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UV-assisted rapid synthesis of high quality CsPbBr₃ perovskite single crystals

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Abstract

Inorganic halide perovskite single crystals have garnered significant interest due to their outstanding optical and electrical properties, making them strong candidates for a wide range of optoelectronic applications. However, conventional crystal growth techniques, such as the Bridgman, antisolvent, and inverse temperature crystallization (ITC) methods, often require complex procedures, high temperatures, and prolonged processing times. Here, we present a straightforward and efficient synthesis method for high-quality CsPbBr₃ perovskite single crystals based on ultraviolet (UV) light irradiation. This method employs UV light to provide the energy required for the CsPbBr₃ crystallization process, which includes molecularization, ionization, and nucleation. The single crystals synthesized using the UV method (UV-grown) exhibit an average carrier lifetime of 17 ns (twice that of ITC-grown counterparts) and a hole mobility of 197 cm²·V⁻¹·s⁻¹ (a six-fold increase over ITC-grown counterparts). Our method not only provides enhanced optoelectronic properties but also simplifies the crystal synthesis process. This advancement paves the way for the scalable production of large-size CsPbBr₃ single crystals for in-depth fundamental studies and a wide range of optoelectronic applications.



KEYWORDS

Inorganic halide perovskite single crystals, photoionization, Optoelectronic applications, Ultraviolet (UV) light irradiation, Crystallization process, Carrier lifetime.

Introduction

Inorganic halide perovskites, particularly CsPbX_3 ($X = \text{Cl}, \text{Br}, \text{and I}$), have gained attention for their superior stability, narrower emission bandwidth, and tunable photoluminescence (PL) wavelength in comparison to organic-inorganic hybrid perovskites^{1–4}. These properties make them ideal for a variety of optoelectronic applications^{5–8}. Although various synthesis methods for CsPbX_3 , including the Bridgman, antisolvent, and inverse temperature crystallization (ITC) methods, have been successfully demonstrated for low defect CsPbBr_3 bulk single crystals, each approach has its own limitations^{9,10}. The conventional Bridgman method is costly and requires sophisticated equipment and high temperatures, while the antisolvent method is time-consuming and demands precise stoichiometry. Although the ITC method significantly reduces synthesis time, it still requires hours or days of regrowth stages to produce large-sized crystals^{11,12}. Therefore, a strategy for the rapid synthesis of high-quality, scalable perovskite single crystals is still necessary.

Recently, interaction between light and the perovskite crystals has emerged as a promising method of enhancing optoelectronic properties of perovskites. UV irradiation has been shown to induce significant changes in perovskite materials, including phase transformation¹³, photo-degradation^{14,15}, and morphology modification¹⁶ in both inorganic and hybrid colloidal 3D nanocrystals and 2D perovskite nano-plates. For instance, UV light exposure can trigger structural phase transition in CsPbBr_3 nanocrystals, leading to changes in their optical and electronic properties¹⁷. These light-induced modifications demonstrate the profound impact of UV



irradiation on perovskite materials at the nanoscale. Studies have revealed that CsPbBr₃ nanocrystals experience a reversible orthorhombic-to-cubic phase transition at moderate excitation light energy and become amorphous at higher light energy, demonstrating that light-driven phase transitions occur in perovskite materials¹⁸. Moreover, a photon-induced reversible phase transition from orthorhombic to tetragonal in CsPbBr₃ perovskite is reported, driven by Pb-Br octahedral torsion and local Coulombic field, enabling fast and controllable response to light¹⁹. Light-induced lattice expansion plays a crucial role in enhancing the efficiency of perovskite solar cells as it was demonstrated that continuous light illumination induces a uniform lattice expansion in hybrid perovskite thin films, leading to improved device performance^{20,21}. Additionally, surface layers of lead halide perovskite crystals exhibit high concentrations of positively charged vacancies and negatively charged halide ions, creating an electric field that increases the optical band gap near the surface after photoexcitation²². Light excitation also enhances the ionic conductivity of methylammonium lead iodide by several orders of magnitude, a key metal halide photovoltaic material, leading to a previously unexplored pathway for photodecomposition of the perovskite²³. Furthermore, light has been shown to prompt the self-assembly of cubic CsPbBr₃ perovskite nanocrystals into nanowires, offering an efficient alternative for the synthesis of nanowires and other perovskite superstructures²⁴. It has also been observed that exposing pre-synthesized nanocrystals to UV irradiation induces an increase in the size of the nanocrystals multiple fold^{25–27}. However, the majority of studies have focused on nanocrystals, with relatively limited exploration into the use of UV irradiation for the growth of bulk single crystals.

