

Quantum oscillations in iron-based superconductors: BaFe₂As₂ vs. KFe₂As₂

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Abstract. We present results of Shubnikov-de Haas oscillation measurements on detwinned BaFe₂As₂ and de Haas-van Alphen oscillation measurements on KFe₂As₂. The Fermi surface of BaFe₂As₂ in the antiferromagnetic phase is found to consist of one hole and two electron pockets, all of which are three-dimensional and closed, and can reasonably be accounted for by LSDA band calculations. We find only moderate mass enhancements m^*/m_{band} of 2–3. In the case of KFe₂As₂, four quasi-two-dimensional Fermi surface cylinders ϵ , α , ζ , and β are observed in qualitative agreement with previous ARPES data. In sharp contrast to BaFe₂As₂, agreement between the observed and LDA-calculated Fermi surface is poor: LDA calculations seem to predict wrong crystal-field splitting of Fe 3d states. Large effective masses up to 20 m_e , m_e being the free electron mass, are found. The Sommerfeld coefficient estimated from the observed Fermi surface and effective masses is consistent with the measured value of 93 mJ/K²mol [H. Fukazawa *et al.*, J. Phys. Soc. Jpn. **80**, SA118 (2011)] and is 8–9 times larger than the band value, indicating strong electronic correlations in KFe₂As₂.

1. Introduction

Since the discovery of superconductivity at $T_c = 26$ K in LaFeAs(O, F) [1], extensive efforts have been devoted to the elucidation of the mechanism and paring symmetry of the iron-based high- T_c superconductivity (for reviews, see Refs. [2, 3, 4]). At the beginning, it looked like



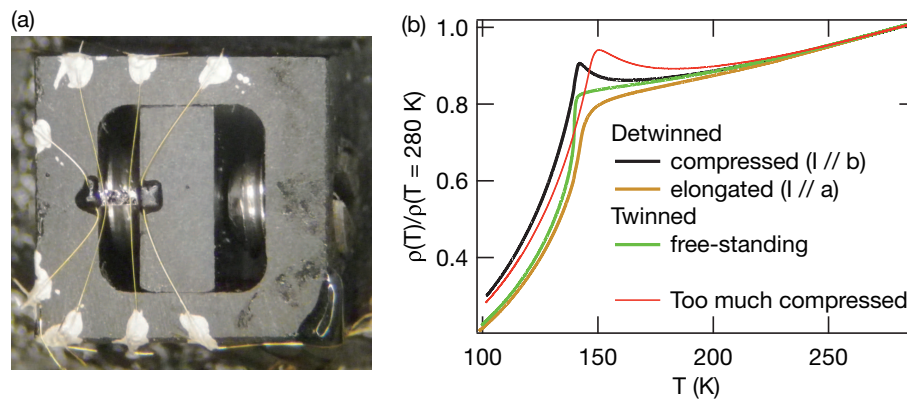


Figure 1. (a) Detwinning device. (b) Temperature dependences of resistivity near the structural/magnetic phase transition measured in compressed, elongated, and free-standing BaFe₂As₂ samples. If a sample is too much compressed or elongated, the transition shifts to higher temperatures.

spin-fluctuations mediated s_{\pm} pairing is the only answer. However, as more and more iron-based superconductors have been discovered, considerable diversity in their superconducting properties has become apparent. There is strong evidence for nodal gap structures in some compounds; nodes may be accidental ones compatible with the s -wave symmetry, but there is also possibility of d -wave pairing. For the pairing mechanism, orbital fluctuations are now seriously considered as possible alternative glue. In this situation, it is necessary to study electronic structures in various iron-based superconductors in detail and systematically to reveal relation between the electronic structures and superconducting properties. (Ba_{1-x}K_x)Fe₂As₂ [5] is a suitable system for such investigations. High-quality single crystals are available for the whole range of composition. Its phase diagram is typical of the iron-based high- T_c : The parent compound undergoes an antiferromagnetic transition and does not exhibit superconductivity. As x is increased, the antiferromagnetism is suppressed and superconductivity emerges. At the optimal doping ($x \sim 0.4$), T_c reaches 38 K, and the superconducting gap is a full gap. At $x = 1$, KFe₂As₂ still has a T_c of 3.4 K but the gap is nodal (see a review [4]). This contrast makes it fascinating to study how the electronic structure evolves with x in (Ba_{1-x}K_x)Fe₂As₂. We here show results of quantum oscillation measurements on the two end members, BaFe₂As₂ [6] and KFe₂As₂ [7].

