Supporting Information

Transport and Thermoelectric Properties of Penta-Sb_2X Monolayers

Nan Wu[a], Xiaofeng Fan[a,*], David J. Singh[c,d] and W.T. Zheng[a,b]

a. Key Laboratory of Automobile Materials (Jilin University), Ministry of Education, and College of Materials Science and Engineering, Jilin University, Changchun, 130012, China

b. State Key Laboratory of Automotive Simulation and Control, Jilin University, Changchun 130012, China

c. Department of Physics and Astronomy, University of Missouri, Columbia, Missouri 65211-7010, USA

d. Department of Chemistry, University of Missouri, Columbia, Missouri 65211 USA

*, Correspondence and requests for materials should be addressed,

Email: xffan@jlu.edu.cn (X. Fan)

Tel: +86-159-4301-3494

Electronic transport model with explicit solution to BTE[1, 2]

In this method, a small driving force $g(k)$ is introduced to perturb the distribution of electrons in solving the Boltzman transport equation (BTE). The actual distribution of electrons under this perturbation can be expressed as,

$$f(k) = f_0[\varepsilon(k)] + xg(k), \tag{s1}$$

where f_0 is the equilibrium Fermi-Dirac distribution and x is the cosine of the angle between the driving force $g(k)$ and **k**. Thus, the f(k) can be considered to be formed under both the elastic and inelastic scattering. In this way, the BTE can be solved self-consistently to obtain the driving force $g(k)$ with the formula,

$$g(k) = \frac{S_i\left[g(k')\right] - \upsilon(k)\left(\frac{\partial f}{\partial z}\right) - \frac{eE}{\mathsf{h}}\left(\frac{\partial f}{\partial k}\right)}{S_O(k) + v_{el}(k)}, \tag{s2}$$

where S_i, S_o are the terms of scattering-in and scattering-out for inelastic scattering mechanism, E is the electric field. v_{el} is the contributions of elastic scattering, including the scattering of ionized impurity, piezoelectric, deformation potential, and dislocation.

The carrier mobility can be obtained based on the driving force $g(k)$ under the electric field E with the formula,

$$\mu = \frac{1}{3E}\frac{\int \upsilon(\varepsilon)D(\varepsilon)g(\varepsilon)d\varepsilon}{\int D(\varepsilon)f(\varepsilon)d\varepsilon}, \tag{s3}$$

where $\upsilon(\varepsilon(k))$ and $D(\varepsilon)$ is the electron group velocity and density of states. $g(\varepsilon(k))$ is the driving force from the formula (s2) with the electric driving force $-\frac{eE}{\mathsf{h}}\left(\frac{\partial f}{\partial k}\right)$ and without the thermal driving force $\upsilon(k)\left(\frac{\partial f}{\partial z}\right)$. With the obtained mobility of electrons, the conductivity of electrons from CBM can be expressed as, $\sigma = ne\mu_e$. With the obtained mobility of holes, the conductivity of holes from VBM can be expressed as, $\sigma = pe\mu_h$.

The Seebeck coefficient can be calculated under just considering the thermal driving force, $\upsilon(k)\left(\frac{\partial f}{\partial z}\right)$ in the formula (s2). The Seebeck coefficient can be expressed

as,

$$S = \frac{k_B}{e}\left[-\frac{\int k^2 f(1-f)\frac{\varepsilon}{k_B T}dk}{\int k^2 f(1-f)dk} + \frac{\varepsilon_F}{k_B T}\right] - \frac{\frac{J}{\sigma}}{\frac{\partial T}{\partial z}}, \quad \text{(s4)}$$

where J is the current density and σ is the conductivity under the temperature gradient $\partial T/\partial z$. The current density can be expressed as,

$$J = -\frac{\sigma}{e}\frac{\partial \varepsilon_F}{\partial z} - \sigma S\frac{\partial T}{\partial z}. \quad \text{(s5)}$$

