Competition between superconductivity and magnetic/nematic order as a source of anisotropic superconducting gap in underdoped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$

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The in-plane London penetration depth, $\Delta\lambda(T)$, was measured using a tunnel diode resonator technique in single crystals of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with doping levels $x$ ranging from heavily underdoped, $x=0.16$ ($T_c=7$ K) to nearly optimally doped, $x=0.34$ ($T_c=39$ K). Exponential saturation of $\Delta\lambda(T)$ in the $T \to 0$ limit is found in optimally doped samples, with the superfluid density $\rho_s(T) \equiv (\lambda(0)/\lambda(T))^2$ quantitatively described by a self-consistent $\gamma$-model with two nodeless isotropic superconducting gaps. As the doping level is decreased towards the extreme end of the superconducting dome at $x=0.16$, the low-temperature behavior of $\Delta\lambda(T)$ becomes non-exponential and best described by the power-law $\Delta\lambda(T) \propto T^2$, characteristic of strongly anisotropic gaps. The change between the two regimes happens within the range of coexisting magnetic/nematic order and superconductivity, $x < 0.25$, and is accompanied by a rapid rise in the absolute value of $\Delta\lambda(T)$ with underdoping. This effect, characteristic of the competition between superconductivity and other ordered states, is very similar to but of significantly smaller magnitude than what is observed in the electron-doped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ compounds. Our study suggests that the competition between superconductivity and magnetic/nematic order in hole-doped compounds is weaker than in electron-doped compounds, and that the anisotropy of the superconducting state in the underdoped iron pnictides is a consequence of the anisotropic changes in the pairing interaction and in the gap function promoted by both magnetic and nematic long-range order.

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INTRODUCTION

The experimental determination of the symmetry of the superconducting gap is important to unravel the mechanism of superconductivity in iron-based superconductors [1,3]. Measurements of the London penetration depth [4,5], thermal conductivity [7,8] and specific heat [9,11] in electron doped $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ (BaCo122) suggest that the superconducting gap changes significantly with doping, developing nodes at both overdoped and underdoped dome edges [8,12,13]. This doping evolution is very similar to what is observed in another electron-doped family, NaFe$_{1-x}$Co$_x$As [14,15] and LiFeAs [16,19]. It is also consistent with the predicted dependence of the gap function with doping in the $s^+$ model [13] [20]. On the other hand, nodal behavior is observed at all doping levels in isovalent-substituted BaFe$_2$(As$_{1-x}$P)$_x$ (BaP122) [21]. This remarkable contrast in two systems that share the same parent compound prompts a detailed study of the hole doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ (BaK122) materials.

In BaK122, a full isotropic gap has been reported in compositions close to optimal [22,28], whereas strongly overdoped compositions with $x \approx 1$ display nodal superconductivity [25,32]. Although these observations suggest a similar trend as compared to the electron-doped BaCo122 materials [33], there has been no systematic studies of the superconducting gap structure in the underdoped BaK122 so far. In this doping regime, superconductivity coexists and competes with long-range magnetic/nematic order [20,34], making this an ideal system to investigate the rich interplay between these ordered states [35,36]. Interestingly, a sizeable asymmetry between the normal state properties of the electron- and hole-doped materials is observed in such underdoped regime [37,39].

In this work we study the evolution of the temperature dependence of the in-plane London penetration depth, $\Delta\lambda(T)$, in high quality single crystals of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ across the underdoped region of the phase diagram $0.16 \leq x \leq 0.34$. We find that the optimally doped samples show a weak exponential temperature dependence in the $T \to 0$ limit, suggesting nodeless isotropic gaps. This conclusion is consistent with the temperature dependence of the superfluid density, which can be well fitted using the self-consistent $\gamma$-model with two full gaps in the clean limit [40]. In contrast, the lowest-$T_c$ samples, deep in the underdoped regime, show a strong power-law temperature dependence of $\Delta\lambda(T)$ in the low-$T$ limit, typical of strongly anisotropic gaps. The onset of this behavior co-
incides with the onset of the coexistence between the superconducting and magnetic/nematic phases, indicating that the anisotropic changes in the pairing interaction arising from this coexistence play a major role in determining the gap structure in the underdoped regime. The magnitude of $\Delta \lambda(0.25T_c)$, which serves as a proxy of the magnitude of the zero-temperature penetration depth, shows a rapid rise in the range of coexisting magnetic/nematic order and superconductivity. Comparison with the penetration depth data in the electron-doped counterpart Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ reveals that the rapid rise of $\Delta \lambda$ in the coexistence state is similar in both materials, as expected for competing ordered states $\text{[41–44]}$. However, the increase in $\Delta \lambda$ is almost three times larger in electron-doped materials, suggesting that the competition between magnetic/nematic order and superconductivity is weaker in the hole-doped materials. Interestingly, this electron-hole asymmetry inside the superconducting state correlates with the asymmetric behavior of the normal state properties, in particular the nematic susceptibility, as measured by the in-plane resistivity anisotropy $\text{[37]}$ and by elastic constant measurements $\text{[45]}$, and pseudogap features in the inter-plane resistivity anisotropy $\text{[37]}$ and by elastic constant measurements $\text{[37]}$ and by elastic constant measurements $\text{[45]}$.

