Muon-spin rotation measurements of the magnetic penetration depth in the iron-based superconductor Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$

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(Received 13 June 2011; published 14 September 2011)

Measurements of the magnetic penetration depth $\lambda$ in the Fe-based superconductor Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ ($x = 0.3, 0.35, 0.4$) were carried out using the muon-spin rotation ($\mu$SR) technique. The temperature dependence of $\lambda$ 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 RbFe$_2$As$_2$, a decrease of the BCS ratio $2\Delta_1/k_BT_c$ 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 $\Delta_1$ 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.

DOI: 10.1103/PhysRevB.84.094513

I. INTRODUCTION

The discovery$^1$ 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 fluoride type [Fe$_2$Pn$_2$] 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 fluoride-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 Pb$_2$O$_2$ structure. With $Ln = Sm$ a critical temperature higher than 55 K was observed.$^2$

Superconductivity with $T_c = 38$ K was also found in the ternary systems AF$_2$Se$_2$ (Refs. 3 and 4) (122 compounds) adopting the tetragonal ThCr$_2$Si$_2$ 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 Fermi-surface 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$_x$Fe$_2$As$_2$ were obtained from the temperature dependence of the magnetic penetration depth $\lambda$ 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 $\lambda$ reflects the topology of the superconducting gap occurring in the density of states of the superconducting ground state. The $\mu$SR technique provides a powerful tool to measure $\lambda$ in type II superconductors.$^9$

As demonstrated in previous works,$^{4,10}$ the value of $T_c$ for hole-doped Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ 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$_x$Fe$_2$As$_2$ and compare the results with those obtained for RbFe$_2$As$_2$ (Ref. 10).

In this paper, we report on $\mu$SR studies of the temperature and field dependence of the magnetic penetration depth of optimally doped Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ ($x = 0.3, 0.35, 0.4$). We compare the present data with the previous results of overdoped RbFe$_2$As$_2$ (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$_x$Fe$_2$As$_2$ were prepared in evacuated quartz ampoules by a solid state reaction method. Fe$_2$As, 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 Fe$_2$As the terminal compounds BaFe$_2$As$_2$ and RbFe$_2$As$_2$ were synthesized at 950°C and 650°C, respectively. Finally, the samples of Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ with $x = 0.3, 0.35, 0.4$ were prepared from appropriate amounts of single-phase BaFe$_2$As$_2$ and RbFe$_2$As$_2$. 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) $\mu$SR experiments were performed at the πM3 beamline of the Paul Scherrer Institute (Villigen, Switzerland), using the general purpose instrument (GPS). The sample was mounted inside of a gas-flow $^4$He 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$_x$Fe$_2$As$_2$ ($x = 0.3, 0.4$) measured in an applied magnetic field of $\mu_0 H = 0.04$ 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$_2$As$_2$ 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 SDW state. Magnetism, if present in the samples, may enhance the muon depolarization rate and falsify the interpretation of the TF-μSR results. Therefore, we have carried out ZF-μSR experiments above and below $T_c$ to search for magnetism (static or fluctuating) in Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ ($x = 0.3, 0.35, 0.4$). 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, 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$_x$As$_2$ and SrFe$_{2−x}$Co$_x$As$_2$ field induced magnetism exists. In the present work TF-μSR 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

