Electronic Supplementary Information: Thermoelectric Transport of Semiconductor Full-Heusler VFe_2Al

Electronic Structure Analysis

To understand the bonding in VFe_2Al we analyse the COHP and partial density of states for Γ point states. The COHP matrices are quite large, with over 900 elements (see the heat maps in the Figure S3, S5, S7, S9, S11, S13, S15). Since these are symmetric matrices, we are only interested in unique elements in the lower triangle of the matrix. Furthermore, for the sake of the MO diagram construction which requires analysis of bonding interactions, we are only interested in *off-site* COHP terms. To easily locate these terms we drew large boxes drawn across the diagonal of the heat map (see thick black lines in Figure S3, S5, S7, S9, S11, S13, S15). The *off-site* COHP terms can be found outside these boxes. Finally, to simplify our analysis in the construction of the Molecular Orbital diagram we consider only those COHP terms for which the atomic orbitals involved have a significant contribution in the orbital projected partial density of states (see Figure S2, S4, S6, S8, S10, S12, S14).

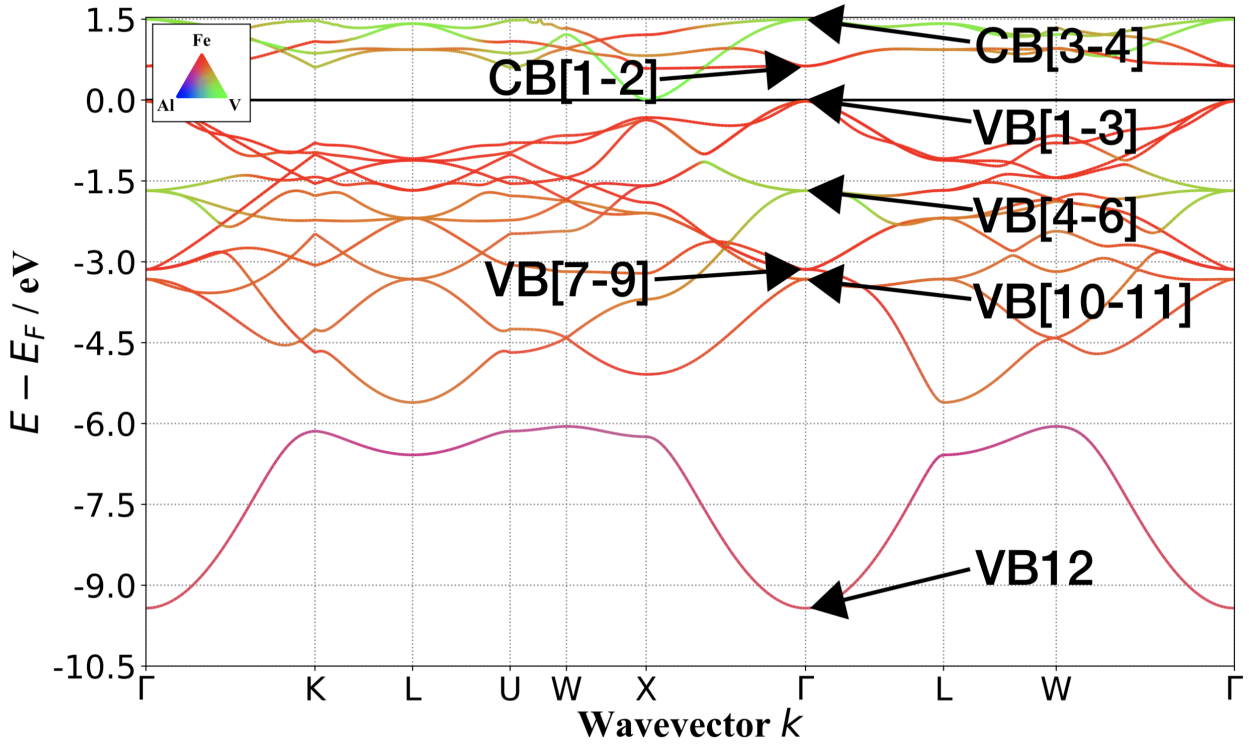

Figure S1 Atom resolved electronic band structure of VFe_2Al. All states containing the 24 valence electrons in the compound are shown. The bands in both the valence and conduction bands are indexed. These indices will be used in the following Figures (Figure S2- S15) which show the orbital projected partial density of states (pDOS) and k-resolved Crystal Orbital Hamilton Population (COHP) analysis of all states at the Γ-point.

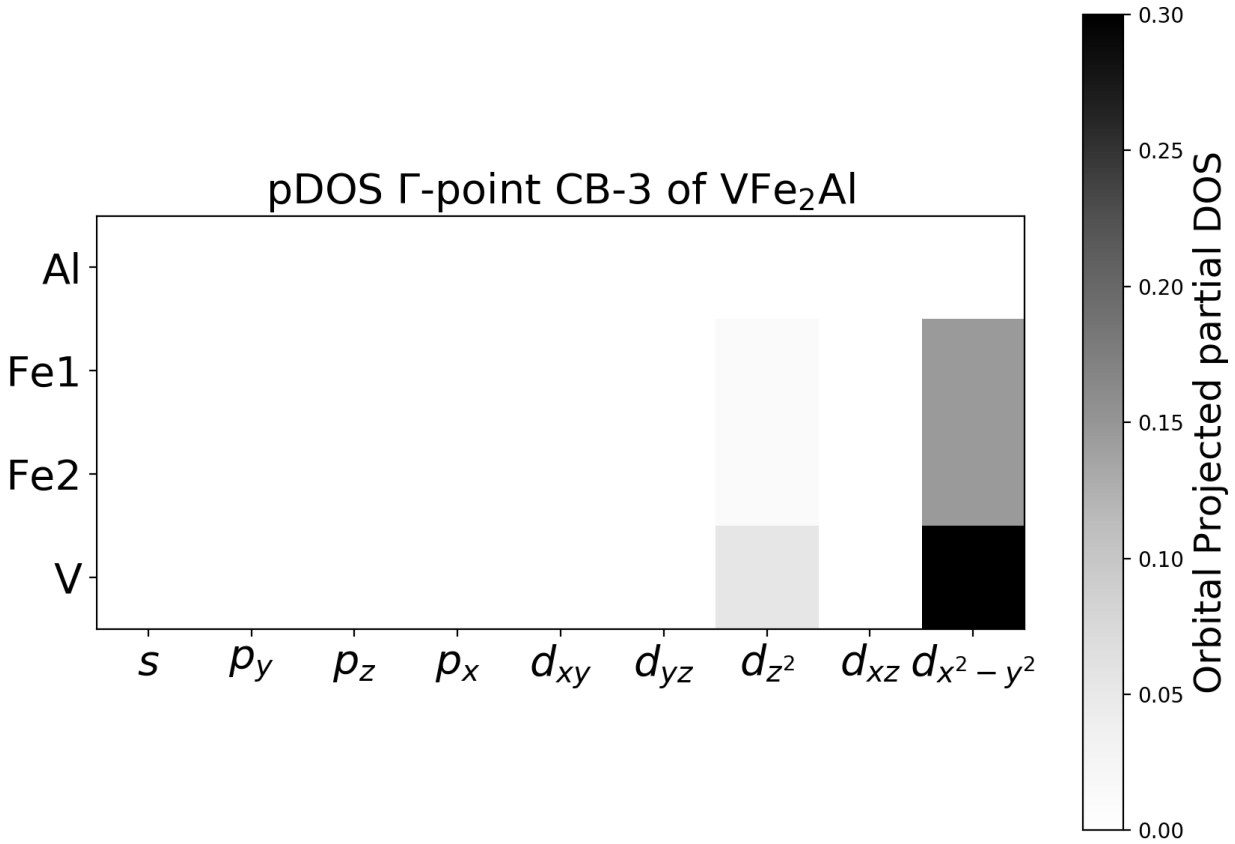

Figure S2 Orbital projected partial density of states heat map for the third Γ point conduction band.

