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Nonexponential London penetration depth of external magnetic fields in superconducting $Ba_{1-x}K_xFe_2As_2$ single crystals

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We have studied the in- and out-of-plane magnetic penetration depths in the hole-doped iron-based superconductor $Ba_{1-x}K_xFe_2As_2$ ($T_c \approx 30$ K). Single crystals grown from different fluxes and by different groups showed nearly identical results. The in-plane London penetration depth λ_{ab} is not exponentially saturating at low temperature, as would be expected from a fully gapped superconductor. Instead, $\lambda_{ab}(T)$ shows a power-law behavior, $\lambda \propto T^n$ ($n \approx 2$), down to $T \approx 0.02T_c$, similar to the electron-doped $Ba(Fe_{1-x}Co_x)_2As_2$. The penetration depth anisotropy $\gamma_\lambda = \lambda_c(T)/\lambda_{ab}(T)$ increases upon cooling, opposite to the trend observed in the anisotropy of the upper critical field, $\gamma_\xi = H_{c2}^{\perp c}(0)/H_{c2}^{\parallel c}(0)$. These are universal characteristics of both the electron- and hole-doped 122 systems, suggesting unconventional multigap superconductivity. The behavior of the in-plane superfluid density $\rho_{ab}(T)$ is discussed in light of existing theoretical models proposed for the iron pnictide superconductors.

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The discovery of superconductivity ($T_c \simeq 26$ K) in fluorine-doped LaFeAsO ("1111") (Ref. 1) has generated remarkable interest in the community. In a short time, the critical temperature was increased by pressure or chemical substitution above 55 K, which is significantly larger than the highest T_c reported in any *s*-wave superconductor, i.e., MgB_2, and comparable to those of the cuprates. Later, superconductivity, with T_c as high as 38 K, was discovered in $Ba_{1-x}K_xFe_2As_2$.[2] This BaK-122 compound is particularly important because, unlike the cuprates or the 1111 iron pnictides, it is not an oxide, downplaying the role of oxygen in this type of high-temperature superconductors. Moreover, large, high quality single crystals of both electron- and hole-doped 122 pnictides were synthesized,[3–6] which is essential for drawing reliable conclusions regarding their physical properties.

One key feature for understanding the origin of the high critical temperature and the pairing mechanism in pnictide superconductors is the symmetry of the order parameter. Phase diagrams of electron-[4] and hole-doped[7] 122 show that superconductivity emerges through doping, suppressing the orthorhombic/antiferromagnetic (AF) ground state in the parent compound. Close proximity to a magnetic state could imply the importance of magnetic fluctuations for pairing and may be reflected in the symmetry of the superconducting gap. At the same time, band-structure calculations and angle resolved photoemission spectroscopy (ARPES) experiments[8,9] show that multiple bands cross the Fermi level, opening the possibility for multiband superconductivity. Several ARPES studies[10–12] on $Ba_{1-x}K_xFe_2As_2$ have found at least two different superconducting gaps without nodes in the *ab* plane. Point-contact spectroscopy[13] and specific-heat[14] data also suggest fully gapped superconductivity and possibly two gaps. However, μ-SR,[15] NMR,[16] and magnetization studies[17] leave open the possibility for a nodal gap. Regarding penetration depth experiments, extensive work on the electron-doped 122 via Co substitution (FeCo-122) (Refs. 18 and 19) showed that at low temperatures, penetration features a robust power-law behavior $\Delta\lambda(T) \propto T^n$, with n being between 2 and 2.5, depending on the doping level. However, recent measurements on $Ba_{1-x}K_xFe_2As_2$, using microwave cavity perturbation technique, claim exponential saturation of the penetration depth at low temperatures.[20]

In this Rapid Communication, we show that contrary to an earlier report,[20] the London penetration $\lambda_{ab}(T)$ in the hole-doped 122 compound, $Ba_{1-x}K_xFe_2As_2$, is not exponential down to at least $0.02T_c$. Instead, similar to Co-doped 122, $\lambda_{ab}(T)$ has a nearly quadratic temperature dependence. To draw more reliable and objective conclusions, single crystals of $Ba_{1-x}K_xFe_2As_2$ with $T_c \approx 30$ K, grown by two different groups, using different fluxes, were studied. A total of six samples was measured, giving highly reproducible results. We show the data for two of them. The crystal labeled *A*, with dimensions of $740 \times 800 \times 70$ μm^3 was grown out of Sn flux. It comes from one of the batches characterized in Ref. 5. Based on elemental analysis from Ref. 5, sample A corresponds to optimal K doping with $x=0.45$, i.e., $Ba_{0.55}K_{0.45}Fe_2As_2$. Sample B, with dimensions of $320 \times 830 \times 70$ μm^3, was grown from self-flux of FeAs and it was characterized in Ref. 6. According to Ref. 6, sample B corresponds to $x=0.3$, i.e., $Ba_{0.7}K_{0.3}Fe_2As_2$.

