Heavy holes as a precursor to superconductivity in antiferromagnetic CeIn₃

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Edited by T. Maurice Rice, Swiss Federal Institute of Technology, Zurich, Switzerland, and approved March 10, 2009 (received for review November 21, 2008)

Numerous phenomenological parallels have been drawn between *f*- and *d*-electron systems in an attempt to understand their display of unconventional superconductivity. The microscopics of how electrons evolve from participation in large moment antiferromagnetism to superconductivity in these systems, however, remains a mystery. Knowing the origin of Cooper paired electrons in momentum space is a crucial prerequisite for understanding the pairing mechanism. Of special interest are pressure-induced superconductors CeIn₃ and CeRhIn₅ in which disparate magnetic and superconducting orders apparently coexist—arising from within the same *f*-electron degrees of freedom. Here, we present ambient pressure quantum oscillation measurements on CeIn₃ that crucially identify the electronic structure—potentially similar to high-temperature superconductors. Heavy hole pockets of *f*-character are revealed in CeIn₃, undergoing an unexpected effective mass divergence well before the antiferromagnetic critical field. We thus uncover the softening of a branch of quasiparticle excitations located away from the traditional spin fluctuation-dominated antiferromagnetic quantum critical point. The observed Fermi surface of dispersive *f*-electrons in CeIn₃ could potentially explain the emergence of Cooper pairs from within a strong moment antiferromagnet.

Fermi surface | heavy fermion | quantum critical point | Lifshitz transition

With a Néel ordering temperature of $T_N \approx 10$ K (1, 2), CeIn₃ belongs to a family of Ce-based antiferromagnets that become superconducting under applied pressure. At first glance, CeIn₃ at ambient pressure appears to exhibit all of the hallmarks of a localized antiferromagnet. As our magnetic oscillation measurements (Fig. 1) confirm, the Fermi surface of CeIn₃ is very similar to that of LuIn₃ and LaIn₃ (3) (in which the *f*-electron shells are filled and empty respectively); the measured *d*- and *k*-orbits closely correspond in magnetic field-orientation dependence and volume to identical sheets measured (and calculated) for these nonmagnetic compounds. Furthermore, the weak mass enhancement of the observed orbits,[†] and large staggered moment between 70% and 90% of the local Ce moment value (a Γ_7 doublet in CeIn₃) (4, 5) point to weak hybridization of the conduction with the *f*-electrons, which minimally participate in the Fermi surface. We are then left with a question as to the electronic origin of superconductivity under pressure in CeIn₃ and not its nonmagnetic analogues.

Indications of the unconventional nature of *f*-electron participation in the Fermi surface of CeIn₃ appear on cooling to dilution refrigerator temperatures $T \ll 500$ mK. The light conduction electron Fermi surface remains largely unchanged [supporting information (SI) Text]. However, we find it to be accompanied by new *r*-orbits with unexpectedly large effective masses $m^* \approx 20m_e$ in magnetic fields between 10 and 20 T, which are absent in the nonmagnetic analogues. The topology of the corresponding sections of Fermi surface is deduced from the magnetic field orientation-dependent frequencies, a few

branches of which were previously observed (5). Our measurements reveal a classic 3-branch form corresponding to spatially separated ellipsoids of revolution situated along the $\langle 111 \rangle$ directions of a cubic system (7). The new section of Fermi surface is thus associated with 8 oblate ellipsoids at $\mathbf{k} = \langle k, k, k \rangle$, where $k = (0.5 \pm 0.1)\pi$ (shown in the *Lower Inset* of Fig. 1*A*), which dominate the thermodynamic mass. In contrast to the conventional picture for the largely uniform Fermi surface mass enhancement by *f-d* hybridization, we argue that selective mass enhancement of ellipsoidal pockets disconnected from the conduction electron Fermi surface (Fig. 1*C*) points to component quasiparticles mainly originating within the *f*-band. These heavy ellipsoidal pockets are primarily responsible for the previously unexplained large residual Sommerfeld coefficient (6) reported within the antiferromagnetic phase of CeIn₃, $\gamma \approx 120$ mJmol⁻¹K⁻²—greatly exceeding that of nonmagnetic LuIn₃ and LaIn₃ (5.3 mJmol⁻¹K⁻²) (8).

To understand the unique ellipsoidal Fermi surface topology, we begin in the noninteracting limit described by the weak coupling picture. Some evidence for a Fermi surface sheet of chiefly itinerant *f*-electrons has been observed in dHvA experiments on the paramagnetic phase of CeIn₃ under pressure (4). A similar Fermi surface topology would be expected from a less-than-half filled band of *f*-character (Fig. 1*A Upper Inset*) within a tight binding approximation, as seen from local density approximation (LDA) band structure calculations in the paramagnetic phase of CeIn₃ (9). The consequence of band folding of such a surface in the antiferromagnetic regime would be to create ellipsoidal hole pockets at $\mathbf{k} = (\pi/2, \pi/2, \pi/2)$ locations in momentum space (Fig. 2*C* and *D*), similar to those observed in CeIn₃. The 2D projection shown in Fig. 2 further reveals the topology of the 3D pockets in CeIn₃ to bear a close resemblance to the recently observed pockets in the 2D Fermi surface of high-temperature superconductors. Within the weak coupling picture, weak Bragg scattering at the antiferromagnetic zone boundaries would imply an occupation number in momentum space $\langle n_{fk} \rangle \leq 2$ within the original Fermi surface, and $\langle n_{fk} \rangle \geq 0$ outside.

