Electronic structure of Ce$_2$RhIn$_8$ 2D heavy Fermion system studied by angle resolved photoemission spectroscopy

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We use angle-resolved photoemission spectroscopy to study heavy fermion superconductor Ce$_2$RhIn$_8$. The Fermi surface is rather complicated and consists of several hole and electron pockets. We do not observe $k_z$ dispersion of Fermi sheets, which is consistent with 2D character of the electronic structure. Comparison of the ARPES data and band structure calculations points to a localized picture of $f$ electrons. Our findings pave the way for understanding the transport and thermodynamical properties of this material.

I. INTRODUCTION

Heavy fermions, first discovered in 1975, are some of the most fascinating materials in condensed matter physics. The name originates from the enhanced effective mass of quasi-particles, which can be two or three orders of magnitude higher than normal metal, while the functional form of resistivity remains the same as Fermi liquid. The behavior of this system is dominated by 4f and 5f electrons and arises due to competition between magnetic and electronic quantum fluctuations, many marvelous phenomena are seen in heavy-fermion superconductors, quantum criticality and many others. Since electrons in heavy fermion materials are an important test-bed for understanding the interplay between magnetic and electronic quantum fluctuations, measurement of the electronic structure is a crucial step for further research.

Ce$_2$RhIn$_8$ with lattice parameters $a =$ 4.665Å, $c =$ 12.244Å and tetragonal crystal structure is a member of Ce$_n$MIn$_{3n+2}$ (M=Co, Rh or Ir, n=1, 2 or $\infty$) family, which are anti-ferromagnetic heavy-fermion materials. Since the structure of Ce$_2$RhIn$_8$ can be viewed as inserting a CeIn$_3$ into CeRhIn$_5$, this matter is expected to share some of the properties of both of these two compounds. The enhanced value of the Sommerfeld coefficient ($\sim 400$ mJ/molCeK$^2$) determined by specific heat measurement is consistent with heavy fermion nature of this material. The resistivity curve follows a $\ln(1/T)$ behavior between 55K and 130K as a result of Kondo screening and single impurity model estimate of Kondo temperature $T_K$ yields a value of 10K. The slope of resistivity changes at $T_N = 2.8K$ and $T_{LN} = 1.65K$ indicating two magnetic transitions. Neutron scattering measurement performed at 1.6K show presence of an anti-ferromagnetic state with ordering vector $Q = (\frac{1}{2}, \frac{1}{2}, 0)$. At this temperature, magnetic moment of Ce is well screened to 0.55$\mu_B$, compared with 2.35$\mu_B$ per Ce at high temperature (200K). The slope of resistivity changes more dramatically at $T_{LN}$ than $T_N$ and $T_{LN}$ is also more sensitive to pressure with a $P_c \sim 0.04$GPa, suggesting that the magnetic structure changes between an incommensurate and a commensurate one at $T_{LN}$. The nature of AFM order seems to fit better with a scenario of local moment ordering rather than SDW ordering. This is because of an additional term would be required to fit the specific heat in order to account for anisotropic gap in SDW state in the case of CeRhIn$_8$. Also, according to conventional models of antiferromagnetic quantum criticality, linear decrease of $T_N$ with pressure points an effective 2D character of spin-fluctuation spectrum. This favors Kondo destruction scenario with local moment ordering.

Ce$_2$RhIn$_8$ becomes superconducting above 1 GPa with maximum $T_c = 2K$ near 2.3GPa. Unlike conventional superconductors, Ce$_2$RhIn$_8$ shows non-Fermi-liquid behavior with $\rho(T) \propto T^{0.95\pm0.05}$ and superconductivity co-exist with anti-ferromagnetism. There are some resorts in published literature indicating that the resistivity of Ce$_2$RhIn$_8$ goes to zero at ambient pressure, which may be related to superstructure in Ce$_2$RhIn$_8$ rather than a sign of the bulk superconductivity.

Due to the layered structure, electronic properties of Ce$_2$RhIn$_8$ are believed to be quasi-2D, which is rarely seen in heavy-fermion superconductors. ARPES data have been previously reported for this compound but only along high symmetry directions. To the best of our knowledge there are no reported measurements of the Fermi surface. To better understand the superconductivity and heavy-fermion phenomenon in this material, we examine the Fermi Surface and detailed band disper-
sion of Ce$_2$RhIn$_8$ using various photon energies.