**RESULTS AND DISCUSSION**

In Fig. 1(a) we show the variation of the London penetration depth $\Delta \lambda(T)$ from the base temperature to $T_c$ for five compositions of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ spanning from $x=0.16$ ($T_c=7$ K, edge of the superconducting dome) to $x=0.34$ ($T_c=39$ K, nearly optimally doped). The data are normalized by $\Delta \lambda(T_c)$ and reveal the high quality of our single crystals as evidenced by the sharpness of the superconducting transitions and the absence of any additional features. In Fig. 1(b) we present the same data plotting the actual $\Delta \lambda(T)$ as function of the reduced temperature $T/T_c$ for $0<T/T_c<0.3$. This is the characteristic temperature range in which the superconducting gap of single-band superconductors can be considered constant, and in which the temperature dependence of $\Delta \lambda(T)$ reflects the nodal structure of the gap. In the clean limit, $\Delta \lambda(T)$ is expected to depend exponen-
The estimated gap values in the $T \to 0$ limit are 6.5 meV and 3.3 meV, the larger gap being in reasonable agreement with the value of ~6 meV found from specific heat measurements \[54\]. This analysis implies that the small superconducting gap, $\Delta_2$, is strongly temperature dependent even for $T < 0.3 T_c$, and the characteristic behavior can be found only at temperatures at least two times lower than $0.3 T_c$. In Fig. 1(c) we show the data over the temperature range $0 < T/T_c \leq 0.1$, presented as a power-law function $T^n$ of the reduced temperature, with exponent $n$ as shown in the main panel.

In Fig. 3 we summarize the doping evolution of the London penetration depth as found in our study. For reference in the top panel Fig. 3(a) we show the doping phase diagram as determined from our TDR and resistivity measurements \[47\], which are in good agreement with the phase diagram determined from neutron scattering and magnetization data on polycrystalline samples of Avci et al. \[56\]. This analysis reveals two clear trends: (i) The exponent $n$ of the power-law temperature dependence of $\Delta \lambda$, Fig. 3(b), as determined from the data analysis for $0 < T/T_{up} \leq T_{up}=0.17 T_c$ and $T_{up}=0.15 T_c$, decreases from $n=3.5$ for $x=0.34$ (which is technically indistinguishable from an exponential dependence) to $n=2$ for $x=0.16$. (ii) The actual variation of the London penetration depth $\Delta \lambda(T_{up})$ with $T_{up}=0.3 T_c$ and $T_{up}=0.15 T_c$ – which mimics the doping dependence of the zero-temperature penetration depth – strongly increases in the same doping regime.

Both effects are more prominent for doping levels $x \leq 0.21$, where magnetism and nematicity are also present. Indeed, these trends can be understood theoretically as a result of the competition and coexistence between magnetic/nematic order and superconductivity. On the one hand, magnetism competes with superconductivity for the same electronic states \[20\] \[34\], which is most directly revealed by the suppression of the magnetic order parameter below $T_c$ seen in neutron scattering \[50\]. Due to its magnetic origin, nematic order inherits this competition and also competes with superconductivity \[37\] \[58\], as manifested by the decrease of the orthorhombic distortion – proportional to the nematic order parameter \[38\] \[58\] – below $T_c$, as measured by x-ray scattering \[56\]. The competition between these electronic ordered states results in a suppression of the zero-temperature superfluid density, and a consequent enhancement of the penetration depth in the low-temperature limit \[41\] \[44\], in qualitative agreement with the experimental data.

