Supplemental Material: Orbital-selective Mott phase and non-Fermi liquid in FePS$_3$

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I. CALCULATIONS OF STRUCTURAL PHASES IN FePS$_3$ UNDER PRESSURE

The layered material FePS$_3$ is composed of a 3-layer unit where a honeycomb lattice layer of Fe with a vertical P dimer at the Fe hexagon center lies in between two S triangular lattices. At ambient conditions, the 3-layer units are stacked in a staggered way with vdW gaps, adopting the monoclinic space group $C2/m$ (#12) which we call HP0-SPD (staggered P dimers) phase. The material is reported to undergo two structural phase transitions under pressure [1]. The first phase transition involves inter-layer sliding (Fig. S1a, b) at ≈ 4 GPa, making the P dimers almost aligned but still preserving the monoclinic symmetry with the monoclinic angle slightly off 90°. This phase will be called HP1-APD (aligned P dimers). The second phase transition involves inter-layer collapse (Fig. S1b, c) at ≈ 14 GPa that gives rise to the formation of P chains (as opposed to separate P dimers) and changes the symmetry to P$\bar{3}$1m (#162). We call this phase HP2-APC (aligned P chain). The sliding transition is a necessary condition for the realization of HP2-APC since the linear alignment of the P dimers is a structural prerequisite for the formation of P chains that stabilize the collapsed phase HP2-APC. Our DFT calculations for enthalpy reproduce the two structural phase transitions (Fig. S2a, b) in agreement with the recent experimental and theoretical studies [1, 2]. Notable changes in the lattice constant c and the volume occur during the transition from HP1-APD to HP2-APC as expected from the formation of P chains (Fig. S2c, d). We also find that the structural phase transitions remain the same for the non-hydrostatic and the hydrostatic conditions.

II. EFFECT OF HUND'S COUPLING IN HP1-APD

The realization of the OSMP in HP1-APD requires sizable strength of the Hund's coupling. If we artificially set $J_H = 0$ in the calculation of HP1-APD, the OSMP does not appear (Fig. S3), and the PDOS becomes similar to that of HP2-APC in which electrons mostly occupy t_{2g} states. The importance of the Hund's coupling in the realization of the OSMP is in agreement with a theoretical report using three-band model Hamiltonian [3]. Thus, considerable strength of the Hund's coupling (compared with the crystal-field splitting) is essential to realize the OSMP in FePS$_3$.
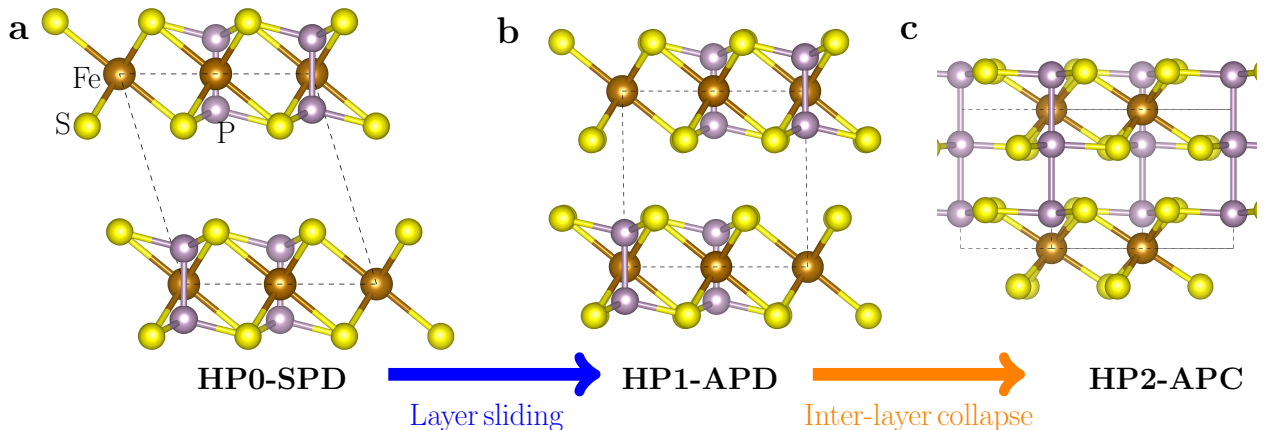

