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Novel exciton dissociation behavior in tin-lead organohalide perovskites

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ABSTRACT

The design of organic-inorganic trihalide perovskite solar cells with higher performance, lower environmental pollution, lower cost and easier of fabrication should be a significant stride towards their practical application. Nevertheless the contribution of exciton dissociation behavior of the perovskite to this strategy has not been recognized comparing with film morphology, device architecture and fabrication process. Here we demonstrated a series of solution-processed solid-state tin-lead organohalide perovskite photovoltaic solar cells using carefully selected fullerene derivatives as the electron transport layer. The hybrid excitonic feature of $\text{CH}_3\text{NH}_3\text{Sn}_{0.5}\text{Pb}_{0.5}\text{I}_3$ was revealed for the first time by comparing the exciton dissociation behaviors of $\text{CH}_3\text{NH}_3\text{Sn}_{0.5}\text{Pb}_{0.5}\text{I}_3$ with conventional excitonic semiconductor in both planar heterojunction solar cells and lateral architecture photosensors. By optimizing the lowest unoccupied molecular orbital level of electron transport layer materials, high open circuit voltage of 0.69 V, short circuit photocurrent density of 22.8 mA cm^{-2} and power conversion efficiency of more than 10% were achieved with the incident photon conversion efficiency spectra onset reaching 1050 nm. These devices may work as high performance photodetectors with a broad spectral response expanding from UV-visible to near-infrared. Our results will contribute to the exciton dissociation behavior to be an efficient perspective for the improvement of perovskite solar cells.

Keywords

Tin-lead organohalide perovskites; Fullerenes; solar cells; exciton dissociation behaviors

Introduction²

The emergence of organohalide perovskites (AMX_3 , $\text{A} = \text{CH}_3\text{NH}_3$ (MA)⁺, $\text{NH}_2\text{CH}=\text{NH}_2$ ⁺, $\text{M} = \text{Pb, Sn, Ge}$, $\text{X} = \text{Cl, Br, I}$) as light harvesters has revolutionized the photovoltaic technologies and underwent fast development in recent years [1-3]. More recently, it was observed that perovskite materials with high optical absorption coefficients, long photoexcited carrier lifetimes, and long diffusion lengths played a critical roles for high photovoltaic performance [4,5]. Photovoltaic devices based on lead organohalide perovskites materials with TiO_2 as the electron transport layer (ETL) material and 2,2',7,7'-tetrakis-(N,N-di-p-meth-oxyphenylamine)-9,9-spirobifluorene (spiro-OMeTAD) as the hole transport layer (HTL) material have been reported to deliver high power conversion efficiency (PCE) even above 20% [6-9].

However, the environmental toxicity of lead may limit commercial applications of lead organohalide perovskite (LOP) solar cells [3]. Finding ways to create lead-free perovskites without sacrificing device performance is especially critical for perovskite photovoltaics. Partial or full replacement of lead with non-toxic tin is a viable alternative. Several groups have reported a broader absorption edge for tin-lead organohalide perovskite (TLOP) (reaching 1060 nm) than the LOP (less than 800 nm) [10-12]. In theory, light harvesting in the near-infrared (NIR) region is required to achieve higher PCE [11]. Unfortunately, recently reported TLOP solar cells with absorption onset more than 1000 nm displayed much lower PCEs as LOP solar cells

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