Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Strain and thickness effects on the electronic structures of low-energy two-dimensional Cd_xTe_y phases

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Structure prediction

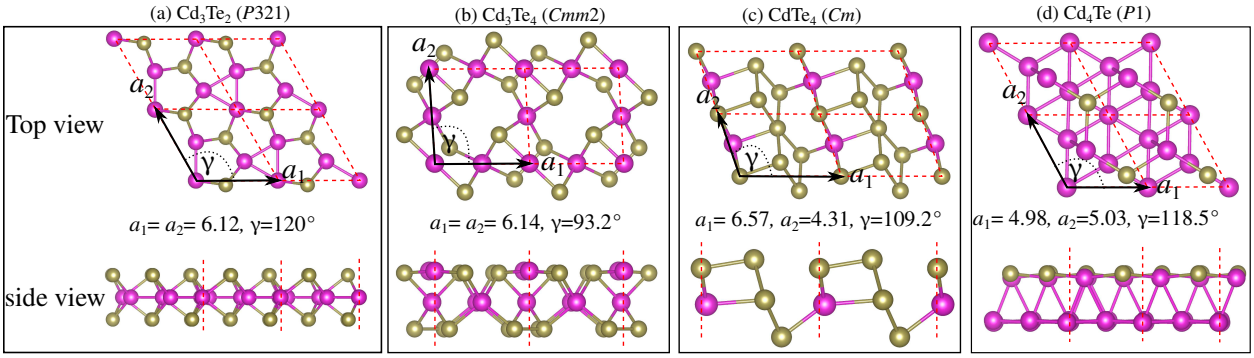

Figure S1: Top and side views of the 2D structures predicted by USPEX in this work, i.e., (a) Cd_3Te_2, (b) Cd_3Te_4, (c) $CdTe_4$, and (d) Cd_4Te. The (red) dashed lines represent the unit cell in each case.

Optimization

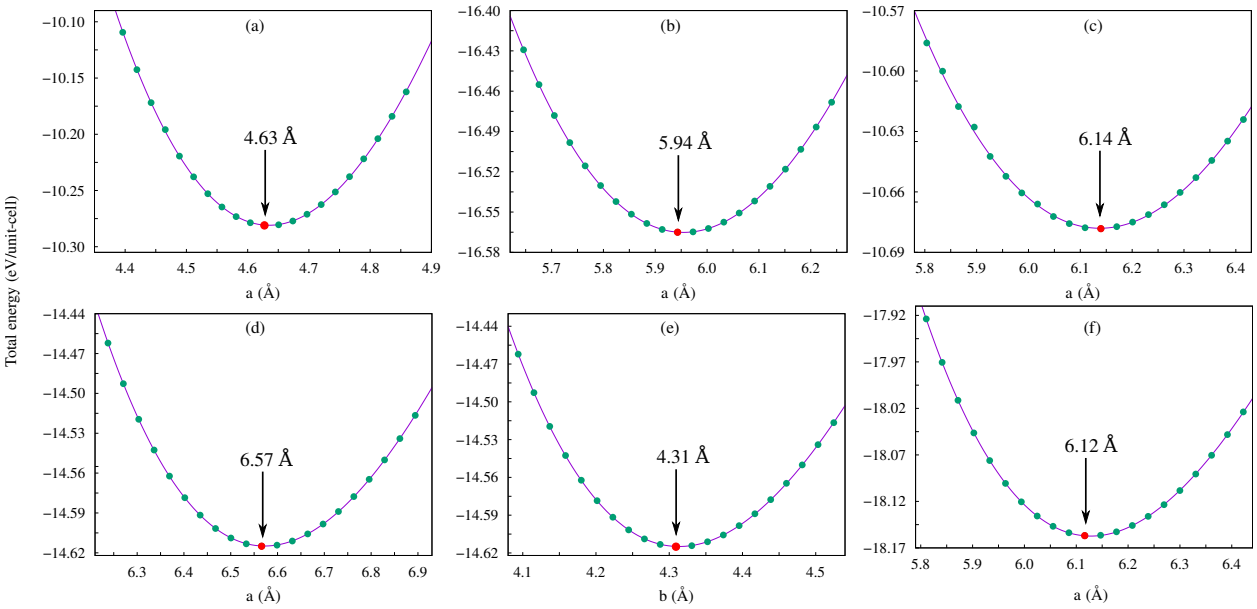

Figure S2: Total energy per unit cell as a function of in-plane lattice constants is plotted for (a) CdTe, (b) $CdTe_2$, (c) Cd_3Te_2, (d, e) $CdTe_4$, and (f) Cd_3Te_4 in their two-dimensional configurations. The solid circles represent DFT data, and the line is the fitting of the Murnaghan equation of state. The solid red circle represents the optimum lattice constant in each figure.

