

Supplementary Information for

**Manipulation of Rashba splitting and thermoelectric performance of MTe (M=Ge, Sn, Pb) via Te off-centering distortion**

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## S.1 RESULTS AND DISCUSSIONS

### S.1.1 Te off-centering influences the electronic band structure

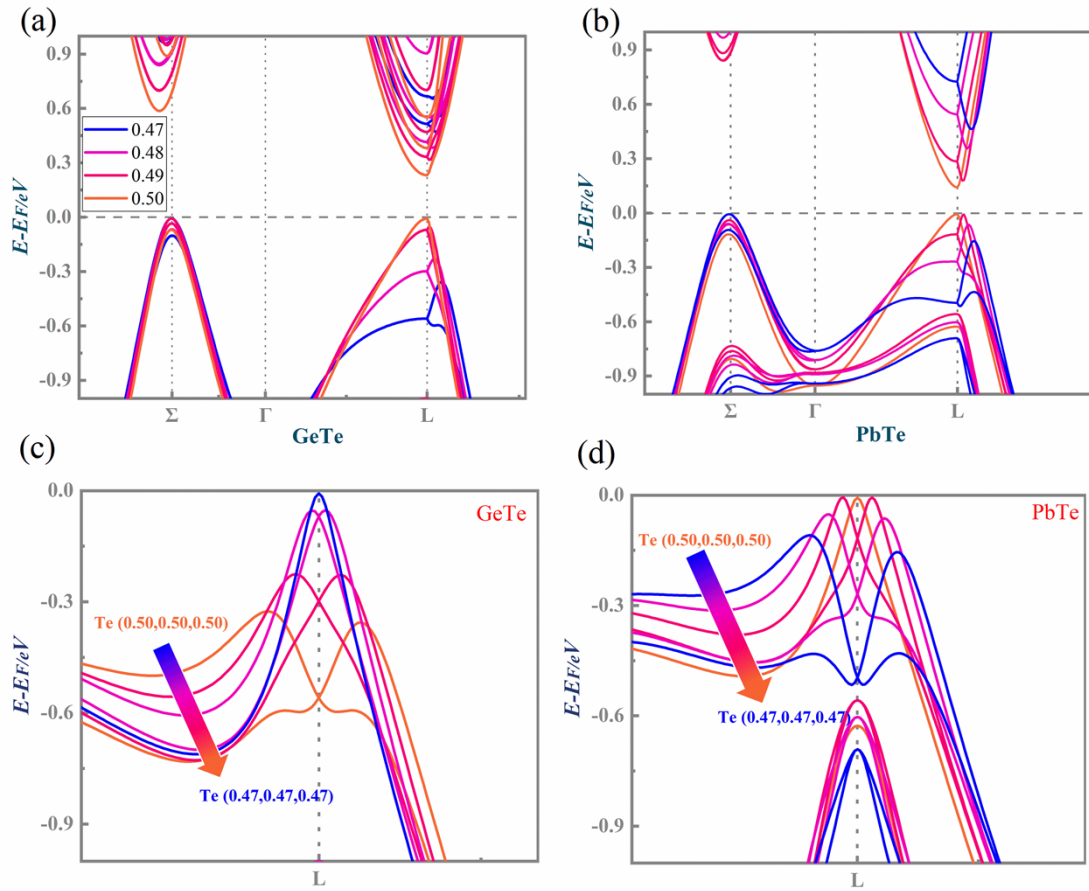


Fig.S1. The enlarged view of the Valence bands(VBs) near the L point and  $\Sigma$  point of (a) GeTe and

(b)PbTe. The amplifying valence bands (VBs) at the L point of (c)GeTe and (d)PbTe to highlight the

