”P-RAD 2023” European Workshop on Perovskite Radiation Detectors

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Book of Abstracts

Contents

Session 4 (Posters) / 2

All-inorganic Cs2AgBiBr6 Double Perovskite Single Crystals for Radiation Detectors: Exploring the Effects of Temperature, Concentration, and Time

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In recent years, all-inorganic double perovskites have gained significant interest as promising candidates for a wide range of optoelectronic applications, particularly in the field of radiation detection. Cs2AgBiBr6, a double perovskite with a low toxicity profile and high chemical stability, has attracted substantial attention due to its potential for enhanced radiation detection performance compared to traditional materials. In this study, we will provide a comprehensive investigation into the effects of temperature, concentration, and time on the synthesis and properties of Cs2AgBiBr6 single crystals, with the ultimate goal of optimizing the material for efficient and reliable radiation detector applications. Through meticulous control and modulation of these parameters, we will uncover crucial insights into the crystal growth process of Cs2AgBiBr6, and elucidate how specific changes in temperature, concentration, and time affect the resulting material properties. We will examine the impacts of these factors on parameters such as defect densities, crystal morphology, and electronic properties, as well as how they influence the overall performance of Cs2AgBiBr6-based radiation detectors. In addition to the experimental results, we will provide a theoretical framework that supports our findings and helps explain the underlying mechanisms governing the crystal growth and performance of Cs2AgBiBr6. This comprehensive study will not only contribute to the understanding of Cs2AgBiBr6 synthesis and optimization, but also serve as a reference for future research on other all-inorganic double perovskite materials. The outcomes of our research pave the way for further development and refinement of Cs2AgBiBr6-based radiation detectors, unlocking the full potential of this promising materials. Ultimately, these advancements will lead to the design and fabrication of high-performance, environmentally friendly radiation detectors, based on all-inorganic double perovskite materials. These offering significant benefits in terms of both safety and efficiency for a variety of applications, including medical imaging, homeland security, and environmental monitoring.

Session 2 / 3

Recent Advancements in Lead Halide Perovskite Nanocrystals and Nanocomposites for Radiation Detection.

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The urgency for affordable and reliable detectors for ionizing radiation in medical diagnostics, nuclear control and particle physics is generating growing demand for innovative scintillator devices combining efficient scintillation, fast emission lifetime, high interaction probability with ionizing radiation, as well as mitigated reabsorption to suppress losses in large volume/high-density detectors. Prized for their solution processability, strong light-matter interaction, large electron-hole diffusion length and tunable, intense luminescence at visible wavelengths, lead halide perovskite nanocrystals (LHP-NCs) are attracting growing attention as highly efficient emitters in artificial light sources and as high-Z materials for next generation scintillators and photoconductors for ionizing radiation detection. Nonetheless, several key aspects, such as the trapping and detrapping mechanisms

to/from shallow and deep trap states involved in the scintillation process and the radiation hardness of LHP NCs and LHP NC-based plastic nanocomposites under high doses of ionizing radiation are still not fully understood, leaving scientists without clear indications of the suitability of LHP-NCs in real world radiation detectors or design strategies for materials optimization. In this talk, I will present on our recent strategies for high performance radiation detection schemes and will report recent spectroscopic results of the scintillation process and its competitive phenomena, ultimately offering a possible path to the realization of highly efficient and extremely radiation hard LHP-NCs. This work is the joint result of numerous scientific teams worldwide and part of the AIDAInnova project.

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Session 2 / **4**

Understanding the role of intra-gap electronic levels in scintillating lead halide perovskite nanocrystals towards effective radiation detection schemes

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Lead halide perovskites (LHP) are rapidly emerging as efficient, low-cost, solution-processable scintillators for radiation detection. Most importantly, LHP can be embedded in suitable polymeric hosts to create composite materials and produce fast, efficient, and more sensitive detectors in a cost-effective way, to meet the specific demands of all up-to-date technological and medical applications. Indeed, when embedded in polymeric matrices, LHP nanocrystals favor the enhancement of the interaction cross-section with ionizing radiation, thanks to their high atomic number, and retain exceptional levels of radiation hardness. The implementation of this novel class of scintillating composites is imperative and aims at the achievements of scintillators with improved performances. This goal is essentially linked to the fundamental understanding of the correlation between the physical-chemical properties and the luminescence features and to the comprehension of the scintillation mechanism in nanosystems, from the primary interaction with the ionizing radiation, through energy transfer and trapping processes, to the emission of light.
An essential stage in the scintillation process is the transport of free carriers generated upon the interaction between ionizing radiation and the scintillating material: it is often largely affected by the presence of trapping sites, which can capture migrating charge carriers and either delay their radiative recombination or decrease the overall scintillation efficiency, according to the characteristics of the traps involved. Therefore, carrier trapping is arguably the most critical limitation to the

