All-inorganic metal halide perovskites are highly attractive materials for optoelectronics and are typically implemented as thin films, nanocrystals, and single crystals with macroscopic and millimeter-size dimensions. Single crystals with regular shapes in the micrometer range are less explored and present an interesting alternative when a well-defined and spatially localized response is desired. Here, CsPbBr₃ microcrystals are fabricated by simple and fast drop-casting of a precursor solution on several substrates that for the growth process are heated to temperatures in the range from 80 to 150 °C. The microcrystals have a cubic shape and feature a pyramidal cavity on their top surface that forms due to faster growth at the edges once the growing crystal is attached to the substrate. Distinct heterogeneity in the photoluminescence (PL) and conductivity of the different facets are measured by µ-PL and conductive atomic force microscopy experiments. Toward device applications, the authors contact single microcrystals on conductive substrates mechanically with a micromanipulator tip. They observe diode-like current–voltage curves and good photodetector functionality, obtaining a high responsivity of 150–300 A W⁻¹ in the blue–green spectral region under forward bias, and a sharp detection band centered around 540 nm with peak responsivity close to 0.7 A W⁻¹ under reverse bias.

1. Introduction

Recently, all-inorganic perovskite materials (CsPbX₃, X = Cl, Br, and I) have gained enormous attention in material science, and their potential technological applications have been intensively explored toward solar cells,[1,2] light-emitting diodes,[3,4] lasers,[5] and photodetectors.[6–8] These low-cost semiconductors exhibit intriguing optoelectronic properties, such as high carrier mobility,[9] narrow emission bandwidth,[10] long carrier diffusion length,[9] and large optical absorption cross section.[11] Their material properties are influenced by the orientation of the perovskite crystals and thus, efforts have been devoted to control their crystallographic orientation in films,[12–14] and single crystals,[15–17] as well as to synthesize them in 1D or 2D microscopic morphologies in order to provide preferential pathways for charge transport.[18–20] Toward single crystals, a few studies have reported that charge mobility and photoresponse depend on the exposed crystallographic facets.[10,21,22] In particular, it was found that the (001) facet of CsPbBr₃ crystals is more efficient for photodetection than the (010) and (100) facets. More recently, K. Wang et al.[22] showed that the optimization of the crystallographic facets, in this case of the (001), provides a route for enhanced in-plane photoresponse under polarized light. Facet-dependent heterogeneity in the photocconductivity of single grains in hybrid perovskite thin films has been also investigated, for example via conductive atomic force microscopy (C-AFM), and revealed how crystal facets impact the key properties for solar cells such as the short-circuit current and open circuit voltage.[23] Similar studies were previously reported for methyl ammonium (MA) based perovskites,[24,25] which show that such anisotropy is strongly related to MA segregation and high PbBr density. High-resolution mapping of the photocurrent revealed spatial variations that were related to fluctuations in the trap density.[24] The advantages of large, micro-, to centimeter-sized single crystal structures for optoelectronics are based on the suppression of grain boundaries and reduced defects.[25] However, in most of the cases these 3D structures with mainly cubic-shapes are formed through synthetic protocols that involve large incubation times and mixtures of solvents,[27,28] and faster fabrication strategies that open access for size and shape control could ultimately boost the impact of such materials in optoelectronic devices. Toward more environmentally friendly devices, the use of Pb-free

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perovskite materials provides appealing solutions for photodetection with high responsivity.[29,30]

