Assessment of carrier transport properties of perovskite CsPbCl3 single crystal with time-of-flight alpha-particle response

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Abstract

All-inorganic CsPbCl3 perovskite is a promising material for high-performance radiation detection owing to its extraordinary photoelectric properties and chemical stability. The critical challenge in developing widebandgap CsPbCl3 perovskite lies in the quantitative assessment of its charge transport properties, which regulate its performance optimization. Herein, we report the first spectroscopic α particles using CsPbCl3 detectors with asymmetric contact. CsPbCl3 single crystals (ϕ 15 × 50 mm3) were successfully grown using the Bridgman melt method and subsequently fabricated into Schottky-type Bi/CsPbCl3/Au detectors. Owing to its high electrical resistivity of 1.25 × 109 Ω · cm, the CsPbCl3 detector demonstrated a low dark current density (870 nA/cm2) and stable performance. The CsPbCl3 detector was also capable of resolving both the α particle (5.5 MeV) and γ-ray (59.5 keV) peaks from the 241Am radioactive isotope. Furthermore, the carrier transport properties of CsPbCl3 were evaluated quantitatively by the time-of-flight technology using 241Am α particle response, revealing the hole and electron mobilities as 11.12 and 12.92 cm2 · V−1 · s−2, respectively. Meanwhile, the hole and electron mobility-lifetime products were obtained as 1.72 × 10−4 and 1.04 × 10−4 cm2 · V−1, respectively. The planar CsPbCl3 detector achieved an excellent energy resolution of 14.9% at 122 keV under γ-ray exposure, which is the highest energy resolution reported to date for CsPbCl3 detectors. This study highlights the considerable potential of inorganic perovskite detectors for radiation detection and provides a practical approach for the future development of perovskite materials.

Full Text

Preamble

Assessment of carrier transport properties of perovskite CsPbCl3 single crystal with time-of-flight alpha-particle response∗ Qi-Hao Sun,1 Zhou Wu,1 Shu-Quan Wei,1 Yu-Quan Wang,1 Xu-Chang He,1 Bao Xiao,1 Nan-Nan Shen,1 Jian-Fu Zhang,2 and Yi-Hui He1, †

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All-inorganic CsPbCl3 perovskite is a promising material for high-performance radiation detection owing to its extraordinary photoelectric properties and chemical stability. The critical challenge in developing widebandgap CsPbCl3 perovskite lies in the quantitative assessment of its charge transport properties, which regulate its performance optimization. Herein, we report the first spectroscopic α particles using CsPbCl3 detectors with asymmetric contact. CsPbCl3 single crystals (ϕ 15 × 50 mm3) were successfully grown using the Bridgman melt method and subsequently fabricated into Schottky-type Bi/CsPbCl3 /Au detectors. Owing to its high electrical resistivity of 1.25 × 109 Ω · cm, the CsPbCl3 detector demonstrated a low dark current density (870 nA/cm2) and stable performance. The CsPbCl3 detector was also capable of resolving both the α particle (5.5 MeV) and γ-ray (59.5 keV) peaks from the 241 Am radioactive isotope. Furthermore, the carrier transport properties of CsPbCl3 were evaluated quantitatively by the time-of-flight technology using 241 Am α particle response, revealing the hole and electron mobilities as 11.12 and 12.92 cm2 · V−1 · s−2 , respectively. Meanwhile, the hole and electron mobility-lifetime products were obtained as 1.72 × 10−4 and 1.04 × 10−4 cm2 · V−1 , respectively. The planar CsPbCl3 detector achieved an excellent energy resolution of 14.9% at 122 keV under γ-ray exposure, which is the highest energy resolution reported to date for CsPbCl3 detectors. This study highlights the considerable potential of inorganic perovskite detectors for radiation detection and provides a practical approach for the future development of perovskite materials.

Keywords

CsPbCl3 perovskite; carrier transport; Bridgman method; radiation detection; semiconductor detector

INTRODUCTION

The accurate detection of X-rays and γ-rays from radioactive sources at room temperature is critical for various applications, including industrial quality control, medical imaging diagnostics, and homeland security monitoring [1-3]. Precise detection enables enhanced imaging resolution, material inspection, and safety protocols. In this context, semiconductors with high radiation-stopping power and large carrier mobility-lifetime products are considered ideal owing to their potential for superior energy resolution. However, their practical implementation has faced persistent challenges for decades [4, 5]. This difficulty originates primarily from the low flux of γ-ray photons, which requires highly sensitive detection materials. Furthermore, most semiconductors are highly sensitive to the structural defects and chemical impurities that are inevitably formed during synthesis. These imperfections create charge carrier traps, which significantly degrade the detector performance by reducing the chargecollection efficiency and energy resolution.