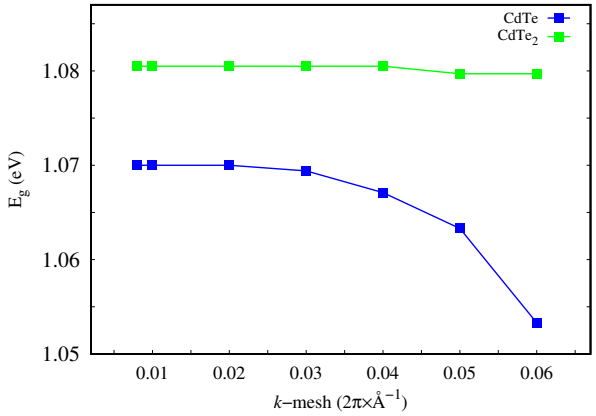

Figure S3: This figure shows the convergence of the band gap with increasing k-mesh in the unit $2\pi \times \text{Å}^{-1}$ for 2D CdTe and $CdTe_2$. The 0.01 is the optimum k-mesh, evident from a negligible variation in the band gap beyond it.

Dynamic stability

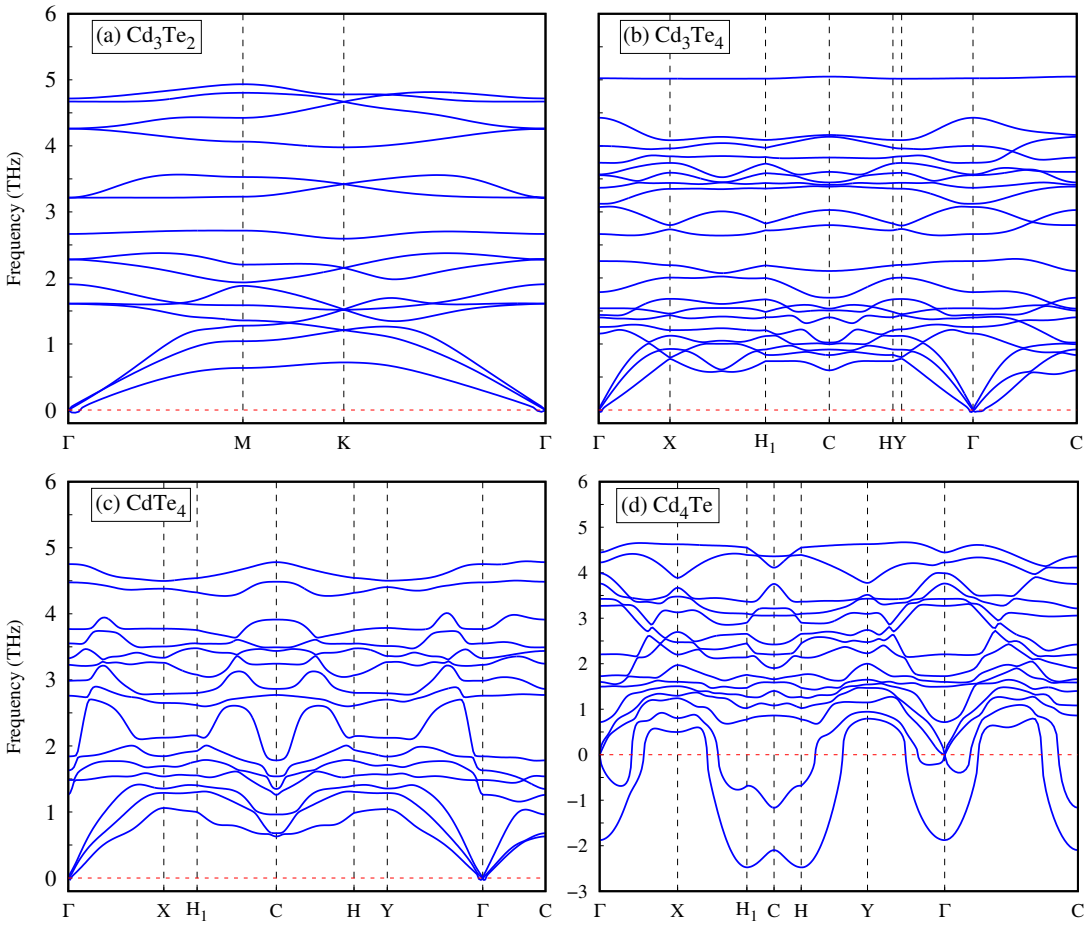

Figure S4: Phonon band structures in the first Brillouin zone of single-layer (a) Cd_3Te_2, (b) Cd_3Te_4, (c) $CdTe_4$, and (d) Cd_4Te. The presence of negative frequencies in phonon spectra of single-layer Cd_4Te shows its dynamical instability.

Thermodynamic stability

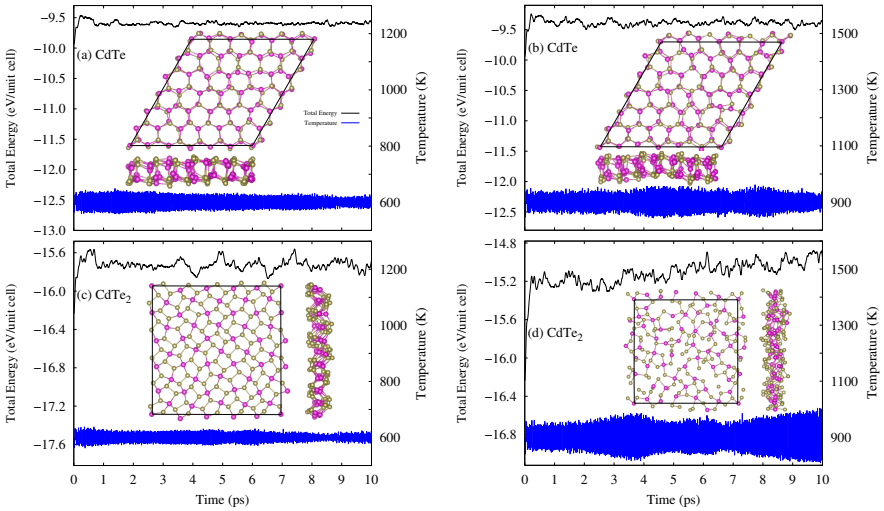

