

Supporting Information

In-situ synthesis of blue-emitting bromide-based perovskite nanoplatelets towards unity quantum efficiency and ultrahigh stability

Hao Huang, Weiren Zhao*, Huanxin Yang, Xiaoli Zhang, Jinnan Su, Kaige Hu,
Zhaogang Nie, Yang Li, Jiyou Zhong

Chemicals: Lead bromide (PbBr_2 , 99%), cesium bromide (CsBr , 99.5%), oleylamine (OAm, 90%), (3-Aminopropyl)triethoxysilane (APTES, 99%), hydrobromic acid (HBr, 40 wt% in H_2O) were purchased from Macklin. n-Hexane (AR) and *N,N*-Dimethylformamide (DMF, AR) were purchased from Tianjin zhiyuan Chemical Reagent Co. Ltd (Tianjin, China). All chemicals were directly used without any further purification.

Synthesis of cross-linking CsPbBr_3 nanoplatelets: First of all, Pb-precursor and Cs-precursor were pre-prepared, respectively. The yellowish Pb-precursor was prepared by mixing PbBr_2 (1mmol 367 mg), DMF (2ml) and OAm (2ml) with continuous stirring for 10 minutes, while Cs-precursor was obtained by solving CsBr in HBr (1 mmol/mL). Then 0.2 ml Pb-precursor and 0.05 ml Cs-precursor were successively injected into 5 ml n-Hexane with different amount of APTES (0.05 – 0.15 ml) under continuous stirring for 1 minute, and the solution becomes turbid immediately. The crude solution was centrifuged at 2000 rpm for 2 minutes, excess reactants and DMF at the bottom of tube were discarded, while the supernatant was collected for further analysis.

Measurements and Characterizations: XRD patterns were obtained using a Bruker D8 Advance X-ray diffractometer with a $\text{Cu K}\alpha$ source. PL spectra were acquired using an Ocean Optics spectrometer (QE 65 Pro) equipped a fiber probe and 395 nm laser light source. The PL decay in solution was recorded with time-correlated single-photon counting (TCSPC) technique on an Edinburgh FLS920 phosphorescence lifetime system equipped with a 320 nm LED light source at room temperature. Absorbance spectra were measured with a UV-3600 plus spectrophotometer. TEM measurements were carried out on a FEI Talos F200S with a Cu grid. SEM measurements were performed using a field-emission scanning electron microscope SU8220. FTIR spectra were measured on a Nicolet IS50 in the region of $4000\text{-}650\text{ cm}^{-1}$. XPS measurements were performed on an Escalab 250Xi photoelectron spectrometer. The absolute PLQYs were determined using a quantum yield measurement system QE

2100 equipped an integration hemisphere. Temperature-dependence of PL and delay measurements were recorded using a He flow cryostation (Montana Instruments C2) with 405 nm laser light source.

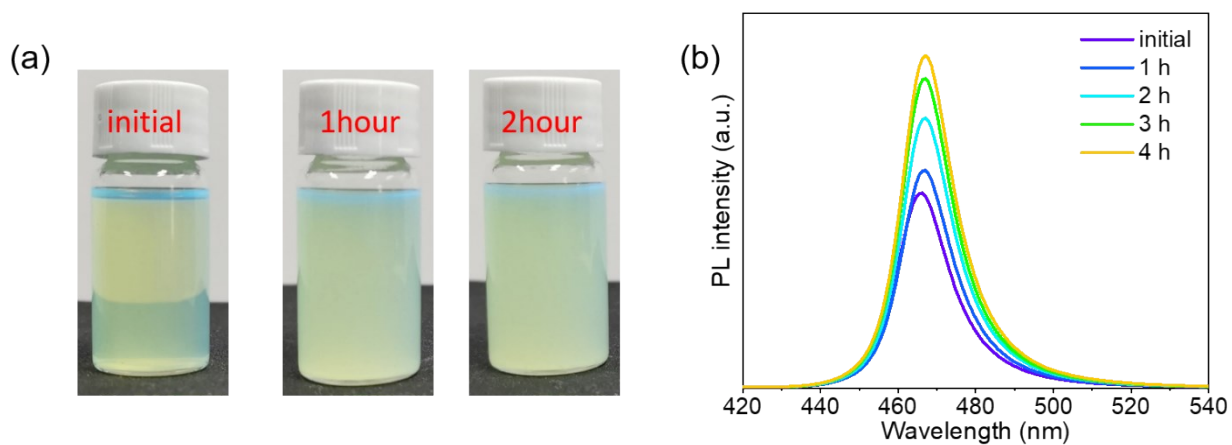


Figure. S1 (a) The images of the state change and (b) the revolution of PL intensity of fresh-prepared APTES-NPLs in the first two hours. (b) of fresh-prepared APTES-NPLs in the first four hours.