Existing γ-ray detector materials are relatively scarce and

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face several critical issues. For instance, traditional materials, such as cadmium zinc telluride (CdZnTe) and high-purity germanium (HPGe), offer promising detection capabilities, but suffer from high production costs and limited bulk fabrication yields [6]. Although HPGe provides excellent resolution, it requires cryogenic cooling to reduce the thermal noise, complicating its operational setup [7]. The fabrication of CdZnTe is constrained by crystal growth challenges, which lead to material inhomogeneity and reduced device efficiency. Overcoming these material and fabrication limitations remains the primary focus of radiation detection research. Efforts have been directed toward developing new materials with fewer intrinsic defects, optimizing crystal growth techniques, and exploring cost-effective fabrication processes to create reliable and scalable γ-ray detectors suitable for room-temperature operation.

In recent years, metal halide perovskites have attracted widespread attention in fields ranging from optoelectronics to radiation detection largely because of their exceptional optoelectronic properties and tunable bandgaps [8–12].

Among them, lead halide perovskites with the general formula CsPbX3 (X = Cl, Br, I) have demonstrated significant potential in high-efficiency solar cells [13], light-emitting devices [14], and photodetectors [15] owing to their high absorption coefficients, efficient charge transport, and remarkable defect tolerance [16, 17]. Notably, these materials can be chemically modified through halogen substitution, enabling the engineering of their electronic and optical properties to meet various application requirements.

Among the CsPbX3 family members, CsPbI3 is prone to a destructive phase transition to a yellow perovskitoid phase

at room temperature, which hinders its long-term stability in practical applications [18, 19]. In contrast, CsPbBr3 has been successfully grown with high crystalline purity and large single-crystal volume [20, 21]. Its intrinsic properties, including suitable bandgap, high absorption efficiency, and exceptional defect tolerance, make it a promising candidate for radiation detection. CsPbBr3 detectors with asymmetric contacts demonstrated high resistivity and a mobility-lifetime ($\mu\tau$) product exceeding 10−3 cm2 /V for holes. Furthermore, these detectors achieved remarkable energy resolution under hard X-rays and γ-rays, achieving an energy resolution of 3.9% under 122 keV 57 Co γ-rays [8].

Its counterpart, CsPbCl3 , which is characterized by a wider bandgap, exhibits strong chemical stability under ambient conditions, making it particularly suitable for high-energy photon detection. Its high effective atomic number (69.8) and density (4.2 g/cm3) contribute to its exceptional radiationstopping power [22]. Although CsPbCl3 is theoretically expected to possess favorable transport characteristics owing to its ionic bonding and robust structural stability, comprehensive experimental data on its carrier mobility and lifetime are limited. Studies on CsPbCl3 mobility–lifetime products and carrier mobility are crucial because these parameters directly influence the charge collection efficiency and overall device performance.

Given its stability and promising theoretical transport properties, CsPbCl3 is emerging as a potential high-performance semiconductor for radiation detection [23]. Its ability to withstand environmental degradation while maintaining its structural integrity makes it particularly appealing for long-term device applications. Understanding and optimizing its transport properties could pave the way for next-generation detectors capable of operating at room temperature with superior energy resolution. Further exploration of crystal growth techniques, defect passivation strategies, and device engineering could reveal the full potential of CsPbCl3 in radiation detection and related optoelectronic applications. Thus, the continued investigation of its fundamental properties is imperative to advance its applicability in cutting-edge technologies.

In this study, we successfully utilized the Bridgman method to grow high-quality and large-volume (ϕ 15 × 50 mm3) single crystals of CsPbCl3 with excellent optical and structural uniformity. The superior quality of the crystal was demonstrated by its remarkable performance as a radiation detector. Employing an asymmetric electrode design, the CsPbCl3 -based detectors demonstrated a stable dark current and an outstanding energy resolution of 14.2% for 241 Am α particles (5.5 MeV) and 14.9% for 57 Co γ-rays at room temperature (25 $\circ$ C). Note that this represents the highest energy resolution reported thus far for a CsPbCl3 detector.

