Multigap superconductivity in RbCa$_2$Fe$_4$As$_4$F$_2$ investigated using µSR measurements

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The superconducting properties of the recently discovered double Fe$_2$As$_2$ layered high-$T_c$ superconductor RbCa$_2$Fe$_4$As$_4$F$_2$ with $T_c \approx$ 30 K have been investigated using magnetization, heat capacity, transverse-field (TF) and zero-field (ZF) muon-spin rotation/relaxation (µSR) measurements. Our low field magnetization measurements and heat capacity ($C_p$) reveal an onset of bulk superconductivity with $T_c \sim$ 30.0(4) K. Furthermore, the heat capacity exhibits a jump at $T_c$ of $\Delta C_p / T_c = 94.6$ (mJ/mole-K$^2$) and no clear effect of applied magnetic fields was observed on $C_p(T)$ up to 9 T between 2 K and 5 K. Our analysis of the TF-µSR results shows that the temperature dependence of the magnetic penetration depth is better described by a two-gap model, either isotropic $s+\text{d}$-wave or $s+\text{d}$-wave than a single gap isotropic $s$-wave or $d$-wave model for the superconducting gap. The presence of two superconducting gaps in RbCa$_2$Fe$_4$As$_4$F$_2$ suggests a multiband nature of the superconductivity, which is consistent with the multigap superconductivity observed in other Fe-based superconductors, including ACa$_2$Fe$_4$As$_2$F$_2$ (A=K and Cs). Furthermore, from our TF-µSR study we have estimated an in-plane penetration depth $\lambda_{\text{in}}(0) = 231.5(3)$ nm, superconducting carrier density $n_s = 7.45 \times 10^{28}$ m$^{-3}$, and carrier’s effective-mass $m^* = 2.45 m_e$. Our ZF µSR measurements do not reveal a clear sign of time reversal symmetry breaking at $T_c$, but the temperature dependent relaxation between 150 K and 1.2 K might indicate the presence of spin-fluctuations.

The results of our present study have been compared with those reported for other Fe pnictide superconductors. The discovery of high temperature superconductivity in cuprates, high temperature superconductivity in cuprates, Sr$_2$CuO$_2$Cl$_2$ (122-family), which have a body centered tetragonal ThCr$_2$Si$_2$-type structure (I4/mmm), where the ubiquitous Fe$_2$As$_2$ layers of the Fe arsenide superconductors lie between the alkaline/alkaline earth atom layers shown in Fig. 1. Highly correlated matters (including Cu, Ni, Rh and Pd) doped BaFe$_2$As$_2$ (122-family), which have a body centered tetragonal ThCr$_2$Si$_2$-type structure (I4/mmm), where the ubiquitous Fe$_2$As$_2$ layers of the Fe arsenide superconductors lie between the alkaline/alkaline earth atom layers shown in Fig. 1. Recently superconductivity with $T_c \sim$ 35 K has been reported in CaFe$_2$As$_4$ (A = K, Rb, Cs, 1144-family), and these materials consist of different arrangements of the layers along the c-axis also displayed in Fig. 1. In this structure, the alternating arrangement of the A and Ca layers leads to two inequivalent As sites either side of the Fe sheets. The crystallographically inequivalent position of the Ca and A atoms changes the space group from I4/mmm (as for the 122-family) to P4/mmm. Furthermore, the different valence attraction from Ca$^{2+}$ and A$^{1+}$ layers to Fe$_2$As$_2$ and the different ionic radii leads to different lengths of the As-Fe bonds, which was proposed to be an important parameter for controlling the $T_c$ of Fe-based superconductors. Stoichiometric CaFe$_2$As$_4$ is intrinsically near optimal hole doping and does not exhibit a high temperature structural phase transition. Similar to the optimally doped 122 compounds, probes of the gap structure and inelastic neutron scattering results strongly suggest the presence of a fully gapped $s^\pm$ state. Further angle-resolved photoemission spectroscopy (ARPES) measurements of CaFe$_2$As$_4$ re-

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I. INTRODUCTION

The discovery of high temperature superconductivity in fluorine-doped LaFeAsO (1111-family) with a transition temperature of $T_c \sim$ 26 K by Kamihara et al. has generated a considerable research interest world-wide to understand the nature of the superconductivity in this new class of compound. It was realized soon after the discovery of the 122 family that the Fe-based superconductors could be increased up to 56 K as observed in Gd$_{0.8}$Th$_{0.2}$FeAsO (2), Sr$_{0.5}$Sm$_{0.5}$FeAs (3), and Ca$_{0.4}$Nd$_{0.6}$FeAs (2). Until this discovery, high temperature superconductivity in cuprates, created the impression that only Cu-O planes are pivotal for understanding the mechanism of high temperature superconductivity. (Of course, the Fe-based superconductors can be understood as two Fe planes and an additional O plane. In any case, the presence of Fe planes is crucial.)

