Supporting Information

Grain Boundary Re-crystallization and Sub-nano Regions Leading to High Plateau Figure of Merit for Bi_2Te_3 Nanoflakes

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Section S1. Parallel model

The parallel model for electrical performance analysis of a composite material can be expressed as:[1]

$$\sigma_c = (1 - x_v) \cdot \sigma_m + x_v \cdot \sigma_v \tag{S1-1}$$

$$S_c = \frac{(1 - x_v) \cdot \sigma_m \cdot S_m + x_v \cdot \sigma_v \cdot S_v}{(1 - x_v) \cdot \sigma_m + x_v \cdot \sigma_v} \tag{S1-2}$$

σ is the electrical conductivity, S is the Seebeck coefficient, x_v is the volume fraction of secondary phase (vacuum here), $x_v=r$ (r is the relative density), c denotes for composite, m denotes for matrix, and v denotes for vacuum. When σ_v=0 S cm^{-1} and S_v=0 μV K^{-1} are taken into account, it can be extracted that:

$$\sigma_c = (1 - r) \cdot \sigma_m \tag{S1-3}$$

$$S_c = S_m \tag{S1-4}$$

As can be seen, the porosity r influence only the σ_c through vacuum compositing. S_c is still determined by the S_m of the matrix material.

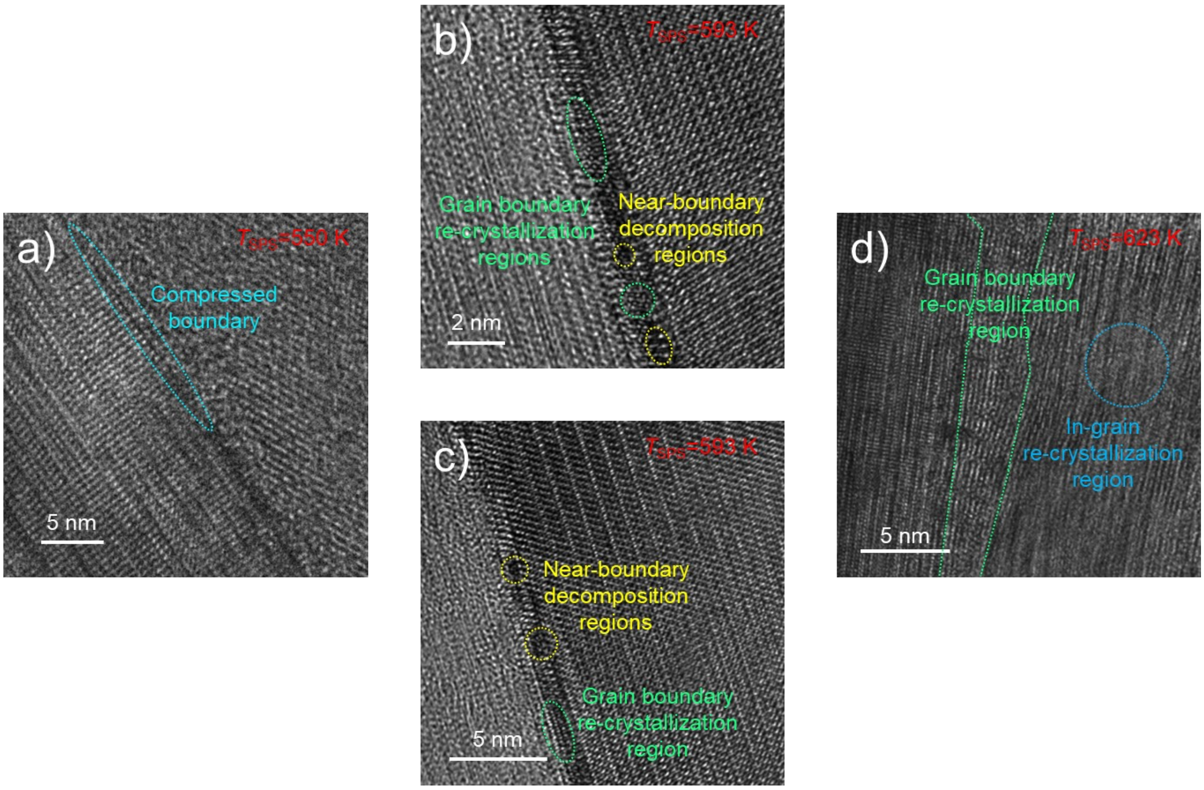

Fig. S1. High-magnification TEM images of as-sintered Bi_2Te_3 pellets sintered under T_{SPS} of a) 550, b) and c) 593, and d) 623 K, respectively.