spin-dependent band splitting subject to Rashba effect,

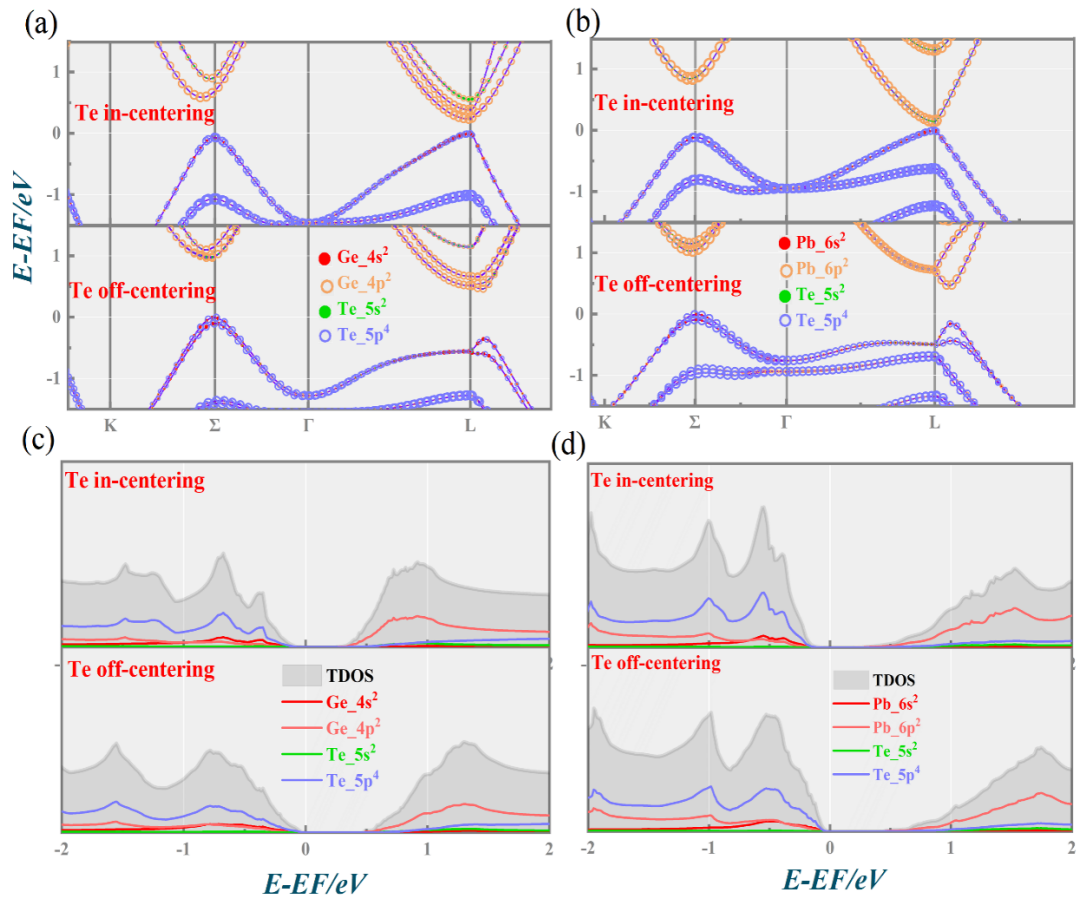


Fig.S2. The calculated PBAND (size of each circle represents atomic orbital weight) of (a) GeTe and (b) PbTe, and PDOS of (c) GeTe and (d)PbTe when Te atomic coordinates locate at (0.47,0.47,0.47) and (0.50,0.50,0.50)