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{(Color online) Transverse field (TF) μSR time spectra obtained in $\mu_0 H = 0.04$ T above and below $T_c$ (after field cooling the sample from above $T_c$): (a) Ba$_{0.7}$Rb$_{0.3}$Fe$_2$As$_2$ and (b) Ba$_{0.6}$Rb$_{0.4}$Fe$_2$As$_2$. The solid and the dashed lines represent fits to the data by means of Eq. (1).}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2.png}
\caption{(Color online) (a) ZF-μSR time spectra for Ba$_{0.7}$Rb$_{0.3}$Fe$_2$As$_2$ 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 $\mu H_{\text{int,SC}}$ measured in the SC state and the one measured in the normal state $\mu H_{\text{int,NS}}$ at $T = 42$ K.}
\end{figure}
Here $A$ denotes the initial asymmetry, $\gamma/(2\pi) \approx 135.5$ MHz/$T$ is the muon gyromagnetic ratio, and $\varphi$ is the initial phase of the muon-spin ensemble. $B_{\text{int}}$ represents the internal magnetic field at the muon site, and the relaxation rates $\sigma_{mc}$ and $\sigma_{\text{mm}}$ 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 $\sigma_{\text{mm}}$ was assumed to be constant over the entire temperature range and was fixed to the value obtained above $T_c$ where only nuclear magnetic moments contribute to the muon depolarization rate $\sigma$. As indicated by the solid lines in Fig. 1, the $\mu$SR data are well described by Eq. (1). The temperature dependence of $\sigma_{mc}$ for Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ ($x = 0.3, 0.35, 0.4$) at $\mu_0 H = 0.04$ T is shown in Fig. 3(a). Below $T_c$ the relaxation rate $\sigma_{sc}$ 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 $\sigma_{mc}(T)$ by the equation

$$\frac{\sigma_{sc}^2(T)}{\gamma^2} = \frac{\Phi_0^2}{\lambda^4(T)},$$

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 $\lambda$. In this case, according to the London model $\sigma_{mc}$ is field independent.\(^20\) We measured $\sigma_{sc}$ 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, $\sigma_{mc}$ strongly increases with increasing magnetic field until reaching a maximum at $\mu_0 H \approx 0.03$ 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 $\sigma_{mc}$ 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:\(^20,22\)

$$\lambda^{-2}(T, \Delta_{0,1}) = 1 + \frac{1}{\pi} \int_0^{\Delta_{0,1}} \int_0^{\infty} \frac{\partial^2 f}{\partial E \partial \varphi} \frac{E dE d\varphi}{\sqrt{E^2 - \Delta_c(T, \varphi)^2}},$$

where $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function, $\varphi$ is the angle along the Fermi surface, and $\Delta_c(T, \varphi) = \Delta_0 \delta(T/T_c) g(\varphi) (\Delta_0 = \max gap value at T = 0)$. 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 $s + i s$-wave gap, and $|\cos(2\theta)|$ for a $d$-wave gap.\(^24\)

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 $\alpha$ 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 $\alpha$ model, the superfluid density is calculated

\[ P(t) = A \exp \left[ -\left( \frac{\sigma_{mc}^2 + \sigma_{mm}^2}{2} \right) t \right] \cos(\gamma t B_{\text{int}} + \varphi). \]
for each component using Eq. (3) and then the contributions from the two components added together

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

(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 $\omega_i$ is a weighting factor which measures their relative contributions to $\lambda^{-2}$ ($\omega_1 + \omega_2 = 1$).

The results of the analysis for Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ ($x = 0.3, 0.35, 0.4$) 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$_x$Fe$_2$As$_2$ ($x = 0.3, 0.35, 0.4$). 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 $\Delta_1$ and a large gap $\Delta_2$, 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.