The penetration depth was measured using a tunnel diode resonator (TDR) technique[21] by inserting the sample into the inductor of a self-resonating tank circuit powered by a tunnel diode. The resonant frequency of the empty resonator was $f_0 \approx 14$ MHz, with a stability better than 5 ppb/hour. The rf magnetic field produced by the inductor was $H_{rf} \sim 10$ mOe, much less than typical values for the lower critical field $H_{c1} \sim 50$ Oe in iron-arsenides. Therefore, when a superconducting sample is placed inside the inductor, the inductance of the coil changes due to Meissner screening, which leads to a change in the resonant frequency. The relative frequency

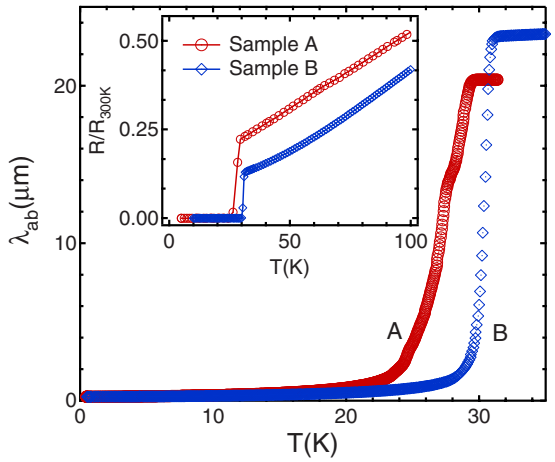

FIG. 1. (Color online) $\lambda_{ab}(T)$ for sample A, grown from Sn flux (Ref. 5), and for sample B, grown from FeAs self-flux (Ref. 6). Inset: temperature dependence of resistivity for each sample, normalized to the room-temperature value.

change is directly proportional to the penetration depth, $\Delta f = -G\Delta\lambda$.[22] The calibration constant G was determined both by following a calibration procedure described in Refs. 22 and 23 and from *in situ* extraction of the sample from the coil, with both methods yielding identical results. First, we mounted the sample with the c-axis along the rf magnetic field ($H_{rf} \| c$). In this geometry, only the in-plane screening currents were induced, and therefore $\Delta f \propto \Delta\lambda_{ab}$. Then, the sample was aligned with $H_{rf} \perp c$. For this geometry, screening currents flow both in the ab plane and along the c direction and $\Delta\lambda_c$ could be obtained in our relatively thick samples using the numerical model described in Ref. 23.

Figure 1 shows $\lambda_{ab}(T)$ (main figure) and resistivity (inset) for samples A and B. We notice that although they correspond to different potassium concentrations, both samples have very similar values for the onset T_c. The widths of the transition at T_c are significantly different, with $\Delta T \leq 3$ K for sample A and $\Delta T \leq 1.5$ K for sample B. Moreover, despite the fact that optimal doping corresponds to $x=0.45$ K for both methods, the maximum T_c reached by using Sn flux is about 30 K,[5] whereas by using FeAs flux it is 38 K.[6] A possible explanation is that some amount of Sn is incorporated in the crystal structure of sample A, as explained in Ref. 5.

Figure 2 shows the low-temperature region of $\lambda_{ab}(T)$ for samples A and B. Despite the differences in potassium content and transition width at T_c, the penetration depth behavior is similar below $0.3T_c$, exhibiting a power law $\lambda_{ab}(T) = \lambda_0 + bT^n$, with the average value of the exponent being $n \approx 1.9 \pm 0.4$. It is important to note that this nearly quadratic temperature dependence of the penetration depth is similar to previous observations in the electron-doped 122.[18,19]

The inset to Fig. 2 shows $\Delta\lambda_{ab}$ plotted against $(T/T_c)^2$ for sample B and for a $Ba(Fe_{1-x}Co_x)_2As_2$ single crystal ($x=0.074$),[18] corresponding to optimal doping. It can be clearly observed that at low temperatures, both traces are well fit by straight lines with similar slopes. This nearly identical behavior suggests that there are common electrodynamic properties within the 122 pnictide superconductors.

