Unconventional sign-changing superconductivity near quantum criticality in YFe$_2$Ge$_2$

Alaska Subedi
Centre de Physique Théorique, École Polytechnique, CNRS, 91128 Palaiseau Cedex, France
(Dated: November 13, 2013)

I present the results of first principles calculations of the electronic structure and magnetic interactions for the recently discovered superconductor YFe$_2$Ge$_2$ and use them to identify the nature of superconductivity and quantum criticality in this compound. I find that the Fe 3$d$ derived states near the Fermi level show a rich structure with the presence of both linearly dispersive and heavy bands. The Fermi surface exhibits nesting between hole and electron sheets that manifests as a peak in the susceptibility at $(1/2, 1/2)$. I propose that the superconductivity in this compound is mediated by antiferromagnetic spin fluctuations associated with this peak resulting in a $s_\pm$ state similar to the previously discovered iron-based superconductors. I also find that various magnetic orderings are almost degenerate in energy, which indicates that the proximity to quantum criticality is due to competing magnetic interactions.

PACS numbers: 74.20.Mn,74.40.Kb,74.25.Jb

Unconventional superconductivity and quantum criticality are two of the most intriguing phenomena observed in physics. The underlying mechanisms and the properties exhibited by the systems in which these two phenomena occur has not been fully elucidated because unconventional superconductors and materials at quantum critical point are so rare. The dearth of realizable examples has also held back the study of the relationship and interplay between unconventional superconductivity and quantum criticality, if there are any.

Therefore, the recent report of non-Fermi liquid behavior and superconductivity in YFe$_2$Ge$_2$ by Zou et al. is of great interest despite a low superconducting $T_c$ of $\sim$1.8 K. This material is also interesting because it shares some important features with the previously discovered iron-based high-temperature superconductors. Like the other iron-based superconductors, its structural motif is a square plane of Fe that is tetrahedrally coordinated, in this case, by Ge. This Fe$_2$Ge$_2$ layer is stacked along the $z$ axis with an alternating layer of Y ions. The resulting body-centered tetragonal structure ($I4/mmm$) of this compound is the same as that of the ‘122’ family of the iron-based superconductors.

The nearest neighbor Fe–Ge and Fe–Fe distances of 2.393 and 2.801 Å, respectively, in this compound are similar to the Fe–As and Fe–Fe distances of 2.403 and 2.802 Å, respectively, found in BaFe$_2$As$_2$. This raises the possibility that the direct Fe–Fe hopping is important to the physics of this material, which is the case for the previously discovered iron-based superconductors.

Furthermore, Zou et al. report that the superconductivity in this compound exists in the vicinity of a quantum critical point that is possibly associated with antiferromagnetic spin fluctuations. A related isostructural compound LuFe$_2$Ge$_2$ that occurs in the same crystal structure exhibits antiferromagnetic spin density wave order below 9 K, and the magnetic transition is continuously suppressed in Lu$_{1-x}$Y$_x$Fe$_2$Ge$_2$ series as Y content is increased, with the quantum critical point lying near the composition Lu$_{0.8}Y_{0.19}$Fe$_2$Ge$_2$. The proximity of YFe$_2$Ge$_2$ to quantum criticality is observed in the non-Fermi liquid behavior of the specific-heat capacity and resistivity. Zou et al. find that the unusually high Sommerfeld coefficient with a value of $C/T \sim 90$ mJ/mol K$^2$ at 2 K further increases to a value of $\sim$100 mJ/mol K$^2$ as the temperature is lowered, although the experimental data is not detailed enough to distinguish between a logarithmic and a square root increase. They also find that the resistivity shows a behavior $\rho \propto T^{5/2}$ up to a temperature of 10 K.

