Electronic supplementary information

Complementary effect of co-doping aliovalent elements Bi and Sb in self-compensated SnTe-based thermoelectric materials

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Table S1 Measured densities for the $Sn_{1.03-x-y}Bi_xSb_yTe$ compositions

Sample	**Density (g/cm^3)**
SnTe	6.4239
$Sn_{1.03}Te$	6.4350
$SnBi_{0.03}Te$	6.4987
$Sn_{0.94}Bi_{0.03}Sb_{0.06}Te$	6.4624
$Sn_{0.92}Bi_{0.03}Sb_{0.08}Te$	6.4553
$Sn_{0.90}Bi_{0.03}Sb_{0.10}Te$	6.4631
$Sn_{0.88}Bi_{0.03}Sb_{0.12}Te$	6.4660
$Sn_{0.86}Bi_{0.03}Sb_{0.14}Te$	6.4449

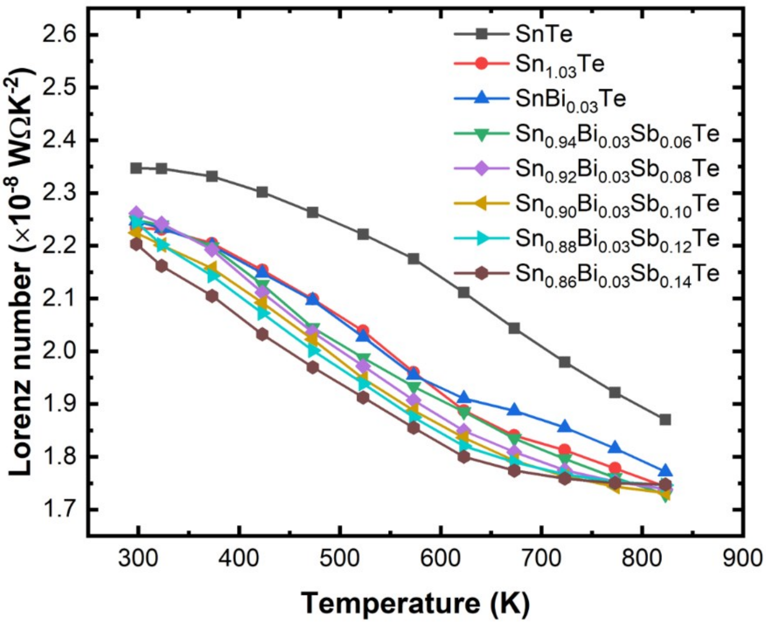

Fig. S1. Temperature dependent Lorenz number for the $Sn_{1.03-x-y}Bi_xSb_yTe$ compositions.

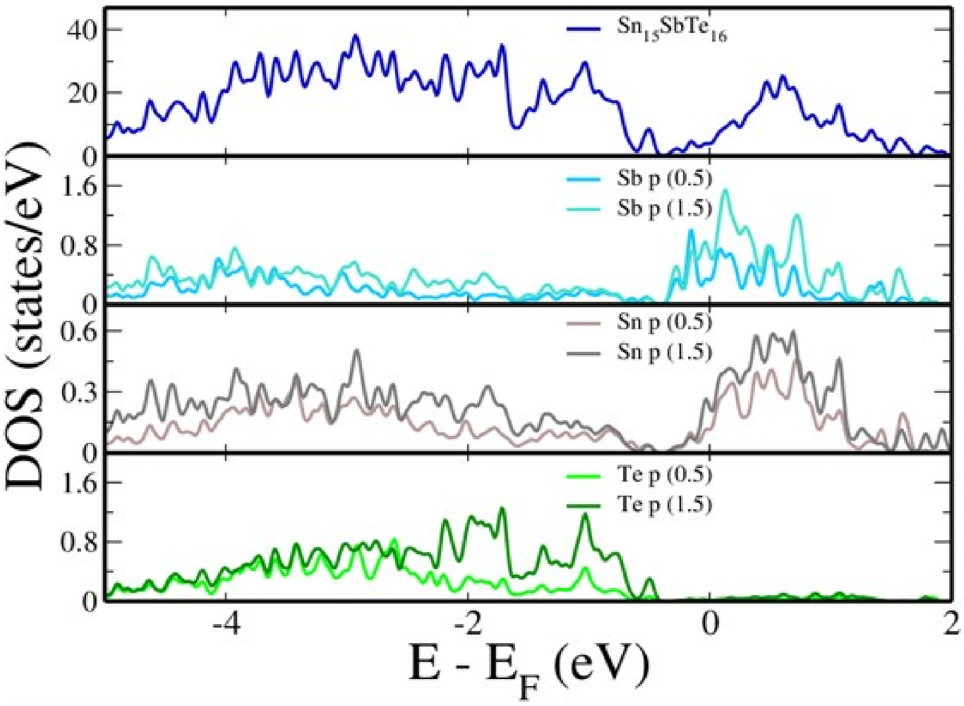

Fig. S2. pDOS of $Sn_{15}SbTe_{16}$. Energies are shifted with respect to Fermi level which is set to zero. Resonance states appear in the form of increased DOS near the Fermi level.

