 Photocarrier dynamics in Cu₂ZnSn(SₓSe_{1-x})₄ single crystals: Composition dependence

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The emerging photovoltaic material Cu₂ZnSn(SₓSe_{1-x})₄ (CZTSₓSe₁₋ₓ) has recently attracted world-wide attention due to its exceptional characteristics for solar energy conversion and earth-abundant composition [1]. Up to date, the highest power conversion efficiencies (PCE) of 8.4 % [2] and 11.6 % [3] were realized for the CZTS-based and CZTSe-based solar cells, respectively. Meanwhile, a world-record 12.6 % PCE [4] was achieved for the S-Se-mixture absorber CZTSSe with the band-gap energy of 1.13 eV that, in fact, is not optimal for the light-electricity conversion in single-junction solar cells. These maximum PCEs of CZTSSe-based solar cells are still rather smaller than those of CIGS-based ones [5] and far below the Shockley-Queisser limit (about 31 %). It is plausible that there are other physical origins determining and limiting the PCE of CZTSSe-based solar cells in addition to the band-gap energy of the absorber. In order to clarify comprehensively these physical origins, a combination of various kinds of optical spectroscopy is particularly useful [6].

In this work, we investigated the composition dependence of optoelectronic properties of CZTSₓSe₁₋ₓ single crystals [7] using photoluminescence excitation, time-resolved photoluminescence, optical pump–THz probe and optical pump–optical probe transient reflectivity spectroscopy. The band-gap energy increases almost linearly with the composition of S. In addition, we found out that the photocarrier recombination dynamics depend strongly on the composition of CZTSₓSe₁₋ₓ crystals, and this composition-dependence gives rise to the different optoelectronic properties of CZTSₓSe₁₋ₓ single crystals. Our results reveal the physics behind the composition-dependent and low PCEs of CZTSSe-based solar cells, and these results are expected to be helpful for directing improvements of CZTSSe-based solar cells.

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