Supplemental materials for:

Understanding the trends of phase transition in d^n ($n =$ 0 to 6) transition metal disulfides under carrier injection: A perspective from d-orbital filling

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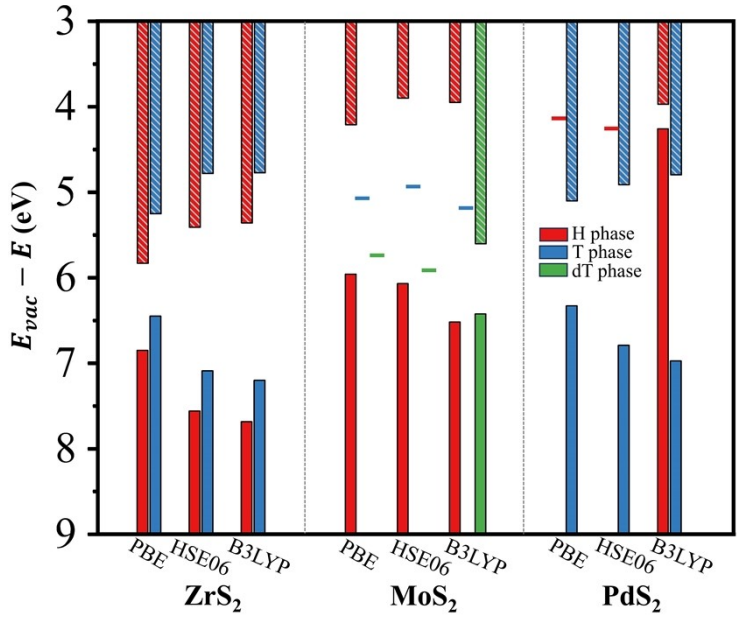

Figure S1. The band edges and work functions (relative to vacuum level E_{vac}) in different phases calculated by PBE, HSE06 and B3LYP methods for ZrS_2, MoS_2, and PdS_2. The solid color-filled regions represent the valence bands, while regions with stripes correspond to the conduction bands for semiconducting phases; and for metalic phases, the energy-position of Fermi level is marked by short lines.

Figure S1 illustrates the band edge relationships of three disulfides (ZrS_2, MoS_2, PdS_2) calculated using different functionals (PBE, HSE06 and B3LYP). It can be observed that although the PBE method is less accurate, the relative energy (the order of band gap sizes) across different phases remains consistent when using PBE, HSE06, and B3LYP methods. This implies that while PBE may underestimate the values of the band gap, it is still capable of correctly reflecting the trends in phase transition.

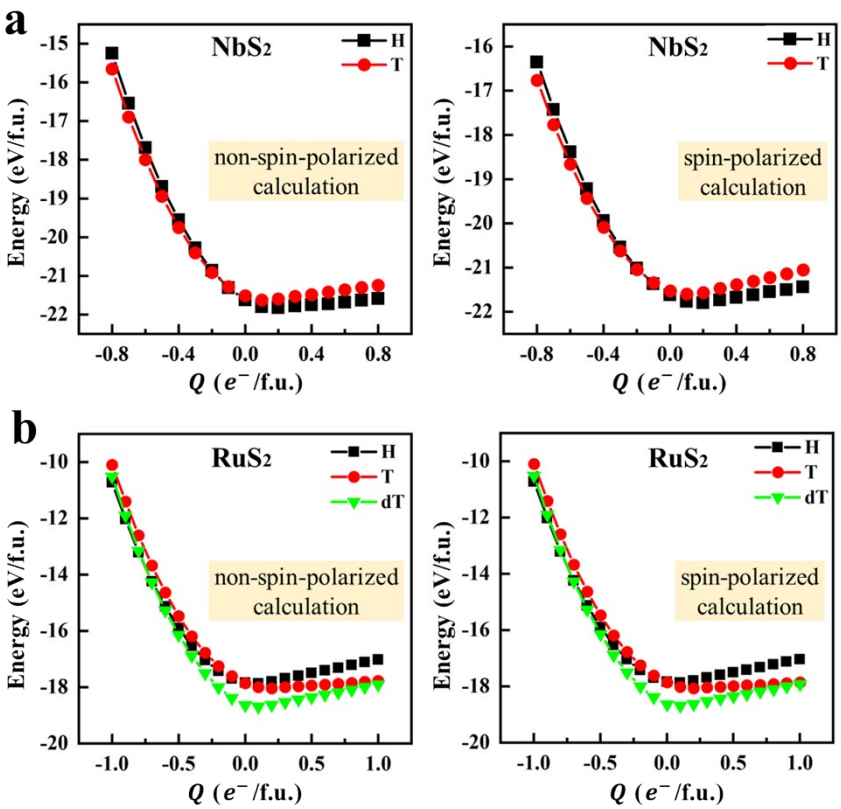

Figure S2. The influence of spin-polarization on PT trends for NbS_2 without spin-polarization (a) and for RuS_2 with spin-polarization (b).

