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Quantum oscillations in antiferromagnetic CaFe$_2$As$_2$ on the brink of superconductivity

N Harrison, R D McDonald, C H Mielke, E D Bauer, F Ronning and J D Thompson

Los Alamos National Laboratory, MS-E536, Los Alamos, NM 87545, USA

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Abstract

We report quantum oscillation measurements on CaFe$_2$As$_2$ under strong magnetic fields—recently reported to become superconducting under pressures of as little as a kilobar. The largest observed carrier pocket occupies less than 0.05% of the paramagnetic Brillouin zone volume—consistent with Fermi surface reconstruction caused by antiferromagnetism. On comparing several alkaline earth AFe$_2$As$_2$ antiferromagnets (with A = Ca, Sr and Ba), the dependences of the Fermi surface cross-sectional area $F_\alpha$ and the effective mass $m_\alpha^*$ of the primary observed pocket on the antiferromagnetic/structural transition temperature $T_s$ are both found to be consistent with the case for quasiparticles in a conventional spin-density wave model. These findings suggest that the recently proposed strain-enhanced superconductivity in these materials occurs within a broadly conventional spin-density wave phase.

(Some figures in this article are in colour only in the electronic version)
In the present paper, we determine the Fermi surface of the most axially compressed member of this series, CaFe$_2$As$_2$, by utilizing the contactless conductivity technique that has been successfully applied to SrFe$_2$As$_2$ [3] and BaFe$_2$As$_2$ [12], as well as the cuprate superconductors in strong magnetic fields [2, 26]. A sample of CaFe$_2$As$_2$ of dimensions $\approx 1 \times 1.5 \times 0.2$ mm$^3$ is cut from a larger Sn flux-grown crystal [15] and mounted with its tetragonal symmetry brought about by magnetic ordering, as was recently established in SrFe$_2$As$_2$ [3].

A significant finding in the current study is that the effective masses in CaFe$_2$As$_2$ exhibit only a small relative change compared to SrFe$_2$As$_2$ [3] or BaFe$_2$As$_2$ [12], in spite of CaFe$_2$As$_2$ being much more easily coerced towards superconductivity in figure 1. Figure 4 shows a fit of the Lifshitz–Kosevich theoretical [27] form $A = A_0 X / \sinh X$ (where $X = 2\pi^2 m_k \hbar \bar{B} / \hbar e B$) to the temperature-dependent quantum oscillation amplitude in CaFe$_2$As$_2$. The fitted values of the effective mass are compared to those of similar pockets in SrFe$_2$As$_2$ and BaFe$_2$As$_2$ in tables 1 and 5.

Figure 2 shows results of quantum oscillation experiments performed on CaFe$_2$As$_2$, after subtracting a linear background and performing Fourier transformation in $1/B$. When $\mathbf{H} || c$, several oscillations are discernible corresponding to a frequency $F_a = 395 \pm 10$ T, with another feature at $F_\beta = 95 \pm 20$ T becoming more clearly discernible at higher $T$ when the amplitude of $F_\alpha$ is thermally suppressed (here we use the frequency labeling scheme adopted for SrFe$_2$As$_2$ in [3]). A third frequency $F_\gamma \sim 200$ T, if present, is strongly reduced in amplitude compared to that in SrFe$_2$As$_2$. On further performing a rotation study (presented in figure 3), both $F_\alpha$ and $F_\beta$ are found to be consistent with small ellipsoidal pockets in a similar fashion to SrFe$_2$As$_2$ [3], with the larger occupying less than 0.05% of the paramagnetic Brillouin zone volume. These findings support a scenario in which the Fermi surface is reconstructed as a consequence of the broken translational symmetry brought about by magnetic ordering, as was recently established in SrFe$_2$As$_2$ [3].

The absence of a divergence in the effective mass on reducing $c$ in figure 5(a), or reducing the antiferromagnetic/structural transition temperature $T_c$ in figure 5(b), is in marked contrast to recent observations made in heavy fermion antiferromagnets [28] and cuprate superconductors [29], where antiferromagnetic correlations strongly suppress the ability of the f- or d-electrons to participate in electrical conduction. Given that the Fermi velocities and/or effective masses in FeAs-layered systems are renormalized by only a factor of two

1 Note that the $\alpha$ and $\beta$ are mistakenly interchanged in figure 2(b) of [3] and in the discussion section of [3] relating the pockets to the bandstructure calculations.
the structural transition temperature $T_a$ comparatively weak enhancement of rotational spin-density wave, antiferromagnetic correlations lead to development of a Mott insulating state. In the case of a conventional antiferromagnetic phase. figure 5(b) can be understood by considering a simplified spin-on account of its larger surface area. The observed trends in which also contributes most to the electronic heat capacity the size of the spin-density wave gap $\Delta_1$.