In this study, we introduce a novel, straightforward, and rapid synthesis method for high-quality bulk CsPbBr₃ inorganic perovskites single crystals using UV light. To the best of our knowledge, this is the first report of utilizing UV light exposure to induce the molecularization, ionization,



nucleation, and crystallization processes for the synthesis of bulk inorganic halide perovskite single crystals. The UV-assisted synthesis method leverages the UV light energy to drive the formative processes of the CsPbBr₃ perovskite precursor, leading to the growth of high-quality crystals. The resulting crystals exhibited rectangular-shaped millimeter-sized morphology with an average length of 3 mm and a maximum of 5 mm achieved within 90 minutes. The optical and electronic characteristics of the UV-grown crystals were investigated using various techniques including X-ray diffraction, UV-Vis absorption, photoluminescence, time-resolved lifetime decay measurements, Hall effect measurement, and Raman spectroscopy. Compared to the ITC-grown crystals, which had an average lifetime of 8.3 ns and a carrier mobility of 30 cm²·V⁻¹·s⁻¹, the UV-grown perovskite crystals demonstrated higher photoluminescence efficiency, a carrier lifetime of 17 ns, and a significantly higher carrier mobility of 197 cm²·V⁻¹·s⁻¹. Therefore, this study presents a compelling contribution to the field of inorganic halide perovskites, and is expected to inspire further research efforts in this direction.

Materials and methods

Cesium bromide (CsBr), Lead (II) bromide (PbBr₂), Methylammonium bromide (MABr), Dimethylformamide (DMF) and Dimethyl sulfoxide (DMSO) were purchased from sigma-Aldrich and used without further purification.

CsPbBr₃ Single Crystal Synthesis

Crystal synthesis was performed in two steps. The first step involved the preparation of the precursor solution, while the second step involved the UV irradiation of the precursor solution. In the first step, CsBr and PbBr₂ compounds were weighed in a molar ratio of 1 to 2 and dissolved in DMSO solvent. The solution was stirred for 3 hours at room temperature to obtain a uniform and



colorless solution. The solution was then filtered using a 0.2 μm Millipore filter to obtain a purified precursor solution. In the second step, 500 μL of the purified precursor solution was exposed to UV light for 1.5 hours to allow photoionization, nucleation, and crystal growth to occur. The irradiation was performed using a UVC-30s UV lamp (Jaesung Engineering CO.) with the following specifications: wavelength of 254 nm, power density of $25 \text{ mW}\cdot\text{cm}^{-2}$. The UV lamp was positioned directly above the uncovered precursor solution in a glass vial, ensuring uniform illumination across the sample surface. The resulting crystals were collected, cleaned with DMF, and dried.

ITC CsPbBr_3 Single Crystal Synthesis

The ITC crystal growth was initiated by placing 500 μL of the precursor solution in a vial and heating it to 85°C in an oil bath similar to earlier reports^{28,29}. The vial was kept in the oil bath for 4 hours to generate crystal seeds. The crystal seeds were then regrown for 16 hours, cleaned with DMF, and dried.

MAPbBr₃ Single Crystal Synthesis

A solution containing equimolar concentrations of MABr and PbBr_2 was prepared by dissolving them in DMF and stirring the mixture at room temperature for 3 hours. Following dissolution, the solution underwent filtration to obtain pure solutions. Subsequently, equal volumes of these solutions were subjected to both UV irradiation and ITC growth method, mirroring the procedures previously employed for CsPbBr_3 .

Characterization.



Powder XRD measurement was carried out using Panalytical (EMPYREAN), with X-ray generator: 4 kW, a vertical goniometer with scan range: $1.5 < 2\theta < 140$, and sample stage (Reflection-Transmission Spinner).

Steady-state absorption and photoluminescence spectra were measured using a UV-Vis spectrometer (Perkin Elmer Lambda 950) and a fluorescence spectrometer (Horiba FluoroMax-4), respectively

Photoluminescence quantum yield (PLQY) measurements were performed using an integrating sphere setup coupled to the fluorescence spectrometer. The absolute PLQY was calculated by comparing the integrated emission intensity to the integrated absorption intensity.

