Fermi Surface and Mass Renormalization in the Iron-Based Superconductor YFe₂Ge₂

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Interaction-enhanced carrier masses are central to the phenomenology of iron-based superconductors. Quantum oscillation measurements in the new unconventional superconductor YFe₂Ge₂ resolve all four Fermi surface pockets expected from band structure calculations, which predict an electron pocket in the Brillouin zone corner and three hole pockets enveloping the centers of the top and bottom of the Brillouin zone. Carrier masses reach up to 20 times the bare electron mass and are among the highest ever observed in any iron-based material, accounting for the enhanced heat capacity Sommerfeld coefficient \(\gamma\) \(\approx 100\) mJ/mol K². Mass renormalization is uniform across reciprocal space, suggesting predominantly local correlations, as in the Hund’s metal scenario.

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Superconductivity in iron pnictides and chalcogenides emerges from a rich interplay of magnetic, structural, and nematic instabilities as well as the associated quantum critical phenomena [1–4]. This diverse phenomenology includes strong quasiparticle mass renormalization, which in the extreme case of the alkali metal iron arsenides (K/Rb/Cs)Fe₂As₂ produces heat capacity Sommerfeld coefficients \(\sim 100–200\) mJ/mol K². Although the Sommerfeld coefficient approaches a level usually associated with rare-earth-based heavy fermion materials, it occurs without their partially filled \(f\) states. Resolving this conundrum would present key insights into the strong correlation physics of iron-based superconductivity. One approach could consider primarily the effects of long-ranged magnetic fluctuations that emerge near the threshold of magnetic order, at a so-called quantum critical point, and which may also underlie the pairing mechanism. An alternative explanation for the strong mass renormalization might instead focus on a more local mechanism like the Kondo effect [5]. In Fe-based materials, this may be amplified by Hund’s rule coupling, which locks the iron \(d\) electrons into a high spin state and thereby boosts electronic correlations [6–11]. Because magnetic fluctuations peak at specific critical wave vectors, they would be expected to enhance quasiparticle masses on certain “hot” regions on the Fermi surface, which are connected by the critical wave vectors, producing nonuniform mass renormalization. The strong correlations induced by on-site interactions as in the Kondo lattice-Hund’s metal picture, by contrast, are local and therefore less selective in reciprocal space. Quantum oscillation measurements present an opportunity to distinguish the two scenarios experimentally.

We examine the origin of mass renormalization in iron-based intermetallics in the iron germanide superconductor YFe₂Ge₂ [12–14]. It shares key aspects with the alkali metal iron arsenides (K/Rb/Cs)Fe₂As₂, such as (i) bad metal behavior at high temperature \(T\), with resistivities \(\rho\) of several hundred \(\mu\)Ωcm, (ii) low transition temperatures \(T_c\) of order a few kelvin, (iii) strongly enhanced heat capacity Sommerfeld coefficient \(\gamma_\text{n} \approx 100\) mJ/mol K², (iv) reduced heat capacity jump at \(T_c\) of order 0.4\(T_c\)\(\gamma_\text{n}\), and (v) residual extrapolated \(C/T\) at low \(T\) of order 0.4\(\gamma_\text{n}\). On the other hand, it lacks the pnictogen or chalcogen constituents of other Fe-based superconductors, and because the Ge layers in YFe₂Ge₂ are covalently bonded along \(c\), it forms a more compressed, collapsed-tetragonal structure. Density functional theory (DFT) calculations [17,18] suggest that this leads to a much more strongly warped Fermi surface (FS) geometry than the cylindrical FS sheets found in other iron-based superconductors (Fig. 1). Key input for any comprehensive theoretical description of iron-based superconductors derives from the experimental determination of the electronic structure by photoemission spectroscopy or quantum oscillation measurements (e.g., [19–21]). In YFe₂Ge₂, an early ARPES study [22] investigated the electronic structure, and core-level spectroscopy indicated a large fluctuating Fe moment \(\approx 1\) \(\mu_\text{B}\) [23], which is consistent with neutron scattering and NMR studies [24,25]. The precise determination of Fermi surface and carrier mass by quantum oscillation techniques, however, has in YFe₂Ge₂ so far been held back by the lack of single crystals with the required level of purity [26]. Here, we present a detailed study of Fermi surface structure and carrier mass by observing de Haas–van Alphen oscillations in a new generation of high-quality crystals of YFe₂Ge₂. We find...
that the Fermi surface consists of three hole pockets and one electron pocket, with shapes broadly in line with numerical calculations [17,18]. Carrier masses are enhanced roughly fivefold over DFT values. The mass renormalization varies only weakly between Fermi surface pockets and within each Fermi surface pocket, suggesting that on site interactions provide the primary mechanism for the profound mass enhancement and boost to the electronic heat capacity recorded in YFe$_2$Ge$_2$.

