Evidence for two-gap superconductivity in $Ba_{0.55}K_{0.45}Fe_2As_2$ from directional point-contact Andreev-reflection spectroscopy

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Directional point-contact Andreev-reflection spectroscopy measurements on the $Ba_{0.55}K_{0.45}Fe_2As_2$ single crystals are presented. The spectra show significant differences when measured in the *ab* plane in comparison with those measured in the *c* direction of the crystal. In the latter case only a reduced point-contact conductance around zero bias has been revealed persisting well above T_c and probably related to the structural and magnetic transitions in the system. Within the *ab* plane two superconducting energy gaps are detected below T_c. Here a reduced conductance above T_c could also be found. The fits of the *ab*-plane data to the superconducting *s*-wave two-gap model indicate that the smaller gap has a size below the BCS value while the large gap reveals much higher coupling strength.

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REFeAsO(F) systems with various rare earth (RE) elements bring a new class of layered high-T_c materials without copper.[1] $NdFeAsO_{0.9}F_{0.1}$ with T_c above 51 K (Ref. 2) together with the Sm and Pr compounds reveal the highest transition temperature at ambient pressure among all oxypnictides. Besides REFeAsO(F), another three groups within pnictides have been identified. Namely, AFe_2As_2 with A=Ba, Sr, and Ca made superconducting by chemical substitution[3] or pressure,[4] Li_xFeAs (Ref. 5) and α-FeSe (Ref. 6) are intensively studied.

One of the important questions to be addressed in these multiband systems concerns the symmetry of the superconducting order parameter(s). There are already many theoretical predictions on this topic but also a body of experimental studies is emerging. Band-structure calculations have shown disconnected sheets of Fermi surfaces with possibly different superconducting energy gaps. A minimal model has to include two bands: the hole band around the Γ point and the electron one around the M point.[7] In contrast to the multiband but conventional *s*-wave scenario in MgB_2,[8] here, the extending *s*-wave pairing with a sign reversal of the order parameter between different Fermi-surface sheets was proposed by Mazin *et al.*[9] Experimental results are providing controversial conclusions. NMR studies on PrFeAsO(F) (Ref. 10) found two superconducting energy gaps with nodes. Also very recent muon spin-relaxation measurements on single crystalline $Ba_{0.5}K_{0.5}Fe_2As_2$ (Ref. 11) point to the gap with nodes. On the other hand, the angle-resolved photoemission spectroscopy measurements[12] have found two nodeless and nearly isotropic gaps in $Ba_{0.6}K_{0.4}Fe_2As_2$ and the penetration depth studies have provided the nodeless superconducting energy gap in $NdFeAsO_{0.9}F_{0.1}$ but with remarkably small coupling $2\Delta/k_BT_c \approx 2$.[13]

By the point-contact Andreev-reflection (PCAR) spectroscopy, Wang *et al.*[14] found a nodal superconductivity with multiple gaps in SmFeAsO(F). Chen *et al.*[15] presented a surprisingly conventional superconducting energy gap with a medium coupling equal to $2\Delta/k_BT_c \approx 3.7$. The recent data of Yates *et al.*[16] obtained on the 45 K $NdFeAsO_{0.85}$ show also an indication of the superconducting energy gap with $2\Delta/k_BT_c=3.6$. In our previous paper[17] two nodeless gaps have been found by the PCAR measurements on the polycrystalline samples of NdFeAsO(F).

In the following we present directional PCAR studies on iron pnictides, namely, on the $Ba_{0.55}K_{0.45}Fe_2As_2$ single crystals. The spectra have shown significant differences when measured in the *ab* plane and in the *c* direction. In the latter case no suitable conditions to observe superconducting energy gaps with coherent gaplike peaks and enhanced conductance in the PCAR spectrum have been found. A predominant feature has been revealed in the form of a reduced point-contact conductance persisting well above T_c which can be attributed to a reduced density of states (DOS) due to a structural phase transition[18] and magnetism found in a partial volume fraction in these samples below 70 K.[11] On the other hand, within the *ab* plane two superconducting energy gaps are found. The pair of peaks due to a small gap has been scattered between $\pm(3-5)$ mV while the humps indicating the presence of the second gap are placed at $\pm(10-12)$ mV. In the superconducting gaplike spectra the reduced conductance above T_c could also be found. The *ab*-plane spectra have been successfully fitted to the *s*-wave two-gap model providing the conventional temperature dependence of the two gaps with the coupling strengths $2\Delta_1/kT_c=2.5-4$ and $2\Delta_2/kT_c=9-10$.