scintillation performance; nonetheless, no clear picture of the trapping and detrapping mechanisms to/from shallow and deep trap states involved in the scintillation process has been reported to date, as well as on the role of the material dimensionality.
We addressed this issue by performing a comprehensive study using temperature-dependent radioluminescence and photoluminescence measurements side-by-side to wavelength-resolved thermally-stimulated luminescence (TSL) and afterglow experiments on $CsPbBr_3$ with increasing dimensionality, namely nanocubes, nanowires, nanosheets, and bulk crystals. All systems are found to be affected by shallow defects resulting in delayed intragap emission following detrapping via a-thermal tunneling. TSL further reveals the existence of additional temperature-activated detrapping pathways from deeper trap states, whose effect grows with the material dimensionality, becoming the dominant process in bulk crystals.
In addition, we prove that $CsPbBr_3$ nanocrystals can be effectively embedded into polymethylmethacrylate matrix to obtain high optical quality flexible and smooth scintillating nanocomposites, whose defectiveness resembles that of LHP bulk crystals, featuring isolated energetically deep defect states that trap carriers which, upon heating, recombine in a specific intragap emission center, as revealed by TSL measurements.
These results highlight that, compared to massive solids where the suppression of both deep and shallow defects is critical, low dimensional nanostructures are more promising active materials for LHP scintillators, provided that their integration in functional devices meets efficient surface engineering.
The effectiveness of this investigation approach coupling scintillation and TSL measurements, traditionally exploited only for classical single component bulk scintillators, is therefore demonstrated also for nanomaterials.

Session 2 / 5

Lead Halide Perovskite Nanocomposite Scintillators

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Lead halide perovskite (LHP) nanocrystals show promise as scintillators due to their high atomic number, bright luminescence and tunable emission wavelength. Here, we present nanocomposite scintillators which consist of formamidinium lead halide perovskite nanocrystals in a polymer matrix. Mixed-halide $FAPbBr_{3x}Cl_{3(1-x)}$ nanocrystals were synthesised by a room-temperature solution-growth method which allowed for rapid, low-cost production. Luminescence spectra, time response and Stokes shifts were measured for each different nanocrystal composition so as to select the most appropriate LHP nanocrystal for inclusion in the nanocomposite scintillator: the ideal nanocrystal would have bright, fast emission at a wavelength close to the to the maximum efficiency of a photomultiplier tube and would also have a large Stokes shift so as to minimise reabsorption of the scintillation light.
Nanocomposite scintillators were then produced which contained LHP nanocrystals in a matrix of either PMMA or the plastic scintillator EJ-290. A significant challenge in producing uniform, transparent nanocomposite materials is the effect of optical scattering, which leads to an increase in the opacity of the scintillator with increased effect when the scintillator is thicker, contains a higher loading of nanocrystals, or contains larger particles. Here, the size of the LHP nanocrystals was minimised and a partial ligand exchange procedure was investigated as a strategy to prevent nanocrystal aggregation. The thickness and nanocrystal loading of the composites were then investigated so as to balance the transparency of the scintillator to visible light with the attenuation of high-energy photons.

Session 4 (Posters) / **11**

Metal Halide Perovskite X-rays Detectors Based on Pressed Polycrystalline Wafers

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Metal halide perovskite is a new semiconductor family with great potential in various applications such as X/γ-ray detectors, solar cells, light emission diodes, and lasers. Especially inorganic metal halide perovskites, such as e.g. CsPbI₃ or CsPbBr₃ feature high atomic numbers (Z_{Cs} = 55, Z_{Pb} = 82, Z_{Br} = 35, and Z_I = 53), an adjustable energy band gap (1.5–3.5 eV), high resistivity (10⁷–10¹² Ωcm), and a large carrier mobility-lifetime product (10^{−5}–10^{−2} cm²/V). These superior characteristics promise perovskite crystals with large radiation attenuation and improved charge collection efficiency, combined with a facile preparation and lower costs. However, the research progress of metal halide perovskite-based X/γ-ray detectors in terms of photoelectric performance and device application is still far behind those of PSCs and LEDs. The development of tailored growth and fabrication methods for radiation detectors and the evaluation of their performance is an important scientific and engineering objective. Among other methods, dry mechanochemical ball-milling synthesis has recently emerged as a very convenient and reliable method to obtain high quality perovskites.
In this work, we study structural, electrical, and spectrometric properties of X/γ-ray detectors based on CsPbI₃, CsPbBr₃, and Cs₂AgBiBr₆ pressed polycrystalline metal halide perovskite wafers produced from dry mechanochemical ball-milling synthesized powder. Ohmic devices were fabricated to correlate grain size with wafer resistivity, and Schottky diodes were fabricated to reduce dark current. Finally, their X-ray detection performance is studied and correlated to the observed microstructure, resistivity, and device architecture. These results provide guidance for further investigations on pressed mechano-synthesized inorganic perovskite crystal as very promising X/γ-rays radiation detection materials.

Session 5 / **12**

Understanding and developing low-dose halide perovskite X-ray detectors

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Halide perovskites are generating excitement for low-cost yet high-performance optoelectronic devices. Recently they have demonstrated enormous potential for radiation detection such as X-rays as scintillator or direct detector device platforms.

Here, I will give a summary of our group's efforts towards understanding and developing halide perovskites for X-ray detector applications. Topics will include studies investigating the fundamental radioluminescence mechanism in perovskite scintillator materials and developing sensitive photon counting perovskite detectors. I will also summarise overall capabilities and directions in the group in this area.