On the other hand, coexistence with magnetic/nematic order leads to strong anisotropies in the gap function, which is also in qualitative agreement with the experimental data. Consider for instance a simplified scenario...
in which the $s^{+-}$ gap function and the pairing interaction are completely isotropic at optimal doping. Because nematic order breaks the tetragonal symmetry of the system, it gives rise to a d-wave component in the original $s^{+-}$ gap function in the coexistence state [59, 60]. Due to the proximity between the d-wave and $s^{+-}$ ground state energies – as manifested by the existence of a Bardasis-Schrieffer mode in the Raman spectrum of the optimally doped samples [61] – this mixing between d-wave and $s^{+-}$ states can be sizeable, leading to strong anisotropies in the gap. Furthermore, long-range magnetic order promotes anisotropy in the pairing interaction itself [65]. Due to the anisotropic reconstruction of the Fermi surface caused by the doubling of the unit cell in the magnetic phase, the electronic states near the Fermi level acquire a significant angular dependence, which is translated to an effectively anisotropic pairing interaction for the states of the reconstructed Fermi surface. As a result, the gap nodes that were fully isotropic in the noncoexistence state develop deep minima, which may even give rise to nodal behavior [65].

It is instructive to compare these observations with the results on electron-doped BaCo122. In Fig. 4 we directly compare the doping phase diagrams (panel (a)) and doping evolutions of the London penetration depth (panel (b)) of the hole and electron-doped BaFe$_2$As$_2$ based superconductors. The data for BaCo122 were taken from Ref. [4]. To take into account the fact that the structural (i.e. nematic) and magnetic transition lines ($T_s$ and $T_m$, respectively) coincide in BaK122, but split with doping in BaCo122, we normalized the compositions of the samples to those with the lowest measured structural transition temperatures, $x=0.25$ in BaK122 and $x=0.063$ in BaCo122. This comparison reveals very interesting similarities and differences between these two families. First, as can be seen from Fig. 4 the $T_s(x)$ boundary in BaK122 terminates very sharply, suggesting a possible first order transition with doping between the orthorhombic/nematic and tetragonal phases. Second, the rapid increase of the London penetration depth $\Delta\lambda(T_{up})$ with underdoping, reflecting the increase of $\lambda(0)$ [62], has very different magnitudes for the two types of doping. For instance, despite using BaK122 samples that are much closer to the edge of the superconducting dome ($T_c=7$ K for $x=0.16$) than in our previous study of BaCo122 compounds ($T_c=7.4$ K for $x=0.038$) [62], we find a three times smaller increase of $\Delta\lambda(0.25T_c)$ on the hole-doped side compared to the electron-doped side. This suggests weaker competition between magnetic/nematic order and superconductivity in the hole-doped BaK122 than in the electron-doped BaCo122, which is in line with the significantly weaker suppression of the magnetic order parameter below $T_c$ found in neutron scattering experiments [54, 55]. Similarly, as shown in the inset of panel (b) in Fig. 4, the normalized resistivity in the normal state is much less affected by the long-range magnetic order in BaK122 than in BaCo122. This suggests that the partial gapping of the Fermi surface by long-range magnetic order is stronger in the latter case, leaving less electronic states available for the superconducting state. We note that the nematic susceptibility is also weaker in the BaK122 family, as evidenced by the in-plane resistivity anisotropy behavior [57] and the softening of the...
shear modulus \[45\]. Therefore, our findings suggest a close relationship between the electron-hole asymmetries of the normal state and superconducting properties. Despite displaying different magnitudes, however, the continuous increase of \(\Delta \lambda(0.25T_c)\) with underdoping in both electron- and hole-doped samples contrasts with the case of isovalent doping, BaP122, in which a sharp peak in the low-temperature penetration depth is observed \[63\].

**CONCLUSIONS**

In conclusion, our measurements of the London penetration depth in high quality single crystals of Ba\(_{1-x}\)K\(_x\)Fe\(_2\)As\(_2\) close to the optimal doping level \(x=0.34\) (\(T_c = 39\) K) reveal a superconducting state with two full gaps \(\Delta_1(0)=6.5\) meV and \(\Delta_2(0)=3.3\) meV. On the other hand, our measurements deep in the underdoped regime, for \(x=0.16\) (\(T_c = 7\) K), demonstrate that the gap develops significant anisotropies without, however, developing nodes. Comparison with the electron-doped compositions Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) reveals a strong asymmetry of the structure of the superconducting state, which is nodeless in hole-doped and nodal in electron-doped compounds. These observations suggest that the competition and the coexistence with magnetic/nematic order is responsible for the anisotropic structure of the superconducting gap in the underdoped regime, and that this competition is stronger in electron-doped rather than in hole-doped compounds.