Another highly investigated family of Fe-based superconductors is hole (i.e. K) and electron (i.e. Co, Mn, Ni, Rh and Pd) doped BaFe$_2$As$_2$ (122-family), which have a body centered tetragonal ThCr$_2$Si$_2$-type structure (I4/mmm), where the ubiquitous Fe$_2$As$_2$ layers of the Fe arsenide superconductors lie between the alkaline/alkaline earth atom layers shown in Fig. 1. Highly correlated matters (including Cu, Ni, Rh and Pd) doped BaFe$_2$As$_2$ (122-family), which have a body centered tetragonal ThCr$_2$Si$_2$-type structure (I4/mmm), where the ubiquitous Fe$_2$As$_2$ layers of the Fe arsenide superconductors lie between the alkaline/alkaline earth atom layers shown in Fig. 1. Recently superconductivity with $T_c \sim$ 35 K has been reported in CaFe$_2$As$_4$ (A = K, Rb, Cs, 1144-family), and these materials consist of different arrangements of the layers along the c-axis also displayed in Fig. 1. In this structure, the alternating arrangement of the A and Ca layers leads to two inequivalent As sites either side of the Fe sheets. The crystallographically inequivalent position of the Ca and A atoms changes the space group from I4/mmm (as for the 122-family) to P4/mmm. Further, the different valence attraction from Ca$^{2+}$ and A$^{1+}$ layers to Fe$_2$As$_2$ and the different ionic radii leads to different lengths of the As-Fe bonds, which was proposed to be an important parameter for controlling the $T_c$ of Fe-based superconductors. Stoichiometric CaFe$_2$As$_4$ is intrinsically near optimal hole doping and does not exhibit a high temperature structural phase transition. Similar to the optimally doped 122 compounds, probes of the gap structure and inelastic neutron scattering results strongly suggest the presence of a fully gapped $s^\pm$ state. Further angle-resolved photoemission spectroscopy (ARPES) measurements of CaFe$_2$As$_4$ re-

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The pairing symmetry of the Cooper pairs in a superconductor is manifested in an energy gap in the single-particle excitation spectrum. The superconducting gap structure is an important characteristic for a superconductor. There is experimental evidence that cuprate-based unconventional superconductors have distinct d-wave nodal gap symmetry compared with conventional phonon-mediated superconductors which have nodeless s-wave gap. On the other hand the superconducting gap symmetry in iron-based superconductors is rather more diverse and the subject of ongoing debate.\textsuperscript{121-123}\textsuperscript{124-126} Whereas nodeless gap structures have been observed in some of the doped 122-families,\textsuperscript{111,112} 1144-families,\textsuperscript{21,22,24-26} \(\text{As}_x\text{Fe}_y\text{Se}_z\) (A=K, Cs)\textsuperscript{27} and Fe\(\text{Te}_{1-x}\text{Se}_x\)\textsuperscript{28-30} the nodalities of small superconducting gaps have been reported in La\(\text{OFe}_2\)\textsuperscript{31}, Li\(\text{Fe}_2\)\textsuperscript{32}, K\(\text{Fe}_2\)\textsuperscript{33}, Rb\(\text{Fe}_2\)\textsuperscript{34}, Ba\(\text{Fe}_2(\text{As}_1-x\text{P}_x)\)\textsuperscript{35}, Ba\(\text{Fe}_2\text{Ru}_x\text{As}_y\)\textsuperscript{36} and Fe\(\text{S}\)\textsuperscript{37}. Furthermore applied pressure and doping or chemical pressure change the gap symmetry from nodeless to nodal in Ba\(0.65\text{Rb}_{0.35}\text{Fe}_2\text{As}_2\)\textsuperscript{38} and in Ba\(\text{Fe}_2\text{Ru}_x\text{As}_y\)\textsuperscript{39}. More interestingly the single crystal \(\mu\)SR study on Fe\(\text{Se}\) reveals a nodeless gap (anisotropic-s-wave) along the c-axis, but one nodal and one isotropic \((s+d\text{-wave})\) gap in the ab-plane\textsuperscript{40}.