The $Ba_{0.55}K_{0.45}Fe_2As_2$ single crystals were grown out of a Sn flux as described in detail in Ref. 18, where the following characteristics have been found: typical dimensions of the crystals are $2\times2\times0.1$ mm^3. The crystallographic *c* axis is perpendicular to the plane of the platelike crystals. The resistive measurements have shown 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. The specific heat as well as the resistivity measurements on these crystals has shown features at about 85 K. Although reduced they are found at the same temperature as on the undoped $BaFe_2As_2$ samples where the tetragonal-to-orthorhombic structural phase transition takes place. This transition associ-

ated also with a reduction in the density of states at the Fermi level (indicated by the Hall effect) has been revealed here at 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 $B_{0.55}K_{0.45}Fe_2As_2$. In the present work a local transition temperature has been measured by the point-contact technique showing superconducting T_c's between 23 and 27 K.

The crystals were cleaved to reveal fresh surface before point-contact measurements. For the measurements in the *c* direction the fresh shiny surface has been 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 usually a reversed tip-sample configuration has been used. The freshly cleaved edge of the single crystal jetting out in *ab* direction has been pressed on a piece of chemically etched copper. A special point-contact approaching system allowed for lateral as well as vertical movements of the point-contact tip by a differential screw mechanism. Details of the technique can be found elsewhere.[8]

PCAR spectra measured on the ballistic microconstriction between a normal metal and a superconductor consists of pure AR and tunneling contributions, respectively.[19] First contribution makes the conductance inside $|V|<\Delta/e$ twice as large as in the normal state or as what is at large bias where the coupling via the gap Δ is inefficient. Tunneling contribution reduces the conductance at the zero bias and two symmetrically located peaks rise at the gap energy. PCAR conductance can be compared with the Blonder-Tinkham-Klapwijk (BTK) model using as input parameters the energy gap Δ, the parameter z (measure for the strength of the interface barrier), and a parameter Γ for the spectral broadening.[20] For a multiband/multigap superconductor the point-contact conductance $G=dI/dV$ can be expressed as a weighted sum of partial BTK conductances. As shown in our previous work[8] on the two-gap superconductor of MgB_2 the total PCAR conductance consists of two parallel contributions, the first originated from a three-dimensional (3D) π band with a small gap Δ_1 and a weight α and the second from a quasi-two-dimensional σ band with a large gap Δ_2, respectively.

Spectral characteristics as the superconducting energy gaps or electron-phonon interaction features can be read from the point-contact data only if the junction is in a ballistic or diffusive regime, where heating effects are avoided. There, both the elastic (l_e) as well as inelastic (l_i) mean-free paths should be bigger than the diameter d of the junction or a diffusion length $\sqrt{l_e l_i}>d$. These requirements should be satisfied in the normal as well as in the superconducting part of the junction.[19] The point contacts made on the $Ba_{0.55}K_{0.45}Fe_2As_2$ single crystals have resistances between tens and hundreds of ohms which corresponds to the contact diameter of tens of nanometers as calculated from the Wexler's formula[21] for the point contact on a highly resistive material as $Ba_{0.55}K_{0.45}Fe_2As_2$.[18] Indeed, the electron mean-free path, although not known in $Ba_{0.55}K_{0.45}Fe_2As_2$ could also be on the order of tens of nanometers. Then the precautions should be made to avoid the junctions with heating effects. In the following only the junctions without the conductance dips and irreversibilities in voltage dependences are presented. Predominantly the junctions with a finite barrier-strength parameter z allowing for tunneling component in the spectrum were examined.

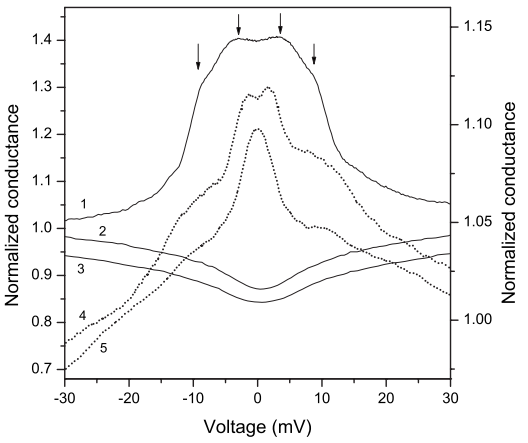

FIG. 1. Typical PCAR spectra measured with the PC current mostly within the *ab* plane of the $Ba_{0.55}K_{0.45}Fe_2As_2$ crystal showing two superconducting gaps indicated by arrows (curves 1, 4, and 5) and in the *c* direction showing reduced PC conductance near zero bias (curves 2 and 3). For the curves 4 and 5 the right ordinate applies.

