Fermi Surface with Dirac Fermions in CaFeAsF Determined via Quantum Oscillation Measurements

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Despite the fact that 1111-type iron arsenides hold the record transition temperature of iron-based superconductors, their electronic structures have not been studied much because of the lack of high-quality single crystals. In this study, we comprehensively determine the Fermi surface in the antiferromagnetic state of CaFeAsF, a 1111 iron-arsenide parent compound, by performing quantum oscillation measurements and band-structure calculations. The determined Fermi surface consists of a symmetry-related pair of Dirac electron cylinders and a normal hole cylinder. From analyses of quantum-oscillation phases, we demonstrate that the electron cylinders carry a nontrivial Berry phase \( \pi \). The carrier density is of the order of \( 10^{-3} \) per Fe. This unusual metallic state with the extremely small carrier density is a consequence of the previously discussed topological feature of the band structure which prevents the antiferromagnetic gap from being a full gap. We also report a nearly linear-in-\( B \) magnetoresistance and an anomalous resistivity increase above about 30 T for \( B || c \), the latter of which is likely related to the quantum limit of the electron orbit. Intriguingly, the electrical resistivity exhibits a nonmetallic temperature dependence in the paramagnetic tetragonal phase (\( T > 118 \) K), which may suggest an incoherent state. Our study provides a detailed knowledge of the Fermi surface in the antiferromagnetic state of 1111 parent compounds and moreover opens up a new possibility to explore Dirac-fermion physics in those compounds.

I. INTRODUCTION

Since the discovery of superconductivity at \( T_c = 26 \) K in \( \text{LaFeAs}(O_{1-x}F_x) \) by Kamihara et al. [1], iron-based superconductors have been studied extensively. By replacing La with smaller rare-earth elements, \( T_c \) of up to 56 K was achieved in the same 1111-type structure immediately after the discovery [2-5]. There are now various structure types: the 122- (e.g. \( \text{BaFe}_2\text{As}_2 \)) [6, 7], 111- (e.g. \( \text{LiFeAs} \)) [8, 9], 11-type (e.g. \( \text{FeSe} \)) [10] structures and so on. Nonetheless, the above \( T_c \) record has not yet been surpassed by other structure types as far as bulk materials are concerned. It is therefore crucial to unravel the normal-state electronic structure of the 1111 iron arsenides from which the highest \( T_c \) among the iron-based superconductors emerges. Unfortunately, however, such studies suffer from difficulty in obtaining high-quality single crystals of the 1111-type iron arsenides.

There is a variant of the 1111 iron arsenides, i.e., \( \text{CaFeAsF} \) (or \( \text{SrFeAsF} \)) [12, 13] that has the same \( \text{ZrCuSiAs} \)-type structure as \( \text{LaFeAsO} \), but the LaO layers in \( \text{LaFeAsO} \) are replaced by the CaF layers (Fig. 1). The electronic band structure near the Fermi level \( E_F \) is predicted to be similar to that of \( \text{LaFeAsO} \) [15]. \( \text{CaFeAsF} \) exhibits a structural and an antiferromagnetic transition as \( \text{LaFeAsO} \) does [13, 16]. The antiferromagnetic structure is the same stripe-type one as \( \text{LaFeAsO} \). By partial substitution of Fe by Co, \( \text{Ca(Fe, Co)AsF} \) exhibits superconductivity with a maximum \( T_c \) of 22 K [12]. Further, \( T_c \) of 56 K has been observed for rare-earth doped compounds [17, 18]. Following initial reports of single-crystal growth of \( \text{(Ca, Na)FeAsF} \) [19] and \( \text{CaFeAsF}_{1-x} \) [20], Ma et al. have succeeded in growing large high-quality single crystals of undoped \( \text{CaFeAsF} \) by self-flux method [21].