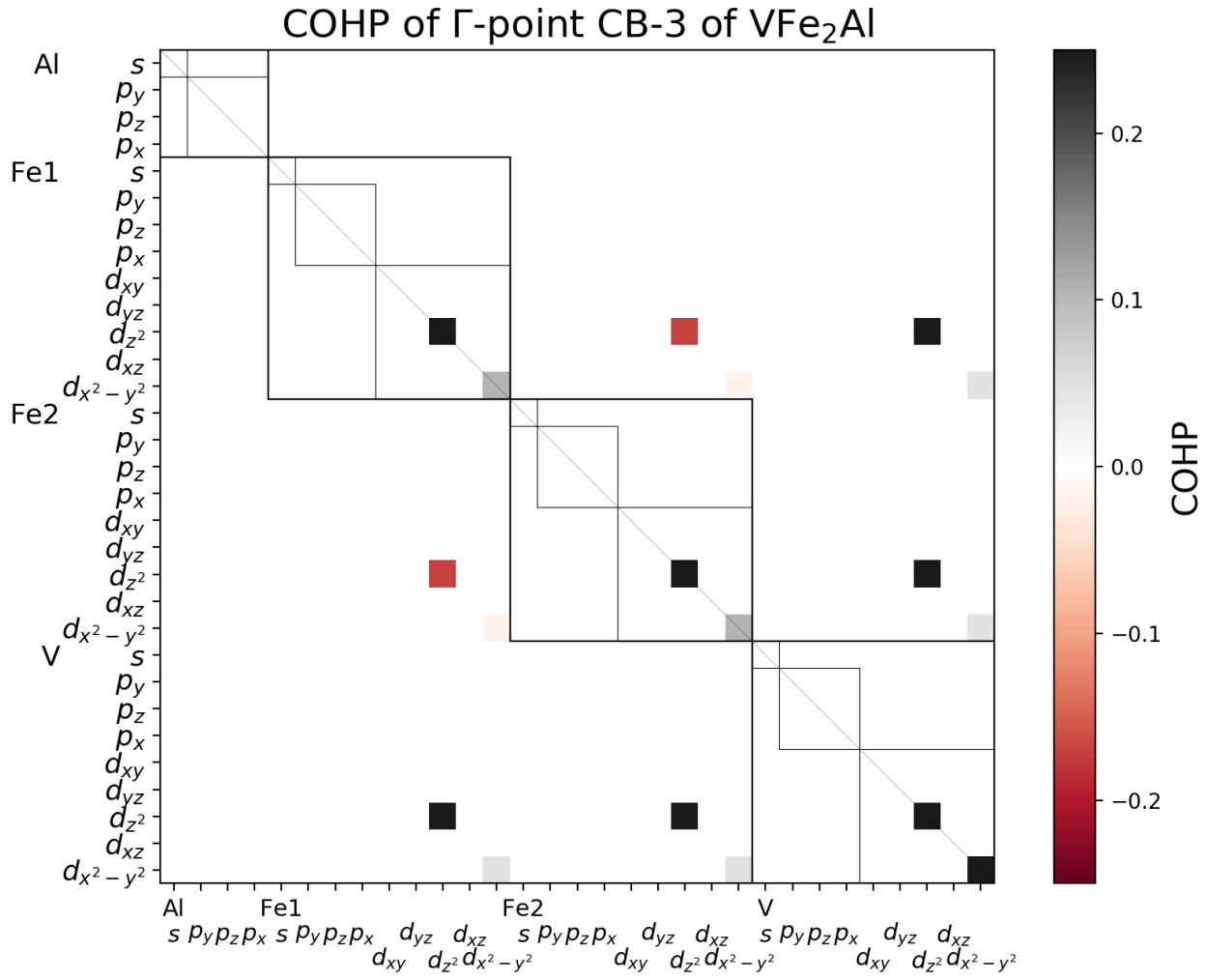

Figure S3 Crystal Orbital Hamiltonian Population (COHP) heat map for the third Γ point conduction band. The *off-site* COHP terms can be found outside the 4 large boxes (in thick black lines) drawn across the diagonal of the heat map.

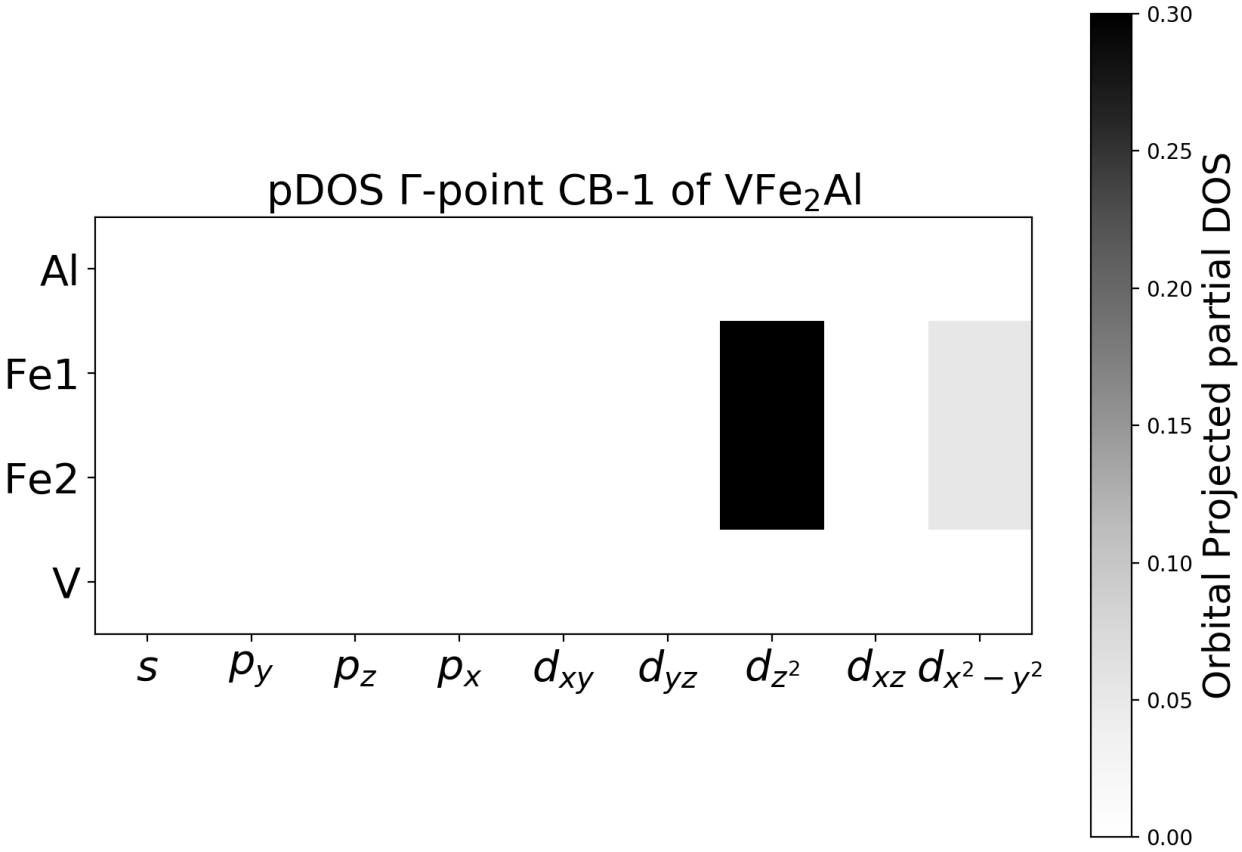

Figure S4 Orbital projected partial density of states heat map for the first Γ point conduction band.