Time-resolved photoluminescence (TRPL) measurement study was carried out using a confocal microscope (MicroTime-200, Picoquant, Germany) with a 10× (air) objective. For the lifetime measurements, a single-mode pulsed diode laser (470 nm with a pulse width of ~30 ps and an average power of 30~100 nW operating in 1 MHz repetition rate) was used as an excitation source. A dichroic mirror (490 DCXR, AHF), a long-pass filter (HQ500lp, AHF), a 150 μm pinhole, a bandpass filter (500~600 nm, Thorlabs), and a single-photon avalanche diode (PDM series, MPD) were used to collect emission from the samples. A time-correlated single-photon counting system (PicoHarp-300, PicoQuant GmbH) was used to count emission photons. Exponential function fittings for the obtained PL decays were performed using Symphotime-64 software (Ver. 2.2).

The Hall mobility and carrier trap density measurements were carried out with the Ecopia HMS3000 system (magnetic field of 0.51 T), using the Van der Pauw Hall effect measurement technique at room temperature.



Discussion

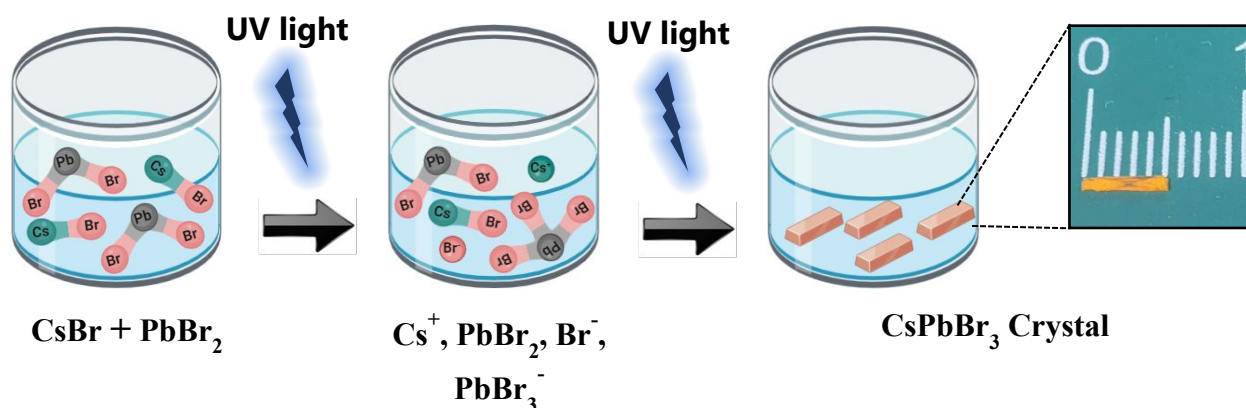
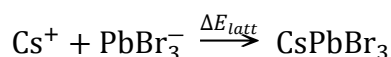
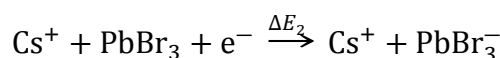
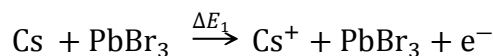
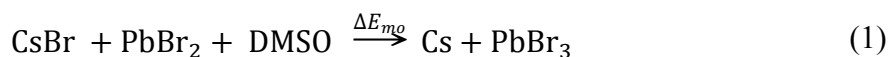


Figure 1. Schematic illustration of chemical processes for the UV-assisted synthesis method of CsPbBr_3 crystal.

The synthesis of CsPbBr_3 single crystals using the UV irradiation technique is illustrated in **Figure 1**. The UV light was directly illuminated onto the uncovered precursor in the preparatory bowl. When molecules absorb a UV light energy greater than its ionization energy, a photoionization process occurs, releasing electron^{30,31}. The ionization energies of the Cs^+ and PbBr_3^- ions in the precursor are reported to be 3.85 eV and 4.31 eV, respectively, which fall within the range of our UV source energy (4.88 eV)^{32,33}. The DMSO serves as a coordinating solvent that modulates the precursor solubility, facilitates ion migration, and contributes to defect passivation. Its interaction with PbBr_2 helps stabilize intermediate complexes that are crucial for uniform nucleation under UV irradiation. Clusters consisting of Cs are particularly susceptible to ionization due to their low ionization energy compared to other elements³⁴. The photoionization reaction process can be represented using the Born-Haber cycle. The Born-Haber cycle has traditionally been employed for analyzing formation enthalpies. In this case, it is utilized for analyzing the reaction enthalpy components of the UV-grown CsPbBr_3 crystal^{35–37}. The formation process of CsPbBr_3 from solid CsBr and PbBr_2 compounds can be subdivided into several consecutive steps, as illustrated in



