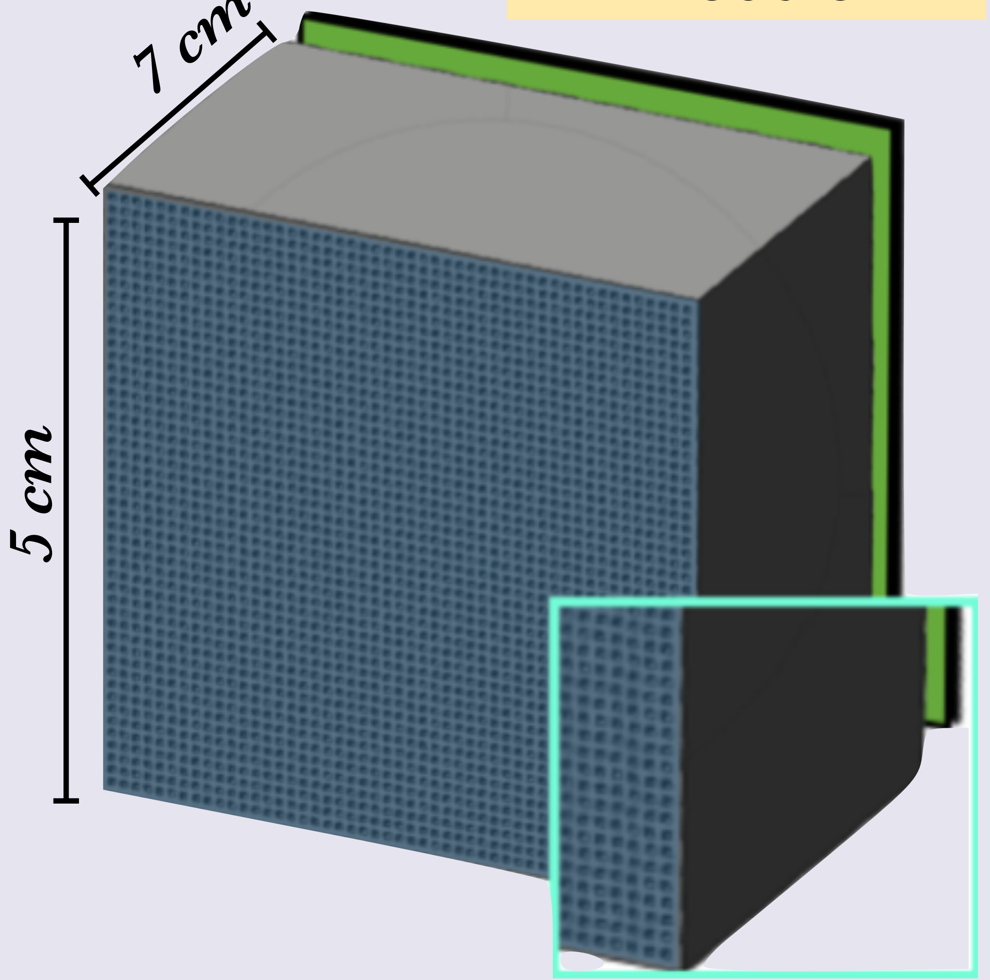
1.



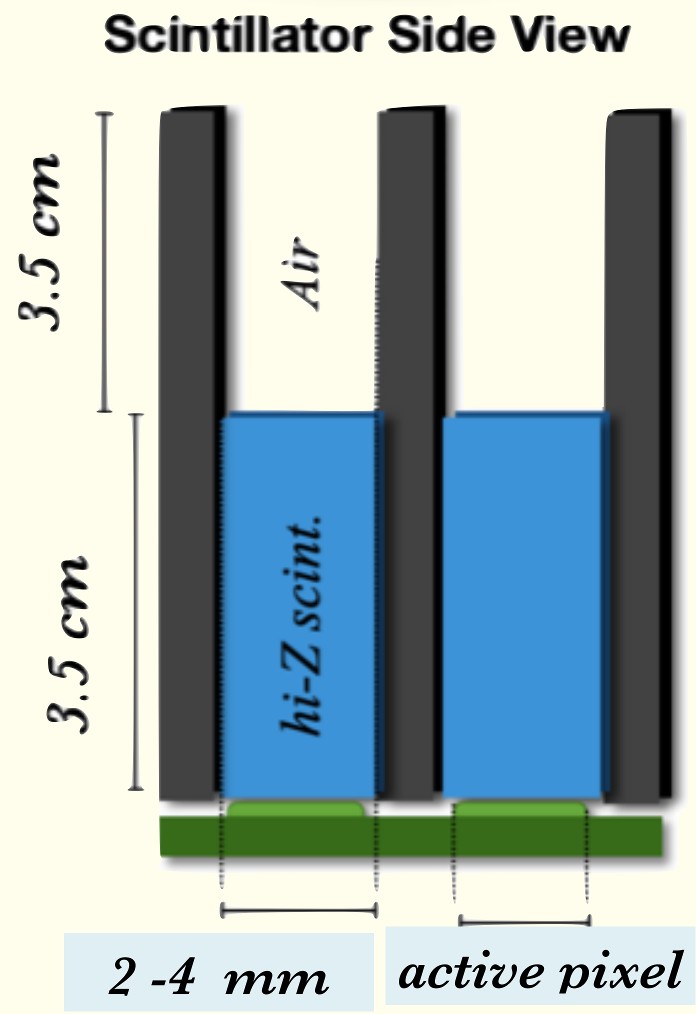
Modular structure that will be able to revolve around the patient, enabling multi-angle data acquisition.

2.

Each module will consist of a 3D-printed **tungsten frame** that serves both as a collimator and as a container of the **scintillator segments**, polymerized directly inside the holes.



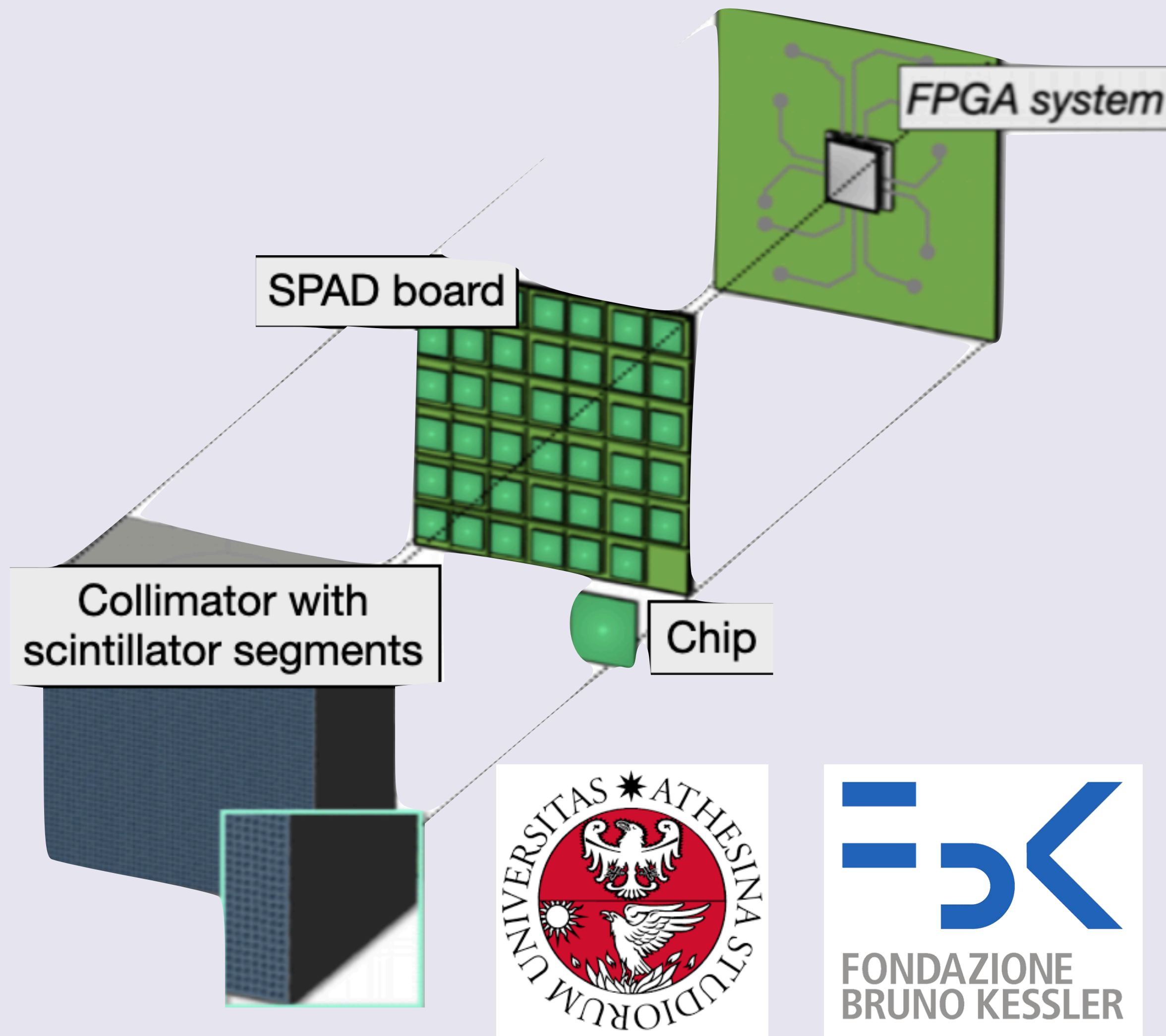
3.



Readout: **Silicon-based photodetectors** arranged in small-size pixels, individually coupled to the scintillator segments.

4.

FPGA matrices will be placed on the back of each module to pre-process the acquired data.



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The granularity of the active material is crucial in order to keep a good space resolution the same pixel size has been maintained for the readout.

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Expected performances of the reSPECT Detection system

Expected performances of reSPECT obtained through MC simulations°

SPECT DETECTION SYSTEM	SENSITIVITY PER MODULE @140 keV [cpm/μCi]	SYSTEM SPATIAL RESOLUTION (FWHM) @10 cm [mm]	DECAY TIME [ns]	RATE CAPABILITY [cps/cm^2]	TOTAL COST	MAGNETIC RESONANCE IMAGING COMPLIANCE	RADIOMETABOLIC DOSIMETRY COMPLIANCE
Anger Camera (NaI) FoV: 53 x 39 cm^2	170	7.4	250	0.25k-3k	$$	✗	✗
CZT FoV: 39 x 51 cm^2	190	7.6	350	30k-700k	$$$	✓	✗
reSPECT 6 rotating blocks, FoV: 35 x 35 cm^2	184*	8.9+	2-5	50M-200M	$	✓	✓

*energy cut at 80 keV
+2 mm pixel
°10% Bismuth concentration

- Unparalleled counting rate capability
- Possibility to conceive a total-body SPECT thanks to the modular structure
- Low cost
- Compliance with MRI and theragnostic studies

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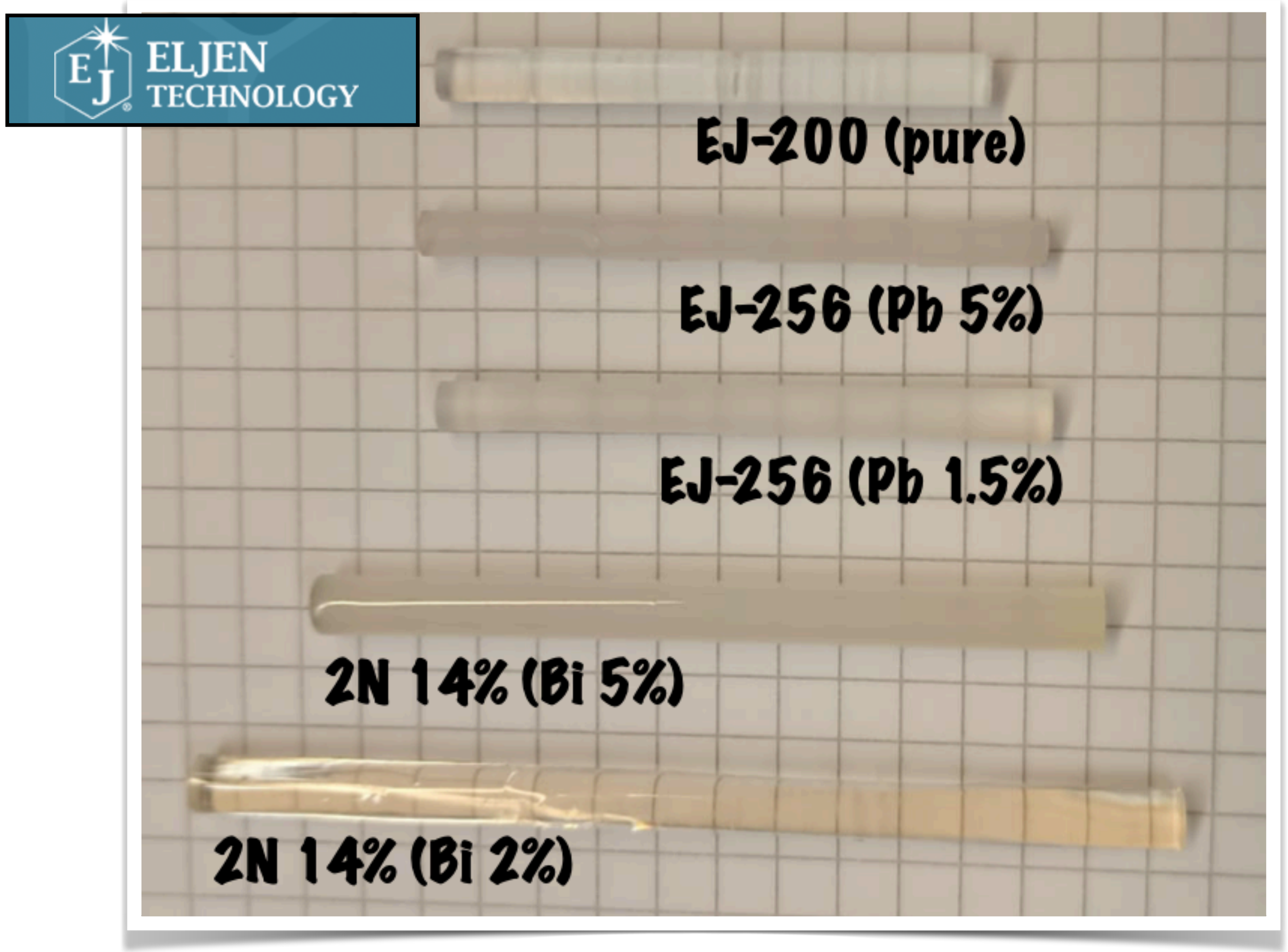
High-Z doped plastic scintillators samples

