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Free carrier dynamics in Cu_2ZnSnS_4 single crystals investigated by optical pump – THz probe spectroscopy

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As being one of the most promising materials for next generation solar cells, Cu_2ZnSnS_4 (CZTS) have recently been studied intensively using both optical and electrical techniques [1-3]. In order to enhance the low efficiencies of CZTS-based solar cells [4], it is important to find out some feasible ways to improve the open circuit voltage V_{OC} and/or the photocurrent. To date, most efforts have been devoted to understand the physics relating to defects, which are believed to limit the V_{OC} of CZTS-based solar cells. On the other hand, free carrier dynamics in CZTS, which strongly correlate to the photocurrent of CZTS-based solar cells, remain unclear.

Here, we report free carrier dynamics in CZTS single crystals at room temperature (RT) investigated by means of optical pump – THz probe spectroscopy. The transient reflectivity (TR) change detected by the 0.5-2.5 THz probe reflects the time-dependent number of free carriers existing in CZTS after photoexcitation. A typical TR decay profile is composed of three exponentials with the time constants of several tens picoseconds, a few hundred picoseconds, and nanoseconds. Basing on the comparisons with results obtained from the conventional pump – optical probe measurements and on the fact that the fastest decay component becomes slower with decreasing the excitation photon energy, we assign the fastest decay time to the trapping time of free carriers to the shallow low-density-of-state band tails. Moreover, we note that time-resolved photoluminescence measurements have revealed a decay time of nanoseconds at RT [1,3]. The long-lived nanosecond decay component of the TR decay profile, thus, is suggested to relate to the thermally released processes of the localized carriers. The details of carrier recombination dynamics will be discussed.

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