Equation 1. The initial step is the molecularization of the CsBr denoted with ΔE_{mo} , which involves breaking the CsBr and PbBr₂ lattice structures and formation of Cs and PbBr₃ molecules. The next



step is the ionization of the Cs molecule (ΔE_1 is Cs⁺ ionization energy), followed by the ionization of PbBr₃ (ΔE_2 is PbBr₃⁻ ionization energy). Finally, the amount of energy to form the inorganic perovskite structure is denoted as the lattice energy (ΔE_{latt}).

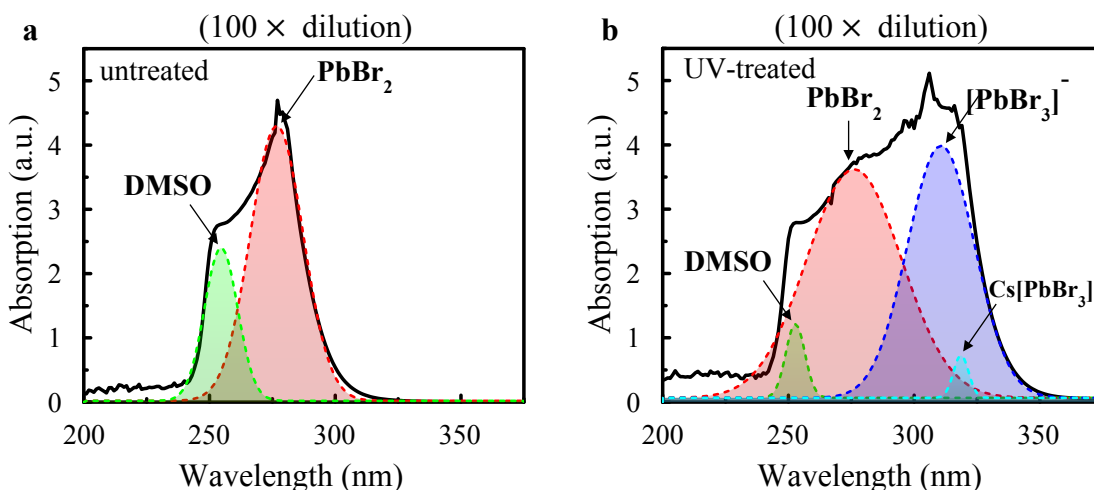


Figure 2. Absorption spectra of CsPbBr₃ Precursor **a.** non-treated with spectra range from 240nm to 310nm and **b.** treated with UV with spectra range from 240nm to 350nm.

To further validate the effect of UV photoionization, two precursor solutions of 0.1 molar solution of CsPbBr₃ precursor, which is 100 times dilution, was prepared. One solution was exposed to UV light (254 nm LED, 5 mW·cm⁻² power) treatment for 3 hours, while the other solution was not exposed to UV light. The optical properties of CsPbBr₃ perovskite precursor solutions were investigated for the UV treated and non-treated samples. **Figure 2a** shows the absorption spectrum



of the untreated precursor solution, spanning from 240 to 310 nm (black line), while the absorption spectrum of the UV-treated CsPbBr₃ precursor solution shown in **Figure 2b** exhibits a broader range from 240 nm to 350 nm (black line). Both untreated and UV-treated solutions displayed an absorption peak around 285 nm, corresponding to the absorption spectrum of PbBr₂ while an additional absorption peaks at approximately 310 nm and 322 nm in the UV-treated solution are attributed to the formation of PbBr₃⁻ ions and Cs[PbBr₃] complex, respectively^{38–40}. The exposure of the CsPbBr₃ precursor to UV facilitated the molecularization, ionization, nucleation and rapid lattice-growth rate of the CsPbBr₃ bulk single crystals. This method produces crystals with fewer defects, yielding higher intensity of the second-order phonon peak in the Raman spectrum of UV-grown CsPbBr₃ single crystals as shown in **Figure S1** in supplementary information^{41,42}. These findings provide valuable insights into the mechanism of UV-grown CsPbBr₃ single crystals. A

