SUPPORTING INFORMATION

Stable Monolayer Transition Metal Dichalcogenide Ordered Alloys with Tunable Electronic Properties

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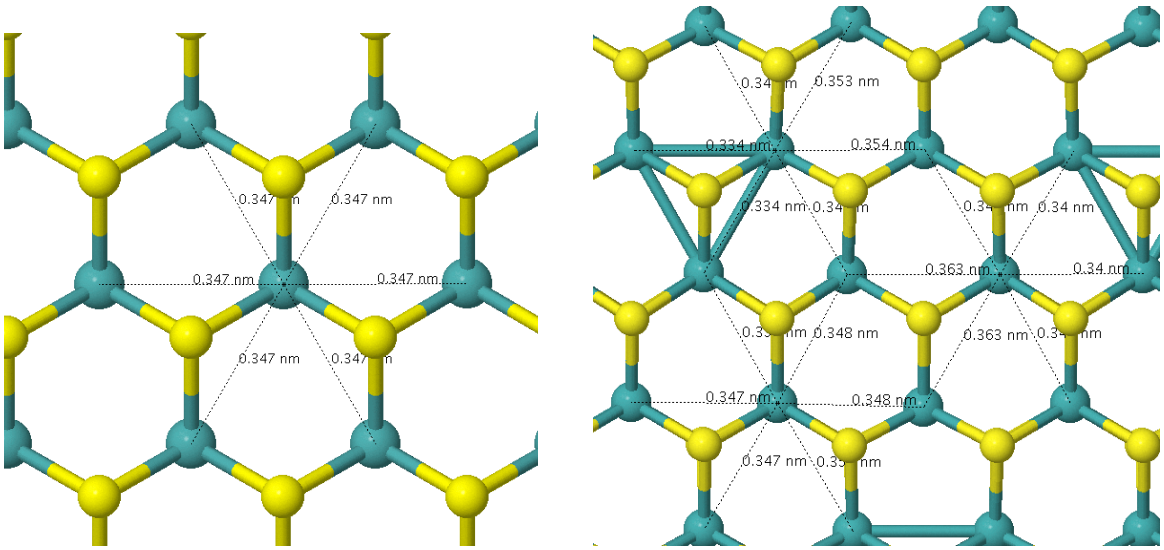
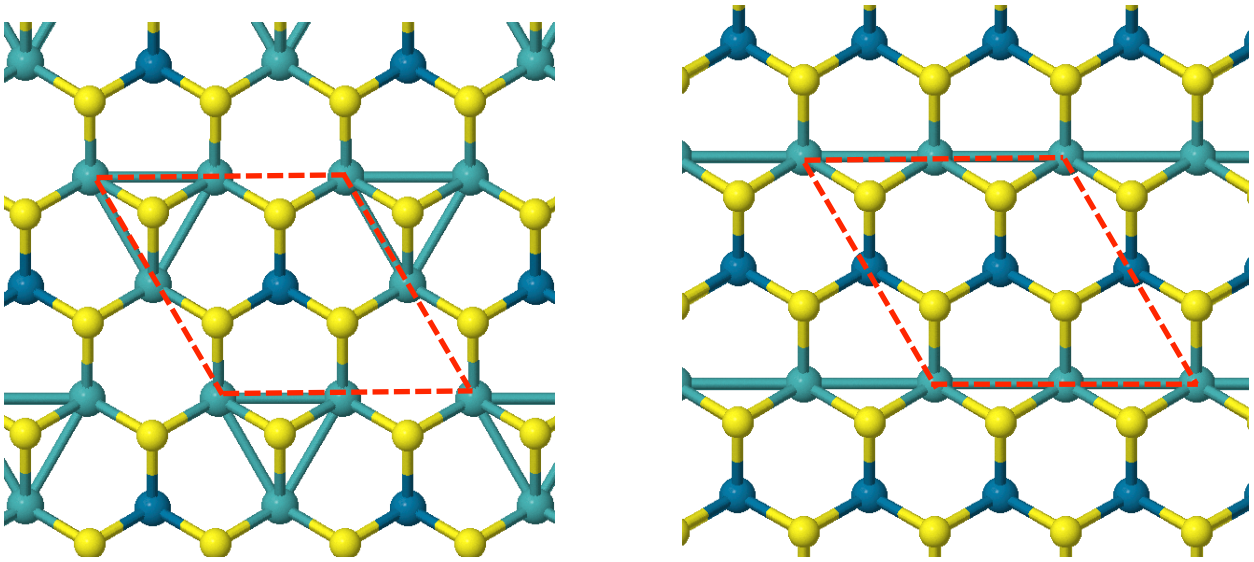
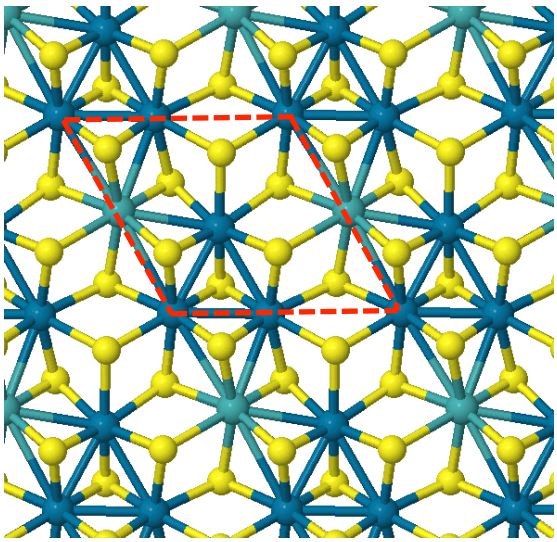