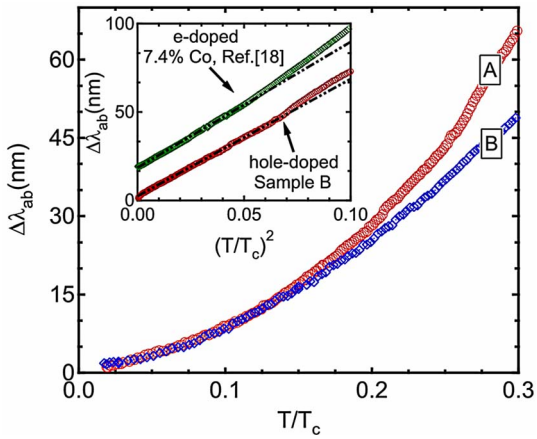

FIG. 2. (Color online) Low-temperature region of $\lambda_{ab}(T)$ for sample A and sample B, respectively. Inset: $\Delta\lambda_{ab}$ vs $(T/T_c)^2$ for hole-doped (sample B from present work) and electron-doped ($x=0.074$ Co from Ref. 18) FeAs-122 (shifted vertically for clarity). Dashed lines represent linear fits.

At this point we defer the discussion of possible symmetries for the superconducting gap consistent with the observed behavior and turn our attention to the anisotropy of penetration depth $\gamma_\lambda = \lambda_c(T)/\lambda_{ab}(T)$. Applying the rf magnetic field both parallel to the c axis and along the ab plane, we were able to directly subtract the change in penetration depth associated with screening currents flowing in the c direction, i.e., $\Delta\lambda_c$. Since we are not aware of independent estimates of λ_c at any temperature, we have used the approach of Ref. 24. High magnetic field experiments on single crystals of $Ba_{1-x}K_xFe_2As_2$ (Refs. 5 and 25) found that $\gamma_\xi = H_{c2}^{ab}(0)/H_{c2}^{c}(0) \approx 3.5$ near T_c. Based on the validity of the Ginzburg-Landau theory in the vicinity of T_c, we consider $\gamma_\xi = \gamma_\lambda \approx 3.5$ at $T=0.9T_c$. Figure 3 shows the resulting $\lambda_c(T)$ on the same graph with $\lambda_{ab}(T)$. Because λ_c has a weaker temperature dependence at low temperatures, it extrapolates

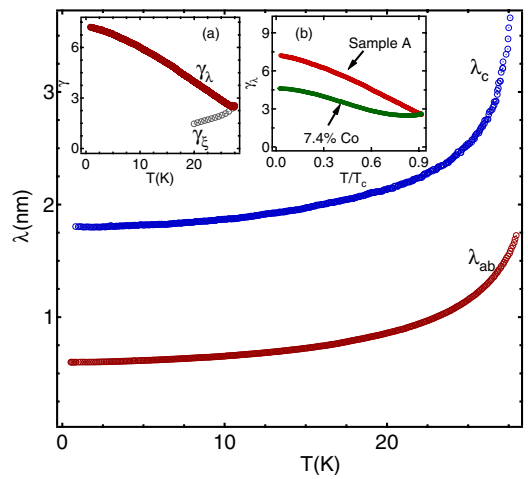

FIG. 3. (Color online) In-plane $\lambda_{ab}(T)$ and out-of-plane $\lambda_c(T)$ penetration depths for sample B. Inset (a): variation in temperature of $\gamma_\lambda = \lambda_c/\lambda_{ab}$ obtained by dividing the values from the main panel and $\gamma_\xi(T)$ from Ref. 25. Inset (b): γ_λ for hole-(current work) and electron-doped (Ref. 24) FeAs-122.

to a significantly higher value at $T=0$, $\lambda_c(0)\approx 1800$ nm. Therefore, we obtain a penetration depth anisotropy $\gamma_\lambda \approx 7$ near $T=0$, which is about twice the value of $\gamma_\lambda(T_c)$. In inset (b) of Fig. 3, we show $\gamma_\lambda(T)$ from the present work together with the result for the electron-doped $Ba(Fe_{0.926}Co_{0.074})_2As_2$ from Ref. 24. In both cases, γ_λ decreases with temperature, although at different rates, which emphasizes again the similarity between the hole-and electron-doped 122 superconductors.