For the first time, the charge transport properties of CsPbCl3 crystals were quantitatively characterized using the time-offlight (ToF) method under 241 Am α particle excitation. The hole and electron mobilities were determined as 11.12

and 12.92 cm2 · V−1 · s−2 , respectively. Moreover, the hole and electron mobility-lifetime products, a critical figure of merit for detector materials, reached 1.72 × 10−4 and 1.04 × 10−4 cm2 · V−1 , respectively. These exceptional transport properties highlight the considerable potential of the allinorganic perovskite CsPbCl3 for room-temperature X/γ-ray radiation detection with high resolution and reliability.

EXPERIMENTAL

Materials Synthesis

CsPbCl3 polycrystalline material was synthesized directly from high-purity precursor compounds, namely, cesium chloride (CsCl, Sigma-Aldrich, 99.999%) and lead chloride (PbCl2 Sigma-Aldrich, 99.999%), through a solid-state reaction with a precise 1:1 stoichiometric ratio. The entire synthesis and crystal growth processes were conducted within the same sealed quartz ampoule (13 mm outer diameter, 10 mm inner diameter) to ensure a controlled environment and prevent contamination. The ampoule was initially evacuated to an ultrahigh vacuum level of less than 5 × 10−5 Pa and subsequently sealed to maintain a clean reaction atmosphere. The sealed ampoule containing the precursor materials was placed in a rocking furnace to facilitate uniform heating and promote the homogeneous mixing of the reactants. The synthesis was performed at 620 ∘ C for 24 h, enabling the formation of highquality CsPbCl3 polycrystals. After the reaction, the resulting material exhibited a pale yellow appearance, indicating successful compound formation. This synthesis method ensured efficient crystal growth while minimizing the introduction of unwanted impurities, thereby rendering the produced CsPbCl3 suitable for advanced optoelectronic and radiation detection applications.

Single-Crystal Growth

After synthesis, CsPbCl3 single crystals were grown using the vertical Bridgman method, which is well suited for producing high-quality crystalline materials. A sealed quartz ampoule containing the synthesized material was carefully placed in a two-zone Bridgman furnace designed for controlled crystal growth. The upper and lower furnace zones were maintained at the temperatures of 650 ∘ C and 300 ∘ C, respectively, establishing a stable temperature gradient estimated to be in the range 15 20 ∘ C/cm. This gradient is crucial for directing crystal growth along the desired axis. During the growth process, the ampoule was lowered at a slow and controlled rate of 0.5 1.0 mm/h to ensure uniform crystal formation while minimizing the incorporation of defects.

After completing the growth process, the furnace was gradually cooled to room temperature at a controlled rate of 4 8 C/h to reduce the thermal stress and prevent cracking. As the melting point of CsPbCl3 is 611 ∘ C, an overheating step was applied by maintaining the temperature at 620 ∘ C for 10 h before starting the crystal growth. This step ensured the complete melting of the raw materials and improved the crystal quality by facilitating better mixing and uniformity of the melt. The ingot was cooled to 60 ∘ C in 100 h. Owing to the existence of phase transitions, the ingot was cooled to room

temperature at a cooling rate of 1 ∘ C/h. Note that excessive temperature gradient, dropping speed, and cooling rate likely resulted in cracks and the final fracture of the ingots.

Structure and Characterization of Photoelectric Properties

Powder X-ray diffraction (XRD) patterns were collected using an X' Pert PRO X-ray powder diffractometer equipped with a Cu Kα radiation source (λ = 1.5406 Å). The diffraction data were recorded over a 2θ range from 10∘ to 70∘ at a tube voltage of 40 kV and a tube current of 40 mA, ensuring high-quality diffraction patterns for phase identification and crystallinity evaluation. This analysis provides detailed structural information regarding the synthesized CsPbCl3 crystals.

The optical properties of the samples were examined using UV–vis spectroscopy performed on a UV-3150 spectrometer, covering a wavelength range of 400–900 nm. This enabled the precise measurement of the optical absorption edge and the determination of the bandgap energy. The electrical properties were evaluated by measuring the resistivity and dark current in the low bias voltage range of -1 to 1 V. The current—voltage (I-V) characteristics were recorded using a Keithley 6517B electrometer under dark conditions. The voltage was swept from 0 to +100 V and 0 to -100 V, following procedures established in our prior studies.

Device Assembly

After slicing, the CsPbCl3 crystals were meticulously polished to achieve smooth surfaces, which are essential for optimal electrode adhesion and device performance. To eliminate any residual surface contaminants, the polished crystals were thoroughly rinsed in toluene, an effective solvent for removing organic impurities and ensuring clean surfaces for subsequent electrode deposition. Asymmetric planar Bi/CsPbCl3 /Au detectors were constructed for device fabrication. Bismuth and gold electrodes were deposited onto the crystal surfaces via thermal evaporation using a precisionengineered shielding mask to define the electrode geometry.