The magnitudes of the large and the small gaps for Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ ($x = 0.3, 0.35, 0.4$) (see Table I) are in good agreement with the results of a previous report. There it was pointed out that most Fe-based HTS’s exhibit two-gap superconducting behavior, characterized by a large gap with $2\Delta/k_B T_c = 7(2)$ and a small one with $2\Delta/k_B T_c = 5(2)$. To reach a more complete view of the superconducting properties of Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$ as a function of the Rb composition (hole doping), we combined the present data with the previous $\mu$SR results on RbFe$_2$As$_2$ (Ref. 10) which presents the case of a naturally overdoped system. Figure 5 shows the small gap to $T_c$, ratio $2\Delta_2/k_B T_c$, the large gap to $T_c$ ratio $2\Delta_2/k_B T_c$, and the weight $\omega_1$ of the small gap to the superfluid density as a function of Rb concentration. The data for RbFe$_2$As$_2$ are taken from Ref. 10. Interestingly, the ratio $2\Delta_2/k_B T_c$ decreases with increasing $x$. On the other hand, the ratio $2\Delta_1/k_B T_c$ for the small gap is essentially independent of $x$. In addition, the weighting factor $\omega_1$ is found to increase with increasing $x$. We note that in the optimally doped 122-system Ba$_{1-x}$K$_x$Fe$_2$As$_2$ several bands cross the Fermi surface (FS). They consist of inner ($\alpha$) and outer ($\beta$) hole-like bands, both centered at the zone center $\Gamma$, and an electron-like band ($\gamma$) centered at the $M$ point. The superconducting gap opened on the $\beta$ band was found to be smaller than those on the $\alpha$ and $\gamma$ bands. It was proposed that the enhanced interband scattering between the $\alpha$ and $\gamma$ bands might promote the kinetic process of pair scattering between these two FSs, leading to an increase of the pairing amplitude. 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 $\alpha$ and $\gamma$ bands would diminish since the $\gamma$ band is in the unoccupied side and concomitantly the size of the $\alpha$ band is increased. According to ARPES results, a decrease of interband scattering will lead to a decrease of the pairing amplitude and the ratio $2\Delta/k_B T_c$ 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.

One of the most interesting results of $\mu$SR 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). 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

![Figure 4](image-url)

**FIG. 4.** (Color online) The temperature dependence of $\lambda^{-2}$ for Ba$_{1-x}$Rb$_x$Fe$_2$As$_2$, 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 lines represent a fit using a two-gap ($s + s'$)-wave model.
FIG. 6. (Color online) Uemura plot for hole- and electron-doped high \( T_c \) 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 \( \text{Ba}_{1-x}\text{Rb}_x\text{Fe}_2\text{As}_2 \) (\( x = 0.3, 0.35, 0.4 \)) obtained in this work. The point for RbFe\(_2\)As\(_2\) is taken from Ref. 10.

IV. SUMMARY AND CONCLUSION

In summary, we performed transverse-field \( \mu \)SR measurements of the magnetic penetration depth \( \lambda \) on polycrystalline samples of the iron-based HTS’s \( \text{Ba}_{1-x}\text{Rb}_x\text{Fe}_2\text{As}_2 \) (\( x = 0.3, 0.35, 0.4 \)). The values of the superconducting transition temperature \( T_c \) and the zero temperature values of \( \lambda \) 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 \( \lambda \) is well described by a two-gap \( s + s \)-wave scenario with gap values similar to \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) (Refs. 5 and 8). ARPES investigations of \( \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \) revealed that the large gap opens on the inner hole-like Fermi surface (\( \alpha \) band) centered at the \( \Gamma \) point and on the electron-like FS (\( \gamma \) band) centered at the \( M \) point (tetragonal structure notations), while the small gap opens on the outer hole-like band (\( \beta \) of the \( \Gamma \) point).\(^{30}\) We found that the large gap to \( T_c \) ratio \( 2\Delta_2/k_B T_c \) decreases with increasing Rb content \( x \). On the other hand, for the small gap opening on the \( \alpha \) and \( \gamma \) bands, the ratio \( 2\Delta_2/k_B T_c \) of the \( \Gamma \) point is practically independent of \( x \). In addition, the contribution of the small gap \( \sigma_0 \) 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 \( \alpha \) and \( \gamma \) bands. This might cause the reduction of \( T_c \) for the overdoped RbFe\(_2\)As\(_2\). We also performed zero-field \( \mu \)SR 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 \( \text{Ba}_{1-x}\text{Rb}_x\text{Fe}_2\text{As}_2 \) (\( x = 0.3, 0.35, 0.4, 1 \)) using the Uemura classification scheme.
ACKNOWLEDGMENTS

Part of this work was performed at the Swiss Muon Source, Paul Scherrer Institut, Villigen, Switzerland. This work was supported by the Swiss National Science Foundation, the Scopus Grant No. IZ73Z0_128242, the NCCR Project MaNEP, the EU Project CoMePhS, and the Georgian National Science Foundation Grant No. GNSF/ST08/4-416.


