Supporting Information

Construction of Nano-Lamellar Expressways and Multidimensional Defects Realizes the Decoupling of Carrier-Phonon Transport in $BiSbSe_{1.25}Te_{1.75}$

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Keywords: Identical chemical composition, n-type $BiSbSe_{1.25}Te_{1.75}$, Hot deformation, Synergistic optimization, Decouple

Experimental details

Material synthesis: High-purity elemental chunks of Bi, Sb, Se and Te with >99.999% were weighed based on the stoichiometric ratio of $BiSbSe_{1.25}Te_{1.75}$ and sealed in a quartz tube of 10^{-3} Pa for the solid-phase reaction. The tube was placed into box furnace, then slowly heated up to 1273 K and soaked at 1273 K for 6 h. The obtained ingot was ground into powders using an agate mortar, and then hot pressed at 1273 K 80Mpa and for 30 minutes in a 15 mm-diameter graphite die, producing the initial bulk samples named as HD0. Subsequently, a hot deformation process was performed by repressing the HD0 samples in a larger graphite die with an inner diameter of 20 mm at 823 K for 30 min using the same stress. Finally, disk-shaped samples of 20 mm called HDx were obtained and x stand for hot deformation times.

Structural Characterization: The phase structure of the samples was examined over the 2θ range of 10-80º using an X-ray diffractometer (XRD, Empyrean, PANalytical, Netherlands) and the texture evolution were carried out by the pole figure measurement using the Schulz back-reflection method (XRD, D8 Advance, Bruker, Germany) The fractographs were determined by scanning electron microscopy (SEM, IT800-SHL, JEOL, Japan). The atomic-scale microstructures were characterized by scanning transmission electron microscopy (STEM, FEI Titan G2 60-300, FEI, USA). The specimen for STEM analysis was prepared by using a focused ion beam (FIB, Helios G4 UX, FEI, USA).

Thermoelectric transport properties: The electrical conductivity and Seebeck coefficient were measured simultaneously on a commercial equipment (LSR-3, Linseis, Germany) under a high-purity helium atmosphere at a temperature range of 300-525 K. And Seebeck coefficients for BSTS-I, BSTS-PB5 and BSTS-PB220 samples were measured by using the Portable Thermoelectric Meter-3 (PTM-3, Joule Yacht) at 300 K, respectively. To ensure the accuracy, each sample was measured ten times at various locations. The thermal diffusivity (D) and specific heat capacity (C_p) of $BiSbTe_{1.5}Se_{1.5}$ samples were determined by laser flash apparatus ((LFA-457, Netzsch, Germany) and differential scanning calorimeter (DSC 404 F3, Netzsch, Germany) in argon flow, respectively. The carrier concentration and carrier mobility were measured under a reversible magnetic field (0.9 T) by the Van der Pauw method by using a Hall measurement system (Lake Shore 8400 Series, Model 8404, USA) at 323 K.The volume density (ρ) was measured using the Archimedes method and the thermal conductivity (κ) was calculated according to the equation $\kappa = DC_p\rho$.

The preferred orientations

The preferred orientations F of the (00l) planes for HDx bulk $BiSbSe_{1.25}Te_{1.75}$ samples are evaluated based on the Lotgering factor[1-3] via analysis of the XRD pattern according to the following equations (1) and (2):

$$F = (P - P_0) / (1 - P_0) \qquad \text{(S1)}$$

$$P = \sum I\,(00l) / \sum I\,(hkl) \qquad \text{(S2)}$$

The single parabolic band (SPB) model

The single parabolic band (SPB) model was adopted to calculate the relationship between Seebeck coefficient and carrier concentrations at 323 K.[4] The Seebeck coefficient and carrier concentration of a degenerate semiconductor can be expressed as followed equations:

$$S = \frac{k_B}{e}\left(\frac{(2+\lambda)F_{1+\lambda}(\eta)}{(1+\lambda)F_{\lambda}(\eta)} - \eta\right) \tag{S3}$$

(S4)

$$n_H = 4\pi\left(\frac{2m^* kT}{h^2}\right)^{\frac{3}{2}} F_{\frac{1}{2}}(\eta) \tag{S5}$$

where k_B, e, η, h, and λ is the Boltzmann constant, electron charge, chemical potential, Planck constant and phonon scattering factor ($\lambda = 0$). $F_j(\eta)$ is the Fermi integral, with x being the reduced carrier energy. The reduced Fermi energy η is determined based on the fitting of the Seebeck coefficients.

The band effective masses m^* in this work at 323 K is obtained by fitting experimental data into Equations S1, S2 and S3. The Lorenz number L can be evaluated by the following Equation S6,

$$L = \frac{k^2}{e^2}\frac{3F_0(\eta)F_2(\eta) - 2F_1(\eta)^2}{F_0(\eta)^2} \tag{S6}$$

Weighted mobility

The weighted mobility μ_W at 323 K was calculated using the measured data of S and the σ according to the following formula[5]:

$$\mu_W = \frac{3h^3\sigma}{8\pi e\left(2m_e k_B T\right)^{3/2}}\left[\frac{\exp\left[\frac{|S|}{k_B/e} - 2\right]}{1 + \exp\left[-5\left(\frac{|S|}{k_B/e} - 1\right)\right]} + \frac{\frac{3}{\pi^2}\frac{|S|}{k_B/e}}{1 + \exp\left[5\left(\frac{|S|}{k_B/e} - 1\right)\right]}\right] \tag{S7}$$

The dimensionless quality factor *B*

The dimensionless quality factor B is calculated using the following relationship[6]:

$$B = 9\frac{\mu_{\mathrm{w}}}{\kappa_{\mathrm{L}}}(\frac{\mathrm{T}}{300})^{5/2} \tag{S8}$$

Table S1. Comparisons of *ZT* at low temperature for HD3 and other state-of-the-art systems.

Material	Operating temperature (K)	Type	*ZT*
$Mg_{3.2}Bi_{1.298}Sb_{0.7}Te_{0.002}$[7]	350	n	0.9
$Mg_{2.475}Zn_{0.5}Li_{0.025}Sb_2$[8]	350	p	0.1
Bi_2Te_3[9]	300	n	0.65
$(Bi_{0.5}Sb_{1.5}Te_3)_{0.97}(MgB_2)_{0.03}$[10]	300	p	1.2
$AgPb_{10}SbTe_{12}$[11]	300	n	0.37
$Ag_{0.83}Sb_{1.06}Te_2$[12]	300	p	0.32
HD3	323	n	0.39

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