Supplementary information for

High-efficiency and stable photoluminescence of $CH_3NH_3PbBr_3$@$CsPbBr_3$ perovskite quantum dot

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1. Element composition of QDs

The scanning electron microscope (SEM) image, the X-ray energy dispersive spectroscopy (EDS), and the element mappings were observed by HIROX SH-4000 with the accelerating voltage of 5-30 kV. The results are shown in Fig. S1. The element mappings (Fig. S1c-e) show that in addition to C, H, and N elements, $CH_3NH_3PbBr_3$@$CsPbBr_3$ QDs also contain Cs, Pb, and Br elements in a uniform distribution. For confirming the composition, EDS of QDs is adopted and shown in Fig. S1b. The atomic composition of Cs, Pb, and Br are 13.1, 21.0, and 65.9 at.%, respectively. Thus, the mole ratio of Pb:Br is about 1:3, which conforms to the stoichiometric ratio.

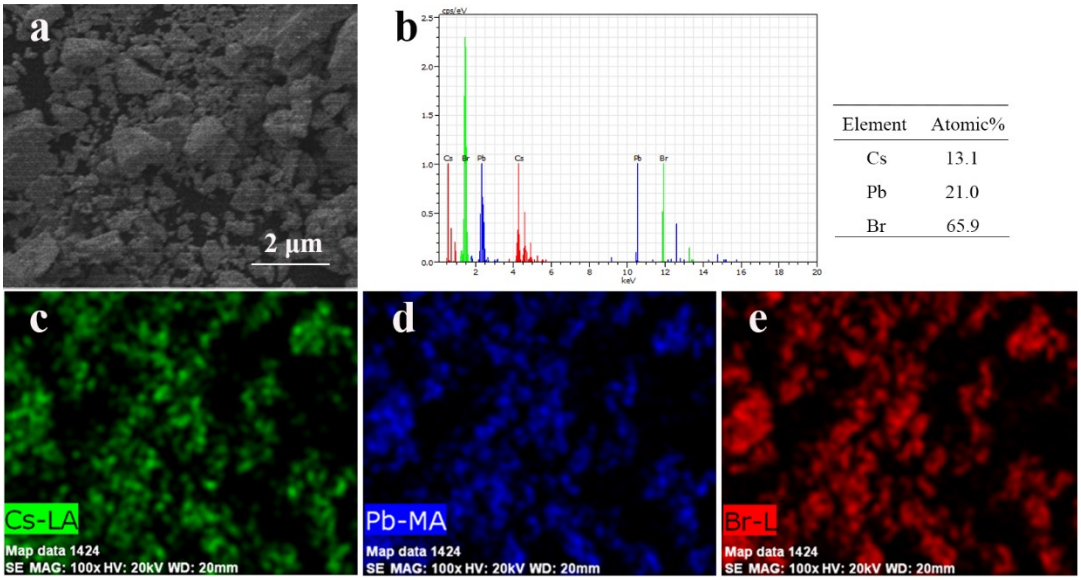

Fig. S1 SEM image (a), EDS spectroscopy (b), and element mappings (c-e) of $CH_3NH_3PbBr_3$@$CsPbBr_3$ QDs.

2. Crystal structure collapse

Experiencing high temperature from the electron beam radiation, the crystal structure of organic shell in $CsPbBr_3$@$CH_3NH_3PbBr_3$ composite QDs collapses and loosens, and the crystallinity is reduced as the following XRD patterns (Fig. S2), so that the size of a single quantum dot expands significantly, as shown in Fig. 1c.