FIG. S1. Crystal structure of FePS$_3$. Atomic structures of (a) HP0-SPD at 0 GPa, (b) HP1-APD at 10 GPa, and (c) HP2-APC at 18 GPa.
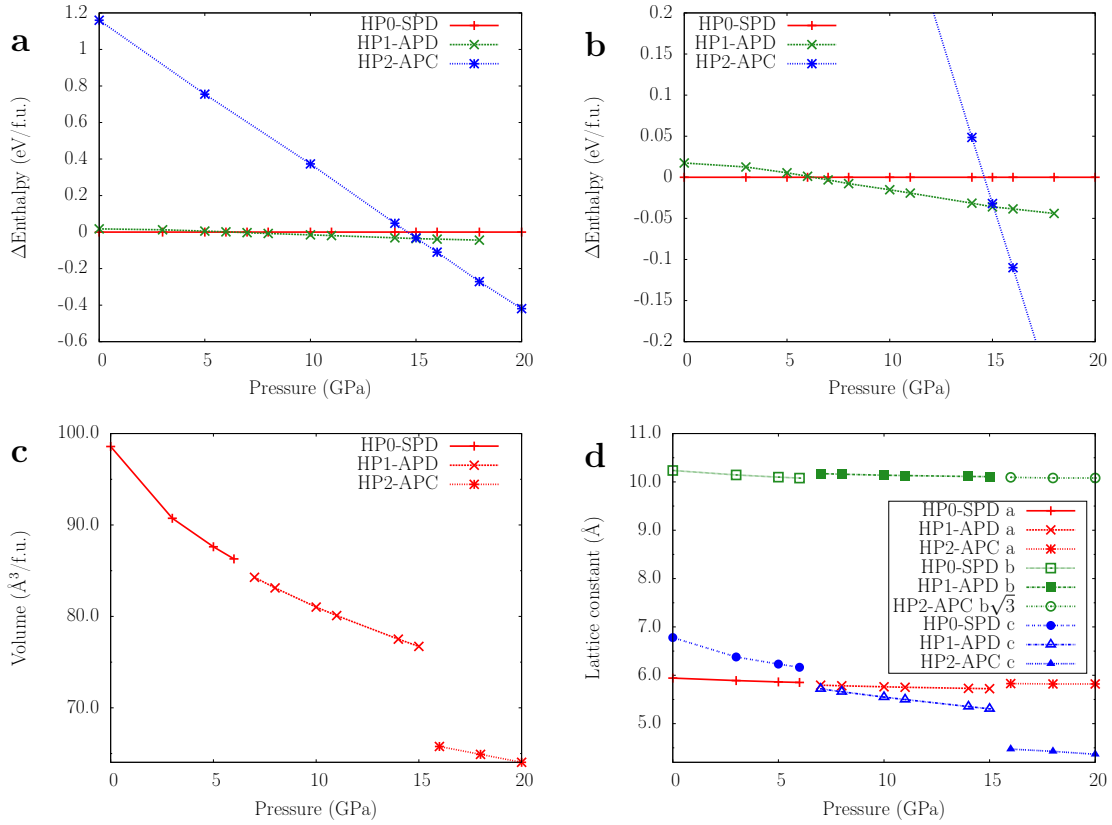
FIG. S2. Structural phase transitions in FePS$_3$. (a) Relative enthalpy with respect to HP0-SPD, and a magnified view in (b). Evolution of (c) volumes and (d) lattice constants under pressure.

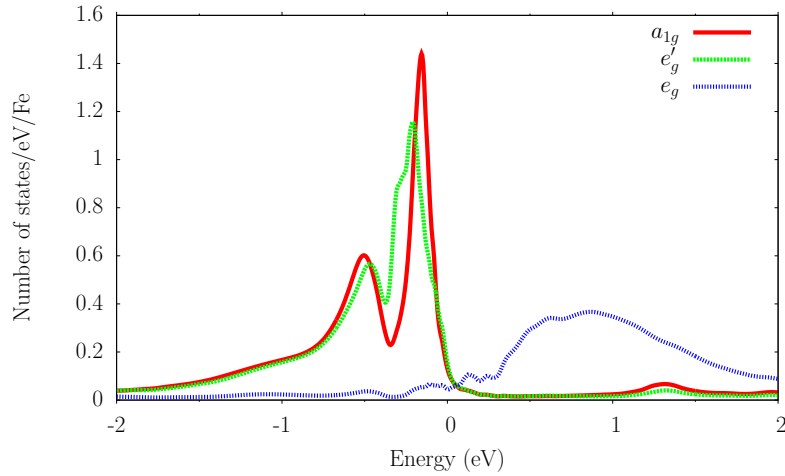FIG. S3. PDOS of HP1-APD at 10 GPa with $U = 8.0$ eV and $J_H = 0.0$ eV.