In this article, we report resistivity and magnetic-torque measurements down to 0.03 K (0.4 T) up to 18 T (45 T) performed on high-quality single crystals of \( \text{CaFeAsF} \). The temperature dependence of the resistivity is nonmetallic down to \( T_s = 118 \) K and exhibits two clearly separated anomalies at \( T_s \) and \( T_N = 107 \) K, which are attributed to a structural and an antiferromagnetic transition, respectively. We observe clear quantum oscillations both in the resistivity and magnetic torque in the antiferromagnetic ground state. Two frequencies \( \alpha \) and \( \beta \) are resolved, and their field-angle dependences indicate that they are from highly two-dimensional Fermi-surface cylinders. From detailed analyses of the oscillation phases, we find a nontrivial Berry phase \( \pi \) associated with the \( \alpha \) oscillation, demonstrating that the \( \alpha \)
cylinder is a Dirac-fermion cylinder. Our band-structure calculations indicate that the Fermi surface consists of a hole cylinder at the Γ point of the Brillouin zone and a symmetry-related pair of electron cylinders originating from Dirac lines located away from Γ. Thus the α and β cylinders are attributed to the electron and hole cylinders, respectively. The carrier density is extremely small, of the order of $10^{-3}$ per Fe. Additionally, we report a nearly linear-in-$B$ magnetoresistivity and an anomalous resistivity increase above about 30 T for $B \parallel c$. Among other things, our work opens up a new possibility to explore Dirac-fermion physics in a branch of the family of the iron-based superconductors with a simple band structure.

II. METHODS

High-quality single crystals of CaFeAsF were prepared by a CaAs self-flux method in Shanghai [21]. The crystals were platelike with an $ab$-plane area of up to $\sim 1 \text{ mm}^2$ and a thickness of $\sim 50 \mu$m or less.

A 20-T superconducting magnet with a dilution refrigerator at the National Institute for Materials Science and the 45-T hybrid magnet with a $^3$He refrigerator at the National High Magnetic Field Laboratory were used. In either magnet, samples can be rotated in situ, and the field angle $\theta$ is measured from the $c$ axis. Electrical resistivity $\rho$ measurements were performed with a four-contact method both for current $I \parallel ab$ and $I \parallel c$ (see Fig. 2 for the contact configuration for $I \parallel c$). The contacts were spot-welded or made with conducting silver paste. A total of six samples were prepared: two $ab$-plane and one $c$-axis samples were used for SdH measurements, while the remaining one $ab$-plane and two $c$-axis samples were used only for zero-field temperature-dependence measurements. Microcantilever torque $\tau$ measurements were performed on another two crystals. Consistent quantum-oscillation results were obtained for the three SdH and two torque crystals.

Fully relativistic electronic band-structure calculations with spin-orbit coupling were performed by using the WIEN2k code with the PBE-GGA potential [22, 23]. We used the experimental lattice parameters of the orthorhombic crystal structure (space group Cmma) at $T = 2$ K: $a = 5.514594 \AA$, $b = 5.476802 \AA$, $c = 8.576845 \AA$, $z_{As} = 0.66329$ [16][24]. The antiferromagnetic ordering with $Q = (1,0,1/2)$ (w.r.t Cmma) [16] was incorporated by using space group Ibam (#72) with a doubled $c$ dimension (Fig. 1). In the self-consistent calculations, the muffin-tin radii $R_{MT}$ of Ca, Fe, As and F atom were set to 2.15, 2.31, 2.19, and 2.26 in units of the Bohr radius, respectively. The maximum modulus of reciprocal vectors was chosen such that $R_{MT} K_{max} = 7.0$. The corresponding Brillouin zone was sampled using a $10 \times 10 \times 10$ $k$-mesh for the self-consistent calculations, and $40 \times 40 \times 40$ $k$-mesh for calculations in the density of states.

III. TEMPERATURE DEPENDENCE OF RESISTIVITY

Figure 2 shows the in-plane and $c$-axis resistivities as a function of temperature $T$. The resistivities increase initially as $T$ is lowered from room temperature as reported in [21]. These measurements were made on the
very same high-quality single crystals that exhibit quantum oscillations at low temperatures. This indicates that the observed non-metallic behavior is intrinsic nature of CaFeAsF.