Session 4 (Posters) / **13**

Investigation of photocurrent relaxation in MAPbBr3 perovskites with Cr/SnO2/, Cr/ contacts

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During the measurement of the hole photocurrent relaxations (illuminated anode), we noticed an unusually high current after the bias polarity was switched. Our setup was designed to suppress the majority of effects tied to parasitic capacitances and is expected to have RC=100ns-1μs depending on the capacitance of the sample. The majority of the observed effects on a scale of 10s of μs, therefore, have to be the relaxation of the sample itself. The most surprising fact is that the photocurrent right before the polarity switch is close to zero once the illumination is switched off. A thorough investigation of this transient event revealed that its width and amplitude are bias dependent. During the measurement, the bias before the polarity switch is always +150V. The amplitude changes only after the polarity switch, which ensures that the illumination always generates the same number of free carriers. The general shape of the transient events looks very similar to hole current waveforms measured by the Laser-induced transient currents technique (L-TCT). Their respective transit times (positions of the falling edges) are a great match too.
We believe that this effect can be explained by the barrier-induced accumulation of the photogenerated holes beneath the cathode. After the bias switch, accumulated holes drift towards the new cathode, which results in the observed transient event. The accumulated electrons can also cause this effect. However, we did not observe any electron signal –L-TCT or photocurrents.
Finally, with this model in mind, we endeavoured to find out the dissipation rate of the accumulated mobile holes and its dependence on contact material. In this experiment, both Cr and Cr/SnO2 electrodes were illuminated and several different switch delays (relative to the end of the optical pulse) were used. Obtained results show that the accumulated mobile holes dissipate with a characteristic time of 600 μs-1.5 ms regardless of the cathode material. For comparison the hole lifetime in the bulk determined by L-TCT is 50μs. There are several mechanisms that can be responsible for the decaying of the transient signal –tunnelling through the barrier, gradual trapping of holes in the surface defects or recombination with injected electrons. Unfortunately, we are not able to distinguish these models.

Session 4 (Posters) / **14**

The development and performance of 1D lead-free Rb-based metal halide scintillators.

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The ability to detect ionising radiation is crucial to many areas of modern-day life spanning use in medical imaging, cancer therapies, security, and environmental monitoring as well as potential in military field applications. Indirect detectors, also known as scintillators, operate by absorbing high energy X-rays then converting into lower energy photons that emit in the UV/Visible region. The development of ideal photoactive materials for low-cost scintillator fabrication has recently seen

a focus towards lead-free metal halides, known for their characteristic broadband emission, large Stoke shifts, nanosecond decay lifetimes and ease of processability. Here we present our latest results on Rb2AgX3, X = Cl, Br, which show broadband emission centred at 515 nm and 580 nm, respectively. These Rb-based materials also possess fast radiative recombination from self-trapped excitons with typical decay times of 10 ns. The performance of the Br and Cl scintillator materials and their suitability for X-ray imaging applications will be compared.

Session 3 / **15**

High Sensitivity Polycrystalline CsPbBr3 Wafer X-ray Detectors

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Here we have fabricated and characterised CsPbBr3 wafers for direct X-ray detection. Gram-scale CsPbBr3 powder was synthesized by milling of CsBr and PbBr2 precursors in a planetary ball mill. 0.4 g of CsPbBr3 power was pressed into a pellet of 10 mm diameter by a hot-pressing method at 70° C. We have characterized the morphology and structural properties of the polycrystalline CsPbBr3 wafer by scanning electron microscopy (SEM) and X-ray diffraction (XRD), respectively. The optimised pellets show highly compact and uniform morphology, which results in superior carrier transport and higher X-ray sensitivity. 80 nm of Au electrodes were deposited on both sides of the pellet by vacuum thermal evaporation. We have studied the X-ray response of the Au/ CsPbBr3 pellet/Au X-ray detector under different X-ray irradiation dose rates and applied bias voltages. The device exhibits an X-ray sensitivity of 151 μC Gyair-1cm-2 under an applied bias of 20 V.

Session 3 / **16**

Growth and characterization of CsPbBr3 single crystals

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Metal halide perovskites with APbX3 crystal structure show rapidly increasing global interest as a new generation of radiation detection materials alternative to CdZnTe. In this work, we present our results of the multiple single crystals growth of all-inorganic perovskite CsPbBr3 using Bridgman method. Crystals were prepared directly from CsBr and PbBr2 starting materials mixed with a various mole ratio. Our optimized growth process produces crystals with diameter 15 mm and length over 50 mm. We found that the quality of grown single crystals strongly depends on the purity of the starting materials, dropping speed of the growth ampule and cooling rate after growth. For characterization of transport properties of as-grown samples, we used pulsed I-V characteristics, Laser-induced transient current technique (L-TCT) at pulsed DC bias or Time-correlated single photon counting (TCSPC). We compared the crystallographic and transport properties of grown crystals depending on the change in the mole ratio of the starting materials, the change in the purity of the starting materials, different degrees of overheating, and different dropping speed of growth ampoule. In case of 1:1 stoichiometric ratio, 40 K overheating and dropping speed of 0.5 mm h-1, we have grown CsPbBr3 crystal with resistivity ρ=2.7·109 Ωcm evaluated from pulsed I-V characteristics. Pulse-biased L-TCT current waveforms show shorter hole lifetime at the center of the wafer than close in its border. The TCSPC signal indicates non single exponential decay with average rapid component of 0.5 ns.