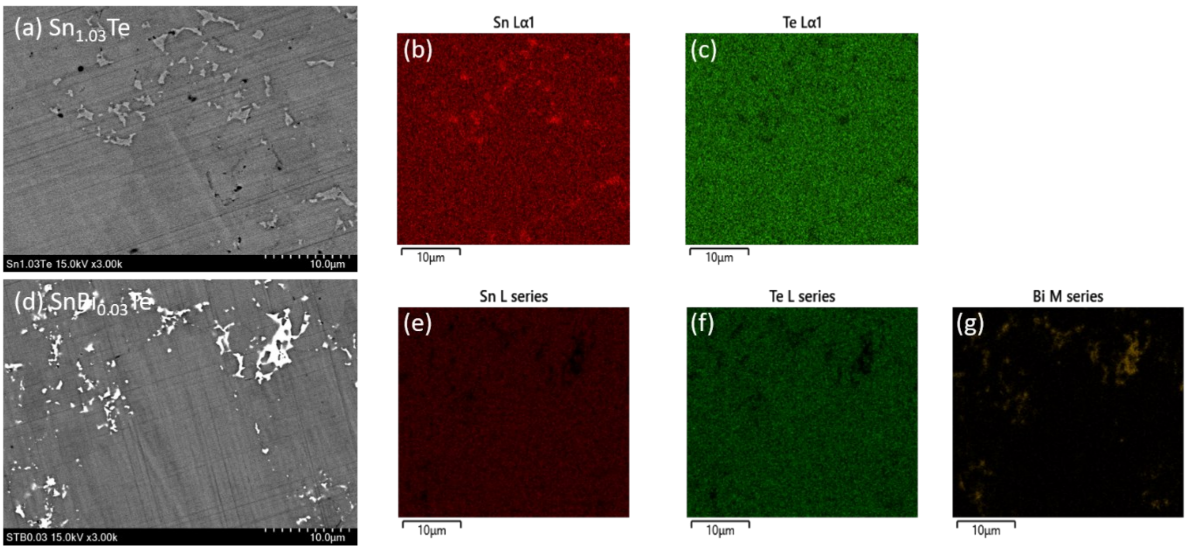

Fig. S3. SEM BSE images for the (a) $Sn_{1.03}Te$ and (d) $SnBi_{0.03}Te$ compositions and respective EDS mapping.