Here, we fabricate CsPbBr$_3$ microcrystals by straightforward drop-casting of a CsBr and PbBr$_2$ precursor solution on substrates that are heated to a temperature in the range of 80–150 °C. In our experiments, the size and shape of the crystals are controlled by the substrate temperature, precursor concentration in dimethyl sulfoxide (DMSO), and the amount of solution that is deposited. Interestingly, we obtain very regular cuboid microcrystals that feature a pyramidal cavity on their top surface. In situ observation of the crystal growth reveals that the cavity forms once the growing microcrystal in solution is attached to the substrate surface. Detailed compositional (via scanning electron microscopy (SEM) combined with energy dispersive spectroscopy (EDS)) and morphological (SEM and AFM) analysis point to a step-like geometry of the internal facets of the cavity. Under above-band gap excitation, we observe a double peak in photoluminescence (PL), and with µ-PL experiments we find that the high-energy component is more pronounced on the internal cavity facets as compared to the cuboid edge and rim. The different facets on the top surface also led to distinct electrical properties. With C-AFM we measure higher current on the inclined surface facets of the cavity and on the external cuboid side facets than on the planar rim and center regions, which indicates that the step-like morphology is advantageous for electrical contact. Finally, we explore the single microcrystals as photodetectors by fabricating them on a conductive substrate, such as Au or indium tin oxide (ITO), that serves as back contact, while the top surface is contacted with a micromanipulator tip. We observe highly asymmetric current–voltage responses that feature a diode-like behavior and hysteresis with respect to forward/backward voltage sweeps. The microcrystal photodetectors show different response for operation under positive (reverse) or negative (forward) voltage bias, with highly competitive photosensitivity under forward bias reaching a responsivity of 300 A W$^{-1}$ and a detectivity of 10$^{14}$ Jones,[8] and narrow-band photodetection under reverse bias with around 0.7 A W$^{-1}$ responsivity at 540 nm.

2. Results and Discussion

2.1. CsPbBr$_3$ Microcrystal Fabrication and Structural Properties

We fabricate CsPbBr$_3$ microcrystals by depositing a solution of CsBr and PbBr$_2$ precursors in DMSO on planar substrates that are suitable for device fabrication such as Si, SiO$_2$, gold on mica, and ITO-coated glass. The precursor solution is deposited by drop-casting, and the substrates are heated to temperatures in the range of 80–150 °C until a dry crystal deposit is formed, which occurs within minutes. Depending on temperature, precursor solution, and reaction time, we obtain cuboid-shaped microcrystals with edge lengths ranging from 10 to 150 µm and height of 1–50 µm, as depicted in Figure 1a. The size of the microcrystals can be controlled by the concentration of the precursors in the DMSO solution, and by the temperature of

Figure 1. CsPbBr$_3$ microcrystals with pyramidal cavities on their top surface. a) Typical microcrystal ensemble obtained by precursor drop-casting on a planar substrate and subsequent heating to 150 °C for ≈15 min. Scale bar is 100 µm. b) Optical microscope image of two crystals showing the typical shape of an approximately square in-plane cross section and a (sometimes truncated) pyramidal cavity with four lateral facets on the surface. Scale bar is 20 µm. The inset displays a cross section along the white line. c) X-ray diffraction pattern of the microcrystals deposited on a zero-diffraction Si substrate together with reflections of orthorhombic CsPbBr$_3$ (ICDS number 97851). d) Crystal structure of orthorhombic CsPbBr$_3$ indicating the (101) and (010) lattice planes, with a scheme of the microcrystal structure assigning crystal planes to the different facets.
the heating process (the time for the reaction is determined by the drying process and thus by the amount of the deposited solution). Figure 1a depicts microcrystals obtained by depositing 10 µL of precursor solution with 0.3 m concentration on an ITO-coated glass substrate that was heated at 150 °C. The sample was left to dry at that temperature (which took approximately 10 min). Images of crystals obtained for different temperatures of the hot plate (for the same precursor concentration [0.3 m] and quantity [10 µL]) together with histograms of the edge lengths are reported in Figure S1, Supporting Information. We obtain larger crystals at higher temperatures, which indicate a much faster crystal growth at higher temperature that outweighs the shorter time available for the growth (due to faster drying of the deposited drop at higher temperature). Typical size distributions that we obtained by drop-cast deposition on hot substrates peak at edge lengths in between 30 and 70 µm.