The resistivities decrease sharply below \( T_s = 118 \) K and exhibit a kink at \( T_N = 107 \) K as reported in [21]. These anomalies at \( T_s \) and \( T_N \) most likely correspond to a structural and an antiferromagnetic phase transition, respectively, as observed in a neutron powder-diffraction experiment by Xiao et al [16]. The \( c \)-axis resistivity of this sample exhibits a sudden faint drop at \( T = 20 \) K, which is not observed in the other samples and hence is probably due to a crack. The resistivity anisotropy is large and basically increases with decreasing temperature.

The room-temperature in-plane and \( c \)-axis resistivities, averaged over the three in-plane and three \( c \)-axis samples, are 1.9(8) mΩcm and 2.3(6) \( \times 10^2 \) mΩcm, respectively. The large errors can be ascribed to errors in the sample dimensions, irregular sample shapes and inhomogeneous current distribution. The large anisotropy \( \rho_c/\rho_{ab} \) of the order of \( 10^2 \) is broadly consistent with the large upper-critical-field anisotropy of 9 observed for CaFe0.85Co0.15AsF [25] as the two anisotropies are approximated by \( m_c^*/m_{ab}^* \) and \( \sqrt{m_c^*/m_{ab}^*} \), respectively.

### IV. QUANTUM OSCILLATIONS

#### A. Theoretical background

Quantum oscillation of the resistivity \( \Delta \rho \) can be described by [26, 27]

\[
\frac{\Delta \rho}{\rho_0} = -C \sqrt{B} R_T R_D R_s \cos \left[ \frac{2\pi}{\beta} \left( \frac{F}{B} - \frac{1}{2} \right) + \phi_D + \phi_B \right],
\]

where \( \rho_0 \) is the background resistivity, \( C \) a positive coefficient, and we have neglected harmonics. We have tacitly assumed that \( \sigma = \rho/(\rho^2 + \rho_H^2) \approx \rho^{-1} \) since the Hall resistivity \( \rho_H \) is much smaller than \( \rho \) [18]. The frequency \( F \) is related to a Fermi-surface cross sectional area \( A \) as \( F = (\hbar/2\pi\epsilon)A \). The temperature and Dingle reduction factors are given by \( R_T = X/\sin X \) and \( R_D = \exp(-X_D) \), where \( X(D) = K\mu^*T(D)/B, \mu^* = m^*/m_e \), and the coefficient \( K \) is 14.69 T/K. The Dingle temperature \( T_D \) is inversely proportional to the carrier scattering time \( \tau \): \( T_D = \hbar/2\pi k_B \tau \). The spin reduction factor \( R_s \) describes the interference of oscillations from up- and down-spin electrons and is given by \( R_s = \cos(\pi g_\mu^*/2) \), where \( g \) is the spin \( g \) factor. \( \phi_D \) is 0 for a two-dimensional (2D) Fermi-surface (FS) cylinder while it is \( +\pi/4 \) when the oscillation is from a minimum or maximum cross section of a three-dimensional (3D) FS pocket. \( \phi_B \) is the Berry phase, which is 0 for normal electrons but \( \pi \) for Dirac fermions [28, 29]. The sign of \( R_s \) is essential to determine the Berry phase in systems where electronic bands are doubly degenerate at each \( k \) due to spin degeneracy of freedom as is the case with the present one [30]. If \( R_s > 0 \) and \( \phi_D \) is neglected, resistivity maxima occur for normal fermions when \( F/B \) takes integer values, while minima for Dirac fermions. If \( R_s < 0 \), the opposite is true.

#### B. Frequency and effective mass

Figure 3(a) shows the in-plane resistivity \( \rho \) and field derivative \( d\rho/dB \) as a function of \( B \) up to 17.8 T applied...
along the c axis. The field derivative shows an initial rise up to \(B \sim 0.6\) T, above which it stays approximately constant except oscillations. Namely, the magnetoresistance is nearly linear in \(B\).

