Muon-spin rotation measurements of the magnetic penetration depth in the iron-based superconductor $Ba_{1-x}Rb_xFe_2As_2$

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Measurements of the magnetic penetration depth λ in the Fe-based superconductor Ba_{1-x}Rb_xFe₂As₂ (x=0.3, 0.35, 0.4) were carried out using the muon-spin rotation (μ SR) technique. The temperature dependence of λ is well described by a two-gap s+s-wave scenario with a small gap $\Delta_1\approx 1$ -3 meV and a large gap $\Delta_2\approx 7$ -9 meV. By combining the present data with those previously obtained for RbFe₂As₂ a decrease of the BCS ratio $2\Delta_2/k_BT_c$ with increasing Rb content x is observed. On the other hand, the BCS ratio $2\Delta_1/k_BT_c$ is almost independent of x. In addition, the contribution of Δ_1 to the superfluid density is found to increase with x. These results are discussed in light of the suppression of interband processes upon hole doping.

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I. INTRODUCTION

The discovery¹ of superconductivity in iron oxypnictide LaFeAsO_{1-x}F_x has generated great interest in the phenomenon of high temperature superconductivity. The basic units responsible for superconductivity are the fluorite type $[Fe_2Pn_2]$ layers where Pn is a pnictogen element (P, As, Sb, and Bi). These layers are separated by spacer layers which play the role of a charge reservoir. In the fluorite-type layers the Fe atoms are surrounded by four pnictogen atoms forming a tetrahedron. The first class of iron-based superconductors studied has the ZrCuSiAs structure (1111 compounds), where the spacer layer $[Ln_2O_2]$ has the "antifluoride" or Pb₂O₂ structure. With Ln = Sm a critical temperature higher than 55 K was observed.²

Superconductivity with $T_c = 38 \text{ K}$ was also found in the ternary systems AFe₂As₂ (Refs. 3 and 4) (122 compounds) adopting the tetragonal ThCr₂Si₂ structure. In this structure the spacer layer is provided by an alkali earth element A =Ca, Sr, or Ba. Doping is realized by the substitution of A by an alkali metal such as K, Cs, or Rb. Several disconnected Fermisurface sheets contribute to superconductivity as revealed by angle-resolved photoemission spectroscopy (ARPES).^{5–7} Moreover, indications of multigap superconductivity in the system $Ba_{1-x}K_xFe_2As_2$ were obtained from the temperature dependence of the magnetic penetration depth λ by means of muon-spin rotation $(\mu SR)^8$ and ARPES.⁵ The magnetic penetration depth is one of the fundamental parameters of a superconductor since it is closely related to the density of the superconducting carriers n_s and their effective mass m^* via the relation $1/\lambda^2 \propto n_s/m^*$. The temperature dependence of λ reflects the topology of the superconducting gap occurring in the density of states of the superconducting ground state. The μ SR technique provides a powerful tool to measure λ in type II superconductors.9

As demonstrated in previous works, 4,10 the value of T_c for hole-doped $Ba_{1-x}Rb_xFe_2As_2$ decreases monotonically upon increasing the Rb content x in the overdoped region. However,

in contrast to the overdoped cuprates, T_c remains finite even at the highest doping level x = 1 with $T_c = 2.52$ K (Ref. 4). A detailed study of the doping dependence of T_c may help to clarify the origin of high- T_c superconductivity in these iron-based systems. It is thus of importance to investigate the superconducting properties of optimally doped $Ba_{1-x}Rb_xFe_2As_2$ and compare the results with those obtained for RbFe₂As₂ (Ref. 10).