The electrode thickness ranged between 50 and 90 nm and was tailored to achieve optimal electrical contact and minimize resistance. The electrodes were designed to cover the entire surface area of the crystal, ensuring a uniform electric field distribution across the detector. Electrical connections to the external measurement circuitry were established by attaching copper wires to the electrodes, facilitating efficient signal transmission during device operation.

Detector Performance Characterization

In this study, 1 µCi 241 Am α particles and a 14 µCi Co γ-ray source were used to evaluate the detector performance. The primary decay from 241 Am to 237 Np generates an α particle with the characteristic kinetic energy of 5.5 MeV, along with a γ-ray photon with the characteristic

energy of 59.5 keV. The pulse shape information was simultaneously recorded by connecting the preamplifier output to a high-speed waveform digitizer card. The signals from the preamplifier (eV-550) were further amplified and shaped using an amplifier (ORTEC 572A) with a gain of 0.8 × 200 and the shaping time constants of 2, 3, 6, and 10 µ s. The final signals were subsequently evaluated using a dual 16 K input multichannel analyzer (ASPEC 927) and analog-to-digital converter, generating a response spectrum displayed on the MAESTRO-32 software. The energy resolution of the characteristic peak was determined using the full width at half maximum (FWHM, in keV) / (peak energy) × 100%. $\mu\tau$ was calculated using the single-carrier Hecht equation. The mobility was estimated using pulse height spectroscopy according to the equation $\mu = d2 /(tr \cdot V)$, where tr is the rise time, d is the thickness of the crystal, and V is the bias. Under a certain bias, the average value of the rise time distribution was used for tr .

RESULTS AND DISCUSSION

Centimeter-sized CsPbCl3 single crystals were grown from the melt using the vertical Bridgman method. Powder XRD experiments on the as-grown CsPbCl3 crystals matched well with the standard card for CsPbCl3 (PDF#18-0366). The ideal crystal composition was estimated to be Cs: Pb: Cl = 1:1:3 from energy-dispersive X-ray spectroscopy (EDS) measurements (Fig. S1 and Table S1). No peaks related to the impurity phases were observed, indicating that the crystal was a pure CsPbCl3 single crystal (Fig. 1a [Figure 1: see original paper]). Both the as-grown crystal ingot and the polished wafer had a pale yellow color with excellent optical transparency (Fig. S2). The optical transmission exceeded 70% when the wavelength of the incident light ranged from 300 to 900 nm (Fig. 1b). The high transmission confirms that the as-grown single crystals possess negligible absorption centers below the bandgap. This observation enabled the calculation of the absorption coefficient α, thereby facilitating the determination of the bandgap Eg of the CsPbCl3 crystals near the absorption cutoff. The optical absorption edge extracted from the transmission spectrum was 434 nm. The inset of Fig. 1b demonstrates the (Eg) calculations for the direct transitions in CsPbCl3 using the following equation: $(\alpha h\upsilon)2 \quad (h\upsilon - Eg)\}$,

where $h\upsilon$ is the photon energy. Extrapolating these calculations revealed that the CsPbCl3 samples have Eg values close to 2.86 eV.

A CsPbCl3 planar detector with asymmetric electrodes was prepared using evaporated Bi and Au metals. Bi metal has a relatively low work function (ϕBi 4.2 eV) and can form a large potential barrier preventing hole injection from the electrode, whereas Au, which possesses a high work function (ϕAu 5.1 eV), generates a large potential barrier for electrons. The I–V curve of the as-prepared Bi/CsPbCl3 /Au device exhibited asymmetrical characteristics (Fig.

1c) because of the dissimilar Schottky barriers under forward and

for the CsPbCl3 single crystal. Inset: Tauc plot showing the direct bandgap of 2.86 eV. (c) Typical I-V characteristic curve of a Schottky-type Bi/CsPbCl3 /Au detector. (d) Temporal characteristics of the Schottky-type Bi/CsPbCl3 /Au detector under positive (forward) and negative (reverse) applied bias conditions

reverse biases. The electrical properties were characterized using I-V tests. As demonstrated by the I-V curve, the results align well with the Ohmic law at a bias of $±$1 V, and these data can be used to calculate the resistivity according to the following equation:

where ρ is the resistivity, S is the contact area of 4 mm × 5 mm, L is the sample thickness (2 mm), and R is the resistance.