Session 4 (Posters) / 17

Lead Acetate Impact on Lead Halide Perovskite (FAPbBr3) Polycrystalline Radiation Detector Performance

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In this study, we analyse the performance of polycrystalline pellets of FAPbBr3 as a semiconductor-based radiation detector, and study the impact of lead acetate (LA) on the FAPbBr3 X-ray detector performance. Using a hot-pressing method various FAPbBr3 pellets were fabricated with different concentrations of lead acetate to investigate the improvement in the pellet microstructure and X-ray sensitivity.
The bulk resistivity and of the pellets was studied by I-V measurements, with the addition of LA showing a significant reduction in dark current and increase in resistivity. The X-ray sensitivity measurements of the device were carried out using X-rays from an Amptek Mini X-ray source with an Au anode and an acceleration voltage of 40 kVp. The X-ray sensitivity of the FAPbBr3 pellet devices with and without LA were studied, and the devices containing LA showed a sensitivity of 27.5 ±0.9 μC/Gy cm2. We will discuss the role of the LA in improving the density and microstructure of the material, and its effect on charge transport and X-ray sensitivity.

Session 4 (Posters) / 18

A perovskite-thermoplastic composite for 3D printing novel radiation detectors.

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The fabrication of perovskite-polymer composites offers improved stability in perovskite radiation detectors alongside properties such as flexibility or improved strength. The facile production of all-inorganic perovskite nanocrystals is a convenient and cost-effective source of radiation detecting material. Incorporating these nanocrystals into the thermoplastic filaments used for fused filament fabrication (FFF) would enable users to design and manufacture custom low-cost 3D printed radiation detectors.

In this work, the characterization of early perovskite-polymer objects made from CsPbBr3 and the thermoplastic polycaprolactone (PCL) are presented. Following the creation of the solid plastic composite material, custom disc-shaped pellets have been formed into test devices with metal contacts. Electrical measurements have been recorded alongside the direct radiation response when exposed to a tungsten target X-ray source. Compositional analysis of early samples has been carried out using microCT to examine the internal structure of samples to reveal defects such as air pockets or cracks. X-ray fluorescence imaging (XRF) and scanning electron microscopy (SEM) has been used to study the uniformity of nanocrystal dispersion throughout the composite and the relation to charge collection. The CsPbBr3/PCL devices presented are composed of ˜7% CsPbBr3 by weight. IV-characterization has been performed with bias sweeps and dark current measurements. Exposure to

the X-ray source at 70 kV across a series of mA values demonstrates a clear photocurrent response of the device under bias. From these measurements, the photocurrent response as a function of dose-rate has been found, demonstrating a linear response at greater dose-rates. Sensitivity and conductivity measurements have been calculated using the known detector area of the metal contacts for these devices. MicroCT and SEM images have been used to demonstrate the improvements made between early and recent generations of samples.

This work lays the foundation for novel 3D printable perovskite-polymer composites for use in direct radiation detection by exploring the compositional makeup of these materials and characterizing their detective properties. Recent device sensitivity to radiation will be presented along with progress that has been made in production of high-quality composite devices for characterization.

Session 4 (Posters) / **19**

Halide Perovskite Composites for High Stability X-ray Detection

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Halide perovskites one of the most promising materials for next-generation, highly sensitive, low-noise, direct high energy X-ray detectors due to their excellent optoelectronic properties including bandgap tunability, high photoluminescence quantum yields (PLQY), defect tolerance and chemical versatility. However, challenges for the successful commercialization of halide perovskites X-ray detectors remain. For example, processing halide perovskites into thick devices for optimal X-ray attenuation, without introducing defects via pressing, or mixing with polymer membranes represents one set of challenges. Further, to realize highly sensitive, low-noise detectors requires novel material choices to reduce dark currents, minimize current drift and enhance stability.

In this work, we present halide perovskite composites with functionality as direct X-ray detector and scintillators. Our scalable processing techniques can be tailored to the required X-ray energies for applications in radiography, CT, and PET scanning. By mixing halide perovskite with intrinsically more resistive scaffolds, we developed composites with high resistivity and significant reduction in dark current. In addition, our composite maintained high current densities and sensitivity resulting from charge carriers generated upon X-ray excitation of perovskite nanocrystals within the composite. Moreover, reproducible medical imaging requires device resistance to current drift. In this regard, halide perovskites often see large dark current drifts due to strong ion migration effects, thus requiring long pre-biasing before optimally functioning. However, our halide perovskite composite shows a significant reduction in dark current drift compared to the equivalent standalone perovskite detector, which we attribute to the rigid scaffold structure limiting ion migration effects. We further present a full direct X-ray detector material characterisation framework using our perovskite composite, demonstrating the full range of device performances which can be achieved by varying incident properties. The methodology of this work will lay the foundations for further device performance characterisation, removing ambiguity from current performance metrics and significantly advancing the application and commercialisation of halide perovskite X-ray detectors.

Session 4 (Posters) / **20**

High quality Perovskite Single Crystal X-ray Detectors

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In this study, we have investigated the impact of ligand-assisted Inverse Temperature Crystallisation (ITC) growth of FAPbBr3 perovskite single crystals on X-ray sensitivity and the charge transport properties. The ligand 3-(Declydimethylammonio)-propane-sulfonate inner salt (DPSI) was used to synthesise these perovskite single crystals from solution and the initial impact of the ligand on the nucleation rate and the growth speed of the single crystals were investigated. The presence of strains in the perovskite single crystals was studied using cross-polar microscopy and the emission properties were investigated using photoluminescence (PL) measurements.

The bulk resistivities of these devices were obtained using IV measurements and the resistivities reached 0.5 GΩcm. The Amptek Mini-X2 X-ray tube with an Au anode and an acceleration voltage of 40kVp was used for X-ray sensitivity measurements of these devices. These devices showed X-ray sensitivities reaching (54.94 ± 0.7) μCGyair-1cm-2. We will further discuss the impact of the DPSI ligand on crystal growth, charge transport and X-ray sensitivity of perovskite single-crystal detectors.