### S.1.2 Te off-centering behavior influences the phonon dispersion

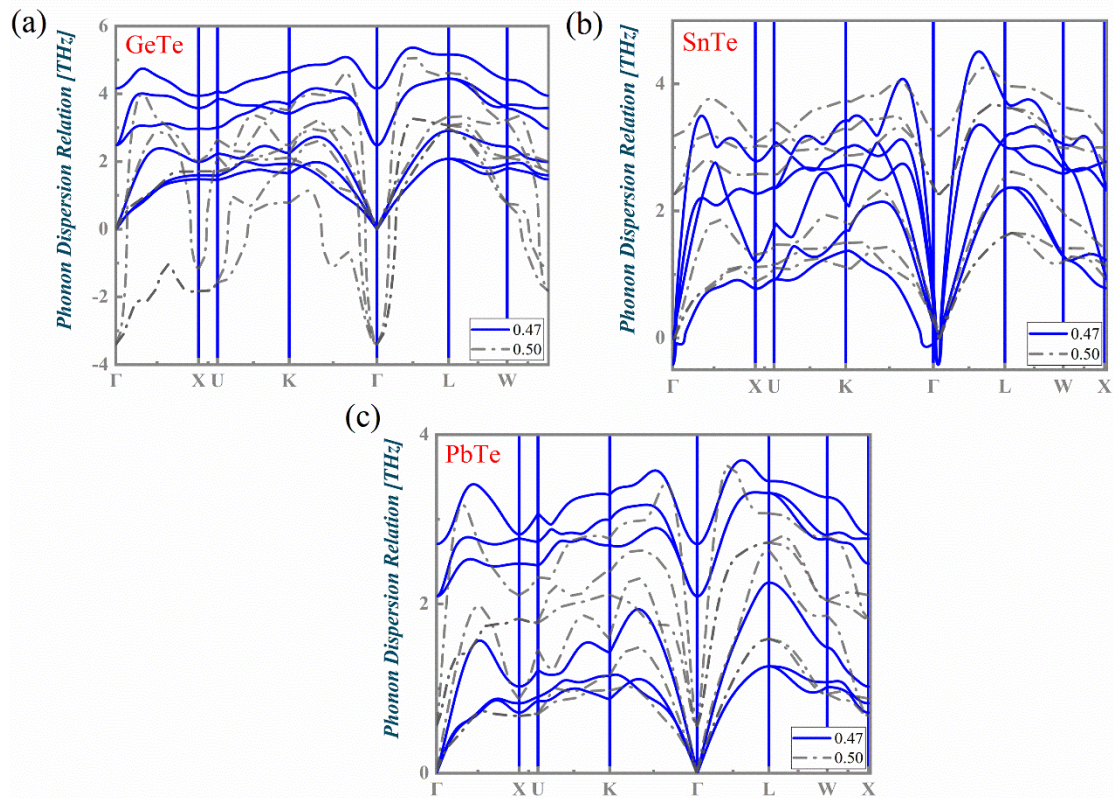


Fig.S3. The calculated phonon dispersion of (a) GeTe, (b) SnTe, (c) PbTe, as a function of carrier atomic reciprocal displacement