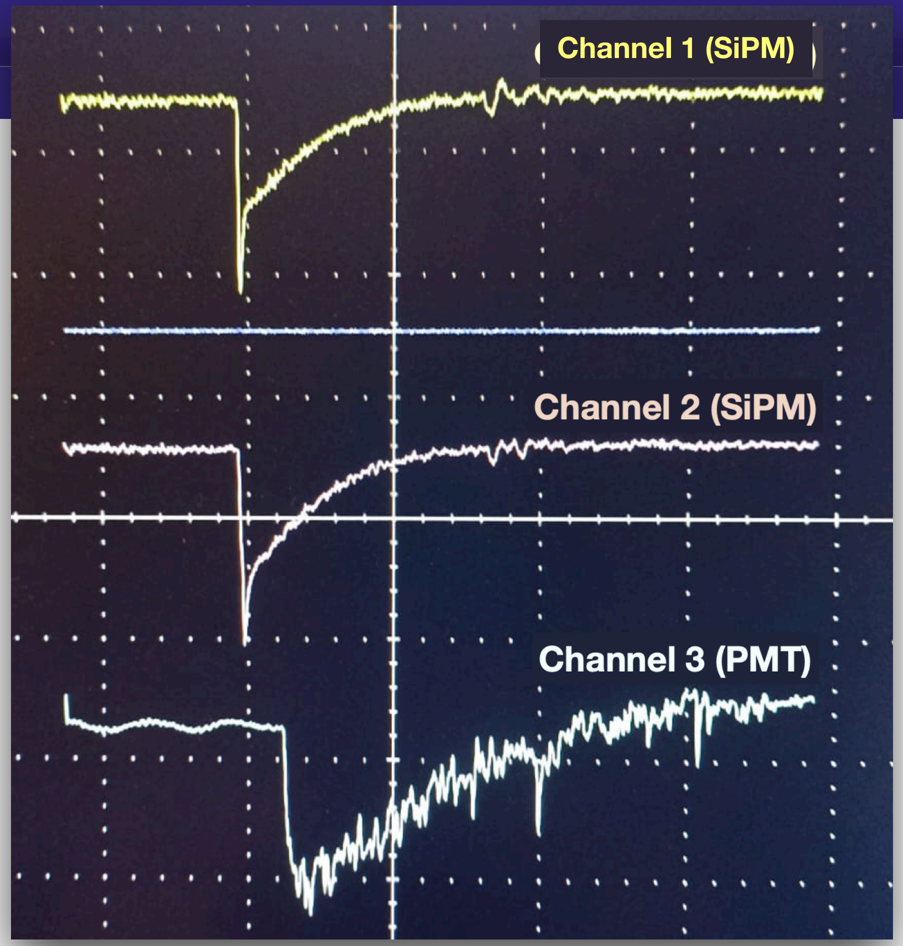


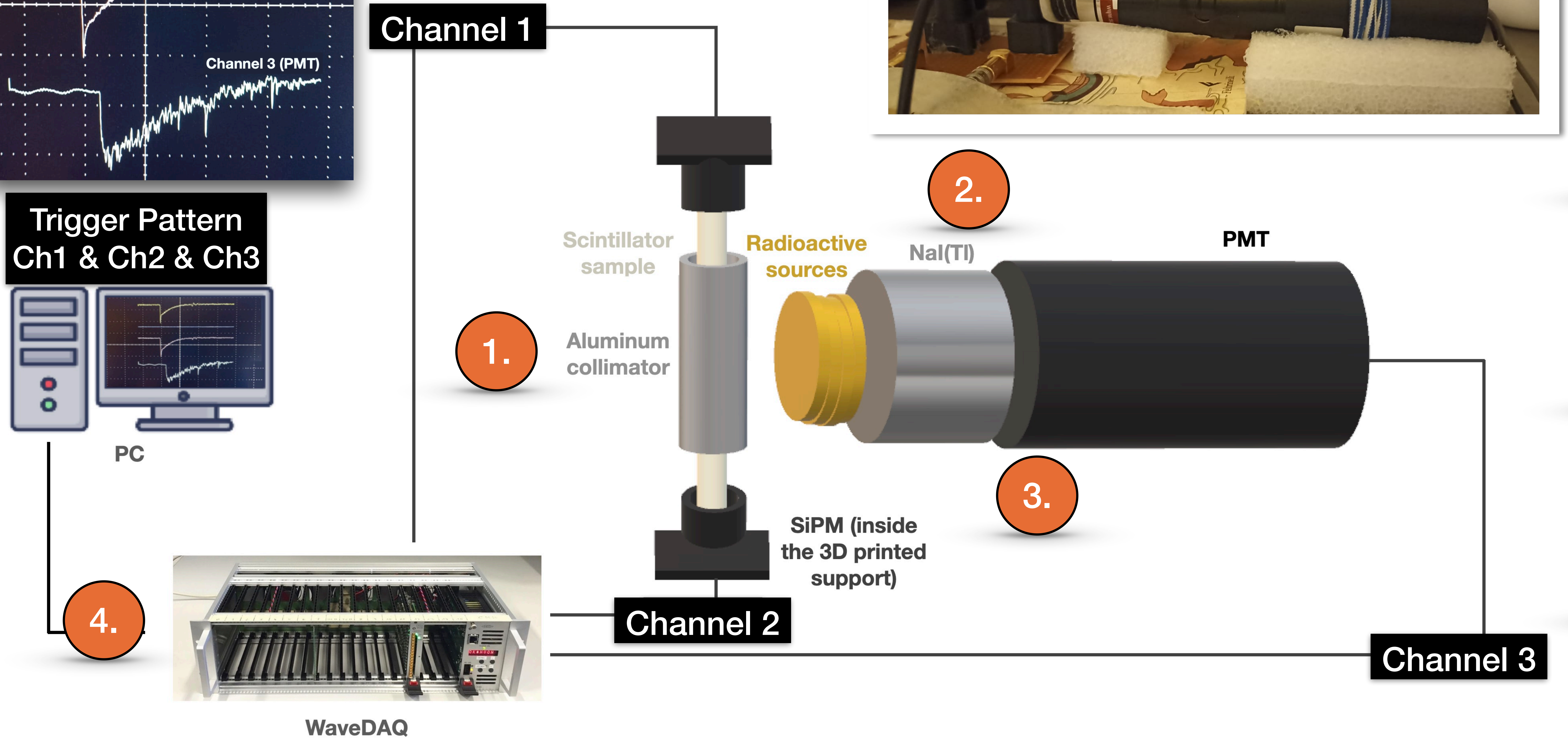
NEW!

Data Sheet	EJ-200 (pure)	EJ-256 (Pb 1.5%)	EJ-256 (Pb 5%)
Light Output	10000 γ/MeV	7700 γ/MeV	5200 γ/MeV
λ of Max Emission	425 nm	425 nm	425 nm
Decay Time	2.1 ns	2.1 ns	2.1 ns
Density	1.023 g/cm³	1.037 g/cm³	1.081 g/cm³
Polymer Base	PVT	PVT	PVT

- The **non-doped scintillator** is used as a **control** for the data acquisition and data analysis procedures.
- The **doped commercial scintillators** are used for **comparison** with the LEOS prototypes.

Experimental set-up





1. For the plastic scintillator: **double-sided readout** performed by **SiPMs**
2. **Triggering mechanism based on a NaI(Tl) scintillator read by a PMT**
3. **Radioactive source that is able to emit back-to-back photons → triple-coincidence**
4. Data acquisition and pre-processing performed by **WaveDAQ**

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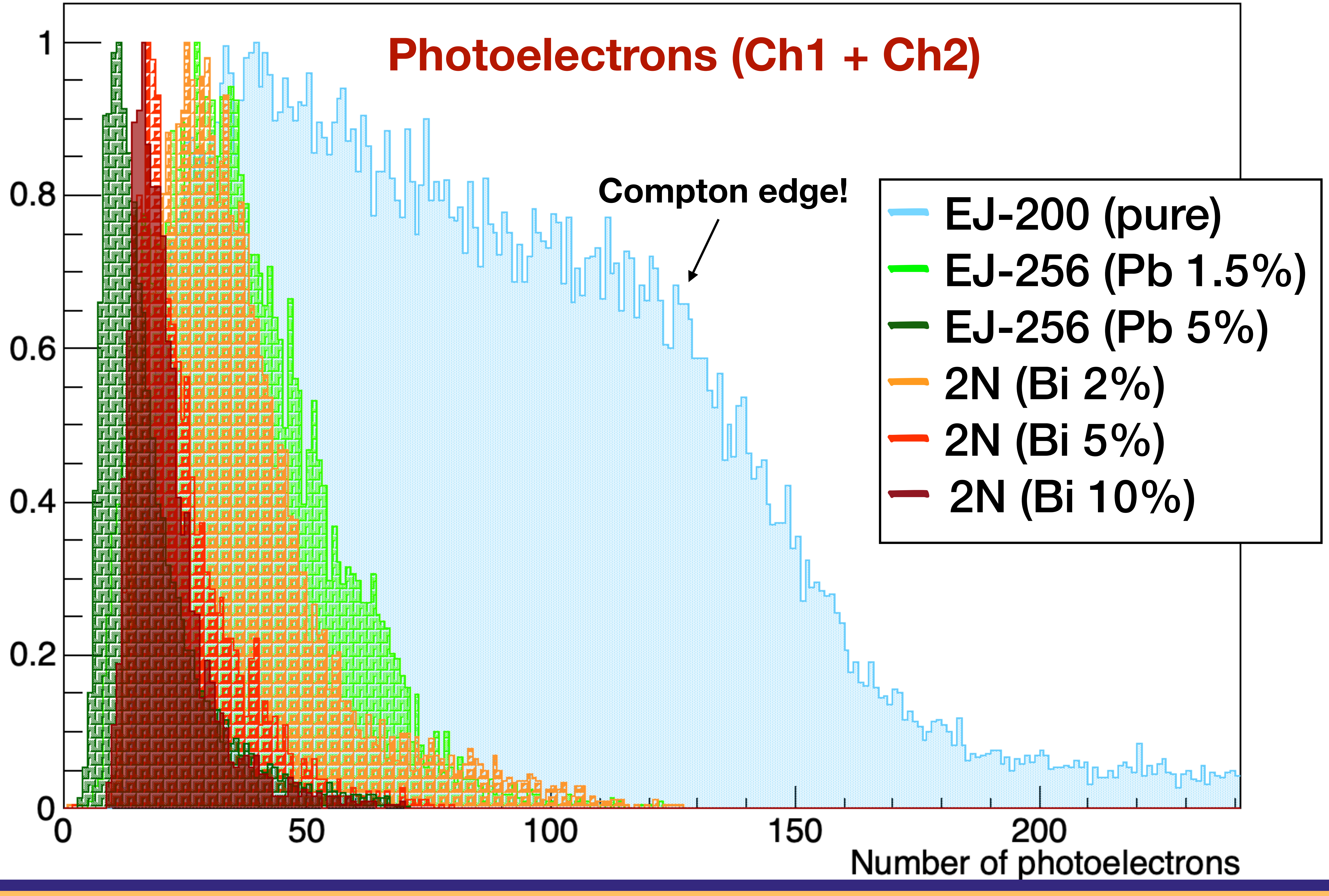
Light output estimation

As expected the histograms exhibit the **Compton spectrum** of the 511-keV photons emitted by Sodium-22.

- Histograms of the number of photoelectrons for each sample.
- Sum of the photoelectrons collected by the two channels in each event.
- Events selected through the "cut" conditions.
- Normalization between 0 and 1.



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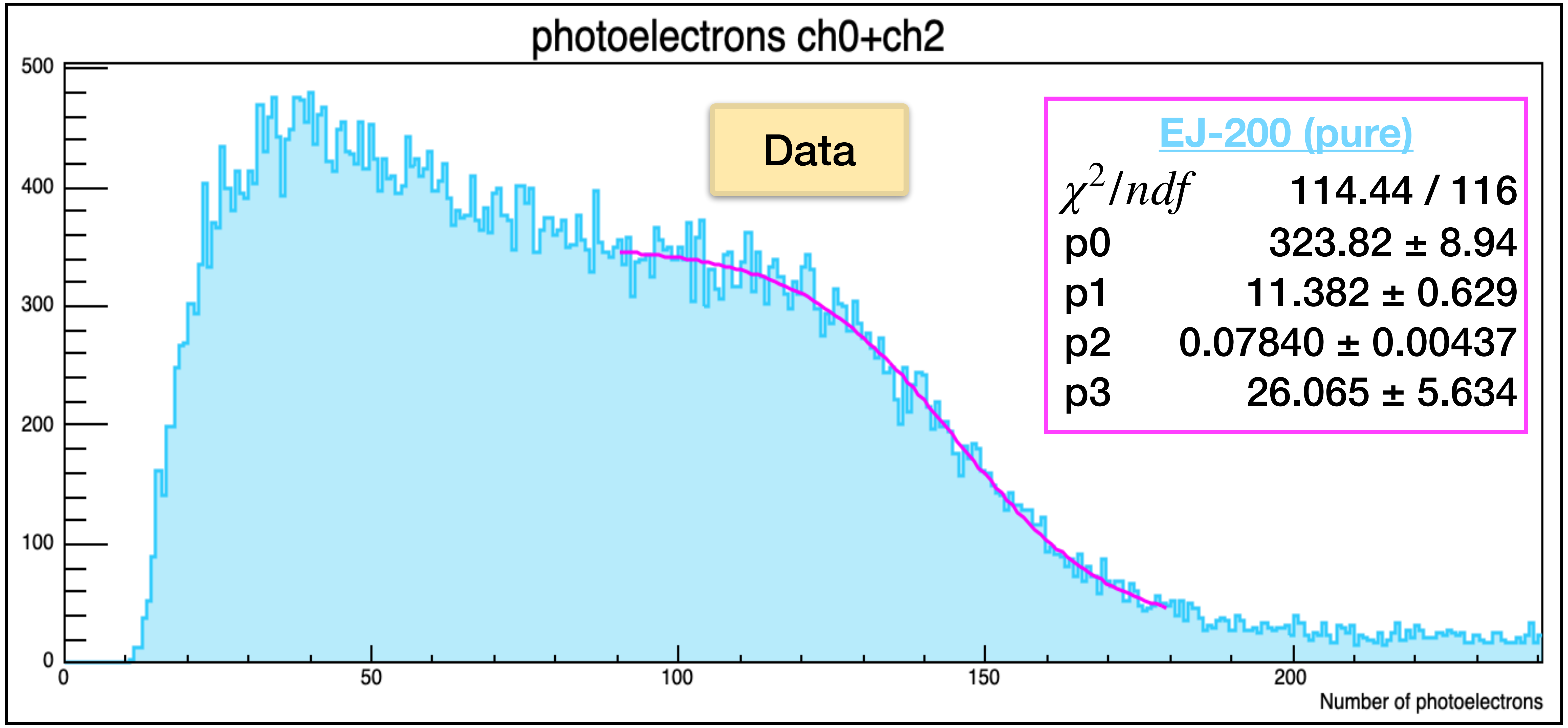
Light output estimation



$$f(x) = \frac{p_0}{1 + e^{p_2 x - p_1}} + p_3$$

- In order to characterize the light output of the samples, it was necessary to **select a specific point on the Compton spectrum**.
- The idea was to **fit the Compton edge with a Fermi-Dirac function** and identify the Compton edge position with the abscissa of the flex (x_0).
- An offset was used to parameterize the tail of the photopeak.

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Light output estimation

	Light Output (Data Sheet)	Expected #photoelectrons	Measured #photoelectrons
EJ-200 (pure)	10000 γ/MeV	~148	145 ± 11
EJ-256 (Pb 1.5%)	7700 γ/MeV	~114	45 ± 12
EJ-256 (Pb 5%)	5200 γ/MeV	~77	14 ± 3
2N (Bi 2%)	—	—	42 ± 5
2N (Bi 5%)	—	—	17 ± 4
2N (Bi 5%)	—	—	21 ± 2

- The number of expected photoelectrons has been calculated with equation $N_{ph.el.} = L.O. \cdot E_{\gamma} \cdot PDE \cdot C.E.$
- The agreement between expected and measured $N_{ph.el.}$ in the case of the non-doped sample suggests that the measurement procedure and the computation can be correct.
- The discrepancy in the cases of the doped samples can be mainly ascribed to the lack of transparency and to the machining of the samples.

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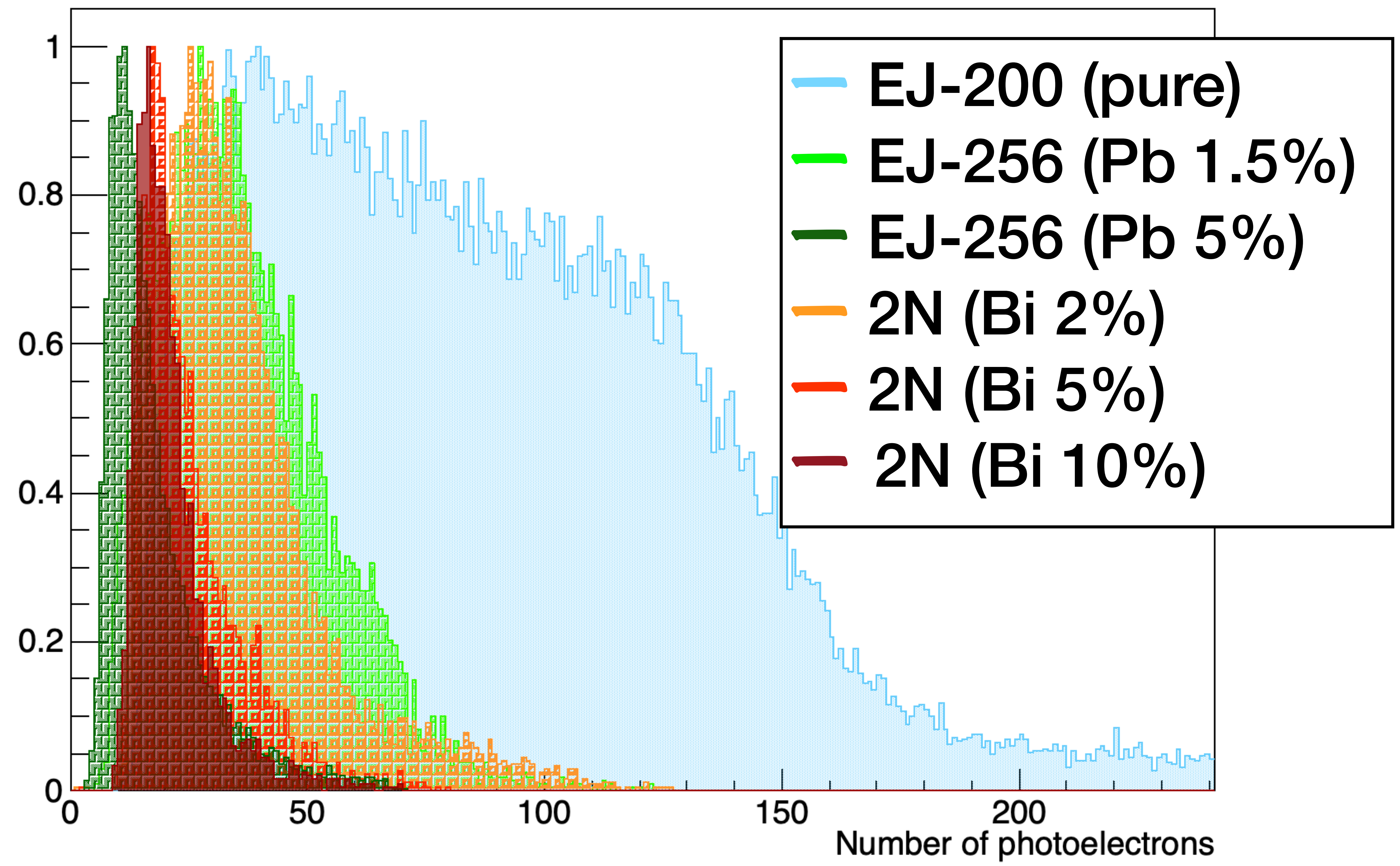
Results

Sample	Measured #photoelectrons
EJ-200 (pure)	145 ± 9
EJ-256 (Pb 1.5%)	45 ± 10
EJ-256 (Pb 5%)	14 ± 1
2N (Bi 2%)	42 ± 3
2N (Bi 5%)	17 ± 2
2N (Bi 10%)	21 ± 1



- The **non-doped sample shows better performances** since it is advantaged by the absence of impurities.
- The **10% doped sample shows promising performances** in terms of light output, time resolution and rise time: this is an important result since samples with this dopant concentration are not available on the market.
- The **2N-based samples have better timing performances** with respect to the commercial counterparts.