Session 4 (Posters) / 21

2D metal-halide perovskites for application as indirect radiation detectors: study of PEA2PbI4 and 4F-PEA2PbI4 polycrystalline films

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The development of high-performance, low-cost scintillators is an important area of research due to their wide application in medical imaging, homeland security, high-energy physics, industrial control, oil drilling explorations, and energy management.[1] The cost and inherent properties of traditional scintillators based on inorganic semiconductors, are their limitations. Low-temperature solution-based processes and high stopping power have shifted the attention on metal halide perovskite (HP). The superior photophysical and electronic properties of 2D-HPs, including large absorption coefficients, high photoluminescence yields, fast decay time and structural stability make them promising candidates for next-generation scintillator. This work aims to develop polycrystalline 2D-HPs for scintillators application with high detection power and fast scintillation response.[2]
Two different 2D-HPs, phenethylammonium lead iodide (PEA2PbI4) and 4Fluoro-phenethylammonium lead iodide (4F-PEA2PbI4,) have been investigated. In particular, different solvent mixtures were studied, and their effect was observed characterizing the different samples by studying their optical, crystalline and morphological properties as a function of the solvents used and the annealing time at fixed temperature (100 ℃).
Despite the irregular growth of 4F-PEA2PbI4, which resulted in a rough and inhomogeneous surface, if compared to PEA2PbI4, both 2D-HPs showed a strong and narrow photoluminescence (FWHM<25 nm) peaked at ˜524 nm with low Stokes-shift. Through XRD characterization it was noted the perfect formation of the polycrystalline film of HPs, the superior crystallinity and order of this compound and the effect of different organic cations.
Fluorescence lifetime measurements were performed on the selected sample using an EPL-405 picosecond pulsed laser operating at 405 nm. It has been shown that these two classes of materials had a fast luminescence decay (˜1ns), making them suitable for potential applications as fast scintillators. The two 2D-HPs in the formulation with the best performance were subjected to Ion Beam-Induced Luminescence (IBIL) experiments using protons (H+) and, in particular, the scintillation performance of the PEA2PbI4 family under alpha particle excitation by an 241Am source was reported in comparison with a standard commercial scintillator.

Session 1 / **22**

Direct detection of 5 MeV protons by mixed 3D-2D Perovskite Flexible Films

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The development of detectors for protons and heavy particles is a long-lasting research topic not only for fundamental applications, but more recently, in the medical field for hadron therapy of cancer. In this application, ion beams are used for the controlled treatment of cancer by focusing them onto small volumes, to avoid the spreading of the radiation to healthy tissues. For this reason, there is an increasing demand of systems optimized for the accurate in-situ, real-time recording and mapping of the dose delivered during a treatment plan.
Metal halide perovskites are rapidly emerging as active materials in lowcost high performing ionizing radiation detectors thanks to strong absorption of ionizing radiation, high charge carrier mobilities, long exciton diffusion, long charge carrier lifetime, and excellent optical properties. [1] Further, they combine the high performance of traditional inorganic semiconductors with the lowcost, large area scalable, deposition methods (i.e., printing technologies) typical of organic semiconductors. However, the direct proton beam detection or dose-monitoring by perovskite based devices has not been explored yet and only one paper has been published so far on direct proton detection, implemented by fully organic flexible devices.[2]
In this work, we propose a novel flexible proton detector based on mixed 3D and 2D perovskites films deposited from solution. Mixed 3D-2D perovskites are formed by mixing 3D (based on methylammonium (MA) cations) and 2D (based on larger organic ammonium (OA) cations) structure perovskites. Their employment has been reported as an effective strategy to retain the exceptional transport properties of 3D perovskites and the high stability induced by the layered structure of 2D perovskites. By adding the 3D phase (MAPbBr3) we here aim to enhance the radiation absorption of protons by the perovskite film, thanks to the higher density of MAPbBr3 and to the higher thickness of the active layer for mixed compounds.
The here proposed devices demonstrate an accurate monitoring of proton dose with instant feedback and low limit of detection, and provide a stable response even after hard and long-lasting proton irradiation.
In fact, the detector exhibited a stable response to repetitive irradiation cycles with sensitivity up to (290 ± 40) nC Gy-1mm-3 and a radiation tolerance is also assessed up to a total of 1.7 × 1012 protons impinging on the beam spot area, with a maximum variation of the detector's response of 14%.
The presented results provide an effective solution to the challenge of identifying novel functional materials and portable devices for real-time accurate monitoring of proton dose, addressing the quest for radiation hardness, low-cost scalability over large areas and mechanical flexibility, still unsolved for a range of application which span from personal dosimetry to large area and lightweight detectors for large accelerators facilities and space missions.
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[2] Fratelli et al., Sci. Adv. 2021; 7 : eabf4462

Session 4 (Posters) / **23**

Multi-layer structure for thermal neutron detection: CsPbBr3 thin films coupled with 10B conversion layers

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Scintillators are versatile indirect-type detectors finding applications in many strategic fields such as nuclear medicine, imaging, high energy physics, and homeland security. Given the outstanding photophysical properties of Metal Halide Perovskite (MHPs) along with the successful application in disparate optoelectronic devices, their use as scintillator detectors is emerging with very promising advantages: low costs, fast response high quantum yield, strong absorption, scalability, flexibility and tunability of the emission wavelength. Given the effectiveness of perovskites as α particle detectors, and the potential of 10B as a neutron converter, in this paper a 10B converting layer was coupled with an all-inorganic lead halide perovskite (CsPbBr3) layer aiming to create an high-performance thermal neutron detector for medium-high fluxes. The response of a bi-layer structure, CsPbBr3 (1 μm thick)/10B (1 μm thick), deposited by laser ablation (LA) on carbon fiber, has been investigated when exposed to a 252Cf neutron source. Monte Carlo simulations have been carried out to support the experimental evidence.