First-principles band structure calculations were performed using spin-polarized density functional theory (DFT) within generalized-gradient approximation (GGA) with projector-augmented wave (PAW) method by VASP code. The GGA exchange correlation functional parameterized by Perdew, Burke and Ernzerhof (PBE) was used. The semi core p states of Rh, as well as the lower lying d states of In, are treated as valences states, while the 4f electron of Ce are treated either as placed in the core or as valence state for comparison. The kinetic energy cutoff was 400 eV and the Monkhorst-Pack’s scheme was used for Brillouin zone sampling with a k-mesh of 17 x 17 x 7 for the FM state and 12 x 12 x 7 for the AFM state in which case a 2x2x1 supercell was used.

Table I. Optimized lattice parameters, energies and magnetic moment of Ce atom in different magnetic states.

| State  | a(Å) | c(Å) | M$_{Ce}$ ($\mu_B$/Ce atom) | E (eV/atom) |
|--------|------|------|-----------------|-------------|
| AFM    | 6.651| 12.283| 0.69             | -4.029      |
| FM     | 4.709| 12.278| 0.68             | -4.030      |
| Non-Mag| 4.705| 12.280| 0               | -4.027      |
| Expriment | 4.665 | 12.244| 0.55            |             |

The calculated lattice parameters, energies and magnetic moment of Ce atoms in different magnetic states are listed in table together with the experimental results. The lattice parameters obtained from the GGA calculations are in good agreement with experiment, with about 1% overestimation. Calculated magnetic moment of the Ce ions also agrees well with the experimental data. From table we can say that the magnetically ordered states have slightly lower energy than the non-magnetic state, with the energy of FM state being slightly lower by $\sim$1 meV per atom.

### II. METHODS

Single crystals were grown using In flux technique and characterized as described in Ref. 9. The ARPES measurements were performed using ARPES system at Ames Laboratory and beamline 7.0.1 of Advanced Light Source (ALS). Samples were cleaved in situ using Torr seal vacuum epoxy and had mirror-like surfaces. All ARPES data were taken at T=16K, above the AFM transition temperature (2.8K) but close to Kondo temperature (10K). Laboratory-based ARPES system consists of GammaData ultraviolet lamp (21.2eV He I$_x$), custom-designed refocusing optics and a Scienta SES2002 electron analyzer. The UV spot size is around 1mm and the energy resolution was set at 10meV. Beamline 7.0.1 is equipped with Scienta R4000 electron analyzer with energy resolution around 40meV.

Figure 1. Fermi Surface measured with (a) He lamp (21.2eV) at 16K. (b) synchrotron (SRC, 80eV) at 20K. (c) synchrotron (ALS, 94eV) at 17K. (d) synchrotron (ALS, 105eV) at 17K. (e) Fermi surface calculated by DFT with f electron set as localized and itinerant respectively.

III. RESULTS AND DISCUSSION

In Fig. (a)-(d) we plot the Fermi surface image of Ce$_2$RhIn$_8$ obtained by integrating photoelectron intensity within ±5meV of the E$_F$ at the at several photon energies. Data in panel (a) was measured using laboratory He source - photon energy of 21.2eV, (b) at SRC using 80eV photons, data in panels (c)&(d) was measured at ALS using 94eV and 105eV photons. Since the cross section of bands can be quite different for various photon energies and polarizations, by performing measurements over large photon energy range, we can reveal the complete Fermi surface topology. To illustrate the orbital contributions to electronic structure, we calculated location of the Fermi sheets with f electrons treated as localized and itinerant as shown in Fig. (e)&(f) respectively. The magnetic moment for itinerant scenario is artificially set to zero, since the temperature for measured data is higher than $T_N$. The Fermi surface for those two scenarios is nearly the same around M point but quite different.
around Γ point. Close to Γ point, the measured Fermi surface more resembles a localized picture as shown in Fig. 2. The localized picture results in a diamond shape (marked by the arrow) of the FS sheet surrounding Γ point, which fits the experimental data well in contrast to a square shape in itinerant scenario. This is not surprising since the data was measured at 16K. Although the temperature is comparable with $T_N ≈ 10K$ and screening of f electron by conduction electron should exist to some extent, it is still much higher than coherence temperature of 5K and most f electron will remain localized.

