# Two-Dome Superconductivity in FeS Induced by a Lifshitz Transition

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Among iron chalcogenide superconductors, FeS can be viewed as a simple, highly compressed relative of FeSe without a nematic phase and with weaker electronic correlations. Under pressure, however, the superconductivity of stoichiometric FeS disappears and reappears, forming two domes. We perform electronic structure and spin fluctuation theory calculations for tetragonal FeS in order to analyze the nature of the superconducting order parameter. In the random phase approximation, we find a gap function with $d$-wave symmetry at ambient pressure, in agreement with several reports of a nodal superconducting order parameter in FeS. Our calculations show that, as a function of pressure, the superconducting pairing strength decreases until a Lifshitz transition takes place at 4.6 GPa. As a hole pocket with a large density of states appears at the Lifshitz transition, the gap symmetry is altered to sign-changing $s$ wave. At the same time, the pairing strength is severely enhanced and increases up to a new maximum at 5.5 GPa. Therefore, our calculations naturally explain the occurrence of two superconducting domes in FeS.

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*Introduction.*—The structurally simplest class of iron-based superconductors with its prime representative FeSe [1] was discovered in the same year as LaFeAsO [2]. FeSe has been intensively studied due to its very large nematic region [3], its interesting magnetism [4], and the complexity of its electronic structure [5]. Only in 2015 was it established that the isostructural FeS is also a superconductor [6]. Even though the replacement of Se by the smaller S appears to be a minor structural modification, it soon became clear that FeSe and FeS behave differently in several respects: The nematic region is absent in FeS [7], the electronic correlations appear to be significantly smaller in FeS [8], and the upper critical field is much smaller [9]. In fact, the possibility to grow high-quality mixed $FeSe_{1-x}S_x$ structures has provided opportunities to study the evolution of properties between FeSe and FeS [8,10–13].

Superconductivity in FeS has been observed below $T_c = 5$ K [6] with some variation due to sample dependence [14]. Scanning tunneling spectroscopy points to strong-coupling superconductivity [15], and Hall conductivities can be fitted with a two-band model [16]. The symmetry of the superconducting gap in FeS has been the subject of some debate. Using scanning tunneling spectroscopy, Yang *et al.* [15] conclude that the superconducting gap of FeS is strongly anisotropic. Specific heat measurements [17] and quasiparticle heat transport studies [18] point to a nodal gap structure. However, muon spin rotation studies found fully gapped behavior in FeS [19,20]. Theoretically, a $d_{x^2-y^2}$ order parameter at ambient pressure has been obtained [21].

Pressure has been shown to suppress superconductivity in FeS [22]. Surprisingly, however, Zhang *et al.* have found that, after the initial suppression, at a pressure of $P = 5$ GPa superconductivity reemerges, and a second superconducting dome is formed, up to a pressure of $P = 22.3$ GPa. Such double-dome superconductivity is known to occur also in alkali iron selenides [23] and in FeSe intercalates [24,25]. In fact, two superconducting domes occur in nearly all classes of unconventional superconductors [26].

In this Letter, we consider the structurally simple FeS as an instructive example system for studying the origin of double-dome superconductivity in iron-based materials. We show that, at a pressure of $P = 4.6$ GPa, a Lifshitz transition occurs, adding a hole pocket to the Fermi surface and boosting the density of states at the Fermi level. Using the spin fluctuation theory in the random phase approximation, we show that the pairing strength of the $d_{x^2-y^2}$ order parameter, which dominates within the low-pressure dome, decreases until a Lifshitz transition of the electronic structure takes place. At the transition, the superconducting order parameter switches to nodeless $s_\pm$, and the pairing strength grows significantly to a new maximum. Our study highlights that, even without a structural phase transition, the pressure-induced changes in the electronic structure trigger the reemergence of superconductivity in FeS.

