Anomalous Fermi surface in FeSe seen by Shubnikov-de Haas oscillation measurements

Taichi Terashima, Naoki Kikugawa, Andhika Kisman, Eun-Sang Choi, James S. Brooks, Shigeru Kasahara, Tatsuya Watanabe, Hiroaki Ikeda, Takasada Shibauchi, Yuji Matsuda, Thomas Wolf, Anna E. Böhmer, Frédéric Hardy, Christoph Meingast, Hilbert v. Löhnaysen, Michi-To Suzuki, Ryotaro Arita, and Shinya Uji

1National Institute for Materials Science, Tsukuba, Ibaraki 305-0003, Japan
2National High Magnetic Field Laboratory, Florida State University, Tallahassee, FL 32310, USA
3Department of Physics, Kyoto University, Kyoto 606-8502, Japan
4Department of Physical Sciences, Ritsumeikan University, Kusatsu, Shiga 525-8577, Japan
5Department of Advanced Materials Science, University of Tokyo, Chiba 277-8561, Japan
6Institute of Solid State Physics (IFP), Karlsruhe Institute of Technology, D-76021 Karlsruhe, Germany

We have observed Shubnikov-de Haas oscillations in FeSe. The Fermi surface deviates significantly from predictions of band-structure calculations and most likely consists of one electron and one hole thin cylinder. The carrier density is in the order of 0.01 carriers/Fe, an order-of-magnitude smaller than predicted. Effective Fermi energies as small as 3.6 meV are estimated. These findings call for elaborate theoretical investigations incorporating both electronic correlations and orbital ordering.

PACS numbers: 74.70.Xa, 71.18.+y, 74.25.Jb, 74.25.Op

I. INTRODUCTION

FeSe is an intriguing material among iron-based superconductors: The FeSe planes are isoelectronic with the (FeAs)\(^-1\) planes of the archetypal parent compounds of iron-based superconductors such as LaFeAsO (Ref. 1) or BaFe\(_2\)As\(_2\).\(^2\) However, FeSe shows only a structural phase transition at \(T_s \sim 100\) K without an accompanying magnetic phase transition and becomes superconducting below \(T_c \sim 8\) K.\(^3\) For comparison, BaFe\(_2\)As\(_2\) has structural and antiferromagnetic phase transitions at 140 K but does not exhibit superconductivity.\(^2\) As both transitions are suppressed by partial substitution of Ba, Fe, or As atoms, superconductivity emerges.\(^2,4\) Although the nature of the transition at \(T_s\) in FeSe is not yet clear, angle-resolved photoemission spectroscopy (ARPES) measurements on FeSe have found a splitting of the \(d_{xz}\) and \(d_{yz}\) bands at the corner of the Brillouin zone below \(\sim 110\) K,\(^5,7\) similar to one found in BaFe\(_2\)As\(_2\),\(^8\) suggesting orbital order.\(^9\) Secondly, the onset temperature of superconductivity can be enhanced up to \(\sim 37\) K by application of pressure.\(^10,11\) Moreover, it has recently been claimed that \(T_c\) in single-layer FeSe films may exceed 50 K.\(^12\) Finally, very recent magnetotransport, penetration depth, and spectroscopic-imaging scanning tunneling microscopy (STM) measurements on vapor-grown high-quality FeSe single crystals suggest that the Fermi energy \(E_F\) is extremely small and comparable to the superconducting energy gap \(\Delta\), as observed previously in Te-substituted alloys Fe(Se, Te) by ARPES measurements.\(^14,15\) FeSe may therefore be in the Bardeen-Cooper-Schrieffer (BCS)–Bose-Einstein-condensation (BEC) crossover regime.

Detailed research into the bulk electronic structure of FeSe is necessary to advance our understanding of these intriguing properties of FeSe, but such research was impeded by difficulties in single-crystal growth. Recently, Böhmer et al.\(^16\) have grown FeSe single crystals of unprecedented quality using a vapor transport technique. X-ray structural refinement has indicated a composition of Fe\(_{3.965(4)}\)Se.\(^16\) The composition very close to stoichiometry has further been confirmed by STM topographs as well as magnetotransport data indicating a nearly perfect carrier compensation.\(^13\) Using those crystals, we were able to observe Shubnikov-de Haas (SdH) oscillations in FeSe. Our central finding is that the observed Fermi surface (FS) is extremely small and strikingly different from band-structure calculations.

