Point contact Andreev reflection spectroscopy of superconducting energy gaps in 122-type family of iron pnictides

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Abstract
A brief overview of the superconducting energy gap studies on 122-type family of iron pnictides is given. It seems that the situation in the hole-doped Ba_{1-x}K_xFe_2As_2 is well resolved. Most of the measurements including the presented here point-contact Andreev reflection spectra agree on existence of multiple nodeless gaps in the excitation spectrum of this multiband system. The gaps have basically two sizes – the small one with a strength up to the BCS weak coupling limit and the large one with a very strong coupling with 2\Delta/kT_c > 6 - 8. In the electron doped Ba(Fe_{1-x}Co_x)_2As_2 the most of the experiments including our point contact measurements reveal in quite broadened spectra only a single gap with a strong coupling strength. The high precision ARPES measurements on this system identified two gaps but very close to each other, both showing a strong coupling with 2\Delta/kT_c ~ 5 and 6, respectively.

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1. Introduction
Iron pnictides [1] – finally a new family of high-T_c superconductors - represent a real challenge in the recent condensed matter physics. Despite an enormous effort during less than one year after their appearance many puzzles remain unsolved. One of the important questions concerns the superconducting order parameter in these systems with a strongly multiband character. In this paper a brief overview of the experimental studies of the order parameter in the 122-type family of iron pnictides is given.

Similarly to the high-T_c cuprates the superconductivity in iron pnictides is enabled by chemical doping of the antiferromagnetic parental compounds which in contrast to the cuprates are metallic. The highest transition temperature (up to 56 K) among different iron pnictides has been achieved in the optimally doped REFeAsO(F), or the 1111 group with Gd, Nd, or Sm [2] standing for a rare earth RE. Considerable interest has also been attracted by another class of iron pnictide superconductors based on AFe_2As_2 with A = Ba, Sr and Ca, referred to as the 122-type group. The 122-type compounds are chemically and structurally simpler and less anisotropic than the 1111 ones. The maximum T_c of 38 K is obtained in the optimally hole doped Ba_{0.6}K_{0.4}Fe_2As_2 system [3] but also the electron doped Ba(Fe_{1-x}Co_x)_2As_2 crystals with T_c about 25 K are available [4]. In contrast to the 1111 systems the 122-type parent compounds show magnetic (from paramagnetic to SDW antiferromagnetic phase) and structural transition (from tetragonal to orthorhombic phase) at the same temperature of about 140 K. This transition is
2. Overview of the superconducting gap studies

High resolution angle resolved photoemission spectroscopy (ARPES) has become extremely effective tool for studies of the FS sheets and superconducting energy gaps in a momentum space. Ding et al. [8] observed three FS sheets in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ crystals with $T_c$ of 37 K: an inner hole-like FS pocket, an outer hole-like Fermi surface sheet, both centered at the Brillouin zone center $\Gamma$ and a small electron-like FS at the M point. A large superconducting energy gap ($\Delta_e = 12$ meV) was detected on the two small hole-like and electron-like FS sheets while a small gap ($\Delta_h = 6$ meV) was found on the large hole-like FS. The gaps closing at the same $T_c$ are isotropic. Two small FS sheets with a very strong coupling strength $2\Delta_c/kT_c \sim 8$ are connected by the $(\pi,0)$ SDW vector in the parent compound indicating an importance of the interband interaction among these two nested FS sheets also for superconductivity. Similar results were obtained also in Ref. 9, 10 and 11. Wray et al. [11] proposed that the observed gap structure oscillating among the FS sheets is consistent with an order parameter that takes the in-plane form of $\Delta \cos(k_x) \cos(k_y)$. Nakayama et al. [12] reported on the observation of the fourth FS sheet in Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. It is another, outer electron pocket centered around the M point with the superconducting energy gap comparable ($\sim 11$ meV) to the gap on the inner electron and hole pockets.

