## **Supporting information:**

## Synergistically optimized electrical and thermal transport properties in SnTe *via* alloying high-solubility MnTe

Haijun Wu,<sup>1,2,#</sup> Cheng Chang,<sup>1</sup> Dan Feng,<sup>2</sup> Yu Xiao,<sup>1</sup> Xiao Zhang,<sup>1</sup> Yanling Pei,<sup>1</sup> Lei Zheng,<sup>1</sup> Di Wu,<sup>2</sup> Shengkai Gong,<sup>1</sup> Yue Chen,<sup>3</sup> Jiaqing He,<sup>2\*</sup> Mercouri G. Kanatzidis,<sup>4\*</sup> Li-Dong Zhao<sup>1\*</sup>

<sup>1</sup>School of Materials Science and Engineering, Beihang University, Beijing, 100191, China
<sup>2</sup>Department of Physics, South University of Science and Technology of China, Shenzhen, 518055, China
<sup>3</sup>Department of Mechanical Engineering, The University of Hong Kong, Pokfulam Road, Hong Kong SAR China
<sup>4</sup>Department of Chemistry, Northwestern University, Evanston, Illinois 60208, United States
<sup>#</sup>Present addresses: Department of Materials Science and Engineering, National University of Singapore, 7 Engineering Drive 1, Singapore 117574, Singapore.

\*Corresponding authors: zhaolidong@buaa.edu.cn; he.jq@sustc.edu.cn; mkanatzidis@northwestern.edu

Table S1. Densities (*d*, g/cm<sup>3</sup>) of all samples  $Sn_{1-x}Mn_xTe$  (x=0, 1%, 3%, 5%, 7%, 9%, 11%, 13%, 15%, 17%, 19%, 20%, 25% and 50%, in mole ratio) investigated in this study.

x	0	1%	3%	5%	7%	9%	11%	13%	15%	17%	19%	20%	25%	50%
d	6.26	6.274	6.212	6.161	5.819	6.075	5.953	5.943	5.63	5.86	5.805	5.811	6.126	5.763



Figure S1. Carrier concentration and mobility in SnTe sample as a function of alloying MnTe fraction at room temperature.



Figure S2. Room temperature hole concentration  $(N_P)$  as a function of Mn, Cd, Mg and Hg fraction alloying in SnTe.



Figure S3. Temperature dependent (a) Heat capacity  $C_p$ , (b) thermal diffusivity D, (c) electronic thermal conductivity, (d) Calculated Lorenz number L for Sn<sub>1-x</sub>Mn<sub>x</sub>Te.



Figure S4. Medium-magnification TEM image of SnTe alloyed with 19% MnTe .