II. EXPERIMENTS

Standard four-contact resistance (\(R\)) measurements were performed with a 35-T resistive magnet and \(^3\)He or \(^3\)He/\(^4\)He dilution refrigerator at the NHMFL. The electrical contacts were spot-welded. The magnetic field (\(B\)) direction \(\theta\) is measured from the crystallographic \(c\) axis. Four samples with \(T_c\) and the resistance ratio (between room temperature and 11 K) of 8.9–9.2 K and 28–32, respectively, were investigated, and consistent results were obtained.

For a purely two-dimensional FS cylinder, there would be a single SdH frequency \(F\), and \(F \cos \theta\) would remain constant as \(\theta\) is varied. However, in real materials, there is some \(c\)-axis dispersion, which modulates the cross-section of the FS cylinder. In simple cases, two frequencies corresponding to the minimum and maximum cross-sections will appear and will exhibit upward and downward variations of \(F \cos \theta\), respectively, as \(|\theta|\) is increased.
and was subtracted from it to obtain $R_{\text{osc}}$ based on the second-harmonic data of sample 2. Samples except for the $\alpha_B$ in FeSe for $T = 0.04$ K. Note that the vertical axis is $F \cos \theta$ dependences of the SdH frequencies for samples 2 and 3. The upper two spectra were obtained for sample 2, for $-17^\circ$ for sample 3. $T = 0.04$ K.

The upper two spectra were obtained for sample 2, while the bottom one for sample 3. We find six frequency branches, $\alpha, 2\alpha, \beta, 2\beta, \gamma, \delta$. They are all small, and the corresponding orbits cover only 0.2–2.3% of the Brillouin zone [Table I and Fig. 3(b)]. Figure 2 shows the angle dependences of the SdH frequencies for samples 2 and 3. Note that the vertical axis is $F \cos \theta$. The two samples show consistent angle dependences. Although the data for sample 2 were those at $T = 0.4$ K because the angular variation was investigated more thoroughly in the $^4$He refrigerator, no new frequency was found in additional measurements on this sample at $T = 0.04$ K. Within experimental accuracy, $F_{2\alpha} = 2F_\alpha$, and $F_{2\beta} = 2F_\beta$, indicating that the $2\alpha$ and $2\beta$ frequencies are the second harmonics (we have also confirmed that $m^*_{2\beta} = 2m^*_{\beta}$ for $B \parallel c$ in sample 2). We have determined effective masses $m^*$ for $B \parallel c$ from the temperature dependences of the oscillation amplitudes as tabulated in Table I. Mean free paths $l$ can be estimated only roughly because of the limited range of inverse field. We find $l \sim 30$ and 80 nm for the $\beta$ and $\delta$ orbits in sample 2, respectively.

![Graph](image1.png)

**FIG. 1.** (color online). (a) Resistance $R$ and normalized oscillatory part $R_{\text{osc}}/R_{\text{background}}$ as a function of $B$. A fourth-order polynomial was fitted to the former above $B = 18$ T and was subtracted from it to obtain $R_{\text{osc}}$. (b) Fourier transforms of SdH oscillations in inverse field vs. $F \cos \theta$. Spectra for $\theta = 0$ and $49^\circ$ were taken for sample 2, for $-17^\circ$ for sample 3. $T = 0.04$ K.

**FIG. 2.** (color online). Angle dependences of the SdH frequencies. The vertical axis is $F \cos \theta$. The circles and crosses are data for sample 2 ($T = 0.4$ K) and 3 ($T = 0.04$ K), respectively. For the former, different frequency branches are indicated by different colors, and harmonics are indicated by hollow marks. The solid curves are hyperboloidal- and ellipsoidal-surface fits to $\alpha$ and $\beta$, and $\gamma$ and $\delta$ in sample 2, respectively (see Appendix B).

