Supplemental Material for

Electronic and thermoelectric properties of semiconducting Bi_2SSe_2 and Bi_2S_2Se monolayers with high optical absorption

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Abstract: Bismuth telluride (Bi_2Te_3) and its derivatives are often focused on as thermoelectric materials around room temperature. In this work, using the density functional theory (DFT) and Boltzmann transport theory, we theoretically predicted two new-type Bi_2Te_3-based two-dimensional materials Bi_2SSe_2 and Bi_2S_2Se. The thermal, dynamic, and mechanical stabilities of Bi_2SSe_2 and Bi_2S_2Se monolayers are confirmed based on ab-initio molecular dynamics (AIMD) simulations, phonon dispersion, and

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elastic constants calculation. The phonon transport properties including lattice thermal conductivity, group velocity, Grüneisen parameter, converged scattering rate, and phonon lifetime contributed by different branches were systematically investigated. The electronic and thermoelectric properties including carrier mobility (μ), Seebeck coefficient (S), electrical conductivity (σ), power factor, and figure of merit (zT) along the zigzag and armchair directions as a function of carrier concentration at different temperatures are also obtained. Bi_2SSe_2 and Bi_2S_2Se monolayers have a moderate indirect band gap (0.92 and 1.08 eV at PBE level) and a low lattice thermal conductivity (4.35 and 5.37 W m^{-1} K^{-1} at 300 K). The largest zT values of Bi_2SSe_2 and Bi_2S_2Se monolayers are 0.50 and 0.28 at 300 K and 1.39 and 0.93 at 700 K for p-doping types, respectively. The Bi_2SSe_2 and Bi_2S_2Se monolayers are predicted to possess high optical absorption peaks of 8×10^5 cm^{-1} in the visible and near-UV light region, respectively. The results indicate that both Bi_2SSe_2 and Bi_2S_2Se could be promising candidates in energy conversion, solar cells, and optoelectronic devices.

Keywords: Bismuth chalcogenides; First-principles; Two-dimensional materials; Electricity transport; Thermoelectric properties

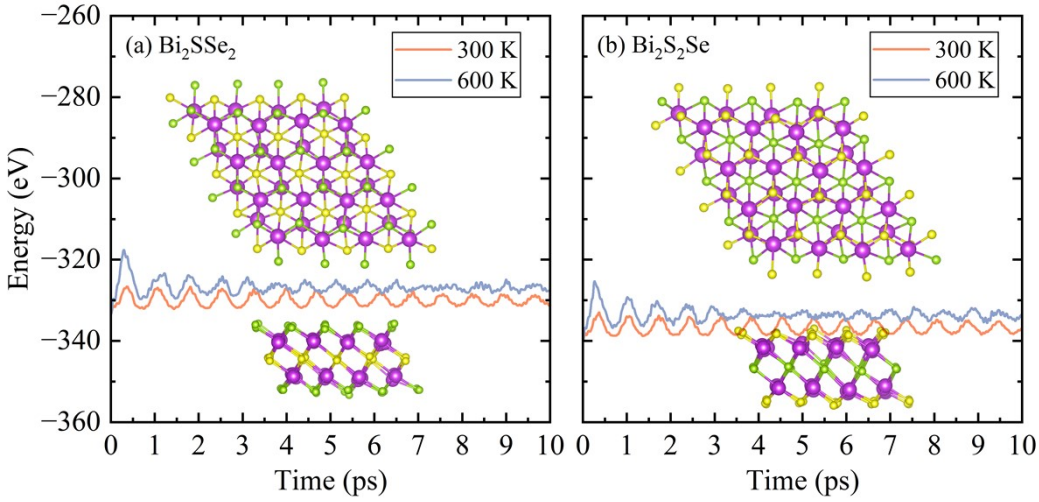

Fig. S1 (Color online) The AIMD simulations of (a) Bi_2SSe_2 and (b) Bi_2S_2Se monolayers at T = 300 K (the blue lines) and T = 600K (the red lines). The inset shows the final structures of the monolayers from the top view (above the lines) and the side view (below the lines) after 10 ps AIMD simulations at T = 600 K.