In this paper, we report on μ SR studies of the temperature and field dependence of the magnetic penetration depth of optimally doped Ba_{1-x}Rb_xFe₂As₂ (x=0.3,0.35,0.4). We compare the present data with the previous results of overdoped RbFe₂As₂ (Ref. 10) and discuss the combined results in light of the suppression of interband processes upon hole doping.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of $Ba_{1-x}Rb_xFe_2As_2$ were prepared in evacuated quartz ampoules by a solid state reaction method. Fe₂As, BaAs, and RbAs were obtained by reacting high purity As (99.999%), Fe (99.9%), Ba (99.9%), and Rb (99.95%) at 800 °C, 650 °C, and 500 °C, respectively. Using stoichiometric amounts of BaAs or RbAs and Fe₂As the terminal compounds BaFe2As2 and RbFe2As2 were synthesized at 950 °C and 650 °C, respectively. Finally, the samples of $Ba_{1-x}Rb_xFe_2As_2$ with x = 0.3, 0.35, 0.4 were prepared from appropriate amounts of single-phase BaFe₂As₂ and RbFe₂As₂. The components were mixed, pressed into pellets, placed into alumina crucibles, and annealed for 100 hours at 650 °C with one intermittent grinding. Powder x-ray diffraction analysis revealed that the synthesized samples are single phase materials. Zero-field (ZF) and transverse-field (TF) μ SR experiments were performed at the π M3 beamline of the Paul Scherrer Institute (Villigen, Switzerland), using the general purpose instrument (GPS). The sample was mounted inside of a gas-flow ⁴He cryostat on a sample holder with a standard veto setup providing essentially a low-background μSR signal. All TF experiments were carried out after a field-cooling procedure.

III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) exhibit the transverse-field (TF) muon-time spectra for $Ba_{1-x}Rb_xFe_2As_2$ (x=0.3, 0.4) measured in an applied magnetic field of $\mu_0H=0.04$ T above (45 K) and below (1.7 K) the superconducting (SC) transition temperature T_c . Above T_c the oscillations show a small relaxation due to the random local fields from the nuclear magnetic moments. Below T_c the relaxation rate strongly increases due to the presence of a nonuniform local field distribution as a result of the formation of a flux-line lattice (FLL) in the SC state. It is well known that undoped $BaFe_2As_2$ is not superconducting at ambient pressure and undergoes a spin-density wave (SDW) transition of the Fe moments far above T_c (Ref. 11). The SC state can be achieved either under pressure 12,13 or by appropriate charge carrier doping 14 of the parent compounds, leading to a suppression of the

of the parent compounds, leading to a suppression of the hold $\mu_0H=0.04\ T$ $Ba_{0.7}Rb_{0.3}Fe_2As_2$ $1.7\ K$ 0.2 0.2 0.2 0.3 0.3 0.4 0.4 0.4 0.5 0.4 0.4 0.5

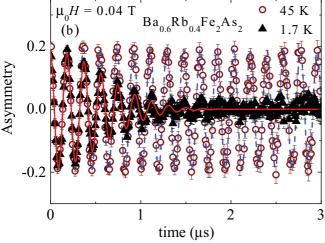


FIG. 1. (Color online) Transverse field (TF) μ SR time spectra obtained in $\mu_0H=0.04$ T above and below T_c (after field cooling the sample from above T_c): (a) Ba_{0.7}Rb_{0.3}Fe₂As₂ and (b) Ba_{0.6}Rb_{0.4}Fe₂As₂. The solid and the dashed lines represent fits to the data by means of Eq. (1).

SDW state. Magnetism, if present in the samples, may enhance the muon depolarization rate and falsify the interpretation of the TF- μ SR results. Therefore, we have carried out ZF- μ SR experiments above and below T_c to search for magnetism (static or fluctuating) in Ba_{1-x}Rb_xFe₂As₂ (x=0.3,0.35,0.4). As shown in Fig. 2(a) no sign of either static or fluctuating magnetism could be detected in ZF time spectra down to 1.7 K. Moreover, the ZF relaxation rate is small and changes very little between 45 and 1.7 K. The spectra are well described by a standard Kubo-Toyabe depolarization function, ¹⁵ reflecting the field distribution at the muon site created by the nuclear moments.