Quantum oscillations are discernible above \(B \sim 5\) T. Figure 3(b) shows Fourier transforms of oscillations as a function of \(F\cos\theta\) for various field directions \(\theta\). Two frequencies \(\alpha\) and \(\beta\) are resolved: \(F_\alpha = 19.6\) T and \(F_\beta = 46.7\) T for \(B \parallel c\), which correspond to 0.14 and 0.34\% of the cross section of the antiferromagnetic Brillouin zone. For both frequencies, \(F\cos\theta\) stays constant for the investigated \(\theta\) range, indicating that they are from highly 2D FS cylinders. The effective masses associated with the \(\alpha\) and \(\beta\) orbits for \(B \parallel c\) are estimated from the temperature dependences of the oscillation amplitudes to be 0.383(4) and 0.92(1), respectively, in units of the free electron mass \(m_e\) [Fig. 3(c)]. Figure 3(d) shows the field dependence of the \(\beta\) oscillation amplitude in the form of a Dingle plot. The amplitudes were estimated from the extremums of the pure \(\beta\) oscillation obtained below in Sec. IV C. From the linear fit to the data, \(T_D = 2.7\) K, corresponding to a carrier mean free path of 21 nm (\(l = \tau\nu_F\), see Table I for \(\nu_F\)).

In order to search for higher frequencies, i.e., larger FS pockets, we extended measurements up to \(B = 45\) T by using the hybrid magnet. The inset of Fig. 4(a) shows semilogarithmic plots of in-plane resistivity vs field. For \(B \parallel c\), the resistivity at \(T = 0.4\) K shows a remarkable increase above \(B \sim 30\) T. The increase is suppressed as the temperature is raised, and almost disappears at \(T = 20\) K. For nearly \(B \parallel ab\) \((\theta = 91^\circ)\), no such an increase is observed. A similar behavior is observed for the \(c\)-axis resistivity (data not shown). Although the origin of the anomalous resistivity increase is not clear at present, it may be related to the quantum limit (i.e., the limit where only the lowest Landau level is occupied) of the \(\alpha\) orbit \((F_\alpha = 19.6\) T). Apart from the anomalous increase, the undulation of the \(T = 0.4\) K curve for \(B \parallel c\) is dominated by the \(\beta\) oscillation: the maxima at \(B \sim 12, 16, 23\) T are ascribable to it, and no faster oscillation is seen.

The main panel of Fig. 4(a) shows Fourier transform analysis of resistivity data measured in the hybrid magnet. We have analyzed the logarithmic derivative \(d\ln\rho dB\) instead of \(d\rho dB\) to suppress huge background due to the anomalous resistivity increase near \(B \parallel c\). The spectra show peaks corresponding to \(\alpha\), \(\beta\), and their harmonics: peaks around 100 T are ascribable to \(2\beta\) and/or \(6\alpha\). No higher fundamental frequency than \(\beta\) exists.

We also measured magnetic torque [Fig. 4(b)]. The undulation of the torque curves is again basically explicable with the \(\beta\) oscillation, and no faster oscillation is seen. The Fourier spectra confirm that there is no frequency higher than \(\beta\). To summarize, our experimental results indicate that the Fermi surface is composed of only two types of extremely small cylinders \(\alpha\) and \(\beta\).

![Figure 4](image)

FIG. 4. (a, inset) In-plane resistivity in CaFeAsF as a function of \(B\) up to 45 T for \(B \parallel c\) and nearly \(B \parallel ab\) \((\theta = 91^\circ)\). (a, main) Fourier transforms of \(d\ln\rho dB\) and \(d\tau dB\) vs \(1/B\) plotted against \(F\cos\theta\).

C. Oscillation phase

We now consider the spin reduction factors \(R_s\) and oscillation phases by analyzing the waveform of the observed oscillations. To avoid ambiguity due to background subtraction, we subtract the same linear background \(a + b\cos\theta\) with \(a = 260 \mu\Omega\) cm and \(b = 89.9 \mu\Omega\) cm/T from the raw \(\rho(B)\) data. The parameters were determined by fitting a line to the \(\rho(B)\) curve of Fig. 3(a) in the field range 5–17.8 T.

For a 2D FS cylinder, the effective mass varies as \(1/\cos\theta\) as the field direction \(\theta\) is varied. \(R_s\) varies accordingly and may cross zero to change sign. For such a “spin-zero” \((R_s = 0)\), \(\mu g\) = 1 + 2\(n\) where \(n\) is an integer.