We now compare the temperature dependence of the penetration depth anisotropy $\gamma_\lambda(T)$ with that of the upper critical field $\gamma_\xi(T)$. In pnictide superconductors, $H_{c2}(0)$ may reach $\sim 100T$, making it difficult to estimate γ_ξ over the entire temperature range.[25] Nevertheless, it has been found that γ_ξ decreases from ≈ 3.5 near T_c to ≈ 1.4 at $0.5T_c$,[25] and we display both $\gamma_\xi(T)$ and $\gamma_\lambda(T)$ in inset (a) of Fig. 3. We suggest that the difference in the temperature dependence of the anisotropies is consistent with the existence of two gaps of different magnitude in 122 superconductors, such as previously observed from ARPES data.[10] In a clean superconductor at $T=0$, $\gamma_\lambda^2(0)=\langle v_a^2\rangle/\langle v_c^2\rangle$, where v_a and v_c are the Fermi velocities along the a and c directions. The averaging is performed over the whole Fermi surface. However, at $T=T_c$, $\gamma_\lambda^2(T_c)=\langle \Delta^2 v_a^2\rangle/\langle \Delta^2 v_c^2\rangle$, where Δ is the superconducting gap. For two gaps with different values, the main contribution to $\gamma_\lambda(T_c)$ is given by the regions of the Fermi surface with a larger gap. If the larger gap exists at a less anisotropic Fermi surface, it will result in $\gamma_\lambda(T_c) < \gamma_\lambda(0)$.

Considering $\lambda_{ab}(0)=180$ nm determined experimentally from optical spectroscopy,[26] Fig. 4(a) shows the in-plane superfluid density obtained for sample B. It can be observed that $\rho_{ab}(T)$ has positive curvature at intermediate temperature and is strongly suppressed close to T_c. Comparing with the experimental result on $Ba(Fe_{1-x}Co_x)_2As_2$ from Ref. 18, we see that superfluid density also behaves similarly in electron- and hole-doped 122 superconductors.

Therefore, the experimental results shown above indicate that any proposed symmetry for the order parameter in FeAs-122 superconductors has to explain at least three particularities of penetration depth: the nearly quadratic temperature dependence at low temperature, the increase in penetration depth anisotropy upon cooling, and the strong suppression of superfluid density close to T_c. Although the temperature dependence of γ_λ is reminiscent of MgB_2, we argue that, unlike in MgB_2,[27] a simple two s-wave gap model cannot correctly describe the superfluid density $\rho_{ab}(T)$ in 122 systems. The continuous line in Fig. 4(a) shows the result from a fit considering two independent s-wave gaps $\Delta_1(T)$ and $\Delta_2(T)$, along with their relative contributions ϵ and $(1-\epsilon)$, respectively: $\rho(T)=\epsilon\rho(\Delta_1)+(1-\epsilon)\rho(\Delta_2)$. Even though the quality of the fit looks acceptable at intermediate temperatures, the model clearly fails to reproduce the low-temperature data, as illustrated in the inset of Fig. 4(a). Moreover, the parameters obtained from the fit, $\Delta_1(0)\approx 0.95k_BT_c$, $\Delta_2(0)\approx 0.25k_BT_c$, and $\epsilon\approx 0.8$, are physically unreasonable. Both gaps are smaller than the BCS weak coupling limit of $1.76k_BT_c$, and they are much smaller than those previously reported from ARPES.[10,11] These two gaps would reproduce measured T_c only if interband coupling is strong, in which case our fitting model of independent gaps is not applicable.[28]