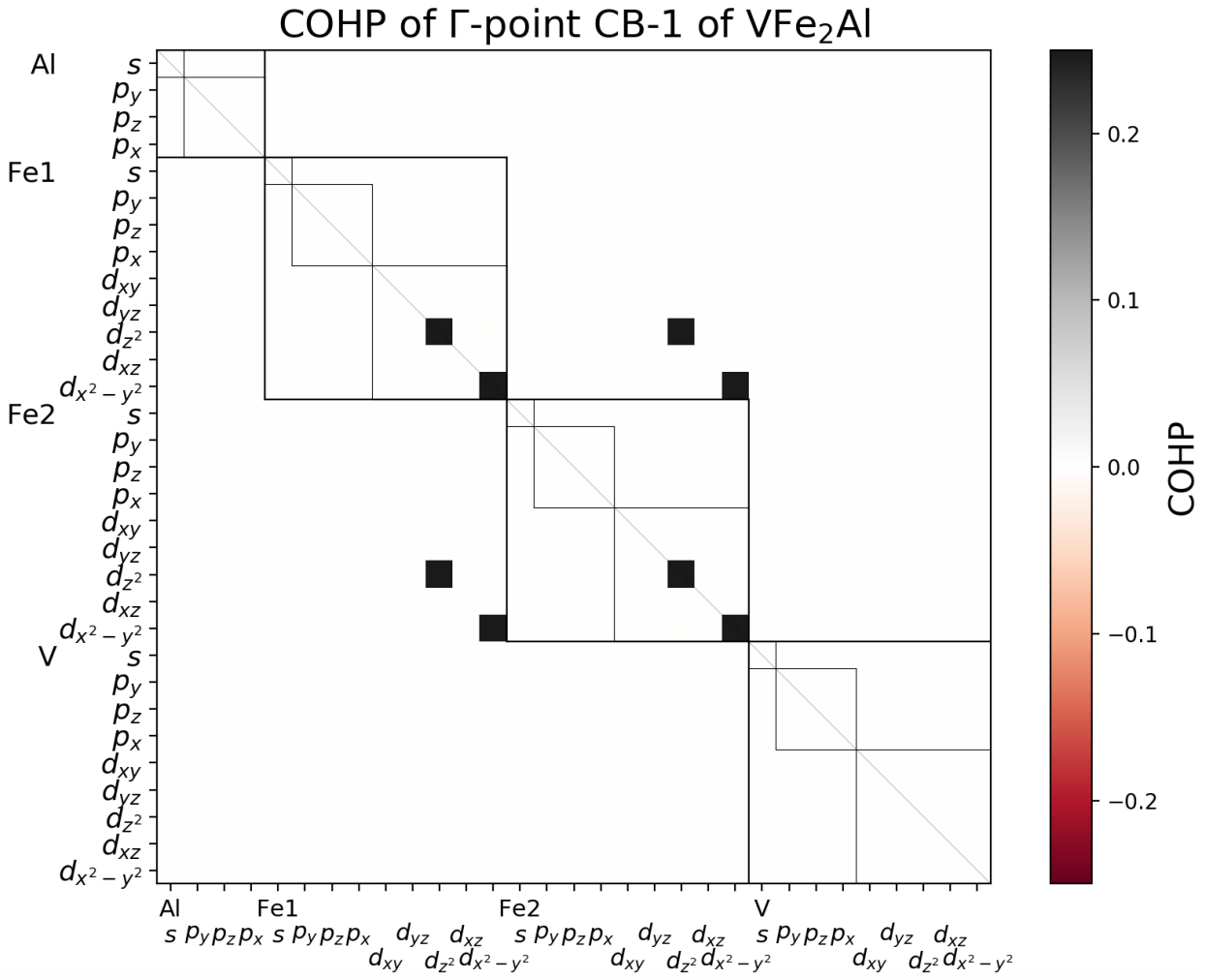

Figure S5 Crystal Orbital Hamiltonian Population (COHP) heat map for the first Γ point conduction band. The *off-site* COHP terms can be found outside the 4 large boxes (in thick black lines) drawn across the diagonal of the heat map.

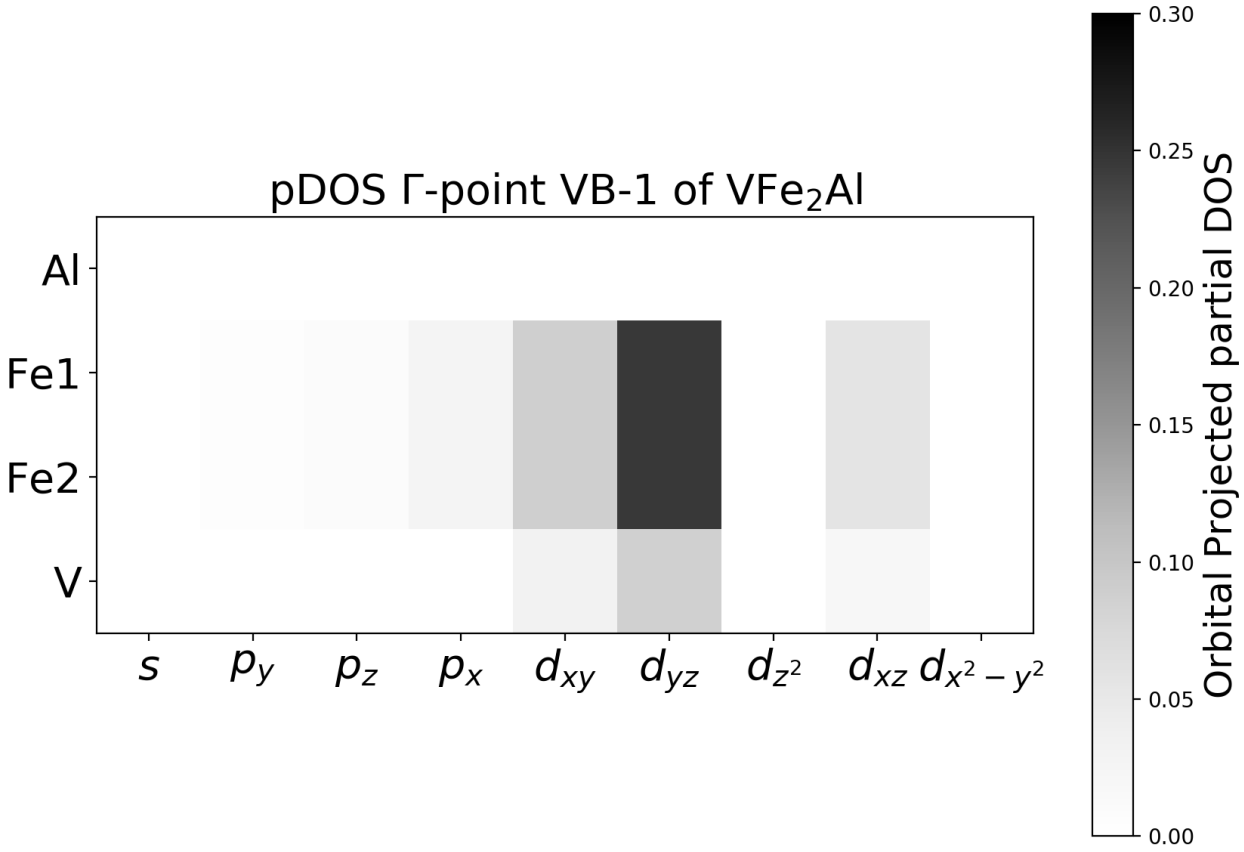

Figure S6 Orbital projected partial density of states heat map for the first Γ point valence band.