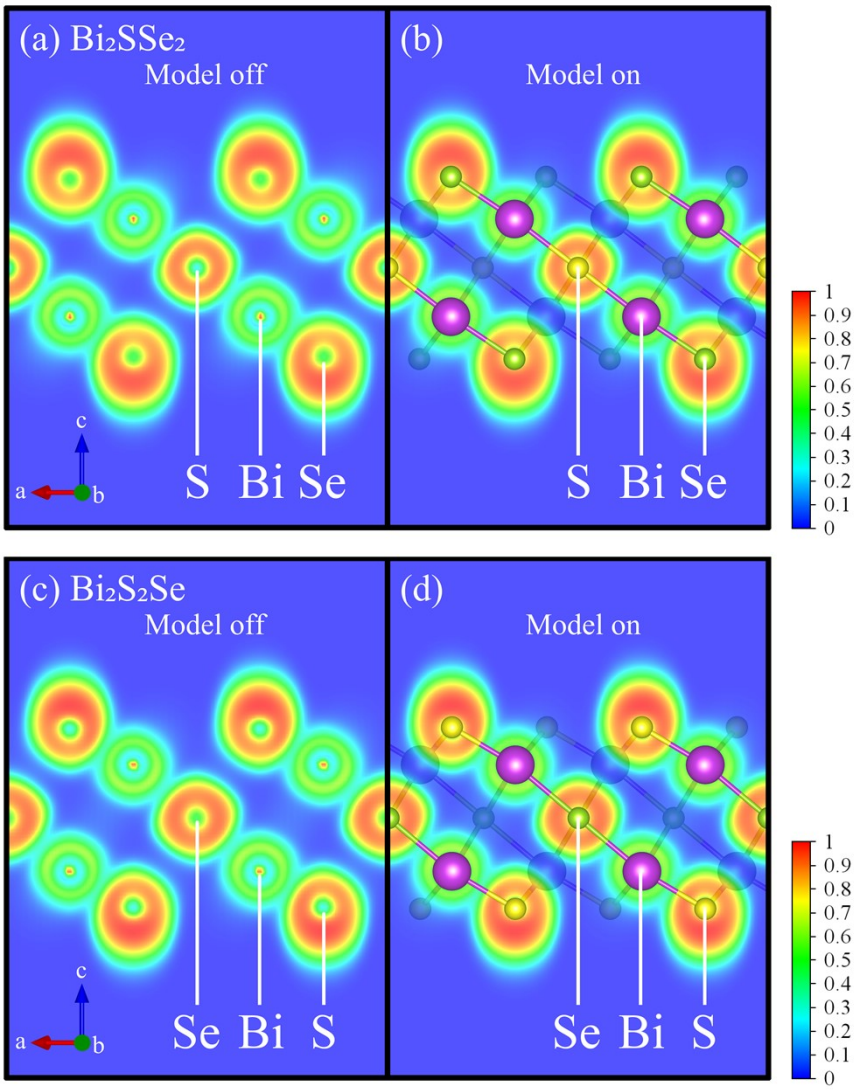

Fig. S2 (Color online) The electron localization functions (ELF) of Bi_2SSe_2 and Bi_2S_2Se monolayers from the side view.

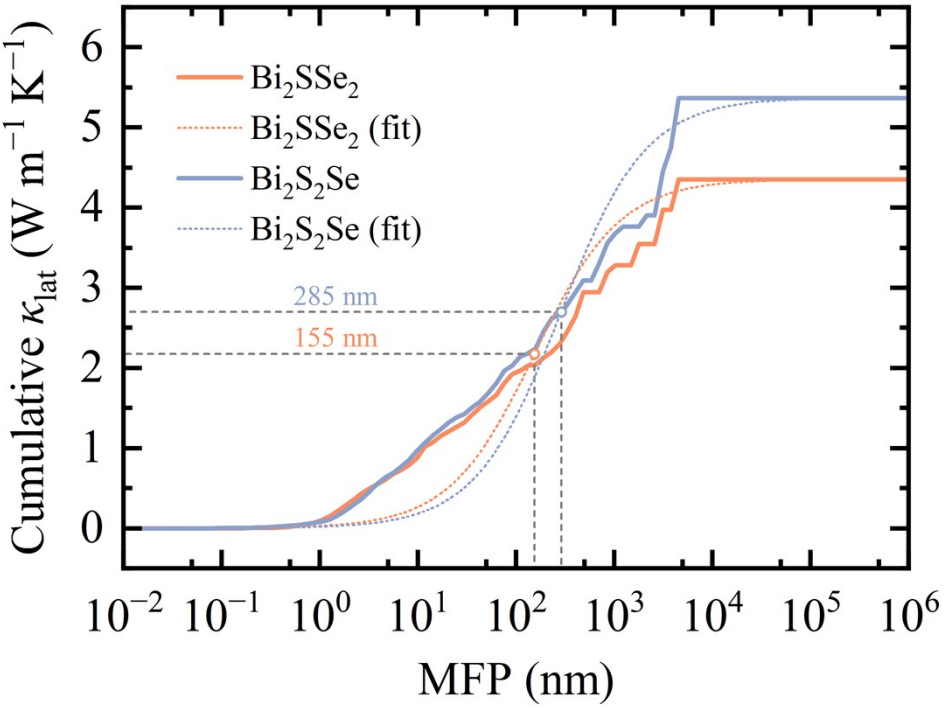

Fig. S3 (Color online) The calculated and fitted cumulative lattice thermal conductivity of Bi_2SSe_2 (red lines) and Bi_2S_2Se (blue lines) monolayers.

