Response to Reviewer 1 Comments

We really thank the reviewer for reviewing this manuscript. Our modifications and clarification in response to the reviewer’s comments are listed in the following.

**Comment #1: Major critic point:**

Fig. 5a) shows EQE larger than 400%!!!

in line 250 an explanation is given:

The photoconduction results from the electron-hole pairs excited by the illumination light with photon energy larger than the semiconductor band gap. The 405 nm light with larger energy can excite more electron-hole pairs in perovskite thin films, leading to a higher photocurrent. But 405 nm = 3.06 eV and the MAPbI$_3$ energy gap is 1.6 eV to excite 2 electrons with 1 photon at least 3.2 eV would be needed.

Possibly the area of the detector, which is given in line 94 as 0.014 cm$^2$ is way too small it would be 0.12 mm x 0.12 mm.

In addition EQE is a term in photovoltaics; for detectors I find in Wikipedia:

Quantum efficiency of Image Sensors: Quantum efficiency (QE) is the fraction of photon flux that contributes to the photocurrent in a photodetector or a pixel. Quantum efficiency is one of the most important parameters used to evaluate the quality of a detector and is often called the spectral response to reflect its wavelength dependence. It is defined as the number of signal electrons created per incident photon. In some cases it can exceed 100% (i.e. when more than one electron is created per incident photon).

Spectral responsivity

Spectral responsivity is a similar measurement, but it has different units: amperes per watt (A/W); (i.e. how much current comes out of the device per incoming photon of a given energy and wavelength). Both the quantum efficiency and the responsivity are functions of the photons' wavelength (indicated by the subscript $\lambda$).

To convert from responsivity ($R_\lambda$ in A/W) to QE$_\lambda$ [6] (on a scale 0 to 1):

$$QE_\lambda = \frac{R_\lambda}{\lambda} \times \frac{hc}{e} \approx \frac{R_\lambda}{\lambda} \times (1240 \text{ W} \cdot \text{nm} / \text{A})$$

where $\lambda$ is the wavelength in nm, $h$ is the Planck constant, $c$ is the speed of light in a vacuum, and $e$ is the elementary charge.

**Response #1:** Thanks for the reviewer’s comments.

ITO substrates used in the experiment were etched to form electrodes with a channel width of
0.02 cm and length of 0.7 cm, respectively. Thus, the active area of the detector is 0.02 cm × 0.7 cm (0.014 cm²). We have added this experimental detail in line 95 of the revised manuscript. Regarding the EQE value, we have recalculated and double confirmed the results. In our calculation, the responsivity and EQE show almost the same evolution as shown in Figure 5 c and d. For photoresistors, the EQE value is also dependent on applied external bias. An external bias will result in an increasing number of electrons collected by the electrode and giving an EQE number higher than 100%. In our experiments, the EQE measurement was calculated from responsivity at 5 V bias. In addition, EQE of perovskite-based photoresistors with a value higher than 100% was also reported by other workers. For example, in Tong’s report, halide perovskite (E_g=1.7 eV) based photodetectors achieved an EQE value of 4000% at 1 V bias under 650 nm illumination with an intensity of 0.2 mW/cm², as shown in the following Figure (Figure 4e of reference 51).

Comment #2: luminescence around PbI₂ gap i.e. 500nm should be shown?
Response #2: Thanks for the reviewer’s kind suggestion. The luminescence of the perovskite films was measured using a 532 nm laser as excitation light. Therefore, the luminescence around 500 nm is not detectable. However, the present of PbI₂ was confirmed by the XRD spectra, as shown in Figure 1a.

Comment #3: Red shift with increasing PbI₂/MAI ratio related to reduced lattice strain: any hint on strain in XRD?
Response #3: Thanks for the reviewer’s comments. We have carefully analysed the XRD pattern again, however for the time being we cannot find direct evidence regarding the reduced lattice strain from XRD. Regarding the PL shift, lattice stain dependence have been discussed in the revised manuscript.
In a recent work, Jones et al demonstrated that lattice strain is directly associated with enhanced defect concentrations and non-radiative recombination on the microscale [44]. Here, the PL intensities are stronger for the PbI$_2$ rich perovskites compared with the pure-phase perovskite, which are consistent with previous reports [38, 39, 45]. The presence of PbI$_2$ can passivate the grain boundaries of the perovskite films and this passivation effect on PL intensity is dominant compared with lattice strain. The sample with MAI: PbI$_2$ ratio of 0.5: 1 (sample S2) shows the strongest PL emission, which suggests an effective suppression of non-radiative recombination.

Comment #4: Line 163: Considering the steady-state PL results as shown in Figure 2b, the observed longer PL lifetime in sample S2 suggests that the grain boundaries of MAPbI$_3$

But: In Fig.2b) higher luminescence is observed, not lifetime

Response #4: Thanks for the reviewer’s comments. The defect states or shallow trapping levels in the grain boundaries of the perovskite will result in PL emission quenching and therefore behave as non-radiative recombination centers. As shown by Figure 2c, the observed longer PL lifetime in sample S2 suggests that the defect states or shallow trapping levels in the grain boundaries of the S2 sample are passivated by PbI$_2$ in the film. Meanwhile, the steady-state PL results in Figure 2b shows a higher PL intensity was observed from the S2 sample, which is consistent with the lifetime measurement. The revision is shown in line 169-175 of the revised manuscript highlight.

Comment #5: Line 18

is: in this contribution, we prepared high …

change to: We prepared high...

Response #5: Thanks for the reviewer’s comments. The change has been made, as shown in line 18 of the revised manuscript highlight.

Comment #6: Line 28

is: the special dependence of photocurrent generation

change to: the spacial dependence of photocurrent generation

or change to: the local dependence of photocurrent generation

Response #6: Thanks for the reviewer’s comments. The change has been made, as shown in line 28 of the revised manuscript highlight.

Comment #7: Line 52
is: For a certain perovskite, composition is a crucial factor…
change to: For a certain perovskite material of AMX3 stoichiometry, chemical composition is a crucial factor…

Response #7: Thanks for the reviewer’s comments. The change has been made, as shown in line 53 of the revised manuscript highlight.

Comment #8: 102
is: photoelectronic performance of the devices were measured in the air
change to: photoelectronic performance of the devices were measured in air

Response #8: Thanks for the reviewer’s comments. The change has been made, as shown in line 103 of the revised manuscript highlight.

Comment #9: 153
is: which suggests an effective suppression of non-irradiative recombination.
change to: which suggests an effective suppression of non-radiative recombination.

Response #9: Thanks for the reviewer’s comments. The change has been made, as shown in line 158 of the revised manuscript highlight.

Comment #10: 244 Figure caption
Figure 5a shows the spectral photoresponse of the device in the wavelength range of 400-800 nm
Is in contradiction to line 238
(a) Spectral photoresponse performance of the S2 device in the wavelength ranging from 238 400 to 700 nm under a fixed incident light intensity.

Response #10: Thanks for the reviewer’s comments. The change has been made, as shown in line 255 of the revised manuscript highlight.