2. BaFe₂As₂

Single crystals were grown by a self-flux method and were annealed [8, 9]. The residual resistivity ratios were 40–60. Because of a structural phase transition from tetragonal to orthorhombic, if samples are cooled without stress, they become a mixture of twin domains [10, 11]. To avoid that, we used a device shown in Fig. 1(a): A single crystal sample with the length along a tetragonal [110] axis is fixed to the outer frame and mobile plate by epoxy glue. By adjusting the screws, the sample can be compressed or elongated. If compressed (elongated), the length direction becomes the orthorhombic b (a) axis. Appropriate strength of stress needs to be found by trial and error so as to avoid appreciable shift in the transition temperature [Fig. 1(b)].

Figure 2 shows magnetoresistance and its oscillatory part for $B \parallel c$ in the compressed sample. SdH oscillations are already visible at $B = 5$ T, indicating high-quality of the sample. Figure 3(a) shows the field-angle dependence of the SdH frequencies. Three fundamental frequencies α , γ and δ are identified. An LSDA band structure calculation indicates that the Fermi surface is composed of relatively large hole and electron pockets and lines of tiny pockets. After

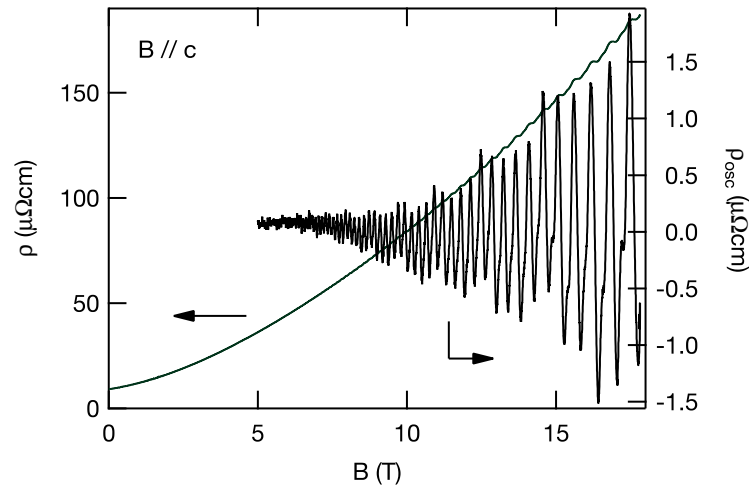


Figure 2. Magnetoresistance and its oscillatory part for $B \parallel c$ in a compressed BaFe_2As_2 sample.