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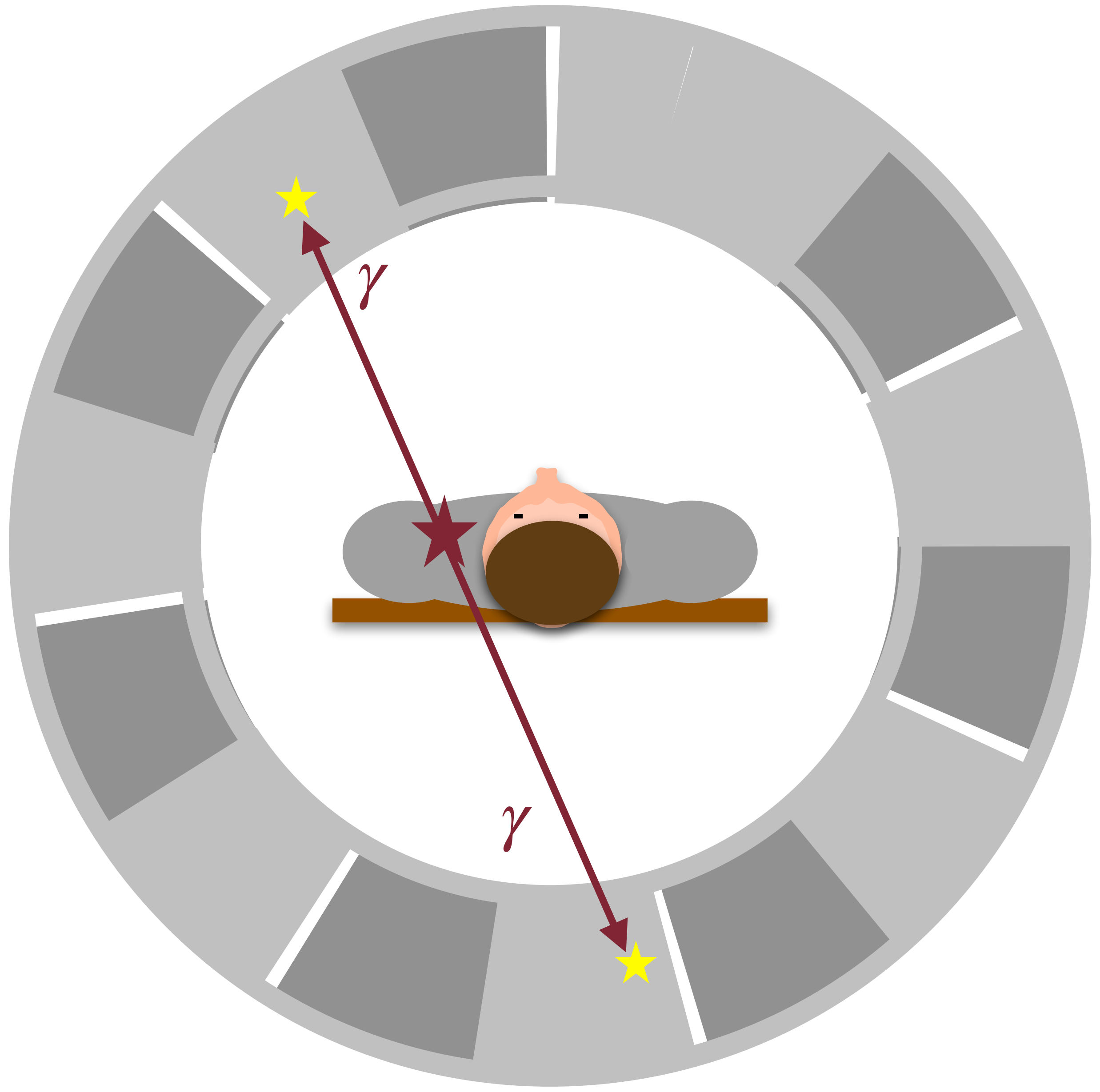
Possible other application: Time resolution

The timing properties of the samples were characterized starting from the **time difference histograms**. The histograms have been **fitted with the sum of two Gaussian functions** with an offset.



- Possible TOF-PET application if the scintillators show optimal timing performances!

- PET = Positron Emission Tomography
- TOF = Time of Flight

- The time difference between the detection of the two gamma rays allows to obtain information about the depth of interaction.

$\sigma(t) = (340 \pm 46)$ ps

Sample	Time resolution
EJ-200 (puro)	(176 ± 7) ps
EJ-256 (Pb 1.5%)	(360 ± 17) ps
EJ-256 (Pb 5%)	(520 ± 31) ps
2N (Bi 2%)	(233 ± 13) ps
2N (Bi 5%)	(278 ± 33) ps
2N (Bi 10%)	(340 ± 46) ps

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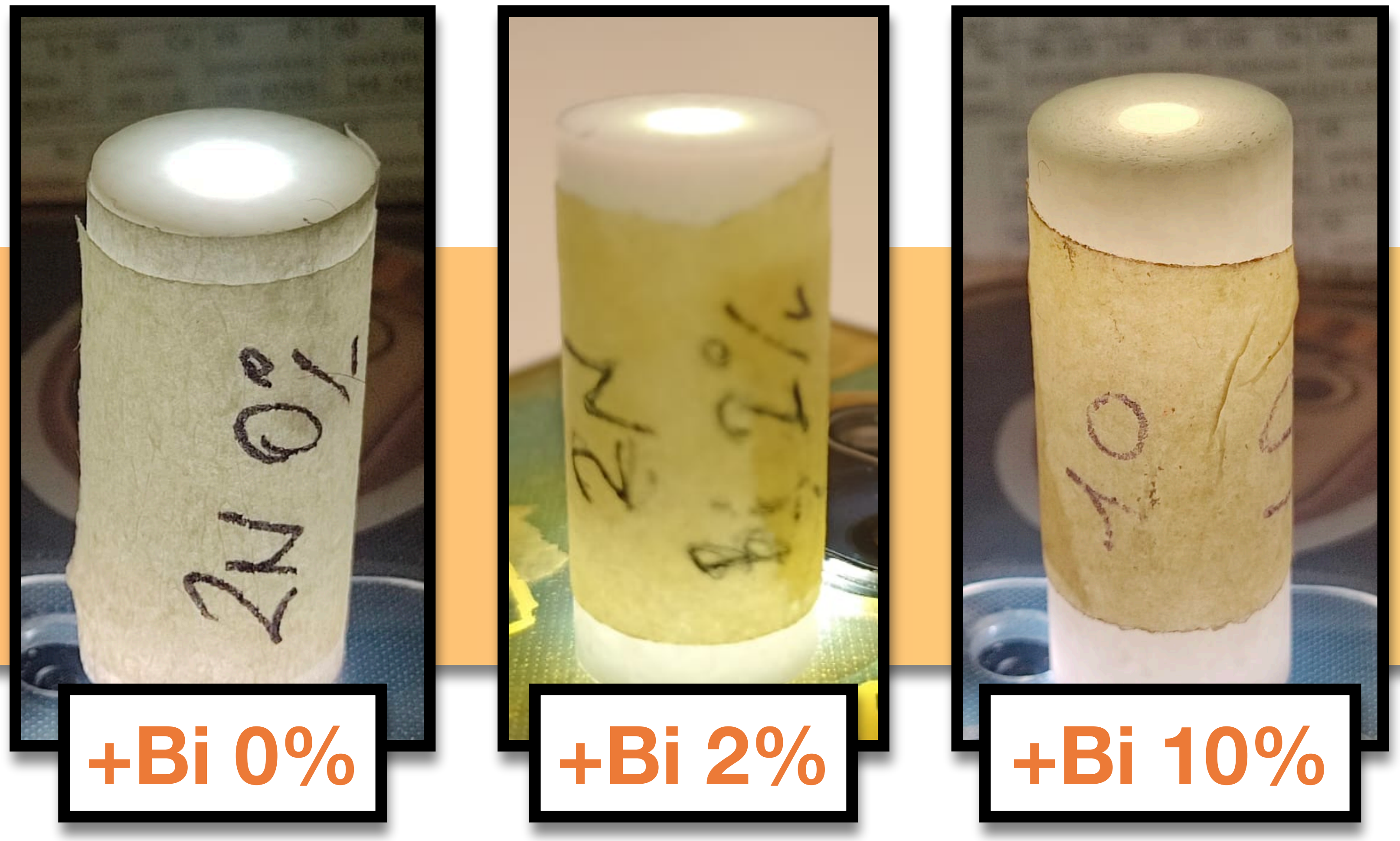
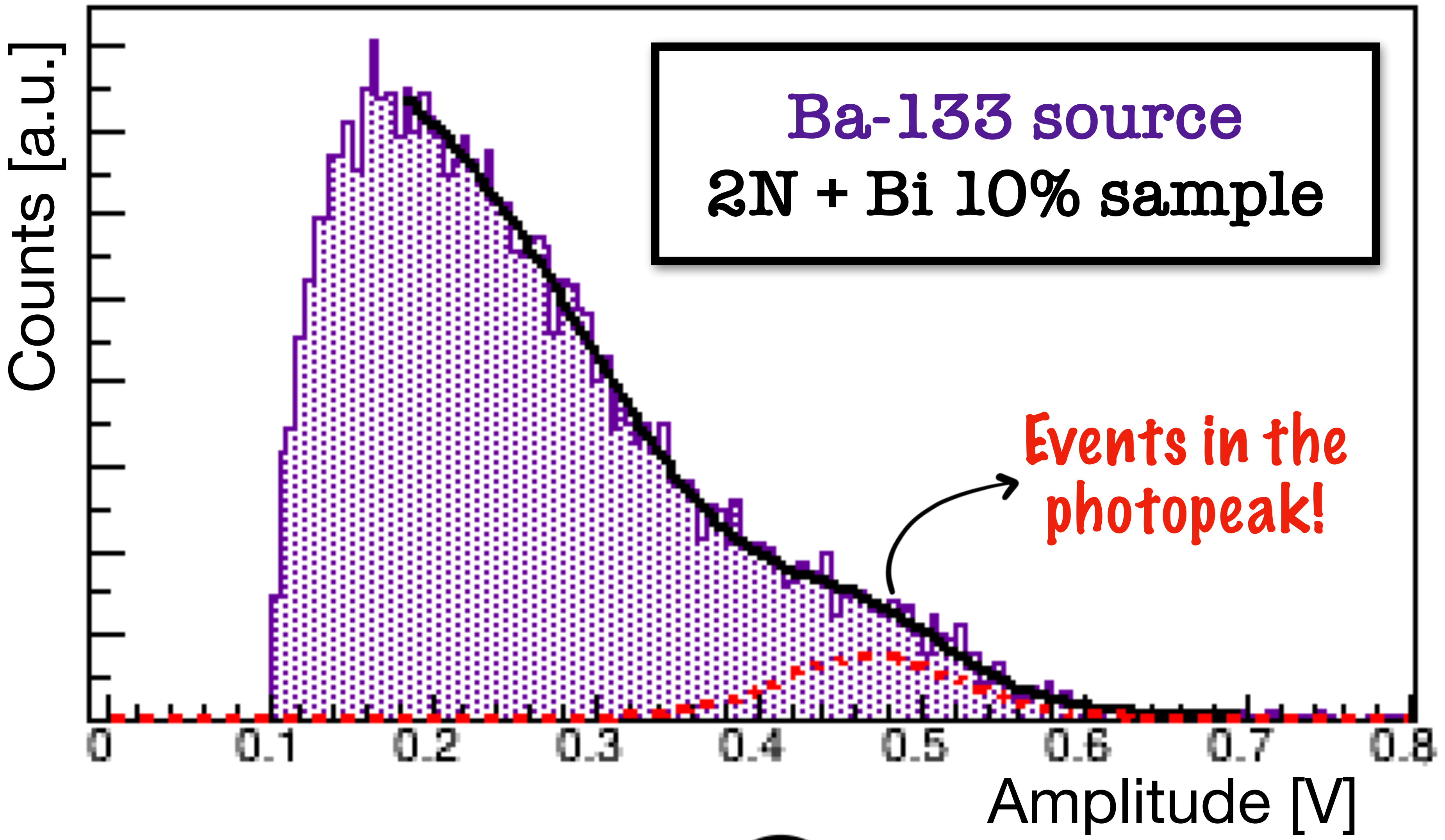
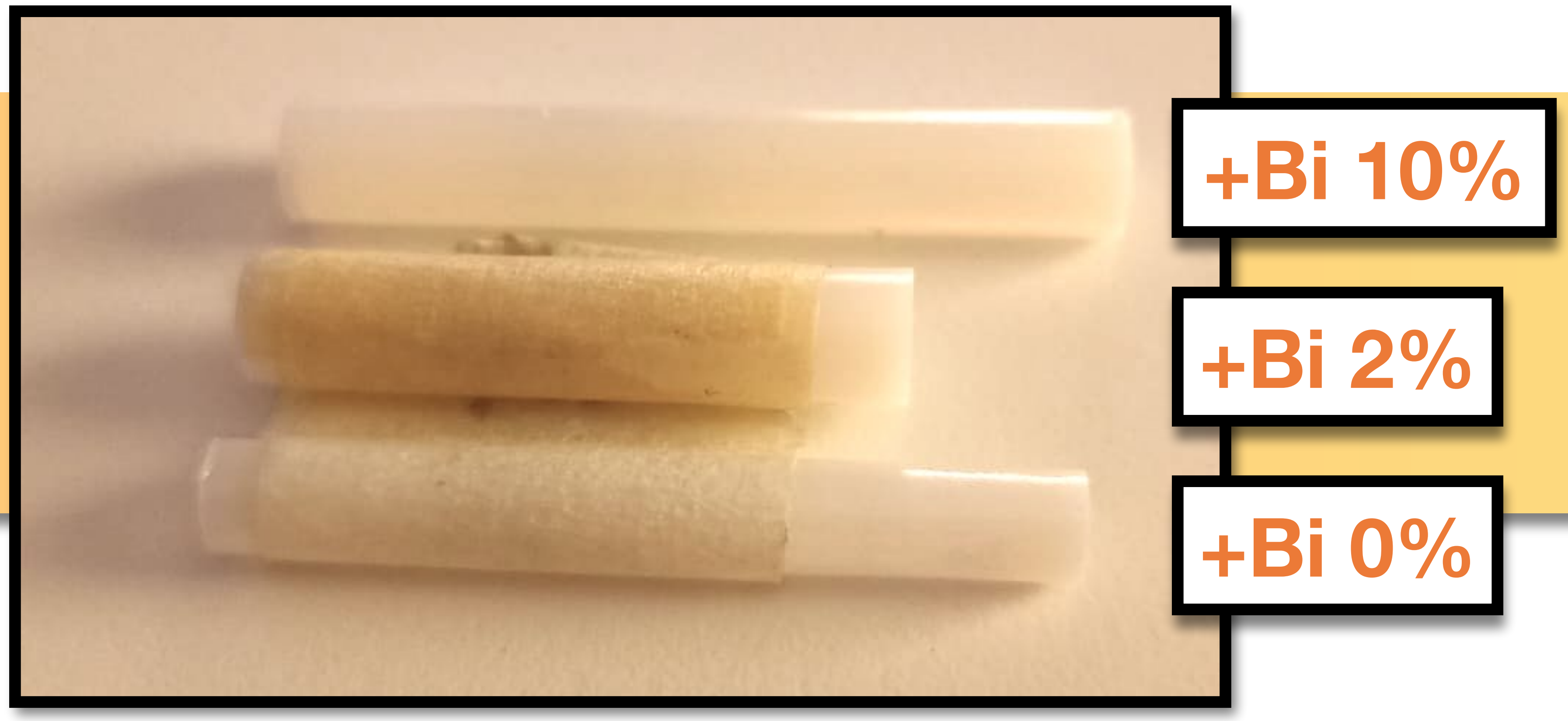
Next Steps

In order to improve the light collection efficiency, we produced samples where the polymerization process happens directly inside a teflon coating.