Session 4 (Posters) / 24

2D-perovskite thin films as gamma-ray detectors for medical applications

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Latest advancements in the field of ionizing radiation detection have been achieved through the application of lead halide perovskites. These organic-inorganic hybrid structures combine low-cost fabrication processes, the scalability and flexibility with desirable intrinsic properties: high stopping power, defect-tolerance, large mobility-lifetime (μτ) product, tunable bandgap and simple solution-based growth. Although promising results have been obtained through single-crystal structures, further efforts are required towards more flexible and stable devices for medical and space-related applications.
Therein we present PEA2PbBr4 thin films as active layers of high-energy, low-flux radiation detectors. Provided the fabrication procedure and techniques for interdigitated single pixels, we emphasize the main physical features to be required for dosimetry: extremely low dark current, fast response, stability over time.
Finally, specific sets of measurements are proposed that include electrical response under X- and Y-rays performed at medical facility centres with radiation sources -emitted photons from radioisotopes-employed for radiopharmaceutical therapy/nuclear medicine ((18F, 99mTc and 177Lu). Specifically, we also show its capability to follow the radioactivity path scheme at the patient skin during the intravenous drugs injection in a dummy phantom.
On the basis of these analyses, we promote PEA2PbBr4 films as the core elements for further insights, outlooks and eventual achievements towards large-area solid-state detectors for low-flux, ultra-fast x- and γ-Ray imaging and dosimetry.

Session 1 / **25**

Enhancing functionalities of blended 3D hybrid perovskite films detectors

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Hybrid organic/inorganic lead halide perovskites represent a breakthrough in the direct detection of ionizing radiation thanks to their solution processability, and their scalability over large areas on flexible plastic substrates. Flexible perovskite X-ray detectors are lightweight devices that can be operated at low-voltages and strongly limit the use of toxic materials and precursors. Polycrystalline films are thus preferred to foresee the implementation of the technology. In this work we present two approaches based on perovskite/polymer mixture, in order to enhance the detection performance of 3D hybrid halide perovskites, targeting the development of flexible and printed detectors employing micrometer thick perovskite active layer. First, we employed methylammonium lead triiodide (MAPbI3) nanocrystals blended with phenyl-C61-butyric acid methyl ester (PCBM) semiconducting polymer, leading to the realization of thick bendable detectors with good ionizing radiation absorption and high quantum efficiency. We performed X-ray nanoanalysis to demonstrate the polymer passivation effect for the traps intrinsically present in perovskite film, boosting its electrical and detection performance. Second, the addition of starch as a polymeric template for the fabrication of perovskite films, is an interesting strategy to confers to the film very high stability in ambient conditions, high homogeneity of the micrometer-thick film, enhanced mechanical flexibility and robustness.

Session 5 / **26**

Perovskites in porous networks for X-ray detection

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Porous materials have started to gather attention as promising X-ray detectors due to their ease of processing and chemical versatility enabling the incorporation of high Z atoms and maximising attenuation efficiencies while keeping functional porosity. Here, I will present our team's activities in functionalising porous structures with perovskites, including high throughput screening for highly dense, mechanically robust composites with fast response time, minimised dark current and outstanding operational lifetimes. A summary of our latest developments on indirect and direct detectors will be given, emphasising their potential for future innovations in the field.

Session 4 (Posters) / **27**

Deposition and Characterisation of Perovskite Films

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This research project investigates the performance of perovskite films and their suitability for use as radiation detectors, either as semiconductors or scintillators. We are particularly interested in films that are 10-100 μm thick, fabricated using various deposition methods.
Using the method of physical vapor deposition, FAPbBr3 films were produced atop a substrate. Photoluminescence carried out on the films showed that the peak emission wavelength of the films is close to that of the bulk single crystal of the same material, at around 557 nm. Line scans of the films were acquired using a profilometer, and by locating the 'step'in the line scan, the films were determined to be a few hundred nanometres thick. Scanning electron microscopy was used to investigate the structure and grain size of the films. We will go on to develop the films so that they reach the desired thickness, alongside investigating the suitability of other source materials, including CsPbBr3.

Session 4 (Posters) / **28**

X-ray nanoanalysis to probe radiation-induced ion migration in hybrid lead-halide perovskite

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X-ray direct detectors based on hybrid organic/inorganic lead-halide perovskite (HOIP) have gained significant attention from the research community over the last decade. In this context, the research has focused on identifying the optimal perovskite material. Among the HOIPs, 3D perovskites have demonstrated very good performances as X-rays detectors, but they are limited by high trap states density and significant ion migration effects leading to large dark currents. To overcome some of these limitations, 2D layered perovskites have gained interest for their lower dark current and reduced ion migration which leads to better environmental stability [1].