We begin with \(R_s\) for the \(\beta\) oscillation. The Fourier transforms in Fig. 4(a) show that the \(\beta\) amplitude almost
FIG. 5. (a) Oscillatory part of the resistivity as a function of $1/(B \cos \theta)$ for field directions near $\theta = 52.6^\circ$. All the curves are from the hybrid measurements. (b) Oscillatory part of the resistivity as a function of $1/(B \cos \theta)$ after box smoothing. The box (window) size is the same as one oscillation period of the $\beta$ frequency, and hence the $\beta$ oscillation is effectively removed. The curves for $\theta = 0, 28.3,$ and $48.3^\circ$ are from the 20-T measurements, while the others from the hybrid measurements.

vanishes at $\theta = 52.6^\circ$ but revives at $57.4^\circ$, suggesting a spin-zero near $52.6^\circ$. Figure 5(a) shows the oscillatory part of the resistivity for field directions around $\theta = 52.6^\circ$. The curves at $\theta = 38.1, 42.9,$ and $47.7^\circ$ show a minimum near $F_{\beta}/B = 2$. Since the fundamental $\beta$ oscillation is weak at these angles ($\theta = 47.7$ and $52.6^\circ$) because of $R_s \approx 0$, the waveform is strongly influenced by the harmonics and the $\alpha$ oscillation, which explains the fact that the peak positions at these angles slightly differ from the other angles. Similarly, looking at $F_{\beta}/B = 3.5$, we notice the second spin-zero between $\theta = 62.2$ and $67.0^\circ$ and the third between $67.0$ and $71.8^\circ$.

Let us assume that the first spin-zero is at $\theta = 50^\circ$, where $\mu^* = 1.43$ and hence $g = 0.699(1 + 2n)$. If $n = 2$ ($g = 3.49$), the second and third spin-zeros occur at $\theta = 62.6$ and $69.1^\circ$, which are consistent with the experimental observations. No other choice of $n$ is consistent with the experimental observation: $n < 2$ cannot explain the second spin-zero while $n > 2$ produces extra spin-zeros. Since $\mu^* = 0.92$ at $\theta = 0$, $\pi g \mu^*/2 = 1.61\pi$ and hence $R_s > 0$. The conclusion $R_s > 0$ at $\theta = 0$ is confirmed by a more general analysis which shows that the experimental observations can be explained with a $g$ value between 3.30 and 3.55, corresponding to the first spin-zero between $49.2$ and $52.6^\circ$. Since $\Delta \rho$ shows a maximum at $F_{\beta}/B = 3$ at low angles where $R_s > 0$ [Figs. 5(a) and 5(a)], the $\beta$ carriers are normal fermions ($\phi_B = 0$).

We now consider $\phi_D$ for the $\beta$ orbit. Figure 6 shows the oscillatory part of the resistivity for $B \parallel c$ as a function of $1/B$. We decompose this data into the $\beta$ oscillation and the rest, the latter of which contains the $\alpha$ oscillation, as follows: We determine the positions of the zeros of the $\beta$ oscillation (crosses in the figure) by interpolating the maximum and minimum positions of $\Delta \rho$. Since $R_s > 0$, the $\beta$ oscillation (lower curve, vertically shifted) is obtained by subtracting the pure $\beta$ oscillation (upper curve, vertically shifted) from the data, and obtain the pure $\beta$ oscillation.
oscillation (lower curve). The Fourier transforms in Fig. 6(b) confirms the success of the decomposition. Figure 6(c) shows a Landau-index plot based on the resistivity maxima and minima of the thus obtained $\beta$ oscillation. Integer and half integer indices are assigned to resistivity maxima and minima, respectively. A linear fit (solid line) gives a slope of 0.0213 T$^{-1}$, which corresponds to $F_\beta = 47.0$ T, in excellent agreement with the Fourier analysis. The extrapolation of the line intercepts the 1/B = 0 line at $n = 0.02(2)$. The obtained intercept is much closer to the 2D value (0) than the 3D values ($\pm 1/8$) and supports the two dimensionality of the $\beta$ cylinder. Note that the intercept also confirms that $\phi_B = 0$

