Two-dome Superconductivity in FeS Induced by a Lifshitz Transition – Supplemental Material –

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A. Structure

We use the crystal structures for tetragonal FeS ($P\,4/nmm$ space group) as given in Ref. [1]. The structure is defined by a and c lattice parameters and the S height h_S above the iron plane as shown in Figure S1 (a), (b) and (c), respectively. Note that the slightly decreasing height of S above the Fe plane as function of pressure (Figure S1 (c)) translates into a slightly increasing S z fractional coordinate. Relaxation of this position using density functional theory within GGA reverses this trend and is thus unreliable for FeS.

Concerning the reliability of the interpolation, we observe that the experimental data points in particular for the sulphur height and to a lesser extent also for the lattice parameters show some scatter. This is, intentionally, not reproduced by our interpolation. For the c lattice parameter, for example, the experimental value at 4.5 GPa is 4.81 Å while our interpolation is 4.78 Å. This is justified by the experimental error bars. Concerning the sulphur height, the overall variation of only 0.06 Å in the entire pressure range is rather small. At 4.5 GPa, our interpolated values differs by 0.01 Å from the experimental data point which is only a modest deviation. Determination of the internal coordinates is also experimentally somewhat less straight-forward than the dermination of the lattice constants. Please note that a less monotonous interpolation than the one we chose would lead to unphysically strong variations in the elastic constants.

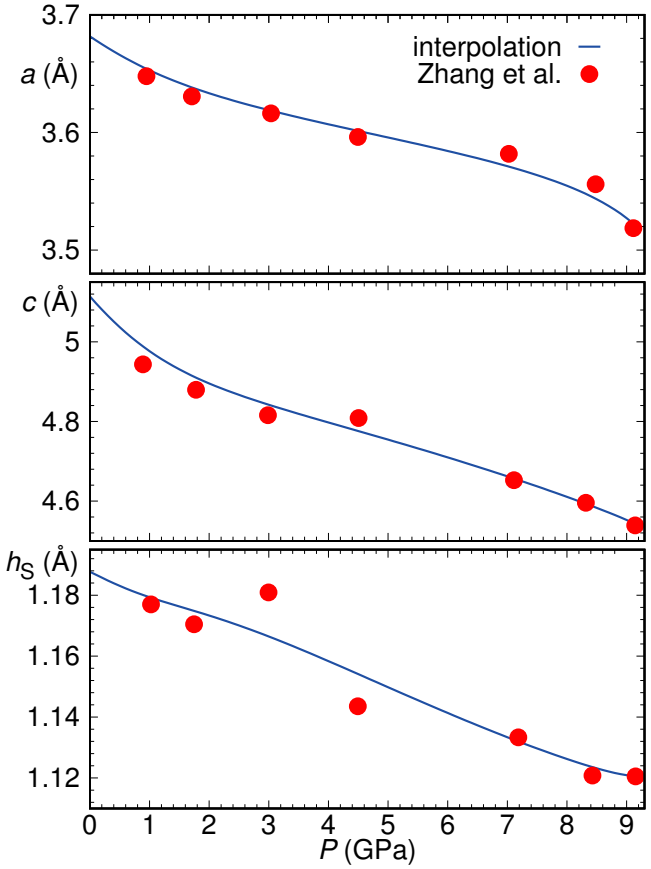

FIG. S1. Experimental crystal structures as measured by Zhang *et al.* [1] (symbols) together with Bézier interpolation (lines). (a), (b) are the tetragonal lattice parameters, and (c) is the height h_S of S above the Fe plane.

B. Origin of the Lifshitz transition

We investigate which structural change in FeS under pressure is most important for the occurrence of the Lifshitz transition at $P = 4.6$ GPa. Bond length $d_{\text{Fe-S}}$ and $\delta \equiv \measuredangle$(Fe-S-Fe) in tetragonal FeS are related to lattice constants a and c and sulfur height h_S via

$$\begin{aligned} d_{\text{Fe-S}} &= \sqrt{\frac{a^2}{4} + {h_\mathrm{S}}^2} \\ \sin\frac{\delta}{2} &= \frac{a}{2\sqrt{2}d_{\text{Fe-S}}} = \frac{a}{\sqrt{2a^2 + 8{h_\mathrm{S}}^2}} \end{aligned} \tag{S1}$$