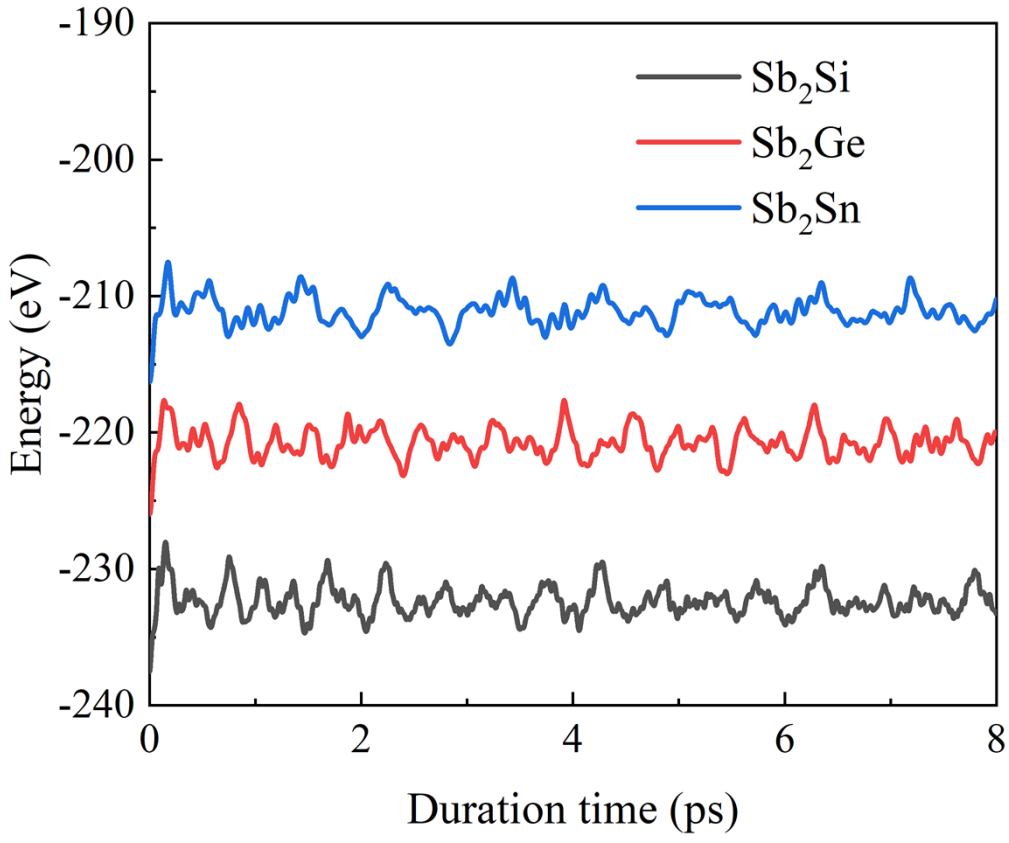

Figure S1. Changes in the energy for penta-Sb_2X (X=Si, Ge, Sn) monolayers over molecular dynamics simulations.