We next examine the $\alpha$ oscillation. Figure 5(b) shows the oscillatory resistivities after box-smoothing. The box (window) size has been matched to the period of the $\beta$ oscillation so that the $\beta$ oscillation has been suppressed effectively. Looking at $F_\alpha/B = 2$, we notice that the curves show a minimum for $\theta < 48.3^\circ$ but a maximum for $\theta \geq 57.4^\circ$ up to 71.8$^\circ$ (it is difficult to classify the curve for $\theta = 52.6^\circ$). This indicates that the first spin-zero for the $\alpha$ oscillation occurs between 48.3 and 57.4$^\circ$. Let us assume that it is at $\theta = 50^\circ$, where $\mu^g = 0.596$ and hence $g = 1.68(1+2n)$. For $n \geq 1$, $R_s$ changes sign at least once between $\theta = 57.4$ and 71.8$^\circ$, which is inconsistent with the experimental observation. Hence, $n = 0$, and $R_s > 0$ at $\theta = 0 (\pi g \mu^g/2 = 0.321\pi)$. A more general analysis indicates that a $g$ value between 1.41 and 1.73 can explain the experimental observation and confirms $R_s > 0$ at $\theta = 0$. Since the curve for $\theta = 0$ shows a minimum near $F_\alpha/B = 2$ and maxima near $F_\alpha/B = 1.5$ and 2.5, $\phi_B = \pi$ for the $\alpha$ oscillation. That is, the $\alpha$ cylinder is a Fermi surface of Dirac fermions. (It is difficult to determine whether $\phi_D = 0$ or $\pm \pi/4$ for the $\alpha$ oscillation: the number of the observed oscillation periods is too small and the resistivity maxima and minima cannot be determined accurately because the oscillation is weak.)

V. COMPARISON WITH BAND-STRUCTURE CALCULATIONS

We now consider results of our band-structure calculations. Figure 7 shows the calculated density of states, band structure, and Fermi surface. Those results are reminiscent of similar results for the antiferromagnetic state of LaFeAsO \cite{31}. The density of states shows that the Fermi level is located at the bottom of a deep pseudo gap. The density of states at $E_F$ is of the order of 0.1 states/eV per spin per cell. The difference in the occupied states below $E_F$ between up- and down-spin at the Fe1 or Fe2 site gives an antiferromagnetic moment of 2.0 $\mu_B$, which is much larger than the experimental value of 0.49(5) $\mu_B$ \cite{16}. We realize that the significant difference in the Fe1 (or Fe2) partial density of states between up- and down-spin is not restricted to near $E_F$ but exists even at 4 eV below $E_F$. This is very different from a textbook picture of a spin-density wave where the spin polarization is expected to occur only near $E_F$ and indicates that the antiferromagnetic order involves states far below $E_F$ as was pointed out early on in e.g. \cite{32}.

The calculated band structure [Fig. 7(b)] shows that two bands cross $E_F$, giving rise to a hole cylinder at the
center of the Brillouin zone Γ and two symmetry-related electron cylinders off Γ toward the antiferromagnetic α direction [Fig. 7(b)]. We note that the electron cylinder encloses a Dirac line: See the band structure along the line between Γ and ‘π/a’. An e and a t2g band cross immediately below E_F without spin-orbit coupling. Spin-orbit coupling induces a gap of about 5 meV, which is too small to be visible in Fig. 7(b). We also note that the two E_F-crossing bands are very flat along the line ΓZ, which indicates a highly two-dimensional electronic state and is consistent with the large anisotropy of the resistivity. This contrasts with the case of antiferromagnetic state of BaFe_2As_2, where the band structure is three dimensional, producing closed Fermi pockets [33, 34].

We attribute the α and β frequencies to the electron and hole cylinders, respectively. The Dirac nature of the α cylinder is thus confirmed theoretically as well. Based on this assignment and from the experimental frequencies and effective masses, we can estimate the carrier densities to be n_ε = 1.7 x 10^{13} holes/Fe and n_e = 1.4 x 10^{13} electrons/Fe, and the Sommerfeld coefficient to be 1.3 mJ/(mol K^2). The slight imperfection of the carrier compensation can be ascribed to possible small corrugation of the α cylinder, error in the α frequency, and/or off-stoichiometry. See table I for other parameters such as Fermi velocity and energy.