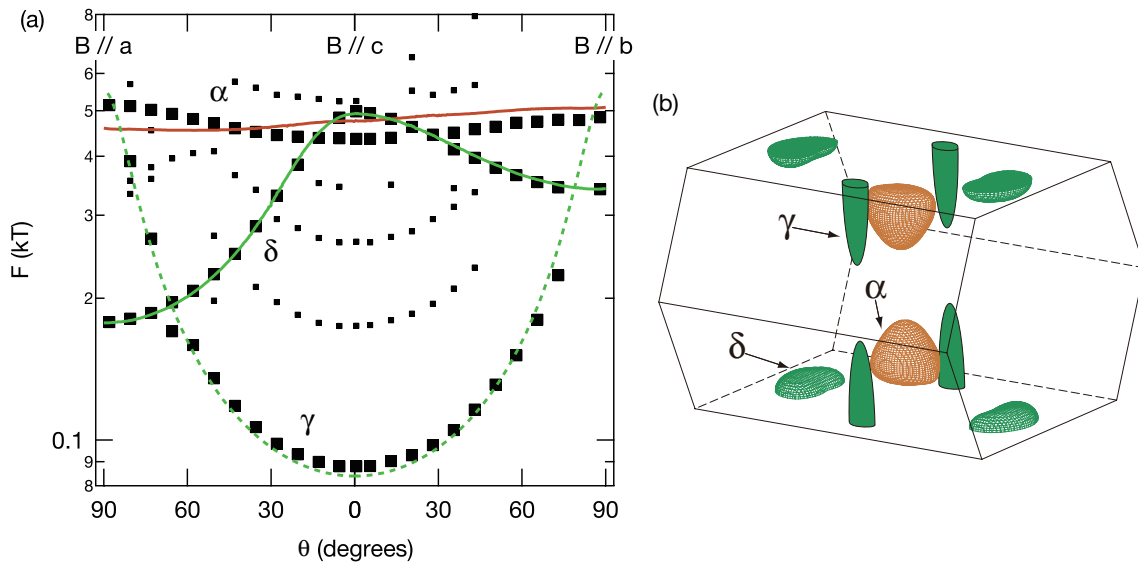


Figure 3. (a) Angle dependence of SdH frequencies in BaFe_2As_2 . The filled squares indicate fundamental frequencies, while the small dots indicate harmonic and combination frequencies. The solid curves indicate calculated frequencies for α and δ with band energy adjustments. The dashed line for γ is a fit to the angle dependence expected for an ellipsoidal Fermi surface pocket. (b) Determined Fermi surface composed of α hole and γ and δ electron pockets.

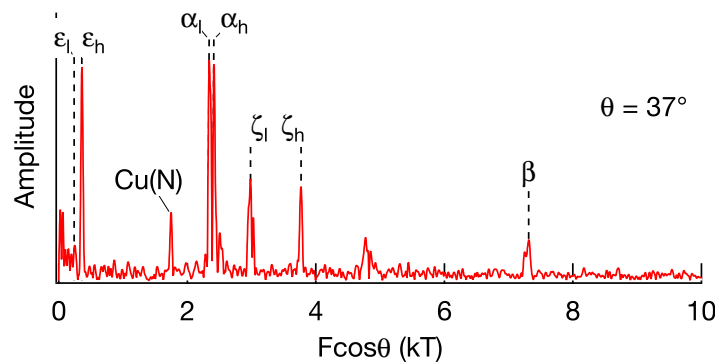


Figure 4. Fourier transform of dHvA oscillations in KFe_2As_2 .

appropriate band energy adjustments, the α and δ frequencies are accounted for by the hole and electron pockets, respectively. The effective masses associated with these frequencies are only moderately enhanced over the band masses by a factor of 2–3. The angle dependence of γ indicates that it arises from an ellipsoidal pocket. Although the tiny pockets predicted by the original calculation disappear after the energy adjustments, it is reasonable to place the γ pocket somewhere along the original tiny-pockets lines. Taking account of laser ARPES data [12] we identify it as an electron pocket and place it in the Brillouin zone as shown in Fig. 3(b). It is to be noted here that, although the observed Fermi surface can be accounted for by the LSDA calculation reasonably well, it overestimates the antiferromagnetic moment ($1.6 \mu_B$ compared to the experimental value of $0.87 \mu_B$ [13]).

Carrier numbers estimated from the determined Fermi surface are 0.0235 holes and 0.0246 electrons per primitive cell, satisfying the carrier compensation within experimental accuracy. The Sommerfeld coefficient estimated from the determined Fermi surface and effective masses is $5.0 \text{ mJ/K}^2\text{mol}$, which agrees well with the measured value of $5.1 \text{ mJ/K}^2\text{mol}$ [14]. These indicate that the determined Fermi surface is complete and that there is no other pocket with a comparable volume to the observed ones.

The determined Fermi surface is composed of three-dimensional closed pockets [Fig. 3(b)]. This is in contrast to k_z -resolved ARPES data [15, 16, 17], which show quasi-two-dimensional Fermi surface cylinders even in the antiferromagnetic phase. This discrepancy arises from the limited k_z resolution of ARPES. The determined Fermi surface is not very anisotropic in the ab plane. Analyses of the measured effective masses and Dingle temperatures also suggest fairly isotropic ab plane electrical transport [6]. This is consistent with recent resistivity and optical measurements on detwinned crystals [8, 9].