In particular, the electrical resistivity of CsPbCl3 obtained by fitting was 1.25 × 109 Ω · cm. The dark current density of this device was 870 nA/cm2 at -1000 V/cm, in contrast to 7.4 µA/cm2 at 1000 V/cm. The temporal stability of the dark current is vital for maintaining the high stability of the perovskite detector. For the Schottky-type Bi/CsPbCl3 /Au detector, the density of the dark current on the reverse side tended to decrease slightly with time, but it appeared to reach a limit at -155 nA under -100 V bias (Fig. 1d). By contrast, the dark

current on the forward side continued to increase for 950 s, which increased the background noise and degraded the energy resolution and detector stability. The photoresponse of the Bi/CsPbCl3 /Au detector was measured using a 420 nm LED at -10 V, where the ratio of the photocurrent to the dark current was larger than 65, as shown in Fig. S3.

A schematic illustrating the charge-carrier transport process induced by α particles in the CsPbCl3 crystal is shown in Fig. 2a [Figure 2: see original paper]. To ensure better collimation of the α radiation emitted from the 241 Am source, rubber putty was employed as the encapsulating material. This configuration enabled a more directional incidence of the emitted α particles onto the detector surface, minimizing scattering and improving measurement precision. Upon irradiation, the interaction of α particles with the CsPbCl3 crystal generates electron-hole pairs owing to ionization. Under the influence of an externally applied electric field, electrons drift toward the anode, whereas holes migrate toward the cathode, facilitating charge collection. The Bi/CsPbCl3 /Au detector, based on a

spectra under negative (b) and positive (c) voltages using a Schottky-type Bi/CsPbCl3 /Au detector. As indicated in the insert, the dimensions of the crystal are 5 mm × 4 mm × 1 mm. (d) 241 Am α particle spectrum resolved concurrently by a Schottky-type Bi/CsPbCl3 /Au detector at -250 V

Schottky-type structure, successfully resolved the full energy spectrum of a collimated 241 Am α radiation source. This marked a significant achievement, as for the first time, the CsPbCl3 detector distinctly resolved the primary high-energy

α particle peak alongside the associated low-energy γ-ray peak. The appearance of these two distinct peaks corresponds to the dual decay modes of the 241 Am α isotope: the emission of energetic α particles and the subsequent generation of lower-energy γ-rays. The ability to separate these two radiation products highlights the potential of CsPbCl3 as a material for high-resolution radiation detection applications. The bias dependence of the energy spectra for α-particle detection was further examined, and the corresponding results are shown in Fig. 2b and 2c for hole and electron collection, respectively. An increase in the applied bias voltage led to an enhanced charge collection efficiency, as evidenced by the prominent peak shift toward higher channel numbers. This behavior indicates improved carrier extraction owing to the stronger electric field, which accelerates charge carriers and

reduces recombination losses. Interestingly, a notable disparity was observed between the electron and hole collection efficiencies. Specifically, the electron response exhibited only weak shoulders in the spectra without an apparent peak resolution. This asymmetry suggests that significant electron trapping occurred within the CsPbCl3 crystal, likely owing to intrinsic defects or trap states. In contrast, the hole collection efficiency was substantially higher, leading to more distinct peak formation in the spectra. This result underscores the importance of mitigating charge-trapping effects, particularly for electrons, to achieve optimal detector performance.

Future improvements in crystal growth techniques and device engineering may further enhance the potential of this material for advanced radiation detection applications. Under an applied voltage of -250 V, the Schottky-type CsPbCl3 detector achieved the energy resolutions of 14.2% for 5.5 MeV α particles and 20.5% for 59.5 keV γ-rays (Fig. 2d), which is the maximum α-particle spectral resolution for a CsPbCl3 single-crystal detector. This is also the highest γ-ray detec-

tion energy resolution currently reported for a CsPbCl3 detector. The excellent spectral responses were attributed to the prominent carrier transport properties, and the ToF technology was adopted to evaluate the charge-carrier mobility of the CsPbCl3 crystal. of α particles with an energy of 5.5 MeV in CsPbCl3 single crystals, obtained through Monte Carlo calculations using the SRIM software. This simulation provides crucial insights into the interaction dynamics of α particles within a crystal lattice.

As shown in Fig. 3b [Figure 3: see original paper], the depth distributions of α particles reveal a shallow penetration depth of approximately 27.4 µm.