Scintillator	Light Output [a.u.]
2N + 0% Bi	100%
2N + 2% Bi	30%
2N + 10% Bi	25%





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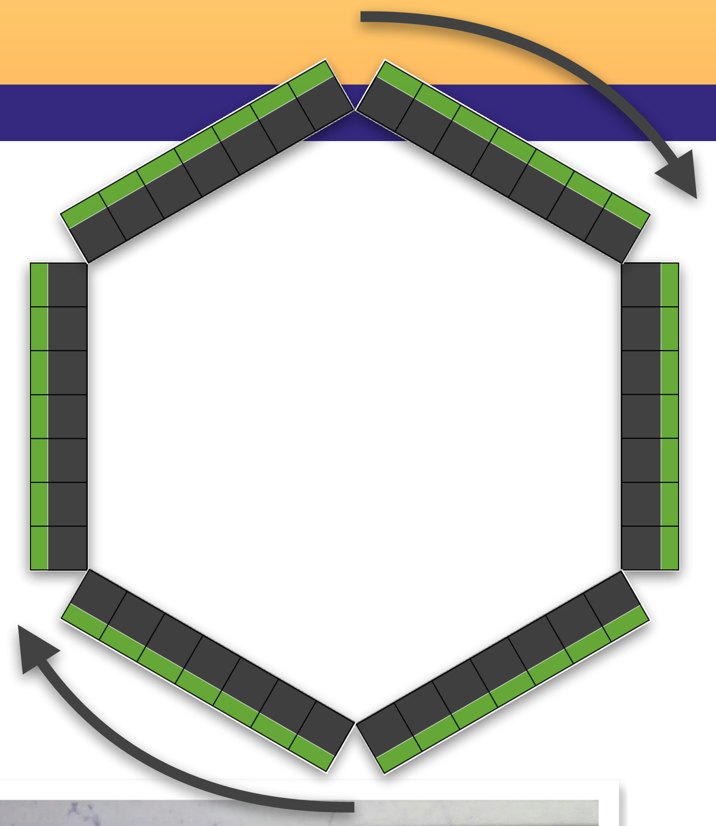
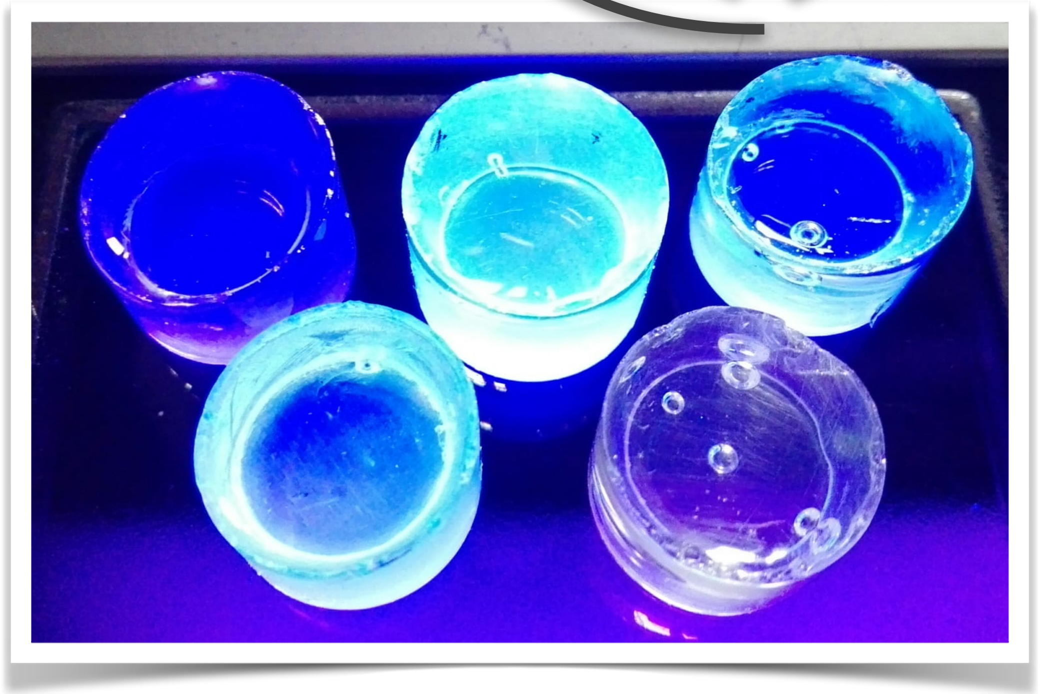
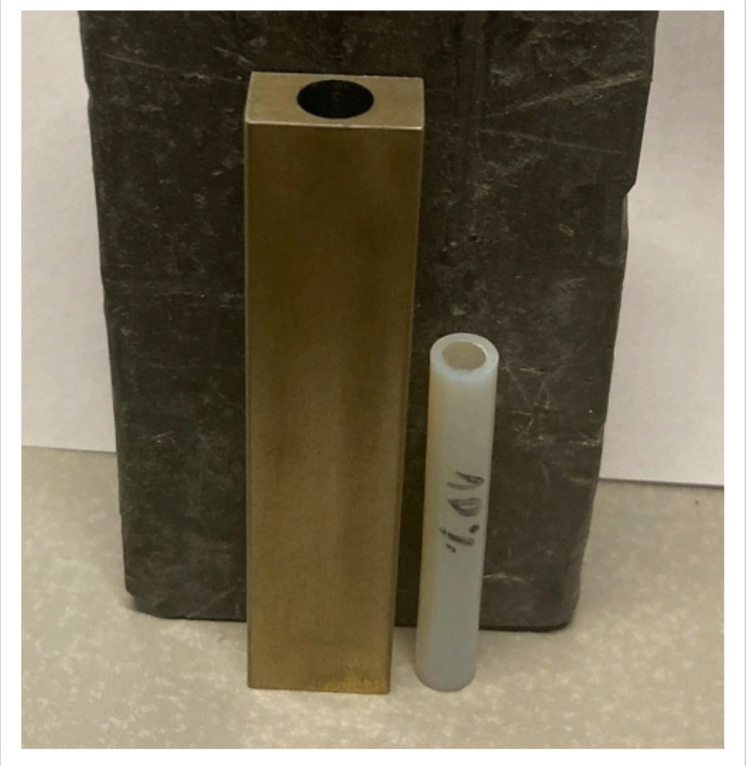
Conclusions



- The reSPECT project aim is to use **plastic scintillators** **enriched with high-Z impurities** for a new total body SPECT modular system, since they are extremely **fast, light, inexpensive and easy to shape**.



- The **2N samples, produced by LEOS laboratory,** has been selected to produce Bismuth-enriched samples, showing **promising performances in terms of light output and timing properties** with respect to commercial samples.



- Next steps are:
  1. New measurements with teflon coating;
  2. Optimization of the manipulation process for the commercial scintillators;
  3. New measurements with tungsten frame.

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THANK YOU FOR THE ATTENTION

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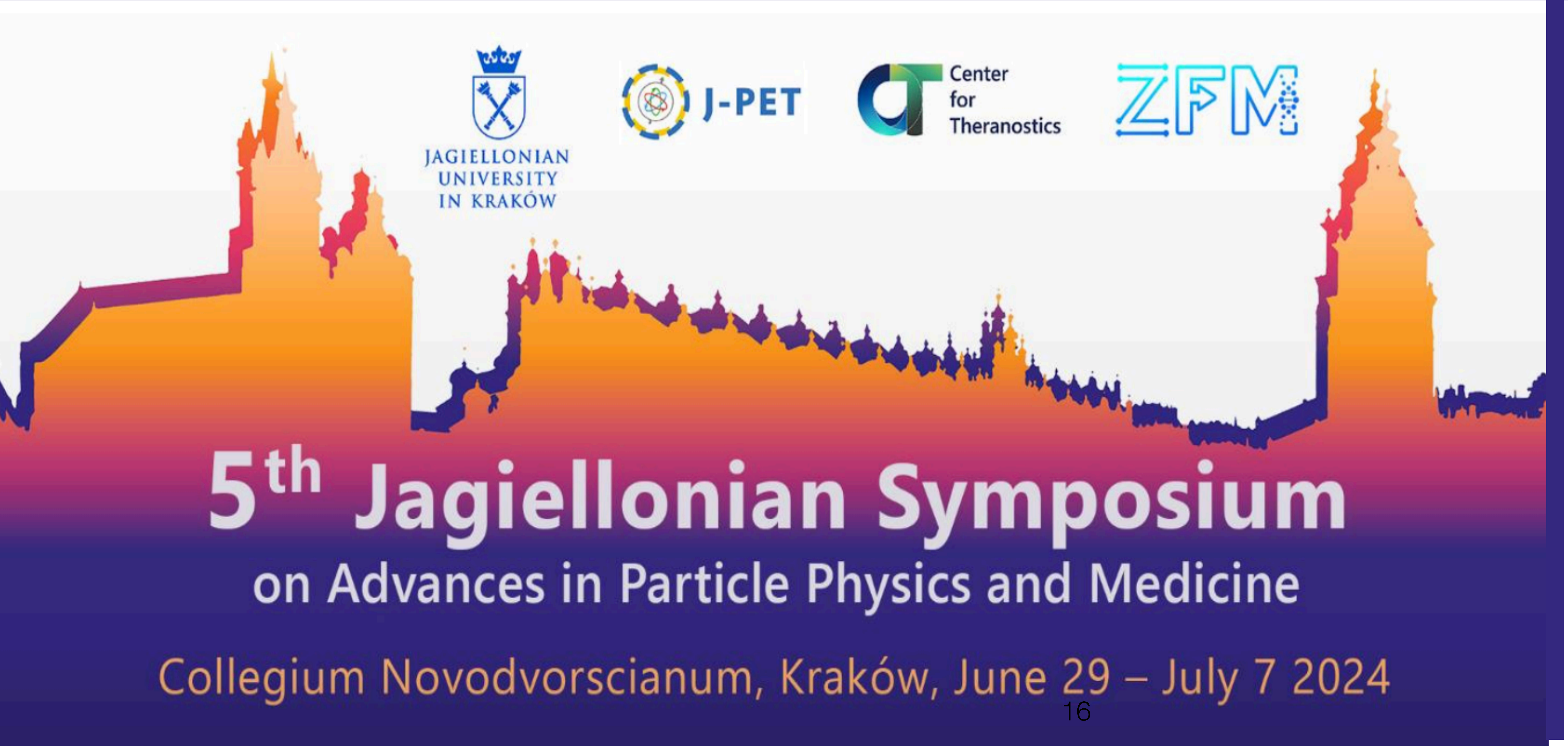

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Results and conclusions

Sample	Measured #photoelectrons	Time resolution	Rise Time (mean)
EJ-200 (puro)	145 ± 9	(176 ± 7) ps	(3.31 ± 0.01) ns
EJ-256 (Pb 1.5%)	45 ± 10	(360 ± 17) ps	(3.91 ± 0.02) ns
EJ-256 (Pb 5%)	14 ± 1	(520 ± 31) ps	(3.67 ± 0.02) ns
2N (Bi 2%)	42 ± 3	(233 ± 13) ps	(3.36 ± 0.01) ns
2N (Bi 5%)	17 ± 2	(278 ± 33) ps	(3.44 ± 0.03) ns
2N (Bi 10%)	21 ± 1	(340 ± 46) ps	(3.44 ± 0.03) ns

- The **non-doped sample shows better performances** since it is advantaged by the absence of impurities.
- The **2N-based samples have better timing performances** with respect to the commercial counterparts.
- The **10% doped sample shows promising performances** in terms of light output, time resolution and rise time: this is an important result since samples with this dopant concentration are not available on the market.

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reSPECT: where we are

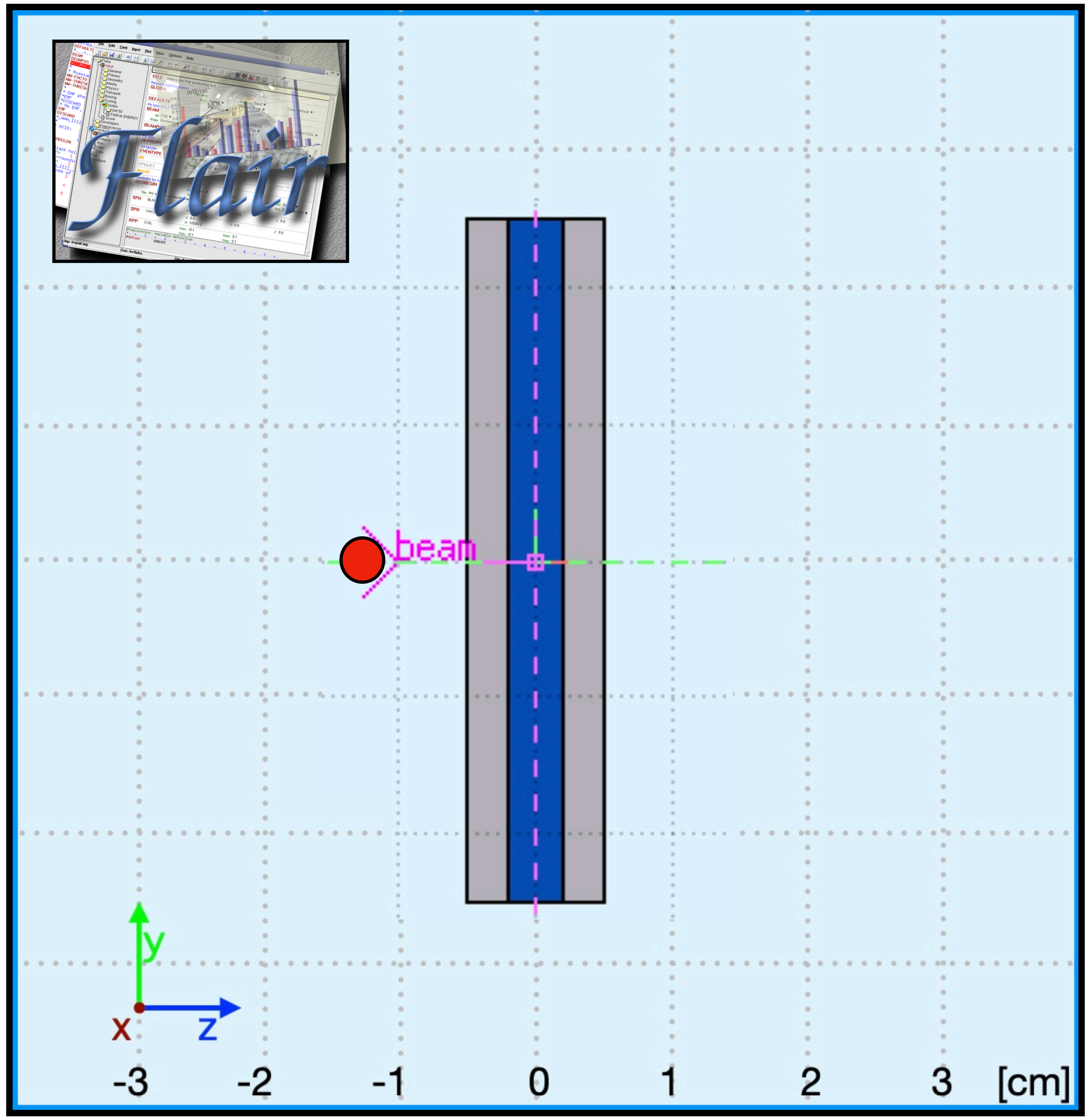
- We synthesized several new organic scintillating molecules (T2, 2N, 1N, 2B…) containing aromatic fragments.
- Plastic scintillators embedding our scintillating molecules in a polyvinyl-tuoluene (PVT) matrix were produced.
- We tested the scintillator prototypes in order to determine their performances in terms of light output and time resolution.

FLUOROPHORES CONCENTRATION

Fluorophores concentrations ranging from 1% to 30% have been studied, but the best performances have been obtained with samples at 14%.

Monte Carlo simulations

- MC simulations are able to provide useful information for experiment design and data analysis.
- **FLUKA** is a general multipurpose tool that allows to **simulate particle transport and interaction with matter** by means of Monte Carlo techniques.



- Plastic Scintillator Sample
- Aluminum hollow cylinder
- Gamma Ray Source

- The simulated geometry reproduces the experimental conditions.
- The experimental resolution can be simulated by adding a **smearing** to the MC truth results.