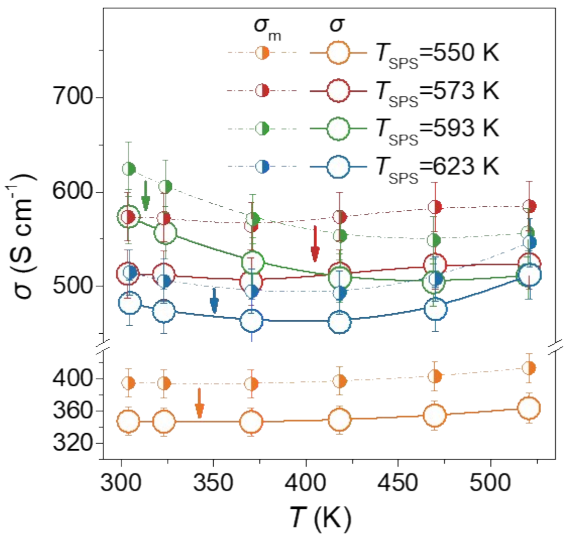

Fig. S2. Measured σ of as-prepared Bi_2Te_3 pellets under different T_{SPS} in comparison with the σ_m extracted by parallel model.

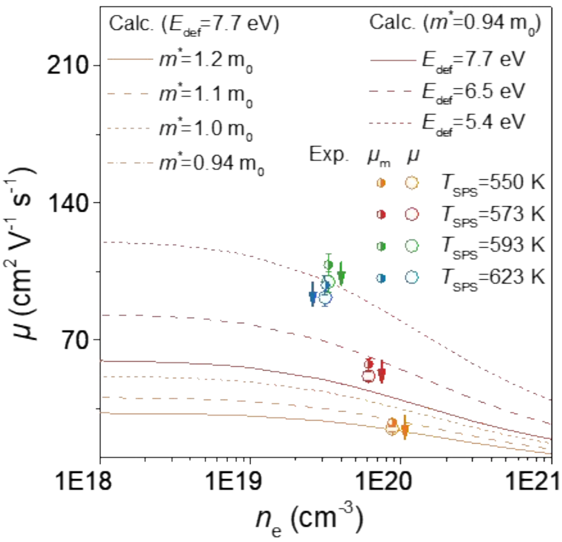

Fig. S3. Comparison between measured (under different T_{SPS}) μ, extracted μ_m and calculated (under different E_{def} and m^*) μ as a function of n_e, at 370 K.

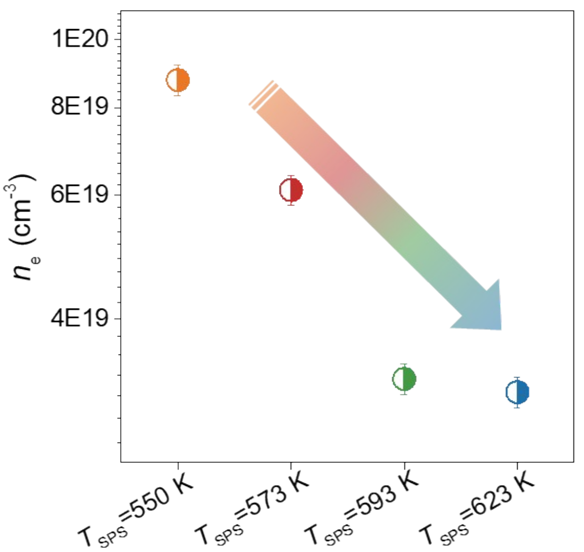

Fig. S4. Measured n_e of Bi_2Te_3 pellets sintered under T_{SPS} of 550, 573, 593 and 623 K, respectively.