This limited penetration results in the majority of electron–hole pairs being generated and concentrated near the cathode region of the crystal. Consequently, the detected charge signal is predominantly attributed to the drift of a single carrier type with either positive or negative polarity, depending on the orientation of the applied electric field. The induced transit current generated by these drifting carriers was converted into a voltage pulse using a charge-sensitive preamplifier. The preamplifier output was subsequently digitized using a high-speed waveform digitizer card for precise signal collection and analysis. The carrier mobility was evaluated by measuring the rise time of the voltage pulse as a function of the applied bias voltage. The drift velocities (υ) of the carriers were determined using the following equation:

where d represents the thickness of the CsPbCl3 crystal, and tr is the rise time measured between the 10% and 90% amplitude points of the signal pulse. Typical voltage pulses extracted from the preamplifier at an applied bias of -300 V are shown in Fig. 3c. These pulses provide direct evidence of charge-carrier transport behavior within the crystal under an external electric field. Furthermore, Fig. 3d shows the voltage pulses induced by α particles at various applied bias voltages, specifically for hole transport. As the applied voltage increased, the amplitude of the preamplifier signal increased, whereas the rise time decreased. This behavior reflects the accelerated drift velocity of the charge carriers owing to the stronger electric field, which enhances the chargecollection efficiency. To gain a deeper understanding of the charge transport properties in the Bi/CsPbCl3 /Au Schottky detector, it is important to emphasize that the device operates in the pulse mode, where each radioactive event is independently collected and processed. This methodology enables the extraction of key charge transport parameters through a statistical analysis of the raw transient pulses produced by the detector. The transient pulses were carefully analyzed using a preamplifier to extract critical characteristics, such as the amplitude and rise times of the signals, which were subsequently categorized into hole and electron drift contributions. To distinguish the charge-collection modes dominated by holes and electrons in the crystal, we irradiated the device with 241 Am α particles and 57 Co γrays from the anode and cathode sides, respectively, as shown in Fig. S4. Notably, the bias voltage applied to the device remained constant throughout the test-

ing process, with only the direction of the incident radiation being altered. Consequently, signal flooding owing to an increase in the dark current under a positive-bias state is not a concern. The device was consistently operated in a low-darkcurrent state with a negative bias voltage.

As shown in Fig. 4 [Figure 4: see original paper] and S5, the distributions of both the amplitude and rise times were analyzed across 2000 events, providing insights into the drift dynamics of the charge carriers within the CsPbCl3 crystals. An interesting trend observed in these analyses was a shift in the peak position of the signal amplitude distribution as the applied voltage increased shifted to higher values with increasing voltage, suggesting that the charge-collection efficiency improved at higher electric fields. This behavior is consistent with the expected response of semiconductor detectors under bias, where higher voltages generally enhance the drift velocity of charge carriers, thus leading to stronger signals. In addition, the corresponding rise-time distributions narrowed as the voltage increased, as shown in Fig. 4b. This narrowing effect indicates faster charge collection times, as the average rise time decreased with increasing applied voltage. These observations are further supported by a detailed analysis of the experimental data, which reveals an evident correlation between the voltage and charge transport properties of the detector. To quantify the charge transport characteristics, the experimental mobility-lifetime product (µτ) was calculated using the single-carrier Hecht equation. This equation provides a relationship between the signal amplitude and the applied voltage, which can be used to deduce the product of the charge carrier mobility (µ) and lifetime (τ). The calculated µτ values for the hole and electron carriers were 1.72 × 10—4 cm2 · V—1 and 1.04 × 10—4 cm2 · V—1 , respectively, as shown in Fig. 4c and S5c. These values are significant because they enable the evaluation of the charge transport efficiency in the CsPbCl3 material, which is crucial for its performance as a radiation detector. Furthermore, the mobility values of the hole and electron carriers were calculated separately as approximately 11.12 cm2 · V—1 · s—2 and 12.92 cm2 · V—1 · s—2 , respectively, as shown in Fig. 4d and S5d. These mobility values are comparable to those reported in a previous study, reinforcing the validity of the experimental approach and the consistency of the material properties.

Note that the mobility values calculated in this study were somewhat lower than those obtained using 241 Am γ-rays in previous experiments. This difference in mobility values can be attributed to the distinct interaction depths between the Am α particles and the material. The interaction depth of α particles is much smaller than that of γ-rays, resulting in a reduced range of charge carrier transport and lower signal amplitude. The penetration length of 241 Am α particles in CsPbCl3 is approximately 27.4 µm, which is much smaller than the thickness of the detector; thus, the 241 Am α particles interact with a relatively small portion of the detector material. This limitation of the interaction depth of α-particles partially explains the lower µ values observed in this study.

