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 (P4/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.

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 \( \delta \equiv \angle(\text{Fe-S-Fe}) \) in tetragonal FeS are related to lattice constants \( a \) and \( c \) and sulfur height \( h_S \) via

\[
d_{\text{Fe-S}} = \sqrt{\frac{a^2}{4} + h_S^2}
\]

\[
\sin \frac{\delta}{2} = \frac{a}{2\sqrt{2d_{\text{Fe-S}}}} = \frac{a}{\sqrt{2a^2 + 8h_S^2}}
\]

Figure S2 shows the \( d_{\text{Fe-S}} \) and \( \delta \) 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 3dz^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 \( \delta \) to the values indicated by the dashed lines in Figure S2, we calculate a lattice parameter and...
\[ d_{\text{Fe-S}} = 2.12 \text{ Å} \]
\[ a = 2\sqrt{2d_{\text{Fe-S}} \sin \delta / 2} \]
\[ z_S = \frac{1}{e} \sqrt{d_{\text{Fe-S}}^2 - a^2 / 4} \] (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 FeS4 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

\[ H = H_0 + U \sum_{i,j} n_{i\uparrow} n_{i\downarrow} + \frac{U'}{2} \sum_{i,s,p\neq s} n_{is\uparrow} n_{ip\downarrow} - \frac{J}{2} \sum_{i,s,p\neq s} S_{is} \cdot S_{ip} \] (S3)

\[ + \frac{J'}{2} \sum_{i,s,p\neq s,\sigma} c_{is\sigma}^\dagger c_{is\sigma}^\dagger c_{ip\sigma} c_{ip\sigma} \]

where \( c_{is\sigma}^\dagger \) (\( c_{is\sigma} \)) are Fermionic creation (annihilation) operators, \( S_{is} \) is the spin operator, \( n_{is\sigma} = c_{is\sigma}^\dagger c_{is\sigma} \). \( U' \) 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

\[ H_0 = - \sum_{i,j} t_{ij}^p c_{ij\sigma}^\dagger c_{ij\sigma} \] (S4)

where \( t_{ij} \) denotes the transfer integral between sites \( i \) and \( j \), \( s \) and \( p \) are the orbital indices, and \( \sigma \) 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

\[ \chi_{ij}^{ss}(q) = - \sum_{k,l,m} a_{i\sigma}^* (k) a_{j\sigma} (k) a_{l\sigma} (k+q) a_{m\sigma}^* (k+q) \]

\[ \times \frac{n_F(E_l(k)) - n_F(E_m(k+q))}{E_l(k) - E_m(k+q)} \] (S5)

where \( E_l(k) \) is the energy value determined by the band index \( l \) and the wave vector \( k \), and \( n_F(E) \) is the Fermi
distribution function. \( a_{ij}^{st} \) 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

\[
\left( \chi_{ij}^{RPA} \right)^{pq}_{st} = \left[ \chi_{ij}^{RPA} \right]_{st}^{-1} - \left( U_s \right)_{pq}^{st}
\]

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

\[
\begin{align*}
\langle U_c \rangle_{aa} &= U, & \langle U_c \rangle_{bb} &= 2U', \\
\langle U_c \rangle_{ab} &= \frac{3}{4} J - U', & \langle U_c \rangle_{ba} &= J' \\
\langle U_s \rangle_{aa} &= U, & \langle U_s \rangle_{bb} &= \frac{1}{2} J, \\
\langle U_s \rangle_{ab} &= \frac{1}{4} J + U', & \langle U_s \rangle_{ba} &= J',
\end{align*}
\]

which enables us to calculate the two-electron pairing vertex. For the interaction parameters, we find that the choice \( U = 1.90 \text{ eV}, U' = U/2, J = U/4 \) 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

\[
\Gamma_{ij}^{st}(k, k') = \left[ \frac{3}{2} U_s \chi_{ij}^{RPA}(k - k') U_s + \frac{1}{2} U_s \right]_{pq}
\]

\[
- \left[ \frac{1}{2} U_c \chi_{ij}^{RPA}(k - k') U_c + \frac{1}{2} U_c \right]_{ps}.
\]

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

\[
\Gamma_{ij}(k, k') = \sum_{s,t, p,q} a_{ij}^*(k) a_{ij}^{*}(k') \text{Re} \left[ \Gamma_{ij}^{st}(k, k') \right] a_{ij}(k') a_{ij}(-k').
\]

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

\[
- \sum_{j} \int_{\mathcal{C}_j} \frac{dk_{ij}}{2\pi} \text{Re} \left[ \Gamma_{ij}(k, k') + \Gamma_{ij}(k, -k') \right] g_{ij}(k') = \lambda_{ij} g_{ij}(k)
\]

where \( \lambda_{ij} \) denotes the pairing eigenvalue and \( g_{ij}(k) \) 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 \( \Gamma \) 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 \( P = 5.5 \text{ GPa}, \) after the Lifshitz transition. At \( P = 0 \), only little \( 3d_{z^2} \) weight is present in the hole pockets near \( \Gamma \). 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

Figure S6 shows a relevant off-diagonal component of the spin susceptibility $\chi^S_{\alpha\alpha}$ with $a = d_{z^2}$ and $b = d_{yz}$. The even more important off-diagonal component of $\chi^S_{\alpha\alpha}$ with $a = d_{z^2}$ and $b = d_{xy}$ is shown in the main text (Figure 2 (d)). $\chi^S_{\alpha\alpha}$ with $a = d_{z^2}$ and $b = d_{yz}$ has a weak maximum near $q = (\pi, \pi)$, due to some nesting between $d_{yz}/d_{xz}$ character hole pockets around $\Gamma$ 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 $P = 4.70$ 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 $q = (\pi, 0), (0, \pi)$ nesting of the $d_{yz}/d_{xz}$ orbitals. Note also the remaining orbital weight of $d_{xy}$ and $d_{z^2}$ around $\Gamma$ in Figs. 3(a) and (c) of the main text, which enables these orbitals to participate in a $s_{\pm}$ solution.