### S.1.3 Te off-centering behavior influences the TE properties of MTe

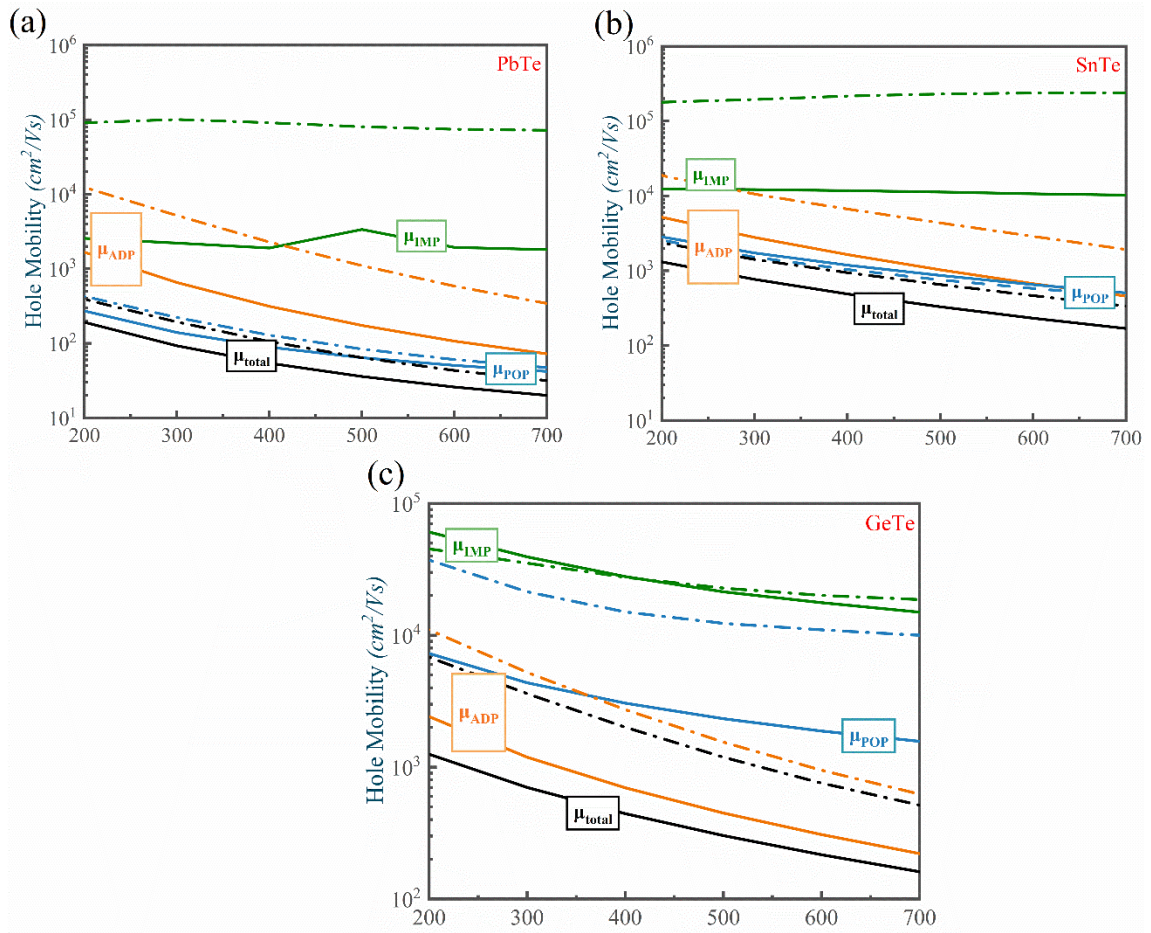


Fig.S4. Electron mobility versus temperature for the doping concentration of  $n = 1 \times 10^{19} cm^{-3}$  of p-type

MTe. The solid and dashed line represent Te coordinate at (0.47,0.47,0.47) and (0.50,0.50,0.50), respectively. Black line is the calculated mobility including all scattering channels, while colored lines correspond to the individual contribution of ADP, IMP and POP.