The magnetic moments of NbS_2's H- and T-phases are both zero for the initial state without carrier injection; in this case, considering spin polarization or not with carrier injection does not change the trend of PT. For RuS_2, although the initial magnetic moment of T-phase is sizable (0.64 μ_B), spin polarization again has a negligible impact on the energy change and still does not change the PT trend.

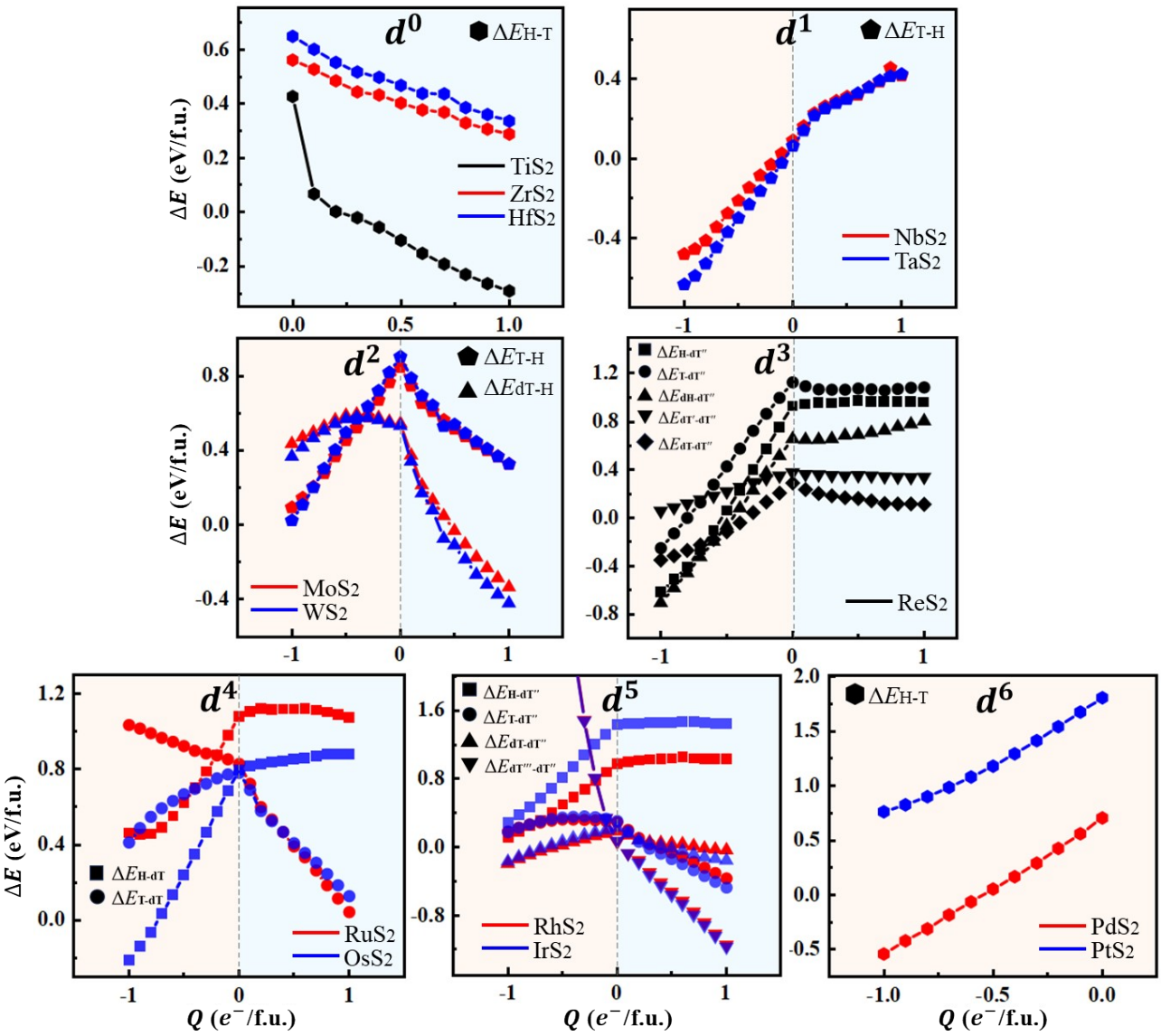

Figure S3. The changes in energy difference (Δ^E) of each phase in MS_2 (d^0 to d^6) with carrier injection by performing full DFT calculations for charged systems. The most stable phase in the initial state was selected as the reference of energy zero. The steepest line in both the electron injection part (Q>0) and the hole injection part (Q<0) corresponds to the favor phase for PT trends. The summary of the PT trends can be found in Table III of the main text. A negative (positive) Q represents hole (electron) injection.