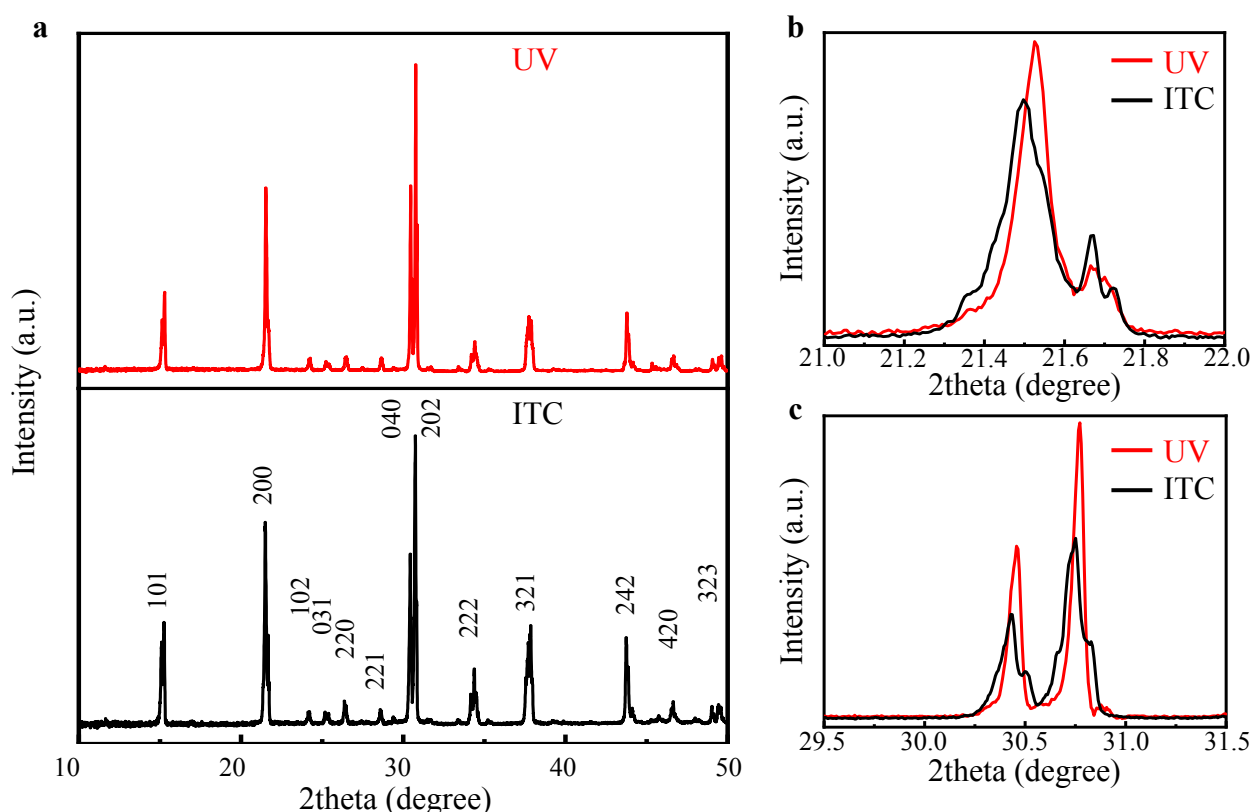


Figure 3 a. XRD plot for UV- and ITC-grown CsPbBr₃ and **b.** enlarged XRD pattern of the peaks with higher intensity and narrower width at 21.0°–22.0° and **c.** 29.5°–31.5°.



