

– flexiBlE hYbrid neutrON Detectors – INFN - Grant Giovani Ricercatori e Ricercatrici 2023-2024

Ilaria Fratelli – associata INFN Bologna, RTT UNIBO DIFA

16 Dicembre 2024

Semiconduttori Innovativi per Rivelazione di Radiazione

Low-cost large-area printing techniques











Space Missions











Semiconduttori Innovativi per Rivelazione di Radiazione



SIEMENS – X-ray detectors – PEROVSKITES



Tedde S. *et al.* High-sensitivity high-resolution X-ray imaging with soft-sintered metal halide perovskites. *Nat Electron* **4**, 681–688 (2021). https://doi.org/10.1038/s41928-021-00644-3

SAMSUNG – X-ray detectors – PEROVSKITES



Kim, Y., Kim, K., Son, DY. *et al*. Printable organometallic perovskite enables large-area, low-dose X-ray imaging. *Nature* **550**, 87–91 (2017). https://doi.org/10.1038/nature24032

Sinergie e Background











Sinergie e Background





Sinergie e Background





NEUTRONI VELOCI – perovskiti a film sottile –

2D hybrid perovskite thin films



Lédée, F., Ciavatti, A., Verdi, M., Basiricò, L., Fraboni, B., (2022). *Advanced Optical Materials, 10*(1), 2101145. Fratelli, I., Basiricò, L., Ciavatti, A., Margotti, L., Cepić, S., Chiari, M., & Fraboni, B. (2024) *Advanced Science, 2401124*. Ciavatti A., Foderà V., Armaroli G., Maserati L., Colantoni E., Fraboni B., Cavalcoli D., Adv. Funct. Mater. 2024, 34, 2405291





2D hybrid perovskite thin films





2D hybrid perovskite thin films





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4/10

Meccanismo di Interazione



10

8

Target Atomic Number

1

16

32

64

Simulazioni – Toolkit Geant4



PPEROVSKITE IBRIDA 2D

 \rightarrow ALTA DENSITÀ DI ELEMENTI A BASSO Z

 \rightarrow MAX TRASFERIMENTO DI ENERGIA A SEGUITO DELL'URTO ELASTICO

CLASS	ACTIVE MATERIAL	DENSITY	HYDROGEN DENSITY	CARBON DENSITY
		8 •	× 10 ²² cm ⁻³	× 10 ²² cm ⁻³
SOLID STATE	MHyPbCl3	3.242	3.79	
	Rubrene	1.26	3.99	
	4MHB	1.46	4.62	
LIQUID SCINTILLATOR	EJ301		4.82	3.98
	EJ309		5.43	4.35
	Stilbene	1.15	4.61	
PLASTIC	EJ276D	1.099	4.65	4.94
SCINTILLATOR	EJ200	1.023	5.17	4.69
THIS WORK	PEA ₂ PbBr ₄	2.27	4.25	2.84

Simulazioni – Toolkit Geant4



100

10

1

1

Neutron Energy = 2.5 MeV PVK thickness = $5 \mu m$

PPEROVSKITE IBRIDA 2D → ALTA DENSITÀ DI ELEMENTI A BASSO Z → MAX TRASFERIMENTO DI ENERGIA A SEGUITO DELL'URTO ELASTICO				¹ H (n,n) ¹ H ¹ H (n,n) ¹ H ¹⁰⁰ ¹⁰⁰ ¹⁰⁰	¹² C (n,n) ¹² C	
CLASS	ACTIVE MATERIAL	DENSITY g cm ⁻³	HYDROGEN DENSITY × 10 ²² cm ⁻³	CARBON DENSITY × 10 ²² cm ⁻³	$ \begin{array}{c} 0 & 0.4 \\ $	0.2 0.4 ¹² C k
	MHyPbCl3	3.242	3.79			
SOLID STATE	Rubrene	1.26	3.99			,,,,,
	4MHB	1.46	4.62		te (a	
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THIS WORK	PEA ₂ PbBr ₄	2.27	4.25	2.84	0 5 10 Berovskite Thickne	15 20





Simulazioni – Toolkit Geant4



Neutron Energy = 2.5 MeV PVK thickness = 5 μm

PPEROVSKITE IBRIDA 2D → ALTA DENSITÀ DI ELEMENTI A BASSO Z → MAX TRASFERIMENTO DI ENERGIA A SEGUITO DELL'URTO ELASTICO 0.6 **HYDROGEN CARBON** DENSITY ACTIVE DENSITY DENSITY **CLASS** MATERIAL g cm⁻³ 0.5 × 10²² cm⁻³ × 10²² cm⁻³ MHyPbCl3 **SOLID STATE** 1.26 1.46 4.62 EJ301 4.82 3.98 LIQUID 5.43 4.35 **SCINTILLATOR** 1.15 4.61 1.099 4.65 4.94 **PLASTIC SCINTILLATOR** EJ200 1.023 5.17 4.69 2.84 **THIS WORK** 4.25 PEA₂PbBr₄ 2.27