*Structure.*—The metastable tetragonal structure of FeS ($P4/nmm$ space group) occurs as a mineral named mackinawite [27]. Single crystals can be synthesized by hydrothermal synthesis [6,27] and by deintercalation of

 

$K_xFe_{2-y}S_2$ [9]. We base our study on the pressure series of tetragonal crystal structures determined by Zhang *et al.* [28]. In this study, mackinawite is found to transform to the hexagonal troilite phase ($P\bar{6}2c$ space group) at high pressures, with a mixed region extending from 5 to 9.2 GPa. However, the high-pressure phase diagram of FeS is complicated, and orthorhombic ($Pnma$ space group) and monoclinic ($P2_1a$ space group) phases have also been described [22,29].

*Methods.*—We perform density functional theory calculations for the tetragonal FeS structures within the full-potential local orbital (FPLO) [30] basis, using the generalized gradient approximation (GGA) exchange correlation functional [31] and fine $k$ meshes of $50 \times 50 \times 50$. We interpolate the experimental crystal structure (as shown in Ref. [32], Fig. S1) so that we can perform calculations employing fine pressure steps of 0.1 GPa. We construct ten-band tight-binding models using the FPLO projective Wannier functions [33], including all Fe $3d$ states. We employ the unfolding method using point group symmetries [34] in order to obtain five-band tight-binding models. We study superconductivity assuming a spin-fluctuation-driven pairing interaction within the multiorbital Hubbard model and use the formalism as detailed by Graser *et al.* [32,35] and as implemented in Refs. [36,37]. We determine the noninteracting susceptibilities on $\mathbf{q}$ meshes of $50 \times 50 \times 10$ points at all pressures, and we use about 5000 $k$ points on the Fermi surface for solving the gap equation in three dimensions; two-dimensional calculations are insufficient for FeS under pressure.

*Results.*—We first determine the electronic structure of tetragonal FeS in small pressure intervals up to a pressure of $P = 9.2$ GPa. Figure 1 shows bands and Fermi surfaces at two representative pressures, $P = 0$ and $P = 5$ GPa. Our results at ambient pressure are in good agreement with angle-resolved photoemission [38,39] and quantum oscillation measurements [40]. The fact that FeS is rather weakly correlated [39] makes the plain GGA calculations a good starting point for our analysis of electronic structure and superconductivity. After a very smooth pressure evolution of the electronic structure, suddenly at $P = 4.6$ GPa a Lifshitz transition occurs, and a hole pocket is added to the Fermi surface [Figs. 1(b), 1(d), and 1(f)]. The reason for this event is the fact that the bands with Fe $3d_{z^2}$ orbital character widen more rapidly with pressure than the other iron bands. A careful analysis of the relationship between geometrical parameters in the FeS structure and its bands reveals that the $3d_{z^2}$ bands are especially sensitive to the Fe-S-Fe angle, much more so than to the Fe-S bond distance [32]. As a consequence, the $3d_{z^2}$ contribution to the density of states at the Fermi level $N(E_F)$ increases gradually below $P = 4.6$ GPa before it rises by more than 100% at the Lifshitz transition, as shown in Fig. 2(a).

We now consider the superconductivity in FeS, assuming a spin-fluctuation-induced Cooper pairing. We use the random phase approximation to calculate the spin susceptibility at all pressures (for details, see Ref. [32]). In iron-based superconductors, the pairing interaction is often dominated by intraorbital nesting (see Ref. [36]) and, in particular, $\chi^S_{xy}$ and $\chi^S_{yz}$ (or $\chi^S_{xz}$), as shown in Figs. 2(b) and 2(c), respectively. These elements of the spin susceptibility are diagonal in the four orbital indices, since we first investigate only intraorbital contributions. The dominant peak in $\chi^S_{xy}$ is near a nesting vector $\mathbf{q} = (\pi, \pi)$ and in $\chi^S_{yz}$ near $\mathbf{q} = (\pi, 0)$. In fact, these nesting vectors can also be extracted easily from a plot of the Fermi surface (Fig. 3).