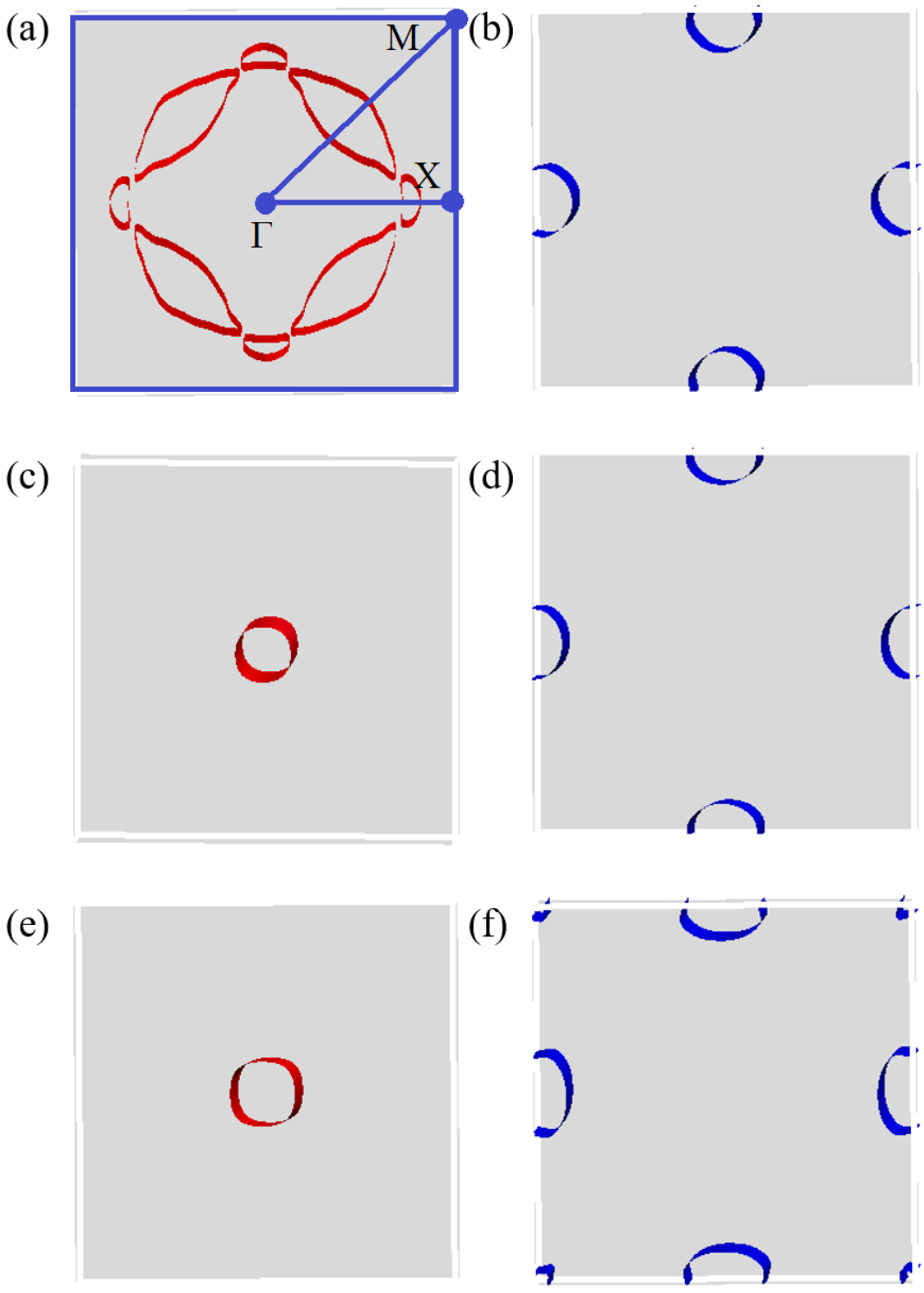

Figure S2. Calculated constant energy surfaces for penta-Sb_2X (X = Si, Ge, Sn) monolayers with the energy levels of 0.07 eV higher than (a, c, e) CBM and with energy levels of 0.04 eV lower than (b, d, f) VBM.

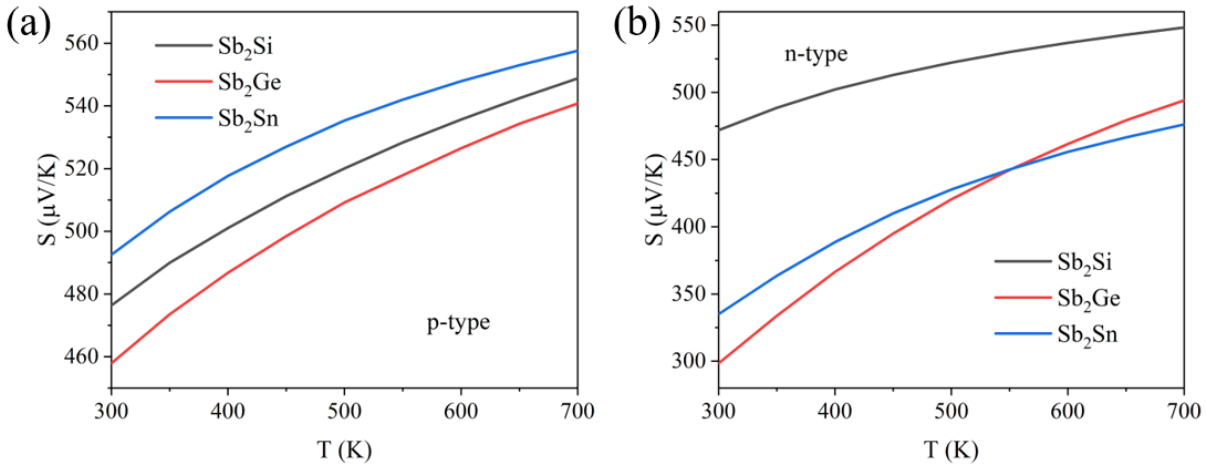

Figure S3. Seebeck coefficient *S* at the carrier concentration of 10^{12} cm^{-2} as the functions of temperature for penta-Sb_2X (X = Si, Ge, Sn) monolayers.