To understand the mechanism of unconventional superconductivity and develop realistic theoretical models of Fe-based superconductors it is very important to study the pairing symmetry and the nature of the superconducting gap. There is no general consensus on the nature of pairing in iron-based superconductors leading to a variety of possibilities ranging from \(s_{\pm}\) wave to \(s_{++}\) to d wave. Furthermore it is also important to investigate whether time-reversal symmetry (TRS) in the superconducting state is preserved or not as well as the role of spin-fluctuations. Broken symmetry can modify the physics of a system and nature of the pairing, thereby resulting in novel and uncommon behavior. Muon-spin rotation and relaxation (\(\mu\)SR) is an ideal and sensitive microscopic technique to investigate the properties of the superconducting state. Transverse field (TF) \(\mu\)SR provides information on the field distribution in the superconducting state and hence information on the penetration depth and gap symmetry. On the other hand zero-field (ZF) \(\mu\)SR allows the detection of very small internal fields and hence can provide direct information about whether TRS is preserved. Recently we have investigated the nature of the superconducting gap and TRS in \(\text{ACa}_2\text{Fe}_4\text{As}_4\text{F}_2\) \((\text{A}=\text{K} \text{and Cs})\) compounds using \(\mu\)SR measurements\textsuperscript{40,41}. We found two superconducting gaps with at least one nodal gap in these compounds, but no clear sign of TRS breaking. It is therefore important to investigate the gap symmetry and TRS in A=\(\text{Rb}\) compound. Here we report TF- and ZF-\(\mu\)SR measurements of the A=\(\text{Rb}\) compound. Our study shows that the superfluid density derived from the depolarization rate of the TF-\(\mu\)SR fits better to two isotropic gaps following a \(s+s\text{-wave}\) model and ZF-\(\mu\)SR does not reveal any clear sign of TRS breaking below \(T_c\).

![Diagram](image)
II. EXPERIMENTAL DETAILS

The sample was characterized using powder x-ray diffraction (XRD), magnetic susceptibility and heat capacity measurements. The heat capacity was measured using a Quantum Design Physical Property Measurement System (PPMS) between 1.8 and 80 K. A standard thermal relaxation method was used with a sample mass of 8 mg. The DC magnetization measurements were carried out using a Quantum Design Magnetic Property Measurement System (MPMS).Muon spin relaxation/rotation (μSR) experiments were carried out on the MuSR spectrometer at the ISIS pulsed muon source of the Rutherford Appleton Laboratory, UK. The μSR measurements were performed in transverse-field (TF), zero-field (ZF) and longitudinal field modes. A powder sample of RbCa$_2$Fe$_4$As$_4$F$_2$ was mounted on a silver sample holder. The sample was cooled under He-exchange gas in a He-4 cryostat operating in the temperature range of 1.5 K−300 K. TF−μSR experiments were performed in the superconducting mixed state in an applied field of 40 mT, well above the lower critical field of $\mu_0 H_{c1} \sim 20$ mT (see Fig.2c) of this material. Data were collected in the field-cooled (FC) mode, where the magnetic field was applied above the superconducting transition temperature and the sample was then cooled down to base temperature. Muon spin rotation and relaxation is a dynamic method that allows one to study the nature of the pairing symmetry in superconductors.

The vortex state in the case of type-II superconductors gives rise to a spatial distribution of local magnetic fields; which demonstrates itself in the μSR signal through a relaxation of the muon polarization. Zero-field (ZF) μSR measurements were performed from 1.2 K to 150 K in the longitudinal geometry. We also performed longitudinal field μSR measurements at 1.2 K and 35 K. The μSR data were analyzed using WiMDA.

III. RESULTS AND DISCUSSIONS

The analysis of the powder x-ray diffraction at 300 K reveals that the sample is single phase and crystallizes in the tetragonal crystal structure with space group I4/mmm (No. 139, Z = 2) as shown in Fig. 2(a). The refined values of the lattice parameters are $a = 3.8716(1)$ Å and $c = 31.667(1)$ Å.

The low-field magnetic susceptibility measured in an applied field of 1 mT shows an onset of diamagnetism below 30 K indicating that superconductivity occurs at 30 K and the superconducting volume fraction is close to 100% at 10 K [Fig. 2(a)]. This result confirms the bulk nature of superconductivity with $T_c = 30$ K in RbCa$_2$Fe$_4$As$_4$F$_2$, which is comparable to $T_c = 33.3$ K and 29 K observed in ACa$_2$Fe$_3$As$_4$F$_2$ (A=K and Cs), respectively.

