CH₃NH₃SnₓPb₁₋ₓI₃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 CH₃NH₃SnₓPb₁₋ₓI₃ perovskite

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. 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 (CH₃NH₃SnₓPb₁₋ₓI₃) materials covering up to 1060 nm and an efficiency of 4.18% was achieved at x=0.5. 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.

In this study, we prepared CH₃NH₃SnₓPb₁₋ₓI₃ on TiO₂ substrate using one step method, 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 CH₃NH₃SnₓPb₁₋ₓI₃ red shifted systematically 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 TiO₂ 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.