Near band-edge optical responses in Cu$_2$ZnSnS$_4$ single crystals
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Recently, Cu$_2$ZnSnS$_4$ (CZTS) is attracting considerable attentions as a less toxic and low-cost absorber material for next generation solar cells [1-3]. However, the best solar energy conversion efficiency of CZTS-based solar cells [1] is still much lower than conversion efficiencies of other thin-film solar cells [4]. To improve the power conversion efficiency of CZTS-based solar cells, it is important to obtain more detailed information on the fundamental physical properties of CZTS material itself.

In this report, we clarify the near band-edge optical responses, and photocarrier localization and recombination dynamics in CZTS single crystals at room temperature (RT) [3]. The band-gap energy was identified consistently by various means, including photoluminescence excitation, photocurrent, and transient reflectivity spectroscopy, and estimated to be approximately 1.58 eV at RT. The large-density band tail states formed below the band edge determine primarily the optical dynamical responses in CZTS. It was found that the photogenerated carriers in CZTS are quickly localized to trap states within a few tens of picoseconds after photoexcitation. The photoluminescence (PL) band was assigned to radiative recombination of photogenerated carriers localized in the band tails. The time-resolved PL measurements revealed the power-law-dependent recombination dynamics, indicating the occurrence of multiple carrier trapping processes near the band edge. The fast photocarrier localization into the band tail states will affect the conversion efficiency of CZTS-based solar cells. Our kinematic studies, therefore, provide essential information relevant to the further improvement of the power conversion efficiency of CZTS-based solar cells.

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