

Supporting Information:

**Realizing high performance n-type PbTe via synergistically
optimizing effective mass, carrier mobility and suppressing bipolar
thermal conductivity**

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Experimental details:

Sample preparation: Raw materials, Pb particle (99.999%, Aladdin element, China), Te chunk (99.999%, Aladdin element, China), Mn powder (99.99%, Aladdin element, China), Sb particle (99.999%, Aladdin element, China), are loaded into carbon-coated silica tubes under an N₂-filled glovebox with nominal compositions, flame-sealed at a residual pressure below $\sim 10^{-4}$ Torr, slowly heated to 1323 K with 50K/h and maintained at this temperature for 10 h. The furnace shuts down and cools to room temperature. The obtained ingots are ground into powder and sintered by spark plasma sintering (SPS-211Lx) at 873 K for 15 min under an axial compressive stress of 50 MPa, resulting in highly densified disk-shaped samples, of which the density are $> 95\%$ of theoretical density.

Structural characterization: The powder diffraction patterns are obtained with Cu K α ($\lambda=1.5418$ Å) radiation in a reflection geometry on a diffractometer operating at 40 KV and 20 mA and equipped with a position-sensitive detector. (Scanning) transmission electron microscopy (STEM and TEM) and energy dispersive X-ray spectroscopy (EDS) studies were conducted using a JEOL ARM200F atomic resolution analytical electron microscope installed in the National University of Singapore equipped with a cold field-emission gun, a new ASCOR 5th order aberration corrector and Gatan OneView camera and an Oxford X-Max 100TLE X-ray detector. Some TEM observations were performed under 300 KV for the microstructure on the FEI Tecnai F30 and SEM observations were performed on Zeiss Merlin.

Hall measurements: Hall coefficient (R_H) is measured under a reversible magnetic field (0.8 T) by the Van der Pauw method by using the Hall measurement system (Lake Shore 8400 Series, Model 8404, USA) at room temperature. Carrier density (n_H) is obtained by $n_H=1/(e \cdot R_H)$, and carrier mobility (μ_H) is calculated using the relationship $\mu_H=\sigma \cdot R_H$ with the electrical conductivity σ .

Optical absorption measurements: The optical band gap was measured using Infrared Diffuse Reflection method in Fourier Transform Infrared Spectrometer

(IRAffinity-1S), and the temperature-dependent band gap from 300-673K was tested in a high-temperature accessory. A KBr standard was used as a reference.

Elastic properties: The longitudinal (v_l) and transverse (v_t) sound velocities are measured using an ultrasonic instrument (Ultrasonic Pulser/Receiver Model 5058 PR, Olympus, USA). The average sound velocity (v_a), Poisson ratio (ν_p), Debye temperature (θ_D) and Grüneisen parameter (γ) are then calculated using the measured velocities.

Electrical transport properties: The obtained spark plasma sintering (SPS) pellets are cut into bars with dimensions $12 \times 3 \times 3 \text{ mm}^3$ for simultaneous measurement of the Seebeck coefficient and electrical conductivity using Cryoall CTA and Ulvac Riko ZEM-3 instrument under a low-pressure helium atmosphere from room temperature to 873K. The uncertainty of the Seebeck coefficient and electrical conductivity measurement is 5%.

Thermal conductivity: The SPS-processed pellets are cut and polished into a disk shape with a sized of $\Phi 6 \text{ mm} \times 2 \text{ mm}$ for thermal diffusivity (D) measurements. The samples are coated with a thin layer of graphite to minimize errors from the emissivity method in a Netzsch LFA457. The density (ρ) is determined using the dimensions and mass of the sample and reconfirmed by Archimedes method. The specific heat capacity (C_p) is estimated with Dulong-Petit law. The thermal diffusivity data is analyzed using the Cowan model with pulse correction. The uncertainty of the thermal conductivity is estimated to be within 8%, considering all the uncertainties from D , ρ and C_p . The combined uncertainty for all measurements involved in the calculation of ZT is less than 20%.