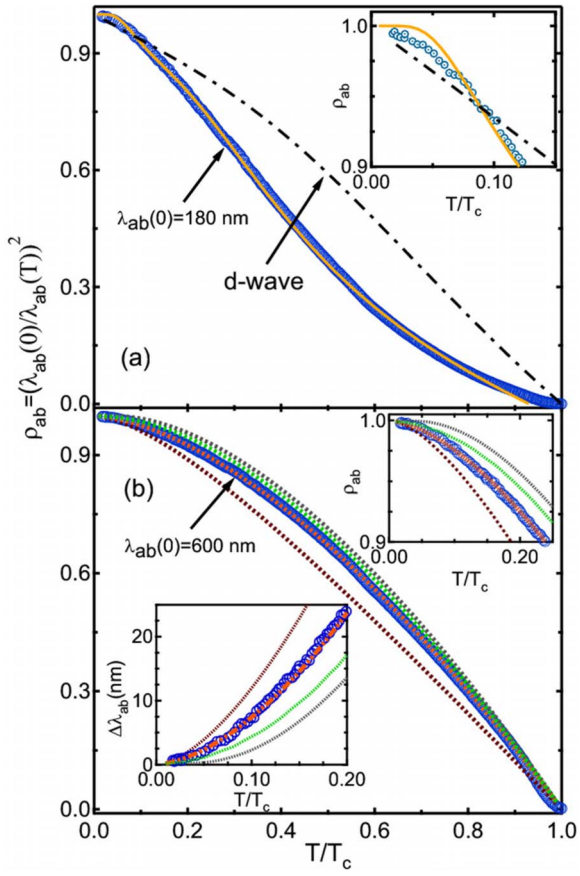

FIG. 4. (Color online) (a) $\rho_{ab}(T)$ for $\lambda_{ab}(0)=180$ nm (symbols), fit with a two-gap model (continuous line), as explained in the text and theoretical calculation for d-wave gap symmetry (dashed lines). Inset: zoom on the low-temperature region. (b) $\rho_{ab}(T)$ for $\lambda_{ab}(0)=600$ nm (symbols) and our theoretical calculations based on the model from Ref. 31 for the following values of dimensionless impurity scattering parameters $(\Gamma_0/2\pi T_{c0}, \Gamma_\pi/2\pi T_{c0})$ (top to bottom): (0,0.064), (3, 0.064), (3,0.068), and (3,0.060), where T_{c0} is the critical temperature in the absence of impurities. Upper inset: the lowest-temperature region showing good agreement between experiment and theory. Lower inset: low-temperature region of $\lambda_{ab}(T)$ for sample B and the theoretical results for the same parameters as in the main panel.

The quadratic temperature dependence of the penetration depth at low temperature is expected for a superconducting gap with line nodes, i.e., d-wave symmetry, in the presence of strong (unitary) impurities, which will create an additional quasiparticle density of states.[29] This deviation from otherwise linear behavior of $\lambda_{ab}(T)$ predicted for a d-wave gap symmetry, is expected to occur only below a characteristic temperature T^*, given by impurity concentration. However, as it can be observed from Fig. 4(a), $\rho_{ab}(T)$ for $Ba_{1-x}K_xFe_2As_2$ is suppressed much stronger than expected for the d-wave symmetry at intermediate temperatures. Therefore, while we cannot rule out the possibility of a superconducting gap with nodes, a single d-wave gap cannot entirely explain our experimental findings.

For iron pnictides, it is gaining support the idea that antiferromagnetic spin fluctuations mediate *interband* pairing. In this case, the order parameter is fully gapped but changes

sign between different Fermi sheets. This situation is referred to as the extended $s^{\pm}$ symmetry.[30] In the lower inset of Fig. 4(b) we plot the low-temperature region of $\Delta\lambda_{ab}(T)$ for sample B together with our calculations considering the extended $s^{\pm}$ symmetry ($\Delta \propto \cos k_x + \cos k_y$) in the presence of nonmagnetic impurities for several values of the intraband (Γ_0) and interband (Γ_π) scattering rates.[31] Details of these calculations are given in Ref. 31, where this model was used for the Co-doped 122 system. With $\lambda_{ab}(0)$ as a fitting parameter, it can be seen that we were able to correctly reproduce the experimental data down to the lowest measured temperature. However, the fit yields $\lambda_{ab}(0) \approx 600$ nm, which is much larger that the experimental estimate. Nevertheless, if we use $\lambda_{ab}(0)=600$ nm in our calibration procedure, Fig. 4(b) shows that there is very good agreement between the theoretical calculation and the experimental result for the superfluid density up to T_c. Also the low-temperature region for $\rho_{ab}(T)$ is correctly reproduced by this model as emphasized in the upper inset of Fig. 4(b). We propose that further theoretical development to include the possibility of two different gaps and/or anisotropy or even a nodal gap[32,33] may be able to reconcile the discrepancy between the experimental and theoretical results for $\lambda_{ab}(0)$ and to capture all experimental results from the present work.[33]

In conclusion we have found that the hole-doped $Ba_{1-x}K_xFe_2As_2$ single crystals exhibit remarkable similarities with the electron-doped FeAs-122 superconductor, such as nearly quadratic temperature dependence of $\lambda(T)$, the anisotropy $\gamma_\lambda(T)$ increasing upon cooling and strong suppression of the superfluid density at elevated temperatures.

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