Interestingly, the top surface of the microcrystals is not planar, but features a pyramidal cavity that can be pointed (right crystal in Figure 1b) or truncated and then has a square planar region at the center (left crystal in Figure 1b). The angle of the pyramidal cavity facets with respect to the horizontal surface plane varies in the range from 5° to 30° (see Figure S2, Supporting Information), which makes a crystallographic plane as the origin of the facet formation unlikely. Furthermore, in some cases we observed asymmetric cavities, as it is the case for the left crystal in Figure 1b and other crystals shown in Figure S2, Supporting Information, which makes a crystallographic plane as the origin of the facet formation unlikely. Furthermore, in some cases we observed asymmetric cavities, as it is the case for the left crystal in Figure 1b and other crystals shown in Figure S2, Supporting Information. X-ray diffraction (XRD) analysis (Figure 1c) yields an orthorhombic CsPbBr3 crystal structure with Pnma space group and unit cell parameters of $a = 8.25$ Å, $b = 11.75$ Å, and $c = 8.20$ Å, in good correspondence with the reference (ICDS 978519). Here the diffraction peaks related to the (101), (202), and (404) lattice planes are particularly dominant, indicating that the exposed (top) facet presents a (101) orientation, and thus the sides correspond to (100) and (010) planes, as outlined in Figure 1d. The structure of the microcrystal is also sketched in Figure 1d.

We have performed SEM-EDS analysis to obtain detailed information on the topography and chemical composition of the microcrystals. Figure 2a–c shows a collection of SEM images of microcrystals which all feature the pyramidal cavity. In particular, in Figure 2c the step-like morphology of the internal cavity facets can be resolved, which points to an alternating structure of the facets of the orthorhombic lattice. For such a surface, also the inclination with respect to the horizontal plane can vary from crystal to crystal and even from facet to facet (see Figure S2, Supporting Information). In literature, similar morphologies were attributed to the formation of dual phases of CsPb2Br5/CsPbBr3.[31] To clarify this point, we investigated six different regions of different microcrystals with EDS. The regions are indicated by the numbers in Figure 2b,c, and the compositional maps are shown in Figure 2d–f. We find the stoichiometric composition of 1:1:3 for Cs:Pb:Br for the internal cavity facets (1–4) and for the outer rim (6) that corresponds well to the CsPbBr3 obtained from XRD, and to a step-like surface that exposes a ratio of elements as expected from the bulk structure. The truncated center of the cavity (5) exhibits reduced Cs content, as reported in Table 1, and such Cs deficiency could indicate a Br-terminated surface.

Concerning the facets of the pyramidal cavities, we conclude that they have a step-like morphology that alternates the horizontal (001) and vertical (100) or (010) planes. To understand the formation of the caved microcrystals, we have recorded their growth in situ using an optical monoscope coupled to a high-resolution CCD camera. The Movie S1 is provided in the Supporting Information, and snapshots taken at different times are reported in Figure 3.

During the growth, we observe that the microcrystals are first formed in solution and float rapidly in the field of view of the camera, until they fall on the substrate surface (see Movie S1,
Table 1. SEM-EDS analysis performed on different regions of a representative microcrystal, as indicated in Figure 2. The red-highlighted numbers indicate the observed depletion of Cs in the center of the crystals. The small differences in signal in between regions 1–4 and region 6 are within the standard variations of EDS analysis from different spots.

| Region | Atomic weight [%] | Atomic weight ratio Cs:Pb Br:Cs Br:Pb |
|--------|-------------------|--------------------------------------|
| 1      | 18.21             | 21.08 60.71 0.9 3.3 2.9              |
| 2      | 22.58             | 20.37 57.05 1.1 2.5 2.8              |
| 3      | 21.23             | 20.64 58.13 1.0 2.7 2.8              |
| 4      | 19.38             | 20.58 60.04 0.9 3.1 2.9              |
| 5      | 14.76             | 21.54 63.7 0.7 4.3 3.0               |
| 6      | 21.36             | 20.69 57.95 1.0 2.7 2.8              |