Figure S2 shows the $d_{\text{Fe-S}}$ and δ calculated for the interpolated series of structures as function of pressure. We now investigate the sensitivity of the electronic structure and in particular the unoccupied band with Fe $3d_{z^2}$ orbital character near the Fermi level to the two relevant structural parameters separately. We focus on structures near $P = 4.6$ GPa. Upon fixing Fe-S distances $d_{\text{Fe-S}}$ and Fe-S-Fe angles δ to the values indicated by the dashed lines in Figure S2, we calculate a lattice parameter and

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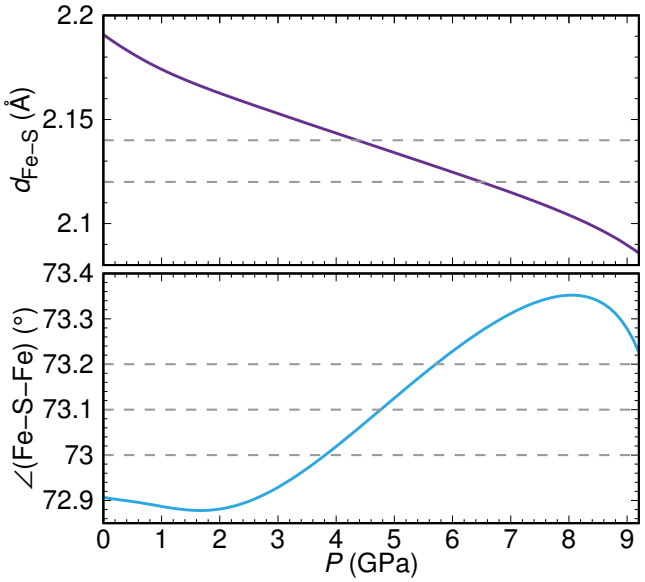

FIG. S2. Variation of (a) Fe-S distance and (b) Fe-S-Fe angle as function of pressure. Dashed lines indicate the bond distances and angles chosen for the plots in Fig. S3.

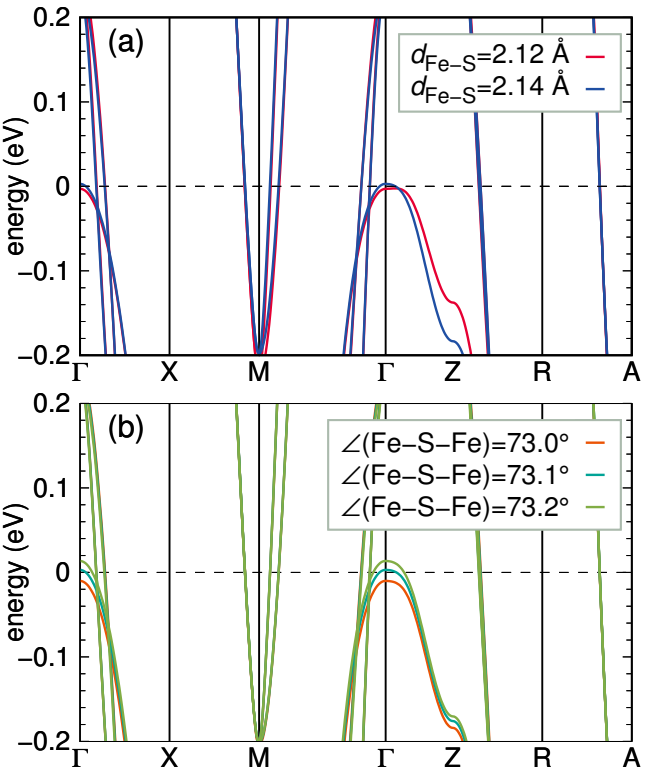

FIG. S3. Change of band structure of FeS for changes in (a) Fe-S distance and (b) Fe-S-Fe angle.

S z coordinate z_S by

$$a = 2\sqrt{2} d_{\text{Fe-S}} \sin\frac{\delta}{2}$$
$$z_S = \frac{1}{c}\sqrt{{d_{\text{Fe-S}}}^2 - \frac{a^2}{4}} \tag{S2}$$

Figure S3 shows the result. A very small change in band structure results from the substantial Fe-S bond length change corresponding to a pressure increase of roughly 2 GPa (Figure S3 (a)) once the Fe-S-Fe angle is kept constant; on the other hand, the effect of a pure angle change as is actually relevant in a 2 GPa window leads to a substantial change in band structure (Figure S3 (b)) even if the Fe-S bond lengths are fixed. Thus, the Lifshitz transition can be considered mainly an effect of the deformation of the FeS_4 tetrahedron rather than its pressure induced volume reduction.