### III. RESULTS AND DISCUSSION

Figure 1(a) shows the resistance at $T = 0.04$ K in sample 2 as a function of $B$ applied parallel to the $c$ axis. After subtracting a smoothly varying background, we see clear SdH oscillations. Figure 1(b) shows Fourier transforms of the oscillations vs. $F \cos \theta$ for three field directions. The upper two spectra were obtained for sample 2, while the bottom one for sample 3. We find six frequency branches, $\alpha, 2\alpha, \beta, 2\beta, \gamma, \delta$. They are all small, and the corresponding orbits cover only 0.2–2.3% of the Brillouin zone [Table I and Fig. 3(b)]. Figure 2 shows the angle dependences of the SdH frequencies for samples 2 and 3. Note that the vertical axis is $F \cos \theta$. The two samples show consistent angle dependences. Although the data for sample 2 were those at $T = 0.4$ K because the angular variation was investigated more thoroughly in the $^4$He refrigerator, no new frequency was found in additional measurements on this sample at $T = 0.04$ K. Within experimental accuracy, $F_{2\alpha} = 2F_\alpha$, and $F_{2\beta} = 2F_\beta$, indicating that the $2\alpha$ and $2\beta$ frequencies are the second harmonics (we have also confirmed that $m^*_{2\beta} = 2m^*_{\beta}$ for $B \parallel c$ in sample 2). We have determined effective masses $m^*$ for $B \parallel c$ from the temperature dependences of the oscillation amplitudes as tabulated in Table I. Mean free paths $l$ can be estimated only roughly because of the limited range of inverse field. We find $l \sim 30$ and 80 nm for the $\beta$ and $\delta$ orbits in sample 2, respectively.

We first consider the issue of the BCS-BEC crossover. An effective Fermi energy $E_F$ can be estimated from experimental values of $F$ and $m^*$ using the following formulae: $E_F = \hbar^2 k_F^2/(2m^*)$, $A = \pi k_F^2$, and $F = \hbar A/(2\pi e)$, where $A$ is the orbit area in the $k$ space and we have assumed circular orbits. The estimated Fermi energies are very small (Table I). Hence the ratio $k_BT_c/E_F$ is large, ranging between 0.04 ($\delta$) and 0.22 ($\alpha$). The proximity to the crossover may also be assessed by the parameter $(\xi k_F)^{-1}$ corresponding to $\sim \Delta/E_F$.\(^{18,19}\) Using $\xi = 5.7$ nm (see Appendix A for the upper critical field and coherence length) and the estimated $k_F$ values (Table I), $(\xi k_F)^{-1} = 0.13$ ($\delta$) and 0.41 ($\alpha$). Since the BCS theory

| Branch | $F$ (kT) | $m^*/m_e$ | $A$ (%BZ) | $k_F$ ($A^{-1}$) | $E_F$ (meV) |
|--------|---------|----------|----------|----------------|-------------|
| $\alpha$ | 0.06 | 1.9(2) | 0.20 | 0.043 | 3.6 |
| $\beta$ | 0.20 | 4.3(1) | 0.69 | 0.078 | 5.4 |
| $\gamma$ | 0.57 | 7.2(2) | 2.0 | 0.13 | 9.1 |
| $\delta$ | 0.68 | 4.2(2) | 2.3 | 0.14 | 18 |

**TABLE I.** Experimental SdH frequencies, effective masses, orbit areas $A$, Fermi momentums and effective Fermi energies in FeSe for $B \parallel c$. The values were averaged over the four samples except for the $\alpha$ branch, for which the values are based on the second-harmonic data of sample 2. $m_e$ is the free electron mass.
We now switch to the Fermi surface. The angular dependences in Fig. 2 indicate that the conductors partially differ from those expected for single-band superconductors. It seems worth looking for possible manifestations of the BCS-BEC crossover in FeSe, though they may substantially differ from those expected for single-band superconductors. Based on these fits, we model the observed FS cylinders as shown in Fig. 3. We attribute α and γ to one cylinder and β and δ to another. This is the only reasonable combination: if β and γ were paired, extra minimum and maximum cross-sections would occur. Each cylinder has hyperboloidal, ellipsoidal, and connecting regions. We assume that the range of ks for both hyperboloidal and ellipsoidal regions is restricted by that covered by orbits for θ = 55°. For larger |θ|, orbits enter the cone-shaped connecting region outside this ks range. The α/γ cylinder contains 0.0093 carriers/Fe and the β/δ 0.015 carriers/Fe. We can also estimate contributions of the observed FS cylinders to the Sommerfeld coefficient. Using the average of the effective masses for the minimum and maximum orbits for each cylinder with a two-dimensional approximation, we obtain 3.2 and 3.0 mJ/molK² for the α/γ and β/δ cylinders, respectively. Since the effective masses for the α and γ orbits differ considerably, the former may not be very accurate, and, if the large effective mass is restricted to the swollen region of the cylinder near the γ orbit, it may be an overestimate.