Figure 1 shows typical PCAR spectra obtained on Pt-$Ba_{0.55}K_{0.45}Fe_2As_2$ point contacts with the current preferably within the *ab* plane (curves 1, 4, and 5) and in the *c* direction (curves 2 and 3) of the crystal. All spectra shown here are raw data, just normalized to their own values at $V=50$ mV eventually vertically shifted for the clarity. The spectra 1, 4, and 5 present (double) enhanced conductances, the typical features of the Andreev reflection of quasiparticles coupled via the superconducting energy gap(s). The first enhancement starts below 20 mV with the gaplike humps at about ± 10 mV while the second one is located below 5–7 mV. On the spectra 1 and 4 also two symmetrical maxima at ± 4 and ± 2 mV, respectively, are displayed. Majority of tens of the spectra measured in the *ab* direction revealed a heavily broadened enhanced conductance near the zero bias as indicated by the spectrum 5.

The measurements with the point-contact current along the *c* axis yield a completely different picture. For this direction we have collected tens of spectra without any enhanced PCAR conductance and coherence peaks due to superconducting energy gap(s). On the contrary, the spectra display a reduced conductance around the zero-bias conductance (ZBC). In Fig. 2 one can find the temperature dependence of such a spectrum. The reduced conductance is gradually smeared and filled up when increasing the temperature. Moreover, as could be seen from the measurements at higher temperatures and from the ZBC vs temperature dependence (inset of Fig. 2) the effect of the reduced conductance persists well above the superconducting transition up to about the Néel temperature $T_N \approx 70$ K found for a similar

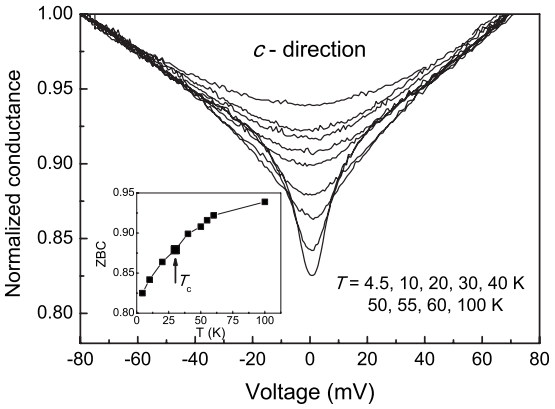

FIG. 2. Spectrum measured in the c direction showing a reduced conductance even above T_c. Inset—zero-bias conductance as a function of temperature.

sample.[11] Thus, within the c direction suitable spectral conditions for observing the superconducting energy gap have not been found, possibly due to a surface reconstruction or contamination. Then the observed reduced conductance can be just a differential-conductance-versus-voltage replica of the increasing resistivity with decreasing temperature below 85 K found in the undoped sample.[18] Another possibility is that it is a spectral characteristic where the superconducting gap features are heavily smeared.

Figure 3 shows the representative spectra measured within the ab planes at different temperatures. The left inset presents the raw data taken at 4.4 and 27 K. As can be seen the spectral background reveals asymmetry, being higher at positive bias voltage, i.e., at negative sample bias. This asymmetry seems to have different sign for the 122 pnictides (see also Ref. 22 for $Sr_{1-x}K_xFe_2As_2$) than for the 1111 superconductors as indicated by Chen *et al.*[15] or in our previous measurements on the polycrystalline NdFeAsO(F).[17] For a better comparison with the BTK model the spectra in the main figure are normalized to the normal-state curve measured at 27 K. The remarkable feature at the lowest tempera-

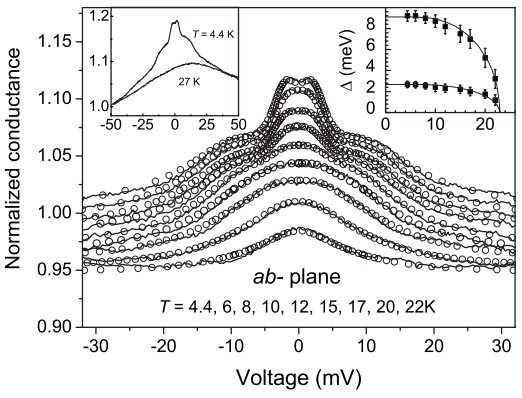

FIG. 3. Normalized PCAR spectra of the $Ba_{0.55}K_{0.45}Fe_2As_2$ single crystal in the ab plane showing two superconducting gaps. Lower curves are vertically shifted for clarity. Circles represent the best fits to the two gap BTK formula. Left inset—raw data taken at 4.4 and 27 K, right inset—temperature dependence of two gaps, solid lines show BCS-type behavior of energy gaps.

