When superconductivity does not fear magnetism: Insight into electronic structure of RbEuFe₄As₄

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In the novel stoichiometric iron-based material RbEuFe₄As₄ superconductivity coexists with a peculiar long-range magnetic order of Eu 4f states. Using angle-resolved photoemission spectroscopy, we reveal a complex three dimensional electronic structure and compare it with density functional theory calculations. Multiple superconducting gaps were measured on various sheets of the Fermi surface. High resolution resonant photoemission spectroscopy reveals magnetic order of the Eu 4f states deep into the superconducting phase. Both the absolute values and the anisotropy of the superconducting gaps are remarkably similar to the sibling compound without Eu, indicating that Eu magnetism does not affect the pairing of electrons. A complete decoupling between Fe- and Eu-derived states was established from their evolution with temperature, thus unambiguously demonstrating that superconducting and a long range magnetic orders exist independently from each other. The established electronic structure of RbEuFe₄As₄ opens opportunities for the future studies of the highly unorthodox electron pairing and phase competition in this family of iron-based superconductors with doping.

INTRODUCTION

Even after many years of intensive research, the mechanism of the electron pairing in iron based superconductors (IBSC) is still not well understood. It is known that emergence of superconductivity (SC) with doping is accompanied by a suppression of the structural and magnetic transitions observed in undoped parent compounds. Theoretical considerations suggest that the pairing mechanism in IBSCs might be due to spin-fluctuation exchange [1–4]. In the case of weakly doped compounds the superconducting gap should have a s± symmetry. However, numerous studies of these materials demonstrate that the physics of the pairing is more complex because of the multiorbital and multiband nature of low-energy fermionic excitations [5–7]. It turns out that both the symmetry and the structure of the order parameter result from a rather non-trivial interplay between spin-fluctuation exchange, intraband Coulomb repulsion, and the momentum structure of the competing interactions [8, 9].

In a newly discovered class of iron based superconductors, the so-called “1144 family” [10], there are particular interesting examples of AEuFe₄As₄ (A=Rb,Cs) compounds where superconductivity coexists with unusual Eu-magnetic order [11–14]. The structure of these compounds (see Fig.1(a)) can be...
viewed as an intergrowth between undoped EuFe$_2$As$_2$ and heavily overdoped AFe$_2$As$_2$. As a result, RbEuFe$_4$As$_4$ becomes intrinsically hole-doped, exhibiting superconductivity with a high transition temperature $\sim$36K. Moreover, the evidence of in-plane ferromagnetic ordering of the Eu$^{2+}$ spins below $\sim$15K was given by magnetization measurements for both polycrystalline [15] and single crystal samples [16, 17].

The optical conductivity measurements [18] revealed a fully opened SC gap of about 5meV, while the inelastic neutron scattering (INS) measurements showed spin resonance at 18meV, and a three-dimensional helical antiferromagnetic order of the Eu atoms was determined from the neutron diffraction [19]. It has been even proposed that this exotic helical magnetic structure is generated by superconductivity due to the interplay of Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange interaction and macroscopic electromagnetic interaction between the superconducting and magnetic subsystems [20, 21]. However, magnetic susceptibility and resistivity studies of RbEuFe$_4$As$_4$ under pressure found that the superconductivity onset is suppressed monotonically by pressure while the magnetic transition was enhanced at higher pressures [13].

In the present study we elucidate the interdependence of Fe-superconductivity and Eu-magnetism in this novel class of materials by combining experimental observations with theoretical calculations. In particular, we investigate the question whether the spatial proximity of the ferromagnetically ordered layer of Eu atoms can induce a non-trivial pairing of the Fe conduction electrons with exotic symmetry of the superconducting gap.

## RESULTS

### Fermi surface topology

In Fig.1(c), we present a Fermi surface (FS) map obtained at 70 eV, corresponding to $k_z$=0 for the $\Gamma$ point where the hole pockets are largest. The measured $\mu$SR surface for the bulk crystal structure is similar to optimally hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ (Ba''122'') pnictides and consists of three hole-like pockets at the centre of the Brillouin zone ($\Gamma$-Z points in Fig.1(b)) and propeller-like electron-like pockets at the corners of the Brillouin zone ($M$-$A$ points in Fig.1(b)) [24]. High symmetry direction band dispersions measured in $M$-$\Gamma$-$M$ direction in Fig.1(d) show the

![Image](image_url)