In addition to the mobility calculations, the lifetime of the hole carriers (τ h) in CsPbCl3 was determined based on the

(b) Depth distributions of α particles in CsPbCl3 . (c) Pulse signal of the preamplifier including amplitude and rise time under α particles at -300 V. (d) Hole pulse signal of the preamplifier under various biases

previously calculated values of µh and µτ h . The hole lifetime was determined to be approximately 15.47 µs, which is significantly longer than the hole lifetime observed in several other compound semiconductors. This value is comparable to the hole lifetime of CsPbBr3 , another perovskite material that has demonstrated strong potential for radiation detection [9]. The longer lifetime of the holes is particularly important because it suggests that the CsPbCl3 material can sustain charge carriers for a longer duration, enabling more efficient charge collection and, ultimately, a higher detection sensitivity. These results demonstrate the excellent charge-transport properties of the CsPbCl3 crystals grown via the Bridgman melting method, confirming their considerable potential for applications in room-temperature radiation detection. The long hole lifetime, high carrier mobility, and strong signal response to the applied voltage suggest that CsPbCl3 is an ideal candidate for advanced radiation detection technologies.

The γ-ray spectroscopic response of the Schottky-type CsPbCl3 detector is shown in Fig. 5 [Figure 5: see original paper], providing detailed insights into its performance when irradiated with γ-rays. The

experiment utilized a noncollimated 241 Am γ-ray source (1 µCi) irradiated from the upper Bi electrode. Remarkably, the fine spectrum of the 241 Am isotope was fully resolved by the Schottky-type Bi/CsPbCl3 /Au detector in both the hole and electron collection configurations. This ability to resolve the γ-ray spectrum highlights the exceptional performance of CsPbCl3 as a radiation detector, and provides a clear and reliable energy spectrum that can be used for various radiation detection applications. The γ-ray energy spectra derived from hole- and electron-dominated charge collection are shown in Fig. 5a and 5b, respectively. These figures illustrate the bias dependence of the γ-ray energy spectra with a noticeable difference between the hole and electron collection configurations. As the applied bias voltage increased, an apparent improvement in the charge transport was observed, as evidenced by the peak shift to greater channel numbers.

This shift indicates an enhanced charge collection efficiency at higher bias voltages, which corresponds to the increased drift velocity of the charge carriers under stronger electric fields. This trend is expected in most semiconductor detec-

particles. (b) Rise time distribution (2000 events) attributed to hole drift based on the transient pulse analysis for 241 Am α particles. (c) Mobility-lifetime product calculation for holes according to the single-carrier Hecht equation. (d) Hole mobility calculation for CsPbCl3

tors where higher voltages facilitate the movement of charge carriers, leading to more efficient signal generation. In the hole collection configuration, the CsPbCl3 detector achieved an impressive energy resolution of 20.1% for 241 Am γ-rays at an applied bias of -180 V. This result is notable because the energy resolution is a critical factor in evaluating the performance of radiation detectors. A high energy resolution ensures that the detector can distinguish between the different energy levels of the incoming radiation, which is essential for identifying and quantifying radioactive isotopes. However, the electron collection configuration demonstrated an inferior performance compared with hole collection. In the electron collection mode, the detector exhibited only a counting response to the γ-rays with visible spectral shoulders but no apparent spectral resolving ability. This lack of resolution in the electron collection mode is presumed to be due to the charge trapping of electrons in the CsPbCl3 material. Charge trapping occurs when charge carriers are captured by defects or impurities in a material, which reduces their mobility and prevents efficient charge collection. This is a known issue in several semiconductor detectors and suggests that the further optimization of the CsPbCl3 material is required to reduce electron trapping and improve its performance in the electron collection mode.