Nanoanalysis is a powerful tool to investigate microscopically the properties of perovskite films used for x-ray detectors. The simultaneous acquisition of X-ray Fluorescence (XRF) and X-ray Beam Induced Current (XBIC) can be exploited to study the correlation between charge collection and elemental distribution in perovskite films [2]. Indeed, through XRF it is possible to map the elemental distribution of the film, obtaining an image of the crystalline formations (nanocrystals or grains) composing the film. Combining XRF with XBIC it is possible to study the collection of radiation-induced charges with the boundary and bulk features of the film.

The stability of perovskite films under ionizing radiation and bias can be probed at the microscopic level to uncover degradation mechanisms and develop more stable films. A known issue in radiation detectors based on perovskites is the migration of metal ions from the electrode into the perovskite. In this work, we mapped the gold electrode degradation in two different perovskite materials: 3D $MAPbI_3$ [3] and 2D $(PEA)_2PbBr_4$ polycrystalline thin films deposited on interdigitated gold contacts. The film stability and the gold migration were probed at the nanoscale by simultaneous XRF and XBIC performed at the ID16B beamline of the European Synchrotron Radiation Facility. Gold ion migration was observed across the perovskite film in both materials. We investigated the origin of metallic ion migration, tying to decouple the effect of bias and radiation damage.

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[2] Stuckelberger, M. et al. *Engineering solar cells based on correlative X-ray microscopy.* J. Mater. Res. 32, 1825–1854 (2017).
[3] Verdi, M. et al. *X-ray nanoanalysis revealing the role of electronically active passivation layers in perovskite X-ray film detectors*; in press in Adv. El. Mater

Session 3 / **29**

On the Detection of High-Energy Protons by Perovskite Single Crystals: Enhanced Device Performance and Healing in MAPbBr3 and MAPbI3 Single Crystal-based Systems after Exposure to Protons

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Proton detectors play a crucial role in various fields, including radiation monitoring, medical imaging, and particle physics research. Recently, metal halide perovskites have emerged as a promising candidate for proton detection, offering high radiation tolerance alongside their demonstrated remarkable capabilities in detecting X-rays, gamma rays, alpha particles, electrons, and neutrons. These materials exhibit excellent charge transport characteristics, fast response times, high stopping power, and cost-effectiveness. Their versatile nature allows them to effectively capture and measure radiation across a wide spectrum, making them valuable for diverse radiation detection and imaging applications.
In this study, we fabricate proton detectors based on inverse temperature crystallization (ITC) grown methylammonium lead tribromide (MAPbBr3) and methylammonium lead triiodide (MAPbI3) single crystals using a coplanar structure. We then measure their response under high energy proton irradiation (68 MeV), commonly used for proton therapy of eye tumors, and confirm their ability to detect protons. Additionally, we investigate their stability up to a total proton dose of 10ˆ13 p/cmˆ2. Interestingly, we observe an initial degradation followed by a healing process that improves sensitivity by 6-fold and 202-fold for MAPbBr3 and MAPbI3-based detectors, respectively.
To confirm this healing effect, we then measure and evaluate transient photoluminescence (TRPL), photoresponse, and external quantum efficiency (EQE) after proton irradiation. We observe an increase in signal-to-noise (S/N) ratio by a factor of 11 for MAPbBr3 and 5 for MAPbI3-based detectors in photoresponse corroborating the improved detection capabilities, while longer TRPL lifetimes suggest improved charge carrier dynamics within the detector material. Moreover, we observe higher EQE indicating an improved efficiency in converting incident radiation into electrical signals. Together, these intriguing findings suggest a proton-induced self-healing of the perovskite proton detectors leading to enhanced functionality and improved performance. The results hence, highlight the suitability of MAPbBr3 and MAPbI3-based detectors for proton detection. Overall, these findings contribute to the ongoing efforts in advancing proton detection technology and pave the way for more accurate and reliable proton-related research and applications in various fields.

Session 4 (Posters) / **30**

Photoconductive Gain Effect: detection of ionizing radiation by highly defective semiconducting thin films-based photoconductors

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Photoconductive gain is a physical phenomenon typical of photoconductors, where a highly defective semiconducting photoactive material is between two ohmic contacts. Such trappingactivated mechanism amplifies the high energy radiation-induced current by a factor G, leading to e quantum efficiency exceeding 100% and providing high sensitivity to the radiation. It occurs when radiation-generated free charge carriers accumulate and pass several times through the semiconductor between the electrodes before recombination sets in. This process of amplification is activated by the trapping of minority charge carriers and the factor G can be expressed as the ratio between the recombination time, characteristic of the trapped states, and the transit time of free moving carriers [1].

Enhancing sensitivity and achieving higher gain are commonly the research focus for photoconductors. In the last years, photoconductive gain mechanism has been observed and deeply studied for the detection of both visible light and ionizing radiation by several semiconductor-based devices such as organic [2]–[4], nanomaterials [5]-[6] and also perovskites (e.g. Cs3Bi2I9 [7],(CH3NH3)3Bi2I9 [8], MAPbBr3 [9]). In all these cases, the common element is the presence of ohmic contacts and a highly defective active material. In particular, a trap-states, interfaces and doping engineering approach have been adopted to boost the gain factor and enhance the sensitivity [10].

Recently we modelled the photoconductive gain mechanism in Organic Field Effect transistor X-Ray direct detectors adopting several strategies and tuning different parameters to reach a full comprehension and control of this effect [11]. Besides, we employed a new technique called Photocurrent Spectroscopy Optical Quenching using both the X-Rays and visible light to study and identify the electrical traps in organic semiconductors which activate the photoconductive gain effect under ionizing radiation.