Density functional theory (DFT) calculations: In this work, density functional theory (DFT) calculations were conducted using the Perdew-Burke-Ernzerhof (PBE) functional of the generalized gradient approximation (GGA)¹ as implemented in the Vienna *ab initio* simulation package (VASP)². We constructed a $3 \times 3 \times 3$ supercell of PbTe containing 54 atoms and substituted one or two Pb atoms with Mn to simulate the $\text{Pb}_{26}\text{Mn}_1\text{Te}_{27}$ and $\text{Pb}_{25}\text{Mn}_2\text{Te}_{27}$ alloys. For $\text{Pb}_{25}\text{Mn}_2\text{Te}_{27}$, we have considered all four atomic configurations, among which the one with the lowest energy was chosen

for further band structure calculation. A plane wave cutoff energy of 350 eV and a k-point mesh density of $2\pi \times 0.03 \text{ \AA}^{-1}$ using the Γ -centered Monkhorst-Pack scheme³ were adopted for all DFT calculations. The structures were fully relaxed until the energy converged within 10^{-5} eV before non-self-consistent band structure calculations. The spin-orbit coupling (SOC) effect was included in our calculations.

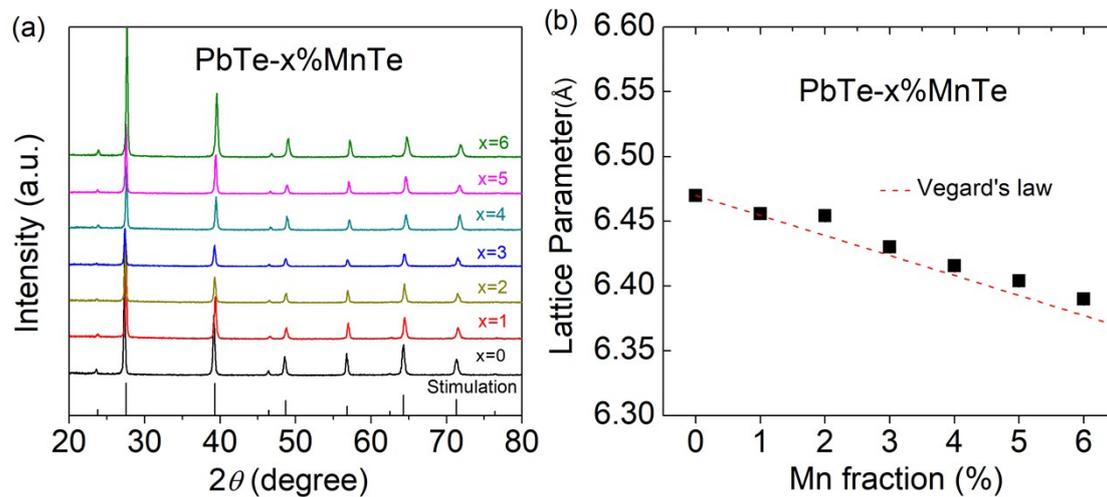


Figure S1. (a) XRD pattern and (b) lattice parameter of PbTe-MnTe.