C. Spin fluctuation formalism

We follow Graser *et al.* [2] in considering the multi-orbital Hubbard model

$$\begin{aligned} H = H_0 &+ U\sum_{i,l} n_{il\uparrow} n_{il\downarrow} \\ &+ \frac{U'}{2}\sum_{i,s,p\neq s} n_{is} n_{ip} - \frac{J}{2}\sum_{i,s,p\neq s} \boldsymbol{S}_{is}\cdot\boldsymbol{S}_{ip} \\ &+ \frac{J'}{2}\sum_{i,s,p\neq s,\sigma} c^\dagger_{is\sigma} c^\dagger_{is\bar{\sigma}} c_{ip\bar{\sigma}} c_{ip\sigma} \end{aligned} \tag{S3}$$

where $c^\dagger_{is\sigma}$ ($c_{is\sigma}$) are Fermionic creation (annihilation) operators, $\boldsymbol{S}_{is}$ is the spin operator, $n_{is\sigma} = c^\dagger_{is\sigma} c_{is\sigma}$, U denotes the intraorbital Coulomb repulsion, U' denotes the interorbital Coulomb repulsion, J denotes the Hund's rule coupling and J' denotes the pair-hopping term. The tight binding part of the Hamiltonian is

$$H_0 = -\sum_{i,j} t^{sp}_{ij} c^\dagger_{is\sigma} c_{jp\sigma} \tag{S4}$$

where t_{ij} denotes the transfer integral between sites i and j, s and p are the orbital indices, and σ denotes the spin index. The five band tight binding Hamiltonian is obtained using projective Wannier functions [3] and unfolding [4]. We first calculate the static noninteracting susceptibility

$$\begin{aligned} \chi^{pq}_{st}(\boldsymbol{q}) = -\sum_{\boldsymbol{k},l,m} & a^{p*}_l(\boldsymbol{k}) a^t_l(\boldsymbol{k}) a^{s*}_m(\boldsymbol{k}+\boldsymbol{q}) a^q_m(\boldsymbol{k}+\boldsymbol{q}) \\ & \times \frac{n_F(E_l(\boldsymbol{k})) - n_F(E_m(\boldsymbol{k}+\boldsymbol{q}))}{E_l(\boldsymbol{k}) - E_m(\boldsymbol{k}+\boldsymbol{q})} \end{aligned} \tag{S5}$$

where $E_l(\boldsymbol{k})$ is the energy value determined by the band index l and the wave vector $\boldsymbol{k}$, and $n_F(E)$ is the Fermi

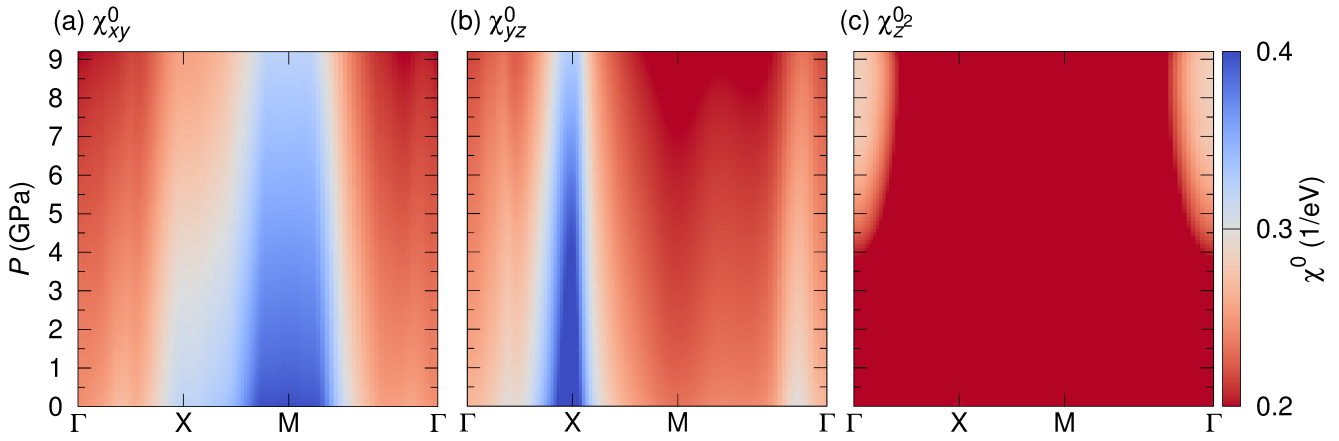

FIG. S4. Pressure dependence of the static noninteracting susceptibility for tetragonal FeS.