### VI. DISCUSSION

First of all, the standard band-structure calculations explain the Fermi surface in the antiferromagnetic state of CaFeAsF reasonably well despite the fact that they significantly overestimate the antiferromagnetic moment. This is very similar to what we saw in the case of the antiferromagnetic state of BaFe_2As_2 [34]. Since the antiferromagnetic ordering involves deep states (at least) down to 4 eV below E_F, band-energy adjustments of order of 100 meV, which are often employed to improve the agreement between the experimental and calculated Fermi surfaces, cannot resolve the disagreement about the moment.

It is interesting to note here that the Fermi surface in the low-temperature nonmagnetic state of FeSe strikingly differs from that expected from band-structure calculations [35–42]: notably, the carrier density is more than one order-of-magnitude smaller than calculated [35]. In the case of FeSe, only the structural (nematic) transition occurs without antiferromagnetic ordering [43]. The stark contrast between the above antiferromagnetic compounds and FeSe seems to suggest that interactions that make the Fermi surface significantly deviate from band-structure calculations are largely suppressed by antiferromagnetic ordering.

The observed antiferromagnetic metallic state of CaFeAsF is unusual in a sense that the carrier density is as low as 10^{-3} per Fe and that one type of the carriers is Dirac fermions. One may ask why CaFeAsF does not become an insulator after the antiferromagnetic ordering despite the strong antiferromagnetic interaction involving states far below E_F. The unusual metallic state is actually a decisive demonstration of an early theoretical idea by Ran et al. [44] (see also [45]). According to Ran et al., CaFeAsF is not allowed to become an insulator. Because of the topological feature of the iron-pnictide band structures, the antiferromagnetic gap has to have two Dirac nodes (in a 2D approximation). The nodes are close to but offset from E_F, giving rise to FS pockets.

The theory by Ran et al. is also applicable to the antiferromagnetic state of RFeAsO (R = rare earth) and AFe_2As_2 (A = Ba, Sr, or Ca). The existence of Dirac fermions in those compounds has been suggested by magnetotransport [46–48], angle-resolved photoemission spectroscopy [33, 49–50], and optical measurements [51, 52]. The Fermi velocity (∼5 × 10^6 m/s) of the Dirac cone in BaFe_2As_2 estimated from angle-resolved photoemission data [49] is close to that of the present α cylinder (Table I). However, quantum oscillation measurements so far have not confirmed a nontrivial Berry phase in those compounds [51, 52, 53, 54]. Thus this is the first observation of a nontrivial Berry phase in iron-based superconductor parent compounds.

It would be interesting if one could eliminate the hole cylinder by slightly electron-doping CaFeAsF. One could investigate transport properties of Dirac fermions. Further, one might be able to induce topological superconductivity of Dirac fermions. In the case of BaFe_2As_2, in addition to the Dirac electron pockets (the γ pockets in [44]), there are hole (α) and electron (δ) pockets, and it is difficult to simultaneously remove both of the latter pockets by doping. Therefore CaFeAsF is best suited to testing those possibilities of Dirac-fermion physics.

We have observed nearly B-linear magnetoresistance [Fig. 3(a)]. Similar B-linear magnetoresistance has been reported for other Dirac-fermion systems such as Bi [60, 61], Cd_3As_2 [62, 63], Na_3Bi [64], TlBiSSe [66], and ZrSiS [67], and further it has been reported for RFeAsO

### Table I. Carrier type, quantum-oscillation frequency F, effective mass m^*, carrier density n, Fermi momentum k_F, Fermi velocity v_F, and effective Fermi energy E_F determined from the quantum-oscillation measurements. B || c for F and m^*. n for the α cylinder refers to the sum of the carriers of the two cylinders occurring in the Brillouin zone. To derive E_F, a linear dispersion (E_F = h v_F k_F) was used for the α cylinder, while a quadratic one (E_F = h^2 k_F^2/(2m^*)) for β.