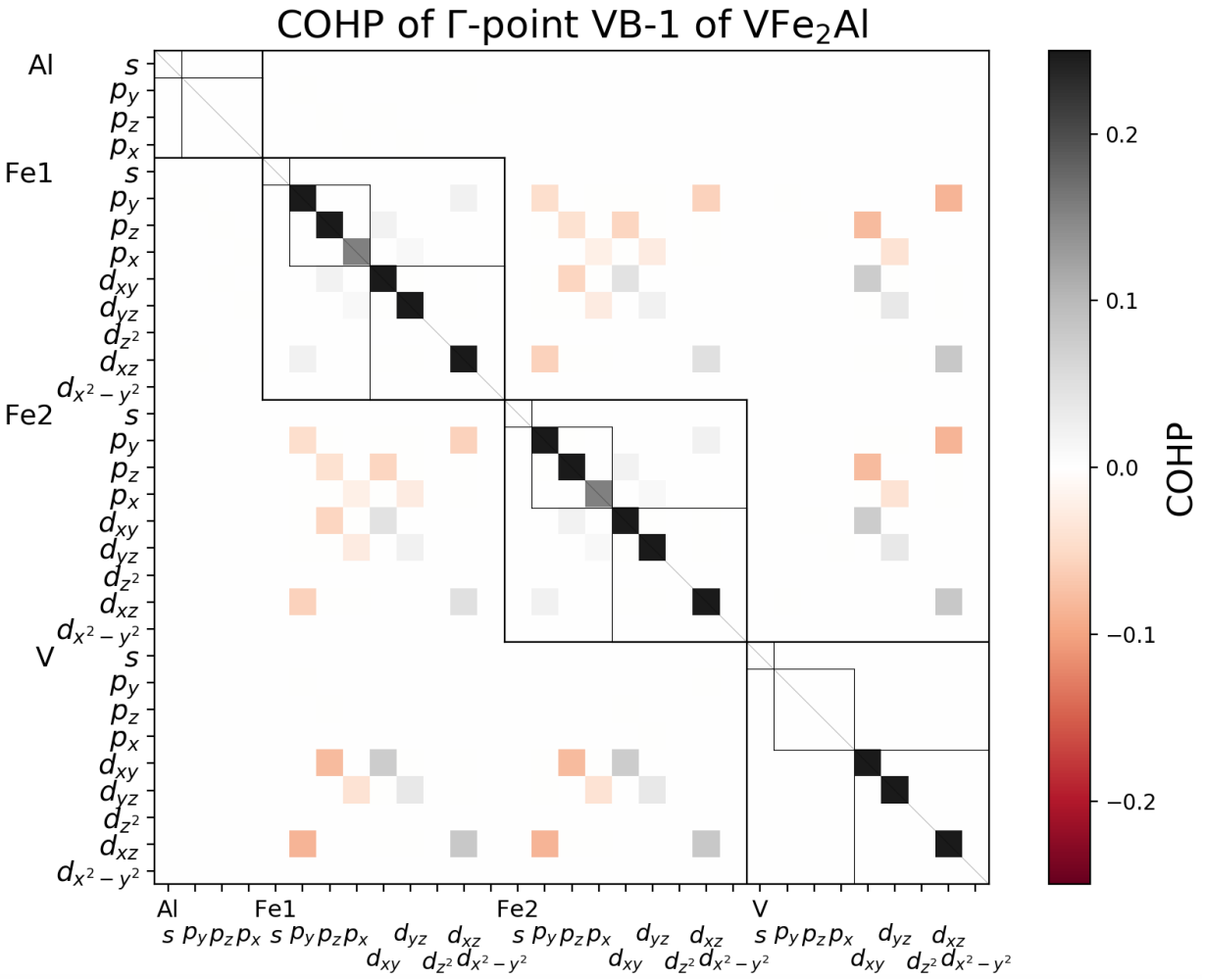

Figure S7 Crystal Orbital Hamiltonian Population (COHP) heat map for the first Γ point valence band. The *off-site* COHP terms can be found outside the 4 large boxes (in thick black lines) drawn across the diagonal of the heat map.

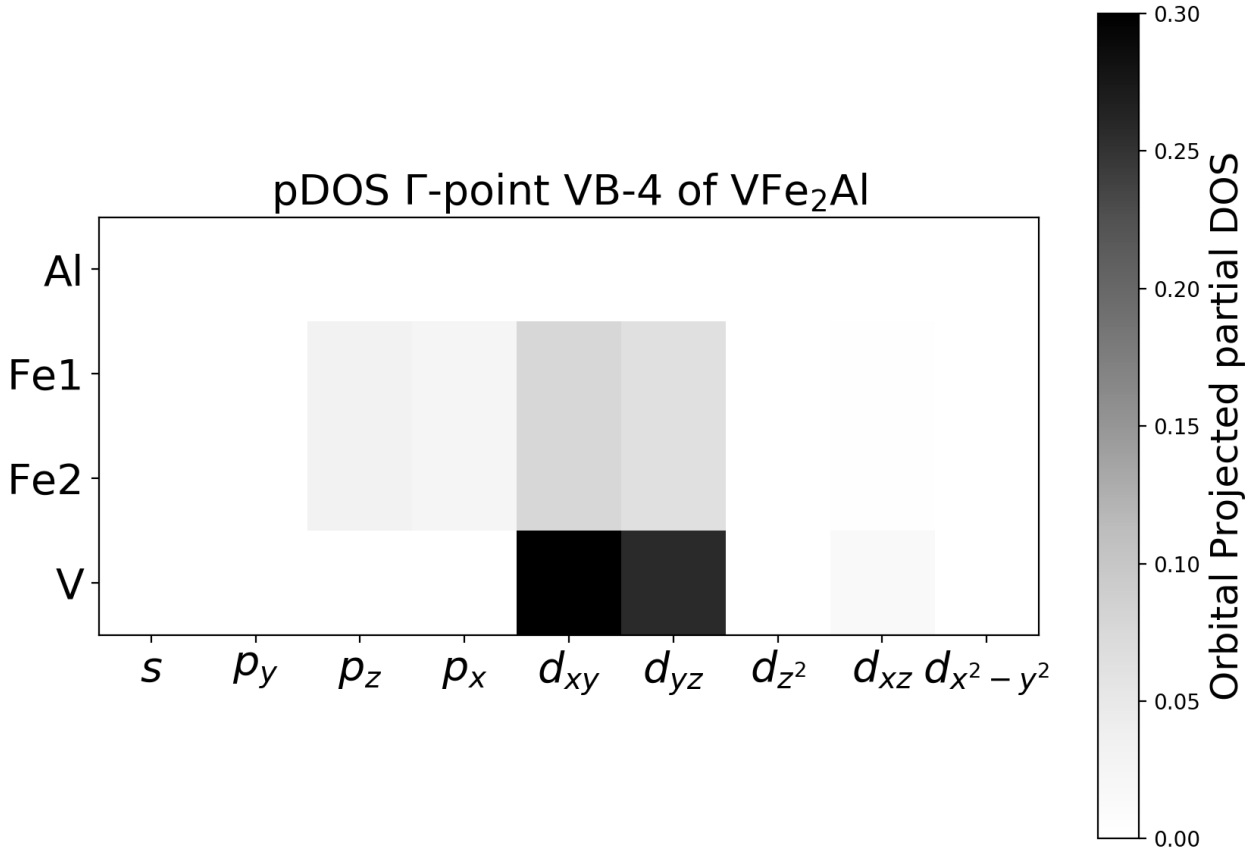

Figure S8 Orbital projected partial density of states heat map for the fourth Γ point valence band.

