Supplemental Information

Band Gap Tuning in $Zn_xCd_{1-x}Te$ Superlattices Through Variable Atomic Ordering

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S1 Modifications to Preconditioner a

Preconditioner a, as given by Teter *et al.* [1], has the form

$$\mathbf{K}_{\mathbf{G}\mathbf{G}'} = \delta_{\mathbf{G}\mathbf{G}'} \frac{27 + 18x + 12x^2 + 8x^3}{27 + 18x + 12x^2 + 8x^3 + 16x^4},$$
$$x = \frac{E_{\text{kin}}(\mathbf{G})}{\sum_{\mathbf{G}'} E_{\text{kin}}(\mathbf{G}')}.$$

When x is a few orders of magnitude away from 1.0, direct implementation of $\mathbf{K}$ can result in numerical imprecisions. We instead use a (nearly) equivalent version:

$$f_1 = \frac{x + 1.5}{(x - 1.14014)x + 1.47904},$$
$$f_2 = \frac{x^2 + 2.25}{(x + 1.64014)x + 1.140944},$$
$$\mathbf{K}_{\mathbf{G}\mathbf{G}'} = \delta_{\mathbf{G}\mathbf{G}'} f_1 f_2 / 2,$$
$$x = \frac{E_{\text{kin}}(\mathbf{G})}{\sum_{\mathbf{G}'} E_{\text{kin}}(\mathbf{G}')}.$$

To modify the above equations to work with the FSM, one replaces the kinetic energy E_{kin} with the 'folded' kinetic energy, E'_{kin}:

$$E'_{\text{kin}}(\mathbf{G}) = \left(E_{\text{kin}}(\mathbf{G}) - E_{\text{ref}}\right)^2.$$

Tables

Interaction	r_0 (Å)	α (eVÅ^{-2})	β (eVÅ^{-2})
Zn-Te	2.7035 (2.6494)	1.377244	0.550420
Cd-Te	2.8518 (2.7948)	1.689228	0.579559

Table S1: Keating force-field parameters used in this study excluding θ_0 (see TABLE S2). r_0 is the ideal bond length, α and β are the bond length and angle force constants, respectively. Two values are listed for r_0 corresponding to the DFT value first and the adjusted empirical value in parentheses.

Interaction	θ_0 (deg.)
Te-Zn-Te	110.61
Te-Cd-Te	108.44
Zn-Te-Zn	113.90
Zn-Te-Cd	109.36
Cd-Te-Cd	105.35

Table S2: Minimum-energy bond angles θ_0 used for the Keating force field. The central atom in the interaction column indicates the central atom in the bonding triplet.

