Supporting information

Large-area 2D Bismuth Antimonide with Enhanced Thermoelectric Property via Multiscale Electron-Phonon Decoupling

Hanliu Zhao[a‡], Yuxin Xue[a‡], Yu Zhao[b‡], Jiayi Chen[a], Bo Chang[a], Hao Huang[a], Tao Xu[c], Litao Sun[c], Yunfei Chen[b] , Jingjie Sha[b,*], Beibei Zhu[a,*], Li Tao[a,*]

a School of Materials Science and Engineering, Jiangsu Key Laboratory of Advanced Metallic Materials, Southeast University, Nanjing 211189, People's Republic of China

b School of Mechanical Engineering, Jiangsu Key Laboratory for Design and Manufacture of Micro-Nano Biomedical Instruments, Southeast University, Nanjing 211189, People's Republic of China

c SEU-FEI Nano-Pico Center, Key Laboratory of MEMS of Ministry of Education, Collaborative Innovation Center for Micro/Nano Fabrication, Device and System, Southeast University, Nanjing 211189, People's Republic of China

E-mail: tao@seu.edu.cn, 101012333@seu.edu.cn, major212@seu.edu.cn

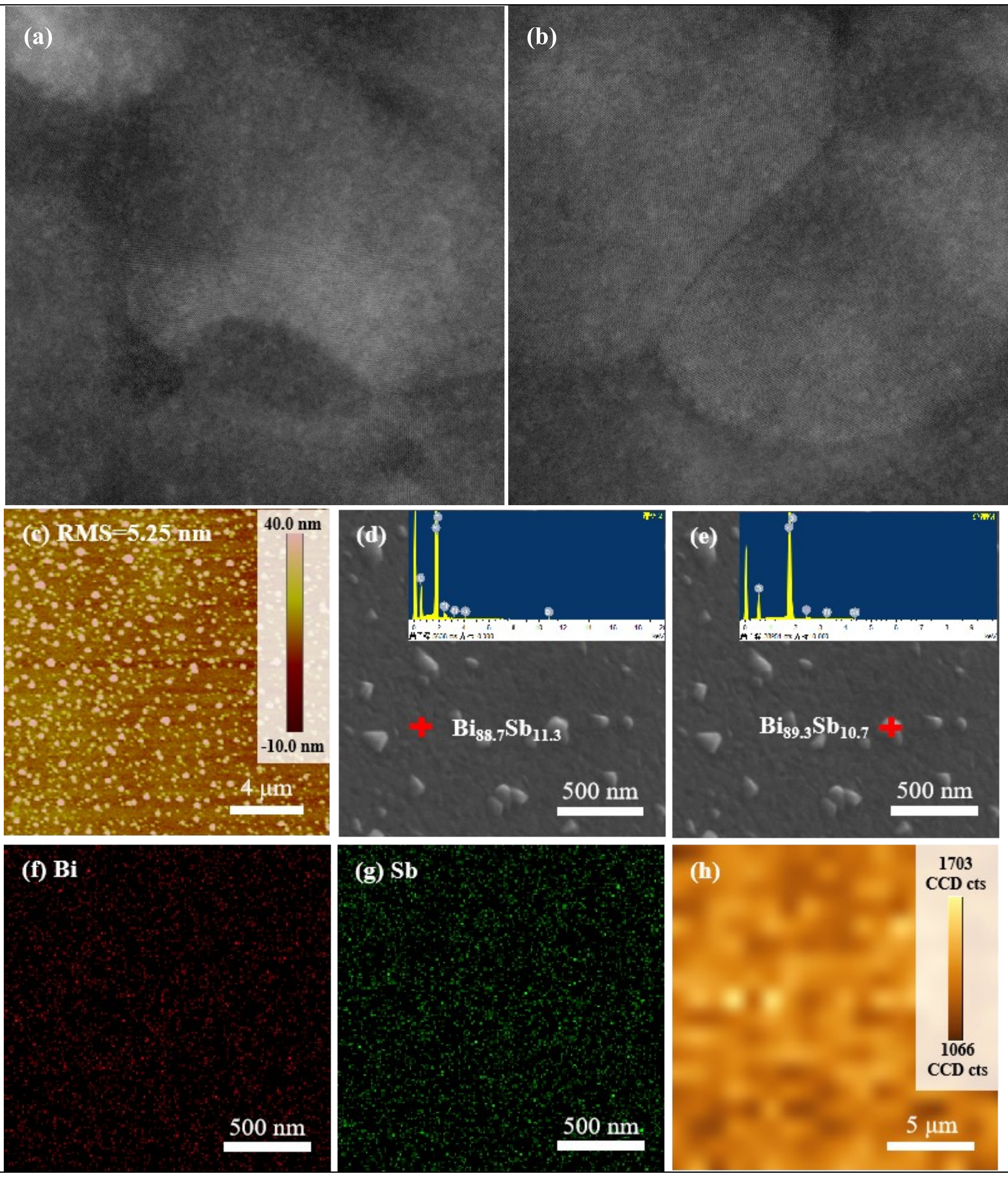

Fig. S1 (a&b) TEM images of 2D $Bi_{82}Sb_{18}$. (c) AFM image of 2D $Bi_{82}Sb_{18}$, showing a roughness of 5.25 nm. (d&e) Point EDS results of flat area and the protruded grains in $Bi_{89}Sb_{11}$. (f&g) EDS mapping of 2D $Bi_{65}Sb_{35}$. (h) Raman mapping of 2D $Bi_{82}Sb_{18}$.

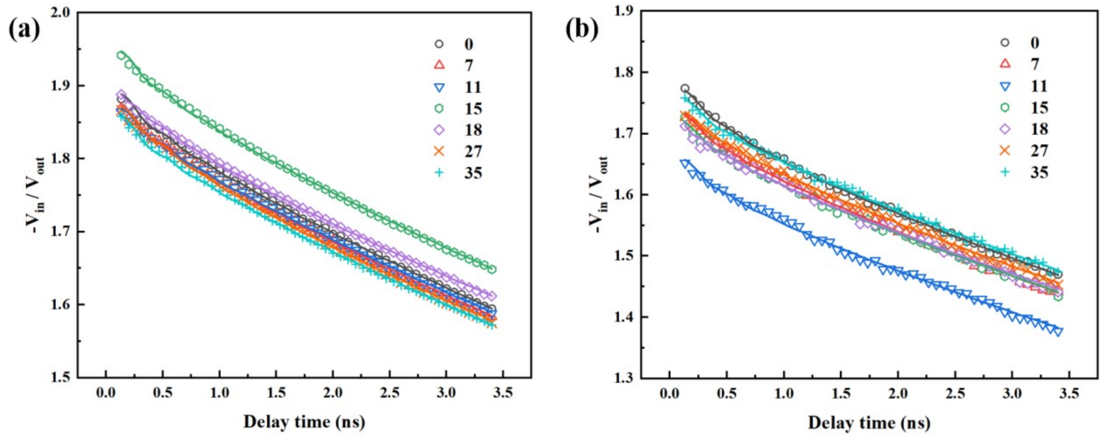

Fig. S2 Fitting curves of $-V_{in}/V_{out}$-delay time for TDTR measurement of (a) as-grown and (b) annealed 2D bismuth antimonide. V_{in} and V_{out} represent in-phase and out-of-phase signals, respectively. The open symbols and the solid lines denote measured data and thermal model fitting result. Measurement conditions: modulation frequency: 1.72 MHz; pump beam spot size: 10 µm; probe spot size: 6.0 µm.