thermocouple inserted into the precursor was used to measure its temperature during crystal growth under UV irradiation. As shown in **Figure S3**, the precursor reached a maximum temperature of 55 °C during the entire growth process, which is lower than the temperature required for ITC, at least 85 °C¹¹. This confirms that UV irradiation, rather than thermal effects, is the primary driver of crystallization. The structural characteristics of the CsPbBr₃ single crystals grown under different conditions were investigated using X-ray diffraction (XRD) analysis. **Figure 3a** presents the XRD patterns of CsPbBr₃ crystals grown using the UV-assisted and ITC methods. The XRD patterns of the two growth conditions exhibited similar diffraction peaks, consistent with previously reported works on CsPbBr₃ single crystals^{28,29}. The major diffraction peaks observed for the UV-grown crystals were located at 15.27°, 21.55°, 30.45°, 30.76°, 37.90°, and 43.78°, which can be indexed to the (101), (121), (040), (202), (123), and (242) planes of the orthorhombic CsPbBr₃ crystal structure (JCPDS No. 96-451-0746), respectively. Notably, the UV-grown crystals exhibited diffraction peaks with markedly higher intensity and significantly narrower width at the signature peaks, corroborating findings from previous studies⁴³. Interestingly, the high-resolution XRD data revealed systematic shifts in the peak positions between the ITC-grown and UV-grown CsPbBr₃ samples. For instance, the (101) peak occurred at $2\theta \approx 15.253^\circ$ for the ITC-grown sample, but shifted to $2\theta \approx 15.274^\circ$ for the UV-grown samples (**Figure S4a**). Similar trends were observed for other characteristic peaks, such as the (121) peak shifting from $2\theta \approx 21.497^\circ$ in the ITC-grown sample to $2\theta \approx 21.528^\circ$ in the UV-grown sample (**Figure S4b**), and the (040) peak position changing from $2\theta \approx 30.432^\circ$ for the ITC-grown sample to $2\theta \approx 30.458^\circ$ for the UV-grown sample (**Figure S4c**). These systematic peak shifts indicate that the UV-grown CsPbBr₃ crystals exhibit a compressively strained lattice structure compared to the ITC-grown samples, resulting from



differences in growth conditions and associated lattice distortions. These findings highlight the structural advantages of the UV-assisted synthesis method, which leads to improved material quality compared to the ITC-grown counterparts. Understanding the origins of these structural variations and their implications for the optoelectronic properties of the materials could provide valuable insights for optimizing the performance of CsPbBr₃ perovskite crystals.

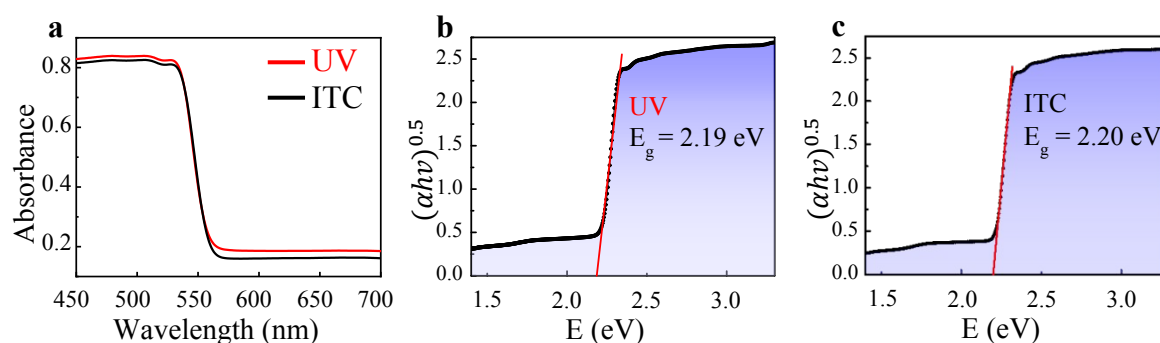


Figure 4 a. Absorbance curve of UV- and ITC-grown CsPbBr₃. Energy band gap from Tauc plot of **b.** UV-grown CsPbBr₃ **c.** ITC-grown CsPbBr₃.

The optical properties of the synthesized CsPbBr₃ crystals were investigated using UV-Vis absorption spectroscopy. The synthesized crystals exhibited a sharp absorption edge at 565 nm, which indicates a highly crystalline structure (**Figure 4a**). The optical band gap of the crystals was estimated using Tauc plots from the UV-Vis absorbance spectra. The band gaps of the UV- and ITC-grown CsPbBr₃ crystals were 2.19 and 2.20 eV, respectively, as shown in **Figures 4b and 4c**. These values are in good agreement with the theoretical values and are consistent with previously reported work⁴⁴. The UV-grown crystal demonstrated a higher absorption compared to the ITC-grown crystal, which could be attributed to its improved crystal quality. These findings indicate that the UV photoionization method offers a favorable approach for synthesizing high-quality CsPbBr₃ crystals.