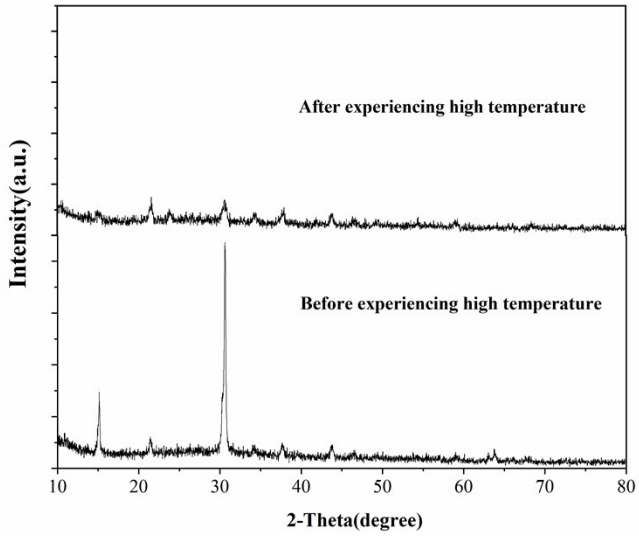

Fig. S2 XRD patterns of $CsPbBr_3$@$CH_3NH_3PbBr_3$ before and after test

3. XRD patterns of $CH_3NH_3PbBr_3$@$CsPbBr_3$

From the enlarged XRD patterns, as shown in the following Fig. S3, the diffraction angles of core-shell structure move to a higher value. According to the Bragg equation $2d_{(h^*k^*l^*)} \cdot \sin\theta_{hkl} = n\lambda$, it can be known that the smaller the interplanar spacing *d*, the higher the diffraction angle θ. The unit cell parameters and the interplanar spacing *d* of $CsPbBr_3$ are slightly smaller than those of $CH_3NH_3PbBr_3$. On the other hand, because the composite QDs include two similar crystal phases, the diffraction peaks overlap with each other and are broadened to a certain extent. Thus, compared with $CH_3NH_3PbBr_3$, $CH_3NH_3PbBr_3$@$CsPbBr_3$ has the higher diffraction angle and the wider full width at half maxima (FWHM) as listed in the Table S1. These observed phenomena also can confirm the core-shell structure of composite QDs.

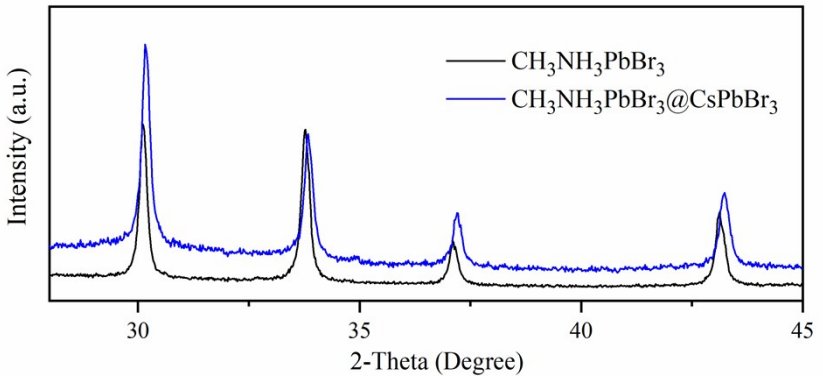

Fig. S3 XRD patterns with the diffraction angle 2θ of 28-45° for $CH_3NH_3PbBr_3$ and $CH_3NH_3PbBr_3$@$CsPbBr_3$

Table S1 Diffraction angle and FWHM of $CH_3NH_3PbBr_3$ and $CH_3NH_3PbBr_3$@$CsPbBr_3$

No.	QDs	2θ (°)	FWHM
1	$CH_3NH_3PbBr_3$	30.1	0.22
	$CH_3NH_3PbBr_3$@$CsPbBr_3$	30.2	0.27
2	$CH_3NH_3PbBr_3$	33.7	0.25
	$CH_3NH_3PbBr_3$@$CsPbBr_3$	33.8	0.30
3	$CH_3NH_3PbBr_3$	37.1	0.30
	$CH_3NH_3PbBr_3$@$CsPbBr_3$	37.2	0.54
4	$CH_3NH_3PbBr_3$	43.1	0.28
	$CH_3NH_3PbBr_3$@$CsPbBr_3$	43.3	0.39