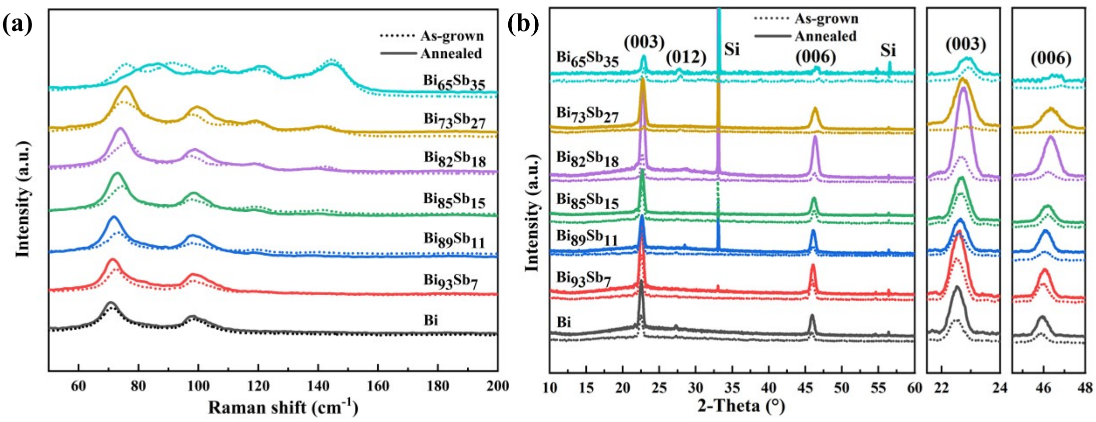

Fig. S3 (a) XRD patterns and (b) Raman spectra of as-grown and annealed 2D bismuth antimonide

After annealing process, the absolute Seebeck coefficient values are increased when x=15, 27 and 35 but decreased when x=0, 7 and 11 (Fig. S4a). There may be two reasons: i) the sublimation in the samples with x=0, 7 and 11 is negligible in SEM image (Fig. S7); ii) the annealing treatment reduces the density of defects. The above situations will reduce the electron scattering and cause the Seebeck coefficient decreasing.