It was reported $^{16-18}$ that in some iron-based superconductors $BaFe_{2-x}Co_xAs_2$ and $SrFe_{2-x}Co_xAs_2$ field induced magnetism exists. In the present work $TF-\mu SR$ spectra measured in different applied fields (see Fig. 1 for $\mu_0 H = 0.04 T$) exhibit a Gaussian-like depolarization above and below T_c that is typical of nuclear moments and the vortex lattice in polycrystalline samples, respectively. In the presence of dilute or fast fluctuating electronic moments one expects an

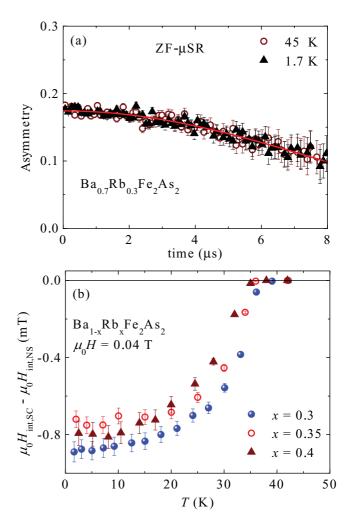


FIG. 2. (Color online) (a) ZF- μ SR time spectra for Ba_{0.7}Rb_{0.3} Fe₂As₂ recorded above and below T_c . The line represents the fit to the data of a standard Kubo-Toyabe depolarization function (Ref. 15). (b) Temperature dependence of the difference between the internal field $\mu_0 H_{\rm int,SC}$ measured in the SC state and the one measured in the normal state $\mu_0 H_{\rm int,NS}$ at T=42 K.

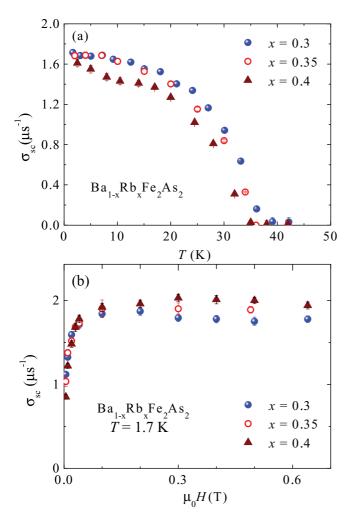


FIG. 3. (Color online) (a) Temperature dependence of the superconducting muon spin depolarization rate $\sigma_{\rm sc}$ measured in an applied magnetic field of $\mu_0 H = 0.04$ T for ${\rm Ba_{1-x}Rb_xFe_2As_2}$ (x=0.3,0.35,0.4). (b) Field dependence of $\sigma_{\rm sc}$ at 1.7 K.

exponential depolarization of the TF-μSR spectrum, which is absent in the present case. Moreover, the SC muon depolarization rate σ_{sc} is constant at high fields as shown in Fig. 3(b). In addition we observed a diamagnetic shift of the internal magnetic field $\mu_0 H_{\text{int}}$ sensed by the muons below T_c . This is evident in Fig. 2(b), where we plot the difference between the internal field $\mu_0 H_{\text{int,SC}}$ measured in the SC state and one $\mu_0 H_{\text{int,NS}}$ measured in the normal state at T = 42 K. Note, that in the systems $BaFe_{2-x}Co_xAs_2$ and $SrFe_{2-x}Co_xAs_2$, where the field induced magnetism was detected, paramagnetic shift was observed 16-18 instead of the expected diamagnetic shift imposed by the SC state. All these observations indicate that there is no field induced magnetism in the system $Ba_{1-x}Rb_xFe_2As_2$ down to 1.7 K. The absence of magnetism in $Ba_{1-x}Rb_xFe_2As_2$ implies that the increase of the TF relaxation rate below T_c is attributed entirely to the vortex

The TF μ SR data were analyzed by using the following functional form:¹⁹

$$P(t) = A \exp\left[-\frac{\left(\sigma_{\rm sc}^2 + \sigma_{\rm nm}^2\right)t^2}{2}\right] \cos(\gamma_\mu B_{\rm int}t + \varphi). \quad (1)$$