Important finding has been reported by Terashima et al. [13] on ARPES measurements on Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ crystals. In the sample with $x=0.15$ and $T_c$ of 25 K due to the electron doping the inner hole-like FS sheets is absent and the nesting conditions are switched from the inner hole FS sheet to the outer one which is connected to the electron FS sheets by the $(\pi,0)$ SDW vector. Strong coupling strengths $2\Delta/kT_c \sim 6$ and $2\Delta/kT_c \sim 4.5$ are found on the hole and electron FS’s, respectively. In heavy Co doped samples only the electron-like FS sheets remain and no superconductivity is present. All this is supporting the inter-FS superconductivity in iron pnictides.

Two-gap nature in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ is supported also by infrared spectroscopy experiments [14] and specific heat measurements [15]. The penetration depth, or the lower critical field measurements by various techniques suggest a multigap picture in 122 as well. In Ba$_{1-x}$K$_x$Fe$_2$As$_2$ Hashimoto et al. [16] found the exponential temperature dependence of the superfluid density compatible with the fully opened two gaps. Particularly interesting was that only in the cleanest crystals this effect was detectable. Similar results are presented also in Ref. 17. Vorontsov et al. [18] argue that also the penetration depth measurements in another isotypic structure of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ can be explained by a two gap scenario with extended s-wave pairing.

Only few tunneling spectroscopy measurements are available so far generally in iron pnictides. Scanning tunneling microscope (STM) measurements were performed on the 32 K Sr$_{1-x}$K$_x$Fe$_2$As$_2$ [19]. On the surfaces with a stripe-like modulation on a square atomic lattice consisting of either Sr/K or As sometimes a gapped spectrum is observed with coherent peaks at 10 mV. This would correspond to the coupling strength $2\Delta/kT_c \sim 7$. Yin et al. [20] in their STM measurements on the Ba(Fe$_{0.6}$Co$_{0.4}$)$_2$As$_2$ crystals found a relatively small variation of the well pronounced gap with the averaged $\Delta \sim 6$ meV, corresponding to $2\Delta/kT_c \sim 6$ (but the local $T_c$ was not established). Also disordered vortices were detected showing neither localized states typical for an s-wave superconductors in the clean limit nor any internal structure predicted for d-wave vortices. Much bigger spatial variations from the averaged gap value were reported by Massee et al. [21] in their STM measurements on Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ with $2\Delta/kT_c$ spanning between 5 and 10.

3. Point contact Andreev reflection spectroscopy on 122-type iron pnictides

Point contact Andreev reflection (PCAR) spectra measured on the ballistic microconstriction between a normal metal and a superconductor consists of pure Andreev reflection and tunneling contributions, respectively [22]. First contribution makes the conductance inside the voltage region $|V|
A co-existence of phase separated regions with a static magnetic order and superconducting islands was demonstrated in single crystalline Ba$_{0.55}$K$_{0.45}$Fe$_2$As$_2$ [24,25]. The size of the phase separated regions of about 50 nm is again comparable with the PC diameter, a region mostly contributing to the PCAR spectrum. This must be taken into considerations to avoid a misinterpretation of the observed data.

In the following we review our point contact spectroscopy study on the Ba$_{0.55}$K$_{0.45}$Fe$_2$As$_2$ single crystals recently published in Ref. 26. The crystals were grown out of a Sn flux. The resistive measurements showed the onset of the superconducting transitions below 30 K and the zero resistance at 27 K. Rather broad transitions in some of the crystals with multiple steps are attributed to a possible different amount of potassium in different layers or different crystals. A local transition temperature measured by the point-contact technique showed superconducting $T_c$’s between 23 and 27 K.

The specific heat as well as the resistivity measurements on these crystals showed features at about 85 K. Although reduced they are found at the same temperature as on the undoped BaFe$_2$As$_2$ samples where the tetragonal-to-orthorhombic structural phase transition takes place. This transition is revealed here at a lower temperature as compared with 140 K found in Ref. 3. The decreased structural transition temperature is due to the amounts of Sn up to 1% incorporated into the bulk. A presence of tin does not significantly effect the high-temperature superconducting phase transition in Ba$_{0.55}$K$_{0.45}$Fe$_2$As$_2$.