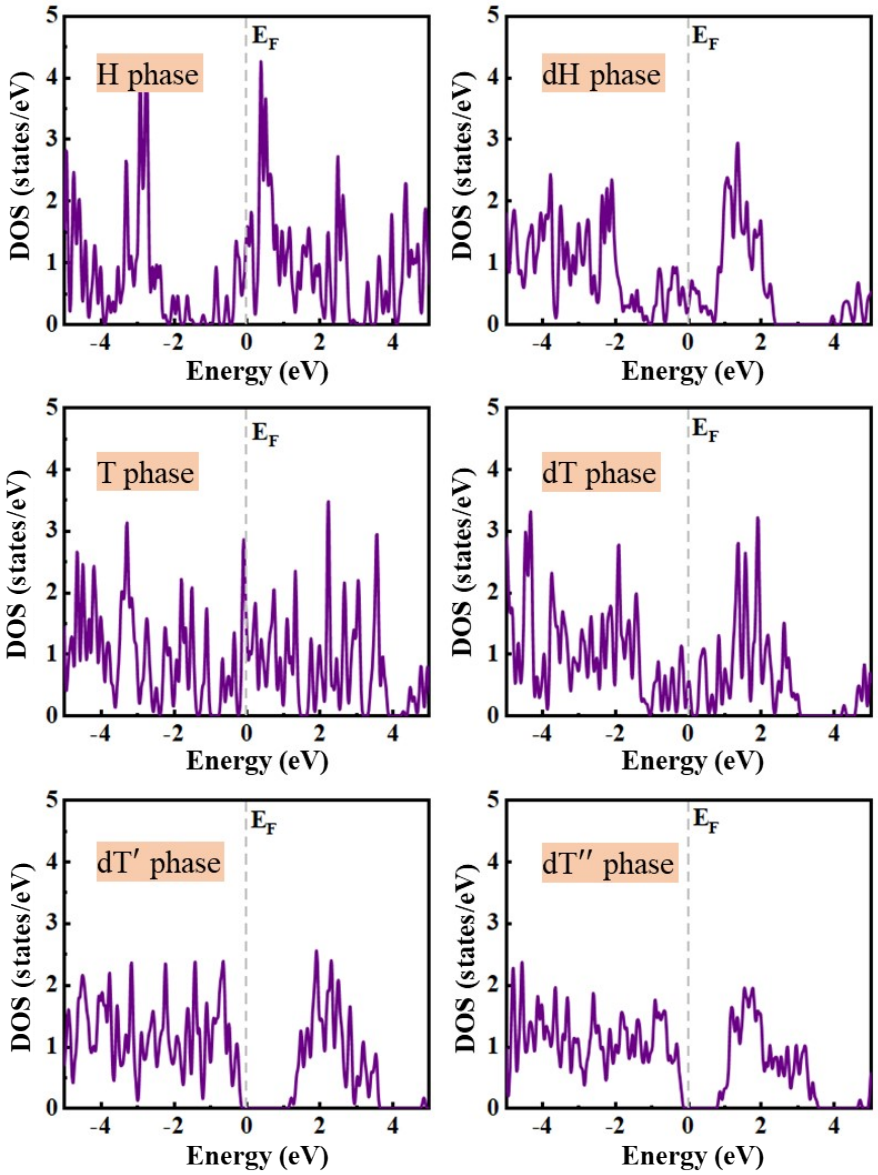

Figure S4. Density of states (DOS) for the six phases (H, dH, T, dT, dT' and dT'') of ReS_2.

Different crystal structures and different distortions exhibit distinct electronic structures. Distortions result in the splitting of degenerate energy levels, leading to the opening of band gaps in the dT' and dT'' phases for ReS_2.