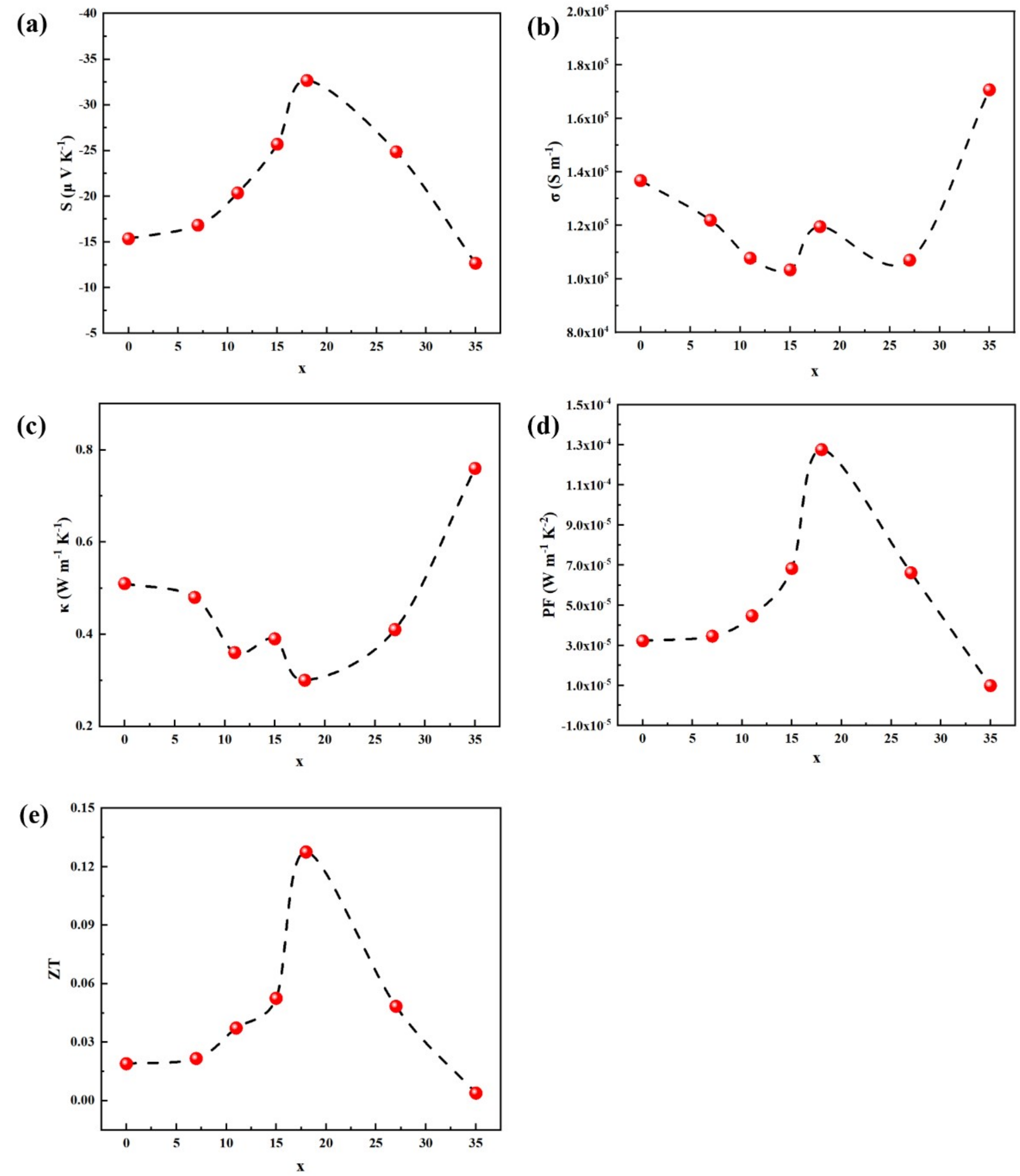

Fig. S4 (a) Seebeck coefficient (b) Electrical conductivity, (c) thermal conductivity, (d) power factor and (e) ZT value of annealed $Bi_{100-x}Sb_x$.

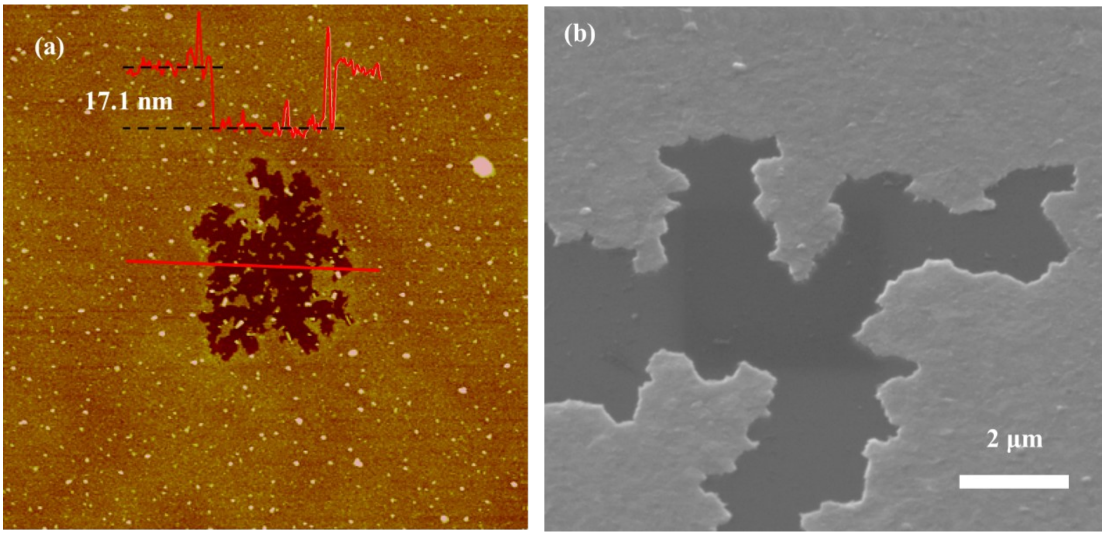

Fig. S5 AFM and SEM images of annealed 2D $Bi_{82}Sb_{18}$.

