Ultra-high average figure of merit in synergistic band engineered Sn1-xNaxSe0.9S0.1 single crystals

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Abstract

Thermal-electricity conversion is one of the most promising routes to harvest heat and convert it as easily storable and deliverable electric energy. Significant progress has been made since the discovery of Seebeck effect in 1821, particularly, the figure of merit $zT$ approached a record high value of 2.6 in 2014. However, for thermoelectric devices, high average $zT$ values ($zT_{ave}$) over the operating temperature range is more important as it is directly related to the conversion efficiency ($g$). Approaching highly stable and repeatable ultra-high $zT_{ave}$ for Te-free materials has been historically challenging over the past century though exciting progress with $zT_{ave}$ well above 1.10 was made recently. Here, through synergistic band engineering strategy for single crystalline SnSe, we report a series of record high $zT_{ave}$ over a wide temperature range, approaching 1.60 in the range from 300 K to 923 K in Na-doped SnSe0.9S0.1 solid solution single crystals, with the maximum $zT$ of 2.3 at 773 K. These ultra-high thermoelectric performance derive from the new multiple valence band extrema near the band edges in SnSe0.9S0.1 and the shift of Fermi level towards the multi-valley bands through Na doping which introduce additional carrier pockets to attend electrical transport. These effects result in an optimized ultrahigh power factor exceeding 4.0 mWm⁻¹K⁻² in Sn0.97Na0.03Se0.9S0.1 single crystals. Combined with the extremely lowered thermal conductivity attributed from the intrinsic anharmonicity and point defect phonon scattering, the series of ultra-high $zT_{ave}$ and a record high calculated conversion efficiency of 21% over a wide temperature range are approached.

Keywords: Average $zT$, carrier pockets, band structure, lattice thermal conductivity

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