For a repulsive interaction, a peak in the spin susceptibility at vector $\mathbf{q}$ induces a sign change of the superconducting gap between Fermi surface pockets connected by $\mathbf{q}$. From the spin susceptibility, it is clear that the electronic structure of FeS leads to the competition between different order parameters, which is typical for iron-based superconductors. The peak at $\mathbf{q} = (\pi, 0)$ in $\chi^S_{yz}$ favors a sign change between hole cylinders around $\Gamma$ and electron cylinders at $X$ and $Y$, i.e., a type of sign-changing $s$-wave order parameter, where the gap has the same sign on all

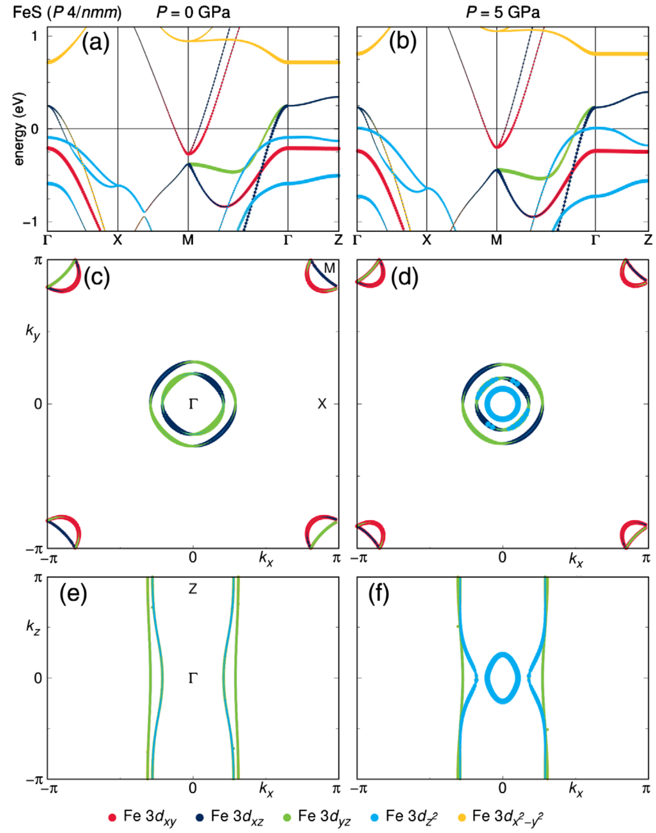


FIG. 1. Electronic structure of FeS at ambient pressure (left column) and at $P = 5$ GPa (right column). Band structures (a), (b), Fermi surfaces in the $k_x - k_y$ plane at $k_z = 0$ (c),(d), and Fermi surfaces in the $k_x - k_z$ plane (e),(f) are all colored with the orbital weights of the Fe $3d$ orbitals. A Lifshitz transition at $P = 4.6$ GPa adds a hole Fermi surface pocket near $\Gamma$.