distribution function. a^s_m is the matrix element of eigenvectors resulting from diagonalization of tight-binding Hamiltonian H_0. Within the framework of the random phase approximation (RPA) the charge and spin susceptibilities can be calculated from the noninteracting susceptibility

$$\begin{aligned}\left[(\chi_{\rm c}^{RPA})^{pq}_{st}\right]^{-1} &= [\chi^{pq}_{st}]^{-1} + (U_{\rm c})^{pq}_{st} \\ \left[(\chi_{\rm s}^{RPA})^{pq}_{st}\right]^{-1} &= [\chi^{pq}_{st}]^{-1} - (U_{\rm s})^{pq}_{st}\end{aligned} \tag{S6}$$

where the nonzero components of the interaction tensors for the multi-orbital Hubbard model are given by [2]

$$\begin{aligned}(U_{\rm c})^{aa}_{aa} &= U & (U_{\rm c})^{aa}_{bb} &= 2U', \\ (U_{\rm c})^{ab}_{ab} &= \frac{3}{4}J - U' & (U_{\rm c})^{ba}_{ab} &= J' \\ (U_{\rm s})^{aa}_{aa} &= U & (U_{\rm s})^{aa}_{bb} &= \frac{1}{2}J \\ (U_{\rm s})^{ab}_{ab} &= \frac{1}{4}J + U' & (U_{\rm s})^{ba}_{ab} &= J',\end{aligned} \tag{S7}$$

which enables us to calculate the two-electron pairing vertex. For the interaction parameters, we find that the choice $U = 1.90$ eV, $U' = U/2$, $J = U/4$ and $J' = U/4$ takes us near the instability for all structures.

Pairing calculations.– The superconducting pairing vertex in the singlet channel is given by

$$\begin{aligned}&\Gamma^{pq}_{st}(\mathbf{k},\mathbf{k}') \\ &= \Big[\frac{3}{2}U_s\,\chi_s^{RPA}(\mathbf{k}-\mathbf{k}')\,U_s + \frac{1}{2}U_s \\ &\quad - \frac{1}{2}U_c\,\chi_c^{RPA}(\mathbf{k}-\mathbf{k}')U_c + \frac{1}{2}U_c\Big]^{tq}_{ps}.\end{aligned} \tag{S8}$$

The vertex in the orbital space description can be projected onto band space using the eigenvector resulting from diagonalization of the tight-binding Hamiltonian,

$$\begin{aligned}&\Gamma_{ij}(\mathbf{k},\mathbf{k}') \\ &\quad = \sum_{s,t,p,q} a_i^{t*}(-\mathbf{k})a_i^{s*}(\mathbf{k})\mathrm{Re}\left[\Gamma^{pq}_{st}(\mathbf{k},\mathbf{k}')\right]a_j^p(\mathbf{k}')a_j^q(-\mathbf{k}').\end{aligned} \tag{S9}$$

Using the vertex $\Gamma_{ij}(\mathbf{k},\mathbf{k}')$, we solve the gap equation

$$\begin{aligned}&-\sum_j \oint_{C_j}\frac{dk'_\parallel}{2\pi}\frac{1}{4\pi v_F(\mathbf{k}')}\left[\Gamma_{ij}(\mathbf{k},\mathbf{k}') + \Gamma_{ij}(\mathbf{k},-\mathbf{k}')\right]g_j(\mathbf{k}') \\ &\quad = \lambda_i g_i(\mathbf{k})\end{aligned} \tag{S10}$$

where λ_i denotes the pairing eigenvalue and $g_i(\mathbf{k})$ is the gap function.

D. Noninteracting susceptibility

Figure S4 shows the static noninteracting susceptibilities of FeS for the orbitals that have significant weight at the Fermi level, (a) d_{xy}, (b) d_{yz} (which is by symmetry equivalent to d_{xz} in tetragonal FeS), and (c) d_{z^2}. The latter only acquires features near Γ after the Lifshitz transition because the orbital weight is concentrated on one hole pocket.