**FIG. 1. Structure and Fermi surface topology:** (a) Layered crystal structure and (b) bulk Brillouin zone of RbEuFe$_4$As$_4$. (c) $k_x$-$k_y$ Fermi surface map, measured using 70 eV photons in linear vertical (LV) polarisation. (d) Band dispersions in $M$-$\Gamma$-$M$ direction measured using 113 eV photons for LV polarisation. All data measured at 40K. (e) Calculated Fermi surface for $k_z$=0 for FM configuration of Eu moments. (f) Calculated band dispersions in $M$-$\Gamma$-$M$ direction showing Fe 3d (green) and Eu 4f (blue) bands.
FIG. 2. Superconducting properties: (a) Hole pocket Fermi surface at Z-point (40K); (b,c) Band dispersions at Z-point below and above $T_c$ measured along high symmetry direction shown as red double arrow in panel (a), white arrows indicate corresponding $k_F$ positions; (d) Electron pocket Fermi surface at M-point (40K); (e,f) Band dispersions at M-point below and above $T_c$ measured along high symmetry direction shown as red double arrow in panel (d); white arrows indicate corresponding $k_F$ positions; (g,h) Photoemission spectra at equivalent $k_F$ positions above and below $T_c$ together with the superconducting gap fit for hole and electron pockets; (i) Temperature dependence of the superconducting gap obtained for three different Fermi surface sheets of the hole pocket at Z-point.

Superconducting properties

Using high resolution ARPES we have observed an opening of the superconducting gap on all sheets of the Fermi surface below $T_c$ (Fig.2). The measured values of the gap at the hole pockets at the centre of the Brillouin zone are significantly varying from one sheet to another, with the largest gap opening on the most inner hole-like Fermi surface, similar to other Ba“122” pnictides [32–34]. We have also observed opening of the superconducting gap at the centre of the electron pocket. The obtained maximum value of the gap at the electron pocket is about $\sim$9 meV according to the fit using the Dynes function [35] and is similar to the largest gap value for the hole pocket (Fig.2(g,h)). This is in a very good agreement with superconducting gap ARPES measurements for optimally doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [32–34]. These maximum gap values derived by ARPES are also in a very good agreement with published data from optical measurements of RbEuFe$_4$As$_4$ [18].

With increasing temperature, the superconducting gap gradually closes for all bands with classical BCS-type dependence, indicating a significant inter-orbital interaction of the Fe 3d bands [36–38], similar to sibling compound CaKFe$_4$As$_4$ [39] and Ba“122” family [34, 40]. By measuring the temperature dependence of the superconducting gap at the hole pockets (Fig.2(i)) within our energy resolution we do not observe any change in the magnitude of the gap at the Eu magnetic transition $T_m \sim$14K. This result suggests that superconductivity in RbEuFe$_4$As$_4$ that involves Fe 3d states near the Fermi level might be fully decoupled from the magnetism on the Eu sublattice.

In order to investigate the effects of Eu 4f magnetism on the Fe 3d electronic states in the vicinity of the Fermi level we performed band structure calculations within DFT for several types of Eu magnetic moment ordering: ferromagnetic (FM), A-type antiferromagnetic ordering similar to EuFe$_2$As$_2$ (AFM180) [41, 42], and for the recently proposed helical antiferromagnetic alignment of Eu moments (AFM90) [19]. As can be seen in Fig.3, irrespective of magnetic ordering the bands forming electron M-pockets at the Fermi level are almost completely determined by $d_{x^2-y^2}$ Fe orbitals, whereas the bands forming hole $\Gamma$-pockets are contributed by a mixture
of different Fe 3d orbitals. In accordance with our calculations the basic band structure at the Fermi level and the orbital character of Fe 3d bands are almost unaffected by different magnetic orderings of Eu 4f magnetic moments. Therefore the underlying spin-fluctuation-mediated interaction of Fe 3d itinerant electrons responsible for superconducting pairing could be completely insensitive to the onset of the local magnetic order on Eu atoms.

**Magnetic properties**

In order to explore the magnetic properties of the Eu sublattice, we performed resonant photoemission measurements at the Eu 4d→4f threshold using 142 eV photons. This allows a resonant enhancement of the 4f emission from divalent Eu ions and gain insight exclusively into the spectral pattern of these states. The experiment was performed as a function of temperature going from the normal state (40K) via the superconducting state (20K) into the magnetic state (7K) as shown in Fig. 4(a,b). Wide binding energy range valence band spectrum in insert of Fig. 4(a) clearly shows that there are no apparent changes in its shape and intensity when passing the onset of the superconducting order. At the same time, our calculations have shown that AFM90 configuration for Eu moments is the most favorable. The total energy of the AFM180 is of 1.8 meV/f.u. lower whereas the FM configuration is less favorable among all considered magnetic configurations.

Furthermore, in Fig. 4(c,d) we performed XMCD measurements to directly probe magnetic moments of both Eu and Fe atoms. Following standard sum rule analysis, we obtained a large magnetic moment 7.04±0.01µB for Eu and a tiny magnetic moment 0.03±0.01µB for Fe. This value of the local magnetic moment on Eu is in close agreement with published transport data [15–17]. The DFT calculated Eu magnetic moment of 6.97µB is in excellent agreement with experiment, and is the same for any considered magnetic configuration.