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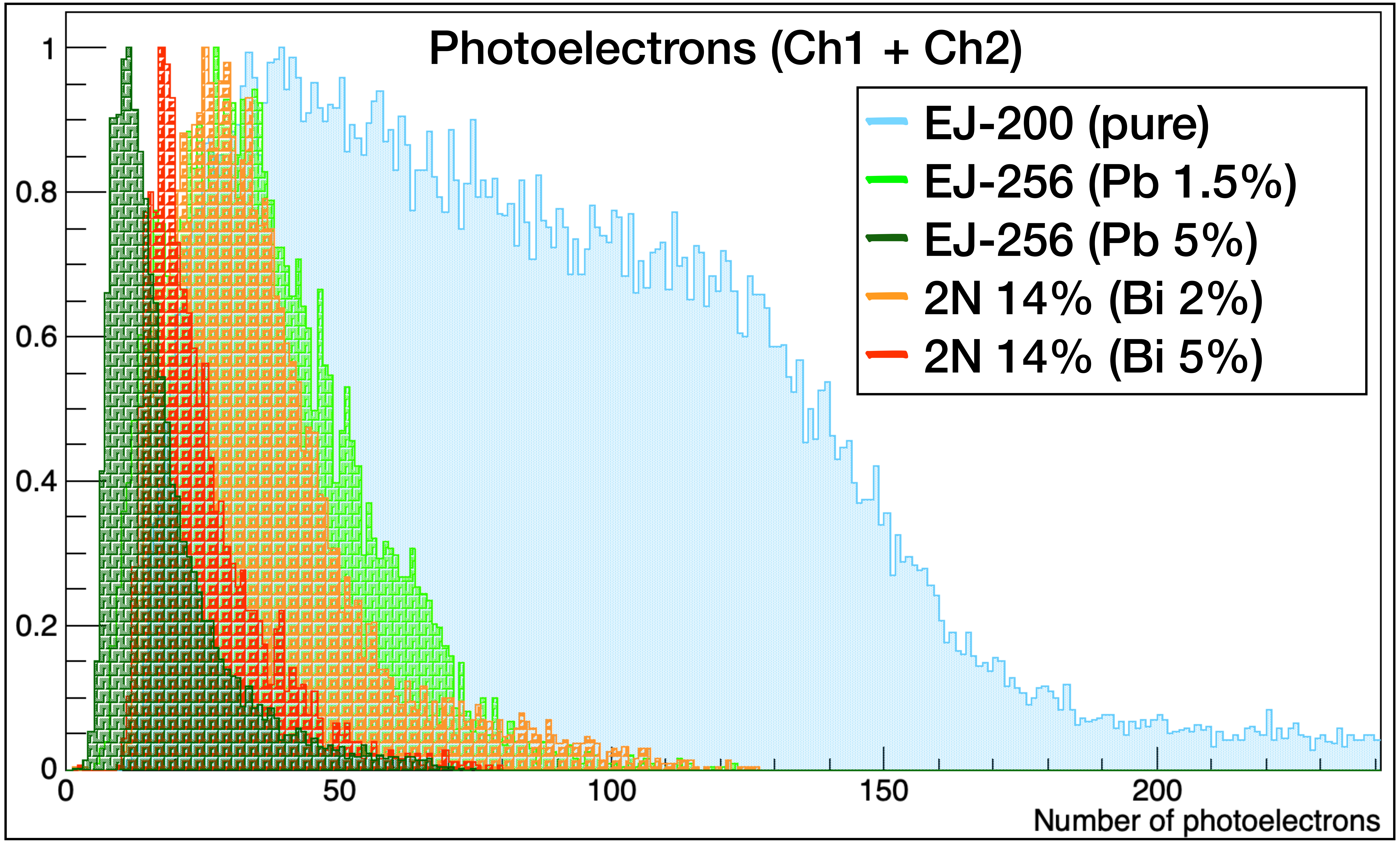
Light output estimation

As predicted by the **Monte Carlo simulations**, the histograms exhibit the **Compton spectrum** of the 511-keV photons emitted by Sodium-22.

- Histograms of the number of photoelectrons for each sample.
- Sum of the number of photoelectrons collected by the two channels in each event.
- Events selected through the "cut" conditions.
- Normalization between 0 and 1.



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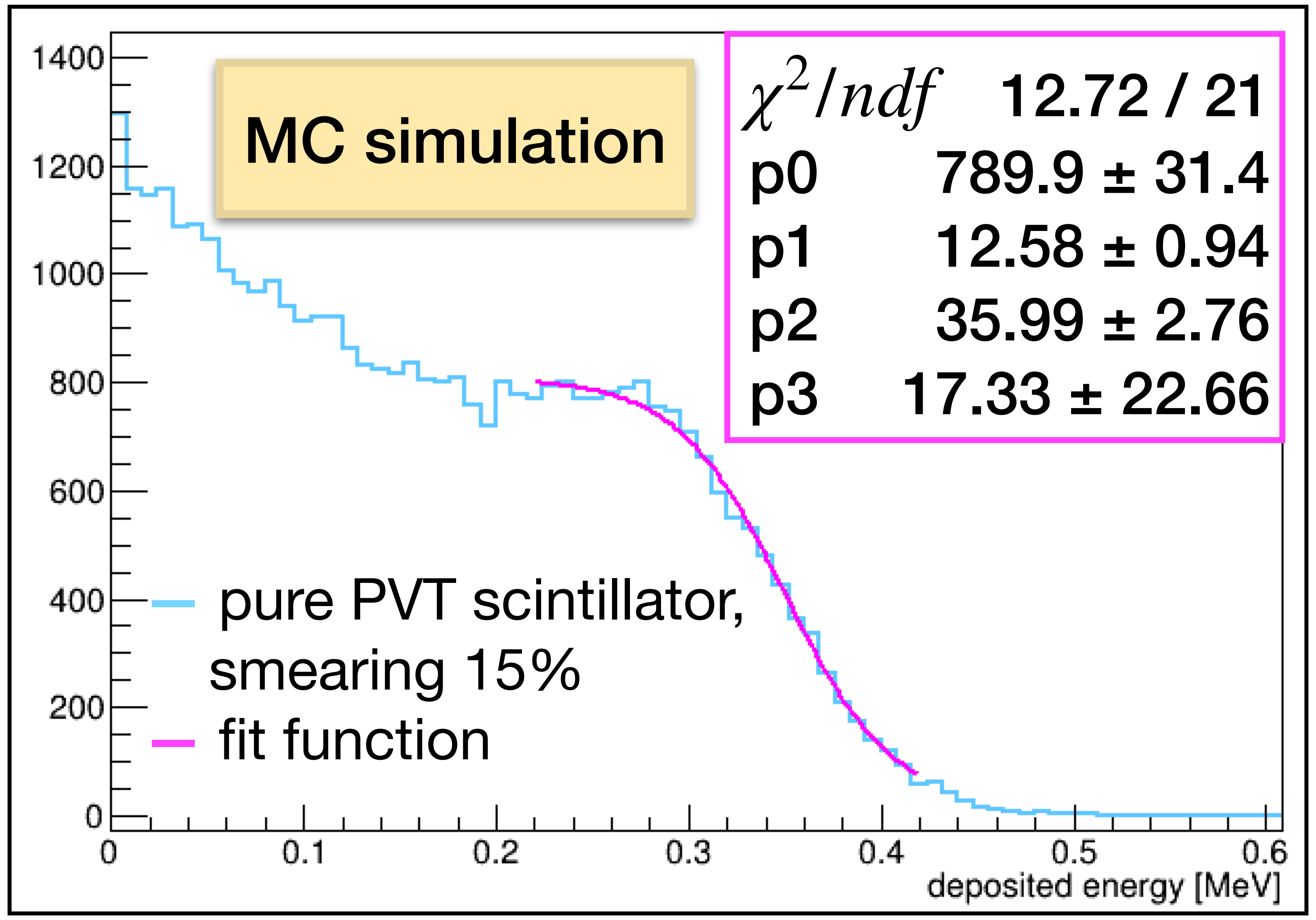
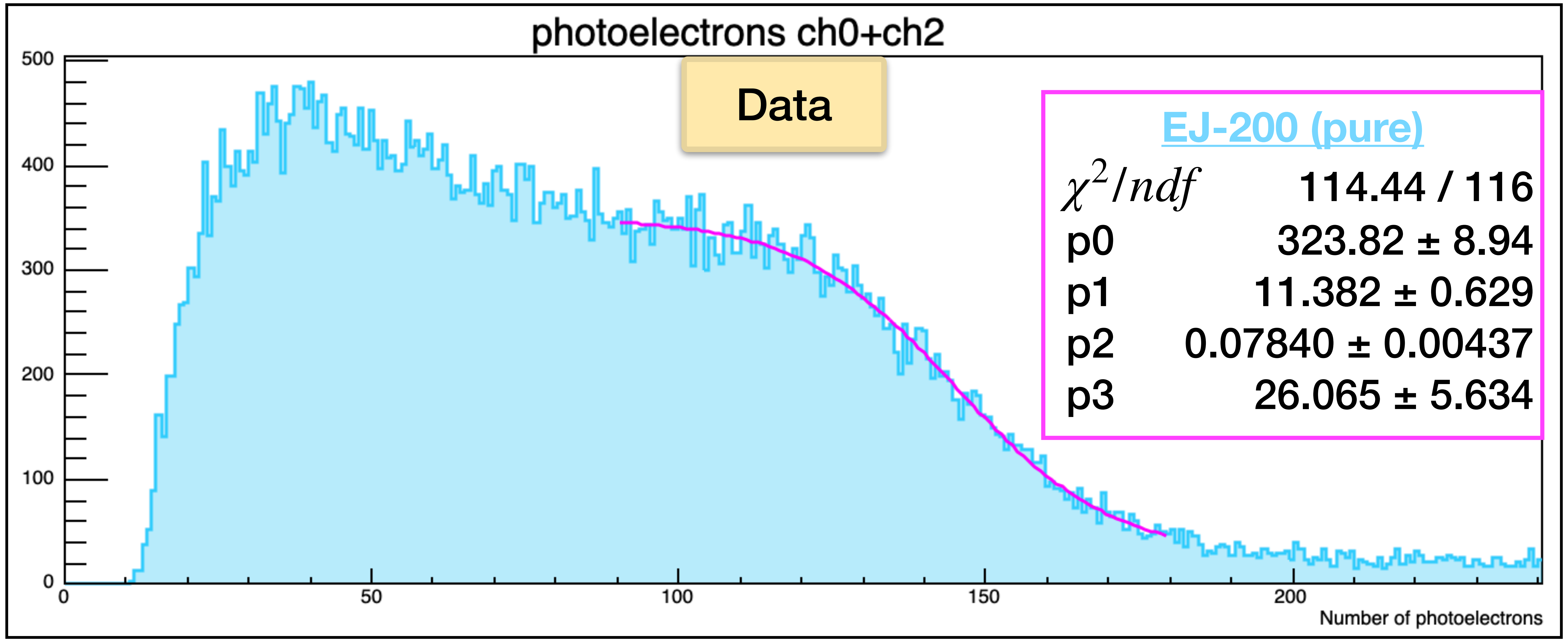
Light output estimation





$$f(x) = \frac{p_0}{1 + e^{p_2 x - p_1}} + p_3$$

- In order to characterize the light output of the samples, it was necessary to **select a specific point on the Compton spectrum**.
- The idea was to **fit the Compton edge with a Fermi-Dirac function** and identify the Compton edge position with the abscissa of the flex (x_0).
- An offset was used to parameterize the tail of the photopeak.