Figure S5: Total energy per unit cell and temperature profiles using *ab initio* molecular dynamics simulations at temperature of (a) 600 K, (b) 900 K for monolayer CdTe and (c) 600K, (d) 900 K for $CdTe_2$. The inset in each case shows the final structure from top and side views after 10 ps.

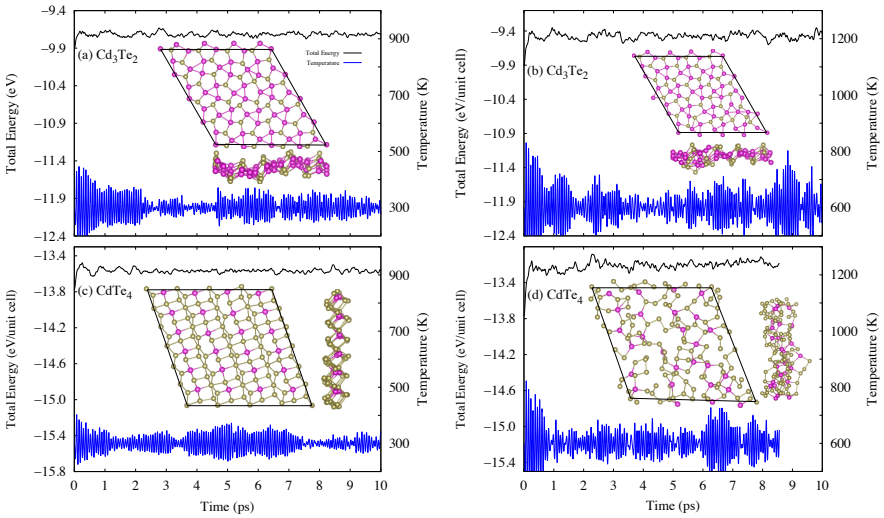

Figure S6: Total energy per unit cell and temperature profiles using *ab initio* molecular dynamics simulations at temperatures of (a, b) 300 K and 600 K for Cd_3Te_2 and (c, d) 300 K and 600 K for monolayer $CdTe_4$, respectively. The inset in each case shows the final structure (supercells of $4 \times 4 \times 1$ for Cd_3Te_2 and $4 \times 6 \times 1$ for $CdTe_4$) after 10 ps. In the case of $CdTe_4$ at 600 K, the AIMD calculations diverge.