To investigate magnetic properties of Fe sublattices, we used a disordered local moment (DLM) approach, in which magnetic moments of individual atoms are randomly oriented as in a paramagnetic state [47, 48]. This gives the magnitude of Fe magnetic moments of 1.47µB. Note however that XMCD is sensitive only to the ordered component of the magnetic moment, which is very small, indicating a large fluctuating moment on the Fe sites. FPLO calculations for the experimental structure with the FM order of Eu moments and U=5 eV and J=1 eV give the Fe moments of 1.7*10^−3 µB and Eu moments of 7.13 µB. Obtained value of the ordered Fe moment is consistent with XMCD measurements, while DLM shows the local moment which can be much larger. Applying the magnetic force theorem [49] we found that the exchange interaction between the Eu and Fe moments is rather weak and negative indicating an antiferromagnetic coupling between the moments. At the same time, our calculations have shown that AFM90 configuration for Eu moments is the most favorable. The total energy of the AFM180 is of 1.8 meV/f.u. higher whereas the FM configuration is less favorable among considered being of 0.1 eV/f.u. higher in energy respecting AFM90.
**DISCUSSION**

Using high resolution ARPES, we observed that the Fermi surface of RbEuFe$_2$As$_4$ consists of multiple hole-like and electron-like sheets, similar to other iron-based superconductors. In this unique compound, the itinerant Fe 3d electrons at the Fermi level are in proximity to the layers of ferromagnetically ordered large local magnetic moments of Eu atoms. Therefore, the observed coexistence of Eu helical antiferromagnetic order with Fe superconductivity raises a widely discussed question about the possibility of non-trivial superconducting pairing in this material. One of the most direct ways to answer the question about prevailing exotic pairing and its relation to co-existing Eu magnetic order is to probe the symmetry of the superconducting order parameter. RbEuFe$_4$As$_4$ is an intrinsically hole-doped iron-based superconductor with a high transition temperature $T_c \sim 35$K. Our ARPES data show the opening of a full superconducting gap on all Fermi surface sheets below $T_c$ with average gap value of $\sim 5$meV, in good agreement with optical gap study [18]. The fact that the temperature dependence of the various hole-like superconducting gaps follows a similar BCS-like relation for different sheets of the Fermi surface unambiguously points towards a significant interorbital coupling in this system [50]. The Fermi surface topology as well as temperature dependence and anisotropy of the order parameter are very similar for RbEuFe$_4$As$_4$ and optimally doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$. This suggests that superconductivity in both compounds is of the same origin and is consistent with theoretically proposed $s^\pm$ pairing due to the spin-fluctuations.

The temperature dependence of the hole-like superconducting gaps does not show any uncommon behaviour below the three-dimensional helical antiferromagnetic ordering of the Eu$^{2+}$ spins at 14K. Moreover, our ARPES data show that weakly dispersing Eu 4f states are at 1-1.7eV below the Fermi level, and our DFT calculations confirm that these localized states do not hybridize with itinerant Fe 3d states and therefore do not contribute to the superconducting pairing directly. Both Fe- and Eu-subsystems are almost decoupled magnetically, since the Fe 3d bands and the Eu 4f states are well separated in energy, despite structural proximity of Fe-As and Eu layers. Besides, the calculations of the band structure in vicinity of the Fermi level for different types of magnetic orders of Eu moments establish that the topology and the orbital character of Fe 3d bands are not constrained by the particular magnetic structure. All these observations suggest that the iron-derived states near the Fermi level are completely independent from the localized Eu 4f electronic subsystem.

Furthermore, we have also addressed the proposition [20] that superconductivity is responsible for the stabilisation of the helical antiferromagnetic ground state. Using high resolution resonant photoemission measurements we not only observed the Eu 4f final state multiplet structure, but, by measuring its temperature and polarisation dependence, confirmed in plane ordering of Eu$^{2+}$ localized moments below $T_m$. These data also show that Eu 4$f^7 \rightarrow 4$f$^6$ final-state multiplet structure and therefore the magnetic arrangement does not change with the onset of superconductivity at $T_c$. DFT calculations not only give correct binding energies of the Eu 4f bands and correct magnetic moments of Eu, but also confirm that the helical magnetic structure is the most energetically favourable one.