E. One-iron Fermi surfaces of FeS

Figure S5 gives the orbital character of FeS Fermi surfaces at two k_z values, $k_z = 0$ and $k_z = \pi$ at ambient pressure and at $P = 5.5$ GPa, after the Lifshitz transition. At $P = 0$, only little $3d_{z^2}$ weight is present in the hole pockets near Γ. The new hole pocket around M has mostly $3d_{z^2}$ character. Comparison of the sizes of the electron pockets around $(\pi, 0, k_z)$ and $(0, \pi, k_z)$ between

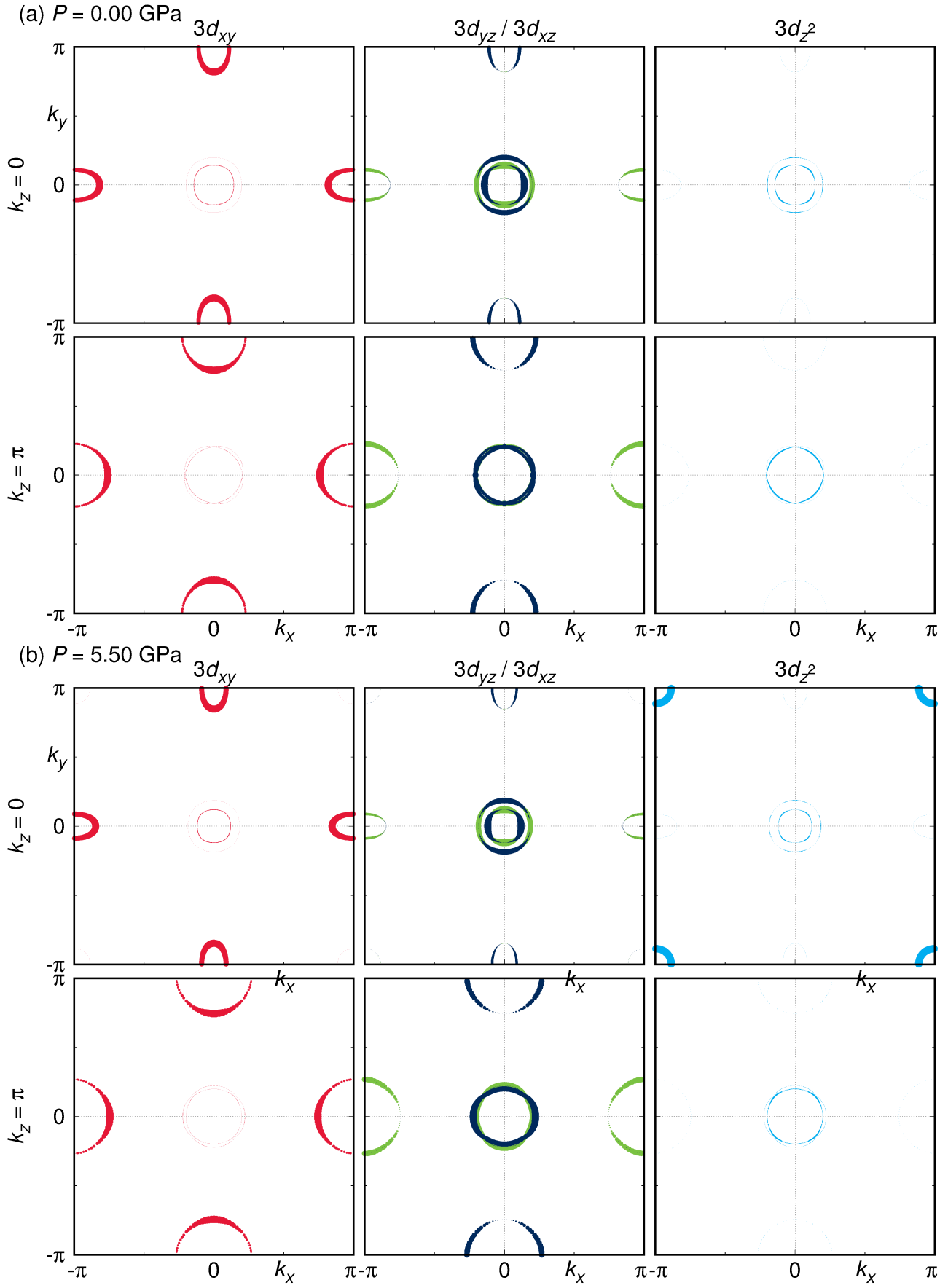

FIG. S5. Fermi surfaces of FeS at two different pressures with orbital weights. They are calculated from the tight binding models which were unfolded to the one-iron Brillouin zone.