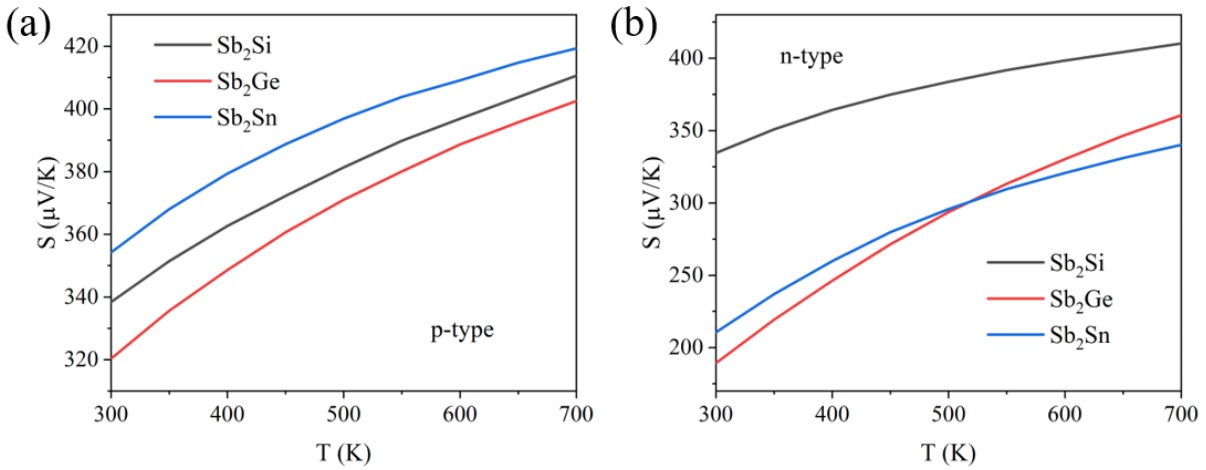

Figure S4. Seebeck coefficient S at the carrier concentration of 5×10^{12} cm^{-2} as the functions of temperature for penta-Sb_2X (X = Si, Ge, Sn) monolayers