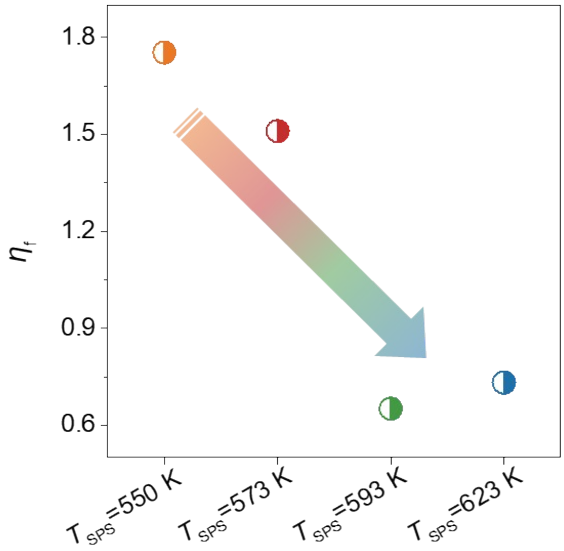

Fig. S5. SPB model extracted η_f of Bi_2Te_3 pellets sintered under T_{SPS} of 550, 573, 593 and 623 K, respectively.

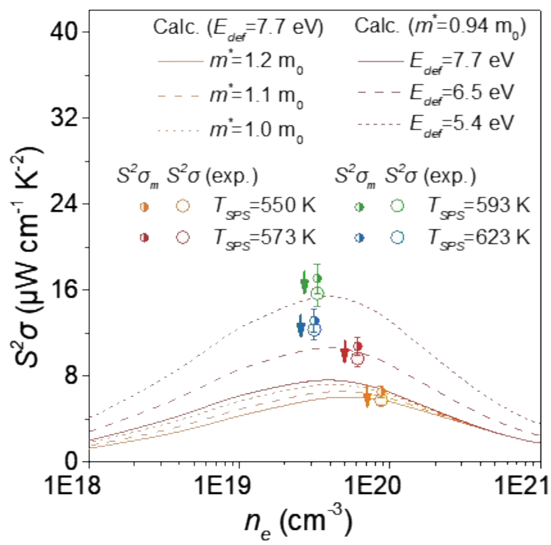

Fig. S6. Comparison between measured (under different T_{SPS}) $S^2\sigma$, extracted $S^2\sigma_m$ and calculated (under different E_{def} and m^*) $S^2\sigma$ as a function of n_e, at 370 K.

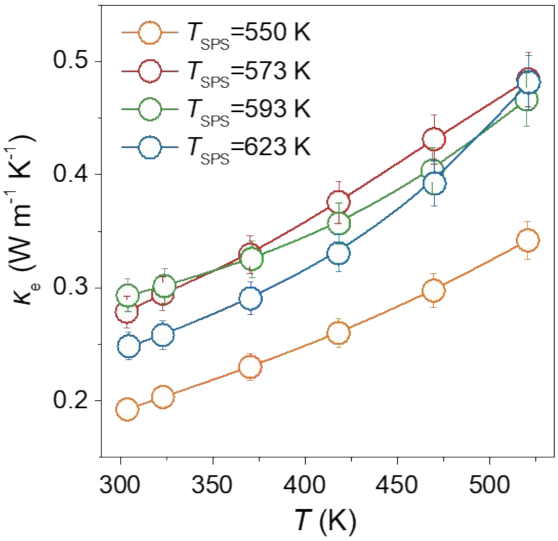

Fig. S7. Extracted κ_e of Bi_2Te_3 pellets sintered under T_{SPS} of 550, 573, 593 and 623 K, respectively.