Here A denotes the initial asymmetry, $\gamma/(2\pi) \simeq 135.5$ MHz/T is the muon gyromagnetic ratio, and φ is the initial phase of the muon-spin ensemble. $B_{\rm int}$ represents the internal magnetic field at the muon site, and the relaxation rates $\sigma_{\rm sc}$ and $\sigma_{\rm nm}$ characterize the damping due to the formation of the FLL in the superconducting state and of the nuclear magnetic dipolar contribution, respectively. In the analysis $\sigma_{\rm nm}$ was assumed to be constant over the entire temperature range and was fixed to the value obtained above $T_{\rm c}$ where only nuclear magnetic moments contribute to the muon depolarization rate σ . As indicated by the solid lines in Fig. 1, the μ SR data are well described by Eq. (1). The temperature dependence of $\sigma_{\rm sc}$ for Ba_{1-x}Rb_xFe₂As₂ (x=0.3,0.35, and 0.4) at $\mu_0H=0.04$ T is shown in Fig. 3(a). Below $T_{\rm c}$ the relaxation rate $\sigma_{\rm sc}$ starts to increase from zero due to the formation of the FLL.

For polycrystalline samples the temperature dependence of the London magnetic penetration depth $\lambda(T)$ is related to the superconducting part of the Gaussian muon spin depolarization rate $\sigma_{\rm sc}(T)$ by the equation²⁰

$$\frac{\sigma_{\rm sc}^2(T)}{\gamma_u^2} = 0.00371 \frac{\Phi_0^2}{\lambda^4(T)},\tag{2}$$

where $\Phi_0 = 2.068 \times 10^{-15}$ Wb is the magnetic-flux quantum. Equation (2) is only valid when the separation between the vortices is smaller than λ . In this case, according to the London model σ_{sc} is field independent.²⁰ We measured σ_{sc} as a function of the applied field at 1.7 K (see Fig. 3(b)). Each point was obtained by field cooling the sample from above T_c to 1.7 K. First, σ_{sc} strongly increases with increasing magnetic field until reaching a maximum at $\mu_0 H \simeq 0.03$ T and then above 0.03 T stays nearly constant up to the highest field (0.64 T) investigated. Such a behavior is expected within the London model and is typical for polycrystalline high temperature superconductors (HTS's).²¹ The observed field dependence of σ_{sc} implies that for a reliable determination of the penetration depth the applied field must be larger than $\mu_0 H = 0.03$ T.

 $\lambda(T)$ can be calculated within the local (London) approximation ($\lambda \gg \xi$) by the following expression:^{19,22}

$$\frac{\lambda^{-2}(T, \Delta_{0,i})}{\lambda^{-2}(0, \Delta_{0,i})} = 1 + \frac{1}{\pi} \int_0^{2\pi} \int_{\Delta(T, \varphi)}^{\infty} \left(\frac{\partial f}{\partial E}\right) \frac{E dE d\varphi}{\sqrt{E^2 - \Delta_i(T, \varphi)^2}},\tag{3}$$

where $f = [1 + \exp(E/k_BT)]^{-1}$ is the Fermi function, φ is the angle along the Fermi surface, and $\Delta_i(T,\varphi) = \Delta_{0,i}\delta(T/T_c)g(\varphi)$ ($\Delta_{0,i}$ is the maximum gap value at T=0). The temperature dependence of the gap is approximated by the expression $\delta(T/T_c) = \tanh\{1.82[1.018(T_c/T-1)]^{0.51}\}$ (Ref. 23), while $g(\varphi)$ describes the angular dependence of the gap and it is replaced by 1 for both an *s*-wave and an s+s-wave gap, and $|\cos(2\varphi)|$ for a *d*-wave gap.