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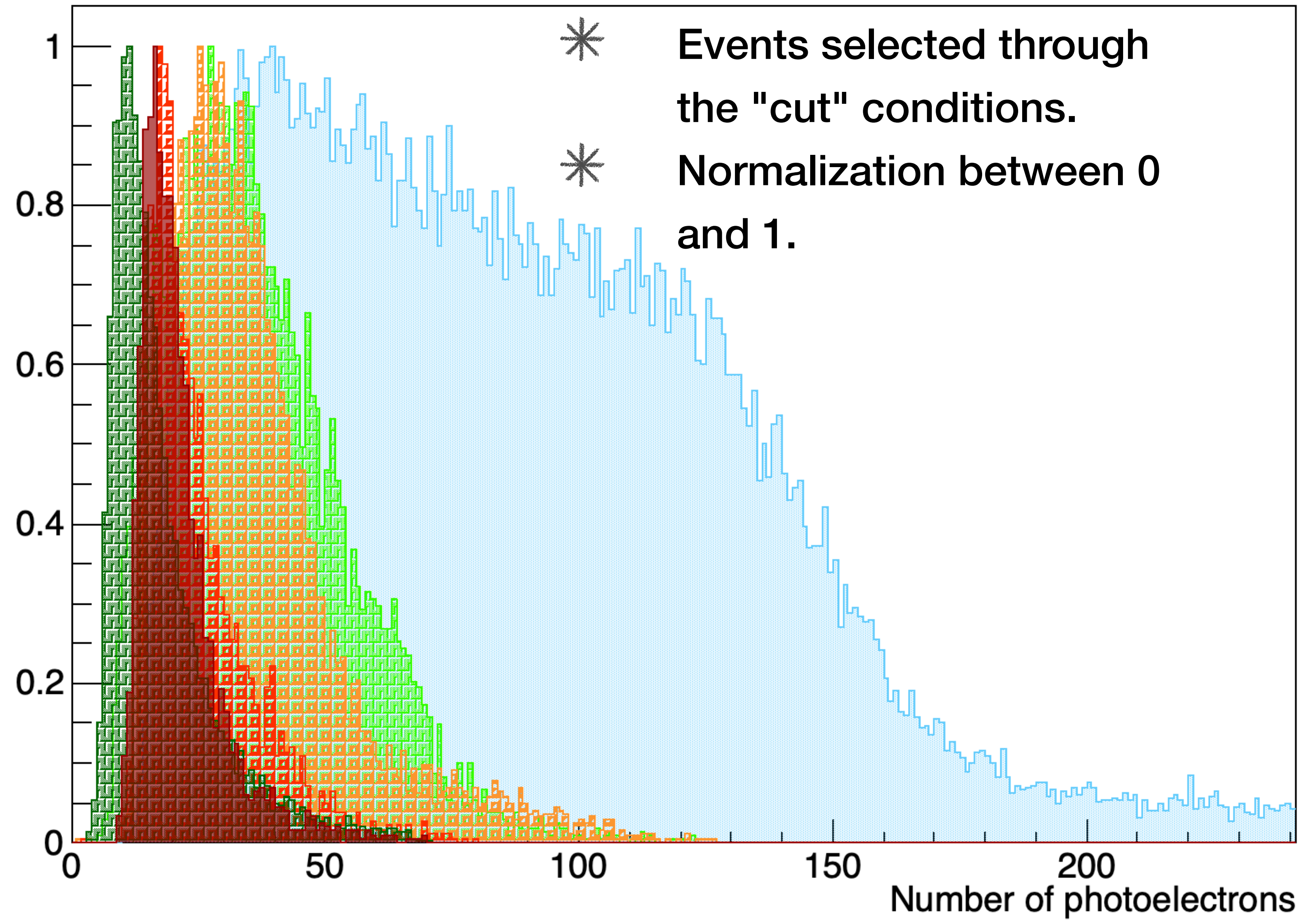
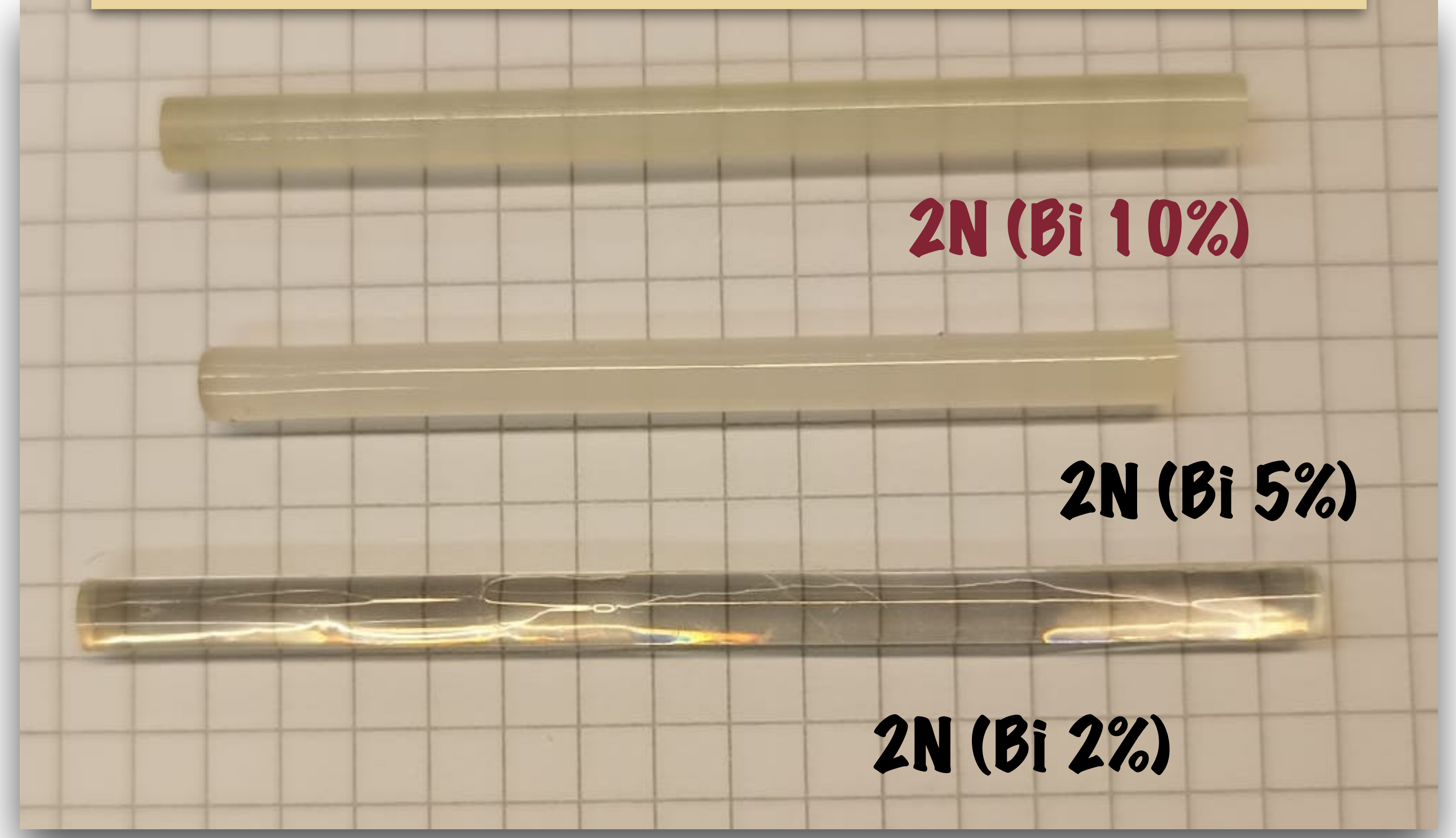
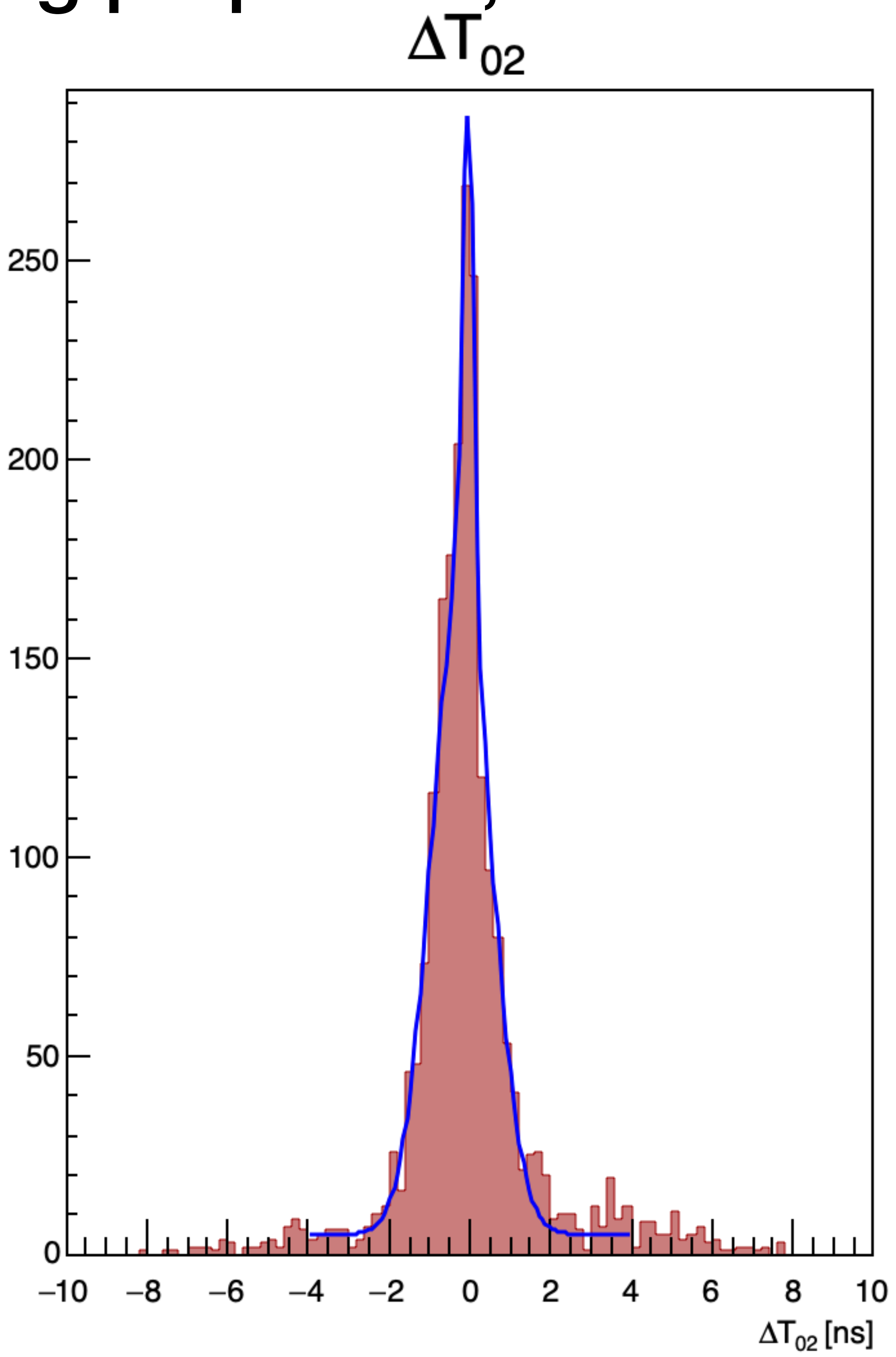
Light output estimation Bi (10%)

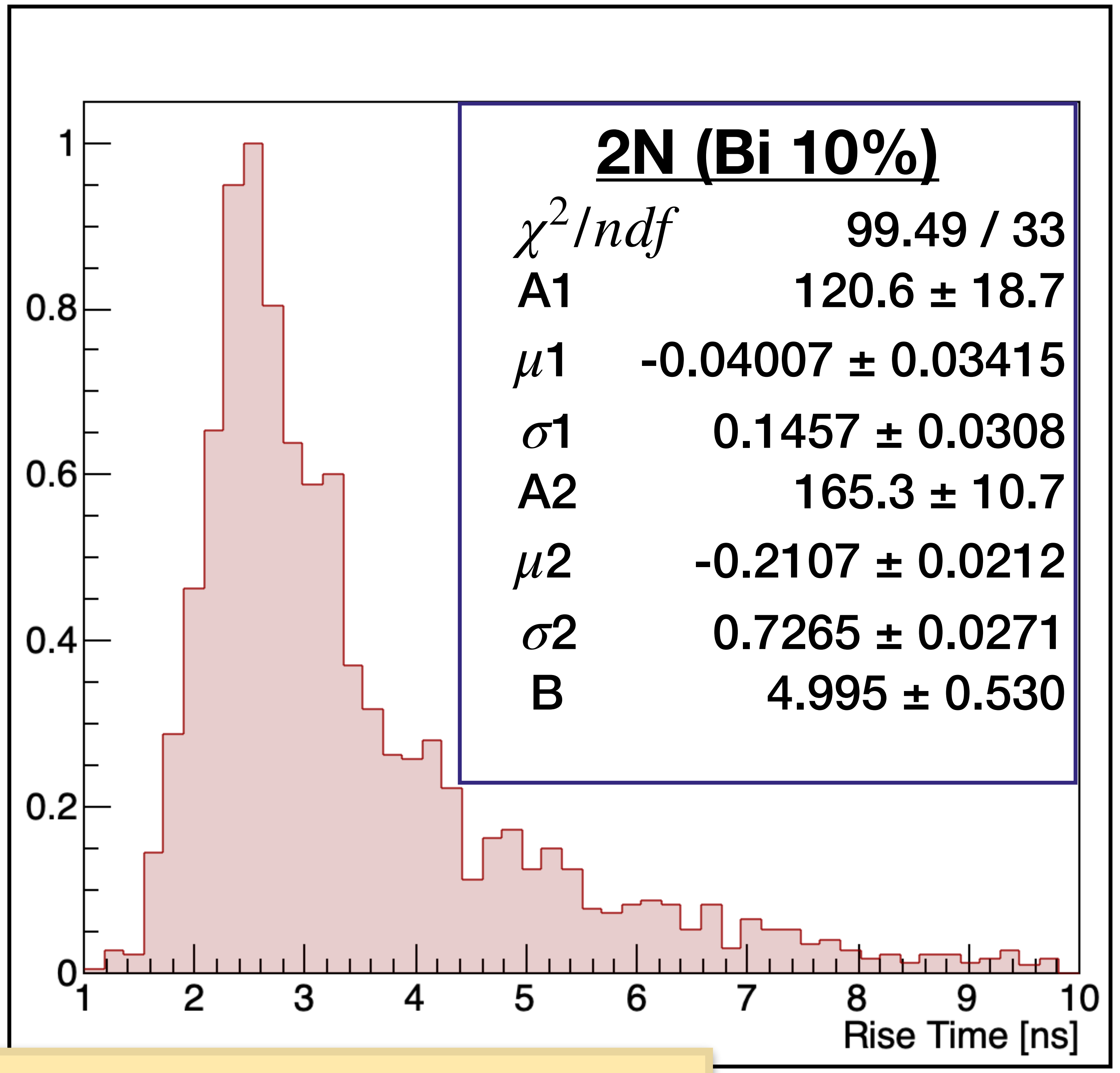


Number of photoelectrons at the Compton edge: 21 ± 2



Despite the high dopant concentration, the sample shows competitive timing properties, both in terms of time resolution and rise time.



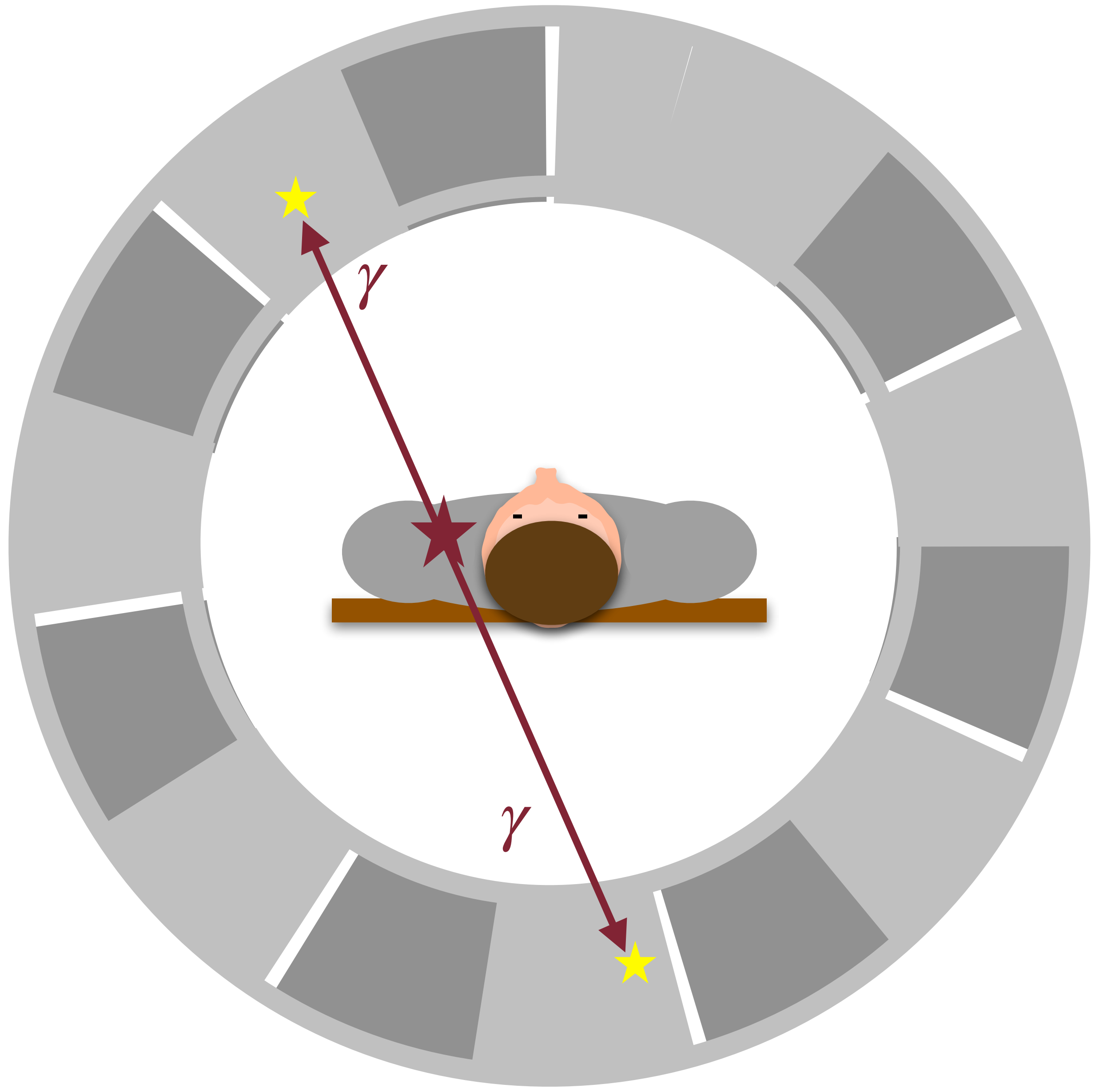


σ(t) = (340 ± 46) ps

- Samples with this dopant concentration are not available on the market due to their poor performances.
- More statistics is needed to look for events in the photopeak.

Time resolution

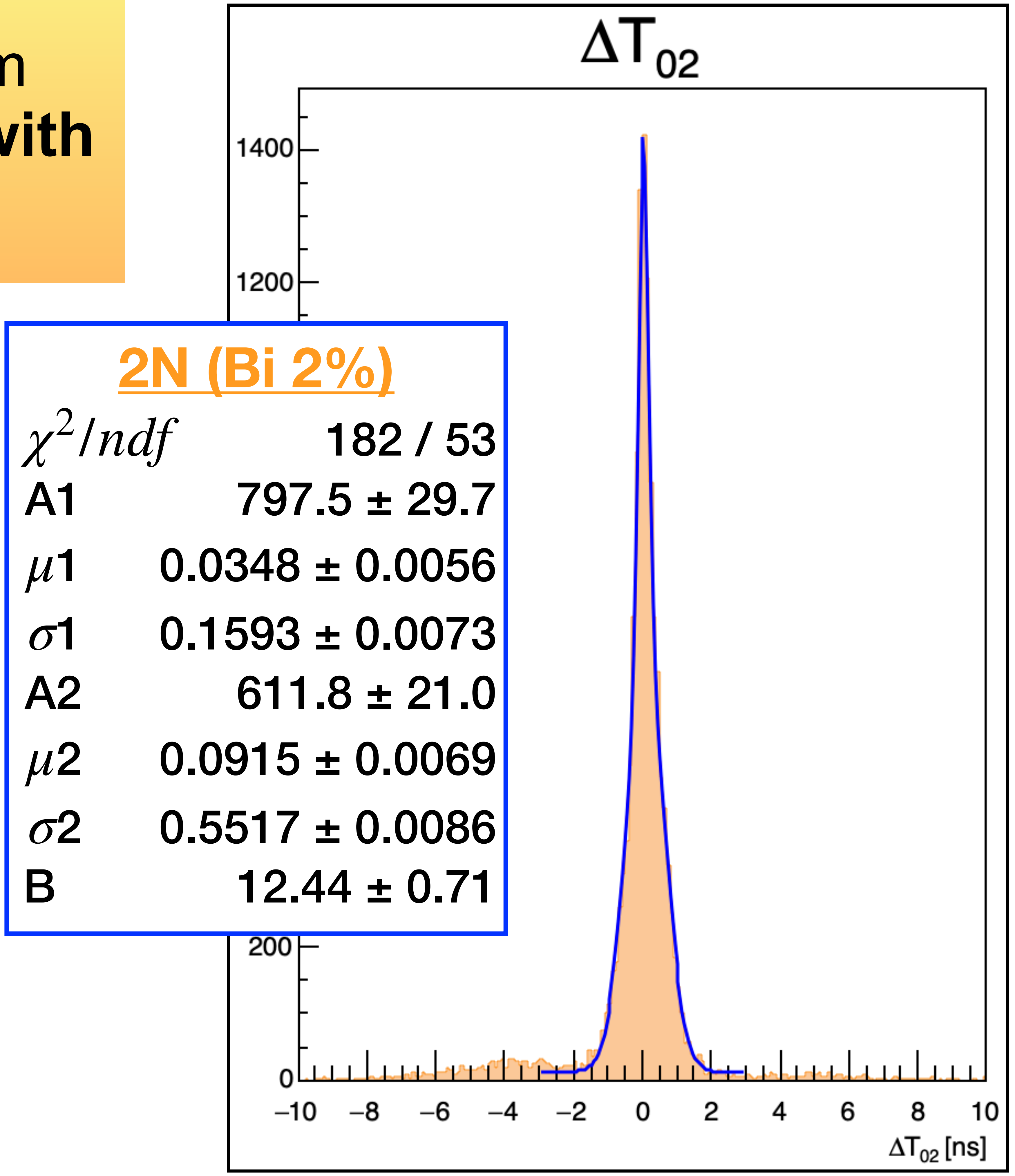
The timing properties of the samples were characterized starting from the **time difference histograms**. The histograms have been **fitted with the sum of two Gaussian functions** with an offset.



