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Interface engineering for improving the performance of metal halide perovskite photodiode

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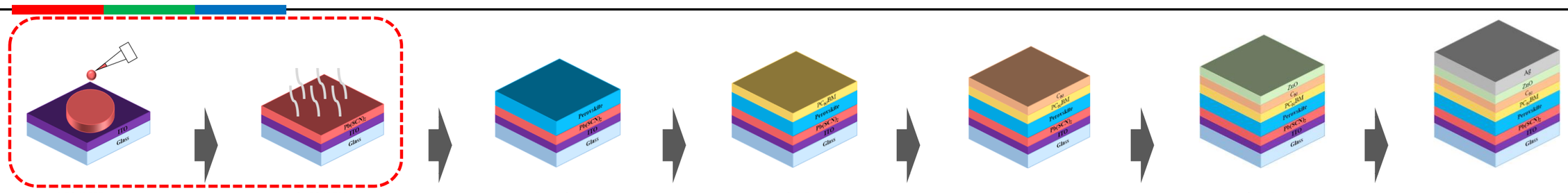
Introduction

Metal halide perovskite has emerged as a next-generation candidate for the active layer of photodiodes by replacing silicon due to its excellent optoelectronic properties such as high absorption coefficient, long carrier lifetime and light absorption selectivity. Perovskite can absorb light selectively because their band gap can be tuned by changing the composition of halide at the X site. However, it is still challenging to synthesize metal halide perovskites absorbing blue light.

It is well known that the ratio of chlorine (Cl) in X site should be increased to absorb light in the short wavelength (400 nm ~ 520 nm) region and lead acetate ($Pb(CH_3COO)_2$) has been widely used in the synthesis process because of its low solubility of Cl. However, lead acetate accelerates the nucleation of perovskite and reduces the grain size, which makes it difficult to improve perovskite photodiodes (PePDs) performance. To enlarge the grain size of the perovskite, some methods, such as solvent engineering, interface engineering, and incorporating additives into the perovskite film have been reported. In this study, we inserted the $Pb(SCN)_2$ layer into the interface between ITO and perovskite to increase the grain size of the perovskite, and we report its effects on the properties of PePDs as a function of the thickness of the $Pb(SCN)_2$ layer.

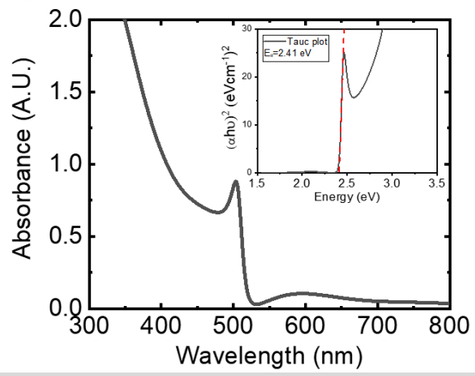
Experiments



a) $Pb(SCN)_2$: 5 mg/ml Spin coating
b) $Pb(SCN)_2$: Annealing at 60℃ for 5 min
c) Perovskite : Anti-Solvent bathing method
d) $PC_{60}BM$: Spin coating
e) C_{60} : Thermal evaporation
f) ZnO : Spin coating
g) Ag : Thermal evaporation

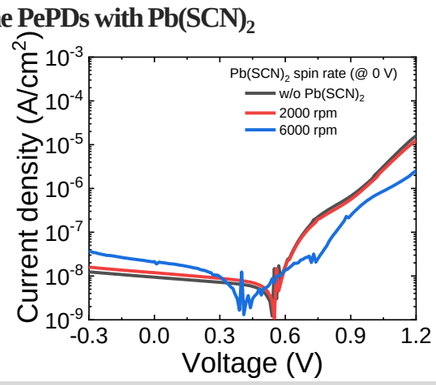
Result & Discussion

- Absorption spectra of the perovskite



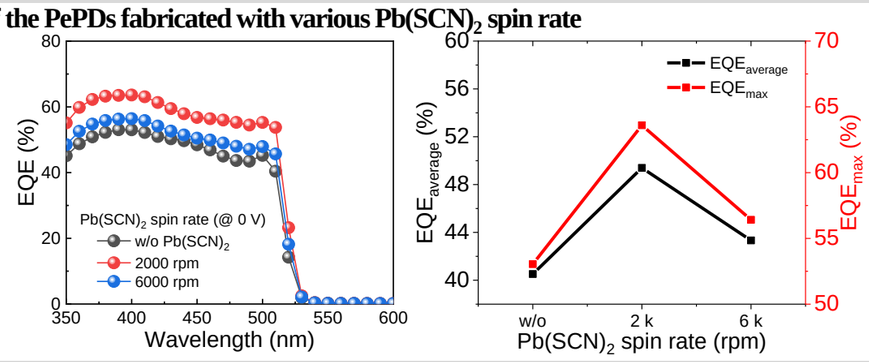
· We synthesized $MA_{0.6}FA_{0.4}PbBr_{2.4}Cl_{0.6}$ perovskite film.
· The absorption onset of perovskite was 528 nm and the absorption peak was located at 503 nm
· The bandgap of the perovskite calculated by using the Tauc plot method is 2.41 eV.