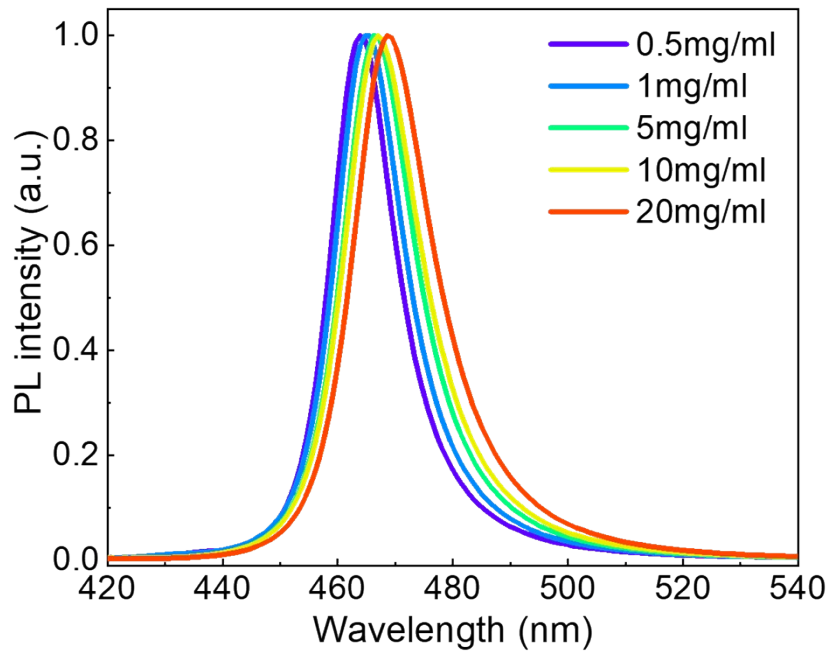


Figure. S2 PL curves of APTES-NPLs (in solution) at different concentration.

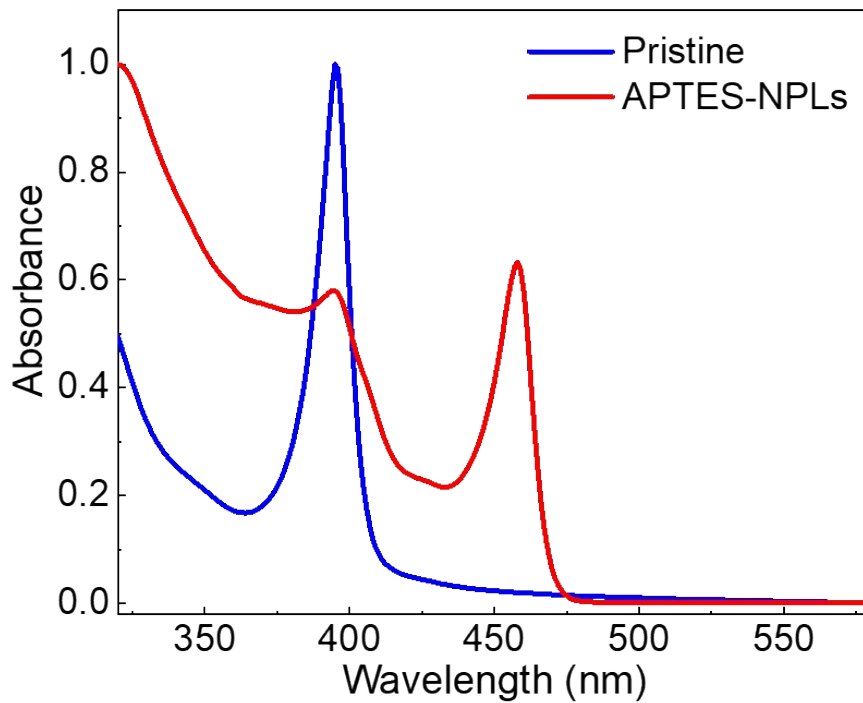


Figure. S3 Absorption spectra of pristine and APTES-NPLs after storage overnight.