The magnetization isotherm $M(H)$ curve at 3 K [Fig. 2(b)] shows typical behaviour for type-II superconductivity. The lower critical field $H_{c1}$ obtained from the M vs H plot at 3 K by linear fitting the data between 0 and 15 mT is about 20 mT [Fig. 2(c)]. The upper critical field ($\mu_0 H_{c2}$) measurements using the field dependent resistivity reveals the slope $d\rho_0 H_{c2}/dT\sim 13.9$ T/K at $T_c$ and the Pauli limit is $\mu_0 H_{P}\sim 1.84 T_c$, which is 55.2 T [23]. Further using the orbital limiting upper critical field $\mu_0 H_{c2}(0)=0.73(dH_{c2}/dT)_{T_c}$, we have estimated $\mu_0 H_{c2}(0)=0.30$ KT. This value of $H_{c2}$ gives the coherence length $\xi=(\Phi_0/(2\pi H_{c2}))^{1/2}=1.04$ nm, where $\Phi_0=2.07\times 10^{-15}$ Tm$^2$ is the magnetic flux quantum. The specific heat ($C_p$) is displayed in Fig. 2(d) for zero field and an applied fields up to 9 T (Fig.2(f)). A clear anomaly is observed in the zero field $C_p$ corresponding to the superconducting transition at around 30.4(4) K. The jump
in $C_p$ was estimated by linearly extrapolating the data above and below $T_c$, yielding a jump of $\Delta C_p/T_c = 94.6$ (mJ/mol K$^2$), which is smaller than 150 (mJ/mol K$^2$) observed in KCa$_2$Fe$_4$As$_4F_2$. To shed light on the nature of the gap symmetry we also performed field dependent heat capacity measurements up to a field of 9 T. We found that the heat capacity is almost independent between 2 K and 5 K.

Figures 3 (a) and (b) show the TF−μSR precession signals above and below $T_c$ obtained in FC mode with an applied field of 40 mT (well above $H_{c1} \approx 20$ mT but below $H_{c2} >> 7$ T, at 3 K, see Fig.2b) and Figs. 3 (c) and (d) show the corresponding maximum entropy spectra, respectively. It is clear that above $T_c$ the μSR spectra show a very small relaxation mainly from the quasi-static nuclear moments, and the internal field distribution is very sharp and centered near the applied field. However at 1.2 K μSR spectra show strong damping and the internal field distribution has two components, one very sharp near the applied field and one very broad which is shifted lower than applied field. The observed decay of the μSR signal with time below $T_c$ is due to the inhomogeneous field distribution of the flux-line lattice. We attribute the narrow component at the applied field to muons stopping in the silver sample holder, which indicates that the field distribution within the vortex lattice is described well by one Gaussian centered at a field below 40 mT. We have used an oscillatory decaying Gaussian function to fit the TF−μSR time dependent asymmetry spectra:

$$A(t) = A_1 e^{-\sigma^2t^2/2} \cos(\gamma_\mu B_1 t + \phi) + A_2 \cos(\gamma_\mu B_2 t + \phi),$$

(1)

where $\gamma_\mu/2\pi = 135.5$ MHz/T is the muon gyromagnetic ratio, $\sigma$ is the Gaussian relaxation rate, $\phi$ is the phase, which is related to the detector geometry, $A_1$ and $A_2$ are the magnitudes of the terms from the sample and silver holder respectively, while $B_1$ and $B_2$ are respective internal fields. We grouped all detectors in 8 groups and all the groups were fitted simultaneously using the WIMDA software. The total amplitudes for each group of detectors were fixed. Furthermore, we first estimated the value of $A_1 \approx 0.7$ and $A_2 \approx 0.3$ by fitting the 1.2 K data and kept them fixed during the analysis allowing us to extract the temperature dependence of the relaxatons rate $\sigma(T)$. Equation 1 contains the total relaxation rate $\sigma$ from the superconducting fraction of the sample; there are contributions from the vortex lattice ($\sigma_{vc}$) and nuclear dipole moments ($\sigma_{nm}$) (see Fig.4b inset), where the latter is assumed to be constant over the entire temperature range $[\sigma = \sqrt{\sigma_{vc}^2 + \sigma_{nm}^2}]$. The contribution from the vortex lattice, $\sigma_{vc}$, was determined by quadratically subtracting the background nuclear dipolar relaxation rate ($\sigma_{nm} = 0.138(5) \mu s^{-1}$) obtained from the spectra measured above $T_c$. As the applied field (40 mT) is much less than the upper critical field ($\mu_0 H_{c2} > 7$ T), $\sigma_{vc}$ can be directly related to the effective penetration depth $\lambda_{eff}$ using the following equation:

$$\frac{\sigma_{vc}}{\gamma_\mu} = 0.0609 \Phi_0 / \lambda_{eff}^2,$$

(2)

where $\Phi_0$ is the magnetic flux quantum. This relation between $\sigma_{vc}$ and $\lambda_{eff}$ is valid for 0.13/$\kappa c^2<<<(H/H_{c2})<1$, where $\kappa=\lambda/\xi>70(43)$. Since RbCa$_2$Fe$_4$As$_4F_2$ has a two-dimensional layered crystal structure with large separation between Fe$_2$As$_2$-layers, the out of plane penetration depth ($\lambda_{c}$) is much larger than that in the plane ($\lambda_{ab}$), so that the effective penetration depth can be estimated as $\lambda_{eff} = 3^{\frac{1}{2}} \lambda_{ab}$. (43)