- Dark Current density of the PePDs with $Pb(SCN)_2$



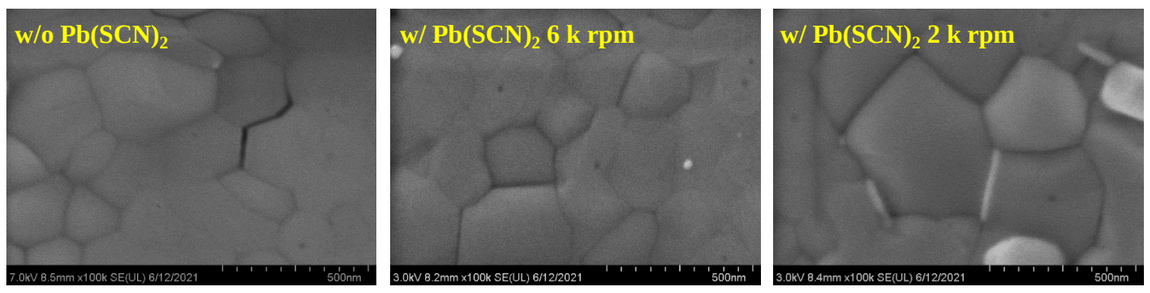
· The devices show low dark current density regardless of $Pb(SCN)_2$ thickness.
. The dark current density is almost identical at -0.3 V. (w/o $Pb(SCN)_2$ 1.44×10^{-8} A/cm², w/ $Pb(SCN)_2$ 6 k rpm 3.67×10^{-8} A/cm² , w/ $Pb(SCN)_2$ 2 k rpm 1.59×10^{-8} A/cm²).

- EQEs of the PePDs fabricated with various $Pb(SCN)_2$ spin rate



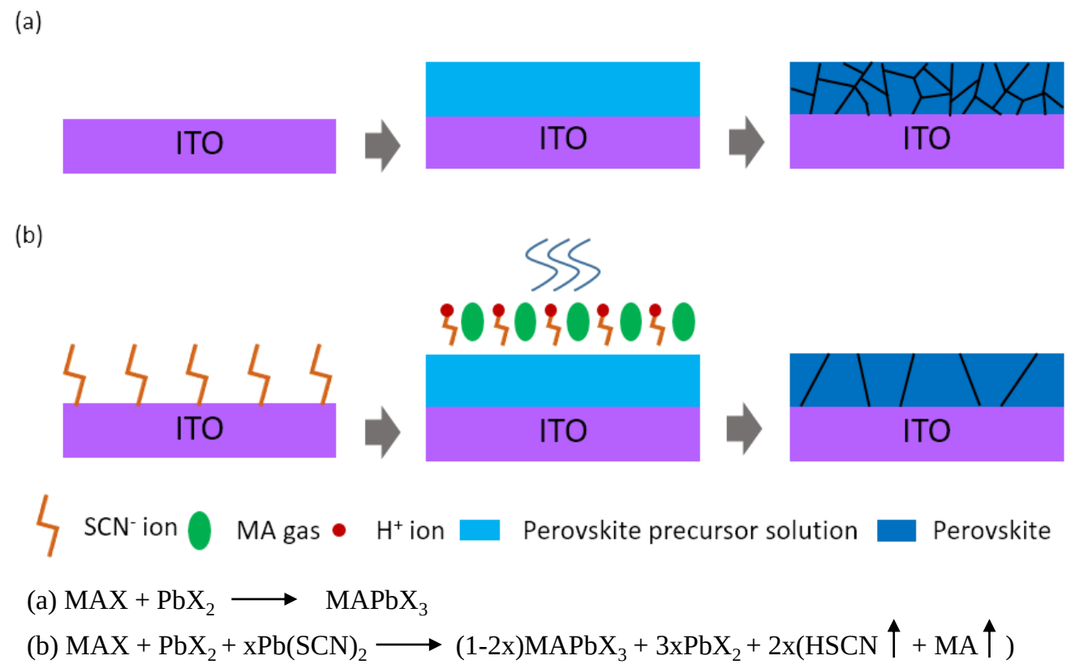
· The EQE of the PePD without $Pb(SCN)_2$ film and the PePD with thin $Pb(SCN)_2$ film(6 k rpm) was almost identical, but that of the PePD with thick $Pb(SCN)_2$ film(2 k rpm) was enhanced.
· The maximum EQE increased from 53.0 % to 63.6 % and the averaged EQE increased from 40.5 % to 49.4% by incorporating $Pb(SCN)_2$ film formed with a spin rate of 2 k rpm.

- FE-SEM image of perovskite film



· The thickness of $Pb(SCN)_2$ was controlled by varying the spin coating speed.
· The grain size of the perovskite film without $Pb(SCN)_2$ and the perovskite film with thin $Pb(SCN)_2$ film(spin coating speed : 6 k rpm) was almost identical, but it was enlarged in the perovskite film with thick $Pb(SCN)_2$ film(spin coating speed : 2 k rpm).

- Mechanism of increasing grain size by coating $Pb(SCN)_2$ at the interface between ITO and perovskite



(a) $MAX + PbX_2 \longrightarrow MAPbX_3$
(b) $MAX + PbX_2 + xPb(SCN)_2 \longrightarrow (1\text{-}2x)MAPbX_3 + 3xPbX_2 + 2x(HSCN\uparrow + MA\uparrow)$

· Grain boundaries reduce charge collection through charge trapping and the reduced charge collection degrades the PePDs performance. The density of grain boundaries can be reduced by increasing grain size.
· The SCN^- ions at the interface react with the MA^+ ions of PVSK to form HSCN and MA gases, and MA gas increases the grain size by delaying the nucleation rate of the perovskite.

Conclusion

- The grain size of perovskite was increased by forming $Pb(SCN)_2$ at the interface between ITO and perovskite.
- EQE of the PePD increased when $Pb(SCN)_2$ was coated at the interface. However, because sufficient $Pb(SCN)_2$ is required for the SCN^- and MA^+ ions to react at the interface, it is necessary to coat $Pb(SCN)_2$ layer over critical thickness.
- These results would be expected in the red or green perovsktie photodiode as well.

Acknowledgement

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