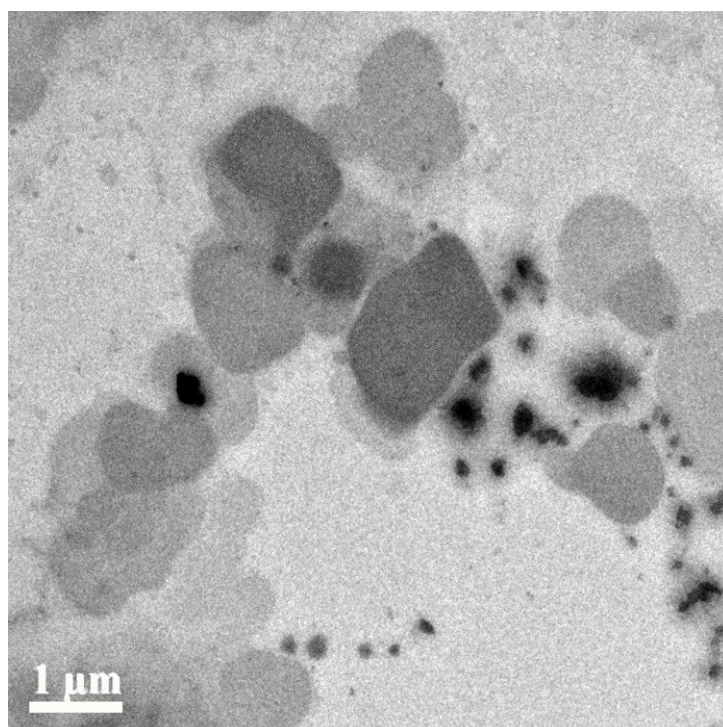


Figure. S4 TEM images of pristine NPLs.

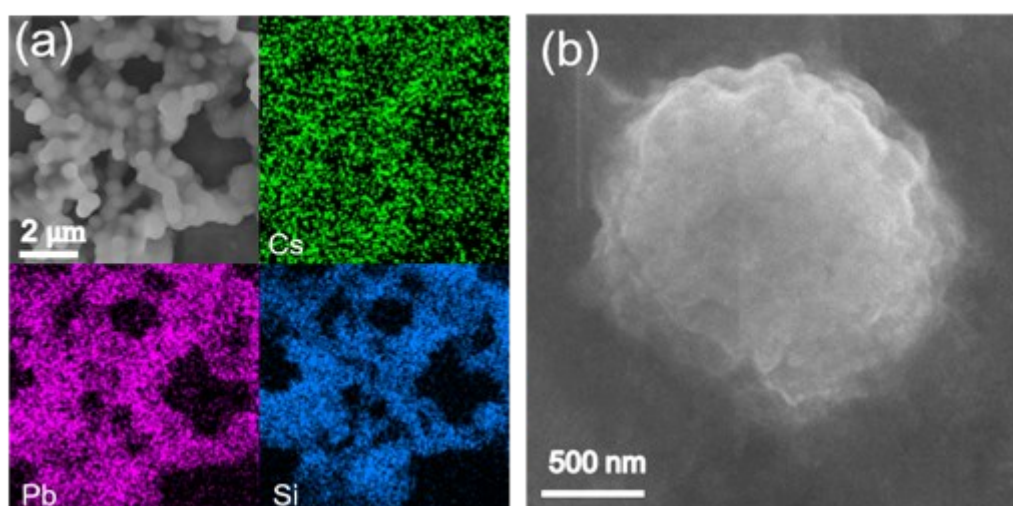


Figure S5 (a and b) SEM and elements mapping of APTES-NPLs powder.