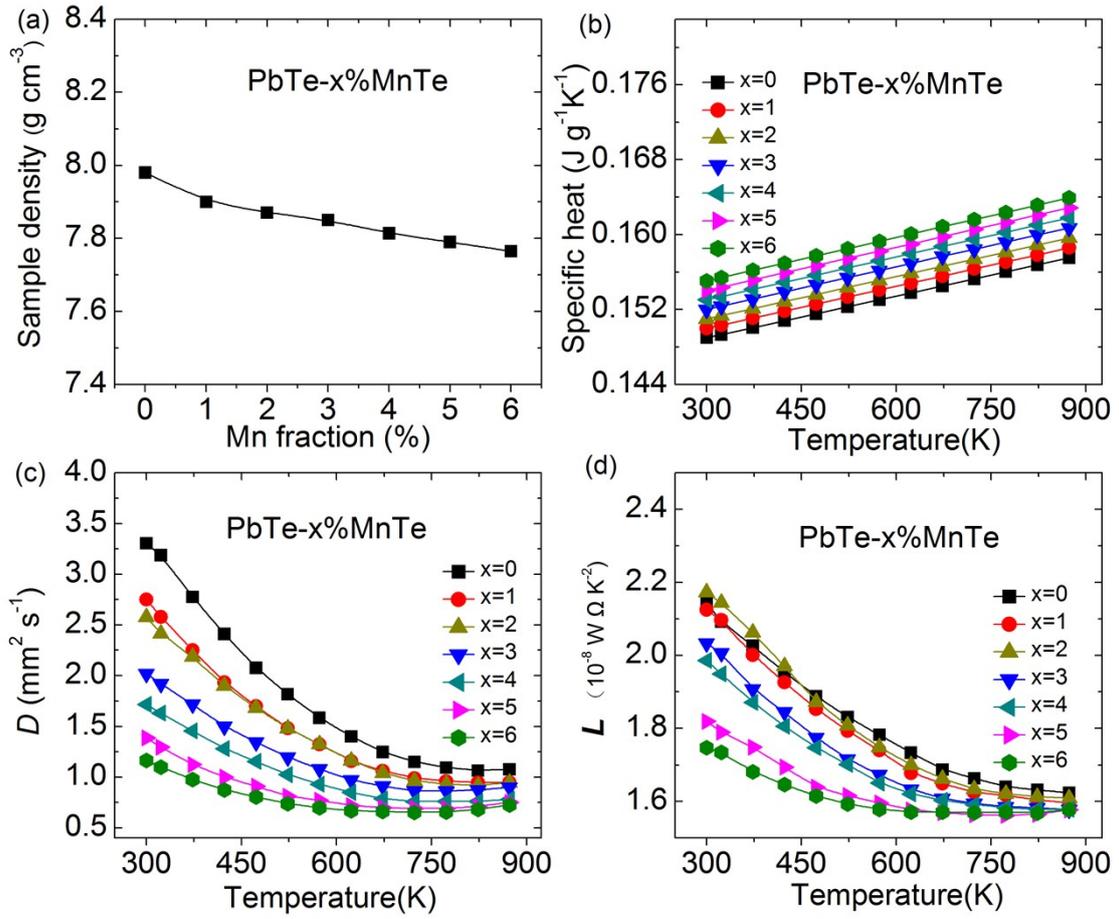


Figure S2. (a) sample density, (b) specific heat, (c) thermal diffusivity, and (d) Lorenz number of PbTe-MnTe.

Table S1. Related parameters in PbTe-x%MnTe (x=0-6), including longitudinal (v_l), transverse (v_t) and average (v_a) sound velocity, Debye temperature (θ_D) and Grüneisen parameter (γ).

Samples	v_l (m/s)	v_t (m/s)	v_a (m/s)	θ_D (K)	γ
PbTe	2807	1559	1736	160	1.64
PbTe-1%MnTe	2801	1565	1742	161	1.62
PbTe-2%MnTe	2782	1543	1718	159	1.64
PbTe-3%MnTe	2792	1548	1724	160	1.65
PbTe-4%MnTe	2798	1540	1716	159	1.67
PbTe-5%MnTe	2794	1543	1719	160	1.66
PbTe-6%MnTe	2797	1540	1717	160	1.67

Reference

1. J. P. Perdew, K. Burke and M. Ernzerhof, *Phys. Rev. Lett.*, 1996, **77**, 3865-3868.
2. G. Kresse and J. Furthmüller, *Phys. Rev. B*, 1996, **54**, 11169-11186.
3. H. J. Monkhorst and J. D. Pack, *Phys. Rev. B*, 1976, **13**, 5188-5192.