Supplementary Information for Phase behaviour of (Ti:Mo)S_2 binary alloys arising from electron-lattice coupling

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1. 1T' Host

Using the same DFT protocol outlined in the manuscript, the total energy of orderings of Mo and Ti within the 1T' prototype has been computed. This host represents a distorted octahedral geometry, and it is the native coordination of the WTe_2 compound. The formation energy of these configurations is reported as orange crosses in Figure 1 along with the convex hull of the 2H and 1T hosts, as shown in the main text. As shown in Figure 1, the formation energy within 1T' host is comparable with the results obtained within the 1T host. Within 1T' host, relaxed geometries in the Ti-rich portion of the composition axis revert to perfect-octahedral 1T coordination while Mo-rich configurations reach distorted geometries similar to the ones obtained within the 1T host (blue hollow symbols in Figure 1). Thus, the phase behaviour of the system is unchanged, as the relaxed configuration from 1T' and 1T are equivalent in the range $x \in [0, 0.6]$ and the 2H host lies lower in energy for higher concentrations of Mo.

Moreover, the large distortions occurring at all concentrations within the 1T' host complicate the mapping onto a fixed lattice Hamiltonian, which is underpinning the CE formalism, and ATAT software used here fails to train a CE model for the system.

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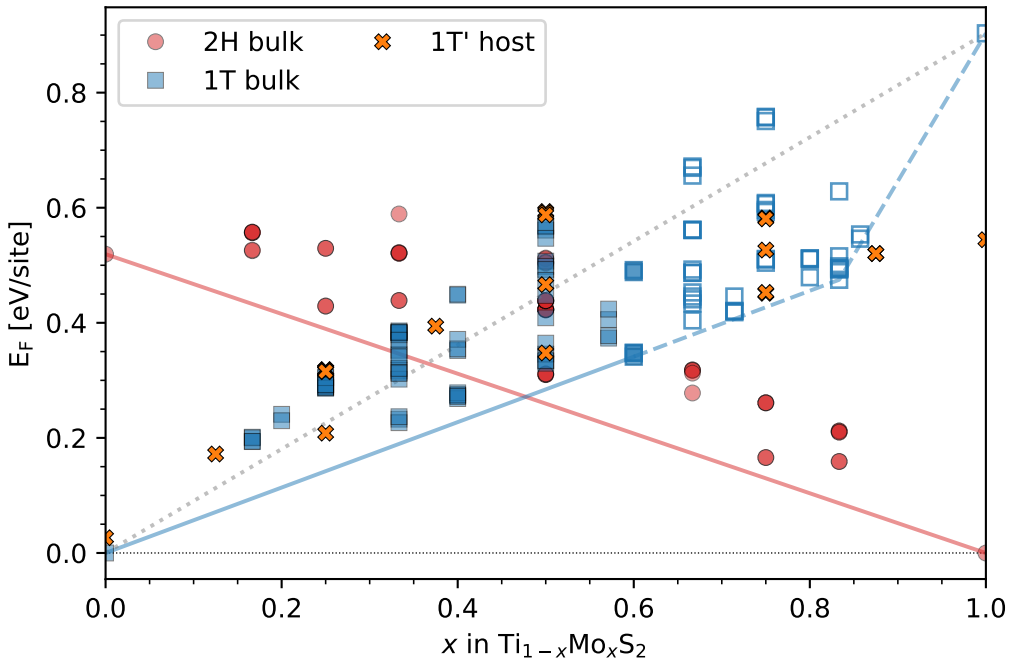

Figure 1: DFT-computed energies for the 2H bulk host (red circles), 1T bulk host (blue squares) and 1T' bulk host (orange crosses). The dashed grey line connects the end-members formation energies of the T host. Red and blue solid lines show the convex hull within the 2H and 1T hosts, respectively. The black dotted line marks the zero-formation energy limit.