$k_z = 0$ and $k_z = \pi$ shows that the $P = 5.5$ GPa electronic structure is more three-dimensional than the ambient pressure electronic structure. Three-dimensional susceptibility and pairing calculations are essential for properly describing the pressure dependence of superconductivity in FeS.

F. Off-diagonal components of the spin susceptibility

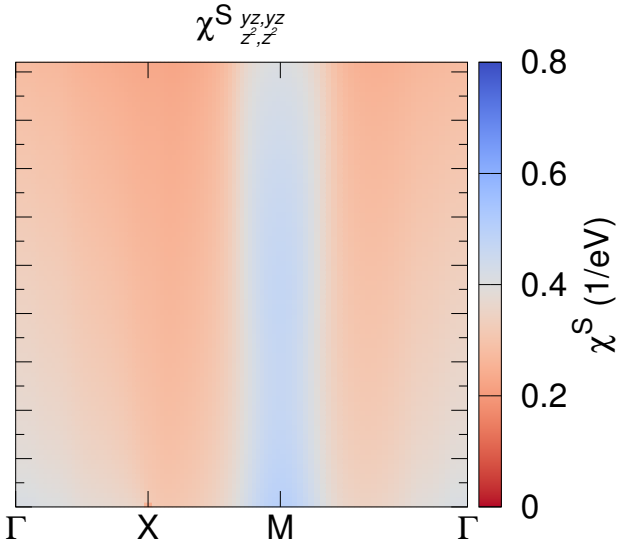

FIG. S6. Pressure dependence of an off-diagonal element of the susceptibility for tetragonal FeS.

Figure S6 shows a relevant off-diagonal component of the spin susceptibility $\chi^{S\,bb}_{aa}$ with $a = d_{z^2}$ and $b = d_{yz}$. The even more important off-diagonal component of $\chi^{S\,bb}_{aa}$ with $a = d_{z^2}$ and $b = d_{xy}$ is shown in the main text (Figure 2 (d)). $\chi^{S\,bb}_{aa}$ with $a = d_{z^2}$ and $b = d_{yz}$ has a weak maximum near $\mathbf{q} = (\pi, \pi)$, due to some nesting between d_{yz}/d_{xz} character hole pockets around Γ and the d_{z^2} character hole pocket around M.

G. Solution of the gap equation with intra-orbital interaction only

In Figure S7, we demonstrate the effect of inter-orbital interaction terms by determining the solution of the gap equation at $U' = J = J' = 0$. The solution at $P = 4.7$ GPa (and higher pressures) is a simple nodeless sign-changing s wave instead of the more complicated $s_\pm$ solution shown in Figure 3 (c) of the main text.

It arises from the $\mathbf{q} = (\pi, 0), (0, \pi)$ nesting of the d_{yz}/d_{xz} orbitals. Note also the remaining orbital weight of d_{xy} and d_{z^2} around Γ in Figs. 3(a) and (c) of the main text, which enables these orbitals to participate in a $s_\pm$ solution.

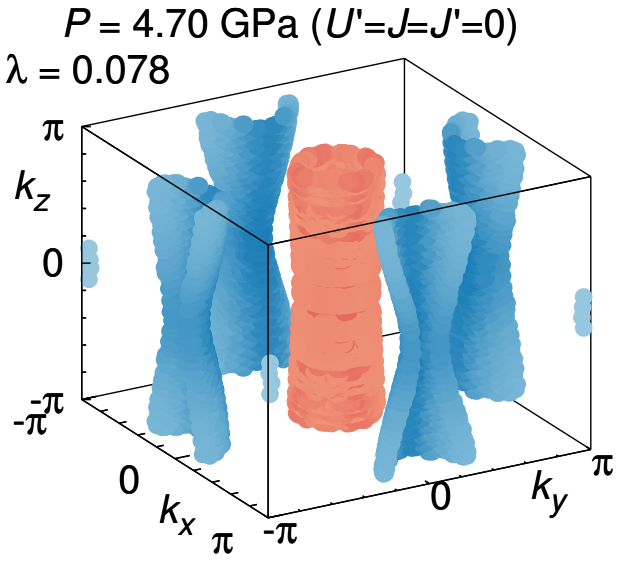

FIG. S7. Hypothetical leading gap function for FeS at $P = 4.7$ GPa for $U = 1.9$ eV and $U' = J = J' = 0$. The three-dimensional Fermi surface is plotted in the one-iron Brillouin zone.

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