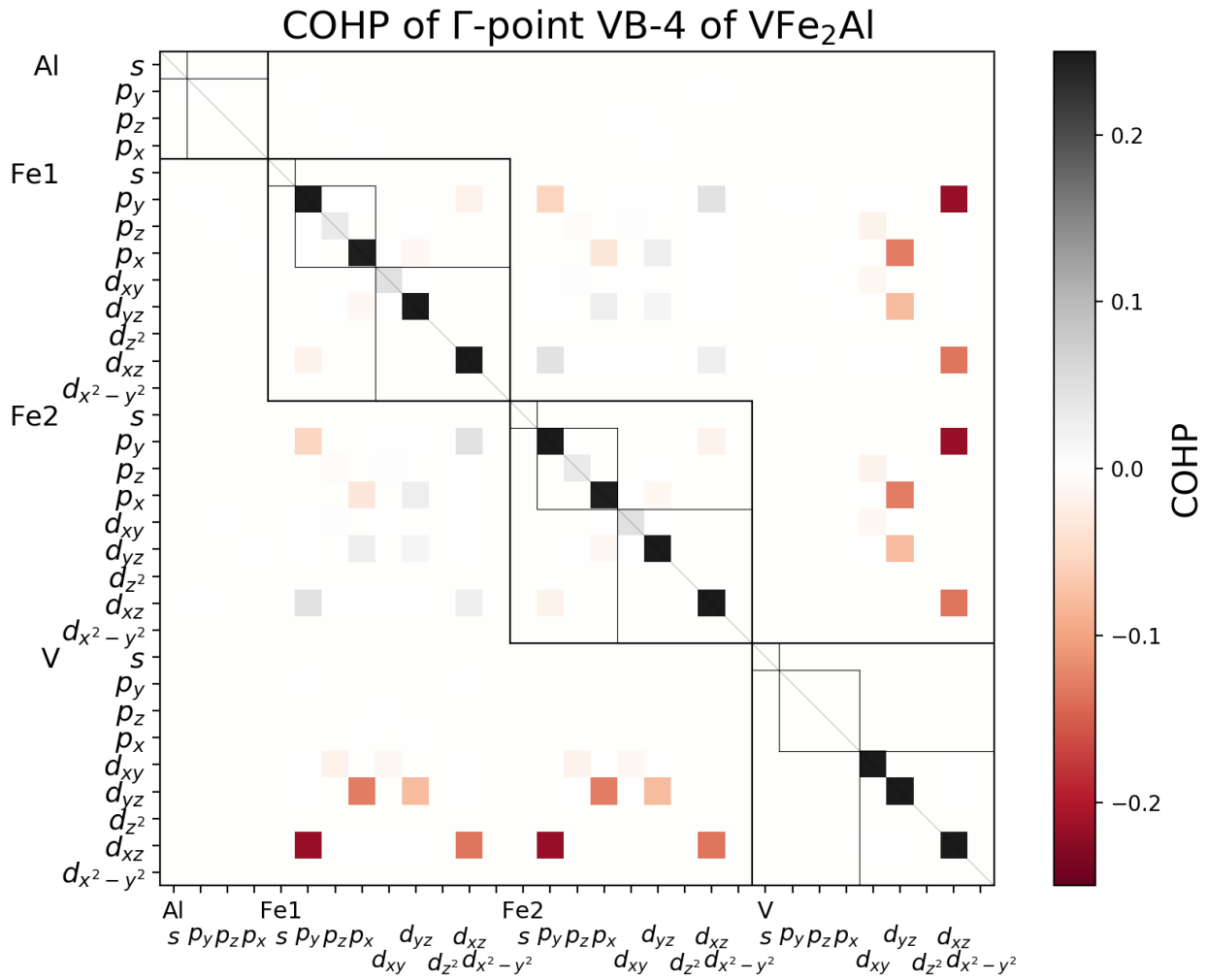

Figure S9 Crystal Orbital Hamiltonian Population (COHP) heat map for the fourth Γ point valence band. The *off-site* COHP terms can be found outside the 4 large boxes (in thick black lines) drawn across the diagonal of the heat map.

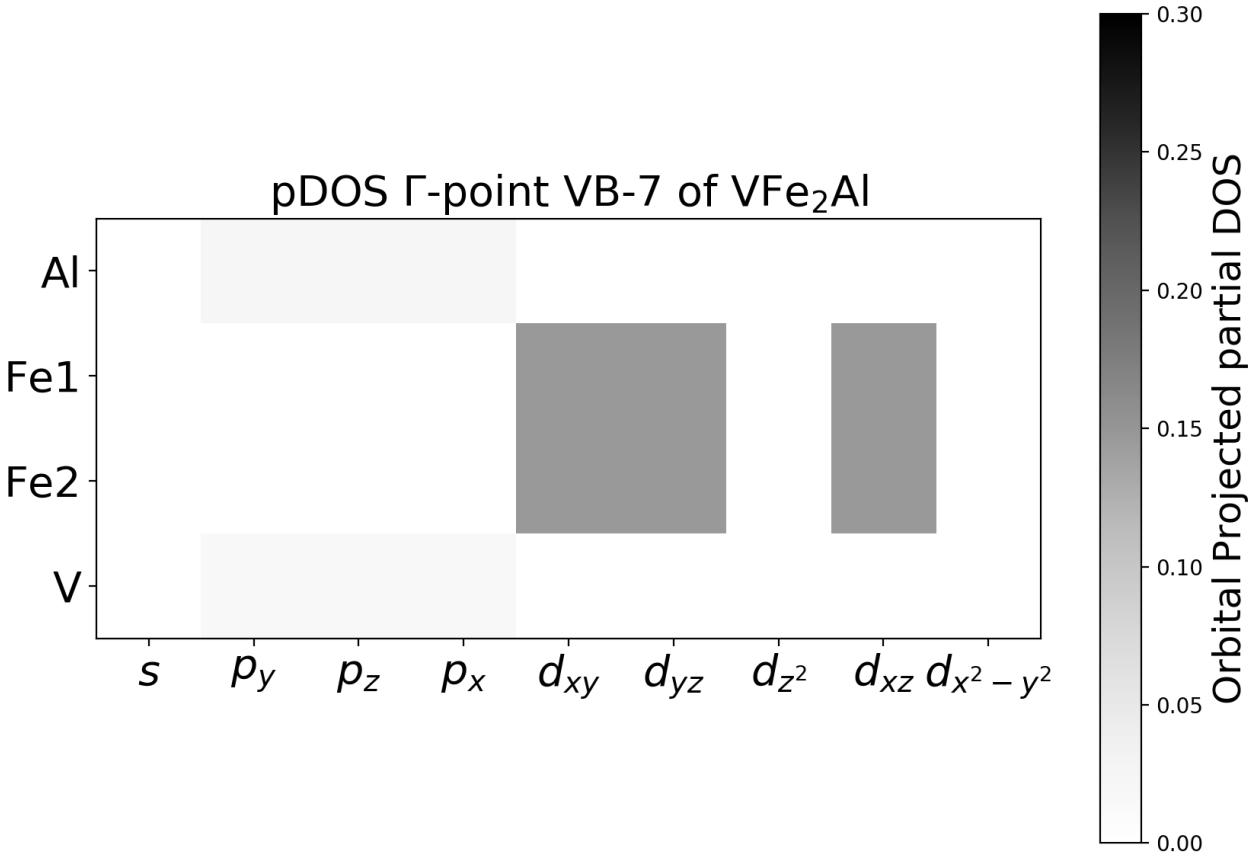

Figure S10 Orbital projected partial density of states heat map for the seventh Γ point valence band.