Furthermore the penetration depth is directly related to the normalized superfluid density, $n_{ns}$. In our analysis we modelled the temperature dependent normalized superfluid density using the following equation:

$$n_{ns}(T) = \frac{\lambda_{ab}^2(T, \Delta)}{\lambda_{ab}^2(0)} = 1 + \frac{1}{\pi} \int_0^{2\pi} \frac{\partial f}{\partial E} \frac{EdE d\phi}{\sqrt{E^2 - \Delta^2(T, \varphi)}}$$

(3)

where $f = [1 + \exp (\Delta/kT)]^{-1}$ is the Fermi function. The temperature and angular dependence of the gap is given by $\Delta(T, \varphi) = \Delta_0 \delta(T/T_c) g(\varphi)$, whereas $g(\varphi)$ refers to the angular dependence of the superconducting gap function and $\varphi$ is the azimuthal angle along the Fermi
FIG. 4. (Color online) (a) Temperature dependence of $\lambda_{\text{ab}}^2$ of RbCa$_2$Fe$_4$As$_4$F$_2$. $\lambda_{\text{ab}}^2$ of FC mode (symbols), where the lines are the fits to the data using Eq. 3 for various two-gap models. The solid black line shows the fit using an isotropic $s$+$d$-wave model with $\Delta_1(0) = 8.15 \pm 0.01$ meV and $\Delta_2(0) = 0.88 \pm 0.01$ meV, the dotted blue line shows the fit to an $s$+$d$-wave model with $\Delta_1(0) = 8.08 \pm 0.02$ meV and $\Delta_2(0) = 0.92 \pm 0.01$ meV and the dashed-dotted green line shows the fit to a $d$+$d$-wave model with $\Delta_1(0) = 14.05 \pm 0.26$ meV and $\Delta_2(0) = 1.26 \pm 0.02$ meV. The inset shows low temperature data in an expanded scale. (b) The normalized internal field shift as a function of temperature. The inset shows temperature dependence of total relaxation rate $\sigma$ and the dotted line shows the temperature independent contribution of nuclear depolarization rate $\sigma_{\text{n.m.}}$.

surface. We have used the BCS formula for the temperature dependence of the gap, which is given by $\delta(T/T_c) = \tanh\left((1.82)(1.018(T_c/T - 1))^{0.5}\right)$. $g(\varphi)$ is given by (a) $1$ for $s$-wave gap [also for $s + s$ wave gap], (b) $\{\cos(2\varphi)\}$ for an $d$-wave gap with line nodes. For the two-gap analysis, we have used a weighted sum of the two components of the resulting normalized superfluid density:

$$n_{\text{ns}} = wn_{\text{ns}}(\Delta_1, T) + (1-w)n_{\text{ns}}(\Delta_2, T)$$

(4)