Structure analyses

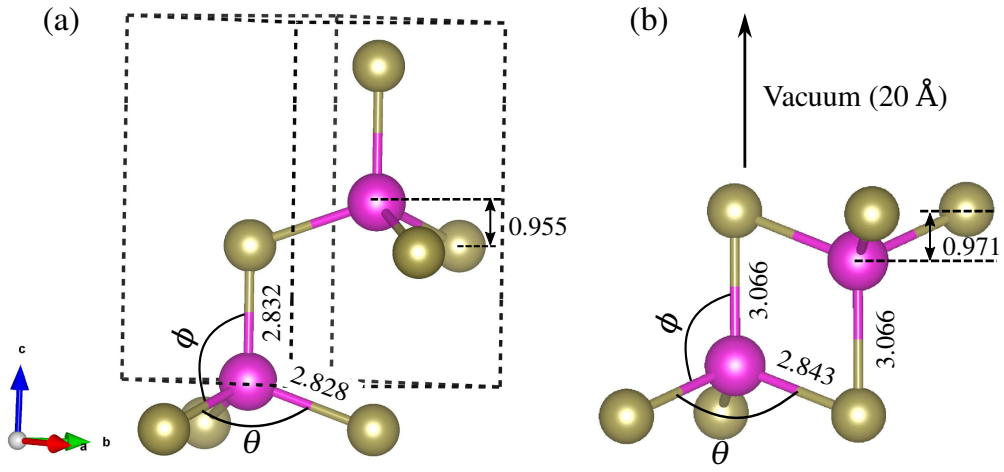

Figure S7: (a) The structure of Bulk CdTe in the wurtzite phase, where the dashed box outlines the primitive unit-cell. (b) The structure of the predicted 2D layer of CdTe by USPEX in this work. The magnitudes of bond lengths and buckling heights are in Å.

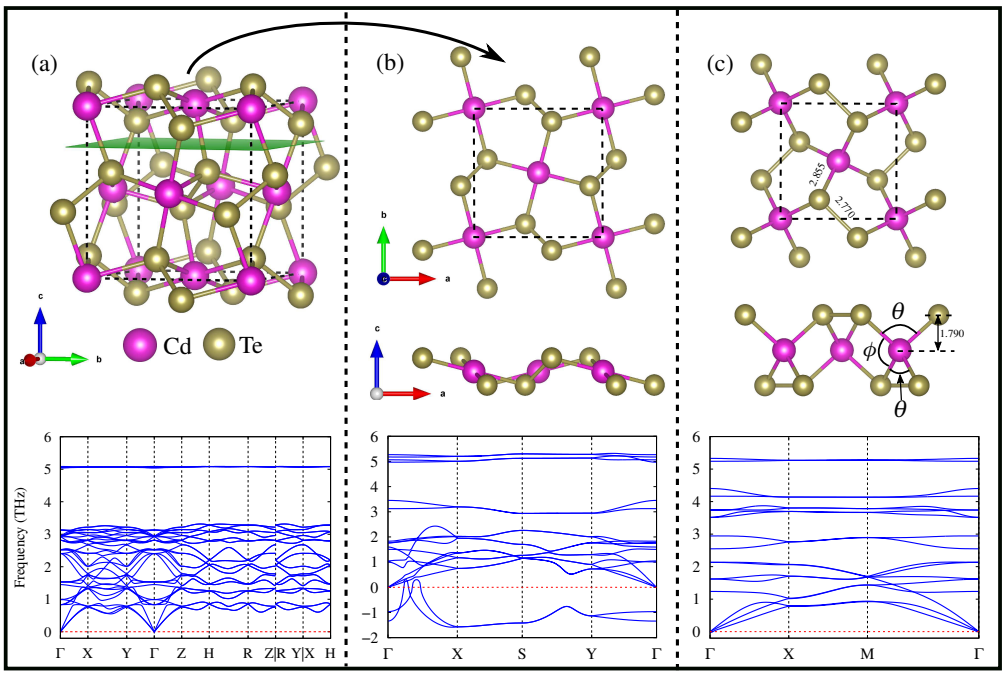

Figure S8: (a) The energetically favorable bulk structure of $CdTe_2$ and its dynamical stability is shown by its phonon spectra. The cleavage plane (001) is indicated by the green color. (b) The top and side views of exfoliated monolayer from the bulk $CdTe_2$, where the negative frequencies in its phonon spectra show the dynamic instability. (c) The top and side views of the stable monolayer $CdTe_2$ predicted in this work. The dashed box in (a) and squares in (b and c) outline the primitive unit cells.