Before point contact measurements the crystals were cleaved to reveal fresh surface. For the measurements in the $c$ direction the fresh shiny surface was obtained by detaching the degraded surface layers by a Scotch tape. The microconstrictions were prepared in situ by pressing a metallic tip (platinum wire formed either mechanically or by electrochemical etching) on a fresh surface of the superconductor. For the measurements with the point contact current in the $ab$ plane a reversed tip-sample configuration was used. The freshly cleaved edge of the single crystal jetting out in $ab$ direction was pressed on a piece of chemically etched copper. A special PC approaching system allowed for lateral as well as vertical movements of the PC tip by a differential screw mechanism. Details of the technique can be found elsewhere [23].

Figure 1 shows typical PCAR spectra obtained on Ba$_{0.55}$K$_{0.45}$Fe$_2$As$_2$ with the PC current preferably within the $ab$ plane. The spectra present double enhanced conductances, the typical features of the
Andreev reflection of quasiparticles coupled via two superconducting energy gaps. The first enhancement starts below 20 mV with the gap-like humps at about 10 mV while the second one is located below ~5-7 mV. On the spectra 2 and 3 also two symmetrical maxima at 2-3 mV are displayed. Majority of the spectra measured in the $ab$ direction revealed a heavily broadened enhanced conductance near the zero bias as indicated by the spectrum 4. This is most probably caused by the sample inhomogeneities on the nanoscale.

The presented spectra are normalized to their respective normal state and fitted to the two gap BTK model (symbols). The resulting values of energy gaps are spread in the range of 2-5 meV and 9-10 meV for the small and large gap, respectively. The values of smearing parameters were 10, 60, 30, and 100% of each energy gap value for curves 1, 2, 3, and 4, respectively. For each presented fit different values of $z$ for the two bands were also necessary. Typically, $z_L$ for the band with a large gap was about 0.4 – 0.8, while $z_S$ was twice smaller. Parameter $\alpha$ varied between 0.4 and 0.8. Although the s-wave two gap BTK formula has been successfully used to fit our PCAR data a possibility of unconventional pairing symmetry cannot be completely ruled out. Obviously, rather strongly broadened spectra as presented here could be in principle fitted also by model taking into account anisotropic or nodal gaps, if an appropriate current injecting angle was selected [27].

In Fig. 2a temperature evolution of the second spectrum from previous figure is presented. All the spectra (lines) were normalized to the conductance measured at 27 K and fitted to the BTK model with a proper temperature smearing involved. Obviously, the spectrum at the lowest temperature reminds the two gap spectrum of MgB$_2$ for a highly transparent junction with conductance enhancements due to Andreev reflection of quasiparticles. As the temperature is increased the double enhanced point contact conductance corresponding to two energy gaps is gradually smeared out and spectrum intensity decreases. Indeed, the spectra could be well fitted to the two gap BTK formula. The best fit for each temperature is shown by open circles. The extracted values of the gaps at different temperatures are shown in Fig. 2b (symbols) following nicely a BCS type temperature dependences of energy gaps.
depicts a position of the critical temperature. Evolution of zero bias conductance with the temperature, arrow direction showing a reduced conductance even above $T_c$. The measurements on the Ba$_{0.55}$K$_{0.45}$Fe$_2$As$_2$ single crystals with the PC current in the c direction yield completely different picture showing just a reduced conductance around zero bias. In Fig. 3 the temperature dependence of such a spectrum is displayed. The zero-bias conductance minimum is step by step smeared and filled up with increasing temperature. However, the filling effect, which cannot be explained just by the spectral broadening by temperature, is not finished at $T_c$ but continues up to about 70 - 80 K, the temperature at which the magnetic transition in the system takes place [28]. Thus, this feature is not related to superconductivity.