4. Theoretical calculation

In order to discuss the electric transition of PL of QDs, the partial density of states (PDOS) of $CH_3NH_3PbBr_3$ and $CH_3NH_3PbBr_3$@$CsPbBr_3$ were calculated by using the density functional theory (DFT) based on pseudopotential plane wave.(*Phys. Rev. B*, 2007, 76, 075401; *Electrochim. Acta*, 2016, 187, 560-566; *Comput. Phys. Commun.*, 1997, 107, 187; *J. Mater. Chem. C*, 2017, 5(31), 7904-7910) The pseudopotential plane wave method is suitable for periodic structure.(*Eur. Phys. J. Appl. Phys.*, 2004, 27, 251-254) All of DFT calculations were performed with Cambridge Sequential Total Energy Package (CASTEP) program package in Materials Studio software.(*Phys. Rev. B*, 2003, 68, 085327; *Z. Kristallogr.* 2005, 220(5-6), 567-570)

The (001) crystal surface of $CH_3NH_3PbBr_3$ and $CH_3NH_3PbBr_3$@$CsPbBr_3$ were cleaved based on the crystal structure of $CH_3NH_3PbBr_3$ and $CsPbBr_3$, as shown in Fig. 5. The fractional thickness were 5 and then 2×2 (U×V) super cells were created. The thickness of vacuum slab were built as 30 Å with the vacuum orientation of *c*. the geometry optimization were performed. In order to calculate accurately, the generalized gradient approximation (GGA) with Perdew-Burke-Ernzerhof (PBE) type exchange-correlation functions was adopted to treat the perovskite-type rare earth ferrite.(*Phys. Rev. Lett.*, 1996, 77, 3865; *Phys. Rev. Lett.*, 1997, 78, 1396) The plane wave basis sets are used with the ultrasoft pseudopotential. The Monkhorst-Pack grid k-points of 2×2×1, the energy cutoff of 300.0 eV were adopted. The convergence criterion for energy was the energy error of 2.0×10^{-6} eV/atom. Based on the optimized models, the band structure, the PDOS, and the electron density isosurfaces of the orbit (wavefunction) of $CH_3NH_3PbBr_3$ and $CH_3NH_3PbBr_3$@$CsPbBr_3$ were calculated.

The calculated band structure and PDOS are shown in Fig. S4. The energy gaps of $CH_3NH_3PbBr_3$ and $CH_3NH_3PbBr_3$@$CsPbBr_3$ are 2.37 and 2.41 eV, respectively. The

valence and the conduction bands are mainly consisted of Br^- 2p, Pb^{2+} 3p electronic state, respectively.