Irraggiamento @ INFN-LNL

Acceleratore Van der Graaff (INFN-LNL, CN, 0° beamline) Fascio 5 MeV di Deuterio, target ⁹Be Fast Neutrons ⁹Be(d,n)¹⁰B (Energia = [1 – 4] MeV Flusso = [0.4 - 3] · 10⁵ n s⁻¹ cm⁻²)







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SELETTIVITÀ (= trasparenza al campo gamma)





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Fratelli I., in preparation



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Tempo di risposta (τ_{rise} = 1,6 s; τ_{fall} = 4,2 s)

🗸 Ripetibilità e Stabilità

✓ **SELETTIVITÀ** (= trasparenza al campo gamma)







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Efficienza a diversi spessori di perovskite



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Fratelli I., in preparation 9/ 10

Efficienza a diversi spessori di perovskite



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E I NEUTRONI TERMICI??













${}^{10}B + n \begin{cases} \xrightarrow{94\%} \alpha (1.47MeV + {}^{7}Li(0.84MeV) + \gamma (0.48MeV) \\ \xrightarrow{6\%} \alpha (1.78MeV + {}^{7}Li(1.01MeV) \end{cases}$





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10x 20x

③ <u>DEPOSIZIONE della</u> <u>PEROVSKITE IBRIDA 2D</u>

> SPIN COATING Thickness ≈ 2 μm

> > INFN



MECCANISMO DI INTERAZIONE



Irraggiamento @ INFN-LNL (linea MUNES)



Cables > 10 m to keep the electronics far away, behind the wall **PMT+ LiBO** placed close to the sample to monitor the flux in the same conditions

Bi window to attenuate gamma , rays



Irraggiamento @ INFN-LNL (linea MUNES)



Cables > 10 m to keep the electronics far away, behind the wall **PMT+ LiBO** placed close to the sample to monitor the flux in the same conditions

Bi window to attenuate gamma



Turno 10 giorni fa...

Stay tuned...



Conclusioni





RIVELAZIONE NEUTRONI VELOCI

PEROVSKITE IBRIDA 2D

- → Alta densità intrinseca di atomi a basso Z (H e C)
- → Migliori proprietà di trasporto rispetto ai materiali organici
- → Maggior <u>stabilità</u> ambientale, soppressione della migrazione ionica, bassa dark current Configurazione a **FILM SOTTILE**
 - → Stampa da soluzione scalabile su larghe aree e substrati flessibili
 - → Trasparenza ai raggi gamma = <u>selettività</u>

RIVELAZIONE NEUTRONI TERMICI

- PEROVSKITE IBRIDA 2D
 - → Stampa da soluzione scalabile su <u>larghe aree</u> e substrati <u>flessibili</u>
 - + possibilità di avere un <u>miglior ricoprimento</u> delle
 - microstrutture 3D

MICROSTRUTTURE 3D ¹⁰B

 \rightarrow <u>Trasferimento di carica</u> al semiconduttore più efficiente





Ilaria Fratelli – 16 Dicembre 2024

INFN-Bologna and UNIBO Department of Physics and Astronomy - DIFA





Prof. B. Fraboni



INFN-Padova and LNL



Dr. I. Fratelli









G. Napolitano

C. Bordoni

PADOVA



Dr. F. Pino



Dr. G. Maggioni





Prof. S. Moretto

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GRAZIE a tutte e tutti voi per l'attenzione!



Dr. J. Delgado





Dr. M. Cinausero





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Starting Point – Neutroni Veloci





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Starting Point – Neutroni Veloci





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4000

Starting Point – Neutroni Termici





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2D layered Perovskites

$\mathbf{PEA_2PbBr_4} (\mathbf{PEA} = \mathbf{C_6H_5C_2H_4NH_3^+})$

3D

VS.

2D



- Lower lon migration
- temperature <150°C
- properties tuning by relative amounts of the components

Boron Neutron Capture Therapy e dosimetria

Incident Beam Quality

- 1) Epithermal neutron flux > 10⁹ n cm⁻² s⁻¹
- 2) Neutron energy range [0,5 eV 10 keV]
- 3) Fast neutrons and gamma rays as low as possible



 ${}^{10}B + n \begin{cases} \stackrel{94\%}{\longrightarrow} \alpha(1.47MeV + {}^{7}Li(0.84MeV) + \gamma(0.48MeV) & (Q = 2,31 \text{ MeV}) \\ \stackrel{6\%}{\longrightarrow} \alpha(1.78MeV + {}^{7}Li(1.01MeV) & (Q = 2,792 \text{ MeV}) \\ & {}^{6}Li + n {\longmapsto} {}^{3}H(2.72MeV + \alpha(2.05MeV) & (Q = 4,78 \text{ MeV}) \end{cases}$





Dymova MA, et al. Boron neutron capture therapy: Current status and future perspectives. Cancer Commun 2020;40(9):406–21.