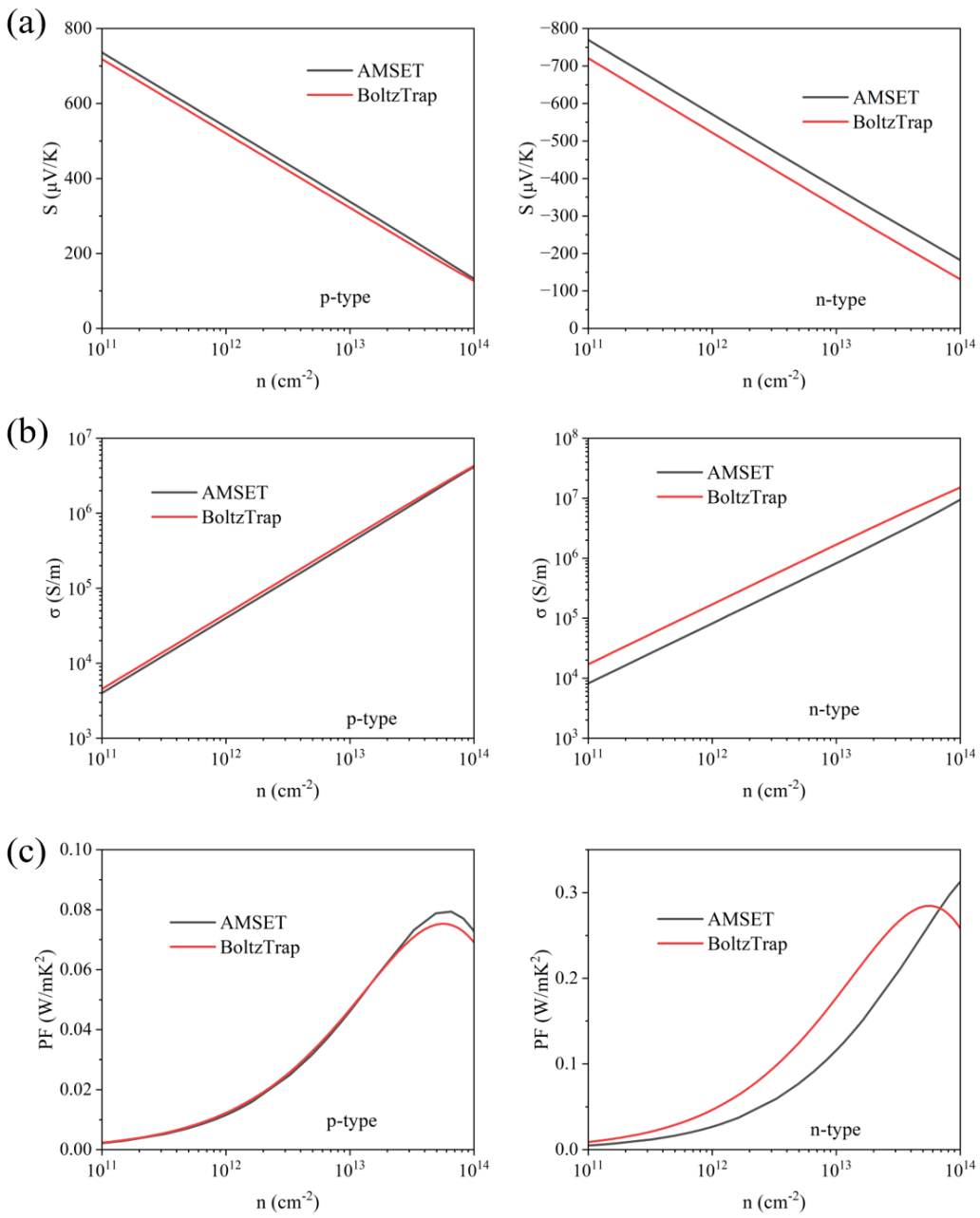

Figure S5. Calculated (a) Seebeck coefficient, (b) conductivity σ and (c) power factor with the methods of AMSET and BoltaTrap as the functions of carrier concentration for Sb_2Si monolayer with the p-type doping and n-type doping at 500 K.

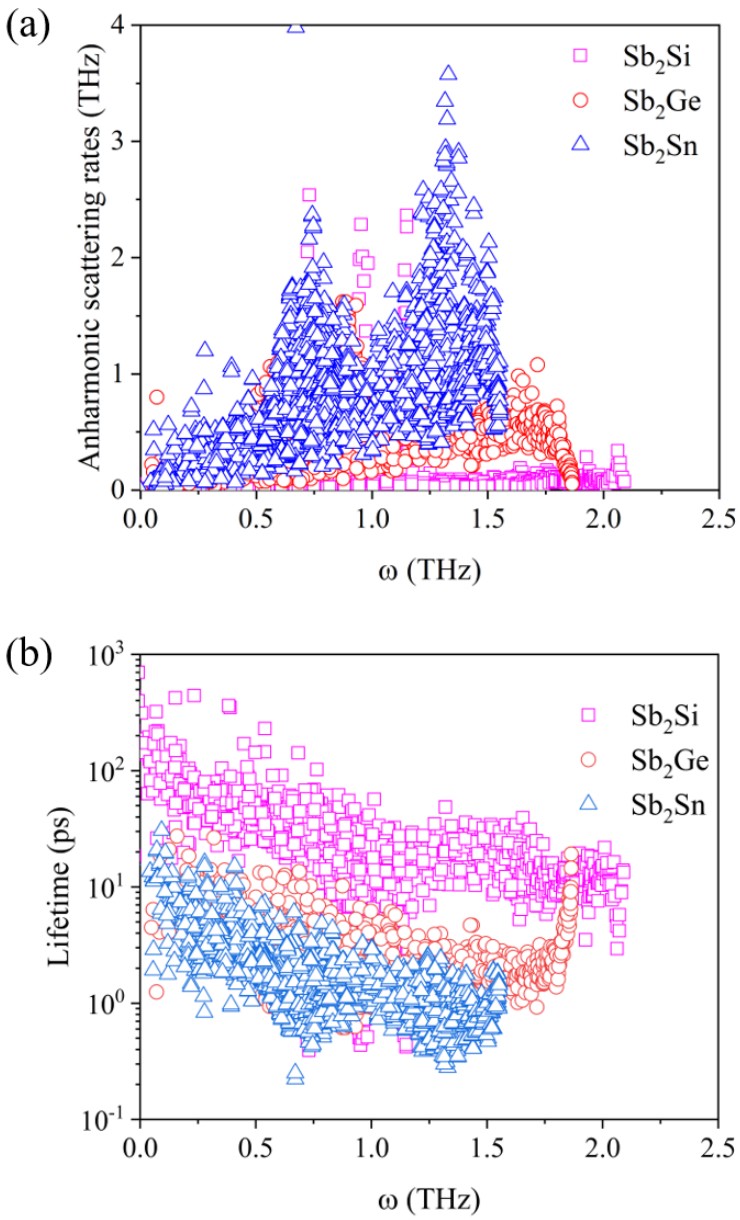

Figure S6. (a) Anharmonic scattering rates and (b) phonon lifetimes of acoustic phonon modes for penta-Sb_2X (X = Si, Ge, Sn) monolayers at 300K.