Figures

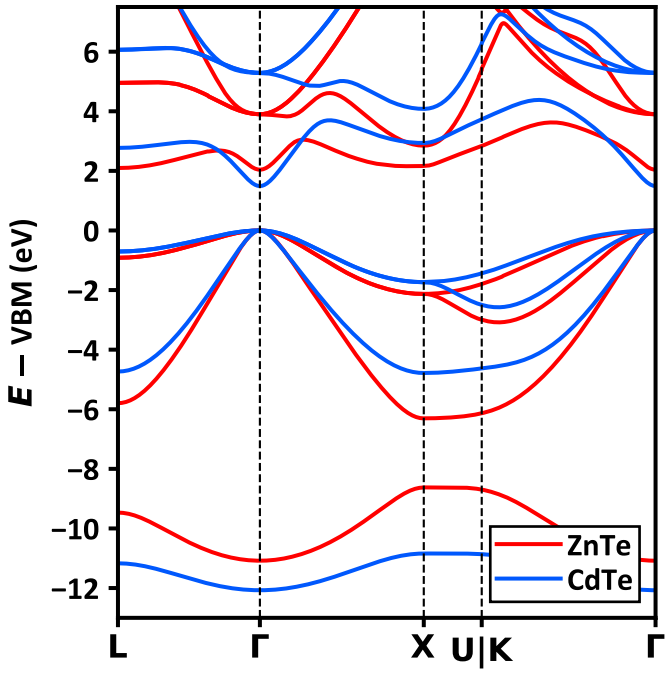

Figure S1: Band structures of ZnTe (red) and CdTe (blue) calculated from empirical pseudopotentials. The special points are as follows: L = $(1/2, 1/2, 1/2)$, Γ = $(0, 0, 0)$, X = $(0, 1/2, 1/2)$, U = $(1/4, 5/8, 5/8)$, K = $(3/8, 3/8, 3/4)$.

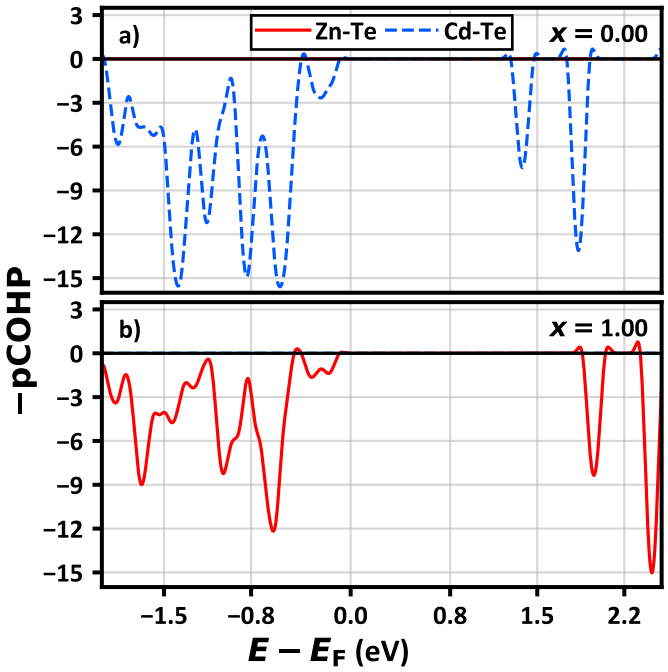

Figure S2: Partial Crystal Orbital Hamiltonian Population (pCOHP) plots for (a) CdTe and (b) ZnTe. The negative pCOHP is plotted so that bonding contributions are below zero on the graphs.

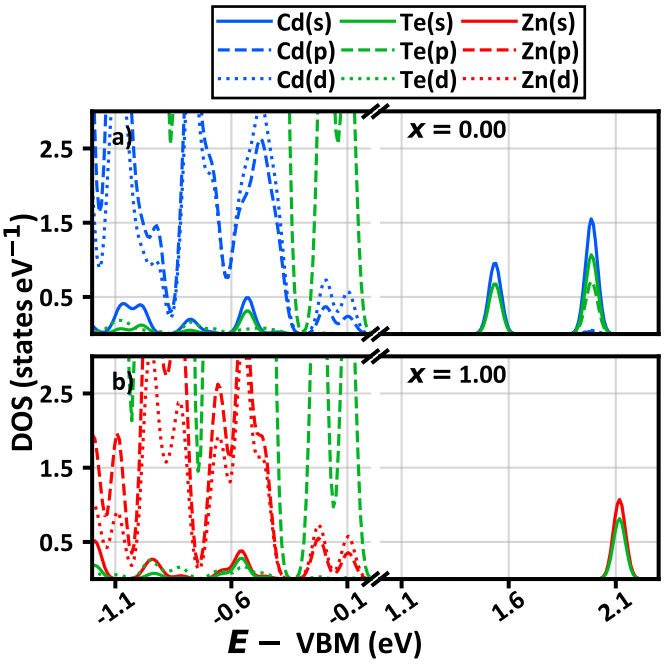

Figure S3: Density of States (DOS) plots for (a) CdTe and (b) ZnTe. The DOS is split into contributions from Zn (red), Cd (blue), and Te (green) atoms as well as their s (solid), p (dashed), and d (dotted) states. Note that the energy axes are broken after the valance band maxima to more clearly show band edge states.