III.   EFFECT OF ANISOTROPY IN PRESSURE

To understand the effect of anisotropy in pressure on the metallicity of HP1-APD, we calculate PDOS for different values of P_r $(= P_{zz}/P)$ with $P = \frac{P_{xx}+P_{yy}+P_{zz}}{3} = 10$ GPa. We find that larger out-of-plane component tends to favor the metallic HP1-APD realizing the OSMP (Fig. S4). Also, under the hydrostatic pressure (i.e., $P_r = 1.0$), the system has a small gap in agreement with the experiment [4].
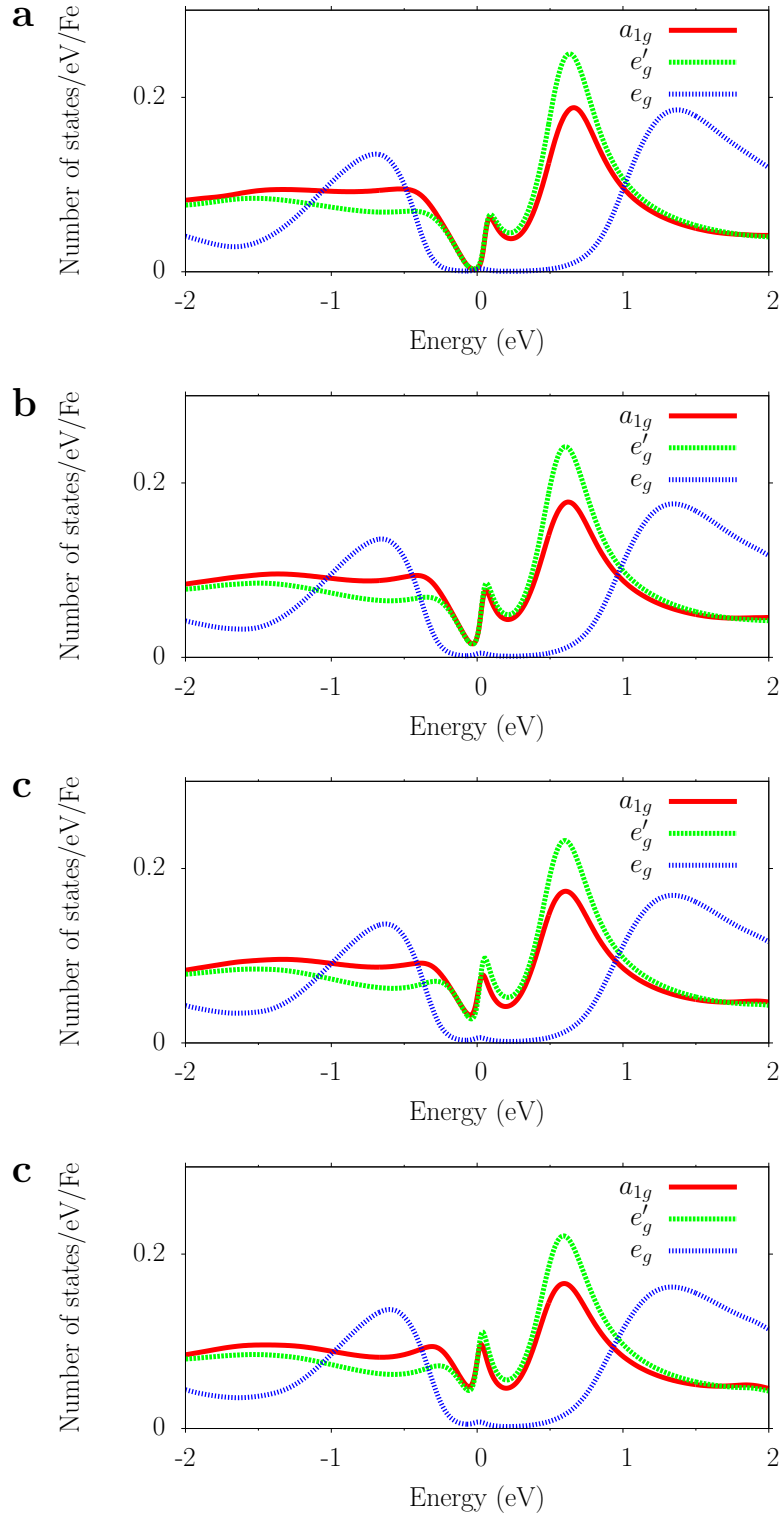
FIG. S4. PDOS of HP1-APD with different anisotropy in pressure at $P = 10$ GPa. PDOS for (a) $P_r = 1.0$, (b) $P_r = 1.2$, (c) $P_r = 1.3$, and (d) $P_r = 1.4$.

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