Figure 4 (a) shows the temperature dependence of $\lambda_{\text{ab}}^2$, measured in an applied field of 40 mT. $\lambda_{\text{ab}}^2$ increases with decreasing temperature confirming the presence of a flux-line lattice and indicates a decrease of the magnetic penetration depth with decreasing temperature. Further below 10 K $\lambda_{\text{ab}}^2$ shows an upturn indicating multigap behavior. The onset of diamagnetism below the superconducting transition can be seen through the decrease in the internal field below $T_c$ as shown in Fig. 4(b). From the analysis of the observed temperature dependence of $\lambda_{\text{ab}}^2$, using different models for the gap, the nature of the superconducting gap can be probed. We have analyzed the temperature dependence of $\lambda_{\text{ab}}^2$ based on five different models, the single gap isotropic $s$-wave and line nodal $d$-wave models, as well as isotropic $s$+$s$-wave, $s$+$d$-wave and $d$+$d$-wave two-gap models. It was clear from the analysis that single-gap models did not fit the data (fits are not shown). The fits to the $\lambda_{\text{ab}}^2$ data with various two-gap models using Eq. (3) are shown by lines in Fig. 4(a) and the estimated fit parameters are given in Table I. It is clear from the goodness of fitted $\chi^2$ values given in Table I that the $d$+$d$-wave model does not fit the data very well. On the other hand the isotropic $s$+$s$-wave, $s$+$d$-wave models show good fits to the $\lambda_{\text{ab}}^2$ data. The value of $\chi^2 = 3.0$ for $s$+$s$-wave model is slightly less than 3.1 for $s$+$d$-wave model. The estimated parameters for the $s$+$s$ ($s$+$d$)-wave model show one larger gap $\Delta_1(0) = 8.15$ (8.08) (meV) and another much smaller gap $\Delta_2(0) = 0.88$ (0.92) (meV). The smaller gap is a nodal gap in the $s$+$d$-wave model. Our $\mu$SR analysis suggests that an
s+s-wave model explains better the temperature dependence of the superfluid density than an s+d-wave model. The value of $\lambda_{ab}(0) = 231.5 \pm 3$ mm and $T_c = 29.19 \pm 0.04$ K were estimated from the s+s-wave fit. The estimated value of $2\Delta_1(0)/k_BT_c = 6.48$ from the s+s-wave fit is larger than the value 3.53 expected for BCS superconductors, indicating the presence of strong coupling and unconventional superconductivity in RbCa$_2$Fe$_4$As$_4$F$_2$. On the other hand for the smaller gap the value $2\Delta_1(0)/k_BT_c \approx 0.7$ is much smaller than the BCS value. The two-gap nature, one larger and another smaller than the BCS value, are commonly observed in Fe-based superconductors as well as in Bi$_2$O$_4$S. The observation of two isotropic gaps and nodeless superconductivity in RbCa$_2$Fe$_4$As$_4$F$_2$ is very similar to that observed in CaKFe$_4$As$_4$, where clear evidence is found for multigap nodeless superconductivity with an $s_\pm$ pairing state. Recently we have observed two gaps in Ca$_2$Fe$_4$As$_4$F$_2$ ($A=K$ and Cs) and ThFeAsN, but at least one gap appears to be nodal in these compounds. Two superconducting gaps (one larger and another smaller) were also observed in SrFe$_{1.85}$Co$_{0.15}$As$_2$, with $T_c = 19.2$ K in an STM study. Moreover combined ARPES and $\mu$SR studies on Ba$_{1-x}$K$_x$Fe$_2$As$_2$ with $T_c = 32.0$ K also revealed the presence of two gaps ($\Delta_1 = 9.1$ meV and $\Delta_2 = 1.5$ meV). The recent $\mu$SR study on FeSe single crystals revealed that the superconducting gap is most probably anisotropic s-wave (nodeless) along the crystallographic c-axis, but it fits better to a two-gap s+d-wave model with one nodal gap in the ab-plane. Furthermore, nodal superconductivity has been observed in cuprate superconductors and the recently discovered quasi-1D Cr-based superconductors, A$_2$Cr$_3$As$_3$ ($A = K$ and Cs) as well.

As with other phenomenological parameters characterizing a superconducting state, the penetration depth can also be related to microscopic quantities. Within London theory $\lambda^{2}_{t} = \lambda^{2}_{\text{eff}} = m^{*}c^2/4\pi\hbar^{2}\lambda_{s}$, where $m^{*} = (1 + \lambda_{e-ph}/m_s)$ is the effective mass and $\lambda_s$ is the density of superconducting carriers. Within this simple picture $\lambda^{2}_{t}$ is independent of magnetic field. $\lambda_{e-ph}$ is the electron-phonon coupling constant, which can be estimated from $\Theta_D$ and $T_c$ using McMillan’s relation

$$\lambda_{e-ph} = \frac{1.04 + \mu^{*} \ln(\Theta_D/1.45T_c)}{(1 - 0.62\mu^{*})\ln(\Theta_D/1.45T_c) + 1.04},$$

where $\mu^{*}$ is the repulsive screened Coulomb parameter and usually assigned as $\mu^{*} = 0.13$. As we do not have heat capacity above 80 K for the present Rb-sample, we first estimated the value of $\Theta^{Rb}_{D} = 366$ K. Then using a scaling factor which incorporates the differing molecular weight and unit-cell volume, we estimated $\Theta^{Cs}_{D} = 351.6$ K (similar for the Cs-sample $\Theta^{Cs}_{D} = 443.4$ K). For RbCa$_2$Fe$_4$As$_4$F$_2$ we have used $T_c = 29.19$ K together with $\mu^{*} = 0.13$ and have estimated $\lambda_{e-ph} = 1.45$. This value of $\lambda_{e-ph}$ is very similar to 1.38 for LiFeAs$^{23}$, 1.53 for PrFeAsO$_{0.60}$F$_{0.12}$ and 1.2 for LaO$_{0.9}$F$_{0.1}$FeAs$^{24}$. On the other hand for many Fe-based superconductors (11-family and 122-family) and HTSC cuprates (YBCO-123)
smaller values of \( \lambda_{\text{e-ph}} = 0.02 \) to 0.2 and 0.02, respectively have been reported. Further assuming that roughly all the normal state carriers \( (n_\text{s}) \) contribute to the superconductivity (i.e., \( n_\text{s} \approx n_\text{e} \)) and using the value of \( \lambda_{\text{ab}}(0) = 231.5 \pm 3 \) nm, we have estimated the superconducting carrier density \( n_\text{s} \) and effective-mass enhancement \( m^* \) to be \( n_\text{s} = 7.45 \times 10^{26} \) carriers/m\(^3\), and \( m^* = 2.45 m_\text{e} \), respectively. We also estimated these parameters for \( \text{ACa}_2\text{Fe}_4\text{As}_4\text{F}_2 \) \((A=\text{K and Cs})\) samples (see Table-II) for comparison.