In addition to the 241 Am γ-rays, the γ-ray energy spectra of the 57 Co γ-ray source (14 µCi) were measured, as shown in Fig. 5c. The 57 Co γ-rays, which have an energy of 122 keV, provide additional insights into the performance of the detector in terms of the energy resolution and spectral features. Notably, the Pb escape γ-ray peak, which is a well-known feature in γ-ray spectra, was observed at a channel number of approximately 336 when the applied bias

was greater than -150 V. The Pb escape X-ray peak occurs when a γ-ray interacts with the detector material and generates an electron that causes the emission of low-energy X-rays from the lead component of the detector. The detection of this peak is a characteristic feature of high-resolution γ-ray spectroscopy, and its observation further confirms the ability of the CsPbCl3 detector to resolve the fine spectral features. At a higher applied bias of -240 V, the Schottky-type CsPbCl3 detector achieved an energy resolution of 14.9% for the 122 keV γ-rays from the 57 Co source, as shown in Fig. 5d. This represents the highest energy resolution for 122 keV γ-rays reported for CsPbCl3 crystals to date, which is a significant achievement for wide-bandgap materials, such as CsPbCl3 . The ability to achieve such a high energy resolution in a material with a relatively wide bandgap is impressive, as wide-gap materials typically struggle with charge transport and spectral resolution owing to the presence of defects and poor charge-carrier mobility. The excellent energy resolution achieved in this study demonstrates the potential of CsPbCl3 for high-performance γ-ray detection. Note that the CsPbCl3 crystals used in this study were prepared without the intentional purification of the starting binary precursor compounds or CsPbCl3 material. This is in contrast to the preparation of other high-performance radiation detectors, such as CdZnTe (CZT), where the intentional purification of the precursor materials is typically required to achieve detector-grade performance [2, 24, 25]. The lack of purification steps during the preparation of CsPbCl3 highlights the inherent defect-tolerant nature of this material, which is a key advantage of perovskite materials. This characteristic makes CsPbCl3 particularly attractive for use in radiation detectors because it simplifies the fabrication process and reduces costs

transport collection under 241 Am γ-rays. (c) 57 Co γ-ray spectrum by the same CsPbCl3 crystal under the same conditions. (d) 57 Co γ-ray response in 3 min under the applied voltage of -240 V with a shaping time of 10 µs

compared with other conventional semiconductor materials.

By contrast, other conventional halide semiconductors, such as HgI2 and TlBr, require multistep and complicated purification processes to achieve the necessary material quality for radiation detection [26]. These purification steps often involve expensive and time-consuming procedures, which increase the overall cost and complexity of the detector fabrication process. The simplicity of preparing CsPbCl3 for radiation detection without the need for extensive purification further emphasizes its potential as a practical and cost-effective material for radiation detection applications. Overall, the results of the γ-ray spectroscopic response demonstrate the exceptional performance of the Schottky-type CsPbCl3 detector. The ability to achieve high energy resolution and spectral features with minimal preparation steps demonstrates the potential of CsPbCl3 as a leading material for radiation detection with possible applications in nuclear security, medical imaging, and environmental monitoring.

SUMMARY

The wide-bandgap CsPbCl3 perovskite has considerable potential for γ-ray detection at room temperature, similar to CsPbBr3 . We successfully utilized the Bridgman melt method to grow centimeter-sized detector-grade crystals with exceptional optical and physical properties. The carrier transport properties of CsPbCl3 crystals were characterized for the first time using α particles via the ToF technology. An asymmetric electrode design (Bi/CsPbCl3 /Au) was implemented to maintain a low and stable dark current. The as-prepared detector exhibited a stable and low dark current (870 nA/cm2), even under a high electric field (1000 V/cm). The Schottkytype Bi/CsPbCl3 /Au detector resolved both the α particle spectrum and coincident γ-ray spectrum from a 241 Am radiation source. The energy resolution for the 5.5 MeV α particle peak was 14.2%. According to the α particle spectrum and corresponding rise time distribution, the hole and electron mobilities were calculated as 11.12 and 12.92 cm2 · V−1 · s−2 , respectively. The CsPbCl3 detector exhibited a high hole mobility-lifetime (μτ)h product of 1.72 × 10−4 cm2 /V

compared with the electron mobility-lifetime (μτ)e product of 1.04 × 10−4 cm2 · V−1 . Furthermore, an excellent energy resolution of 14.9% was achieved using 57 Co γ-rays with a characteristic energy of 122 keV. Overall, CsPbCl3 is the first chloride-based semiconductor reported to date with excellent γ-ray detection capabilities in the bulk form. Most other materials considered promising for γ-ray detection exhibit poor counting properties, lack energy resolution, and ultimately fail to satisfy the required standards. CsPbCl3 is not only the second member of the all-inorganic perovskite family, but also one of the few semiconductor materials across various families that can achieve an outstanding energy resolution. These results highlight the exceptional detector performance and charge transport properties of CsPbCl3 single crystals, highlighting their potential for broad applications in room-temperature radiation detection.

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Note: Figure translations are in progress. See original paper for figures.