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Variation in the Photocurrent Response Due to Different Emissive States in Methylammonium Lead Bromide Perovskites

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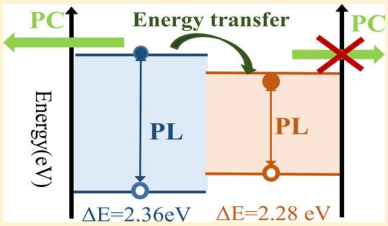
Supporting Information

ABSTRACT: Thin films and crystals of methylammonium lead bromide ($MAPbBr_3$) perovskites have strong photoluminescence (PL). Previous studies have shown that the emission arises from different states. However, the role of these states in the performance of a solar cell has not been reported. We have used photocurrent and photoluminescence microscopies (PCM and PLM) to investigate the correlation between the photocurrent (PC) and the PL behavior in the different regions of $MAPbBr_3$ thin film solar cells. Our results show that the PC and the PL responses from the different regions in the thin film show poor correlation compared to the correlation between those of a high efficiency GaAs solar cell. Furthermore, we establish a relationship between the different emissive states and the PC and the PL responses. Out of the two emissive states at 2.34 and 2.28 eV that have been reported, only the state at 2.34 eV has a dominant contribution to the PC. Our results suggest that the emission at 2.28 eV is related to traps, which can lower the performance of the solar cells. Finally, the correlation analysis of the PC and the PL responses we have presented can be used in any solar cell made from direct band gap semiconductor to identify the loss channels in the device.



1. INTRODUCTION

Metal halide perovskites have emerged as excellent materials for photovoltaic technology because of ease of fabrication as well as favorable properties such as high absorption coefficient[1−5] and carrier mobilities.[6−10] The most efficient perovskite solar cells have a power conversion efficiency (PCE) of 22.1%.[11,12] Although solution-processed perovskites are inherently inhomogeneous, they are still highly emissive.[13−16] The high PCE as well as the PL yield suggests that the perovskite solar cells may be operating close to the Shockley−Queisser limit.[17] However, the high degree of correlation between the PC and the PL yields of efficient solar cells made out of direct band gap materials, such as GaAs, is achieved only in the samples that have extremely low defect density. As the solution-processed perovskite solar cells have a high density of defects,[18,19] one does not expect high PC or PL from them. Thus, the role of heterogeneity and defects in the photovoltaic performance of the perovskite solar is being actively investigated.[20−25]

It has been shown that the structural inhomogeneity affects the PL yield and the spectra in $MAPbBr_3$. At least two different kinds of emissions at energies of about 2.28 and 2.34 eV have been identified at different regions of $MAPbBr_3$ crystals and thin films.[21,22,26,27] Although the impact of heterogeneity on the carrier lifetimes and the photocurrent response have been studied previously,[20,24,28−38] the role of the different emissive states on the yield of the PC has not been systematically investigated.

Here, we have used PCM and PLM to simultaneously measure the PC and the PL, respectively, from different regions in a $MAPbBr_3$ thin film solar cell. The PCM and the PLM allow us to correlate the PC and the PL from the different emissive states. Our results show that the major contribution to the PC is from the emissive state at 2.34 eV, whereas the other emissive state at 2.28 eV does not show a clear contribution. We have also used the PCM and the PLM to investigate the correlation between the PC and the PL from GaAs, which shows similar behavior to the PC response from the state at 2.34 eV in $MAPbBr_3$ thin film. Our results also indicate that the state at 2.28 eV is populated by energy transfer from the state at 2.34 eV. Therefore, although its presence helps to increase the PL, it decreases the yield of the PC.

2. EXPERIMENTAL METHODS

2.1. Device Fabrication. Fluorine-doped tin oxide (FTO)-coated glass substrate with a sheet resistivity of 7 Ωm was purchased from Sigma-Aldrich, USA. The substrates were etched with zinc powder (Merck, USA) and 2 M HCl for the intended electrode pattern (2 mm × 25 mm, $W \times L$). Patterned substrates were sequentially cleaned in detergent (5% labolene solution), deionized water, acetone, isopropyl alcohol (IPA), and ethanol for 20 min each in an ultrasonic bath. The

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