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Photocarrier dynamics in perovskite $CH_3NH_3PbI_3$ thin films revealed by time-resolved THz spectroscopy

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Solar cells developed based on lead halide perovskites have demonstrated an excellent performance for solar-to-electricity conversion with efficiencies greater than 20% presently [1]. Despite the rapidly increasing power conversion efficiency of perovskite-based solar cells, the physical nature of photocarriers in the lead halide perovskite absorber is still not clear yet [2,3]. Some studies reported the excitonic nature of the excitations that are derived primarily from large excitonic binding energies observed by steady-state absorption measurements [2]. In contrast, other studies using time-resolved optical spectroscopy have revealed the free-carrier nature of the photoexcitations [3]. The steady-state photoelectrical responses of photocarriers in complex photovoltaic materials might be very different with the time-resolved behaviors [4]. Therefore, utilizing both steady-state and time-resolved techniques is essentially necessary to gain thorough and precise understandings of photophysical properties of emerging solar-cell materials [5-6].

In this work, we clarify the intrinsic nature of photoexcitations in methylammonium lead iodide $CH_3NH_3PbI_3$ thin films using a combination of temperature-dependent transmission, steady-state and time-resolved photoluminescence (PL), transient absorption (TA) and THz transient absorption (THz-TA) spectroscopy. We discuss their excitonic and free-carrier recombination dynamics at low and high temperatures.

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