Figure S1. Relaxed structures of $TaSe_2$. Using a 2x2 cell results in a perfect 1H structure (left). A 3x3 cell results in a slightly distorted structure (right), which is 2 meV/atom lower in energy than the 1H structure.



(a) (b)



(c)

Figure S2. Stable structures of (a) $M_{0.75}Re_{0.25}S_2$ and $M_{0.75}Re_{0.25}Se_2$, (b) $M_{0.5}Re_{0.5}S_2$ and $M_{0.5}Re_{0.5}Se_2$ and (c) $M_{0.25}Re_{0.75}S_2$ and $M_{0.25}Re_{0.75}Se_2$, where M is either Nb or Ta (green spheres). Re atoms are blue spheres and S/Se atoms are in yellow. Structures in (a) and (b) are derived from the 1H structure while the structure in (c) is derived from the DT structure.

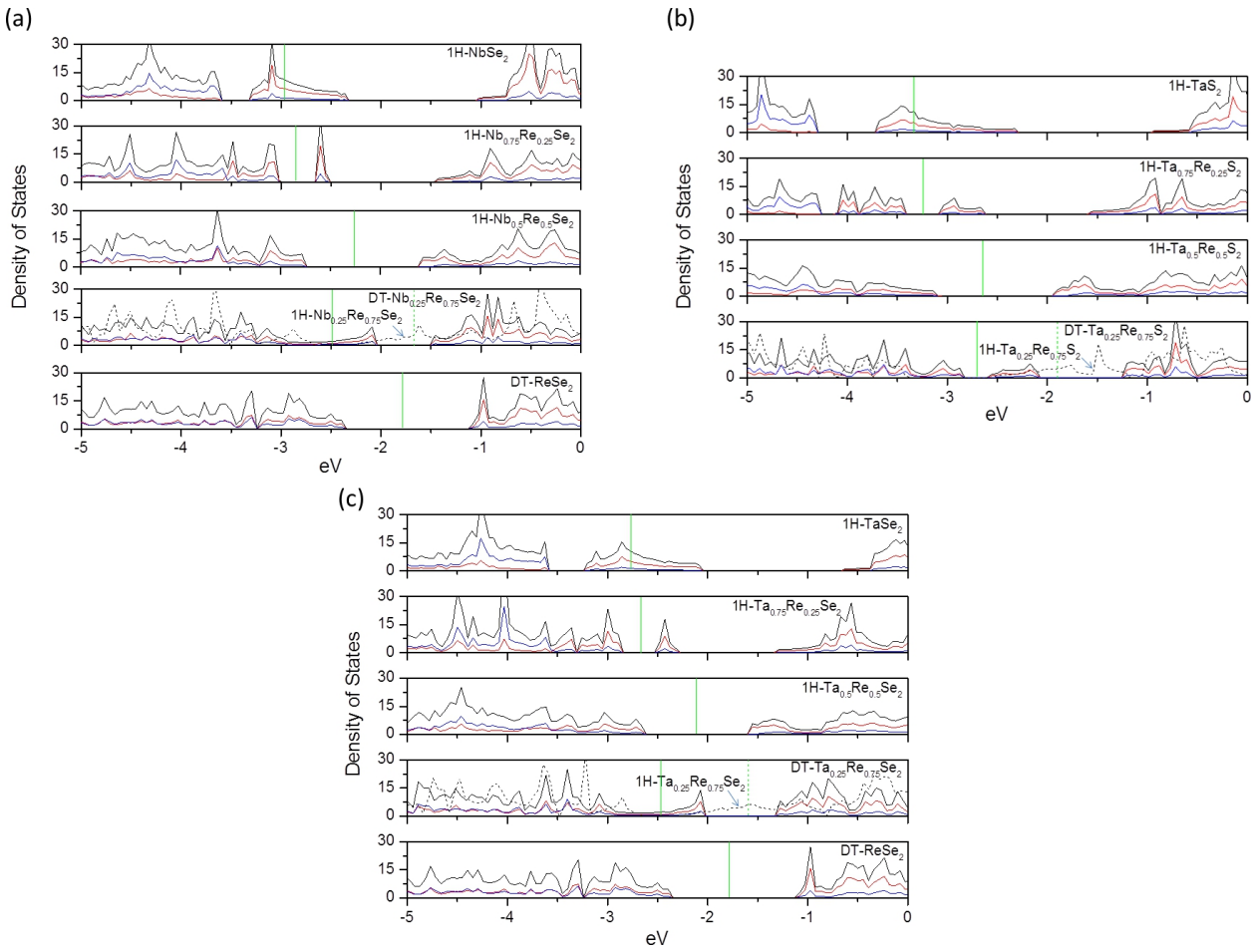


Figure S3. Density of states (DOS) of (a) $Nb_xRe_{1-x}Se_2$, (b) $Ta_xRe_{1-x}S_2$, and (c) $Ta_xRe_{1-x}Se_2$.

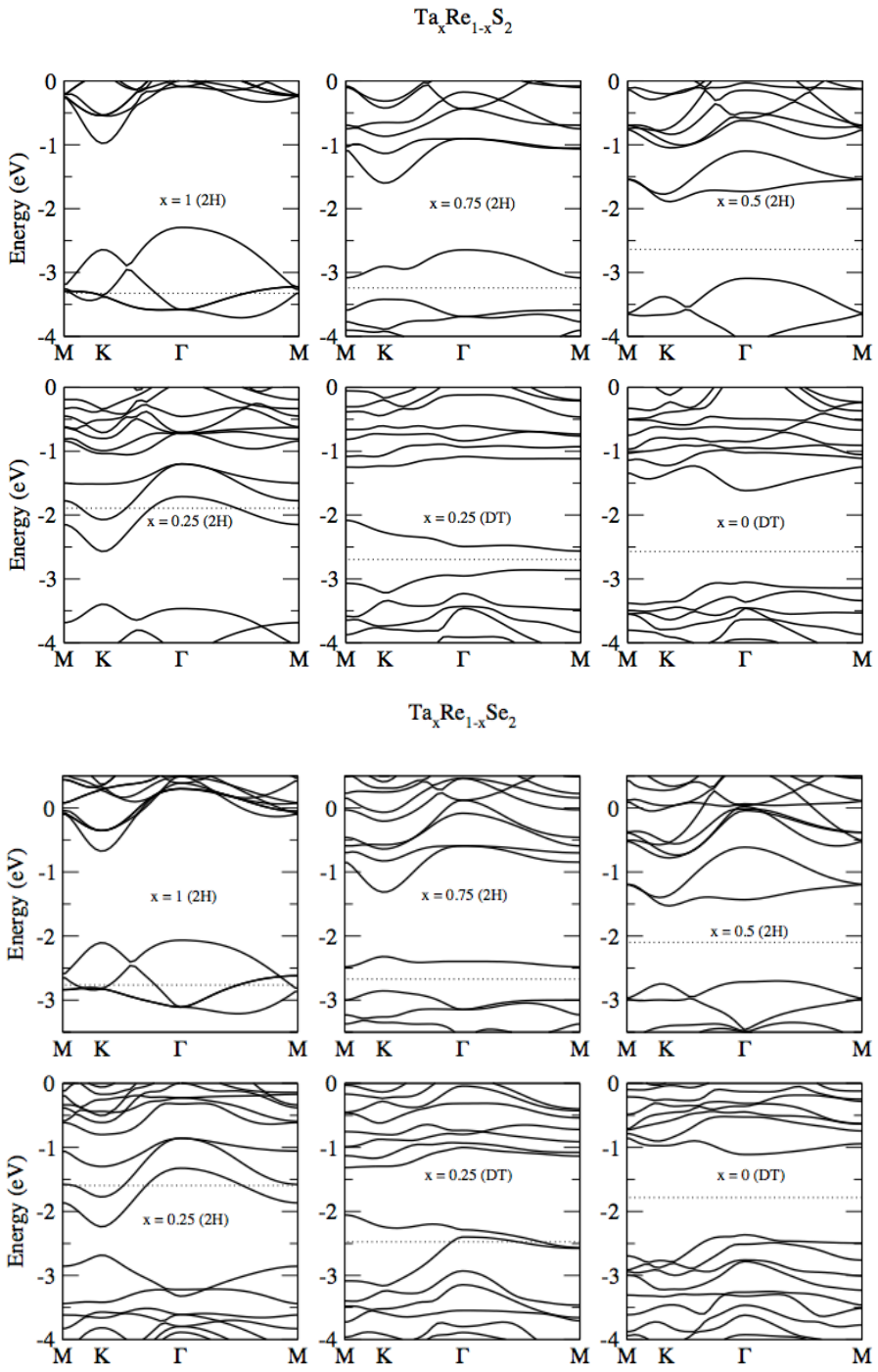


Figure S4. Bandstructures of $Ta_xRe_{1-x}S_2$ and $Ta_xRe_{1-x}Se_2$ systems. The structures used correspond to the lowest energy structures at each composition, except for the 2H structure at x = 0.25, which has a higher energy than the corresponding DT structure.