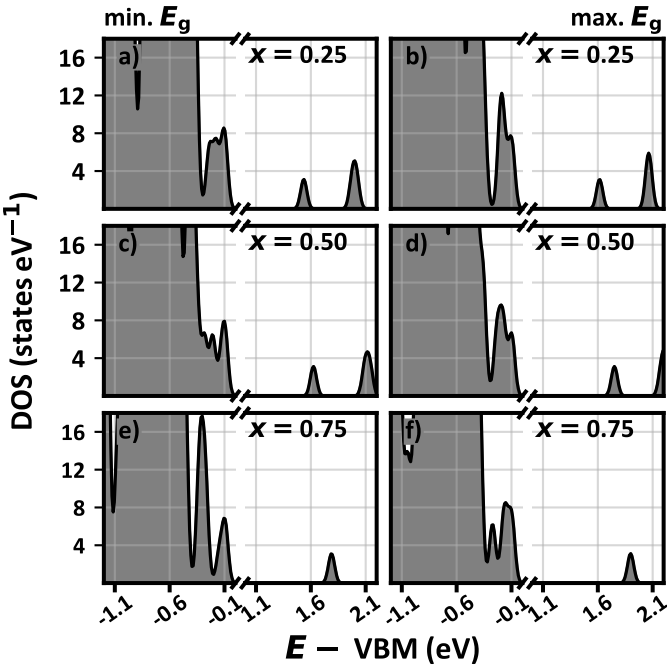

Figure S4: Total DOS plots for minimum ((a), (c), (e)) and maximum ((b), (d), (f)) band gap $Zn_xCd_{1-x}Te$ configurations calculated with HSE06. Note that the energy axes are broken after the valence band maxima to more clearly show band edge states.

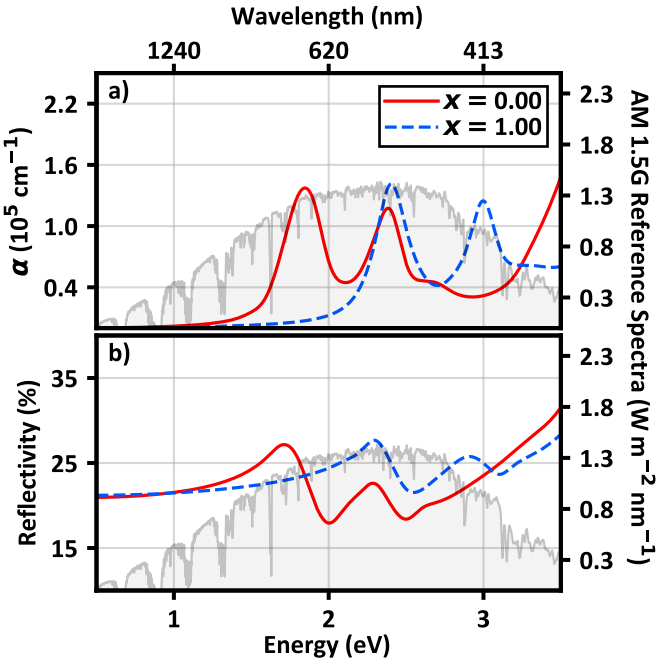

Figure S5: (a) Absorption coefficient α and (b) reflectivity spectra for CdTe (red, solid lines) and ZnTe (blue, dashed lines). The response functions correspond to the left-hand axes. They overlay the AM1.5G solar spectra, shown in grey, the scale of which are on the right-hand axes.

Acknowledgments

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References

[1] Michael P. Teter, Michael C. Payne, and Douglas C. Allan. Solution of Schrödinger's equation for large systems. *Physical Review B*, 40(18):12255–12263, December 1989.