For the sake of comparison, we have performed band-structure calculations for the orthorhombic structure using the WIEN2K code as shown in Fig. 4. The used lattice parameters are a = 5.3078 Å, b = 5.3342 Å, c = 5.486 Å, and zSe = 0.26689. The calculated FS consists of two electron cylinders at the Y point of the Brillouin zone and three hole cylinders at the Γ point, similar to the iron-pnictide parent compounds. The calculated carrier density and Sommerfeld coefficient are $n_e = n_h = 0.17$ carriers/Fe and $\gamma_{\text{band}} = 4.6$ mJ/mol K².

The question now is: are the two observed cylinders electrons or holes? Quantum oscillation measurements on the iron-pnictide parent compounds so far indicate that electron surfaces are generally easier to observe. It is thus tempting to assign the observed cylinders to the two calculated electron ones. However, considerations on the Sommerfeld coefficient are unfavorable to this sc
nario. A previous specific-heat measurement on a single crystal of FeSe0.963 in magnetic fields up to 9 T reported a Sommerfeld coefficient of 5.73±0.13 mJ/molK². A recent measurement on vapor-grown FeSe at B = 14 T has found a similar value (∼5.9 mJ/molK²). On the other hand, the sum of the above estimated coefficients for the observed cylinders is already 6.2 mJ/molK². Further, at least one unobserved hole cylinder would have to exist in this scenario to satisfy the carrier compensation, and effective masses for the hole cylinder would most likely be no smaller than those for the electron ones (otherwise oscillations from the hole cylinder would have been detected). Hence the total would become still larger and be difficult to reconcile with the specific-heat data.

The above considerations lead us to assume that we have observed the whole Fermi surface consisting of an electron and a hole cylinder. We may assign the α/γ cylinder to electrons (n_e = 0.0093 carriers/Fe) and β/δ to holes (n_h = 0.015 carriers/Fe). Then, the small carrier imbalance is consistent with the Fe-deficient composition within error. As shown below, this second scenario means radical changes to the calculated band structure, but it can be reconciled with reported ARPES data.

We first consider the holes. Inspection of the calculated band structure along the ΓZ line [Fig. 4(a)] suggests that, because of the k_z dispersion of bands, it is difficult to have a single hole cylinder at Γ by simple constant band-energy shifts. On the other hand, ARPES measurements on FeSe indicate that only one hole band crosses the Fermi level at low temperatures to form a single hole sheet at ΓZ. Further, Ref. 9 suggests that the k_z dispersion of the hole band along the ΓZ line is ∼10 meV. This is consistent with our β/δ cylinder, for which the k_z dispersion can be estimated from the difference in the effective Fermi energies of the β and δ orbits to be 13 meV. Strictly, Ref. 9 claims that the hole band sinks below E_F in parts of the ΓZ line to form a closed pocket rather than a cylinder. However, this discrepancy could be attributed to surface effects such as surface band-bending.