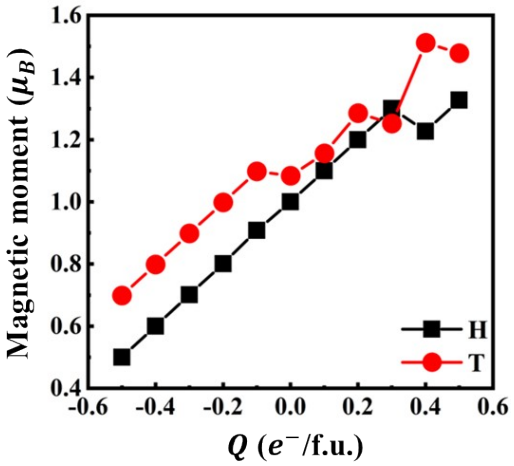

Figure S5. The variation of the total magnetic moment with the carrier injection (Q) for the H- and T-phases of VSe_2, showing an approximate linear relationship between the magnetic moment and Q. A negative (positive) Q represents hole (electron) injection.

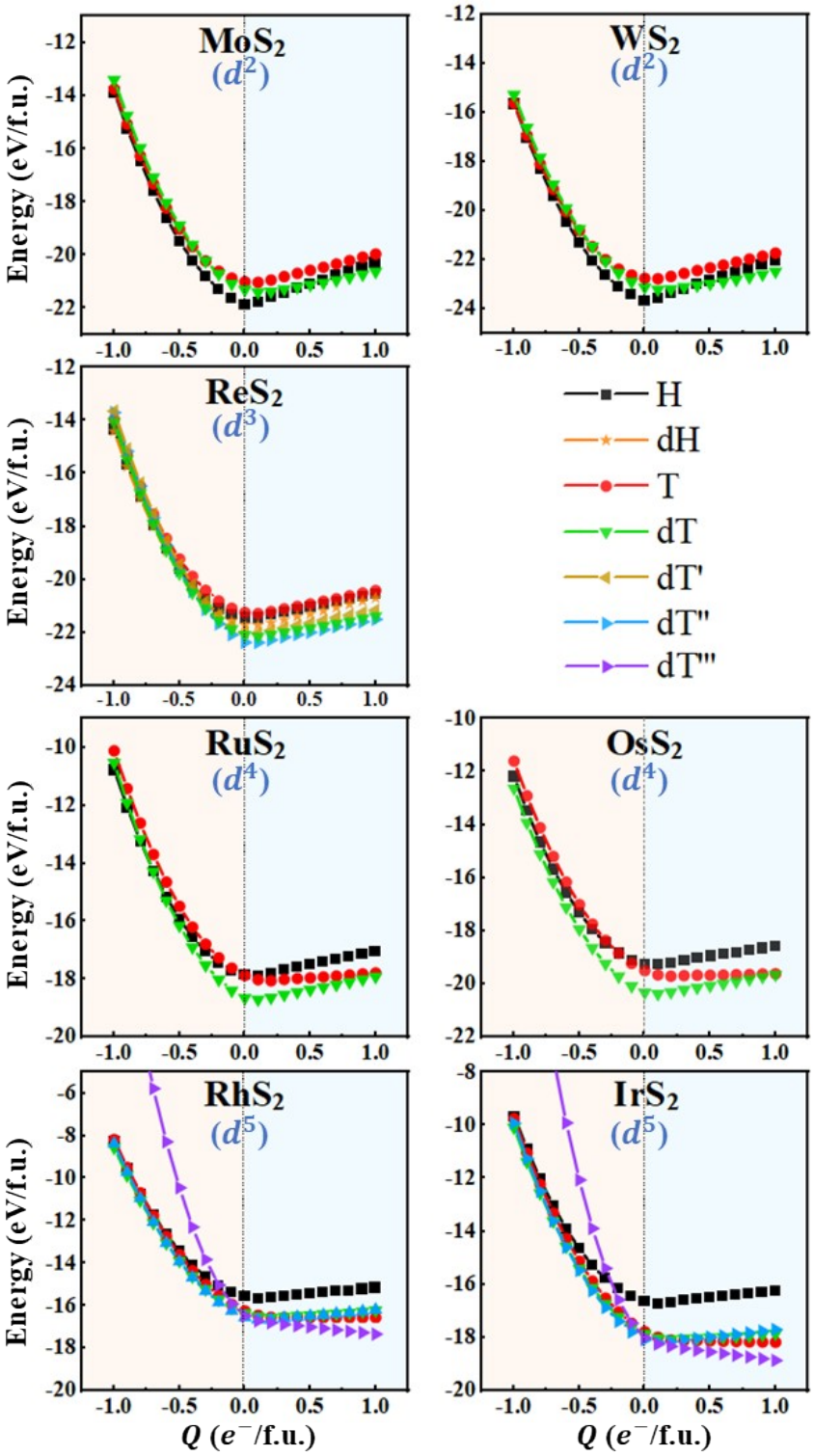

Figure S6. The total energy as a function of carrier injection quantity (Q) for different phases of MS_2 from d^3 to d^5. A negative (positive) Q represents hole (electron) injection.