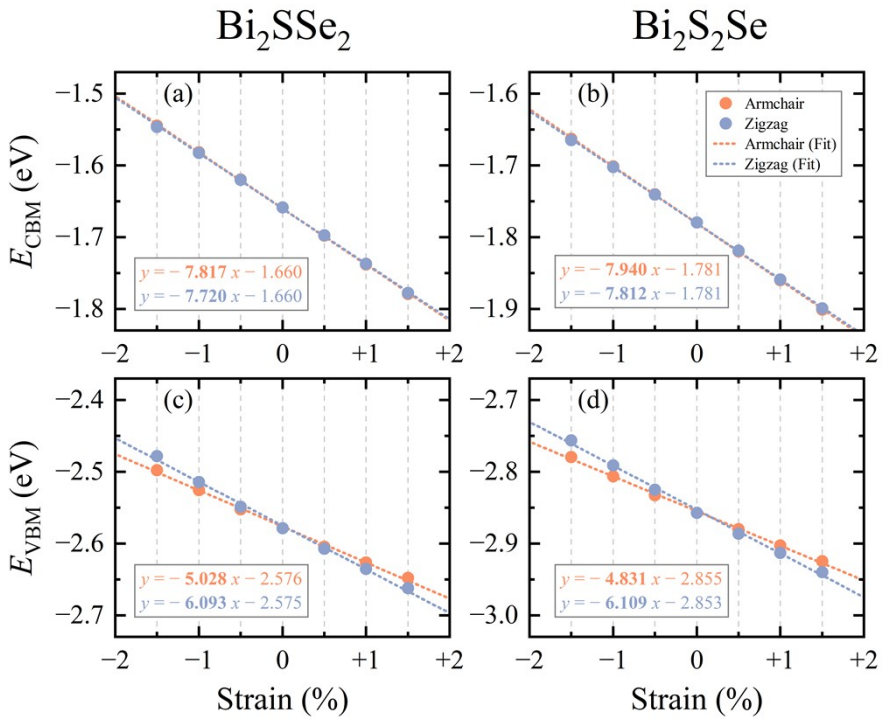

Fig. S4 (Color online) The calculated and fitted deformation potential of Bi_2SSe_2 and Bi_2S_2Se monolayers.