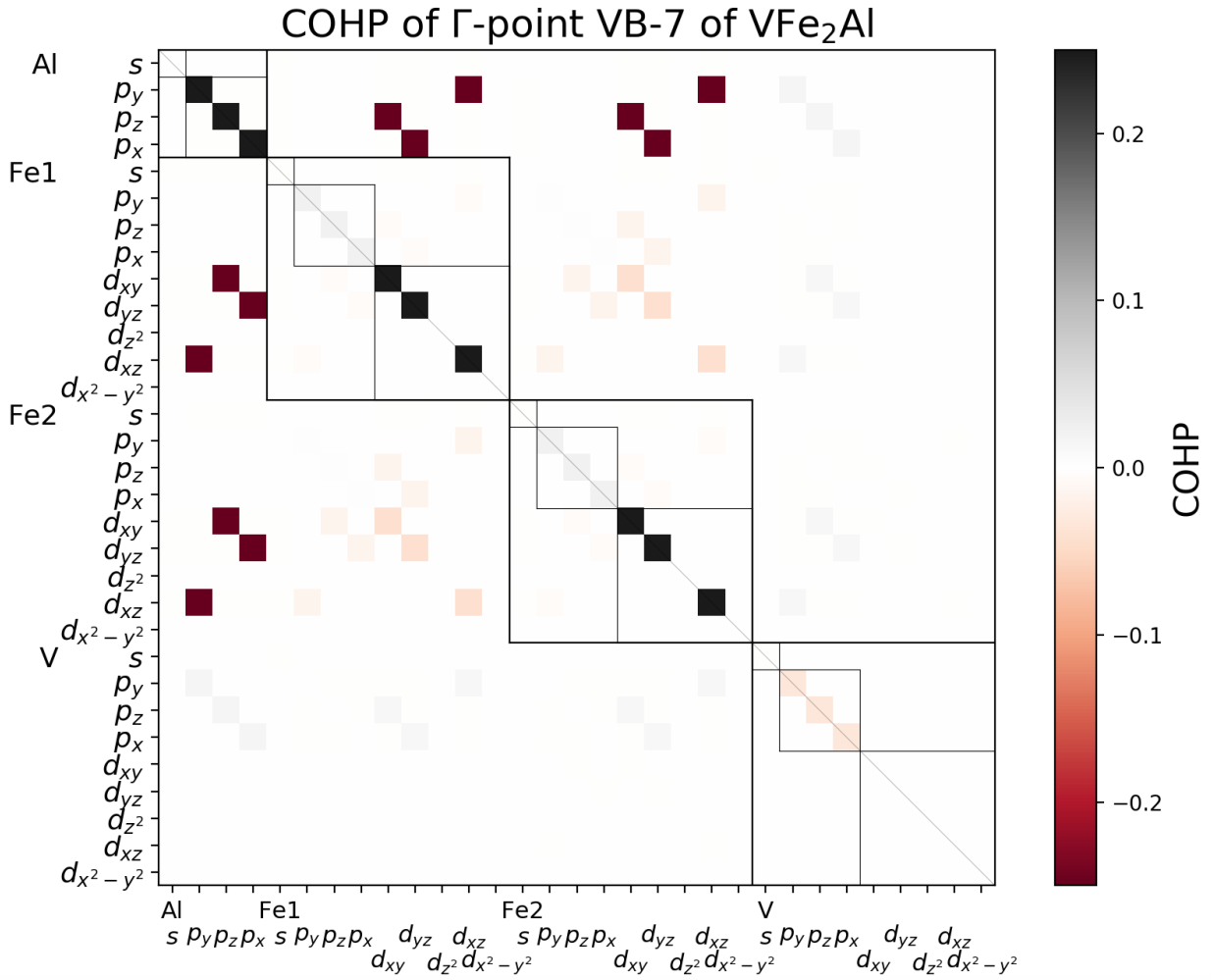

Figure S11 Crystal Orbital Hamiltonian Population (COHP) heat map for the seventh Γ point valence band. The *off-site* COHP terms can be found outside the 4 large boxes (in thick black lines) drawn across the diagonal of the heat map.

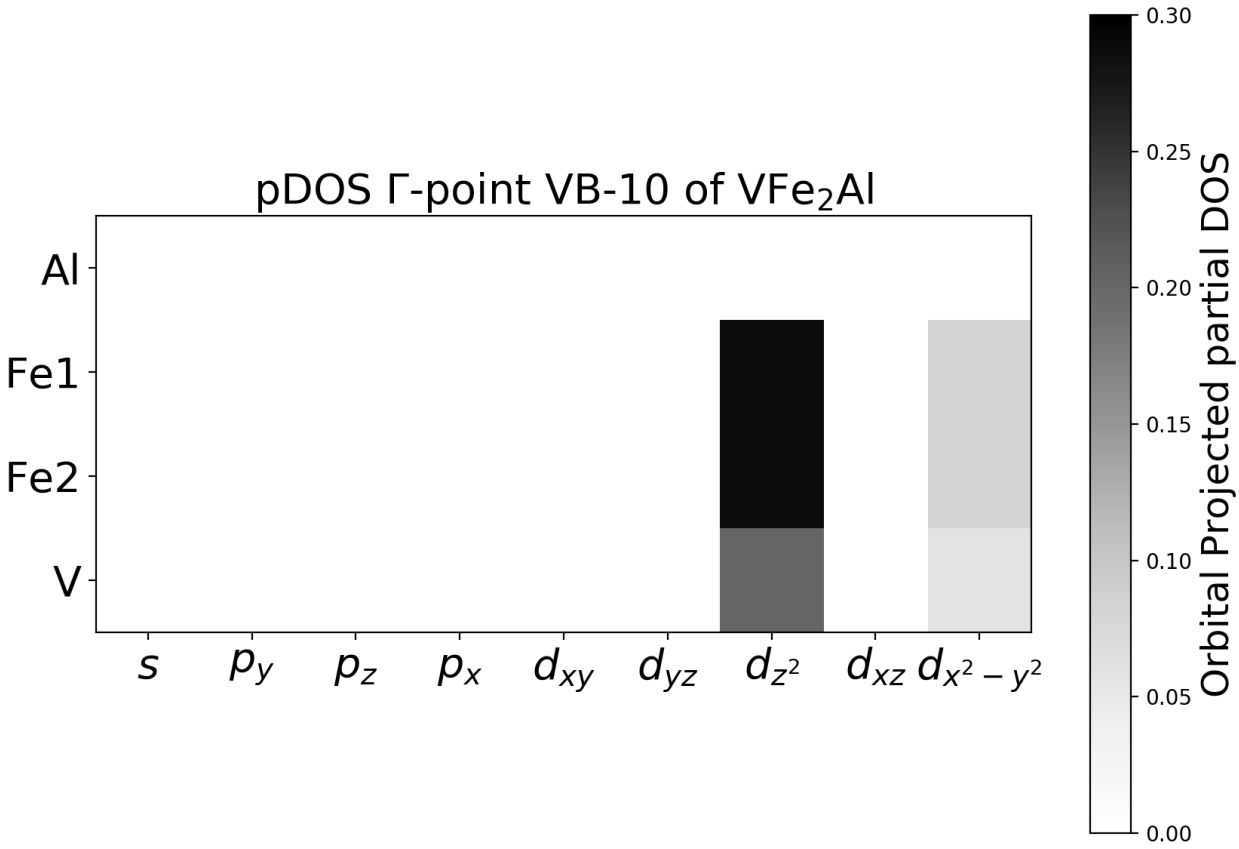

Figure S12 Orbital projected partial density of states heat map for the tenth Γ point valence band.

