ペロブスカイト $CsPbI_3$ の遅いホットキャリア緩和と P3HT への高速ホール移動

SLOW HOT CARRIER COOLING IN $CsPbI_3$ PEROVSKITE AND ULTRAFAST HOLE TRANSFER TO P3HT

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The interest in organic-inorganic hybrid solid-state solar cells based on organolead halide perovskite has increased over the past three years following the recently reported power conversion efficiency (PCE) of 20.1%. Organolead halide perovskites in the form of AMX_3 (A=organic molecule, e.g., CH_3NH_3(MA), B=Pb, X=Cl, Br, and I) can be simply crystallized from solution at low temperature (≤100 °C), which enables them to be utilized as light absorbing materials in various solar cells. Recently, inorganic caesium lead halide ($CsPbX_3$) perovskite have been reported in working solar cells [1-3], which suggests a potential for making even more stable inorganic perovskite solar cells than the hybrid organic-inorganic materials currently displaying the highest efficiencies. For future application such as hot-carrier solar cells, electrically pumped lasers, the understanding of photoexcited carrier dynamics, especially hot carrier cooling dynamics are very important. Recently, hot-carrier cooling dynamics in $MAPbI_3$ was reported [4].

In this study, for the first time, we studied ultrafast photoexcited carrier relaxation dynamics, especially hot carrier cooling, in CsPbI3 using a transient absorption (TA) spectroscopy. A clear bleach peak can be observed at the bandgap energies in the TA spectra. We found that during the hot carrier cooling process within a few picoseconds: (1) a sub-bandgap transient absorption signal arises at about 1.75 eV, which can be explained by bandgap renormalization and hot-carrier distribution; (2) the high-energy tail (1.85 eV-2 eV) of the TA spectrum is broadened, which is attributed to the presence of a quasi-equilibrium carrier distribution at a temperature Tc higher than the lattice temperature of the sample. Then we calculate the carrier temperature Tc versus time and the hot carrier cooling rate from the time-resolved TA spectra. It is very interesting that the hot carrier cooling is as slow as 20 ps for higher excitation densities, which is attributed to a 'phonon bottleneck'. Meanwhile, we found an ultrafast hot hole transfer from CsPbI3 to P3HT. Our findings indicate a potential of $CsPbI_3$ for application to hot carrier solar cells.

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