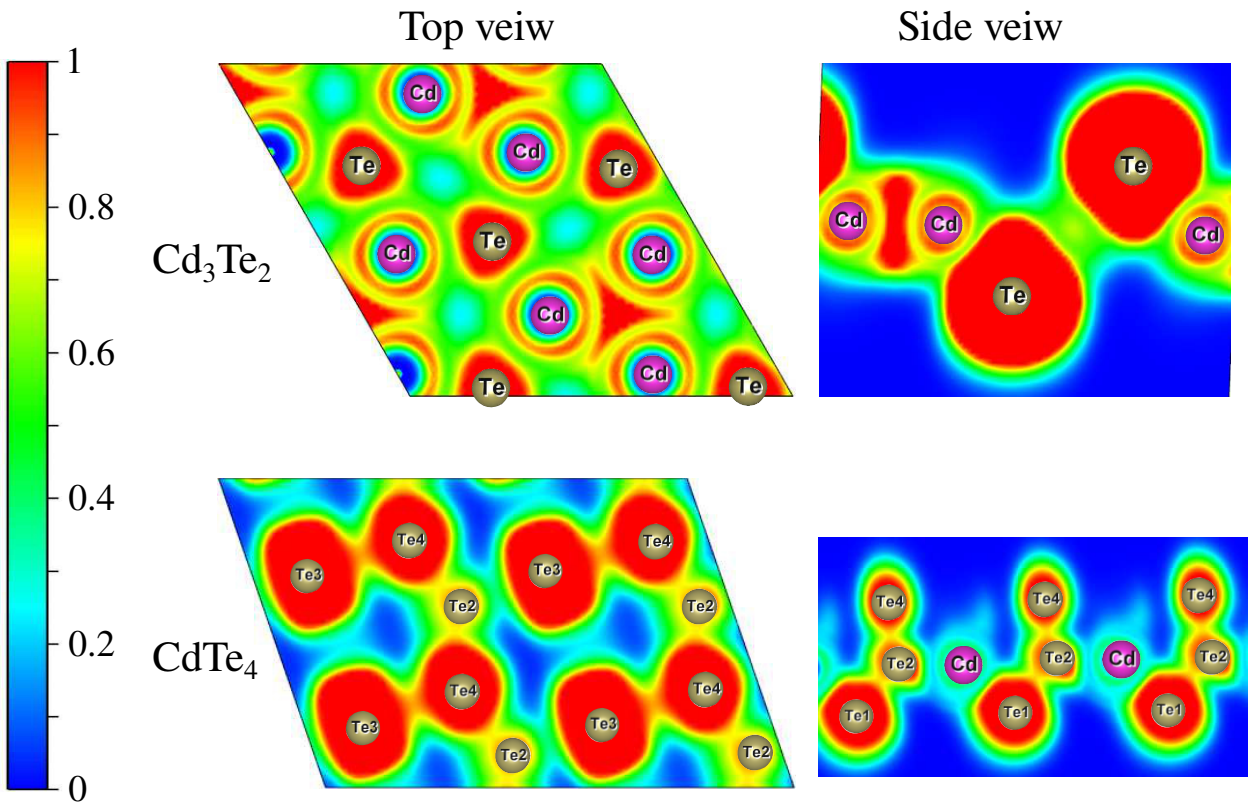

Figure S9: This figure shows the electron localization function (ELF) in crystal planes (001) and (100) of single-layer Cd_3Te_2 and $CdTe_4$. The multi-color bar (left side) scales the value of ELF, where zero and one value correspond to no localization and complete localization of electrons, respectively.