3. KFe_2As_2

Single crystals grown by a self-flux method show residual resistivity ratios of ~ 600 [18]. de Haas-van Alphen oscillations were detected using the field-modulation technique. Figure 4 shows an example of a Fourier transform of observed dHvA oscillations. The field angle θ is measured from the c axis. We identify four fundamental frequencies ϵ , α , ζ , and β . Their angle dependences indicate that they correspond to quasi-two-dimensional Fermi surface cylinders with different degrees of warping. The former three appear in pairs as expected, but the other pair to β is unclear at present. An LDA band structure calculation predicts three hole cylinders at the zone center, small hole cylinders near the zone boundary, and a tiny hole pocket. Using this as a guide, we place the α , ζ , and β cylinders at the center and the ϵ cylinders near the corner

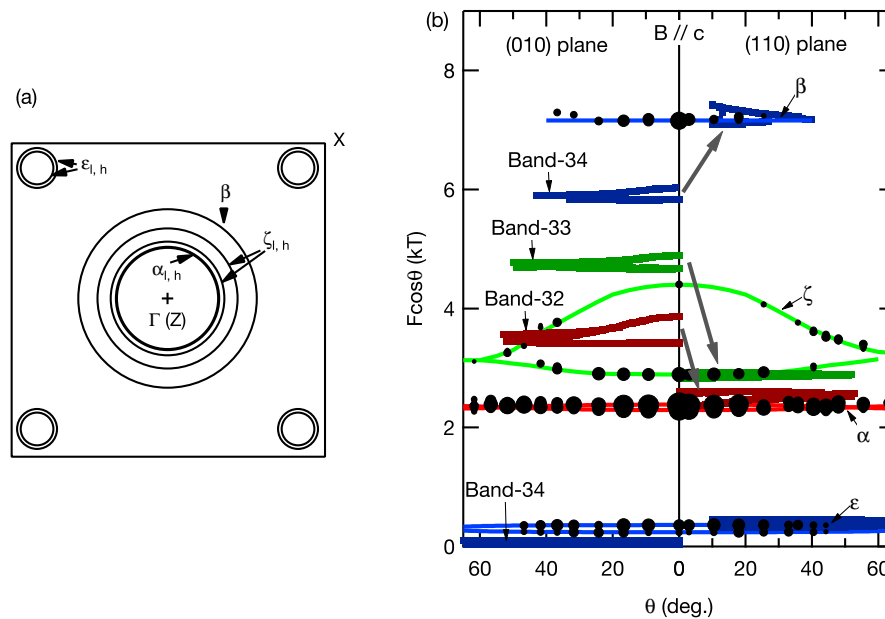


Figure 5. (a) Observed orbits projected along the c axis. In-plane anisotropy is neglected. (b) Comparison between the experimental (ϵ , α , ζ , and β) and calculated frequencies (Bands-32, 33, and 34). The right panel shows results of band-energy adjustments.

[Fig. 5(a)]. The Fermi surface volume is estimated to be 51.4% or 50.5% from the frequencies observed at $\theta = 0$ or 37 degrees, respectively. These estimates are in satisfactory agreement with the stoichiometry (50%). The Sommerfeld coefficient is estimated to be 94 or 82 mJ/K²mol from the effective masses determined at $\theta = 0$ or 37 degrees, respectively. These values are consistent with the measured value of 93 mJ/K²mol [19]. These indicate that the determined Fermi surface is complete. It is to be emphasized that the estimated Sommerfeld coefficients are 8–9 times larger than the band value, indicating strong electronic correlations in KFe₂As₂.

Quantitative agreement between the measured and calculated frequencies is poor [Fig. 5(b)]. In contrast to the case of BaFe₂As₂, band energy adjustments do not work as shown on the right side of the figure: the three dimensional shape of the ζ cylinder can not be reproduced, for example. Band structure calculations predict that the xz and yz bands are above the xy band at the Γ point. The former two are degenerate at Γ without spin-orbit coupling. On the other hand, experimentally, we have two similarly-sized cylinders, α and ζ , and a large cylinder, β . To explain this, it seems necessary to invert the calculated level scheme. Namely, the xz and yz bands are lower than the xy band and produce the two similarly-sized cylinders, α and ζ . Recent AREPS measurements have confirmed this conjecture [20].

4. Summary

We have determined the Fermi surface in BaFe₂As₂ and KFe₂As₂ based on quantum oscillation measurements. The Fermi surface in BaFe₂As₂ is composed of small three-dimensional electron and hole pockets and is explained by an LSDA calculation reasonably well despite the overestimated antiferromagnetic moment. The mass enhancements are only moderate, approximately 2–3. The Fermi surface in KFe₂As₂ is composed of quasi-two-dimensional hole cylinders. The Fermi surface calculated with LDA is not very consistent with the observed one.

It seems that LDA calculations yield wrong crystal field splitting of the Fe 3d states. Effective masses are heavy, up to $20m_e$, indicating strong electronic correlations in this compound.

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