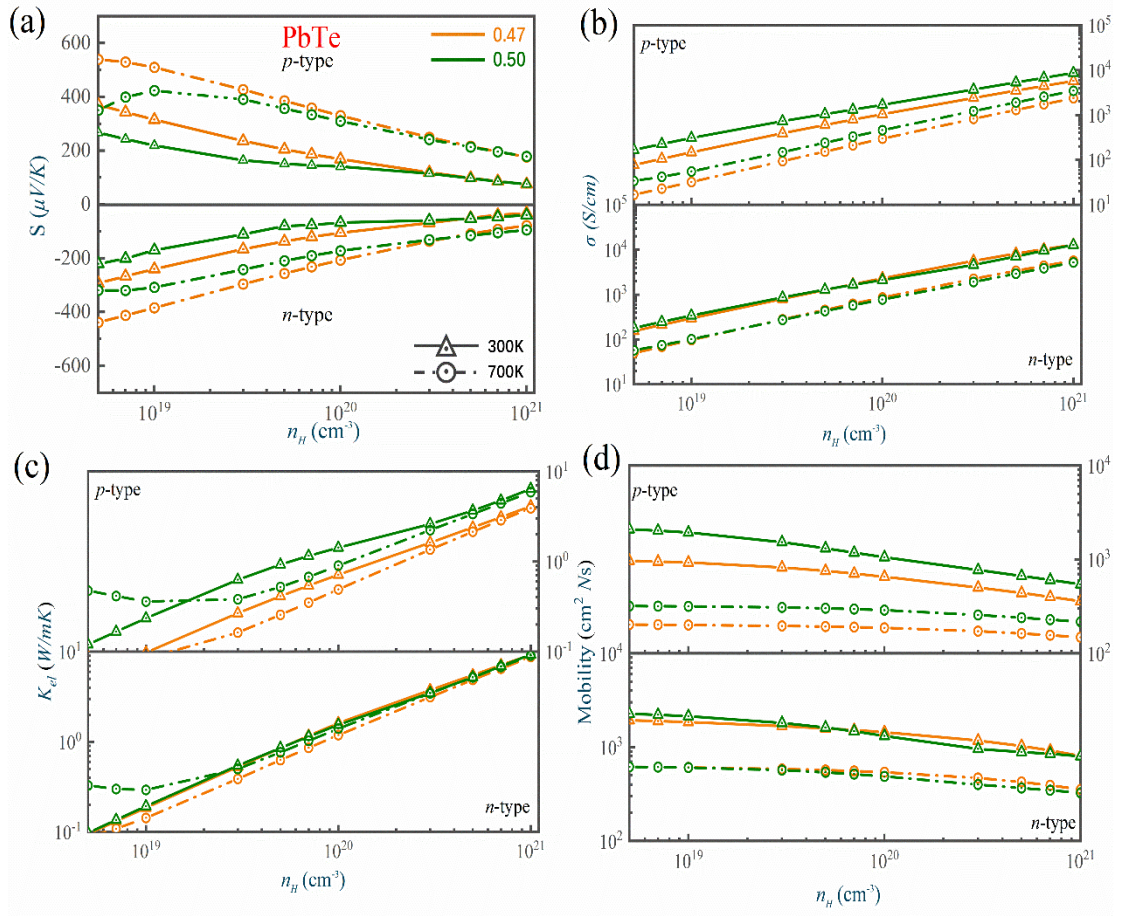


Fig.S5. The calculated (a) Seebeck coefficient, (b) electrical conductivity, (c) thermal conductivity, and (d) mobility of PbTe as a function of carrier concentration and atomic reciprocal displacement at 300K

and 700K

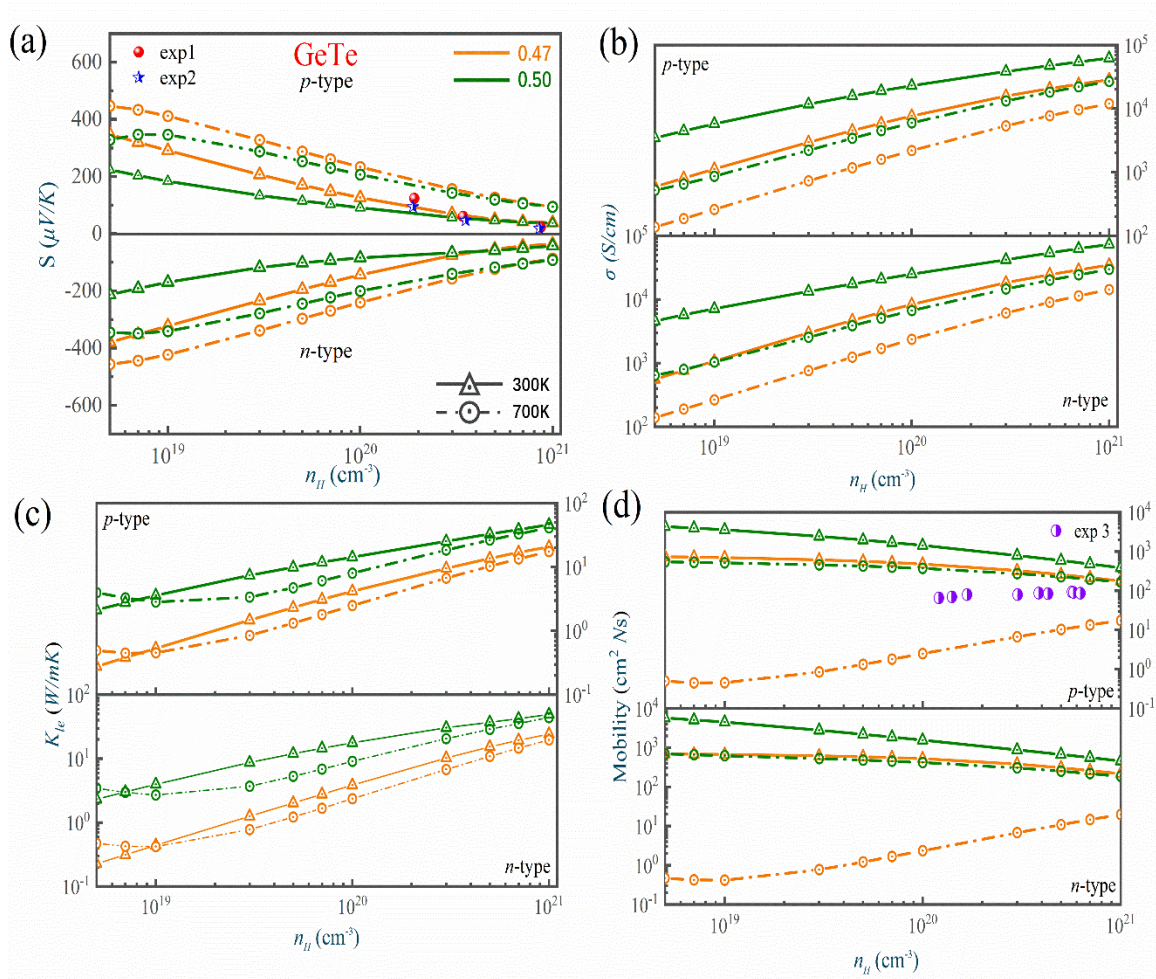


Fig.S6. The calculated (a) Seebeck coefficient, (b) electrical conductivity, (c) thermal conductivity, and