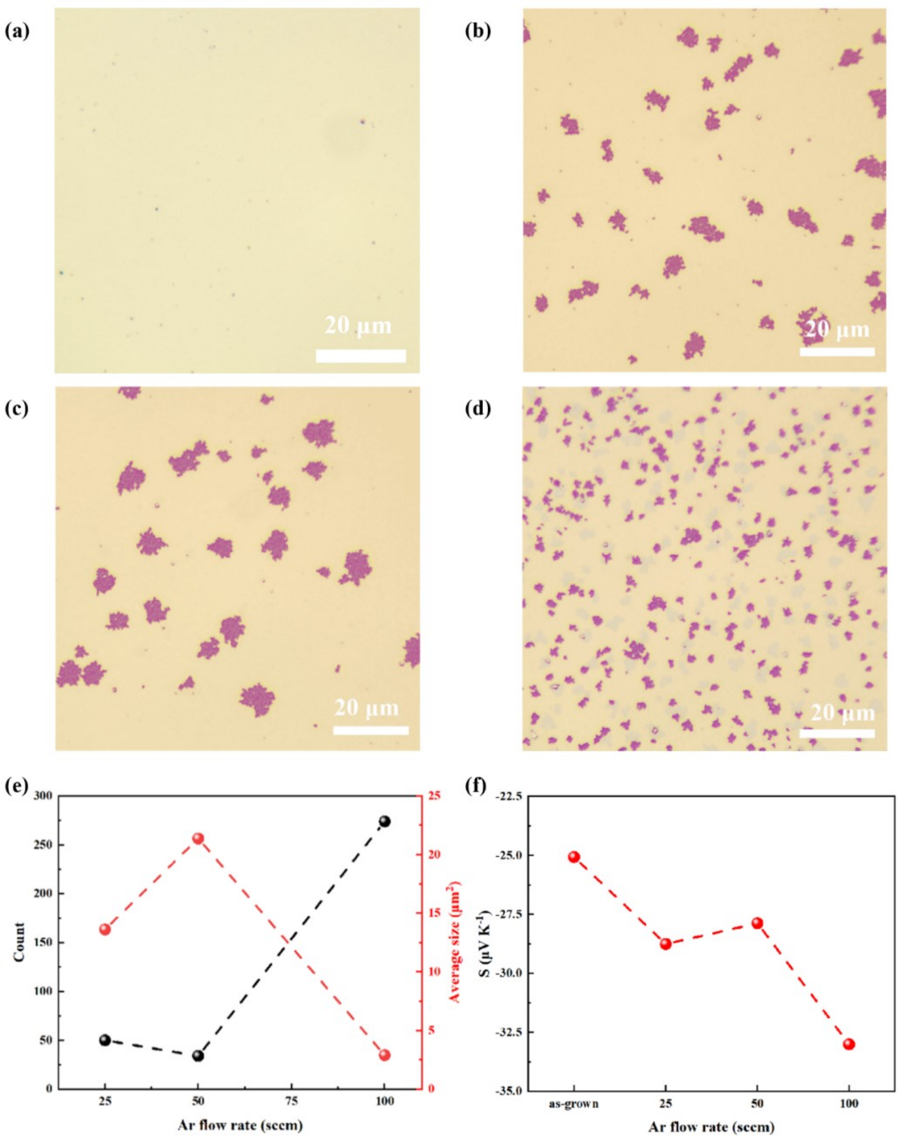

Fig. S6 Microporous structures of $Bi_{82}Sb_{18}$ films before and after annealing at different Ar flow rates. (a)as-grown (b) 25 sccm. (c) 50 sccm. (d) 100 sccm. (e)

number of micropores and average size. (f) Seebeck coefficient.

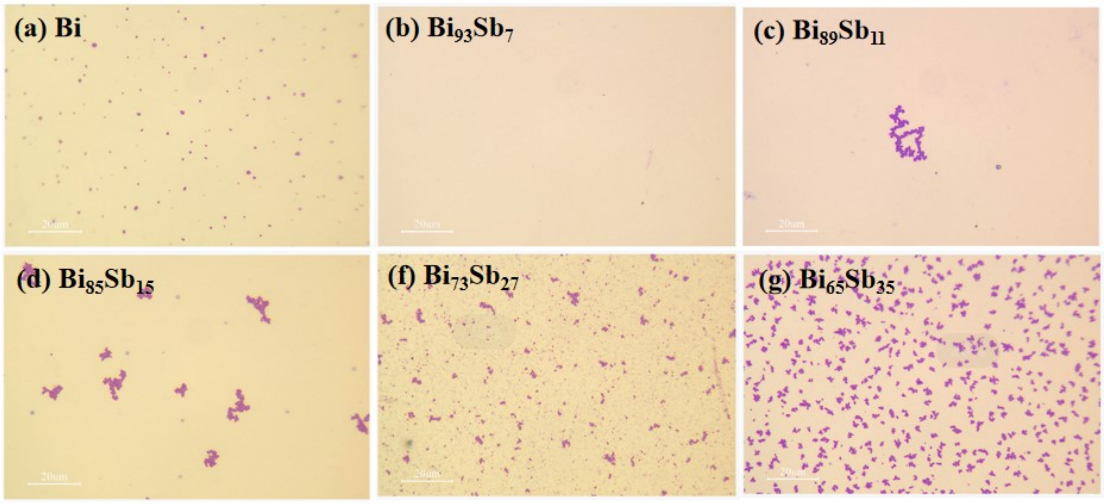

Fig. S7 OM images of annealed 2D $Bi_{1-x}Sb_x$.