Supporting Information). The image sequence in Figure 3 shows the growth of one selected (typical) microcrystal after it touched down on the substrate (ITO at 120 °C in this case). In the early stage, no distinct pyramidal cavity can be discerned (first two frames). Then starting from frame 4 on we observe a ring-like contrast around the microcrystal that we relate to the formation of localized precursor solution droplets around the growing microcrystals, which leads to a rapid growth, in particular at the edges, resulting in the formation of the pyramidal cavity on the top surface. Therefore, it is reasonable that the angle of the cavity facets depends on the local precursor solution volume and concentration in the last phase of the microcrystal growth, in which a local droplet bubble is formed around the growing crystal on the substrate surface (frames 7–9 in Figure 3). The optical microscope image recorded from this crystal (last frame in the bottom line in Figure 3) evidences the cuboid shape with the pyramidal cavity on the top surface.

2.2. Photoluminescence Properties

The PL recorded under above-band gap excitation from such microcrystals ensembles manifests a dominant emission peak in the green spectral range (here at 520 nm), accompanied with a red-shifted sideband, as shown in Figure 4a. Such a double peak is frequently observed in the emission from metal-halide perovskite macrocrystals, and the different emission wavelengths are typically attributed to PL from the surface region that appears at smaller wavelengths, and emission from the bulk at longer wavelengths. Such behavior has been confirmed by one-photon and two-photon PL spectroscopy (see Figure S3, Supporting Information), where one-photon excitation leads to emission from the surface region, while two-photon excitation, with wavelength below the optical band gap, penetrates the bulk and can therefore generate emission from far deeper regions in the crystal.[32,33] The origin of the different PL wavelength from these regions is attributed to different crystal structure at the surface (leading to a different band gap energy) and/or self-absorption and re-emission of the light within the crystal that leads to a red-shift.[33] Accordingly, we attribute the PL peak at 520 nm in Figure 4a to emission from the surface, and the one at 540 nm to PL from the bulk. The PL decay traces, and thus the dynamics of the emission process for both peaks are very similar, as demonstrated in Figure S4, Supporting Information. Interestingly, the onset in absorption is red-shifted with respect to the PL spectra in Figure 4a. Therefore, the absorbance is dominated by the bulk, while the PL under above-band gap excitation is dominated by the surface. Indeed, the PL measured with two-photon excitation (femtosecond pulsed laser at 800 nm wavelength) that originates from the bulk is slightly red-shifted with respect to the absorbance band edge, as it would be expected.

The faceted surface of the microcrystals provides an interesting opportunity to investigate the effects of the surface morphology on the emission properties in more detail. To this aim, we have performed µ-PL measurements on single crystals with one-photon excitation (at 350 nm), where the excitation spot had a diameter of few micrometers. For the one-photon excitation with UV light that we used here, we can estimate a penetration depth of less than 500 nm,[34] thus dominated by the crystal structure at the surface. We probed the characteristic regions of the top surface, and the PL recorded from the center...
The different facets of the perovskite microcrystals can have a significant impact on their conductive properties.\cite{16,18,21,22} For example, Leblebici et al.\cite{35} demonstrated facet-dependent heterogeneity of the conductance of the grains in MAPbI\textsubscript{3} perovskite thin films. We fabricated the CsPbBr\textsubscript{3} microcrystals on a Au-coated mica substrate that acted as the counter electrode, and investigated the spatial dependence of the current with C-AFM, with a tip bias of $-3.5$ V, under ambient conditions. For these experiments, we selected small and very regular microcrystals, as the one depicted in Figure 5a. Interestingly, most microcrystals that we investigated by AFM, thus with small lateral dimensions to be compatible with our scanning range (thus edge lengths smaller than 20 $\mu$m), show a well-defined planar rim around the edges. Such small microcrystals in the ensemble are likely formed in regions where the precursor solution retracted/evaporated earlier with respect to the majority of larger crystals, and thus the rim is less pronounced and flat. The current map that corresponds to the topography image in Figure 5a is shown in Figure 5b. We observe current from the inclined facets of the pyramidal cavities, and from edges of the crystal, but interestingly, there is no current from the planar rim region. This shows that a surface with the (101) plane is detrimental for electrical contact. We attribute this behavior to the good stability and surface termination of such planar surface, while the inclined surfaces with step-like morphology, on the contrary, should be more reactive and thus can be expected to be favorable for the formation of good electrical contact. Figure 5c correlates the surface topography and measured current along the line shown in Figure 5b, evidencing the higher conductivity of the inclined facets. Figure S6, Supporting Information, shows more C-AFM images from a small ensemble of microcrystals, confirming the observation that the facets show good conductance, while from the planar rim nearly no current is measured.