Electronic structures

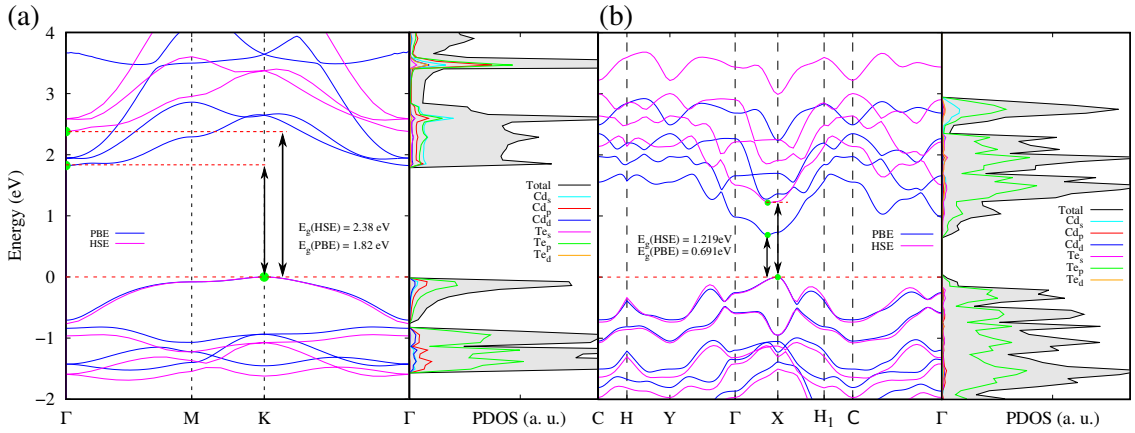

Figure S10: Electronic band structure calculated with PBE and HSE06 functionals and the corresponding PBE atom-projected density of states (PDOS) of Cd and Te atoms for single-layer (a) Cd_3Te_2 and (b) $CdTe_4$. The zero in the energy axis is indicated by the horizontal dashed line, which is set at the valence band maximum (VBM). The band edges (i.e., VBM and CBM) are highlighted with solid green circles, and the fundamental band gaps (PBE and HSE06) are mentioned.

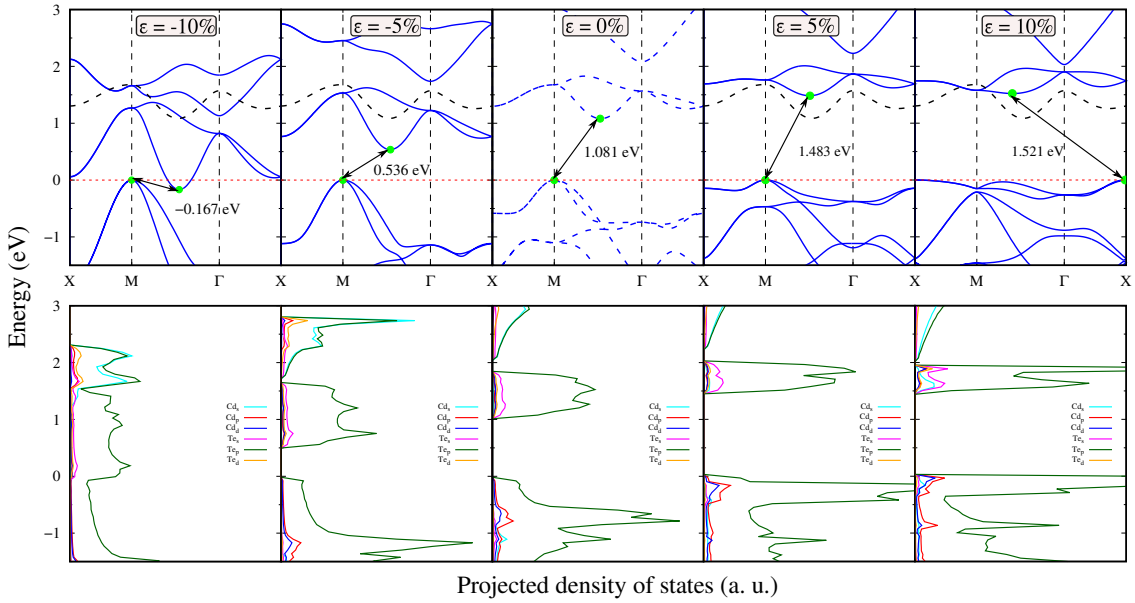

Figure S11: Electronic band structure and atom-projected density of states (PDOS) of unstrained and strained (i.e., tensile and compressive strained up to ±10%) monolayer $CdTe_2$ using PBE functionals. VBM and CBM are indicated with solid green circles, and the zero in the energy axis (red dashed line) is set at VBM in each case. The lowest conduction band of the unstrained band structure (represented by dashed lines) is plotted on all strained band structures for comparison.