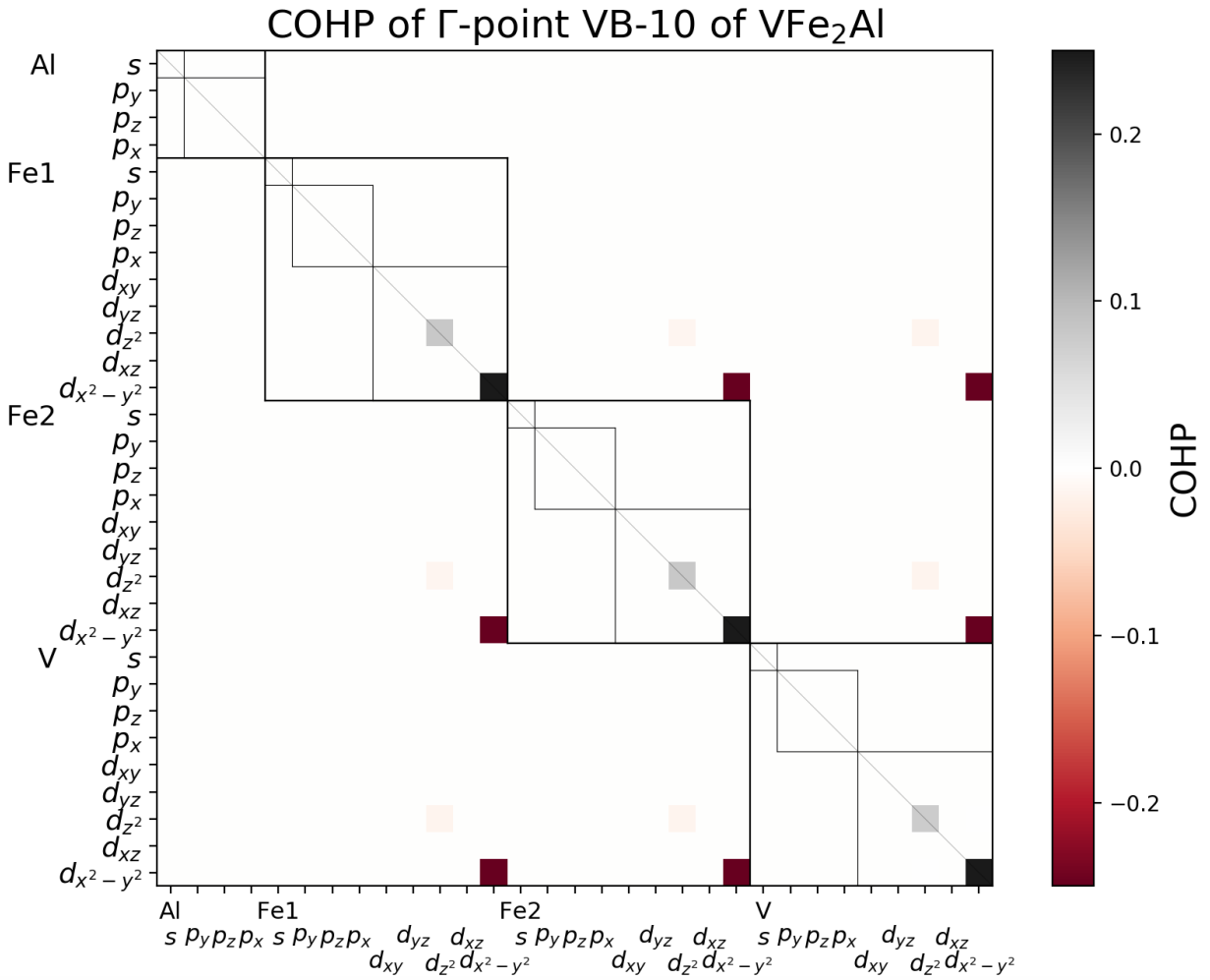

Figure S13 Crystal Orbital Hamiltonian Population (COHP) heat map for the tenth Γ point valence band. The *off-site* COHP terms can be found outside the 4 large boxes (in thick black lines) drawn across the diagonal of the heat map.

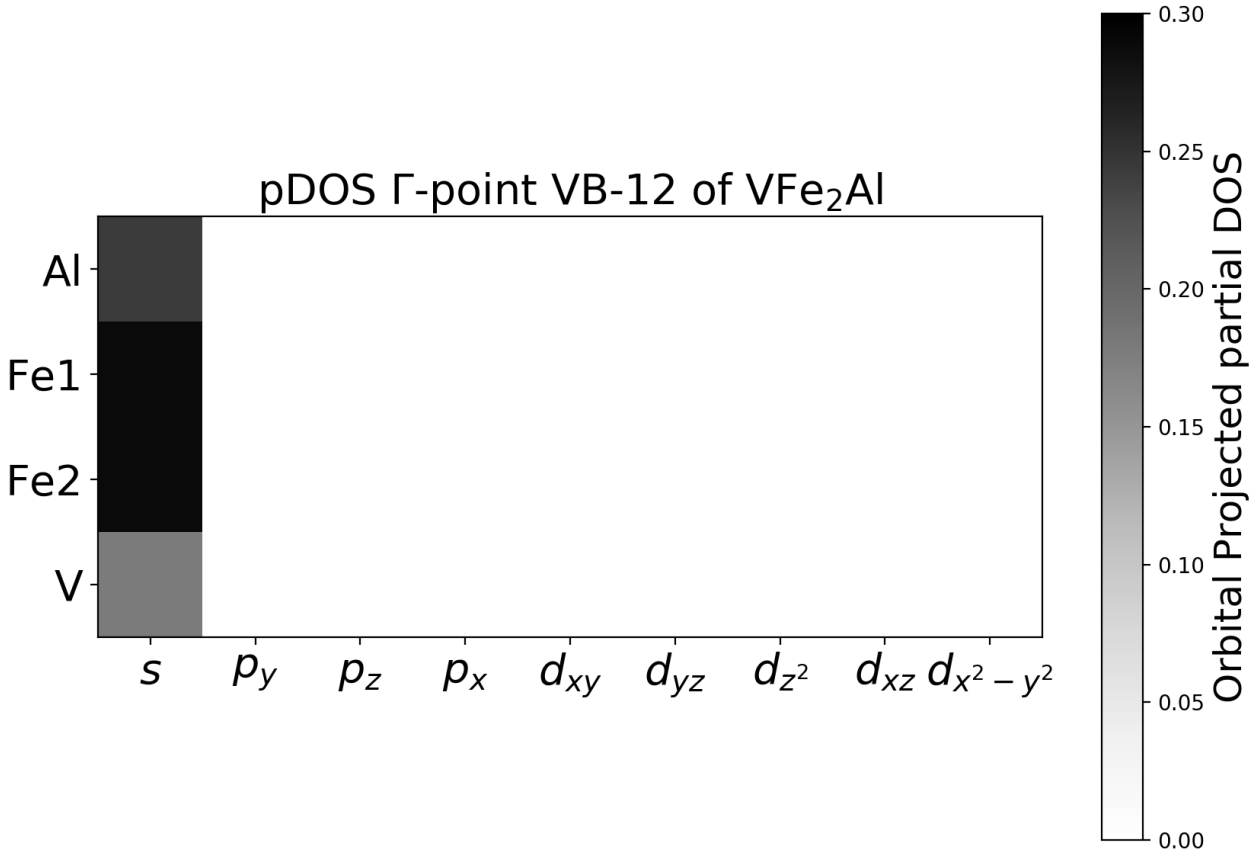

Figure S14 Orbital projected partial density of states heat map for the twelfth Γ point valence band.