Figure 5a shows the steady-state photoluminescence (PL) spectra of CsPbBr₃ crystals grown by both the UV and ITC methods. The PL spectra for both samples exhibited a peak at 528 nm, consistent with previously reported value⁴⁵. The observed divergence between the absorption onset at 565 nm and the PL peak at 528 nm can be rationalized by considering the influence of defect-mediated absorption tails and exciton-phonon interaction. Such spectral offsets have been previously reported in CsPbBr₃ crystals, where the apparent red-shift in the absorption edge does not correspond to the bandgap^{46,47}. Liu et al. demonstrated that the strong exciton-phonon coupling leads to absorption broadening while PL remains centered at the band-edge emission energy⁴⁸. Similarly, Falsini et al. attributed the extended absorption tails to the dynamic disorder and Urbach tail formation, whereas the PL emission reflected direct band-to-band transition⁴⁹. In addition, Su et al. showed that CsPbBr₃ crystals grown by anti-solvent precipitation exhibit morphology-dependent optical responses, further indicating that synthesis methods influence defect states and excitonic behavior⁵⁰. Collectively, these findings support our assertion that the PL peak accurately represents band-edge recombination, while the absorption edge includes sub-bandgap contribution from the tail states. Furthermore, **Figure 5b** shows the TRPL measurements were conducted to calculate the carrier recombination lifetime of the CsPbBr₃ crystals by fitting the curve with an



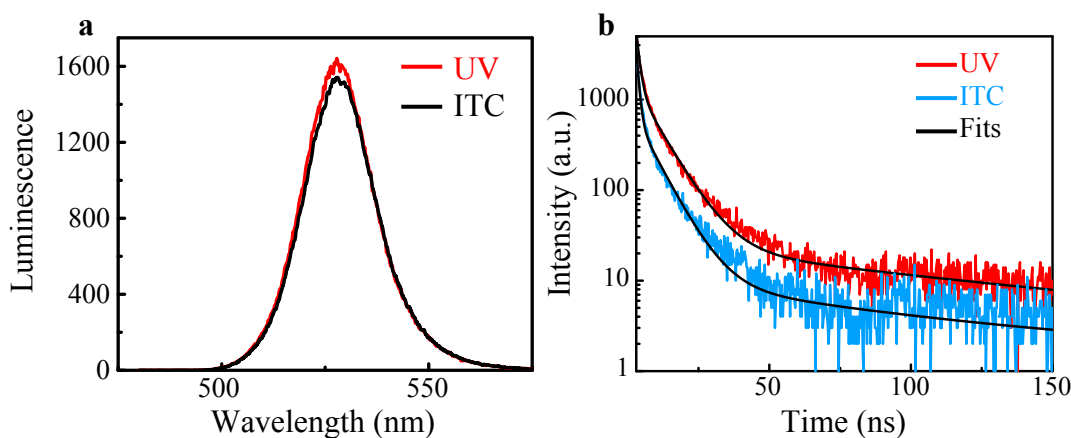


Figure 5 a. Photoluminescence spectra and b. carrier lifetime decays of UV- and ITC-grown CsPbBr₃.

exponential time (τ_{ave})⁵¹ function in equation (2), where τ_i represents the carrier lifetime component and A_i represents the amplitude of the corresponding carrier lifetime. The average decay time (τ_{ave}) reflects the recombination dynamics of carriers in the perovskite materials.

$$\langle \tau_{ave} \rangle = \frac{\sum_i A_i \tau_i^2}{\sum_i A_i \tau_i} \quad (2)$$

The parameters of the triexponential function fitted to time-resolved photoluminescence spectra are listed in **Table S2** in supplementary information. In comparison to the average lifetime of 8.3 ns for ITC-grown crystal, the UV-grown crystal exhibits a noticeably longer carrier lifetime decay of 17 ns at an excitation wavelength of 470 nm. The compact lattice structure, as evidenced by the angular shifts in the XRD data, reduces the density of defects and trap states, thereby suppressing non-radiative recombination and extending the carrier lifetime. This result shows that UV-grown crystals not only acquire higher crystallinity and lower defect density but also suppressed nonradiative recombination near crystal surface, which will result in enhanced optoelectronic performances⁵².