We next turn to the electrons. In the tetragonal structure, if the spin-orbit coupling is neglected, the two electron bands responsible for the electron cylinders are degenerate by symmetry along the MX and AR lines, which correspond to the YS and TR lines of the orthorhombic Brillouin zone. Even if the spin-orbit coupling and tiny orthorhombic distortion (|(a−b)/(a+b)| ∼ 2 × 10⁻³) are included in band-structure calculations, they remain quasi-degenerate along these lines and produce two electron cylinders as shown in Fig. 4. On the other hand, if we take the splitting of the d_xx and d_yy bands observed in ARPES measurements5-7 and anticrossing of bands into account, it is possible to have a single electron cylinder at the zone corner as illustrated in Fig. 5. Note however that this figure is very conceptual and that realistic models would have to include band renormalization and shifts, especially those of the d_{xy} band. Although the ARPES papers on FeSe do not state whether there is a single electron cylinder or two, there is an ARPES study on NaFeAs which shows that, while two electron cylinders exist at the zone corner in the tetragonal phase, only one exists in the orthorhombic phase due to the electronic reconstruction at T_s. We now discuss a remarkable disparity between the calculated and observed carrier densities: n_e = n_h = 0.17 carriers/Fe vs. n_e = 0.0093 and n_h = 0.015 carriers/Fe. In iron-based superconductors, upward and downward shifts of electron and hole bands, respectively, relative to band structure calculations are often found, resulting in smaller Fermi surfaces. This FS shrinking has been attributed to electronic correlation effects, especially interband scattering. For example, the Fermi surface of BaFe2(As1−xPx)x shrinks as x is decreased from 1, while the mass enhancement, a measure of the correlations, and T_c increase. The carrier density at x = 0.63 is 0.05 carriers/Fe. At x = 0.41, where T_c ∼ 25 K, the Fermi surface is roughly twice smaller than calculated. However, the magnitude of the present shrinking is the largest ever observed. It is interesting to note that the observed carrier density is fairly comparable to that in the antiferromagnetic state of BaFe2As2 (n_e = n_h = 0.006 carriers/Fe), where most of the paramagnetic FS has been destroyed by the reconstruction at the antiferromagnetic transition. There are some theoretical works on the electronic structure of FeSe where the dynamical mean-field theory (DMFT) or GW approximation is used to treat the electronic correlations beyond the level of conventional band-structure calculations. They predict slightly modified Fermi surfaces compared to conventional calculations but can not explain our extremely small Fermi surface.

In conclusion, we have observed SdH oscillations in FeSe. Our analyses indicate that the Fermi surface in the orthorhombic state is very different from that expected from band-structure calculations, most likely consisting of one hole and one electron tiny cylinders. To elucidate how this radical deviation occurs is an urgent task, when effects of both the electronic correlations and the orbital order have to be considered. It will be very interesting to see how this anomalous Fermi surface evolves as T_c increases with pressure.

ACKNOWLEDGMENTS

This work has been supported by Japan-Germany Research Cooperative Program, KAKENHI from JSPS and Project No. 56393598 from DAAD, and the Topological Quantum Phenomena (No. 25103713) KAKENHI on Innovative Areas from MEXT of Japan. A portion of this work was performed at the NHMFL, supported by NSF Cooperative Agreement DMR-1157490, the State of Florida, and the US DoE. JSB acknowledges support from NSF-DMR 1309146.
For $B \parallel c$, $B_0$ increases approximately linearly with decreasing $T$. A similar nearly linear or even convex variation of $c$-axis $B_{c2}$ has been reported for other iron-based superconductors and has been explained by multiband effects.\textsuperscript{37–41} For $B \parallel ab$, $B_0$ shows a tendency to saturation down to $\sim 2$ K, suggesting paramagnetic limiting, but shows an anomalous enhancement below. A similar low-$T$ enhancement has been reported for Fe(Se, Te)$\textsuperscript{32,43}$ and (Ba,K)Fe$_2$As$_2$.\textsuperscript{40}

The initial slopes are -1.6 and -6.9 T/K for $B \parallel c$ and $ab$, yielding coherence lengths $\xi$ of 1.3 and 5.7 nm for the $c$ and $ab$ directions, respectively.

The mass anisotropy $m_{||}/m_{\perp}$ is estimated to be 19, which is larger than 9.5 found in LiFeAs,\textsuperscript{44} contrary to the expectation that FeSe is more three-dimensional.

Appendix B: ellipsoidal and hyperboloidal-surface fits

For an ellipsoidal (+) or a hyperboloidal (−) Fermi surface $k_{ab}^2/(k_{oc}^2) = 1$, where $k_{ab}$ and $k_c$ are the $ab$ plane and $c$ axis components of the $k$ vector, respectively, the angle dependence of the frequency is given by $F(\theta) = F(0) \cos^2 \theta \pm (k_{ab}^2/k_{oc}^2)^2 \sin^2 \theta^{-1/2}$. The fitting results shown in Fig. 2 are $k_{ab}^2 / k_{oc}^2 = 1.3$ and $1.6$, sign) = (0.042, 0.086, −), (0.078, 0.17, −), (0.13, 0.24, +), and (0.14, 0.37, +) for $\alpha$, $\beta$, $\gamma$, and $\delta$, respectively.

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