A similar Fermi surface topology is obtained for the more physical large Coulomb repulsion limit applicable to the large-moment antiferromagnet CeIn₃, by treatment within a strong

Author contributions: N.H. designed research; S.E.S., N.H., V.F., M.J., T.P.M., E.C.P., and T.E. performed research; S.E.S., N.H., C.D.B., S.A.T., and H.H. analyzed data; and S.E.S., N.H., and C.D.B. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

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[†]Some mass enhancement is observed in concentrated regions along the $\langle 1, 1, 1 \rangle$ direction (5, 6), which we explain in SI Text.

This article contains supporting information online at www.pnas.org/cgi/content/full/0811859106/DCSupplemental.

Fig. 1. Fermi surfaces of CeIn_3 . (A) Measured d-, k-, and r-orbit quantum oscillation frequencies F (obtained by magnetic torque), related to the Fermi surface (FS) cross-sectional area A_k ([SI Text](#)), versus the orientation of H. Electrons undergoing cyclotron motion in real-space circumnavigate the FS in momentum space k. Measured d- and k-band frequencies are similar to those in nonmagnetic LuIn_3 (red lines) calculated by an augmented plane wave method, whereas r-orbit frequencies, found only in CeIn_3 , correspond to simulations (purple lines) of the 8 ellipsoidal pockets depicted in the *Lower Inset*. The *Upper Inset* shows a less-than-half filled f-electron Fermi surface sheet from tight binding calculations in the paramagnetic phase of CeIn_3 (8). (B) Schematic representation of the conduction electron Fermi surface (d-sheet) expected within the strong moment Kondo lattice model. In this picture, any mass enhancement would occur because of conduction-f electron hybridization over the entire conduction electron Fermi surface (see [Figs. S1 and S2](#)). (C) Schematic for unconventional participation of f-electrons in Fermi surface indicated in CeIn_3 . A distinct heavy surface (argued to be of principally f-electron character) appears, causing an enhancement of the conduction electron density of states only in the regions of conduction-f electron band proximity ([SI Text](#)).

coupling picture. In this representation, the less-than-half filled band of chiefly f -character is modeled as a background of antiferromagnetically aligned local moments interspersed with a finite concentration of holes. The interplay between the kinetic energy (t) and electron–electron interaction (approximated by the on-site Coulomb interaction U) of this band can be treated within a single-band Hubbard model. A similar ellipsoidal Fermi surface topology is obtained, whereas the large Coulomb repulsion means that double occupancy is inhibited, and strong scattering by the antiferromagnetic periodic potential results in an occupation number $\langle n_{f\mathbf{k}} \rangle \approx 1$ throughout the Brillouin zone. Holes aggregate about the $(\pm\pi/2, \pm\pi/2, \pm\pi/2)$ points in momentum space (Fig. 2E), which are nested by the antiferromagnetic ordering wave vector $\mathbf{Q} = (\pi, \pi, \pi)$ at half filling (10, 11). The interior of the resulting pockets has a lower occupation number, approaching zero within the mean field approximation. Within this picture, “heaviness” results predominantly from the magnetic distortion accompanying hole motion in an antiferromagnetic “background” (12).

The heavy ellipsoidal pockets in CeIn₃ show further surprises in their magnetic field tuned properties. Although a modification in Fermi surface properties would be natural in the vicinity of the critical field $B_c \approx 61\text{T}$ where antiferromagnetism is suppressed, we find that, astonishingly, the ellipsoidal mass diverges at an intermediate field $B_f \approx 0.7B_c \approx 40\text{ T}$ accompanied by an upturn in frequency. Divergences in the effective masses of the r1₁₁₀ and r2₁₁₀ orbits are observed as the field approaches 40 T (Fig. 2A), well inside the antiferromagnetic phase and unaccompanied by