In this paper, I use the results of first principles calculations to discuss the interplay between superconductivity and quantum criticality in YFe$_2$Ge$_2$ in terms of its electronic structure and competing magnetic interactions. I find that the fermiology in this compound is dominated by Fe 3$d$ states with the presence of both heavy and linearly dispersive bands near the Fermi level. The Fermi surface consists of five sheets. There is an open tetragonal electron cylinder around $X = (1/2, 1/2, 0)$. A large three dimensional closed sheet that is shaped like a shell of a clam is situated around $Z = (0, 0, 1/2) = (1, 0, 0)$. This sheet encloses a cylindrical and two almost spherical hole sheets. The tetragonal cylinder sheet around $X$ nests with the spherical and the cylindrical sheets around $Z$, which manifests as a peak at $(1/2, 1/2)$ in the bare susceptibility. I propose that the superconductivity in this compound is mediated by antiferromagnetic spin fluctuations associated with this peak, and the resulting superconductivity has a sign-changing $s_\pm$ symmetry with opposite signs on the nested sheets around $X$ and $Z$. This superconductivity is similar to the one proposed for previously discovered iron-based superconductors. Furthermore, I find that there are competing magnetic interactions in this compound, and the quantum criticality is due to the fluctuations associated with these magnetic interactions.

The results presented here were obtained within the local density approximation (LDA) using the general full-potential linearized augmented planewave method as implemented in the WIEN2k software package. Muffin-
The non-spin-polarized LDA band structure and density of states (DOS) are shown in Figs. 1 and 2, respectively. The lowest band that starts out from $\Gamma$ at $-5.2$ eV relative to the Fermi energy has Ge 4$p_z$ character. There is only one band with Ge 4$p_z$ character below Fermi level, and there is another band with this character above the Fermi level. This indicates that the Ge ions make covalent bonds along the $c$ axis, which is not surprising given the short Ge–Ge distance in that direction.

The four bands between $-1.2$ and $-4.8$ eV that start out from $\Gamma$ at $-1.5$ and $-2.6$ eV have Ge 4$p_x$ and 4$p_y$ character. Rest of the bands below the Fermi level have mostly Fe 3$d$ character. Similar to the other iron-based superconductors, there is no gap-like structure among the Fe 3$d$ bands splitting them into a lower lying $e_g$ and higher lying $t_{2g}$ states. This shows that Fe–Ge covalency is minimal and direct Fe–Fe interactions dominate. Almost all of the Fe 4$s$ and Y 4$d$ and 5$s$ character lie above the Fermi level. This indicates a nominal occupation of Fe 3$d^{6.5}$, although the actual occupancy will be different because there is some covalency of Fe 3$d$ states with Y 4$d$ and Ge 4$p$ states.

The electronic states near the Fermi level come from Fe 3$d$ derived bands and show a rich structure. The electronic DOS at the Fermi level is $N(E_F) = 4.50$ eV$^{-1}$ on a per formula unit both spin basis corresponding to a calculated Sommerfeld coefficient of 10.63 mJ/mole K$^2$. The Fermi level lies at the bottom of a valley with a large peak due to bands of mostly $d_{xz}$ and $d_{yz}$ characters on the left and a small peak due to a band of mostly $d_{x^2-y^2}$ character on the right. (The local coordinate system of the Fe site is rotated by 45° in the $xy$ plane with respect to the global cartesian axes such that the Fe $d_{x^2-y^2}$ orbital points away from the Ge $p_x$ and $p_y$ orbitals.) There is a pair of linearly dispersive band with mostly $d_{xz}$ and $d_{yz}$ as well as noticeable Ge $p_z$ characters either side of Z. If they are not gapped in the superconducting state, they will provide the system with a massless excitation. In addition to this pair of linearly dispersive bands, there is also a very flat band near the Fermi level along $X$–$\Gamma$. This band has an electron-like nature around $X$ and crosses the Fermi level close to it. Along the $X$–$\Gamma$ direction, it reaches a maximum at 0.08 eV above the Fermi level, turns back down coming within 0.01 eV of touching the Fermi level, and again moves away from the Fermi level. It may be possible to access these band critical points.
that have vanishing quasiparticle velocities via small perturbations due to impurities, doping, or changes in structural parameters. The role of such band critical points in quantum criticality has been emphasized recently\textsuperscript{12} and similar physics may be relevant in this system.

The Fermi surface of this compound is shown in Fig. 3. There is an open very two dimensional tetragonal electron cylinder around \( X \). This has mostly \( d_{xz} \) and \( d_{yz} \) character. There are four closed sheets around \( Z \). One of them is a large three dimensional sheet with the shape like the shell of a clam with \( d_{xz}, d_{yz}, d_{xy}, \) and \( d_z \) characters. There are two almost spherical hole sheets. These have mostly \( d_{xz} \) and \( d_{yz} \) characters, with the smaller one also containing noticeable Ge \( p_z \) character. These two spherical sheets are enclosed by a closed cylindrical hole sheet that has mostly \( d_{xy} \) character.