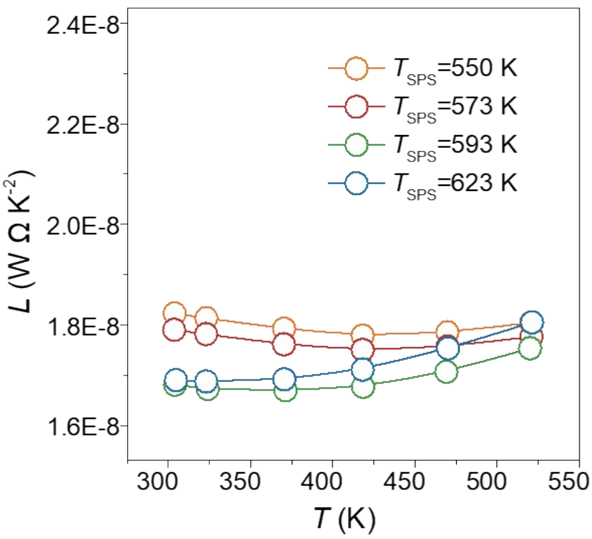

Fig. S8. Extracted L of Bi_2Te_3 pellets sintered under T_{SPS} of 550, 573, 593 and 623 K, respectively.

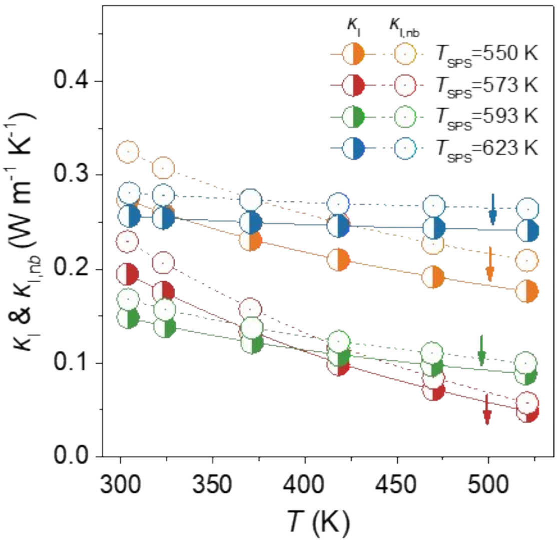

Fig. S9. Comparison between evaluated κ_l of Bi_2Te_3 pellets prepared under different T_{SPS} and the gray medium model-evaluted $\kappa_{l,nb}$ of nanobulks without pores.

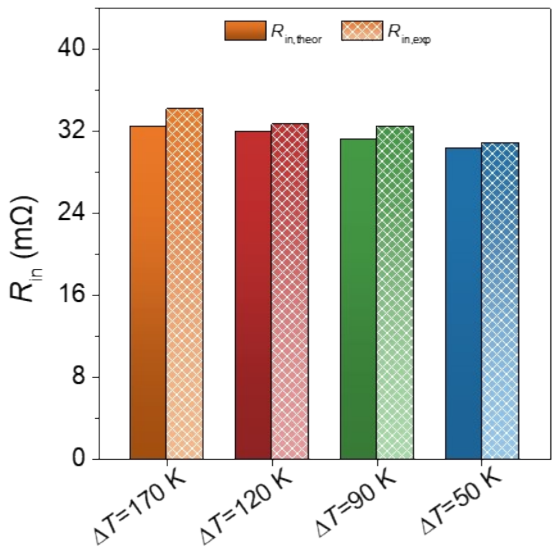

Fig. S10. Measured R_{in} of the of a single-leg device prepared based on the Bi_2Te_3 pellet sintered under the T_{SPS} of 593 K under different ΔT, comparing with the theoretical values.

Table S1. Comparison of key carrier transport properties evaluated by SPB model of Bi_2Te_3 pellets prepared under different T_{SPS} and those of Bi_2Te_3 matrixes (denoted by m) extracted by parallel model.

T_{SPS} (K)	n_e (cm^{-3})	μ (cm^2 V^{-1} s^{-1})	μ_m (cm^2 V^{-1} s^{-1})	m^* (m$_0$)	m^*_m (m$_0$)	E_{def} (eV)	$E_{def,m}$ (eV)
550	8.76E+19	24.7	28.1	1.17	1.17	7.68	7.20
573	6.11E+19	51.6	57.6	1.02	1.02	6.51	6.16
593	3.29E+19	99.6	108.4	0.94	0.94	5.44	5.22
623	3.15E+19	91.9	98.0	0.84	0.84	6.58	6.37

References

1. H. Yao, Z. Fan, H. Cheng, X. Guan, C. Wang, K. Sun and J. Ouyang, *Macromol. Rapid. Comm.*, 2018, **39**, 1700727.