Absence of apparent superconducting gap features in the c axis junctions characteristics is probably caused by the surface contamination and reconstruction.

Remarkably a similar reduced conductance background around the zero bias is displayed sometimes also on the ab plane junctions revealing the two gaps at low temperatures [26]. The reduced conductance is then detectable near $T_c$ and persists again well above $T_c$ similarly to the case of the c axis spectra. This indicates that the reduced conductance is a spectral feature related to a reduced DOS in the normal state. A pseudogap in the quasiparticle excitation spectrum responsible for superconductivity is one of possible interpretations. But since there is an evidence for a mesoscopic phase separation of antiferromagnetically ordered and non-magnetic/superconducting regions in the similar single crystals [25] another possibility would be that there are parallel PC currents to these two regions. A rather large broadening of the PCAR spectral features revealed by $\Gamma$ parameters could also be related to the pair breaking effect originating from static magnetic moments.

Recently we extended our PCAR measurements to the hole doped system of the 122-type family, namely to the optimally doped Ba(Fe$_{0.6}Co_{0.07}$)$_2$As$_2$ samples with $T_c$ of approximately 23 K determined from transport and susceptibility measurements. The single crystals were grown from FeAs/CoAs flux from a starting load of Ba, FeAs and CoAs precursors. Details of the preparation resulting to large crystals of a few mm size can be found elsewhere [29].

In Fig. 4 the typical spectra of the Pt-Ba(Fe$_{0.6}Co_{0.07}$)$_2$As$_2$ junctions measured at 4.5 K are presented. The spectra show enhanced differential conductances and the bottom curve reveal also a single pair of peaks at about 5 mV. In no case a double enhanced conductance spectrum indicative for two gap superconductivity was found. Indeed, the spectra on Ba(Fe$_{0.6}Co_{0.07}$)$_2$As$_2$ can be fitted just by a single s-wave gap BTK formula. This is in strong contrast to the PCAR spectra of the hole doped Ba$_{0.55}$K$_{0.45}$Fe$_2$As$_2$. The fits on the electron doped system have yielded the superconducting energy gap
of around 5-6 meV. In all spectra a significant broadening was observed which is witnessed by a large value of the $\Gamma$ parameter spreading between 50% and 100% of the respective gap value. The superconducting transition temperature at the junctions was 22 K very close to the bulk $T_c$ determined from transport measurements. With $T_c$ the coupling strength $2\Delta/k_BT_c$ between 5.3 and 6.3 can be determined. This is remarkably consistent with the above mentioned results obtained by the ARPES measurements. Although our results are only preliminary and the statistics is not yet sufficient, the data seem to prove that if there are multiple gaps in the electron doped $\text{Ba(Fe}_{0.93}\text{Co}_{0.07})_2\text{As}_2$ superconductors they are quite close to each other.

**Note:** We are not aware of any other PCAR measurements done on 122-type iron pnictides so far but it is worth to note that on the 1111 superconductors similar results to ours were obtained but it is worth to note that on the 1111 superconductors similar results to ours were obtained by several groups. For example on $\text{LaFeAs(O,F)}$ Gonnelli et al. [30] observed very pronounced two gap PCAR spectra. Their size of the large and small gaps on the samples with a similar $T_c$ are remarkably alike to those presented here.

**Conclusion**

In contrast to cuprates which are single band and $d$ wave superconductors, 122-type iron pnictides seem to be proven as the multiband systems with multiple nodeless gaps. In the hole doped $\text{Ba}_x\text{K}_y\text{Fe}_2\text{As}_2$ the available data point to an existence of two distinct superconducting energy gaps with the strength below and much above the single band BCS weak coupling limit, respectively. In the electron doped $\text{Ba(Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ if there are two gaps present they are very close to each other having a strong coupling $2\Delta/k_BT_c$ between 5 and 6. Further experimental and theoretical effort is certainly needed to elucidate physics in these extremely interesting systems.

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