The cylindrical and larger spherical sheets centered around \( Z \) touch at isolated points. Otherwise, the Fermi surface is comprised of disconnected sheets. If one considers the \( \Gamma-Z-\Gamma \) path along the \( k_z \) direction, there is a series of box-shaped cylindrical hole sheet that encloses the two spherical sheets. Although there are no sections around \( \Gamma \), these sheets around \( Z \) enclose almost two-third of the \( \Gamma-Z-\Gamma \) path. Therefore, there is likely to be substantial nesting between the sheets around \( Z \) and \( X \) that will lead to a peak in the susceptibility at the wave vector \( (1/2, 1/2) \).

The bare Lindhard susceptibility is further enhanced due to the RPA interaction, and its real part is related to magnetism and superconductivity. It is found experimentally that pure YFe\(_2\)Ge\(_2\) does not order magnetically down to a temperature of 2 K although it shows non-Fermi liquid behavior in the transport and heat capacity measurements that is likely due to proximity to a magnetic quantum critical point.\textsuperscript{14} As the temperature is lowered further, superconductivity manifests in the resistivity measurements at \( T_c = 1.8 \) K and DC magnetization at \( T^m_{\text{mag}} = 1.5 \) K. This superconductivity can be due spin fluctuations associated with the peak in the susceptibility\textsuperscript{14,15} The pairing interaction has the form

\[
V(q = k - k') = -\frac{I^2(q)\chi_0(q)}{1 - I^2(q)\lambda^2(q)}
\]

in the singlet channel and is repulsive. (In the triplet channel, the interaction is attractive and also includes an angular factor.) Here \( I(q) \) is the Stoner parameter which microscopically derives from Coulomb repulsion between electrons.

In the present case, the structure of the calculated susceptibility leads to the off-diagonal component of the interaction matrix to have a large negative value \(-\lambda \) for the pairing between the hole sheets at \( Z \) and electron cylinder.
at \( X \) in the singlet channel. The diagonal component of the interaction matrix \( \lambda_{ij} \) pairing interactions on the hole and electron sheets are small and ferromagnetic. (For simplicity, I have assumed that the density of states are same for the hole and electron sections.) The eigenvector corresponding to the largest eigenvalue of this interaction matrix has opposite signs between the hole sheets around \( Z \) and electron cylinder around \( X \), and this is consistent with a singlet \( s_{+} \) superconductivity with a wave vector \((1/2, 1/2)\). This superconductivity is similar to the previously discovered iron-based superconductors. 

The proposed superconductivity in YFe\(_2\)Ge\(_2\) and the previously discovered iron-based superconductor is similar, but the \( T_c \) = 1.8 K for YFe\(_2\)Ge\(_2\) is much smaller than those reported for other iron-based superconductors. One reason for this may be the smaller nesting in this compound leading to a smaller peak in susceptibility. The hole cylinder around \( Z \) has mostly \( d_{xy} \) character whereas the hole spheres around \( Z \) and the electron cylinder around \( X \) have mostly \( d_{xz} \) and \( d_{yz} \) character. These factors should lead to a slightly smaller and broader peak at \( X \). I note, however, that nesting in the other iron-based superconductors is also not perfect and the band characters between the nested sheets also vary.

Another reason for the smaller \( T_c \) in YFe\(_2\)Ge\(_2\) may be due to the existence of competing magnetic fluctuations associated with the proximity to quantum criticality. The DOS from non-spin-polarized calculation is \( N(E_F) = 1.125 \text{ eV}^{-1} \) per spin per Fe, which puts this material on the verge of a ferromagnetic instability according to the Stoner criterion. Ferromagnetism is pair-breaking for the singlet pairing and will suppress the \( T_c \) in this compound. Furthermore, there is a peak in the susceptibility at \( Z \) as well. The presence of additional antiferromagnetic interactions might reduce the phase space available for the spin fluctuation associated with the pairing channel and may be pair-breaking as well.