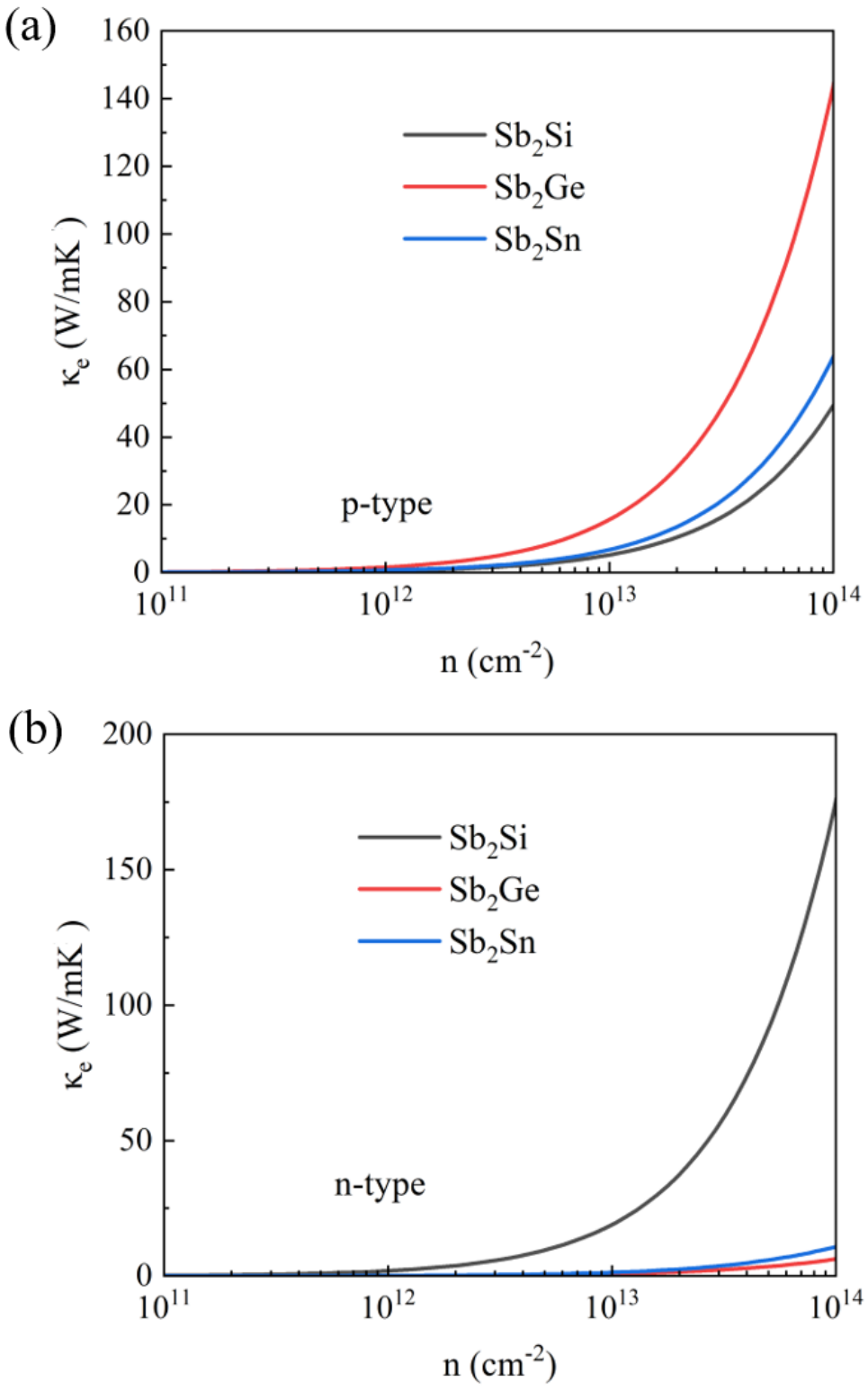

Figure S7. Calculated electronic thermal conductivity (κ_e) of penta-Sb_2X (X = Si, Ge, Sn) monolayers for (a) the p-type doping and (b) n-type doping at 700 K as a function of carrier concentration

References

1 A. Faghaninia, J. W. Ager and C. S. Lo, Ab initio electronic transport model with explicit solution to the linearized Boltzmann transport equation, *Phys. Rev. B*, 2015, **91**, 235123.

2 D. L. Rode, Semiconductors and Semimetals (*Academic Press*, *New York*, *1975*), Chapter 1.