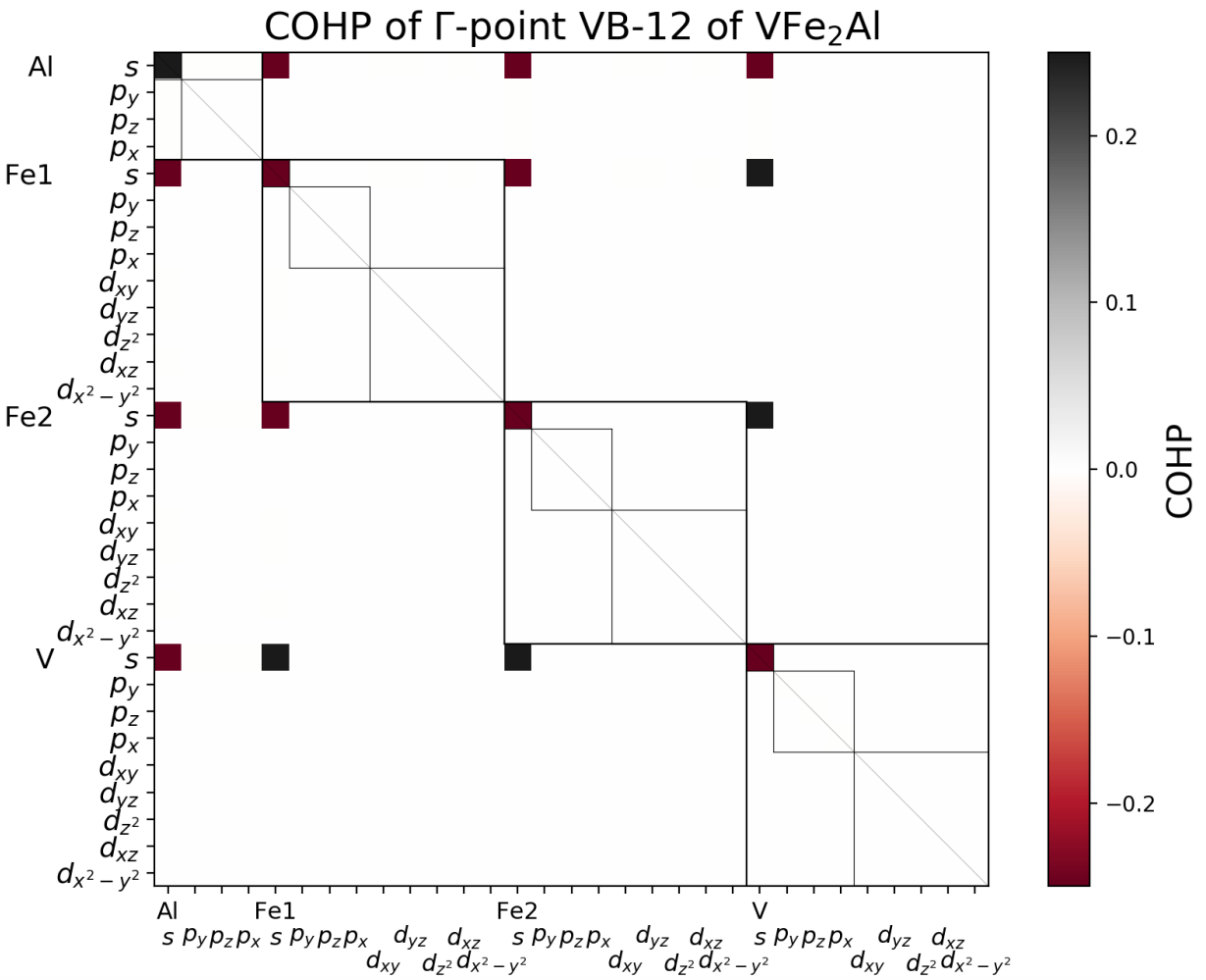

Figure S15 Crystal Orbital Hamiltonian Population (COHP) heat map for the twelfth Γ point valence band. The *off-site* COHP terms can be found outside the 4 large boxes (in thick black lines) drawn across the diagonal of the heat map.

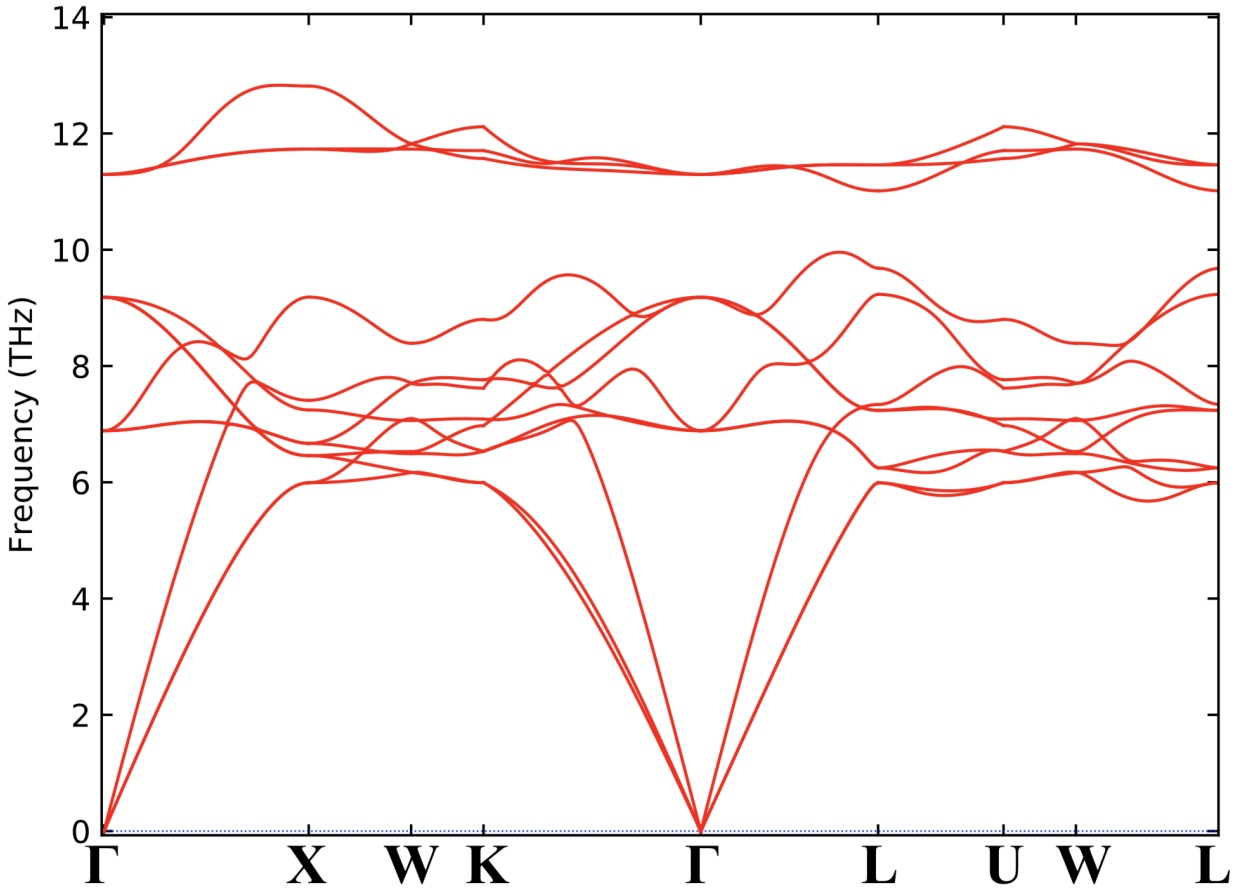

Figure S16 Phonon dispersion curves for VFe_2Al.