Stato dell'Arte per la rivelazione di neutroni



Interazione neutrone termico-materia



$${}^{10}B + n \begin{cases} \stackrel{94\%}{\longrightarrow} \alpha (1.47 MeV + {}^{7}Li(0.84 MeV) + \gamma (0.48 MeV) & (Q = 2,31 MeV) \\ \stackrel{6\%}{\longmapsto} \alpha (1.78 MeV + {}^{7}Li(1.01 MeV) & (Q = 2,792 MeV) \\ & {}^{6}Li + n {\longmapsto} {}^{3}H(2.72 MeV + \alpha (2.05 MeV) & (Q = 4,78 MeV) \\ \end{cases}$$

	Cross Section (barn) @ 0.025 eV	Natural Abundance (%)
⁶ Li	942	7,6%
¹⁰ B	3842	19,9%

Spettrometria con PVK - letteratura





He, Y., Matei, L., Jung, H.J. et al. Nat Commun 9, 1609 (2018).

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Quevedo-Lopez M., et al. Adv Mater Technol. 2020;5(12):3–9.



Electron/hole pair creation mean energy

empirical model of Devanathan $W = 2E_G + 1.43 \text{ eV}$

The energy gap value of (PEA)₂PbBr₄ of $E_G = (3.00 \pm 0.03) \rightarrow W_{(PEA)_{2}PbBr_4} = (7.43 \pm 0.06) \text{ eV}.$





REQUIRED PROPERTIES

• HIGH SENSITIVITY

MATERIAL	ELECTRIC FIELD (V μm ⁻¹)	PROTON ENERGY (MeV)		SENSITIVITY	REF.
bBr ₃ + (PEA) ₂ PbBr ₄	0.2	5	(1	12 ± 0.01)·10 ⁻¹⁸ C H ^{+ -1}	[1]
PGe-pentacene	0.03	5		(6.4 ± 0.2)·10 ⁻²⁰ C H ^{+ -1}	[2]
MAPbBr ₃	0.01	3	(2	2.19 ± 0.03)·10 ⁻¹⁸ C H ^{+ -1}	[3]
CsPbCl ₃	2	100-228		4·10 ⁻²⁰ C H ^{+ −1}	[4]
(PEA) ₂ PbBr ₄	0.2	5	(2	I.25 ± 0.02)·10 ⁻¹⁸ C H ^{+ −1}	This work
	response (> 2 G Basiricò, Fraboni et al., <i>Adv. Sci.</i> 2022 , <i>2204815</i> , 1.				04815, 1.
	I. Fratelli, Fraboni et al., <i>Sci Adv</i> 2021 , 7, eabf4462. H. Huang, et al., <i>ACS Appl Electron Mater</i> 2022 , DOI 10.1021/acsaelm.2c01406.				
	M. Bruzzi, et al., Front Phys 2023 , 11, 1.				
				L	





Basiricò, Fraboni et al., *Adv. Sci.* **2022**, *2204815*, 1.

REQUIRED PROPERTIES

• HIGH SENSITIVITY

- → HIGH Signal to Noise Ratio → dark current has to be lower than 1% of the signal current
- FAST RESPONSE for Real-Time monitoring
- **TRANSPARENT** to be placed in-line avoiding perturbation of the primary beam
- <u>RADIATION TOLERANT</u> for reliable and stable response (> 2 Gy)
- <u>LARGE AREA</u> and <u>FLEXIBLE</u> (> 10 x 10 cm²) and good <u>SPATIAL RESOLUTION</u>



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Fratelli et al., Adv. Sci. 2024, 101002/advs.202401124



Each 5 MeV proton passes through the 2D perovskite layer releasing 12 keV $\mu m^{\text{-1}}$



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Flexible and large area beam monitor based on 2D perovskite thin film



Voltage (V)

12

Pixel dimension down to 0.5 mm

50 X-ray Photocurrent variation (%) 25 -Current (A) —■— device 1 œ Ω device 2 device 3 H device 5 -25 device 6 10⁻¹³ – Mevice 7 device 8 ---- device 9 -50 10 7 8 9 -10 -5 1 2 3 5 5 6

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Device Fratelli et al., Adv. Sci. 2024, 101002/advs.202401124

Flexibility









Studio di diversi spessori per aumentare l'efficienza









Fig. 4. A summary of the ⁹Be(d, n) thick-target spectra for deuteron energies of 2.6 to 7 MeV in steps of 0.4 MeV.

Meadows, J. W. (1993). The 9 Be(d, n) thick-target neutron spectra for deuteron energies between 2.6 and 7.0 MeV. In *Nuclear Instruments and Methods in Physics Research* (Vol. 324).