(d) and mobility of GeTe as a function of carrier concentration and atomic reciprocal displacement at

300K and 700K. Experimental data are shown in spheres<sup>1</sup>, circles<sup>2</sup>, stars<sup>3</sup>

#### **S.1.4 CRTA calculation Te off-centering behavior influences the TE properties of MTe**

Fig. S7, Fig. S8 and Fig. S9 show the calculated TE properties as a function of the carrier concentration for n-type and p-type MTe from 300 K to 800 K using a constant scattering time approximation. MRTA (momentum relaxation time approximation) can calculate MTe thermoelectric values more accurately, which basically matches the experimental data. However, the conclusion of (constant relaxation time approximation) CRTA still proves the Rashba effect can significantly enhance the Seebeck coefficient because the Seebeck coefficient does not involve carrier relaxation time. The pristine PbTe has a superior Seebeck coefficient than pristine SnTe (GeTe in between). Yet as both bandgap and band topology (effective mass) change with Te off-centering the relaxation time will also change. Therefore, using CRTA cannot obtain accurate electron transport properties, especially for electronic conductivity and electronic thermal conductivity.



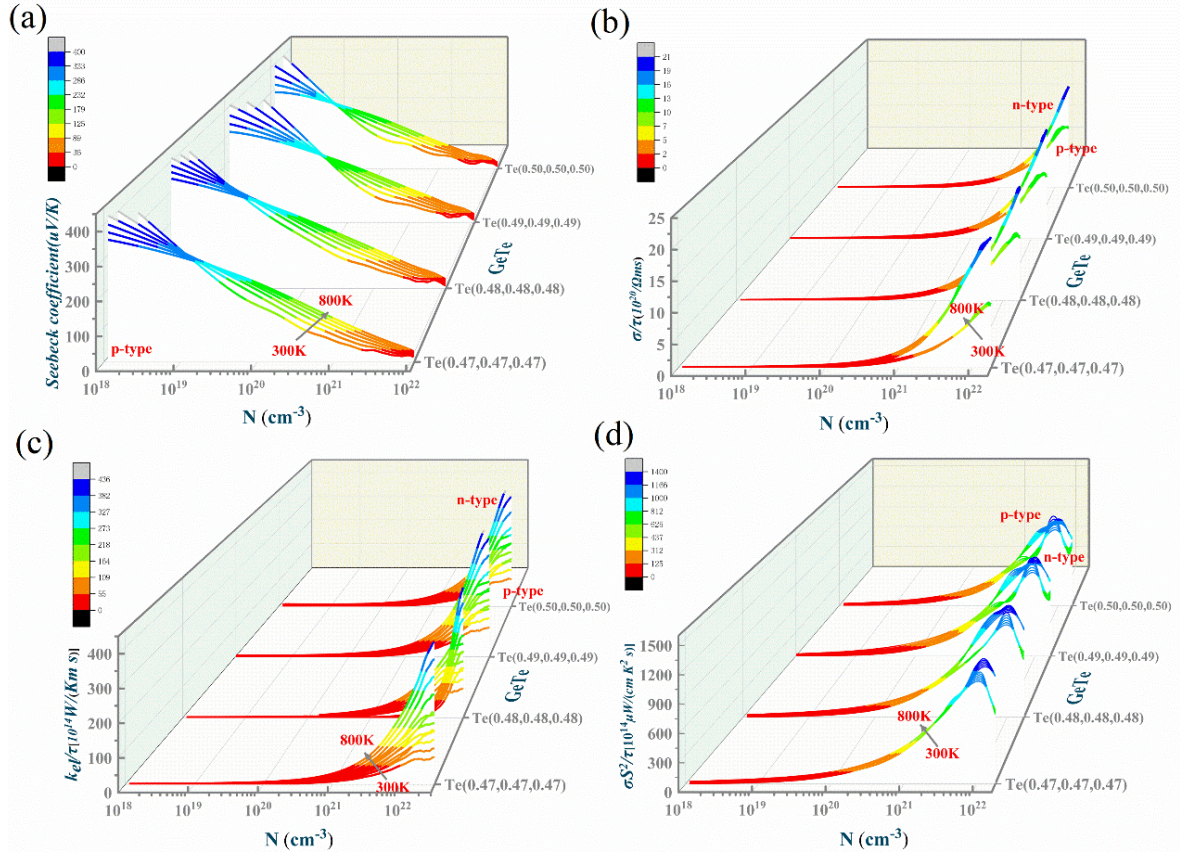


Figure.S7. The calculated (a) Seebeck coefficient, (b) electrical conductivity, (c) thermal conductivity, and (d) power factor of GeTe as a function of carrier concentration and atomic reciprocal displacement at the temperatures 300 K to 800 K

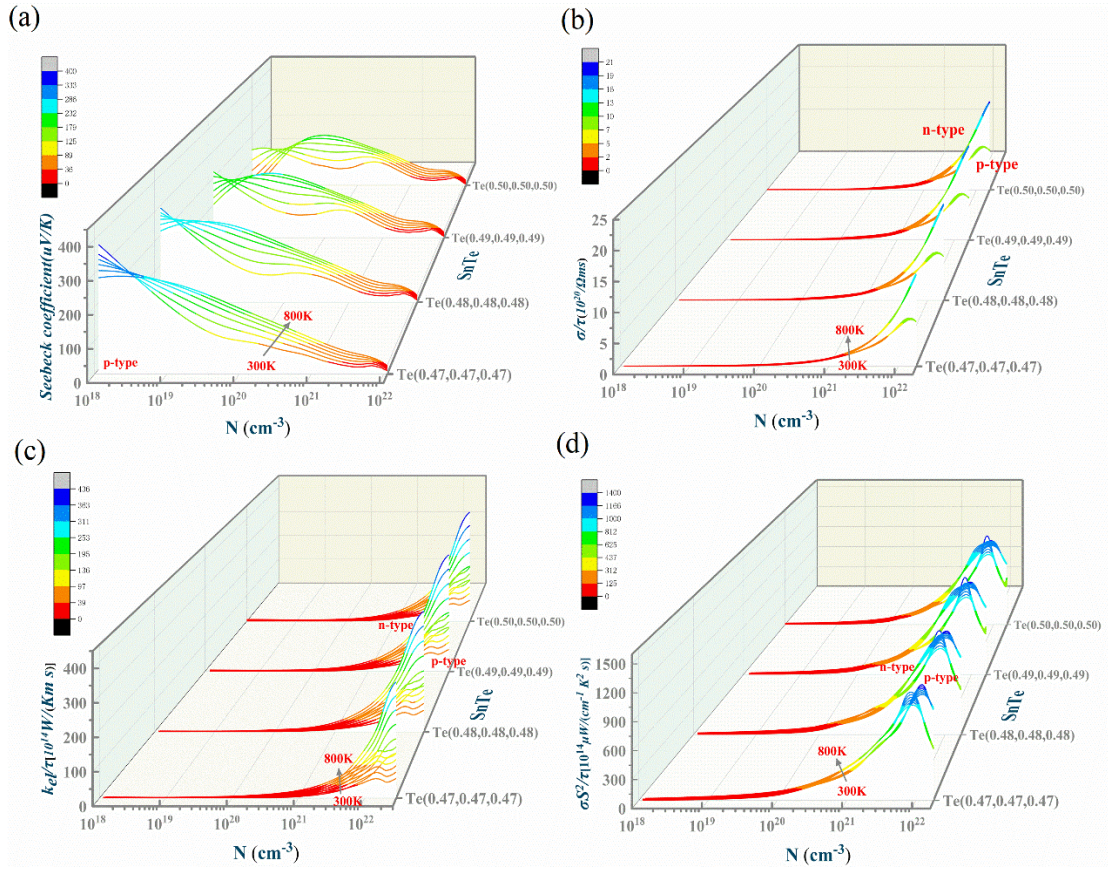


Fig.S8. The calculated (a) Seebeck coefficient, (b) electrical conductivity, (c) thermal conductivity, and (d) power factor of SnTe as a function of carrier concentration and atomic reciprocal displacement at the temperatures 300 K to 800 K