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1. I. I. Mazin, Nature 464, 183 (2010).
2. F. Wang and D.-H. Lee, Science 332, 200 (2011).
3. P. J. Hirschfeld, M. M. Korshunov, and I. I. Mazin, Rep. Prog. Phys. 74, 124508 (2011); A. V. Chubukov, Annu. Rev. Cond. Mat. Phys. 3, 57 (2012).
4. R. T. Gordon C. Martin, H. Kim, N. Ni, M. A. Tanatar, J. Schmalian, I. I. Mazin, S. L. Bud’ko, P. C. Canfield, and R. Prozorov, Phys. Rev. B 79, 100506 (2009).
5. R. T. Gordon, N. Ni, C. Martin, M. A. Tanatar, M. D. Vannette, H. Kim, G. D. Samolyuk, J. Schmalian, S. Nandi, A. Kreyssig, A. I. Goldman, J. Q. Yan, S. L. Bud’ko, P. C. Canfield, and R. Prozorov., Phys. Rev. Lett. 102, 127004 (2009).
6. C. Martin, H. Kim, R. T. Gordon, N. Ni, V. G. Kogan, S. L. Bud’ko, P. C. Canfield, M. A. Tanatar, and R. Prozorov., Phys. Rev. B 81, 060505 (2010).
[45] A. E. Bohmer, P. Burger, F. Hardy, T. Wolf, P. Schweiss, R. Fromknecht, M. Reinecker, W. Schranz, and C. Meingast, Phys. Rev. Lett. 112, 047001 (2014).
[46] M. A. Tanatar, N. Ni, A. Thaler, S. L. Bud’ko, P. C. Canfield and R. Prozorov, Phys. Rev. B 82, 134528 (2010).
[47] M. A. Tanatar, W. E. Straszheim, H. Kim, J. Murphy, N. Spyrisoin, E. C. Blomberg, K. Cho, J.-P. Reid, B. Shen, L. Taillefer, H.-H. Wen and R. Prozorov, Phys. Rev. B 89, 144514 (2014).
[48] H. Q. Luo, Z. S. Wang, H. Yang, P. Cheng, X. Zhu, and H.-H. Wen, Supercond. Sci. Technol. 21, 125014 (2008).
[49] C. T. van Degrift, Rev. Sci. Instrum. 46, 599 (1975).
[50] R. Prozorov and R. W. Giannetta, Supercond. Sci. Technol. 19, R41 (2006).
[51] R. Prozorov, R. W. Giannetta, A. Carrington, and F. M. Araujo-Moreira, Phys. Rev. B 62, 115 (2000).
[52] R. Prozorov and V. G. Kogan, Rep. Prog. Phys. 74, 124505 (2011).
[53] G. Li, W. Z. Hu, J. Dong, Z. Li, P. Zheng, G. F. Chen, J. L. Luo, and N. L. Wang, Phys. Rev. Lett. 101, 107004 (2008).
[54] G. Mu, Huiqian Luo, Zhaosheng Wang, Lei Shan, Cong Ren, and Hai-Hu Wen, Phys. Rev. B 79, 174501 (2009).
[55] M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. 101, 107006 (2008).
[56] S. Avci, O. Chmaissem, E. A. Goremychkin, S. Rosenkranz, J.-P. Castellan, D. Y. Chung, I. S. Todorov, J. A. Schlueter, H. Claus, M. G. Kanatzidis, A. Daoud-Aladine, D. Khalyavin, and R. Osborn, Phys. Rev. B 83, 172503 (2011).
[57] E. G. Moon and S. Sachdev, Phys. Rev. B. 85, 184511 (2012).
[58] R. M. Fernandes, S. Maiti, P. Woelfle, and A. V. Chubukov, Phys. Rev. Lett. 111, 057001 (2013).
[59] R. M. Fernandes and A. J. Millis, Phys. Rev. Lett. 111, 127001 (2013).
[60] G. Livanas, A. Aperis, P. Kotetes, and G. Varelgiannis, arXiv:1208.2881
[61] F. Kretszschmar, B. Muschler, T. Bohm, A. Baum, R. Hackl, H.-H. Wen, V. Tsurkan, J. Deisenhofer, and A. Loidl, Phys. Rev. Lett. 110, 187002 (2013).
[62] R. T. Gordon, H. Kim, N. Salovich, R. W. Giannetta, R. M. Fernandes, V. G. Kogan, T. Prozorov, S. L. Bud’ko, P. C. Canfield, M. A. Tanatar, and R. Prozorov, Phys. Rev. B 82, 054507 (2010).
[63] K. Hashimoto, K. Cho, T. Shibauuchi, S. Kasahara, Y. Mizukami, R. Katsumata, Y. Tsuruhara, T. Terashima, H. Ikeda, M. A. Tanatar, H. Kitano, N. Salovich, R. W. Giannetta, P. Walmsley, A. Carrington, R. Prozorov, and Y. Matsuda, Science 336, 1554 (2012).