The higher wavelength one at 545 nm. The insets show optical microscope images of the microcrystal where the excitation spot can be noticed by the bright green color. c) Intensity ratio of the small wavelength (high energy [HE]) and large wavelength (low energy [LE]) peaks for the different spot positions.
2.4. CsPbBr$_3$ Microcrystal Photodetectors

We explored the functionality of the single microcrystals as photodetectors by directly contacting the top surface with a Au-coated micromanipulator, while the metalized substrate was used as the second electrode. For these experiments, we worked with microcrystals grown on ITO-coated glass and Au-coated mica (as in the C-AFM studies). The micromanipulator that had roughly 15 µm tip curvature was mechanically placed on top of the selected microcrystal as illustrated in Figure 6a (see Figure S7, Supporting Information, for a CCD camera image of a contacted crystal). Figure 6b shows typical current–voltage ($I$–$V$) curves of a microcrystal on Au in the dark and under white light illumination that reveals strong photoconductivity of the crystals (see $I$–$V$ curves of a similar microcrystal on ITO in Figure S8, Supporting Information). Generally, we observe strongly asymmetric and nonlinear IV curves with much more signal at negative bias. This asymmetry can be rationalized by a different contact formation on the top and bottom surfaces, and it is reasonable to assume that the planar interface that was formed during microcrystal growth at elevated temperatures results in a smaller contact resistance than the mechanically placed micromanipulator tip. We can therefore understand the electrical circuit as two metal-semiconductor junctions with opposite polarity connected in series (see Figure 6d,e), where the current is determined by the junction under reverse bias. When the more resistive junction (tip/microcrystal) is under reverse bias (positive voltage in Figure 6b, and scheme in Figure 6g), conduction is significantly suppressed, while when the microcrystal/substrate junction is reverse-biased (negative voltage in Figure 6b and scheme in Figure 6e), charge injection from the metal into the microcrystal becomes possible at sufficiently high voltage across the barrier. Furthermore, at high bias, the field gradient in perovskite microcrystal should lead to depletion of the majority carriers in the vicinity of the reverse-biased junction, which reduces the barrier potential. The latter can explain the continuing increase in current at high bias voltage shown in the inset of Figure S8a, Supporting Information (that is more prominent at negative bias in our measurements). Accordingly, the $I$–$V$ curves recorded with different sweep direction (depicted by the full and dashed lines for increasing and deceasing bias voltage, respectively) manifest a hysteretic behavior and two crossing points, one at negative and one at positive bias. Similar hysteresis curves have been recorded from copper-halide perovskite crystals and from Ag$_2$S nanocrystals films, and were explored for memristor functionality.[36–38] Under illumination, a similar behavior is observed, albeit with much higher currents, which points to photoconductive gain, as is indeed the case (Figure S8b,c, Supporting Information).