High-purity crystals of YFe$_2$Ge$_2$ were grown by a liquid transport technique [27,28] and characterized by electrical transport, magnetic, and thermodynamic measurements. They display sharp superconducting transition anomalies (Fig. 2), and their residual resistivities are $\simeq 0.3 \mu \Omega \text{cm}$, corresponding to residual resistance ratios RRR $= \rho(300 \text{ K})/\rho_0 \simeq 700$. Angle-dependent de Haas–van Alphen measurements at fields of up to 18 T were performed using a mutual inductance technique with modulation field amplitude $\simeq 0.2 \text{ mT}$ at a frequency of $\sim 29 \text{ Hz}$ on two samples ($S1, S2$) in a superconducting cryomagnet-dilution refrigerator system at the Cavendish Laboratory, and using piezo-resistive torque magnetometry on a third sample ($S3$) at fields of up to 38 T in a resistive electromagnet-dilution refrigerator system at HFML Nijmegen. Sample $S1$ ($S2$) was mounted with the crystallographic $\hat{c}$ ($\hat{a}$) direction aligned with the axis of the respective pickup coil, and $S3$ was mounted with $\hat{c}$ perpendicular to the surface of the cantilever. Quantum oscillation data were extracted by subtracting a low-order polynomial background from the raw data, and oscillations periodic in $1/B$ were identified from peaks in the power spectrum. The peak frequencies $F$ relate to extremal cross-sectional areas $A_k$ of the Fermi surface via the Onsager relation $A_k = (2\pi e/\hbar) F$. The dependence of the signal amplitude $\tilde{y}$ on temperature $T$ at a fixed magnetic field $B$ provides the effective carrier mass $m^*$ via the Lifshitz-Kosevich expression $\tilde{y} = \alpha T \sinh[14.639 \text{TK}^{-1}(T/B)(m^*/m_e)]^{-1}$, where $\alpha$ is a temperature-independent factor and $m_e$ is the bare electron mass. The field dependence of the signal envelope provides an estimate of the electronic mean free path [29,30]. The electronic structure was calculated using the generalized gradient approximation [32] in WIEN2k [33] with 100 000 $k$ points in the BZ (6768 $k$ points in the irreducible BZ) and $R_{\text{Kmax}} = 7$, using the experimentally determined crystal structure at 100 K with $a = 3.95917(3) \text{ Å}$, $c = 10.39754(13) \text{ Å}$, and the fractional vertical Ge position $z = 0.378331(7)$ [27]. Extremal orbits, band masses, and pocket-resolved contributions to the density of states (DOS) were extracted using SKEAF [34], and Fermi surfaces were plotted in FermiSurfer [35].

The calculated Fermi surface (Fig. 1) consists of three roughly ellipsoidal hole pockets ($A–C$) nested around $Z$ at the top and bottom of the Brillouin zone and a roughly cylindrical electron pocket ($D$) around $X$ at the zone corner, with a duckbill-shaped outgrowth toward the center of the BZ at $\Gamma$. Moreover, our DFT calculations produce an unrenormalized Sommerfeld coefficient $\gamma_0 \simeq 16.7 \text{ mJ/mol K}^2$, a factor of six less than the experimentally observed value but significantly larger than values previously obtained in calculations based on numerically relaxed $z$ parameters [17,18].

The quantum oscillation (QO) signal observed in $S1$ (Fig. 3) displays three fundamental frequencies, $\alpha$, $\beta$, and $\gamma$. The same set of frequencies is present in sample $S2$, as shown in the Supplemental Material [30]. The results of a rotation study from $H||\hat{c}$ ($\theta = 0^\circ$) to $H||\hat{a}$ ($\theta = 90^\circ$) are summarized in Figs. 3(b) and 4(a). The three fundamental
frequencies $\alpha$, $\beta$, and $\gamma$ were tracked over most of the angular range. By comparing to the DFT results, they can be assigned to extremal orbits on the three hole pockets. The highest frequency, $\gamma$, is unambiguously associated with the largest hole pocket, C. The next highest frequency, $\beta$, matches predictions for the second hole sheet (B), with good quantitative agreement near $H \parallel \hat{a}$. The third frequency, $\alpha$, depends weakly on tilt angle, as expected for the smallest, nearly ellipsoidal hole pocket (A), but at roughly half the predicted frequency. The effective masses extracted from Lifshitz-Kosevich fits [Fig. 4(b)] are high, with $m^* \gg m_e$ on the $\gamma$ and $\beta$ sheets. These values markedly exceed the highest masses measured in the BaFe$_2$(As/P)$_2$ series [36] and are as high as those observed in (K/Rb/Cs)Fe$_2$As$_2$ [21,37,38]. Whereas effective masses for $\alpha$ and $\beta$ show little angle dependence, that of $\gamma$ rises sharply near $H \parallel \hat{c}$ [Fig. 4(c)]. A uniform renormalization of DFT band masses by $\sim 5$ produces good agreement with experimental values on all three hole pockets.