In this work we will discuss in deep all the advantages related to the presence of the photoconductive gain mechanism but we also highlight the main limits it imposes. In particular, we will focus on the most relevant challenges related to the presence of the gain and that have to be overcome to target the employment of these devices in real applications.

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Session 4 (Posters) / 31

Optimization of 2D perovskite thin films for photodetectors

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2D perovskite thin films have attracted significant attention for their potential use as high-performance detectors due to their unique optical and electronic properties. These materials crystallize in a natural self-assembled quantum well-like structure and possess several interesting features, including synthesis at relatively low temperatures (below 100℃) and low defect density.1 Furthermore, 2D perovskites have shown good stability in various environments, such as ambient conditions, humidity, and temperature, making them suitable for use in everyday applications. In photodetection, 2D perovskites have been used to create high-performance photodetectors with low dark current, high responsivity, and fast response time.2 The ability to deposit 2D perovskites using low-cost solution processing techniques, such as spin-coating and spray-coating, also makes them attractive for large-scale fabrication.
However, one of the main challenges in developing high-performance detectors is the deposition of high-quality thin films. The morphology and crystalline quality of the thin films can significantly affect their optical and electronic properties, which in turn can affect their sensitivity, response time, and stability.3 Therefore, the deposition conditions must be carefully controlled to ensure the formation of uniform and defect-free thin films.

In this study, we present the optimization of 2D perovskite thin films for photodetectors using chemical deposition in an inert nitrogen atmosphere. We focused on the hybrid perovskite PEA2PbBr4 (PEA = C6H5C2H4NH3+), deposited using the spin-coating method. The optimization of these thin films was achieved by changing the deposition speed, solvent for the perovskite solution, use of plasma cleaning before the deposition, addition of an anti-solvent during deposition, and changing the number of deposited layers. The deposition was carried out on Kapton samples on a glass substrate, at 800, 2000, and 6000 rpm. DMF and DMSO were used as solvents for preparing the perovskite solution. Samples were made with one and two layers of perovskite thin film. Additionally, we explored the impact of adding chlorobenzene as an anti-solvent. Before deposition, samples were treated with oxygen plasma. Our results showed that the best performance was achieved using a deposition speed of 2000 rpm with the addition of an anti-solvent. Moreover, the perovskite solution with DMF as a solvent showed better results in terms of adhesion of microcrystals to Kapton compared to the perovskite solution with DMSO. Our findings demonstrate the importance of optimizing the deposition process for 2D perovskite thin films to achieve better photodetector performance. Further research is needed to develop large-area films with even better uniformity which could be integrated into existing and new technologies for practical applications.

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Session 1 / **32**

Record stability for fully passive perovskite-based X-ray detectors through the use of a polymeric template

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High sensitivity and efficient X-ray detectors are needed to promote and boost their application as tools in medical diagnostics and radiotherapy. Lead halide perovskites have emerged recently as a novel class of material for efficient X-ray detection. Although 3D perovskites possess very interesting optoelectronic properties they suffer from low environmental and operational stability. Here we report a strategy based on using starch as a polymeric template for the fabrication of stable thin film perovskite X-ray detectors. The proposed p-i-n photodiodes can operate with no external

bias applied (fully passive devices), reaching a top sensitivity of 5.5±0.2 μC Gy-1 s-1. The device degradation was monitored for sample stored in air for a time window of 630 days, demonstrating an exceptional stability: 97% of the initial sensitivity was retained for the best perovskite-starch composite formulation making it the most stable unencapsulated perovskite X-ray detector reported so far.

Ref: Verdi et al, Advanced Materials Interfaces, 2023, doi 10.1002/admi.202300044

Session 2 / **33**

Overview of perovskite detectors research at Surrey

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I will present a summary of the perovskite detector research that is currently being carried out at Surrey. Our work is broadly organised into 3 themes, (1) single crystal lead halides for gamma spectroscopy, (2) polycrystalline perovskite thick films for X-ray imaging, and (3) perovskite scintillators. In this talk I will focus on the challenges of single crystal lead halides, and discuss recent results that compare the electronic and charge transport properties of FAPbBr3 and CsPbBr3 single crystals. The goal to produce gamma spectroscopy detectors from perovskite single crystals remains a key challenge, and I will present some of the current material problems which limit gamma ray detection performance, and strategies to solve these issues.

Session 1 / **34**

Perovskite thin film detectors for high energy photons and particle at the University of Bologna

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The research on perovskite radiation detectors at the University of Bologna is focused on thin film devices for the direct detection of radiation. We investigate the interaction between thin films and high energy photons and protons and we explore the best device geometries and fabrication process that allow scalability over large area and onto flexible substrates.

I will discuss the major goals we are targeting and the open issues we are presently facing in the development of detectors based on 2D, 2D/3D and polymer/3D perovskite blends with the aim to maximize their sensitivity, limit of detection and stability in time.

Session 6 / **36**

Closing Session and Future Workshop Planning

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Session 5 / **38**

Perovskite research at Freiburg

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A review of perovskite detector research as Freiburg.

Session 6 / **39**

Discussion Session: Thin Film Perovskite Detectors

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An open discussion about key results, challenges and future work around thin film and polycrystalline perovskite detectors

Session 3 / **40**

Discussion: single crystal perovskite detectors

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An open discussion about key results, challenges and future work around single crystal perovskite detectors