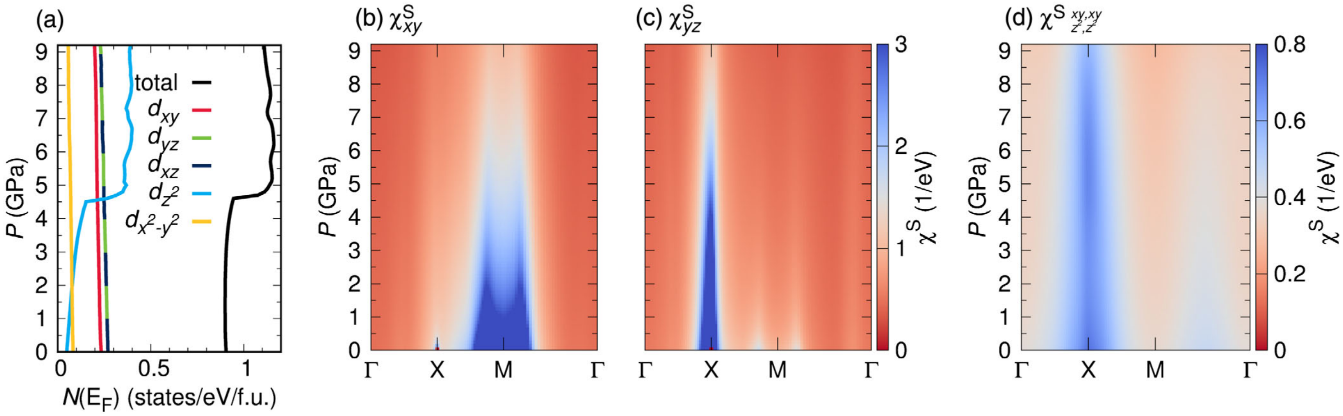


FIG. 2. Pressure dependence of (a) the density of states at the Fermi level and (b),(c) diagonal elements and (d) off-diagonal elements of the spin susceptibility for tetragonal FeS.

electron pockets. On the other hand, the strong $\mathbf{q} = (\pi, \pi)$ peak in $\chi^S_{xy}$ favors a sign change between the electron pockets; this is most easily fulfilled by a $d_{x^2-y^2}$ order parameter. As a compromise between these two possibilities, a nodal sign-changing $s$-wave order parameter sometimes occurs (see Ref. [36]).

Because of the increased bandwidth and smaller density of states at the Fermi level, the spin susceptibility generally decreases with increasing pressure [Figs. 2(b) and 2(c)]. Therefore, we find a general decline of the pairing strength with increasing pressure [Fig. 4(b)]. From a quantitative solution of the superconducting gap equation, we find

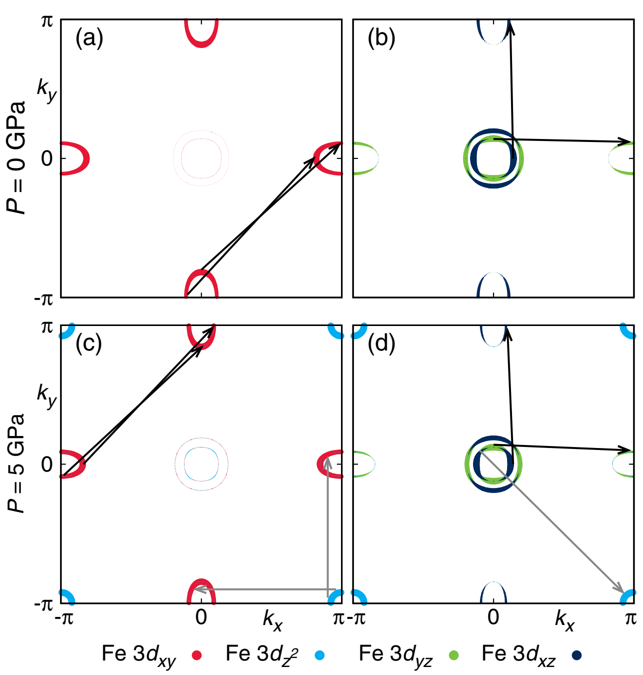


FIG. 3. Fermi surfaces in the one-iron Brillouin zone at $k_z = 0$ at ambient pressure and at $P = 5$ GPa (after the Lifshitz transition). Black arrows indicate important intraorbital nesting vectors, gray arrows significant interorbital nesting vectors.

[Fig. 4 (a)] that, at $P = 0$, the $d_{x^2-y^2}$ solution wins, while several $s_\pm$ solutions are in competition but subleading [Fig. 4 (b)]. This result is in agreement with Ref. [21]. As a function of pressure, the eigenvalues of the gap equation are suppressed rapidly. Initially, no change in the symmetry of the superconducting gap is found. This corresponds well to the first superconducting dome that was observed experimentally [28].

However, very close to the Lifshitz transition, the nature of the superconducting order parameter changes dramatically. At $P = 4.65$ GPa, a new sign-changing $s$-type order parameter appears [Fig. 4 (c)] and becomes the dominating solution up to the highest pressure $P = 9.2$ GPa, at which the tetragonal phase is completely replaced by the hexagonal phase of FeS. The eigenvalue of the gap equation increases rapidly for this solution, in very good agreement with the experiment, up to a maximum at $P = 5.4$ GPa. Thus, our calculation provides clear evidence for the existence of two-dome superconductivity in FeS under pressure. Note that at the present level of theory we cannot compare superconducting and nonsuperconducting ground states, which means that we have a strong suppression of the superconducting pairing strength but cannot capture a $T_c = 0$ pressure interval.