| Energy (meV/Fe) | Moment (\( \mu_B \)/Fe) |
|----------------|------------------------|
| NSP            | 0                      |
| FM             | −6.29                  |
| AFM (0,0,1/2)  | −11.63                 |
| SDW (1/2,1/2,0)| −6.52                  |
two hole spheres around $Z$. There is a peak in the bare susceptibility at $(1/2, 1/2)$ due to nesting between the hole sheets around $Z$ and the electron cylinder around $X$. I propose that the superconductivity in YFe$_2$Ge$_2$ is due to antiferromagnetic spin fluctuations associated with this peak. The resulting superconducting state has a $s_\pm$ state similar to that of previously discovered iron-based superconductors. I also find that different magnetic configurations are close in energy, which suggests the presence of competing magnetic interactions that are responsible for the proximity to quantum criticality observed in this compound.

I am grateful to Antoine Georges for helpful comments and suggestions. This work was partially supported by a grant from Agence Nationale de la Recherche (PNIC-TIDES).

1. Y. Zou, Z. Feng, P. W. Logg, J. Chen, G. I. Lampronti, and F. M. Grosche, arXiv e-print, 1311:0247.
2. G. Venturini, and B. Malaman, J. Alloys Compounds 235, 201 (1996).
3. M. Rotter, M. Tegel, D. Johrendt, I. Schellenberg, W. Hermes, and R. Pöttgen, Phys. Rev. B 78, 020503(R) (2008).
4. D. J. Singh, and M.-H. Du, Phys. Rev. Lett. 100, 237003 (2008).
5. M. Avila, S. Bud’ko, and P. Canfield, J. Magn. Magn. Mater. 270, 51 (2004).
6. J. Ferstl, H. Rosner, and C. Geibel, Physica B: Condens. Matter 378-380, 744 (2006).
7. S. Ran, S. L. Bud’ko, and P. C. Canfield, Philosophical Magazine 91, 4388 (2011).
8. I. I. Mazin, D. J. Singh, M. D. Johannes, and M. H. Du, Phys. Rev. Lett. 101, 057003 (2008).
9. K. Kuroki, S. Onari, R. Arita, H. Usui, Y. Tanaka, H. Kotani, H. Aoki, Phys. Rev. Lett. 101, 087004 (2008).
10. P. Blaha, K. Schwarz, G. Madsen, D. Kvasnicka, and J. Luitz, “WIEN2k, An Augmented Plane Wave + Local Orbitals Program for Calculating Crystal Properties” (K. Schwarz, Tech. Univ. Wien, Austria) (2001).
11. http://elk.sourceforge.net
12. B. P. Neal, E. R. Ylvisaker, and W. E. Pickett, Phys. Rev. B 84, 055133 (2011).
13. J. T. Park, D. S. Inosov, A. Yaresko, S. Graser, et al., Phys. Rev. B 82, 134503 (2010).
14. N. F. Berk and J. R. Schrieffer, Phys. Rev. Lett. 17, 433 (1966).
15. D. Fay and J. Appel, Phys. Rev. B 22, 3173 (1970).
16. T. Moriya, Spin Fluctuations in Itinerant Electron Magnetism (Springer, Berlin, 1985).
17. A. Z. Solontsov and D. Wagner, Phys. Rev. B 51, 12410 (1995).
18. A. Ishigaki and T. Moriya, J. Phys. Soc. Jpn. 67, 3924 (1998).
19. A. Aguayo, I. I. Mazin, and D. J. Singh, Phys. Rev. Lett. 92, 147201 (2004).
20. P. Larson, I. I. Mazin, and D. J. Singh, Phys. Rev. B 69, 064429 (2004).
21. D. J. Singh, Phys. Rev. B 78, 094511 (2008).
22. F. Ning, K. Ahilan, T. Imai, A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, D. Mandrus, J. Phys. Soc. Jpn. 78, 013711 (2009).
23. S. Jiang, H. Xing, G. Xuan, C. Wang, Z. Ren, C. Feng, J. Dai, Z. Xu, G. Cao, J. Phys. Condens. Matter 21, 382203 (2009).
24. S. Kasahara, T. Shibauchi, K. Hashimoto, K. Ikada, S. Tonegawa, R. Okazaki, H. Shishido, H. Ikeda, H. Takeya, K. Hirata, T. Terashima, Y. Matsuda, Phys. Rev. B 81, 184519 (2010).