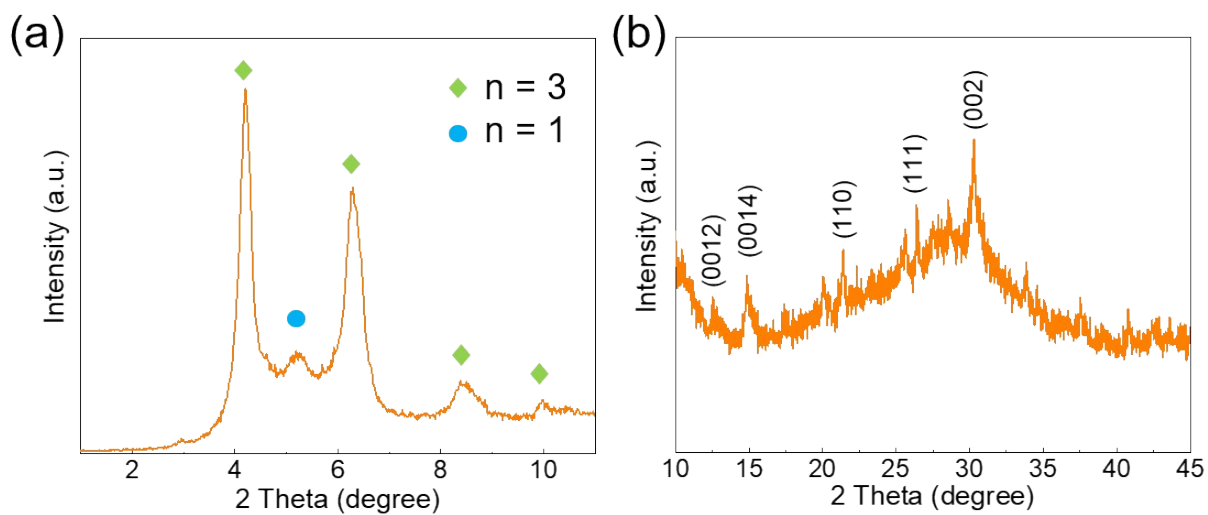


Figure. S6 XRD data of the crosslinking CsPbBr₃ NPLs at (a) small range and (b) local zoom : $2\theta = 10^\circ \sim 45^\circ$.

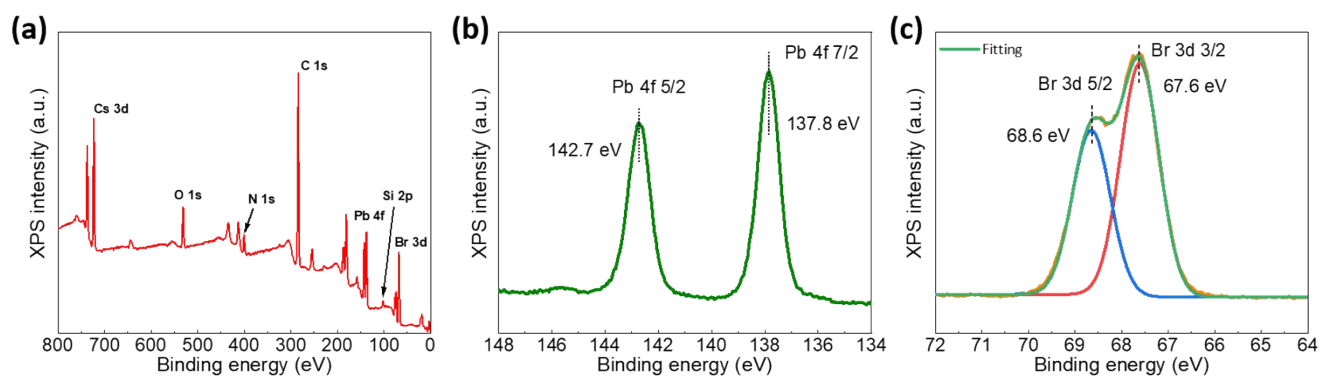


Figure. S7 (a) Full XPS spectrum of APTES-NPLs; high-resolution XPS spectra of (b) Pb 4f and (c) Br 3d regions for APTES-NPLs.