Note that the eigenvalue of the nodal $s_\pm$ solution is also enhanced at the Lifshitz transition, while the eigenvalue of the $d_{x^2-y^2}$ solution is not affected at all [Fig. 4(b)]. This is the case because the symmetry-required nodes of the $d_{x^2-y^2}$ solution are located exactly where the $d_{z^2}$ hole pocket emerges. Therefore, it is naturally excluded from the $d_{x^2-y^2}$ solution.

As we have not studied superconductivity in the hexagonal phase, which is presumably of nonmagnetic, BCS origin, we cannot complete the second superconducting dome at the higher pressures investigated experimentally. Predicting the internal coordinates for the $P\bar{6}2c$ space group FeS structures at high pressures based on experimental lattice parameters and analyzing the superconducting mechanism is

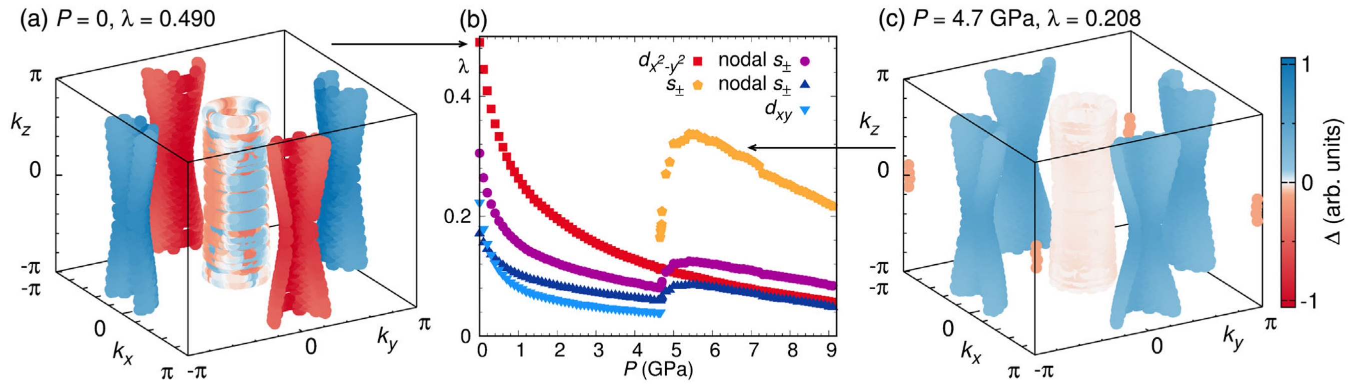


FIG. 4. Leading gap functions for FeS at (a) ambient pressure and (c) $P = 4.7$ GPa. The three-dimensional Fermi surfaces are plotted in the one-iron Brillouin zone. (b) Leading eigenvalues $\lambda$ of the linearized gap equation as a function of pressure. Up to $P = 4.6$ GPa, the $d_{x^2-y^2}$ order parameter dominates, but its eigenvalue decreases, marking the right half of the first superconducting dome. At the Lifshitz transition pressure $P = 4.6$ GPa, an $s_\pm$ solution takes over, with its eigenvalue forming a second superconducting dome.

an interesting endeavour which is beyond the scope of the present study.

*Discussion.*—So far, we have demonstrated two important effects that occur in pressurized FeS without any structural discontinuity: a Lifshitz transition, which creates a hole pocket and significant Fe $3d_{z^2}$ weight at the Fermi level, and a change of superconducting order parameter from $d$ to sign-changing $s$ wave, which occurs at almost exactly the same pressure. The important question of the connection between the two events remains to be answered.