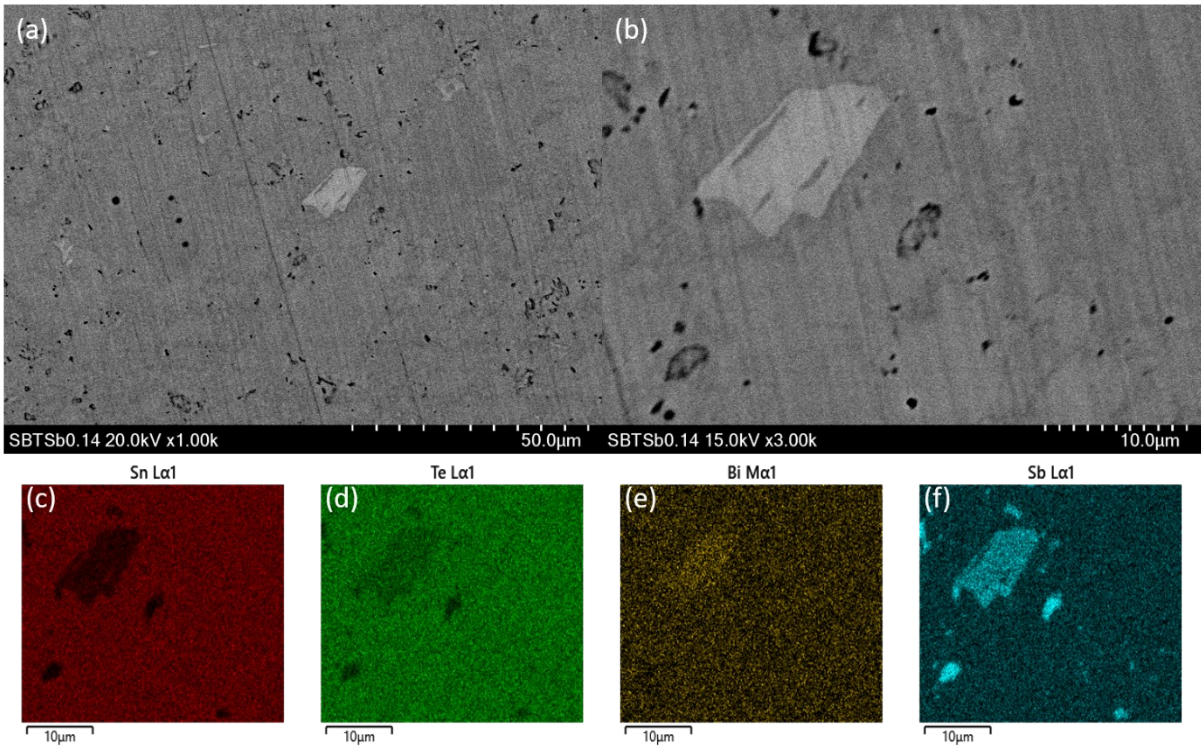

Fig. S4. SEM BSE images for the $Sn_{0.86}Bi_{0.03}Sb_{0.14}Te$ composition and respective EDS mapping.

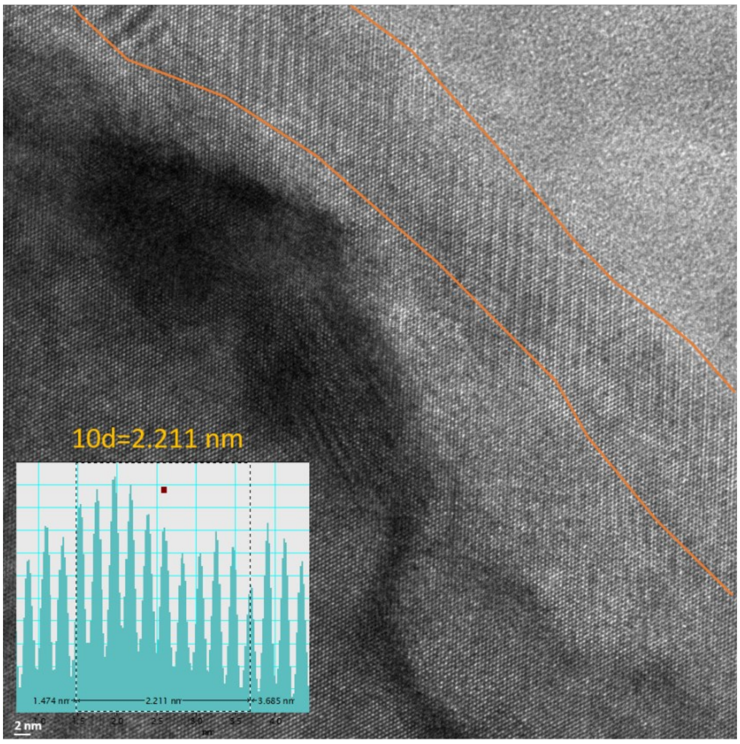

Fig. S5. HRTEM image for the $Sn_{0.86}Bi_{0.03}Sb_{0.14}Te$ composition showing lattice spacing for the SnTe (220) plane and a layered structuring bound by the orange lines.

Lattice parameter calculation

The lattice parameter for all compositions shown in this were extracted from the XRD data.

From Bragg's law:

$$\lambda = 2dsin\theta \tag{S1}$$

Where λ is the wavelength for the x-ray radiation (1.5406 Å), d is the inter planar distance and θ is the diffraction angle obtained from XRD as $2\theta/2$.

For a cubic crystal system:

$$\frac{1}{d^2} = \frac{(h^2 + k^2 + l^2)}{a^2} \tag{S2}$$

Where d is the previously calculated inter planar distance from XRD data, *hkl* are the Miller indices of a diffraction plane and *a* is the lattice parameter to be determined.