To explore the photodetector functionality, we either used a negative bias in the regime where the dark current is stable at fixed bias (Figure 6c), or we worked at positive bias where the hysteretic behavior is negligible, but where also the photocurrent is much lower (Figure S8b,d, Supporting Information). With ITO as transparent substrate in the visible, we could also exploit illumination from the bottom by using a reflecting sample stage in the setup, and in this case, the bottom cross section of the microcrystal (i.e., the square of the edge length) is the active area $A$ of the photodetector. Under these conditions, we obtain a highly competitive responsivity of 150–300 $\text{A W}^{-1}$ in the green–blue spectral range (Figure 6c),[6,40] with a band edge peak at around 540 nm, and hysteresis in the spectral...
range above the peak, from 520 to 540 nm. Interestingly, the band edge peak is much more pronounced and sharp under positive bias, as depicted in Figure S8, Supporting Information, and therefore enables narrow-band photodetection with a full-width-at-half-maximum of less than 20 nm and maximum responsivity of 0.7 A W$^{-1}$. This band edge peak in photoconductivity roughly coincides with the absorption band edge and the PL observed with two-photon excitation (Figure 4a), which is reasonable for the vertical electrode configuration that we used here. The high responsivity for wavelengths smaller than 500 nm under negative bias could be related to efficient harvesting of photogenerated high above the band gap at the substrate/microcrystal interface. Concerning the other figures of merit (FOM), we estimate the spectral noise density as the sum of the thermal and shot noise, and the response under negative bias obtain a noise-equivalent power NEP = 10$^{-14}$ W$\sqrt{\text{s}}$ (see Supporting Information for details and values under positive bias) and a detectivity $D^*$ = $\sqrt{A}$/NEP = 10$^{11}$ Jones in the green to blue spectral region, where A is the area of the photodetector. These FOM are highly competitive with perovskite-based photodetectors reported in literature, especially considering the simple contact fabrication and vertical device architecture, as well as with respect to the narrow detection band that can be achieved at positive bias.[6,40] The light on/off response is reported in Figure S9, Supporting Information, with rise and fall times of around 100 and 20 ms, respectively.

3. Conclusion

We reported the facile fabrication of regular CsPbBr$_3$ microcrystals with a fast (few minutes) crystal growth process at relatively low temperatures (80–150 °C). The crystals have a cubic shape with lateral edge lengths ranging from 10 to 100 µm, and height from 1 to 30 µm. The size of the crystals can be tuned by the reaction temperature, solution concentration, and reaction time. The fast growth process on a substrate leads to a pyramidal cavity on their top surface that exposes different crystal facets with different optical and conductive properties. In particular, the emission is strongest from the center of the cavity, and the step-like pyramidal facets show stronger conductance as compared to planar horizontal crystal facets. This heterogeneity is particularly interesting toward the integration of lead-halide microcrystals in optoelectronic devices such as solar cells, light-emitting diodes (LEDs), and photodetectors. Concerning photodetectors, we obtain excellent responsivity in the green–blue spectral of several hundred A W$^{-1}$ when measuring in forward bias, and narrowband detection at 540 nm.
4. Experimental Section

Synthesis of the Precursor Solution and Microcrystal Fabrication: The precursor solution was prepared by adding 127.68 mg of CsBr and 220.2 mg of PbBr₂ salts to 2 mL of DMSO and heated to 70 °C for 10 min. The solution was then cooled to room temperature and stirred for 24 h. For the microcrystal fabrication, the precursor solution was deposited by drop-casting with a micropipette on planar ITO, glass, or gold-coated mica substrates that were either placed on a hot plate or into an oven set to the desired temperature in the range from 80 to 150 °C. The substrate was removed after the solvent was evaporated and the dry crystal deposit was formed. All chemicals were purchased from Sigma Aldrich.