The temperature dependence of the penetration depth was analyzed using either a single gap or a two-gap model which is based on the so-called α model. This model was first discussed by Padamsee *et al.*²⁵ and later on was successfully used to analyze the magnetic penetration depth data in HTS's. ^{23,26} According to the α model, the superfluid density is calculated

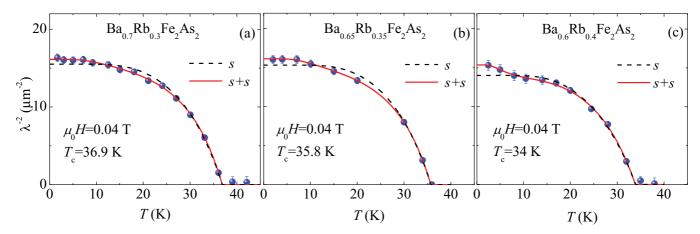


FIG. 4. (Color online) The temperature dependence of λ^{-2} for Ba_{1-x}Rb_xFe₂As₂, measured in an applied field of $\mu_0 H = 0.04$ T: (a) x = 0.3, (b) x = 0.35, and (c) x = 0.4. The dashed lines correspond to a single gap BCS s-wave model, whereas the solid ones represent a fit using a two-gap (s + s)-wave model.

for each component using Eq. (3) and then the contributions from the two components added together

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \omega_1 \frac{\lambda^{-2}(T, \Delta_{0,1})}{\lambda^{-2}(0, \Delta_{0,1})} + \omega_2 \frac{\lambda^{-2}(T, \Delta_{0,2})}{\lambda^{-2}(0, \Delta_{0,2})},\tag{4}$$

where $\lambda^{-2}(0)$ is the penetration depth at zero temperature, $\Delta_{0,i}$ is the value of the *i*th (i = 1, 2) superconducting gap at T = 0 K, and ω_i is a weighting factor which measures their relative contributions to λ^{-2} ($\omega_1 + \omega_2 = 1$).

The results of the analysis for $Ba_{1-x}Rb_xFe_2As_2$ (x = 0.3, 0.35, 0.4) are presented in Fig. 4. The dashed and the solid lines represent a fit to the data using an s-wave and a s + s-wave models, respectively. The analysis appears to rule out the simple s-wave model as an adequate description of $\lambda(T)$ for $Ba_{1-x}Rb_xFe_2As_2$ (x = 0.3, 0.35, 0.4). A *d*-wave gap symmetry was also tested, but was found to be inconsistent with the data. The two-gap s + s-wave scenario with a small gap Δ_1 and a large gap Δ_2 , describes the experimental data remarkably well. The results of all samples extracted from the data analysis are summarized in Table I. A two-gap scenario is in line with the generally accepted view of multigap superconductivity in Fe-based HTS.^{5,6,8,27-29} The magnitudes of the large and the small gaps for $Ba_{1-x}Rb_xFe_2As_2$ (x = 0.3, 0.35, 0.4) (see Table I) are in good agreement with the results of a previous report.⁵ There it was pointed out that most Fe-based HTS's exhibit two-gap superconducting behavior,

TABLE I. Summary of the parameters obtained for polycrystalline samples of $Ba_{1-x}Rb_xFe_2As_2$ (x=0.3,0.35,0.4,1) by means of μ SR. The data for x=1.0 are taken from Ref. 10.

	x = 0.3	x = 0.35	x = 0.4	x = 1.0
$T_{\rm c}$ (K)	36.9	35.8	34	2.52
Δ_1 (meV)	3.2(7)	2.9(8)	1.1(3)	0.15(2)
$2\Delta_1/k_{\rm B}T_{\rm c}$	2.0(5)	1.9(4)	0.8(6)	1.4(2)
Δ_2 (meV)	9.2(3)	8.8(3)	7.5(2)	0.49(4)
$2\Delta_2/k_{\rm B}T_{\rm c}$	5.8(6)	5.7(5)	5.1(4)	4.5(4)
ω_1	0.19(5)	0.21(4)	0.15(3)	0.36(3)
λ (nm)	249(15)	250 (17)	255 (9)	267(5)