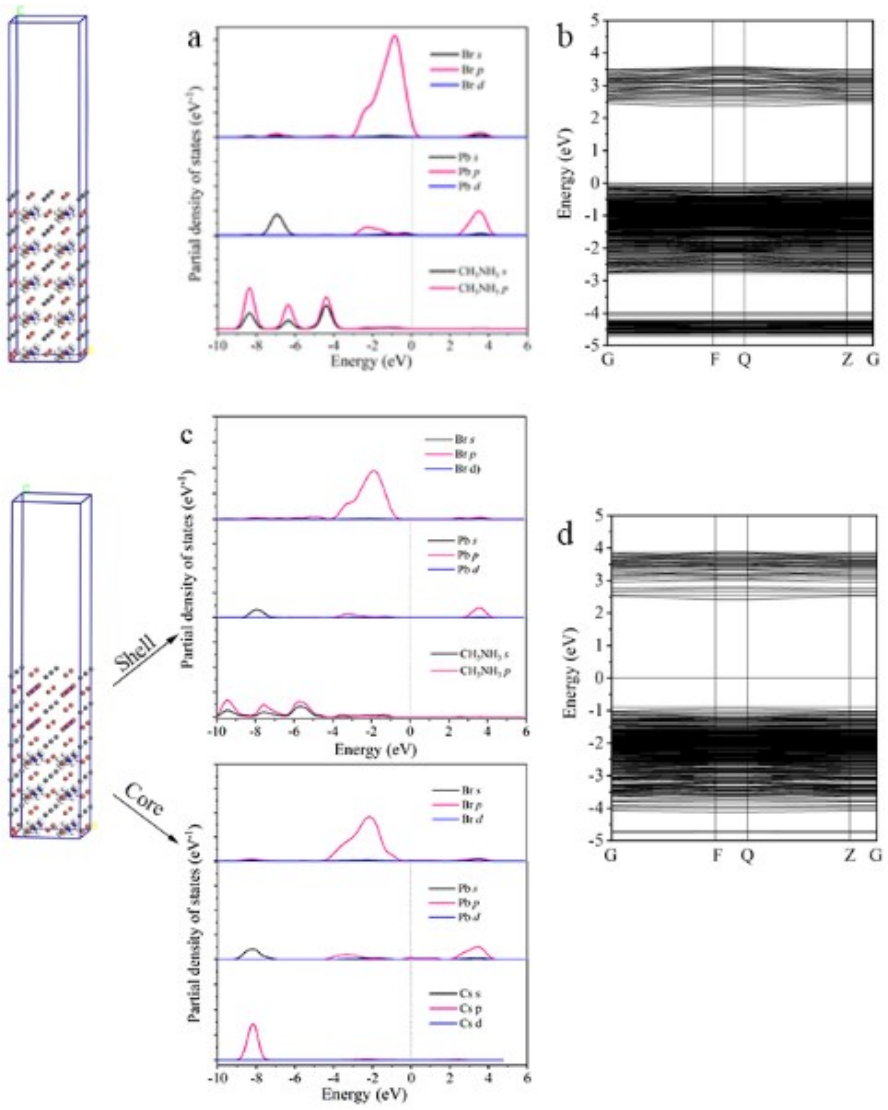

Fig. S4 PDOS (a) and band structure (b) for $CH_3NH_3PbBr_3$. PDOS of core and shell (c), band structure (d) for $CH_3NH_3PbBr_3$@$CsPbBr_3$

5. Instability

After QDs were placed in natural light, room temperature of about 25-35 ℃ and humidity of about 50-60% for more than four months, the XRD patterns of $CsPbBr_3$@$CH_3NH_3PbBr_3$ agree with those of the orthorhombic $PbBr_2$ (JCPDS PDF#31-0679), as shown in Fig. 4a and Fig. S5, which confirms the formation of $PbBr_2$.

$CH_3NH_3PbBr_3$ may have decomposed into CH_3NH_3Br and $PbBr_2$, and $PbBr_2$ emits a weak PL at about 400-500 nm. (*Chem. Commun.*, 2016, 52, 7118) Additionally, the organic shell of has been collapsed and decomposed, and the core has agglomerated. Thus the $CH_3NH_3PbBr_3$ and $CsPbBr_3@CH_3NH_3PbBr_3$ QDs only have a weak PL at about 400-500 nm.

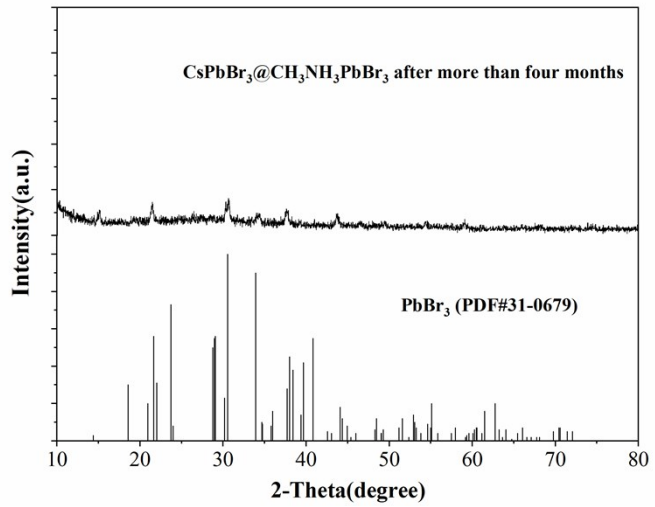

Fig. S5 XRD patterns of $CsPbBr_3@CH_3NH_3PbBr_3$ after more than four months and the orthorhombic $PbBr_2$