The electron pocket D could not be resolved at fields of up to 18 T. Its $ab\ initio$ volume corresponds to 0.142 electrons/formula unit (f.u.), and its expected dominant QO frequency is $\sim 0.9\, \text{Kt}$. This estimate can be refined by using the QO data on the three observed hole pockets in combination with charge neutrality, which imposes an overall hole count of 1/formula unit. Assuming that the $ab\ initio$ geometry and associated hole count $n_0$ of each hole pocket can be scaled to make them consistent with the measured QO frequencies, we estimate the actual hole count for each pocket as $n = n_0 q_a q_c (q_\alpha q_\beta).$ Here, $F_\alpha \propto q_a q_c$, the QO frequency for field along $\hat{a}$, is given by the extent $q_a$ of the pocket along $\hat{a}$ and $\hat{c}$, and $F_c \propto q_\alpha^2$, the QO frequency for field along $\hat{c}$, is determined by the extent of the pocket $\perp \hat{c}$, or along $\hat{c}$. $F_{\alpha\beta}$ and $F_{\alpha\alpha}$ are the $ab\ initio$ frequencies for the same pocket, and $q_{\alpha\beta}, q_{\alpha\alpha}$ the corresponding dimensions. In the case of pocket C, the $\gamma$ frequency was extrapolated to $\theta = 0^\circ$, and the resulting hole count checked for consistency against a second calculation in which $F_c$ and $F_{\alpha\alpha}$ values were taken at $\theta = 7.5^\circ$, the lowest angle at which the $\gamma$ oscillation could be observed. The upper half of Table 1 summarizes the results of these calculations, which suggest that the total hole count per f.u. arising from A, B and C is about 1.07. This implies that the volume of the electron pocket is smaller than expected from $ab\ initio$ calculations. It corresponds to about $0.07 e^2$ per f.u., which leads to a dominant QO frequency of order 500 T.

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Our initial observation exploited the phenomenon of torque interaction \cite{29-31}, which mixes quantum oscillation frequencies via the nonlinear cantilever response at high magnetic fields. The $\gamma$ peak develops several side lobes, some of which can be indexed as $\gamma \pm n\delta$ with $\delta \sim 450$ T, depending on the field angle. At some angles, a distinct low-frequency peak at the corresponding frequency $\delta$ can also be resolved directly (Fig. 5). Further work will be required to narrow down the current mass estimate $m^* \approx 8.8 \pm 1.8 \, m_e$ obtained at a $60^\circ$ tilt angle from the temperature dependence of the directly observed peak intensity [Fig. 5(c)], and to track the mass as a function of angle. Because the angle dependence [Fig. 5(b)] is consistent with a small cylindrical pocket, we interpret $\delta$ as a signature of the elusive electron pocket $D$. The DOS contribution of the electron pocket is highly sensitive to details such as band filling and the structural $z$ parameter. This sensitivity is caused by a flat region in the dispersion along the BZ diagonal, which produces a duckbill outgrowth toward the center of the BZ. As this feature is quasi-1D (little dispersion within the symmetry plane of the BZ), its emergence produces a van Hove-like singularity in the DOS [Fig. 5(d)]. YFe$_2$Ge$_2$ appears to be situated close to the cusp of this anomaly, similar to the situation of the $\gamma$ sheet in Sr$_2$RuO$_4$ (e.g., Ref. \cite{39}).

The lower part of Table I tracks the contributions of the different Fermi surface pockets to the overall DOS. When the bands are shifted to bring the DFT pocket volumes into line with QO data, the resulting “QO-corrected” DOS is dominated by the electron pocket, which contributes nearly half of the total DOS. Applying a uniform mass renormalization of 4.7 across all pockets $A$–$D$, as suggested by the high-resolution measurements for pockets $A$–$C$ (Fig. 4) would account for a normal-state Sommerfeld ratio $C/T \approx 76 \, \text{mJ/mol K}^2$ at $\approx 18$ T, where QO masses are extracted. The shortfall of about 20% compared to $C/T$ at zero field could be attributed in part to the experimentally observed reduction in $C/T$ in high magnetic field [inset Fig. 4(a)], as magnetic fluctuations are increasingly suppressed and in part to uncertainty in ascertaining the precise DOS contribution associated with the electron pocket, as mentioned above.

Our findings establish the electronic structure of YFe$_2$Ge$_2$ as consistent with expectations from \textit{ab initio} calculations, but with strong, uniform mass renormalization on all Fermi surface sheets. This suggests an underpinning, robust mass enhancement caused by on site correlations, as in the Kondo lattice-Hund’s metal scenario \cite{5-11}. Further
to this local mechanism, long-ranged magnetic correlations observed in neutron scattering [24] and consistent with the magnetic order observed in the antiferromagnetic iso-electronic sister compound LuFe$_2$Ge$_2$ [40] may underlie the superconducting pairing interaction as well as the non-Fermi liquid $T^{1.5}$ resistivity power law. The combination of a robust mass renormalization and a fine-tuned pairing interaction is strongly reminiscent of rare-earth-based Kondo lattice heavy fermion superconductors [41] and may apply more generally in strongly correlated transition metal compounds.

All data needed to evaluate the conclusions in the paper are present in the paper, the Supplementary Materials, and the Data Repository at the University of Cambridge and can be downloaded from [42].

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