characterized by a large gap with $2\Delta/k_BT_c = 7(2)$ and a small one with 2.5(1.5). To reach a more complete view of the superconducting properties of Ba_{1-x}Rb_xFe₂As₂ as a function of the Rb composition (hole doping), we combined the present data with the previous μSR results on RbFe₂As₂ (Ref. 10) which presents the case of a naturally overdoped system. Figure 5 shows the small gap to T_c ratio $2\Delta_1/k_BT_c$, the large gap to T_c ratio $2\Delta_2/k_BT_c$, and the weight ω_1 of the small gap to the superfluid density as a function of Rb concentration. The data for RbFe₂As₂ are taken from Ref. 10. Interestingly, the ratio $2\Delta_2/k_BT_c$ decreases with increasing x. On the other hand, the ratio $2\Delta_1/k_BT_c$ for the small gap is essentially independent of x. In addition, the weighting factor ω_1 is found to increase with increasing x. We note that in the optimally doped 122-system $Ba_{1-x}K_xFe_2As_2$ several bands cross the Fermi surface (FS).⁵⁻⁷ They consist of inner (α) and outer (β) hole-like bands, both centered at the zone center Γ , and an electron-like band (γ) centered at the M point. The superconducting gap opened on the β band was found to be smaller than those on the α and γ bands. It was proposed that the enhanced interband scattering between the α and γ bands might promote the kinetic process of pair scattering between these two FSs, leading to an increase of the pairing amplitude.³⁰ Hole doping may cause a shift of the band bottom of the electron pockets above the Fermi level E_F . As a result, the interband scattering between α and γ bands would diminish since the γ band is in the unoccupied side and concomitantly the size of the α band is increased. According to ARPES results, a decrease of interband scattering will lead to a decrease of the pairing amplitude and the ratio $2\Delta/k_BT_c$ in agreement with the results presented in Fig. 5(a). These results suggest the possible role of interband processes in optimally hole-doped iron-based 122 superconductors.^{6,30}

One of the most interesting results of μSR investigations in HTS's is the observation of a remarkable proportionality between T_c and the zero-temperature relaxation rate $\sigma(0) \propto 1/\lambda^2(0)$ (Uemura relation).³¹ This relation $T_c(\sigma)$, which seems to be generic for various families of cuprate HTS's, has the features that upon increasing the charge carrier doping T_c first increases linearly in the underdoped region

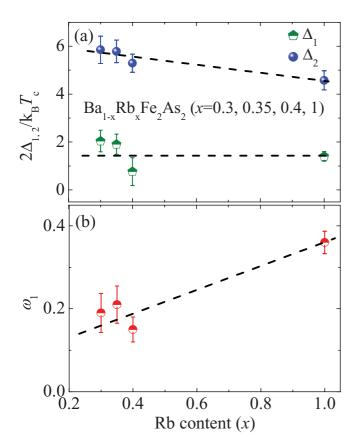


FIG. 5. (Color online) (a) Superconducting gap to T_c ratios $2\Delta_{1,2}/k_BT_c$ and (b) the contribution ω_1 of the small gap to the superfluid density as a function of the Rb composition for $\mathrm{Ba}_{1-x}\mathrm{Rb}_x\mathrm{Fe}_2\mathrm{As}_2$ (x=0.3, 0.35, 0.4, 1.0). The measurements were performed in an applied magnetic field of $\mu_0H=0.04$ T. The data for RbFe₂As₂ are taken from Ref. 10. The dashed lines represent guides to the eyes.