Calculation of sound velocity

The plane acoustic longitudinal (V_l) and transverse (V_t) velocities of all compositions were estimated in the [100], [110], and [111] crystal orientations using elastic constants of SnTe (C_{ij})[1] and the measured density (ρ) as explained below.[2]

For wave propagating in the [100] direction:

$$V_l = \sqrt{C_{11}/\rho} \tag{S3}$$

$$V_t = \sqrt{C_{44}/\rho} \tag{S4}$$

For wave propagating in the [110] direction:

$$V_l = \sqrt{(C_{11} + C_{12} + C_{44})/2\rho} \tag{S5}$$

$$V_{t1} = \sqrt{(C_{11} - C_{12})/2\rho} \tag{S6}$$

$$V_{t2} = \sqrt{C_{44}/\rho} \tag{S7}$$

For wave propagating in the [111] direction:

$$V_l = \sqrt{(C_{11} + 2C_{12} + 4C_{44})/3\rho} \tag{S8}$$

$$V_t = \sqrt{(C_{11} - C_{12} + C_{44})/3\rho} \tag{S9}$$

A summary of the plane acoustic wave velocities is shown in Table S2.

Table S2 Plane acoustic wave velocities in m/s for the $Sn_{1.03-x-y}Bi_xSb_yTe$ compositions in different crystal orientation directions

	[100]		[110]			[111]	
Composition	V_l	V_t	V_l	V_{t1}	V_{t2}	V_l	V_t
SnTe	4185	1351	3202	2859	1351	3008	2461
$Sn_{1.03}Te$	4181	1350	3199	2856	1350	3005	2459
$SnBi_{0.03}Te$	4161	1343	3183	2842	1343	2991	2447
$Sn_{0.94}Bi_{0.03}Sb_{0.06}Te$	4172	1347	3192	2850	1347	2999	2454
$Sn_{0.92}Bi_{0.03}Sb_{0.08}Te$	4175	1347	3194	2852	1347	3001	2455
$Sn_{0.90}Bi_{0.03}Sb_{0.10}Te$	4172	1347	3192	2850	1347	2999	2454
$Sn_{0.88}Bi_{0.03}Sb_{0.12}Te$	4171	1346	3191	2849	1346	2998	2453
$Sn_{0.86}Bi_{0.03}Sb_{0.14}Te$	4178	1349	3197	2854	1349	3003	2457

For an isotropic cubic structure, the longitudinal and transverse sound velocities were estimated from the calculated bulk modulus (B) and shear modulus (G) using the below relations.[2]

$$V_l = \sqrt{(B + 4G/3)/\rho} \tag{S10}$$

$$V_t = \sqrt{G/\rho} \tag{S11}$$

Where both B and G are obtained by solving equations S12-15.[2]

$$B = (B_R + B_v)/2 \tag{S12}$$

$$G = (G_R + B_R)/2 \tag{S13}$$

$$B_V = B_R = \frac{1}{3}(C_{11} + 2C_{12}) \tag{S14}$$

$$G_R = \frac{5(C_{11} - C_{12})C_{44}}{4C_{44} + 3(C_{11} - C_{12})} \tag{S15}$$

The average sound velocity, V_m was then obtained using equation S16.[2,3]

$$V_m = \left[\frac{1}{3}\left(\frac{2}{V_t^3} + \frac{1}{V_l^3}\right)\right]^{-1/3} \quad \text{(S16)}$$

Table S3 Longitudinal, transverse, and average sound velocities in m/s for the $Sn_{1.03-x-y}Bi_xSb_yTe$ compositions

Composition	V_l	V_t	V_m
SnTe	3576	2152	2380
$Sn_{1.03}Te$	3573	2150	2378
$SnBi_{0.03}Te$	3556	2140	2366
$Sn_{0.94}Bi_{0.03}Sb_{0.06}Te$	3566	2146	2373
$Sn_{0.92}Bi_{0.03}Sb_{0.08}Te$	3568	2147	2374
$Sn_{0.90}Bi_{0.03}Sb_{0.10}Te$	3566	2146	2373
$Sn_{0.88}Bi_{0.03}Sb_{0.12}Te$	3565	2145	2372
$Sn_{0.86}Bi_{0.03}Sb_{0.14}Te$	3571	2149	2376

References

1 A. G. Beattie, *J. Appl. Phys.*, 1969, **40**, 4818–4821.

2 Z. Jia, P. Wang and W. Smith, *Open Phys.*, 2018, **16**, 826–831.

3 Z. Chen, Q. Sun, F. Zhang, J. Mao, Y. Chen, M. Li, Z. G. Chen and R. Ang, *Mater. Today Phys.*, 2021, **17**, 100340.