Zero-field \( \mu\text{SR} \) measurements were performed from 1.2 K to 150 K and the results are displayed in Fig.5(a) for four selected temperatures. The data were fitted with the sum of a Lorentzian and Gaussian relaxation function

\[
A_0(t) = A(\exp(-\Delta t) + (1 - a)\exp(-\sigma_{ZF}(t^2/2))) + A_{bg},
\]

(6)

where \( A_{bg} \) is the temperature independent background arising from muons stopping on the sample holder. The value of \( A_{bg}=5.898(8)% \) and \( a=0.367 \) were estimated by fitting the 150 K data and were kept fixed during the analysis. At high temperature the relaxation is dominated by Gaussian decay, while at low temperature the relaxation changes to a Lorentzian decay. Moreover, there is a gradual decrease of initial asymmetry \( (A) \) with decreasing temperature, which suggests the development of fast component, which relaxes faster than the resolution of the experiment. The asymmetry exhibits a small drop below 70 K, which could be due to a competing magnetic/structural phase or related to some unknown phase transition and needs further investigation. The temperature dependence of \( \Lambda \) and \( \sigma_{ZF} \) increases with decreasing temperature between 150 K and 75 K, followed by a weak temperature dependence between 75 K and 25 K. Below 25 K both \( \Lambda \) and \( \sigma_{ZF} \) show a moderate temperature dependence. These results suggest the presence of weak magnetic fluctuations, but neither quantity shows a detectable anomaly upon passing through \( T_c \), indicating an absence of time reversal symmetry breaking. However, since \( \Lambda(T) \) and \( \sigma_{ZF}(T) \) show some temperature dependence, a weak increase of the relaxation due to time reversal symmetry breaking cannot be entirely ruled out. Furthermore we also performed longitudinal fields (LF) measurements at 1.2 K and 35 K in applied LF of 25, 40 and 50 mT and the data of 40 mT field are shown in the inset of Fig. 5a. At all applied longitudinal fields, the data showed negligible relaxation (i.e. the asymmetry is almost constant with time), indicating very weak spin-fluctuations which require a very small LF field to decouple the \( \mu\text{SR} \) signal.

The correlation between \( T_c \) and \( \sigma_{ZF}(0) \) (or \( \lambda_{\text{ab}}(0) \)) observed in \( \mu\text{SR} \) studies has suggested a new empirical framework for classifying superconducting materials. Here we explore the role of muon spin relaxation rate/penetration depth in the superconducting state for the characterisation and classification of superconducting materials as first proposed by Uemura et al. In particular we focus upon the Uemura classification scheme which considers the correlation between the superconducting transition temperature, \( T_c \), and the effective Fermi temperature, \( T_F \), determined from \( \mu\text{SR} \) measurements of the penetration depth. Within this scheme strongly correlated “exotic” superconductors, i.e. high \( T_c \) cuprates, heavy fermions, Chevrel phases and the organic superconductors, form a common but distinct group characterised by a universal scaling of \( T_c \) with \( T_F \) such that \( 1/10 > (T_c/T_F) > 1/100 \) (Fig. 6). For conventional BCS superconductors \( 1/1000 > (T_c/T_F) \). Considering the value of \( T_c/T_F = 0.04 \) for \( \text{RbCa}_2\text{Fe}_4\text{As}_4\text{F}_2 \) (see Fig. 6), this material can be classified as an exotic superconductor, according to Uemura’s classification. Furthermore we have also plotted the data of \( \text{ACa}_2\text{Fe}_4\text{As}_4\text{F}_2 \) \((A=\text{K and Cs})\) in Fig. 6, which also belong to the same class.

It has been found that the jump in the heat capacity \( \Delta C_p/T_c \) at \( T_c \) is also related to \( T_c \) for electron and hole doped \( \text{BaFe}_2\text{As}_2 \) superconductor. We have plotted the heat capacity jump of \( \text{ACa}_2\text{Fe}_4\text{As}_4\text{F}_2 \) \((A=\text{K and Rb})\) on the scaling plot shown in Fig.7. It is clear that for \( A=\text{K} \) and Rb compounds the heat capacity jump also follows this trend suggesting a common relation between \( \Delta C_p/T_c \sim T_c^2 \), the so-called BNC scaling.