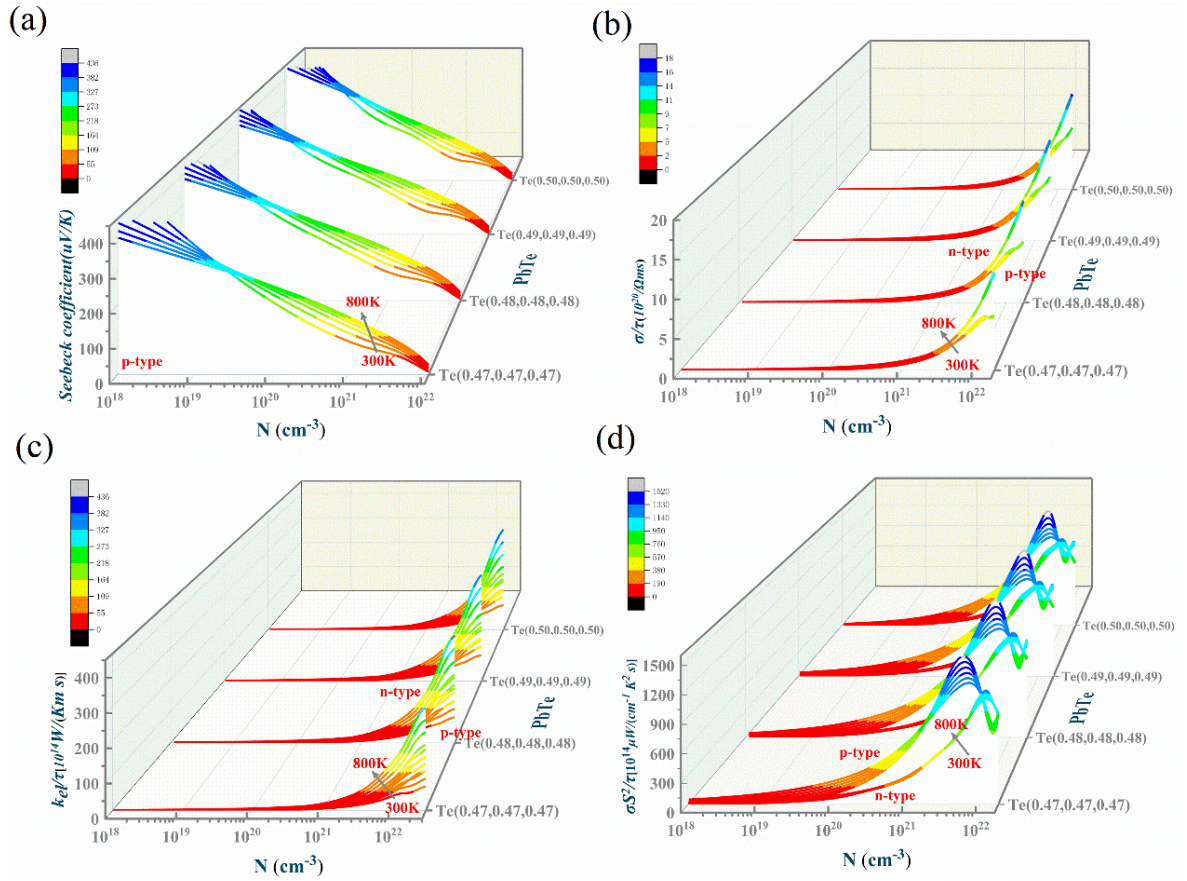


Fig.S9. The calculated (a) Seebeck coefficient, (b) electrical conductivity, (c) thermal conductivity, and (d) power factor of PbTe as a function of carrier concentration and atomic reciprocal displacement at the temperatures 300 K to 800 K

## References for Supplementary Information

1. M. Hong, Z. G. Chen, L. Yang, Y. C. Zou, M. S. Dargusch, H. Wang and J. Zou, *Adv. Mater.*, 2018, **30**, 1705942.
2. J. Dong, F.-H. Sun, H. Tang, J. Pei, H.-L. Zhuang, H.-H. Hu, B.-P. Zhang, Y. Pan and J.-F. Li, *Energy Environ. Sci.*, 2019, **12**, 1396-1403.
3. J. Li, Z. Chen, X. Zhang, Y. Sun, J. Yang and Y. Pei, *NPG Asia Mater.*, 2017, **9**, e353-e353.