Finally, the electrical properties of UV- and ITC-grown crystals are compared in **Table 1**, along with the results from previous studies. The dominant charge carriers were found to be hole in both crystals. The trap density was significantly lower in the UV-grown crystal compared to the ITC-grown crystal, indicating fewer defects that could hinder hole mobility and conductivity. Additionally, the hole mobility was significantly higher in the UV-grown ($197 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$) crystal than the ITC-grown crystal ($30 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$). This suggests that the UV-grown crystal provide a more favorable transport environment with reduced charge scattering caused by defects. The synergistic effect of reduced lattice strain, due to reduced interplanar spacing, and a more compact lattice structure in the UV-grown CsPbBr_3 crystals may be responsible for the simultaneous enhancement in carrier mobility and lifetime, key metrics for high-performance optoelectronic devices. The findings also highlight the importance of the growth method in determining the quality of semiconductor crystals and their suitability for various electronic applications. PLQY measurements conducted under uniform excitation conditions revealed that UV-grown CsPbBr_3 crystals exhibited a superior PLQY of 0.1%, markedly higher than the 0.07% observed in samples synthesized via ITC. The enhanced PL performance is indicative of suppressed non-radiative recombination pathways, attributed to reduced intrinsic defect densities and minimized deep level trap states, as substantiated by time-resolved photoluminescence spectroscopy. Extended carrier lifetime observed in the UV-grown samples further validate the efficacy of UV irradiation in mitigating defect formation during crystal nucleation and growth. These findings underscore the critical role of photonic treatment in engineering high-purity CsPbBr_3 crystals for advanced optoelectronic applications. The intrinsic properties of the UV-grown crystals, such as high crystallinity, sharp photoluminescence emission, and low trap density, which are directly linked to



their suitability for photodetector and light-emitting applications. These characteristics align well with the performance metrics reported by Gupta et al.¹⁰, where similar CsPbBr₃ crystals were successfully integrated into photodetector devices.

The UV-assisted synthesis method was applied for the growth of MAPbBr₃ single crystals. However, the photoluminescence and lifetime measurements, detailed in **Figure S5** of the supplementary information, revealed a noticeable degradation in the lifetime performance of UV-grown crystals when compared to their counterparts grown through ITC method. The diminished performance observed in the UV-grown crystals can be attributed to the degradation of the organic component within the methylammonium (MA) site induced by UV irradiation^{53–55}.

Table 1. Comparison of optoelectronic parameters and photoluminescent carrier lifetime of CsPbBr₃ crystals grown with various techniques.

Crystal growth method	Lifetime (ns)	Trap density (cm ⁻³)	Mobility (cm ² · V ⁻¹ · s ⁻¹)	Charge Type	Ref.
ITC ^a	233	1.1 × 10 ¹⁰	52	electron	44
Bridgeman	10.5	1.9 × 10 ⁹	2300	electron	56
Bridgeman	10.9	6.36 × 10 ⁹	11.6	electron	57
AVC ^b	6.8	2.8 × 10 ¹⁰	13.6	---	58
AVC ^b	---	4.5 × 10 ⁷	143	hole	59
ITC ^a	6.2	7.1 × 10 ¹⁰	28.5	electron	28

LTC ^c	16.8	7.1×10^{10}	160	electron	28
UV-photoionization	17	1.46×10^9	197	hole	This work
ITC ^a	8.3	9.63×10^9	30	hole	This work

^aInverse temperature crystallization method

^bAntisolvent vapor-assisted method

^cLow-temperature crystallization method

Conclusion

In conclusion, this study demonstrated the novel growth method of high-quality CsPbBr₃ perovskite single crystals using UV irradiation. The UV-driven method facilitates the molecularization, ionization, nucleation, and lattice growth of CsPbBr₃ bulk single crystals, resulting in a high hole mobility of 197 cm² V⁻¹ s⁻¹ and extended carrier lifetime of 17 ns. These findings provide significant insights into the fundamental understanding of perovskite single crystals and represent an important step forward in advancing all-inorganic halide perovskite materials for optoelectronic applications. Further research on the development of thin films with this method could lead to the emergence of novel perovskite-based devices with improved performance and stability, thereby enabling the realization of next-generation optoelectronic technologies.

Associated Content



Supplementary Information is available.

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Conceptualization: J.P, M.A.A, A.M.O, Methodology: J.P, M.A.A, A.M.O, Supervision: J.P, funding acquisition: J.P. Experiment and measurement: M.A.A, A.M.O, H.L, G.K. Data analysis and investigation: J.P, M.A.A, A.M.O, and H.K. The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

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Conflict of Interest.

The authors declare no conflict of interest.

Data Availability Statements.

The data supporting this article have been included as part of the Supplementary Information.

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Data availability statements

The data supporting this article have been included as part of the Supplementary Information