X-Ray Diffraction: XRD measurements were carried out on PANalytical Empyrean X-ray diffractometer equipped with a 1.8 kW Cu Kα ceramic X-ray tube and a PixiFlex[3] 2 × 2 area detector, operating at 45 kV and 40 mA. The diffraction pattern was collected under ambient conditions using a parallel beam geometry and symmetric reflection mode. HighScore 4.1 software from PANalytical was used to analyze the XRD data. Samples were prepared by drop-casting 2.5 µL precursor solution at 0.3 m concentration onto a zero-diffraction silicon substrate that was placed on a hot plate at 150 °C.

Optical Microscopy Analysis: Optical images and 3D profilometry measurements were obtained with a confocal Zeta-20 profilometer (Zeta instruments, USA) and with an Olympus microscope (BX10). The movies were recorded with a CCD camera from Thorlabs connected to an optical mono-zoom (12×) at maximum magnification (24×).

Optical Spectroscopy: Steady-state and time-resolved PL experiments were performed on macroscale ensembles of microcrystals with an Edinburgh FLS 920 spectrophotometer that includes a time-correlated single photon counting (TCSPC) module and a pulsed laser diode at 375 nm wavelength. The optical spot diameter was 2 mm.

For micro-PL measurements on single crystals, the authors used a home-built optical setup with a nanosecond pulsed laser at 350 nm wavelength (Spectra-Physics) and a UV extended 20× objective (Olympus) with NA = 0.5. The sample was placed on a piezo stage from PI, imaged with a CCD camera from Thorlabs, and the emitted light was detected through the same objective with an Ocean Optics HR4000 spectrometer. A dichroic beam splitter reflecting in the range from 325 to 375 nm and transparent in the range from 415 to 1600 nm was placed before the objective to separate illumination and detection signals.

Absorbance measurements were performed on Edinburgh Instruments (FLS920) fluorescence spectrometer equipped with a calibrated integrating sphere and operating in reflection geometry. The collected reflectance/transmittance setup was converted to an absorbance spectrum through the inbuilt function in the instrument. The absorbance of a single microcrystal was investigated through a customized micro-reflectance/transmittance setup in the wavelength range between 450 and 900 nm. A tungsten-halogen light source (Spectral Products ASBN-W) was used to illuminate the sample, while the transmitted signal was focused into a fiber-optic spectrometer (Ocean Optics USB4000-VIS-NIR) by a 20× microscope objective (Nikon Plan Achromat, NA = 0.4).

Energy Dispersive Spectroscopy: The SEM analysis was performed using a HRSEM JEOL JSM-7500LA (JEOL, Tokyo, Japan) equipped with a cold field-emission gun, operating at 25 kV acceleration voltage. No additional conductive coating was needed. EDS (Oxford Instrument, X-Max, 80 mm²) operating at 25 kV was utilized to distinguish the presence of Pb, Br, and Cs. The Br K-edge and Cs and Pb L-edges were used for the EDS quantification.

Conductive Atomic Force Microscopy: The C-AFM measurements were performed with a XE 100 (Park Systems, Korea) microscope under ambient atmosphere, using 25P300B probe tips from Rocky Mountain Nanotechonologies (USA). The gold film on the mica substrate acted as the sample electrode, and the voltage was applied to the tip.

(Phot-) Electrical Characterization: Current–voltage curves were recorded with a micromanipulator probe station (Suss, Germany), using a Keithley 2612 source meter, and the microcrystal top surface and the gold-coated substrate were contacted with gold-coated manipulators having a tip radius of 10 µm. White light came from a microscope lamp with black body radiation at 3200 K. For the spectral photocurrent measurements, a fiber coupled supercontinuum laser from NKT (EX-4) with an acousto-optic coupler working in the range from 480 to 650 nm was used as a light source, and focused to a spot of a few mm diameter on the sample with a lens with 10 cm focal length. The reference spectrum was recorded with a calibrated DET 210 photodetector from Thorlabs.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

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