The tiny magnetic moment of Fe obtained in our XMCD experiment is in agreement with results of Mössbauer spectroscopy measurements [51]. These Mössbauer experiments also suggest that Fe-subsystem might not be magnetically ordered despite the long-range order of Eu-subsystem. Using DLM approximation, we show that exchange interaction between Fe and Eu is small and negative. Since existing DFT functionals can not describe correctly charge and spin fluctuations, we can not determine magnetic order in the Fe sublattice and assume here that the Fe moments are disordered. The magnetism of iron in RbEuFe$_4$As$_4$ could be different from isostructural CaKFe$_4$As$_4$ [12, 14] due to the possible biquadratic Eu-Fe coupling [52]. Nevertheless, both magnetic RbEuFe$_4$As$_4$ and non-magnetic CaKFe$_4$As$_4$ compounds have similar Fermi surface topology [39]. In both cases the superconducting gaps for different Fermi surface sheets have no clear nodes and are roughly isotropic. These observations once again demonstrate that superconductivity in RbEuFe$_4$As$_4$ is completely independent from the magnetic order on Eu.

RbEuFe$_2$As$_4$ is one of the few examples of the iron-based superconducting compounds with high transition temperature without additional doping. The superconductivity in this material is enhanced for several reasons: due to the stoichiometric chemical composition there is no defect scattering detrimental for superconductivity; due to the charge carriers from Rb layers it has maximized density of states at the Fermi level beneficial for superconductivity; and it has no long-range magnetic order of iron moments, which is a well-known competitor of the superconductivity. Moreover, a recent study of the doped RbEu(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ system shows that by introducing extra electrons with Ni substitution at doping levels above $x \sim 0.07$ the ferromagnetic superconductor with $T_c > T_m$ transforms into a superconducting ferromagnet with $T_m > T_c$ [53]. This doping induced interchange opens an unique opportunity to study the underlying change of electronic structure and possible existence of unconventional superconductivity. Therefore, understanding the electronic structure of the parent compound RbEuFe$_4$As$_4$ becomes extremely important for any future studies of prospective superconducting magnets among iron-based pnictides.

To summarize, we have performed high resolution ARPES and ResPES studies of a newly discovered iron based superconductor RbEuFe$_4$As$_4$ with helical magnetic order. We observed three hole-like Fermi surface pockets around $\Gamma$-point and small electron-like pockets around M-point, all formed by Fe derived bands. Our DFT calculations show that topology and the orbital character of the Fe 3d band does not strongly depend on the particular magnetic ordering of the Eu 4f states. A full nodeless superconducting gap with BCS-
like temperature dependence has been observed on both hole- and electron-like bands below $T_c$. In particular, no deviations in the temperature dependence of the order parameter have been detected below the magnetic transition $T_m$ despite of a clear indication for an in plane magnetic order of Eu$^{2+}$ localized moments. All these facts unambiguously indicate that Eu-magnetism and Fe-superconductivity are almost fully decoupled in RbEuFe$_4$As$_4$.

**METHODS**

**Single crystal sample growth**

High quality single crystals of RbEuFe$_4$As$_4$ were grown by the self-flux technique. Superconducting and magnetic transition temperatures $T_c \sim 35$K and $T_m \sim 14$K were confirmed by magnetisation measurements [54].

**X-ray spectroscopy measurements**

High resolution angle-resolved photoemission spectroscopy (ARPES) and resonant photoemission spectroscopy (ResPES) measurements were performed at the I05 beamline at the Diamond Light Source, UK [55]. The photoelectron energy and angular distributions were analysed with a SCI- ENTA R4000 hemispherical analyser. The angular resolution was $0.2^\circ$, and the overall energy resolution was better than 10 meV for Fermi surface mapping and $\sim 2$-3 meV for SC gap measurements. In order to increase the contrast in Fig.4(b) we have used a procedure described in Ref. [56]. X-ray magnetic circular dichroism (XMCD) measurements were performed at the HECTOR end-station of the BOREAS beamline at the ALBA synchrotron radiation facility [57]. Absorption spectra in total electron yield mode have been recorded at the Fe L$_{3,2}$ and Eu M$_{5,4}$ edges in magnetic fields up to 6 T and at 5K sample temperature, well below $T_m \sim 14$K.

**First-principles calculations**

Density functional theory (DFT) calculations were performed considering various basis sets [58]: the projector augmented-wave (PAW) method for representation of core electrons [59, 60] as implemented in the VASP code [61], the all-electron full-potential localized orbitals (FPLO) basis set code [62], and a full potential Green’s function method within the multiple scattering theory [63]. Calculations were benchmarked with various codes. The generalized gradient approximation (GGA-PBE) [64] to the exchange-correlation potential was applied. To perform the calculations we have used the experimental crystal structure parameters [15]. The Eu 4f states were treated employing the GGA+$U$ approach [65] within the Dudarev scheme [66]. The $U_{eff}=U-J$ value (where $U$ and $J$ are the effective on-site Coulomb and exchange interaction parameters, respectively) for the Eu 4f states was chosen to be equal to 5.5 eV. Using this $U_{eff}$ value we found a good agreement with the experimentally obtained binding energy of the Eu 4f states of RbEuFe$_4$As$_4$ [67].

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