Fig. 2. Field dependence of the ellipsoidal r -orbits observed in CeIn_3 , evidenced by torque measurements made by using a portable dilution refrigerator in the 45-T hybrid magnet at the National High Magnetic Field Laboratory, Tallahassee (SI Text). (A) Effective masses obtained on fitting the temperature-dependent quantum oscillation amplitude to the Lifshitz-Kosevich theoretical form $\text{Amp}(T) = \text{Amp}_0 X / \sinh X$, where $X = 4\pi^2 k_B m^* T / \hbar e B$ (30) for $33 < T < 500$ mK ($\approx 5\%$ uncertainty in T) (see Fig. S3. A Hanning window is used in Fourier transforming over 5- to 8-T intervals. Symbols denote frequencies plotted in B. Subscripts denote the orientation of H in Fig. 1 (Fig. S4). (C) Weak coupling picture: 2D projection of f -electron surface of topology shown in Fig. 1 (*Upper Inset*) folded at the antiferromagnetic zone boundary to give rise to hole pockets. Within this picture, an f -electron occupancy $\langle n_{\mathbf{k}} \rangle \lesssim 2$ inside the original Fermi surface and $\langle n_{\mathbf{k}} \rangle \geq 0$ outside (indicated by the shading) would be implied because of weak Bragg scattering. (D) Migration of ellipsoids toward the zone center because of Zeeman splitting in the weak coupling picture. (E) Strong coupling picture: Strong scattering leads to an f -electron occupancy of $\langle n_{\mathbf{k}} \rangle \approx 1$ everywhere in the Brillouin zone. Away from half filling, the residual holes aggregate at the nesting wave-vector ends ($\pm \pi/2$, $\pm \pi/2$, $\pm \pi/2$). The occupancy number in the interior of the resulting pockets drops to a lower value approaching zero in the mean field limit. (F) Migration of ellipsoidal pockets because of motion of bands in opposing directions with field in the strong coupling picture.

divergent features in the spin susceptibility (6). We show that these signatures can be interpreted in terms of magnetic field tuned correlation effects within a single band of chiefly f -character.

In the noninteracting picture, a magnetic field applied perpendicular to the Néel axis would result in the 2 overlapping bands moving in opposite directions toward the left and right, causing the ellipsoids to migrate toward the zone center (upper right quadrant of the original Brillouin zone shown in Fig. 2 *D* and *F*). Equivalently, in the single-band Hubbard model, the migration of ellipsoids toward the zone center with magnetic field (Fig. 3) is associated with the transformation of background periodic potential from antiferromagnetic to ferromagnetic. In the ferromagnetic (saturated) limit, the concentration of holes moves in a background of all parallel aligned spins, from which we readily see that the quasiparticle dispersion minimum moves from $\mathbf{k} = (\pi/2, \pi/2, \pi/2)$ at low fields to $\mathbf{k} = (0, 0, 0)$ in the

The contribution of the heavy f -hole pockets at $\mathbf{k} = (\pi/2, \pi/2, \pi/2)$ to the electronic heat capacity γT within the antiferromagnetic phase dwarfs that of the conduction bands. An integral $\gamma = k_B^2/6\hbar f dS/|v_F|$ over the Fermi surface of the f -hole pockets (where $v_F = 2\pi\hbar k_F/h$ is the Fermi velocity) reveals that they alone account for at least 80% of the experimental electronic heat capacity at zero field (8), revealing a concentration of density-of-states at the hole pockets in the antiferromagnetic phase. With the emergence of superconductivity from antiferromagnetism under pressure (1, 2), the Fermi surface of the regular conduction bands remains essentially unchanged (4), indicating the likely formation of Cooper pairs almost exclusively at or near these hole pockets situated at $\mathbf{k} = (\pi/2, \pi/2, \pi/2)$.

The underdoped state of high-temperature superconductors exhibits small pockets at the equivalent $\mathbf{k} = (\pi/2, \pi/2, q)$ locations, as revealed by quantum oscillations (15–18) and photoemission (19). Whereas previous literature and studies have suggested phenomenological commonalities between d - and f -superconductors (20–28), here, we show a potential parallelism at the microscopic level between the electronic structures of the precursor states to superconductivity (12, 19, 20). Doped Mott insulator physics is mimicked in CeIn_3 , resulting in a 3D Fermi surface with topological similarities to the 2D cuprate Fermi surface. The effective charge reservoir provided by the conduction bands in CeIn_3 could play a similar role to the magnetically

inert oxygen chains in the YBCO cuprates. The emergence of a new magnetic field tuned mass divergence of the hole pockets in CeIn_3 points to a softening of a distinct class of quasiparticles associated with these pockets of f -character. The separation of critical fields in CeIn_3 where the 2 distinct modes (spin and potentially charge) soften appears to contrast with recent experimental findings in CeRhIn_5 under pressure, where the 2 are reported to coincide (29). Further experiments on the hole pockets in CeIn_3 by tuning along the pressure axis will be of interest in exploring where the softening of the new mode occurs in relation to superconductivity and the suppression of antiferromagnetism. CeIn_3 provides us with a structurally simple model system to explore the relationship between the evolution of the hole pocket regions in momentum space under pressure (or doping), and the concomitant appearance of superconductivity.

ACKNOWLEDGMENTS. We thank G. G. Lonzarich and P. B. Littlewood for valuable inputs, and R. D. McDonald and A. C. H. Cheung for technical assistance with simulations. This work was performed under the auspices of the National Science Foundation, the U.S. Department of Energy, and the State of Florida. Support was provided by a Grant-in-Aid for Scientific Research on Priority Areas, "High Field Spin Science in 100T" and the Ministry of Education, Culture, Sports, Science and Technology. S.E.S. acknowledges discussions arising at the Trieste Miniworkshop on Strong Correlations 2008 and support from the Mustard Seed Foundation, the Institute for Complex Adaptive Matter, and Trinity College (Cambridge University, Cambridge, U.K.).

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