While the noninteracting diagonal susceptibility $\chi^0_{d_{z^2}}$ acquires some weak maximum near $\mathbf{q} = 0$ (see Ref. [32]), the weak feature near $\mathbf{q} = 0$ in the diagonal spin susceptibility $\chi^S_{d_{z^2}}$ does not help to explain any change in the superconducting order parameter. Note that the hole Fermi surface around $M$ in the unfolded one-iron Brillouin zone [see Fig. 4 (c)] is of a different nature from the $\gamma$ Fermi surface feature at $M$ described in Ref. [41]; in their case, electron doping populates a pocket of $d_{xy}$ orbital character (in the local coordinates chosen for the present analysis), and the pocket can contribute to pairing via the $\mathbf{q} = (\pi, \pi)$ peak in $\chi^S_{xy}$.

Our case highlights the importance of the interaction terms proportional to $U'$, $J$, and $J'$: They mediate participation of the $d_{z^2}$ orbital in the pairing via the off-diagonal components of the spin susceptibility, such as $\chi^{Sbb}_{aa}$, $\chi^{Sba}_{ab}$, and $\chi^{Sab}_{ab}$ with $a = d_{z^2}$ and $b = d_{xy}$ (or, in principle, also $b = d_{xz/yz}$) [see Fig. 2(d) and Ref. [32]], which are significantly peaked at $\mathbf{q} = (\pi, 0)$. The interorbital nesting between $d_{z^2}$ and $d_{xz}$ is much weaker and does not contribute significantly to the pairing. Figure 3 shows the relevant intra- and interorbital nesting vectors before and after the Lifshitz transition. This figure indeed confirms that there is considerable interorbital nesting between the $d_{xy}$ and $d_{z^2}$ orbitals.

Although Figs. 3(c) and 3(d) show that there is only a small pocket of $d_{z^2}$ character, its strong influence on the pairing interaction is explained by the extremely large density of states at the Fermi level in this orbital after the Lifshitz transition [Fig. 2(a)].

Finally, we also comment on the negligible gap size on the central hole pockets in Fig. 4(c). The intraorbital spin susceptibility of the $d_{xz/yz}$ orbitals, which is peaked at $X$, should lead to a sign change between the electron pockets and the central hole pockets as discussed before, with a negative sign on the central hole pockets. However, the interorbital spin susceptibility between $d_{z^2}$ and $d_{xz/yz}$, which is peaked at $M$, should lead to a sign change between the emergent hole pocket and the central hole pockets, with a positive sign on the central hole pockets. Therefore, these interactions are frustrated. As a compromise, the gap on the central hole pockets remains close to zero.

Since our analysis highlights the importance of interorbital interactions, one could expect that superconductivity breaks down once interorbital Coulomb interaction, Hund's rule coupling, and the pair-hopping term are neglected. We corroborate the significance of the interorbital interaction terms by solving the gap equation at finite intraorbital Coulomb interaction $U = 1.9$ eV with other interactions set to zero ($U' = J = J' = 0$). The order parameter we obtain in this case is nodeless $s_\pm$, but, more importantly, the associated pairing eigenvalue is close to zero; i.e., superconductivity vanishes without interorbital interactions.

It would be very interesting to probe the Lifshitz transition by performing quantum oscillation experiments in FeS at pressures around 5 GPa; also, the predicted superconducting order parameter change could be observed in low-temperature specific heat measurements under pressure.

*Conclusion.*—We investigated the superconducting order parameter of tetragonal FeS using a combination of density functional theory calculations and spin fluctuation theory for the multiorbital Hubbard model. We showed that a Lifshitz transition occurs in FeS at a pressure of about

$P = 4.6$ GPa, which changes the superconducting order parameter from $d_{x^2-y^2}$ to a sign-changing $s$ wave, with significantly enhanced pairing strength right after the Lifshitz transition due to the enhanced density of states at the Fermi level. While superconducting pairing within the first dome is dominated by intraorbital nesting of $d_{xy}$ states, the second dome features unusual interorbital nesting between $d_{xy}$ and $d_{z^2}$ states. In conclusion, our calculations explain the recently found double-dome superconductivity in FeS.

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