Table 1. Values of the Fermi surface frequencies $F_i$ and effective masses $m_\epsilon$ for $H||c$, taken from this work and [3, 12]. Also shown, is the structural transition temperature $T_s$ into the orthorhombic, antiferromagnetic phase.

| $A$ | $T_s$ (K) | $F_{\alpha}$ (T) | $m_\epsilon^*$ (meV) | $F_{\gamma}$ (T) | $m_\epsilon^*$ (meV) |
|-----|-----------|-----------------|---------------------|-----------------|---------------------|
| Ca  | 170       | 395             | 1.8 ± 0.2           | 95              | 1.0 ± 0.2           |
| Sr  | 200       | 370             | 2.0 ± 0.1           | 70              | —                   |
| Ba  | 138       | 430             | 1.2 ± 0.3           | 95              | 0.9 ± 0.1           |

compared to bandstructure estimates [3, 12], the physics of $\text{AFe}_2\text{As}_2$ appears to be much closer to that of a conventional spin-density wave [30], with the on-site Coulomb repulsion between the d-electrons causing some degree of departure from the BCS relation, $\Delta = 1.76k_B T_s$ [32]2. On the other hand, quantitative consistency with the spin-density wave model is found on comparing the reduction $\delta E$ in the filling $E$ of the pocket with the increase $\delta \Delta$ in $\Delta$ on going from $A = \text{Ca}$ to $\text{Sr}$ (which have the smallest percentage error bars in $m_\epsilon^*$). If we assume a parabolic dispersion whereby $E = h e F_{\epsilon}/m_\epsilon^*$ and a BCS gap (as a first approximation), we arrive at quantitatively similar incremental changes $\delta E \approx 4 \pm 2$ meV and $\delta \Delta \approx 4.5$ meV (estimated using the BCS relation [32] (see footnote 2)) as expected for the simple spin-density wave model presented in figure 5(c).

Figure 3. Magnetic field orientation-dependence of the $\alpha$ and $\gamma$ quantum oscillation frequencies determined by Fourier transformation, where $\theta = 0$ corresponds to $H||c$ and $\theta = 90^\circ$ corresponds to $H||a$ at room temperature. The sample and coil are rotated in situ.

Figure 4. Temperature-dependence of $A/T$ for $H||c$ together with fits to the Lifshitz–Kosevich theory as described in the text. Fitted values of the effective mass are shown, where $m_\epsilon$ is the free electron mass.

Possible evidence for conventional spin-density wave ordering in $\text{AFe}_2\text{As}_2$ is provided in figure 5(b) by the reduction in the size of the pocket $F_{\alpha}$ and weak enhancement of $m_\epsilon^*$ on increasing $T_s$ (which varies non-monotonically with $A$ and $c$). Here, we concentrate on the larger pocket that is observed consistently in all antiferromagnetic $\text{AFe}_2\text{As}_2$ compounds, which also contributes most to the electronic heat capacity on account of its larger surface area. The observed trends in figure 5(b) can be understood by considering a simplified spin-density wave reconstruction model (see figure 5(c)) in which the spin-density wave gap $2\Delta$ increases with the transition temperature $T_s$. If the unreconstructed Fermi surfaces are assumed to be similar throughout [3], the progressive increase in $2\Delta$ incurred on going from Ba to Ca to Sr improves the effectiveness of nesting, resulting in a progressive reduction in the sizes of the pockets and a gradual flattening of the reconstructed band leading to the enhancement in $m_\epsilon^*$.

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We are neglecting the effect of the coupling to the lattice, which likely causes the transition to become of first order near $T_s$. Coupling to the lattice typically increases the size of the gap compared to that obtained from the BCS estimate [32].
Given that superconductivity can occur within orthorhombic phase of AFe$_2$As$_2$ [18, 19], the existence of small pockets of carriers (now identified to be a universal feature of these compounds) are crucial prerequisite for its coexistence with antiferromagnetism, as recently found in CeIn$_3$ [36]. The present experiments reveal the magnitude of the superconducting transition temperature to be more closely correlated with the interlayer spacing (i.e. [16] and figure 1) than it is with the degree of effective mass enhancement. Such a correlation between transition temperature and layer spacing is found in other families of layered superconductors [33–35]. The primary effect of the close proximity to the ‘collapsed tetragonal phase’ in CaFe$_2$As$_2$ therefore appears to be in instigating superconductivity by precipitating strain under non-ideal hydrostatic conditions. If the orthorhombic distortion is a necessary prerequisite for long range antiferromagnetism in this series of compounds, by suppressing the geometric frustration between neighboring Fe moments that would otherwise occur [37], then its vulnerability to an in-plane strain component could be an important factor in tuning superconductivity.

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Figure 5. Plots of the largest quantum oscillation frequency $F_0$ for $H||c$ (left-hand axis) and effective mass $m^*_c$ (right-hand-axis) versus the bilayer spacing (a) and the structural transition temperature $T_s$ (b) from the high temperature tetragonal phase into the low temperature orthorhombic, antiferromagnetic phase. (c) A one-dimensional schematic showing how the size of the pocket ($\alpha F$) is inversely related to the size of the spin-density wave gap ($\Delta_1$). In the simplest model, an increase $\Delta$ in $\Delta$ leads to an equal and opposite reduction $\Delta E$ in the filling $E$ of the pocket (for a constant Fermi energy $\epsilon_F$).