Supplementary Information

Synergistic Enhancement Optoelectronic Performances and Stability of MA and Cs in $FA_xMA_yCs_{1-x-y}PbI_zBr_{3-z}$ Single Crystal

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1. Thermal stability of $FA_xMA_yCs_{1-x-y}PbI_zBr_{3-z}$ SCs

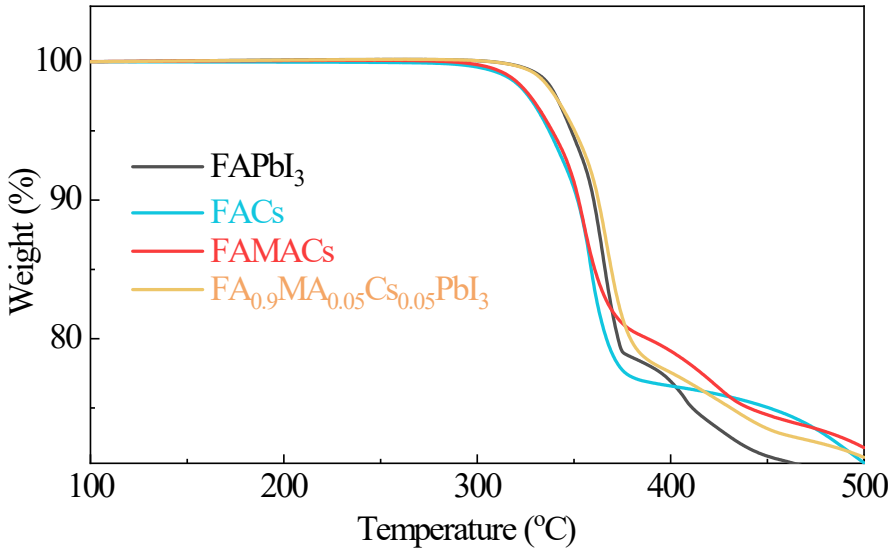

Figure S1. Thermogravimetric analysis (TGA) curves of $FA_xMA_yCs_{1-x-y}PbI_zBr_{3-z}$ SCs.

2. Halide Content in FAMACs and FACs

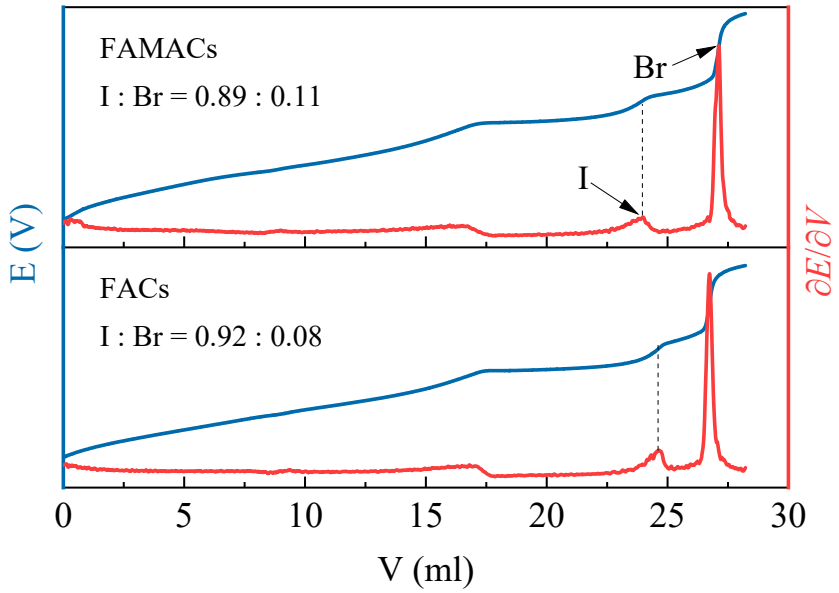

Figure S2. Solution potential curve of FAMACs and FACs.

Potentiometric titration is the method to determine the end point of titration by measuring the change of potential in the process of titration. When titration reaches the end point, the concentration of ions to be measured will often change by several orders of magnitude, causing the potential jump. Specific test method: 50ml of FAMACs and FACs solution with a halogen concentration of about 0.01M was prepared, and $AgNO_3$ standard solution was dropped into the solution at a constant rate (0.7mL/min), and the change of solution potential was recorded. The first potential leap point is the titration end-point of I^-, and the second potential leap point is the titration end-point of Br^-. The reactions involved are as follows:

$$Ag^+ + I^- = AgI\downarrow$$

$$Ag^+ + Br^- = AgBr\downarrow$$

3. Pb-I bond lengths

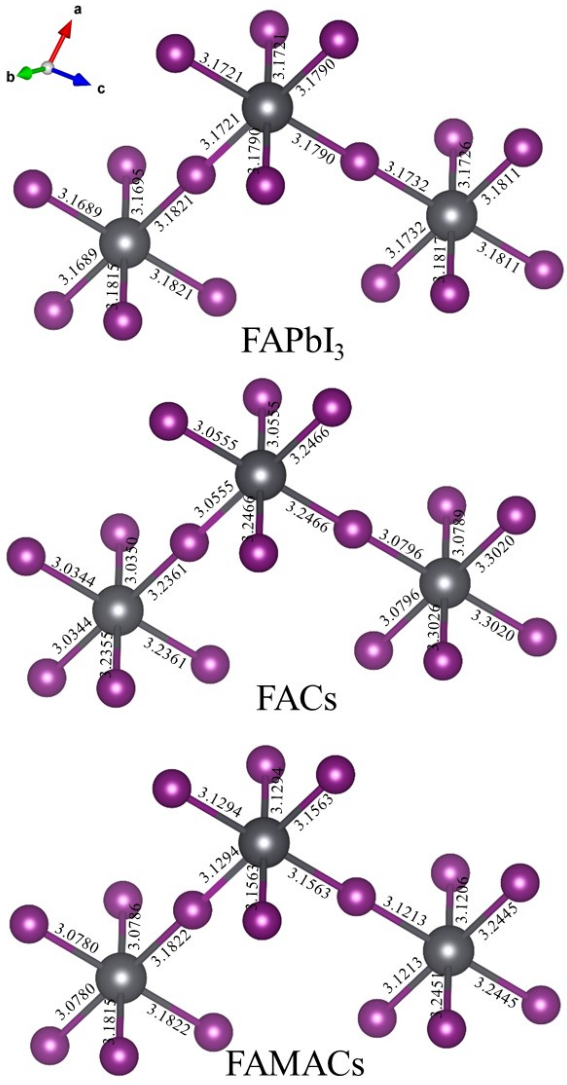

Figure S3. Pb-I bond lengths of $FAPbI_3$, FACs and FAMACs

4. Dielectric constant of $FA_xMA_yCs_{1-x-y}PbI_zBr_{3-z}$

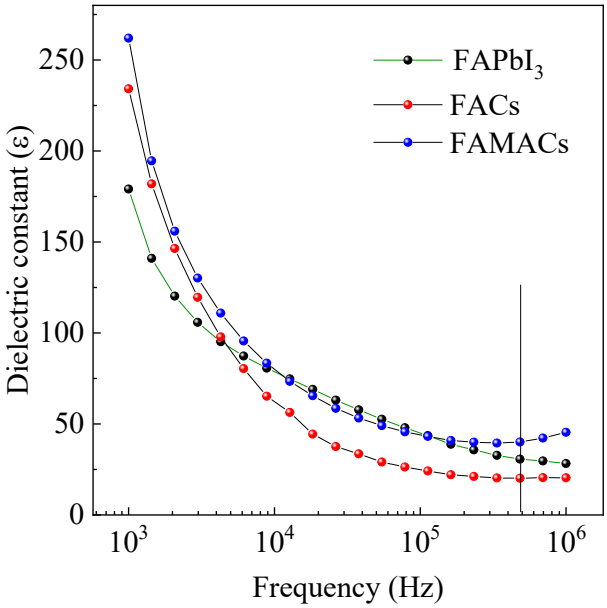

Figure S4. Dielectric constant (ε) of $FA_xMA_yCs_{1-x-y}PbI_zBr_{3-z}$ SCs in the frequency range of 1 kHz to 1 MHz.

5. Dark current of $FA_xMA_yCs_{1-x-y}PbI_zBr_{3-z}$ photodetector

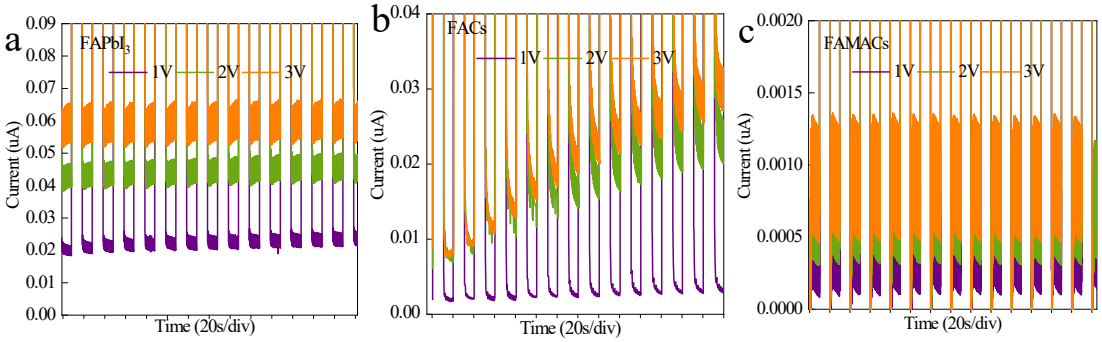

Figure S5. (a-c) Variation of dark current of $FA_xMA_yCs_{1-x-y}PbI_zBr_{3-z}$ SCs after continuous cycling under various applied voltages.