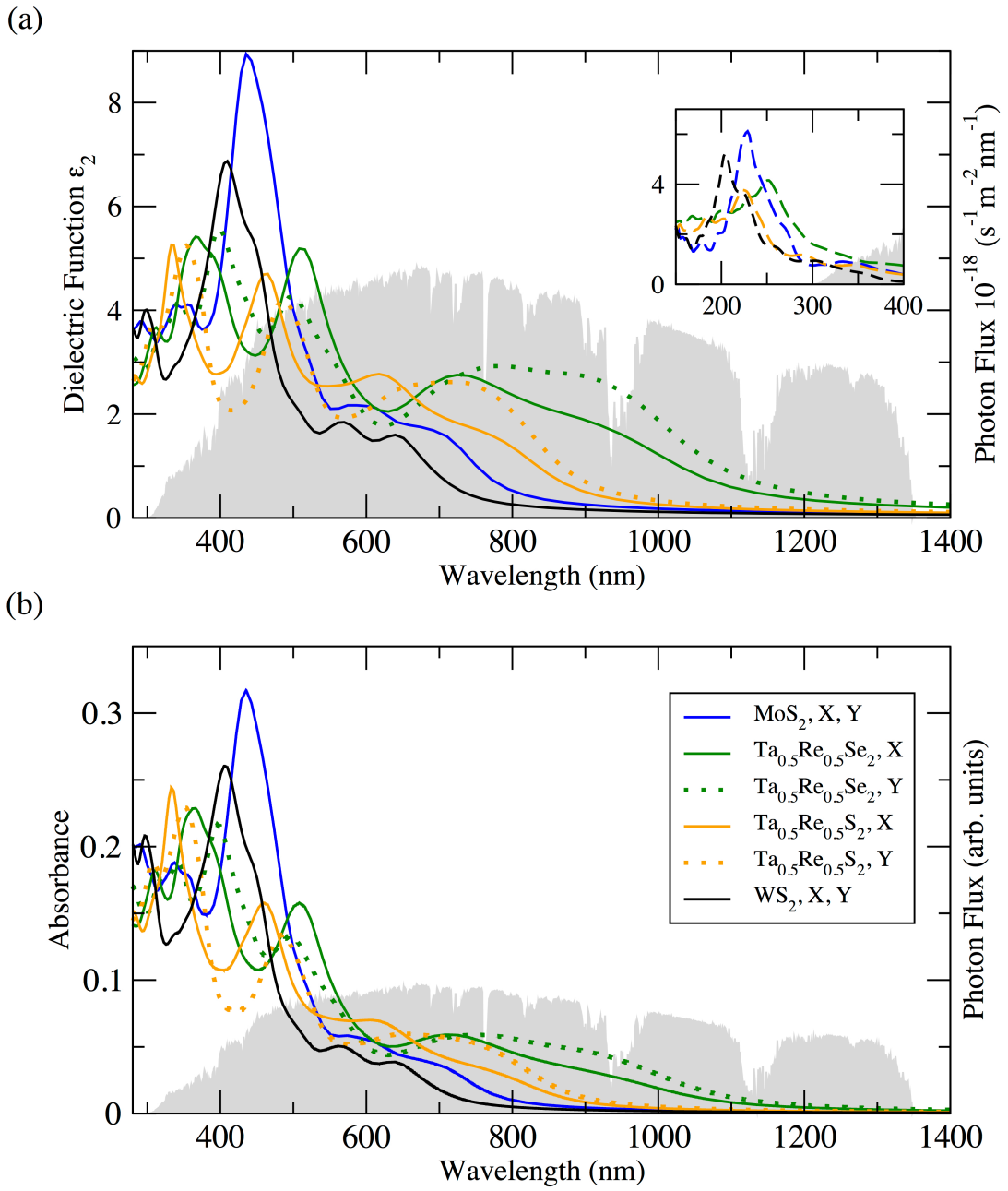


Figure S5. Calculated (a) imaginary part of the dielectric function, ε_2, for MoS_2 (blue), WS_2 (black), $Ta_{0.5}Re_{0.5}Se_2$ (green) and $Ta_{0.5}Re_{0.5}S_2$ (orange) along in-plane X (full) and Y (dotted) directions. Inset shows ε_2 along the direction normal to the monolayer. (b) Derived absorbance along in-plane directions. The photon flux of AM1.5G solar spectrum is shown in grey.