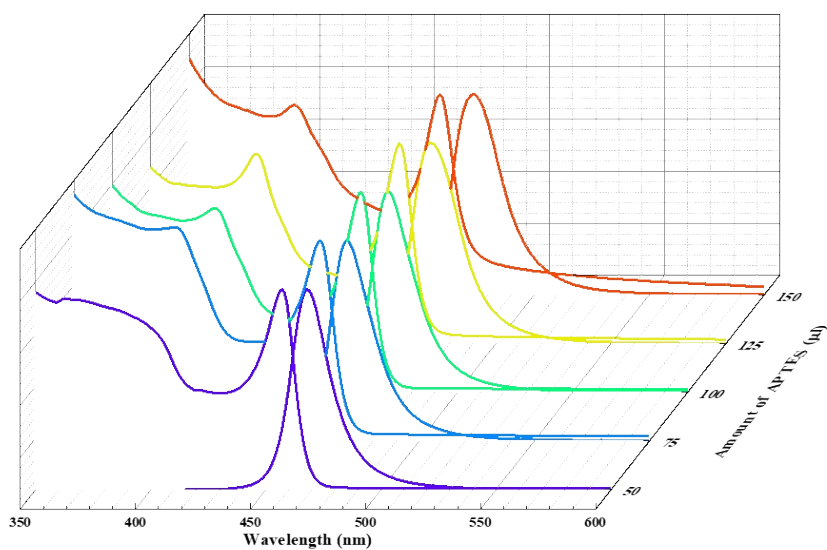


Figure. S8 PL and absorption spectra of crosslinking passivated NPLs prepared with different amount of APTES.

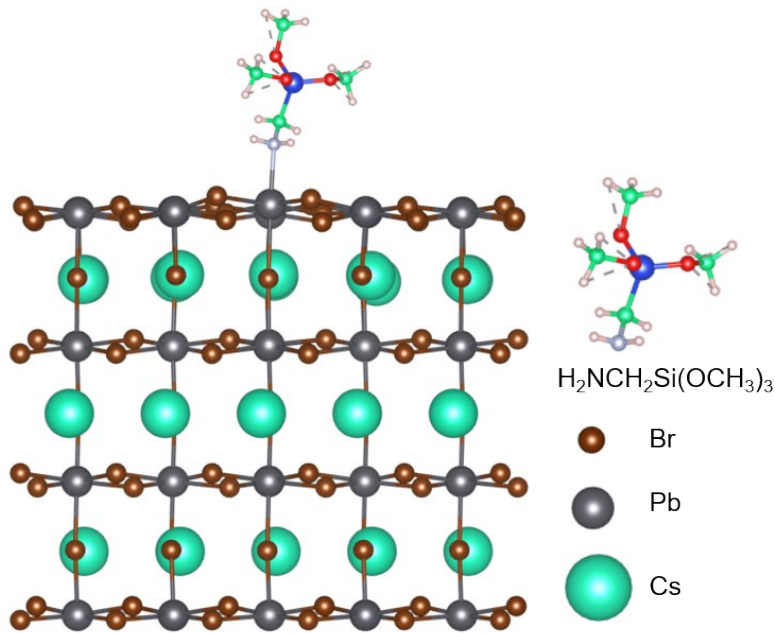


Figure. S9 DFT optimized structures for PbBr-terminated surface $\text{H}_2\text{NCH}_2\text{Si}(\text{OCH}_3)_3$ passivation.

Discussion: We employ density functional theory using the Vienna Ab initio Simulation Package (VASP) for all calculations in this paper. The projector augmented-wave method (PAW) is adopted to describe the electron-ion potential. The generalized gradient approximation (GGA) in the form of Perdew–Burke–Ernzerhof (PBE) approach is used for describing the exchange correlation interaction. The kinetic energy cutoff of the plane wave basis set was set to be 400 eV. In order to eliminate the interactions between adjacent layers, the vacuum layer is 20 Å. The geometric structures were fully relaxed until the total energy converged to 10^{-4} eV, and the Hellmann–Feynman force on each atom was less than 0.05 eV/Å. A long-range van der Waals interaction is described by the DFT-D3 method. Based on the orthorhombic CsPbBr_3 , we constructed a 2×2 supercell consisting of 7 atomic layers.

Table S1. Lifetime and Fractional Contribution of Different Decay Channels for Pristine and In SituPassivated CsPbBr₃ NPLs

Temperature	$A_1(\%)$	$\tau_1(ns)$	$A_2(\%)$	$\tau_2(ns)$	$\tau_{av}(ns)$
5K	0.166	1.7	0.834	9.41	9.14
65K	0.11	1.27	0.89	7.28	7.16
110K	0.09	0.96	0.91	5.92	5.84
155K	0.121	0.76	0.879	5.46	5.37
200K	0.151	0.73	0.849	5.33	5.22
245K	0.08	0.64	0.92	4.84	4.79
290K	0.216	0.74	0.784	4.47	4.31