6. Response speed under various applied voltages of the devices

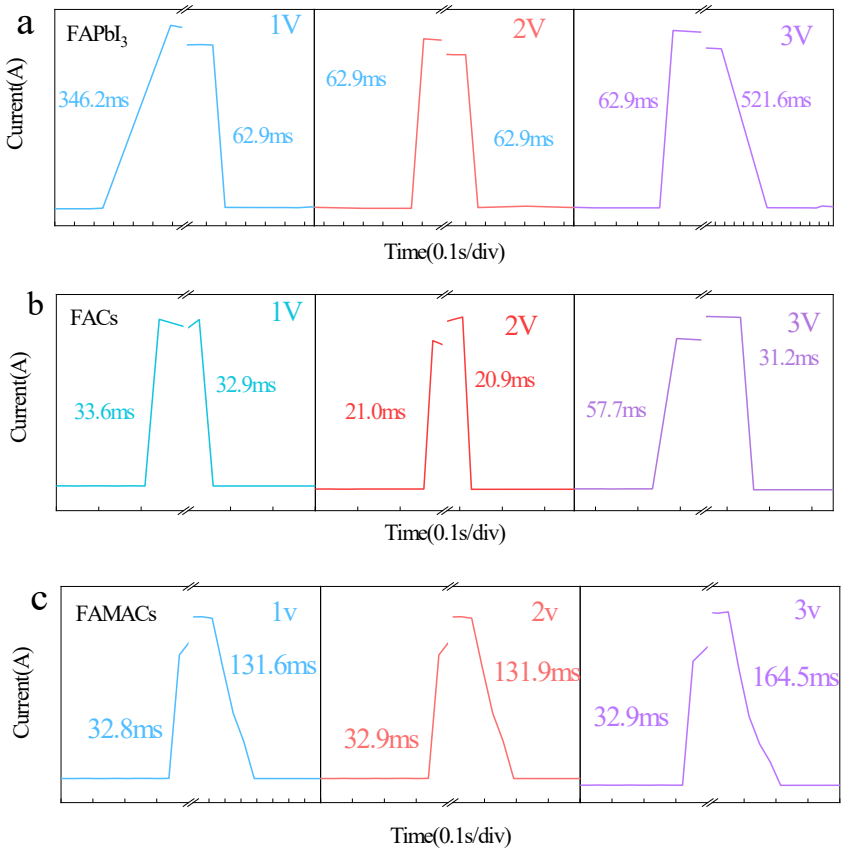

Figure S6. (a-c) Detailed response times under various applied voltages of the devices.

7. In suit PL spectra of FACs and FAMACs

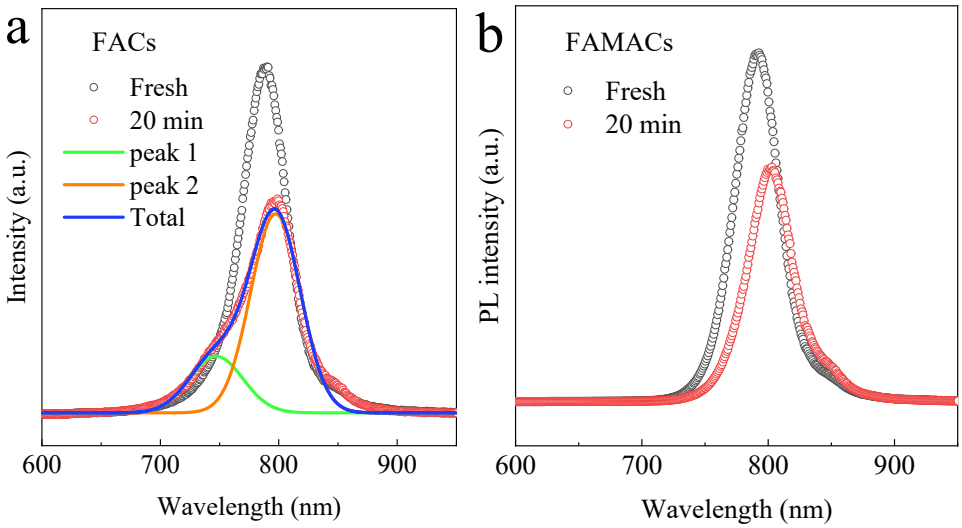

Figure S7. (a-b) In suit PL spectra of FACs and FAMACs