## IV. CONCLUSIONS

In conclusion, we have presented magnetization, heat capacity and transverse field (TF) and zero-field (ZF) muon spin rotation (\( \mu\text{SR} \)) measurements in the normal and the superconducting state of \( \text{RbCa}_2\text{Fe}_4\text{As}_4\text{F}_2 \), which has a double Fe\(_2\)As\(_2\) layered tetragonal crystal structure. Our magnetization and heat capacity measurements confirmed the bulk superconductivity with \( T_c = 30.0 \) (4) K. From the TF \( \mu\text{SR} \) we have determined the muon depolarization rate in the FC mode associated with the vortex-lattice. The temperature dependence of the superfluid density fits better to a two-gap model, with either an isotropic \( s+\text{d-wave} \) or an \( s+\text{d-wave} \) gap, than to single gap isotropic \( s \)-wave or \( d \)-wave models. The \( s+\text{s} \) and \( s+\text{d-wave} \) model fits give a goodness of fit \( (\chi^2) \) value of 3.0 and 3.1, respectively, suggesting that an \( s+\text{s-wave} \) model is an appropriate for the gap structure of \( \text{RbCa}_2\text{Fe}_4\text{As}_4\text{F}_2 \). Furthermore, the value (for the larger gap) of \( 2\Delta_1(0)/k_B T_c = 6.48 \pm 0.08 \) obtained from the \( s+s \)-wave model fit is larger than 3.53, expected for BCS superconductors, indicating the presence of strong coupling superconductivity, that is supported through a larger value of \( \lambda_{\text{e-ph}} \), in \( \text{RbCa}_2\text{Fe}_4\text{As}_4\text{F}_2 \). Moreover, two superconducting gaps have also been observed in the Fe-based families of superconductors, including in other \( \text{ACa}_2\text{Fe}_4\text{As}_4\text{F}_2 \) \((A=\text{K and Cs})\) compounds and hence our observation of two gaps is in agreement with the general trend observed in Fe-based superconductors. It is an open question why the \( A=\text{Rb} \) material is more consistent with two isotropic gaps, while \( A=\text{K} \) and Cs have at least one nodal gap despite the ionic size (lattice pa-
TABLE I. Fitted parameters obtained from the fit to the $\sigma_{\text{sc}}(T)$ data of RbCa$_2$Fe$_4$As$_4$F$_2$ (as shown in Fig. 4(a)) using different gap models.

| Model     | $T_c$ (K) | $\Delta_1(0)$ | $\Delta_2(0)$ (meV) | $2\Delta(0)/k_BT_c$ | $\lambda_{ab}^s(0)$ | $\chi^2$ |
|-----------|-----------|----------------|---------------------|---------------------|---------------------|---------|
| s+s wave  | 29.19(4)  | 8.15(1)        | 0.88(1)             | 6.48                | 0.70                | 0.87(1) |
| s+d wave  | 29.19(5)  | 8.08(2)        | 0.92(1)             | 6.42                | 0.73                | 0.87(2) |
| d+d wave  | 28.57(7)  | 14.05(26)      | 1.26(2)             | 11.41               | 1.02                | 1.02    |

TABLE II. The comparison of various estimated parameters, transition temperature $T_c$, electron-phonon coupling constant, $\lambda_{\text{e-ph}}$, carrier effective mass, $m^*$, superfluid density, $n_s$, and Fermi temperature, $T_F$ of RbCa$_2$Fe$_4$As$_4$F$_2$ (A=K, Rb and Cs).

| Compound               | $T_c$(K) | $\lambda_{\text{e-ph}}$ | $m^*(m_e)$ | $n_s(10^{26}\text{m}^{-3})$ | $T_F$(K) |
|------------------------|----------|-------------------------|------------|-----------------------------|----------|
| KCa$_2$Fe$_4$As$_4$F$_2$| 33.36    | 1.588                   | 2.588      | 8.01                        | 741.77   |
| RbCa$_2$Fe$_4$As$_4$F$_2$| 29.19    | 1.451                   | 2.451      | 7.45                        | 727.41   |
| CsCa$_2$Fe$_4$As$_4$F$_2$| 28.31    | 1.438                   | 2.438      | 6.66                        | 652.32   |

parameters and unit cell volume) increasing, while $T_c$ decreases linearly, going down the alkali atom group from K to Cs. Further confirmation of the presence of two gaps and their symmetry in RbCa$_2$Fe$_4$As$_4$F$_2$ could be found from angle-resolved photoemission spectroscopy (ARPES) study and TF-µSR study on single crystals, for H$\parallel$c-axis and H$\parallel$ab-plane, of RbCa$_2$Fe$_4$As$_4$F$_2$.

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