## ARTICLE

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# Synergistic passivation effect of functional doped Povidone-Iodine on quasi-2D perovskite solar cells

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 $\begin{array}{c} A_1 \times \tau_1^2 + A_2 \times \tau_2^2 \\ \hline \text{Table S1. Summaries of fitting parameters of time-resolved photolumines} cence for the GAMA_{s}Pb_{s}I_{16} films before and after PVP and PVP-I treatment. The average life is calculated according to the following formula^{1.2:} \\ \hline A_1 \times \tau_1 + A_2 \times \tau_2 \end{array}$ 

| Sample   | $\tau_1$ (ns) | Intensity τ <sub>1</sub><br>(%) | $	au_2$ (ns) | Intensity<br>τ <sub>2</sub> (%) | $\frac{\tau_{ave}}{(\text{ns})}$ |
|----------|---------------|---------------------------------|--------------|---------------------------------|----------------------------------|
| Pristine | 6.23          | 4.01                            | 221.77       | 95.99                           | 213.13                           |
| PVP      | 4.01          | 3.00                            | 253.83       | 97.00                           | 246.34                           |
| PVP-I    | 7.45          | 1.61                            | 584.60       | 98.39                           | 575.31                           |



Figure S1. Reverse- and forward-scanned J-V curves of PSCs (a) without (pristine) and with (b) PVP and (c) PVP-I treatment.



Figure S2. Statistical photovoltaic parameters of (a)  $J_{scr}$  (b)  $V_{ocr}$  (c) FF and (d) PCE depending on PVP concentration.



Figure S3. Statistical photovoltaic parameters of (a) J<sub>sc</sub>, (b) V<sub>oc</sub>, (c) FF and (d) PCE depending on PVP-I concentration.



Figure S4. UPS spectra of the ACI GAMA<sub>5</sub>Pb<sub>51<sub>16</sub></sub> perovskite films (a, b) without(pristine), with (d, e) PVP and (g, h) PVP-I treatment and the corresponding (Ahv)<sup>2</sup> vs hv curves of 2D perovskites films (c, f, i).



Figure S5. Absorbance spectra of the quasi-2D ACI perovskite films before and after PVP and PVP-I treatment.



Figure S6. C1s XPS spectra for perovskite film (a) without, with (b) PVP-I and (c) PVP-I. The binding energy was calibrated using the C-C binding energy of 284.8 eV. N1s XPS spectra for perovskite film (d) without, with (e) PVP-I and (f) PVP-I. The binding energy was calibrated using the C-NH<sub>3</sub> binding energy of 400.0 eV. Empty circles and lines represent the measured and the fit data, respectively.



Figure S7. (a) The illumination stability of the devices fabricated before and after PVP and PVP-I treatment tested at 20 ± 5 °C, under AM 1.5G one sun illumination in the N<sub>2</sub> glove-box. (b) The humidity stability of the devices fabricated before and after PVP and PVP-I treatment tested at 20 ± 5 °C, 40±5% RH in the air.

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| Device structure               | Perovskite composition                                | PCE (%) | Ref       |
|--------------------------------|---|---------|-----------|
| ITO/PEDOT:PSS/PVSK/PCBM/LiF:A1 | $(GA)(MA)_3Pb_3I_{10}$                                | 16.65   | [3]       |
| ITO/PTAA/PVSK/PCBM/BCP/Ag      | (GA)(MA)5Pb5I16                                       | 15.73   | This work |
| ITO/PEDOT:PSS/PVSK/PCBM/BCP/Ag | $(GA)(MA)_4Pb_4I_{13}$                                | 14.3    | [4]       |
| ITO/ PEDOT:PSS/PVSK/C60/BCP/Ag | $(GA)(MA)_4Pb_4I_{13}$                                | 13.82   | [5]       |
| ITO/PEDOT:PSS/PVSK/PCBM/BCP/Ag | (GA)(MA) <sub>4</sub> Pb <sub>4</sub> I <sub>13</sub> | 13.53   | [6]       |
| ITO/ PEDOT:PSS/PVSK/C60/BCP/Ag | (GA)(MA) <sub>4</sub> Pb <sub>4</sub> I <sub>13</sub> | 12.8    | [7]       |
| FTO/PEDOT:PSS/PVSK/PCBM/A1     | $(GA)(MA)_3Pb_3I_{10}$                                | 7.26    | [8]       |

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