| FS cylinder | α     | β     |
|-------------|-------|-------|
| carrier type | Dirac electron | normal hole |
| F (T)       | 19.6  | 46.7  |
| m^*/m_e     | 0.383(4) | 0.92(1) |
| n (10^{-3} per Fe) | 1.4  | 1.7   |
| n (10^{19} cm^{-3}) | 2.2  | 2.6   |
| k_F (Å^{-1}) | 0.024 | 0.038 |
| v_F (10^4 m/s) | 7.4  | 4.7   |
| E_F (meV)   | 12    | 5.9   |
The nonmetallic conductivity above $T_s$ is enigmatic. Band-structure calculations for the paramagnetic state predict a large Fermi surface similar to that in the paramagnetic state of other 1111 or 122 compounds [25]. On the other hand, it has theoretically been suggested that as the $\text{As}$ height, the vertical distance of $\text{As}$ from the Fe plane, increases, the Fe $3d$ band width decreases, leading to enhanced electronic fluctuations and stronger electronic correlations [70, 71]. The $\text{As}$ height in CaFeAsF is $1.41 \text{ Å}$ [21], which is appreciably larger than $1.32 \text{ Å}$ in LaFeAsO [72]. Therefore, because of enhanced fluctuations, CaFeAsF above $T_s$ may be in an incoherent state where the Fermi surface is not well-defined.

We also note that the conductivity becomes metallic below the structural (nematic) transition at $T_n$, not the antiferromagnetic one at $T_N$. Since no band folding occurs at $T_n$ and the orthorhombic distortion is tiny (0.3%), almost no change is expected for the calculated Fermi surface at $T_n$. This may indicate that the metallic conductivity below $T_n$ is owing to the suppression of nematic fluctuations, which in turn may indicate the importance of those fluctuations in driving CaFeAsF into an incoherent state.

It is interesting to note that the conductivity near room temperature becomes metallic with Co doping: the resistivity of Ca(Fe$_{0.88}$Co$_{0.12}$)AsF decreases from room temperature down to about 100 K, where it exhibits a broad and shallow minimum [16, 73]. This behavior is very similar to that observed in doped $\text{RF}$eAsO, e.g., LaFeAsO($1-x$)$_2$F$_x$ with $x = 0.03$ or 0.05 and Sm(Fe$_{0.92}$Co$_{0.08}$)AsO [74, 76].

Finally, we mention a very recent report of quantum oscillation measurements on SmFeAsO [59]. Only one frequency was observed for $B \parallel c$, and its angular dependence was not reported. The reported frequency $F = 65(5)$ T is close to the $\beta$ frequency, but the effective mass $0.5(1) m_e$ is about half of the $\beta$ mass.

**VII. SUMMARY**

We have comprehensively determined the Fermi surface of antiferromagnetic CaFeAsF: it consists of the $\beta$ hole cylinder at the $\Gamma$ point and a pair of the $\alpha$ Dirac electron cylinders symmetrically located at positions off $\Gamma$ toward the antiferromagnetic direction. We have proved that a nontrivial Berry phase $\pi$ is associated with the $\alpha$ Dirac cylinders. The carrier density is extremely small, of the order of $10^{-3}$ per Fe. This unusual metallic state is a consequence of the previously studied topological nature of the band structure [44]. A nearly linear magnetoresistance is observed as in other Dirac-fermion systems. The anomalous resistivity increase observed above $\sim 30$ T for $B \parallel c$ is likely related to the quantum limit of the $\alpha$ orbit. The band-structure calculations describe the Fermi surface reasonably well. The same calculations however largely overestimate the antiferromagnetic moment. Electron interactions that preserve the Fermi surface have to be sought to resolve this problem. The nonmetallic conduction observed above $T_s$ is intriguing given that the low-temperature antiferromagnetic state is metallic with a well-defined Fermi surface. It may suggest that CaFeAsF is in an incoherent state above $T_s$. Finally, slight electron-doping can make CaFeAsF a system composed solely of Dirac fermions. Investigations of transport and other properties there would be fruitful, and moreover Dirac-fermion superconductivity might be induced.

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