(Uemura line), then saturates, and finally is suppressed for high carrier doping. The initial linear trend of the Uemura relation indicates that for these unconventional HTS's the ratio T_c/E_F (E_F is the Fermi energy) is up to two orders of magnitude larger than for conventional BCS superconductors. Figure 6 shows T_c vs $\sigma(0)$ plot for various hole- and electrondoped high T_c Fe-based superconductors (after Ref. 28), including the present results. The solid line shows the Uemura relation in hole-doped cuprates³¹ and the dashed line corresponds to electron-doped cuprates as observed by Shengelaya et al. 32 The Uemura relation for Fe-based superconductors was already discussed in Ref. 28. Here, we demonstrate that the data points for $Ba_{1-x}Rb_xFe_2As_2$ (x = 0.3, 0.35, 0.4) are located in the Uemura plot close to those of the other Fe-based superconductors. On the other hand, for naturally fully overdoped RbFe₂As₂, the ratio $T_c/\sigma(0)$ is strongly reduced. The small value of ratio $T_c/\sigma(0)$ is characteristic for conventional superconductors. For comparison the point for conventional BCS superconductor Nb is also shown on the Uemura plot. This suggests that superconductivity in the compound RbFe₂As₂ has a more conventional character. Additional experiments are in progress to clarify this point.

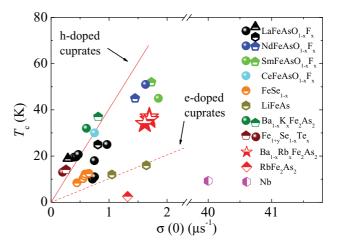


FIG. 6. (Color online) Uemura plot for hole- and electron-doped high $T_{\rm c}$ Fe-based superconductors (after Ref. 28). The Uemura relation observed for underdoped cuprates is also shown (solid line for hole doping and dashed line for electron doping) (after Ref. 32). The point for conventional BCS superconductor Nb is also shown. Data points for the pnictides are taken from Refs. 27–29, 33–39. The stars show the data for $Ba_{1-x}Rb_xFe_2As_2$ (x=0.3,0.35,0.4) obtained in this work. The point for RbFe₂As₂ is taken from Ref. 10.

IV. SUMMARY AND CONCLUSION

In summary, we performed transverse-field μ SR measurements of the magnetic penetration depth λ on polycrystalline samples of the iron-based HTS's $Ba_{1-x}Rb_xFe_2As_2$ (x = 0.3, 0.35, 0.4). The values of the superconducting transition temperature T_c and the zero temperature values of λ were estimated to be $T_c = 36.9$, 35.8, 34 K and $\lambda(0) = 249(15)$, 250(17), 255(9) nm for x = 0.3, 0.35, and 0.4, respectively. The temperature dependence of λ is well described by a two-gap s + s-wave scenario with gap values similar to $Ba_{1-x}K_xFe_2As_2$ (Refs. 5 and 8). ARPES investigations of Ba_{1-x}K_xFe₂As₂ revealed that the large gap opens on the inner hole-like Fermi surface (α band) centered at the Γ point and on the electron-like FS (γ band) centered at the M point (tetragonal structure notations), while the small gap opens on the outer hole-like band (β) of the Γ point.³⁰ We found that the large gap to T_c ratio $2\Delta_2/k_BT_c$ decreases with increasing Rb content x. On the other hand, for the small gap opening on the α and γ bands, the ratio $2\Delta_1/k_BT_c$ is practically independent of x. In addition, the contribution of the small gap ω_1 to the total superfluid density increases with increasing x. These results may be interpreted by assuming a disappearance of the electron pocket from the Fermi surface upon the high hole doping, resulting in a suppression of the scattering processes between the α and γ bands. This might cause the reduction of T_c for the overdoped RbFe₂As₂. We also performed zero-field μSR experiments and found no evidence of either static or fluctuating magnetism, implying that the spin-density wave ordering of the Fe moments is completely suppressed upon Rb doping. The absence of field induced magnetism in the investigated compounds is also demonstrated. Finally, the correlation between T_c and the zero-temperature relaxation rate $\sigma(0) \propto 1/\lambda^2(0)$ is discussed for the samples $Ba_{1-x}Rb_xFe_2As_2$ (x = 0.3, 0.35, 0.4, 1) using the Uemura classification scheme.

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