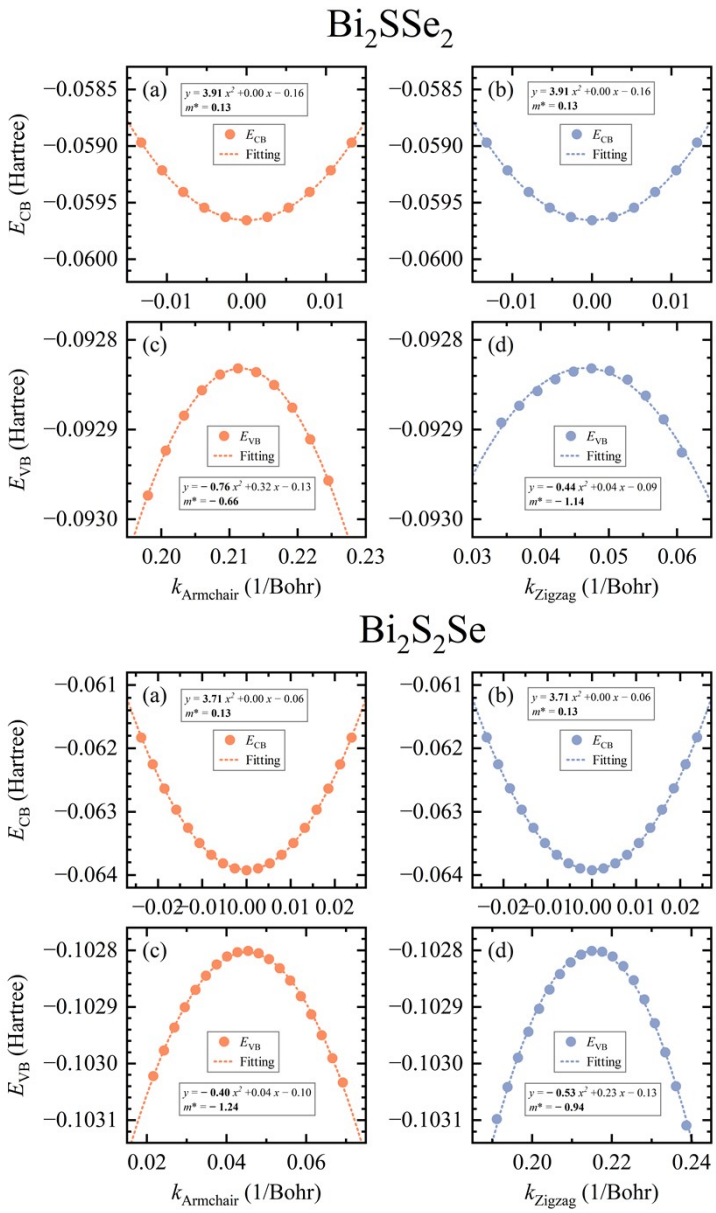

Fig. S5 (Color online) The calculated and fitted effective masses of Bi_2SSe_2 and Bi_2S_2Se monolayers.

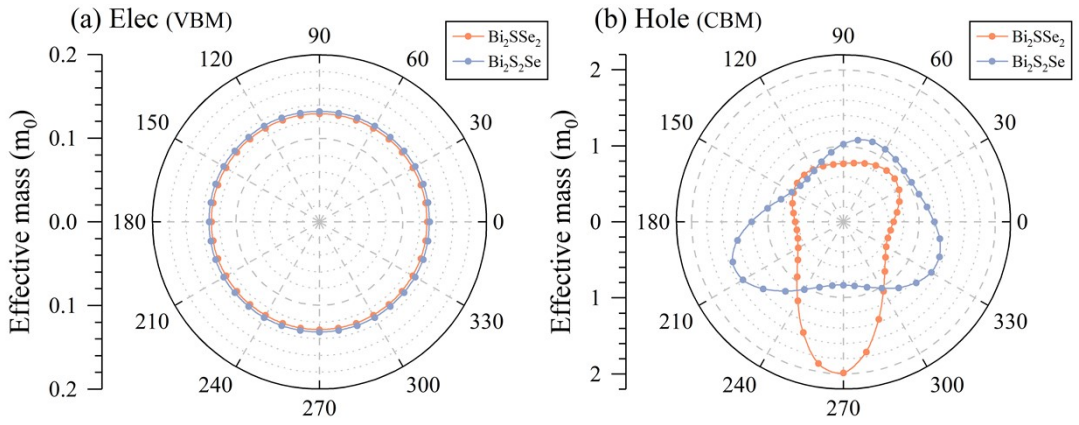

Fig. S6 (Color online) The calculated angle-dependent effective mass of (a) electrons and (b) holes for Bi_2SSe_2 (red lines) and Bi_2S_2Se (blue lines) monolayers in the square cells, where the 0 and 180 degrees are armchair directions, and 90 and 280 degrees are zigzag directions.

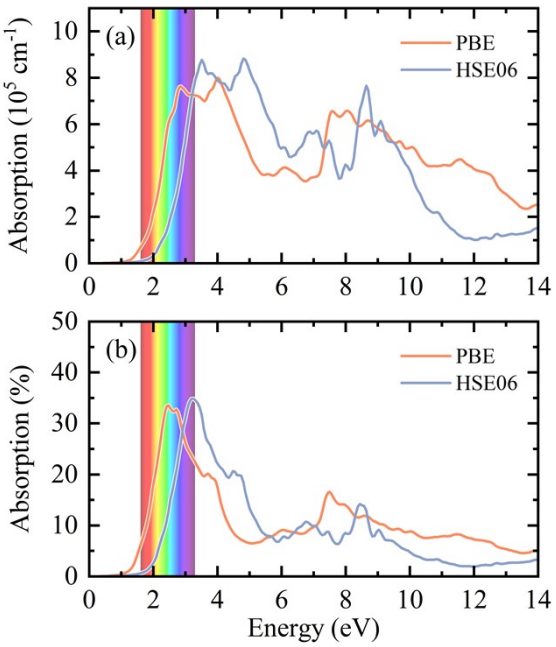

Fig. S7 (Color online) The calculated (a) traditional absorption spectra and (b) two-dimensional absorption spectra of Bi_2SSe_3 monolayers at PBE (red lines) and HSE06 (blue lines) levels, respectively. The simulated colors of the visible light are shown in the corresponding energy regions in the figure.