Possible TOF-PET application if the scintillators show optimal timing performances!

PET = Positron Emission Tomography
TOF = Time of Flight

The time difference between the detection of the two gamma rays allows to obtain information about the depth of interaction.



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Italiadomani PIANO NAZIONALE DI RIPRESA E RESILIENZA

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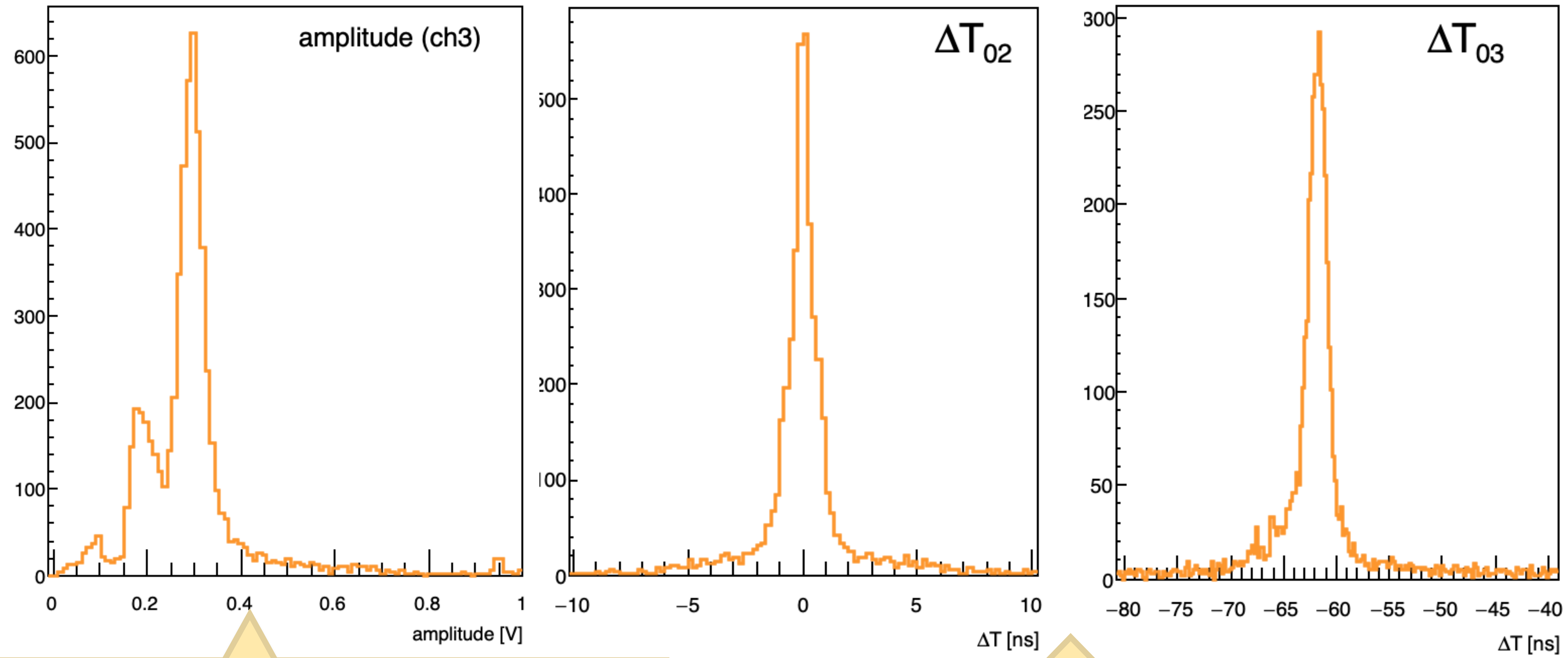
Event selection conditions

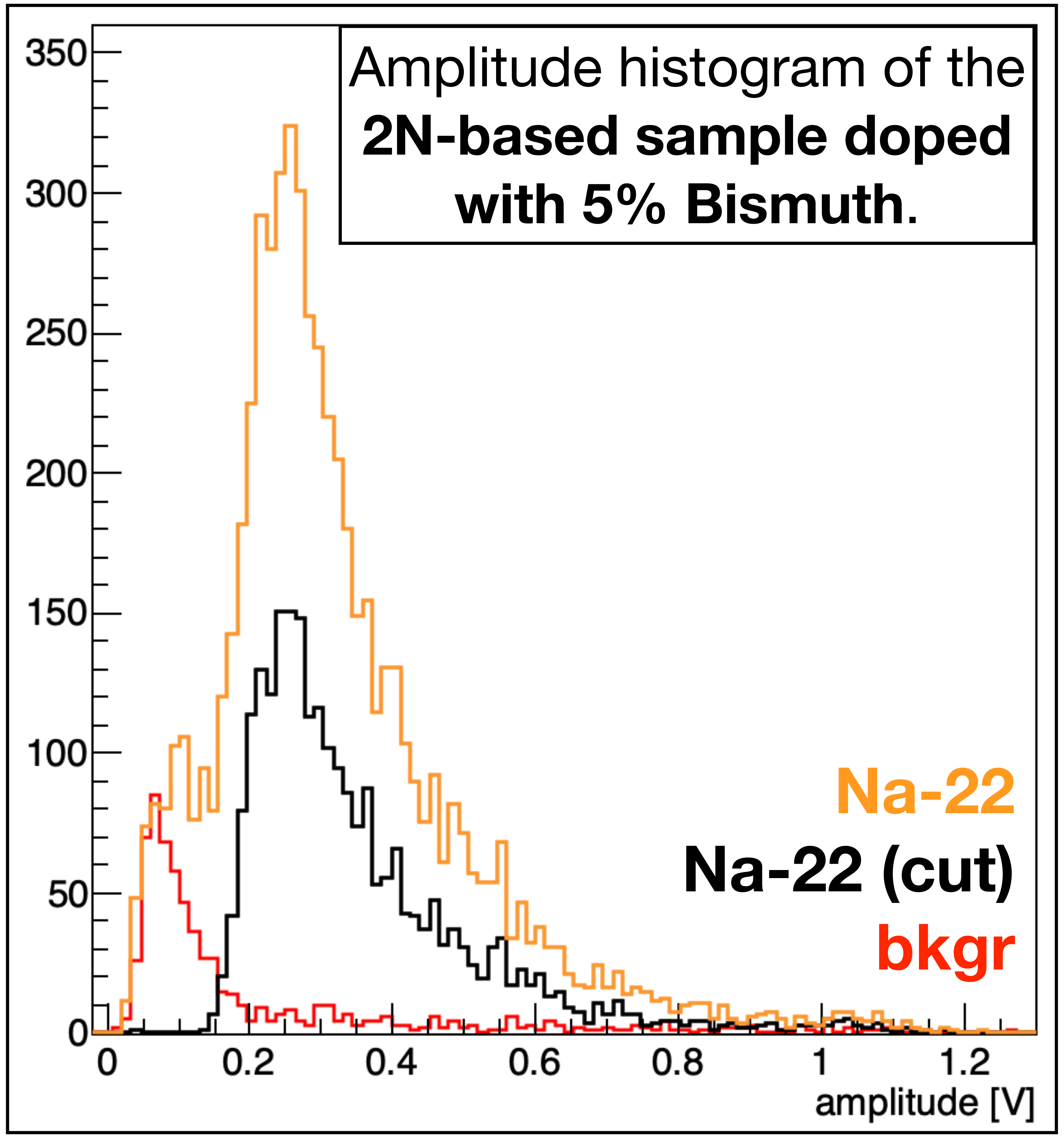
For each sample, two measurements were carried out:

- **one with the Sodium-22 radioactive sources** placed between the plastic scintillator and the NaI(Tl)
- **the other without any radioactive source to study the background**.
- Among the data acquired with the Na-22 sources, **the "good" events were selected exploiting two conditions ("cut")**.

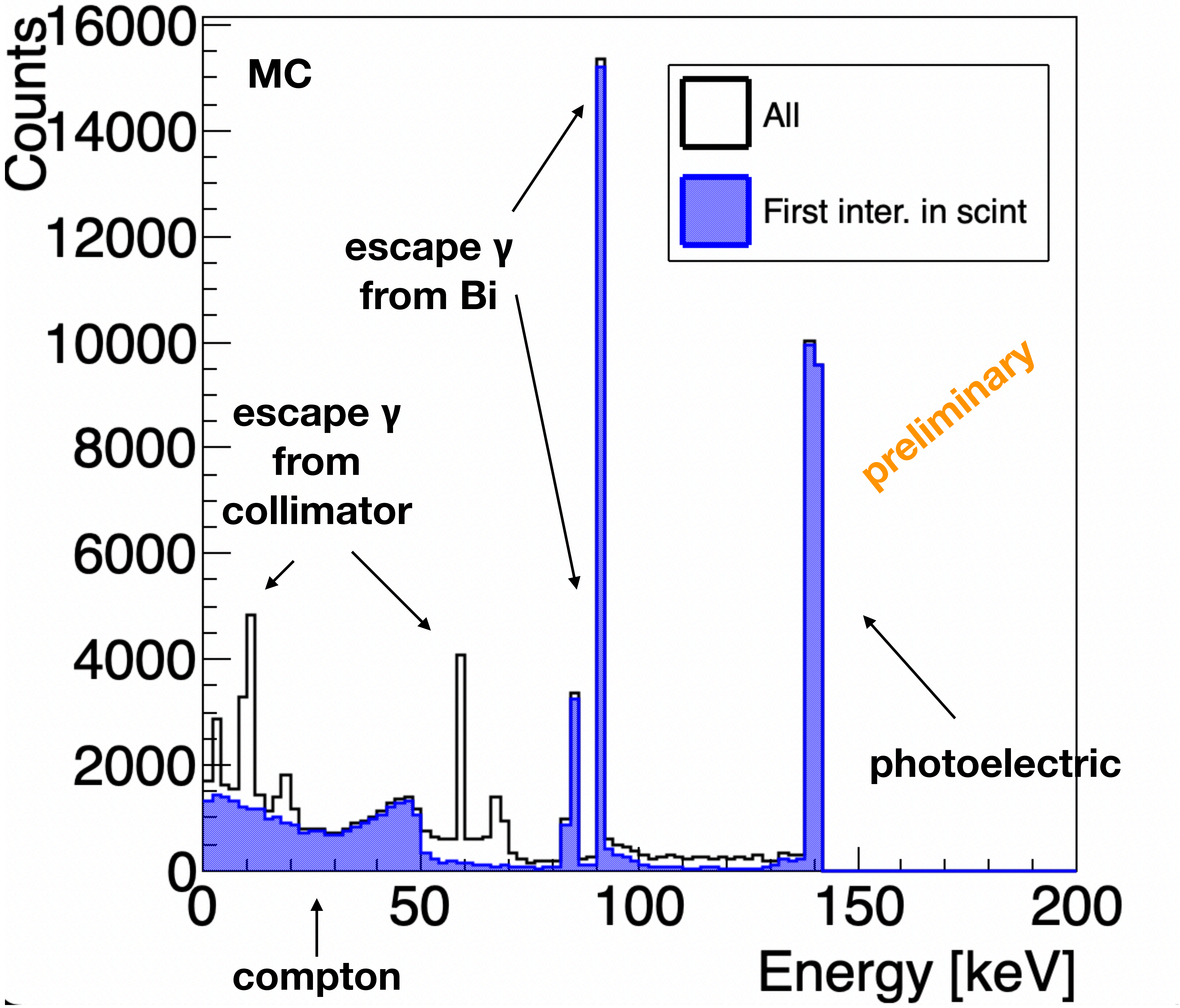


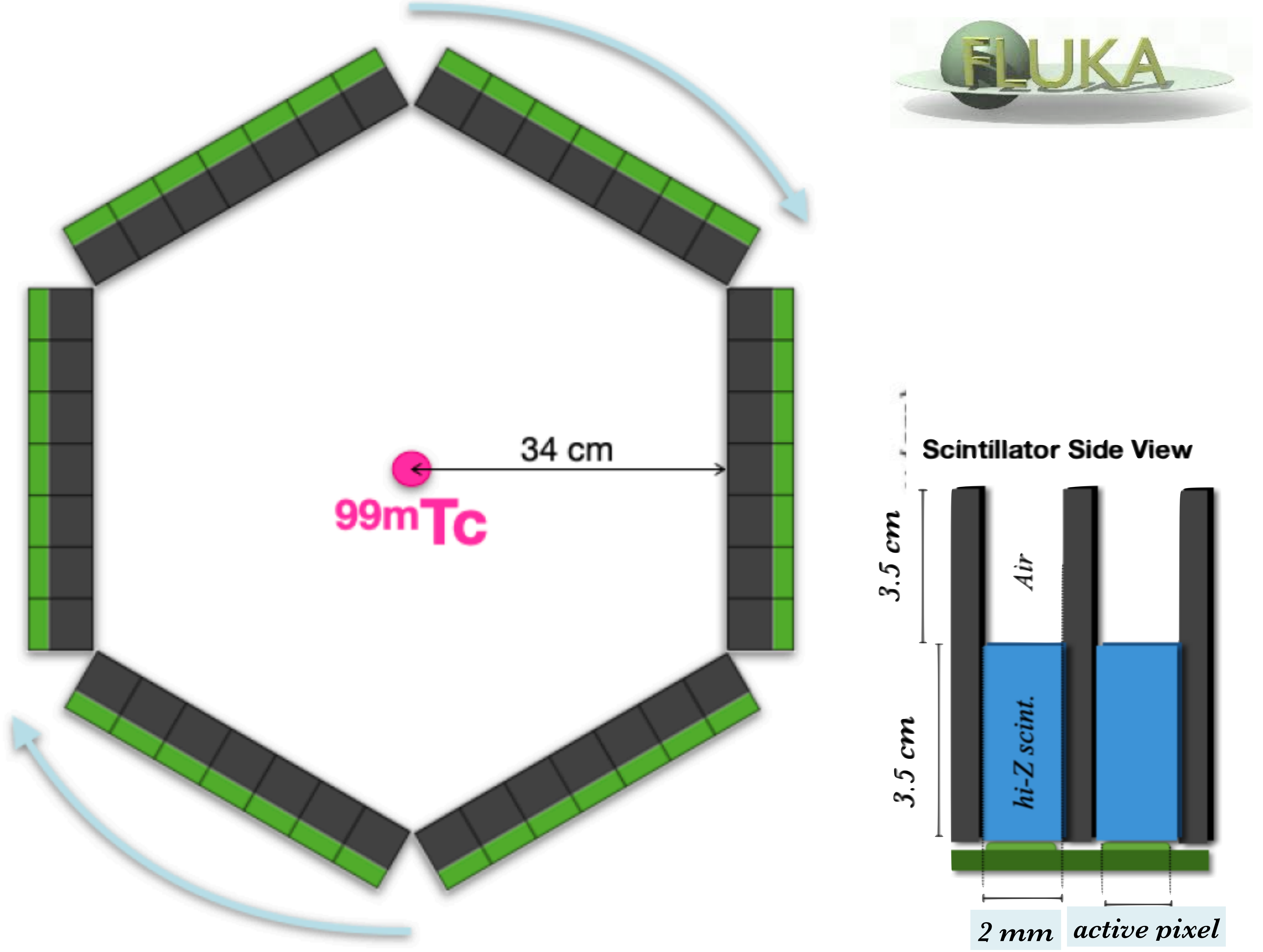
1st Condition: the event must fall within the **511-keV photopeak region seen by NaI(Tl).**

2nd Condition: the event must be seen simultaneously in the three reading channels.



