$CH_3NH_3Sn_xPb_{1-x}I_3$ ペロブスカイトの Sn 混合比 x による光吸収特性と電荷分離・再結合特性の変化

Optical absorption, charge separation and recombination dependences of the Sn mixing ratio x of $CH_3NH_3Sn_xPb_{1-x}I_3$ perovskite

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Organometal trihalide perovskite-based solid-state hybrid solar cells have attracted unexpected increasing interest because of the high efficiency (the record power conversion efficiency has been reported to be over 20%) and low cost for preparation.[1)] The high efficiency was thought to mainly originate from the strong optical absorption over a broader range (up to 800 nm for Pb) and longer lifetimes of photoexcited charge carriers (in the order of 10 ns – 100 ns) of the organometal trihalide perovskite absorbers. Recently, Hayase and coworkers have succeeded in harvesting energy in the NIR region by using Sn/Pb cocktail halide based perovskite ($CH_3NH_3Sn_xPb_{1-x}I_3$) materials covering up to 1060 nm and an efficiency of 4.18 % was achieved at x=0.5.[2)] They also found that the photovoltaic properties depended greatly on x. To improve the photovoltaic performance of Sn/Pb halide based perovskite solar cells, optical absorption property and charge separation and recombination mechanism, especially their dependences on x, are key factors and should be understood deeply.

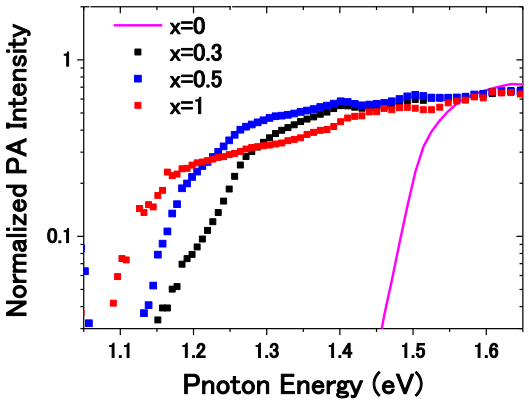

Fig. 1 Changes of the PA spectra of $CH_3NH_3Sn_xPb_{1-x}I_3$ as x increases from 0 to 1.

In this study, we prepared $CH_3NH_3Sn_xPb_{1-x}I_3$ on TiO_2 substrate using one step method,[2)] where x was changed from 0 to 1. We have investigated the optical absorption properties using photoacoustic spectroscopy (PAS) and charge separation and recombination dynamics using transient absorption (TA) techniques. Firstly, we found that the bandgap E_g of $CH_3NH_3Sn_xPb_{1-x}I_3$ red shifted systemically from 1.52 eV to 1.16 eV as x increased from 0 to 1 as shown in Fig. 1. On the other hand, the Urbach energy E_u increased from 22 meV to 54 meV as Sn was mixed in the Pb perovskite with x=0.3, but decreased to be 34 meV at x=0.5 and then increased again to be 43-45 meV for x>0.5. This result suggests that the crystalline property of the Sn/Pb cocktail perovskite is best at x=0.5. Secondly, we found that the recombination dynamics at the interface of TiO_2 and the perovskite interface depended on x. For the sample with x=0.5, the recombination time was much larger compared to those with other x. Relationship with these properties and the photovoltaic properties and the mechanism will be studied in detail.

References:1) http://www.nrel.gov/ncpv/; 2) Y. Ogomi et.al., J. Phys. Chem. Lett., 2014, 5, 1004.