![Figure 2. Fermi surface image close to Γ point with overlay of Fermi surface calculation using (a) localized or (b) itinerant scenario.](image)

The shape of the Fermi surface remains constant over studied range of the photon energies, which covers large range of $k_z$ values from $19π/c$ to $25π/c$. This demonstrates a quasi-2D character of the electronic structure of Ce$_2$RhIn$_8$, which is consistent with 2D effective dimensionality of the spin-fluctuation spectrum from phase diagram[^2]. Such 2D character is further illustrated by $k_z$ dispersion shown in Fig. 3 (a)&(c) along Γ-X ($k_y = 0$) and Γ-M ($k_z = k_y$) directions. This was accomplished by changing photon energy form 80eV to 157eV. The band dispersion shown as high photoelectron intensity is almost vertical with little or no observable dispersion. The $k_z$ dispersion calculated within localized scenario, shown in Fig. 3 (b)&(d), also predicts a nearly 2D electronic structure especially for the bands along Γ-M direction.

By performing measurements with different photon energies, we can detect all sheets of FS. We find that it consists of 2 pockets around Γ point, 4 pockets around M point and one pocket located in between Γ and X point. The main difference between measured and calculated Fermi surface is that the band forming the pocket marked by arrow in Fig. 2(a) crosses the Fermi level along Γ-X direction at around 0.35π, which would cut one pocket into two smaller ones. This is more obvious in intensity plot shown in Fig. 4. The α band clearly crosses Fermi level along both Γ-X and Γ-M direction, which forms two electron pockets, one around Γ point and one close to X point, while the calculated result using itinerant scenario places this band below Fermi energy along Γ-X direction. Band α also shows a nearly zero intensity along Γ-M direction at all photon energy at ALS data, which may due to the selection rules for horizontal polarization of photons. Band δ forms a tiny electron pocket centered at Γ point.

Calculations and experiment agree well on the topology of Fermi surface around M point with four pockets $\beta_1 - \beta_4$. Based on intensity plot shown in Fig. 4, we can conclude that all four bands form electron pockets. Although the calculation shows a crossing between $\beta_2$ and $\beta_3$ pockets, there is no strong evidence for this in experimental data. It is however possible that the momentum resolution may not be sufficient to detect such a crossing.

To reveal the nature of the AFM order, one needs to establish the presence of a nesting vector equal to AFM ordering vector $\mathbf{q}=(0.5,0.5,0)$. In Fig. 2(b) the high intensity spot at the corner of bands $\beta_2$ and $\beta_4$ forms a vector slightly larger than ordering vector $\mathbf{q}$, which may decrease due to injection of f electrons below coherence temperature. Since the Fermi surface of Ce$_2$RhIn$_8$ contains multiple pockets, when nesting occurs for one of the pockets, large part of Fermi surface is not affected. This is consistent with specific heat measurement[^3] where the change of Sommerfeld coefficient γ above and below $T_N$ (from 400 to 370 mJ/molCeK$^2$) is much smaller than CeRhIn$_5$ (400 to 56 mJ/molCeK$^2$), indicating that only a small part (~8%) of Fermi surface becomes gapped be-

![Figure 3. $k_z$ dispersion along (a)&(b) Γ - X direction and (c)&(d) Γ - M direction from 80eV to 157eV comparing with local f electron calculation.](image)
IV. CONCLUSIONS

We used angle-resolved photoemission spectroscopy to measure the electronic properties of Ce$_2$RhIn$_8$. The lack of significant $k_z$ dispersion confirms the quasi two dimensionality of the electronic structure. Fermi surface is quite complicated and consists of several hole and electron pockets. By comparing our data with DFT calculation, we find our results consistent with a localized picture of f electrons. This provides clues to understanding of unusual transport and thermodynamical properties of this important material.

V. ACKNOWLEDGMENTS

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Figure 4. Intensity plot along high symmetry direction overlay with localized f electron band structure calculation. Green arrow mark the band character and location.