Energy spectrum (Bi 5%)





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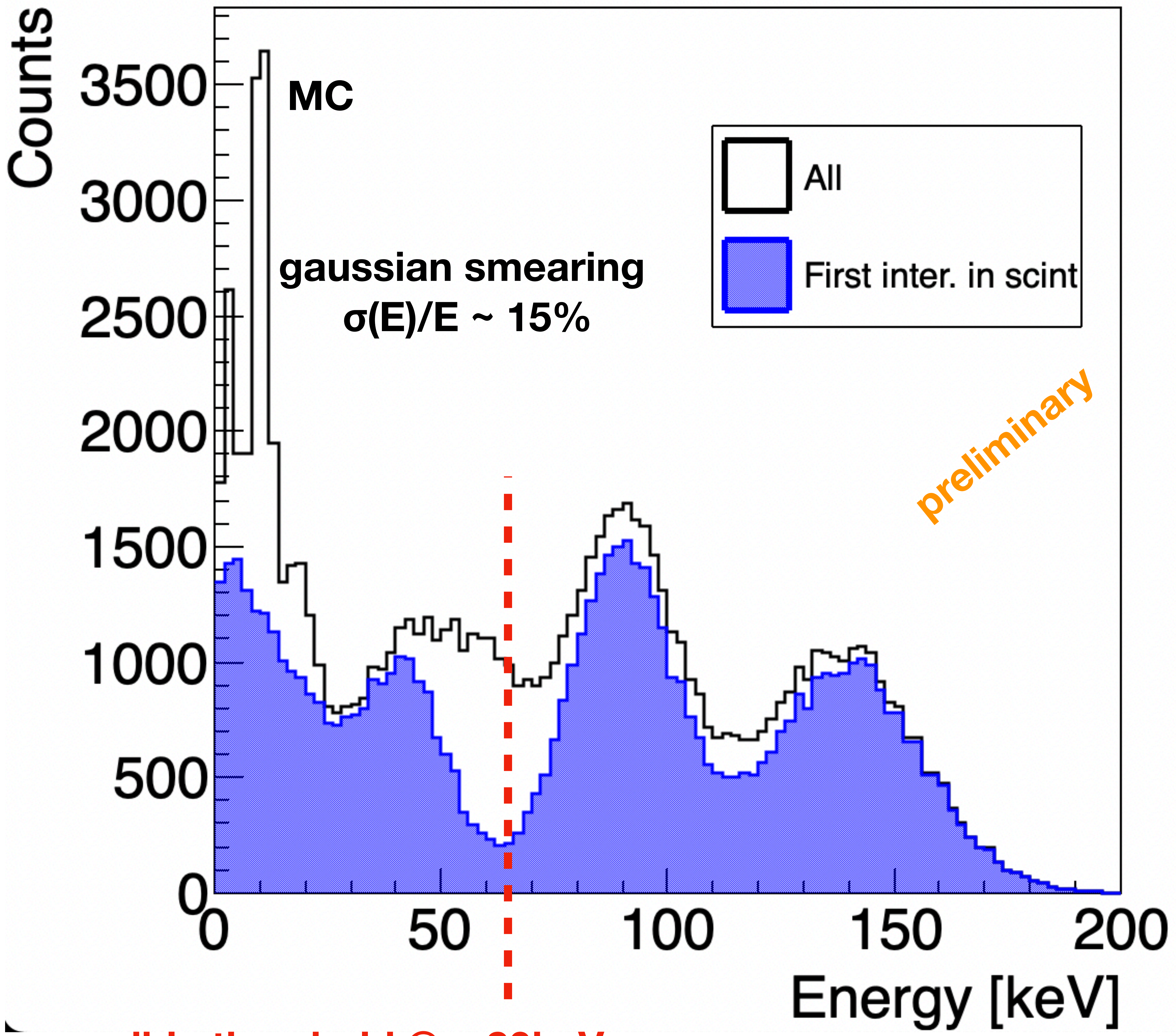
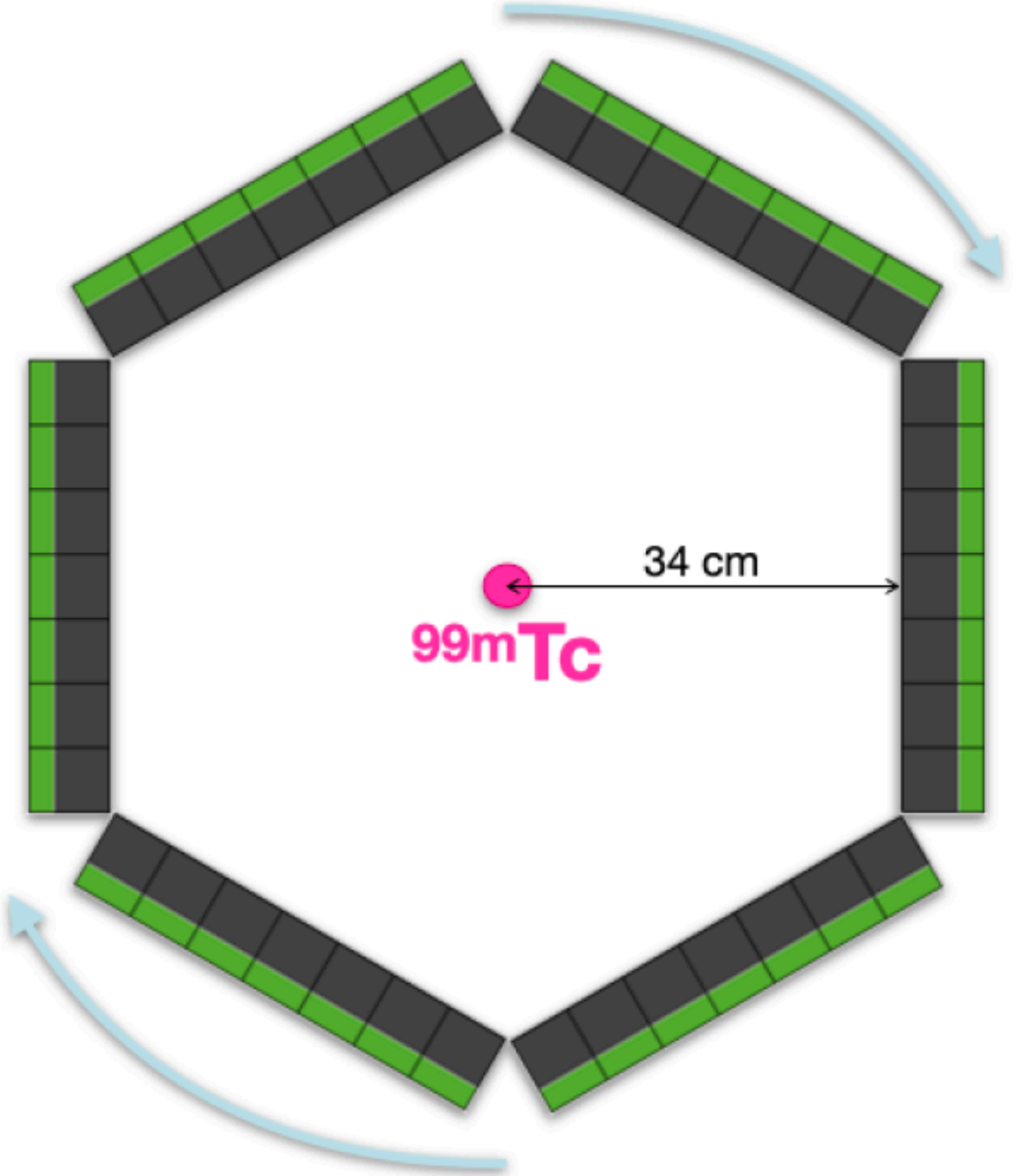
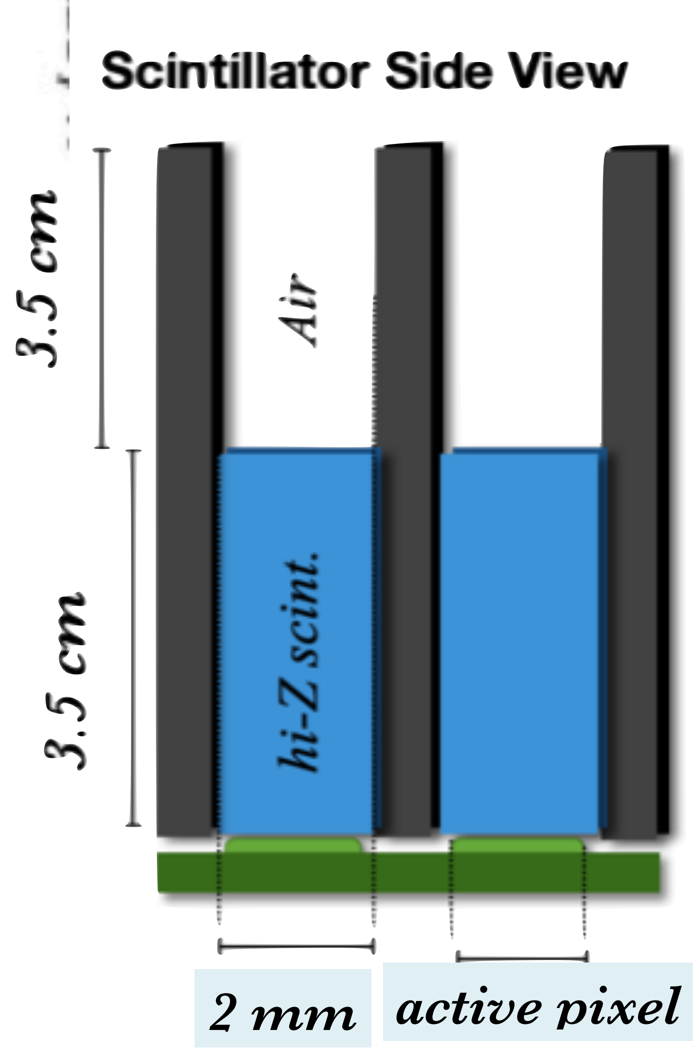
Energy spectrum (Bi 5%)



possible threshold @ ~ 80keV





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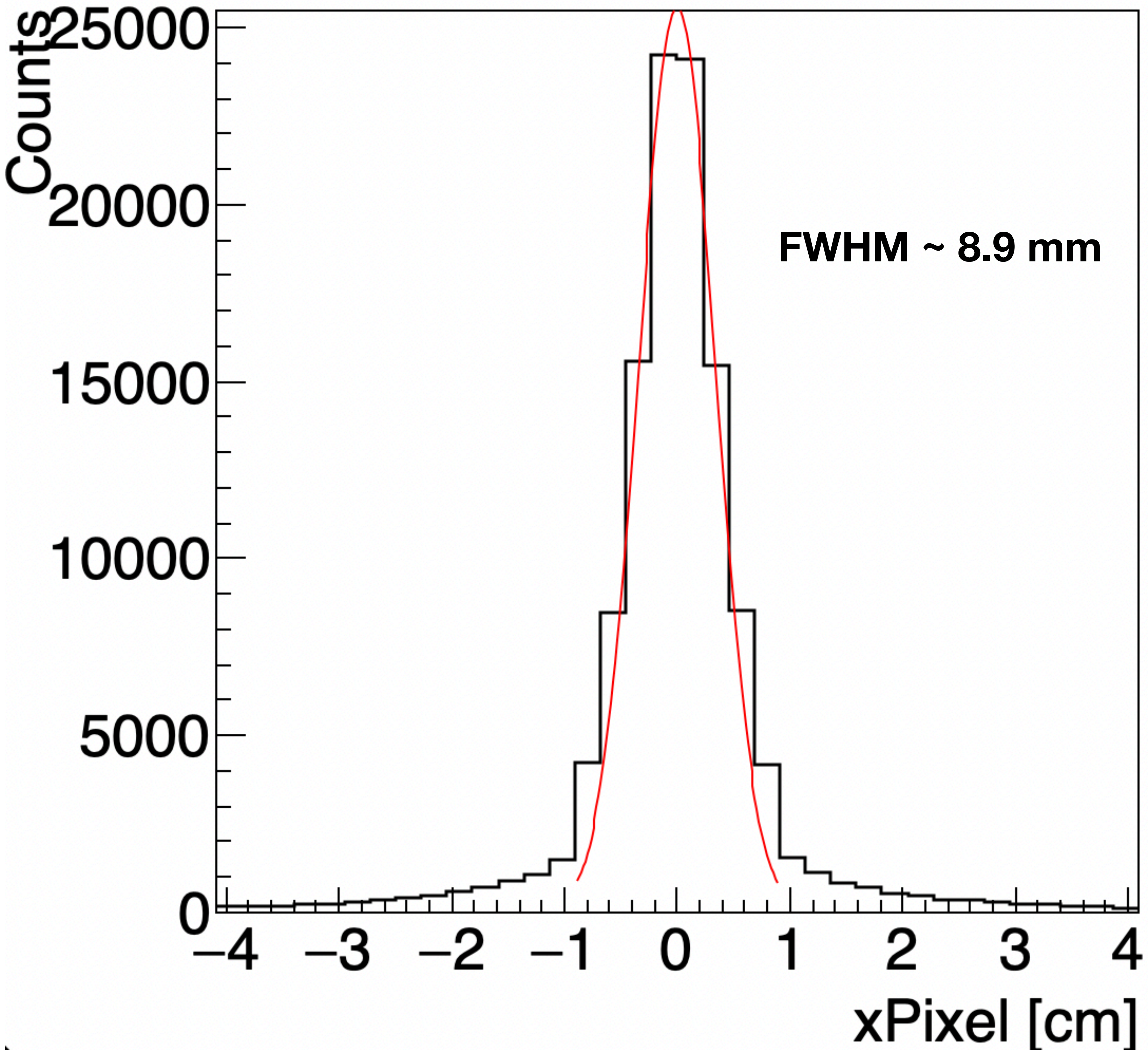
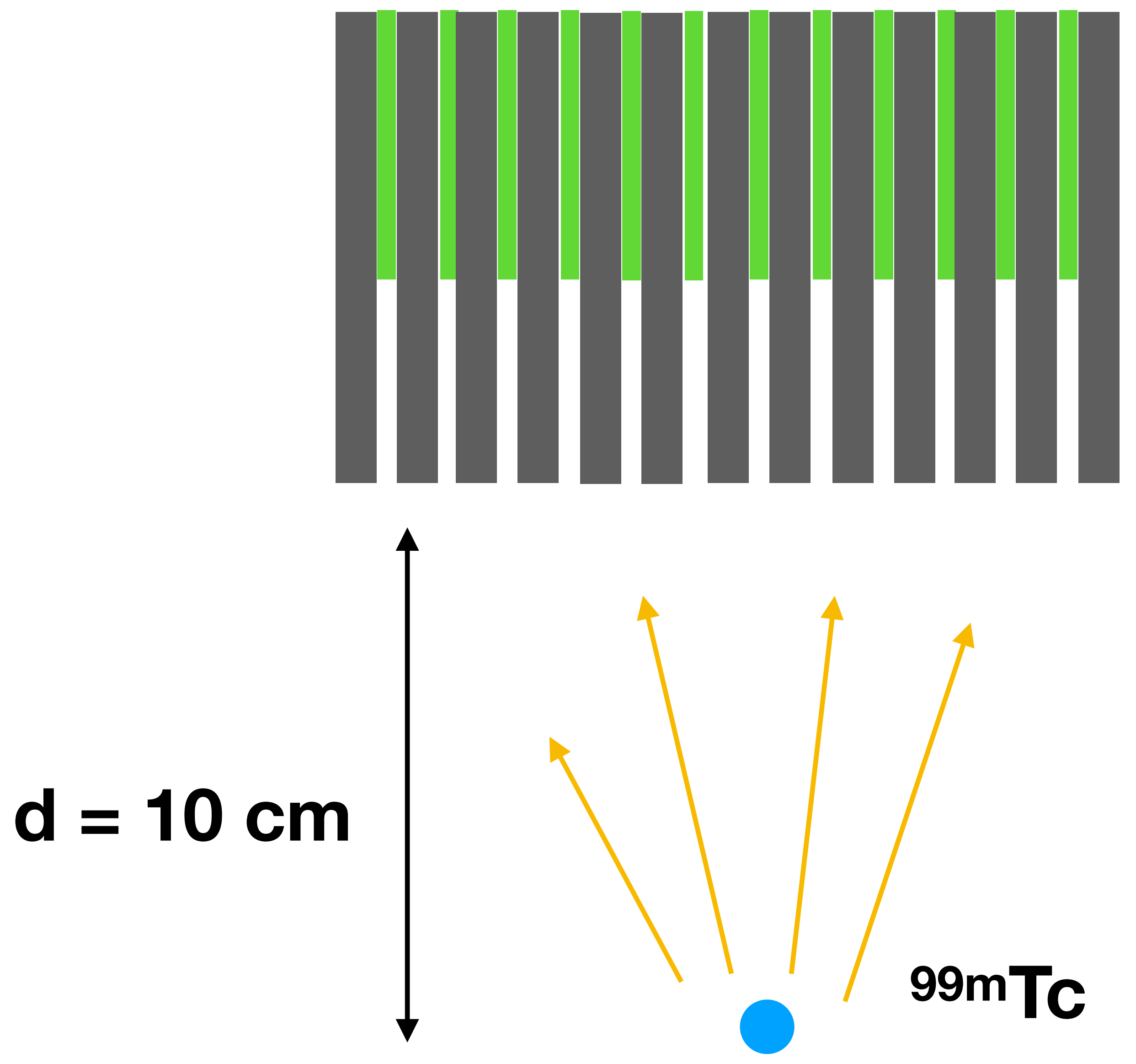
Spatial resolution @ 10 cm





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