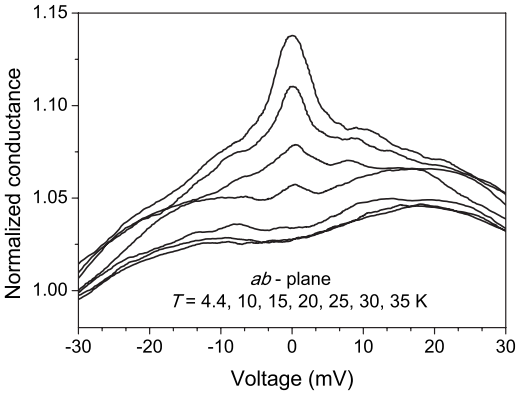

FIG. 4. PCAR spectra of the $Ba_{0.55}K_{0.45}Fe_2As_2$ single crystal showing the gap as well as the pseudogap features. The spectra have been divided by their value at 60 mV.

ture is a double enhanced point-contact conductance. The form of the spectrum obviously reminds the two-gap spectrum of MgB_2 for a highly transparent junction with conductance enhancements due to Andreev reflection of quasiparticles on the small and large superconducting energy gaps. With increasing temperature the two-gap structure is smoothly smeared out. Indeed, the spectra from Fig. 3 could be easily fitted to the two-gap BTK formula. The best fit for each temperature is shown by open circles. The right inset of Fig. 3 shows the temperature dependence of the superconducting energy gaps resulting from the fitting procedure. For comparison we also show by solid lines the BCS-type temperature dependence of Δ's. The values of the energy gaps at lowest temperature are for the small $\Delta_1 \approx 2.7$ meV and the large one $\Delta_2 \approx 9.2$ meV, which corresponds to the coupling strengths $2\Delta_1/kT_c=2.7$ and $2\Delta_2/kT_c=9.3$ for $T_c=23$ K. The smearing parameters $\Gamma_1=1.8$ meV and $\Gamma_2=6$ meV, the barrier strengths $z_1=0.34$ and $z_2=0.5$ as well as the weight factor $\alpha=0.5$ obtained at 4.4 K were kept constant at higher temperatures (Γ's changing less than 10%). From the data obtained on many junctions we observe that the gaps are scattered as $2\Delta_1/kT_c=2.5-4$ and $2\Delta_2/kT_c=9-10$.

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 anisotropic or nodal gaps, if an appropriate current injecting angle was selected.[23] On the other hand, in no case the ZBC peak in the low-temperature PCAR conductance as a fingerprint of the Andreev bound states due to d- or p-wave symmetry has been observed.

Figure 4 presents another point contact measured in the ab plane documenting that besides the broadened Andreev-type enhancements of the conductance also a reduced conductance background around the zero bias is revealed on the same junction. The latter feature detectable near T_c has persisted 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.

Does the simultaneous occurrence of the superconducting energy gaps and reduced conductance on the same junction

mean that these two effects are due to quasiparticles from the same Fermi surface? Goko *et al.*[11] brought evidence for coexistence of superconductivity and strong static magnetic order in a partial volume fraction of $(Ba,K)Fe_2As_2$ and similar systems. Recently Park *et al.*[24] found a mesoscopic phase separation on the single crystals of $(Ba,K)Fe_2As_2$ with a similar $T_c \approx 32$ K as in our case. This phase separation leads to antiferromagnetically ordered and nonmagnetic/superconducting regions, both with characteristic sizes of several tens of nanometers. Our point contacts have about the same size. Then even if the reduced point-contact conductance is observed on the same junction which reveals also the superconducting energy gaps one has to admit that these two effects are caused by two spatially separated phases and not by the quasiparticles originating from the same Fermi surface. To resolve the problem of a possible coexistence of superconductivity and magnetism in a single electronic system the spectroscopic studies on the system without a mesoscopic phase separation are necessary.

Very recently Gonnelli *et al.*[25] also obtained pronounced two-gap spectra on the $LaFeAsO_{1-x}F_x$ polycrystals as well as shown an existence of the reduced DOS in the normal state up to about 140 K, close to the Néel temperature of the undoped specimen. Their size of the large and small gaps are remarkably similar to those found in the present work on the 122 sample with a similar T_c.

In conclusion, directional PCAR studies have been performed on the single crystalline $Ba_{0.55}K_{0.45}Fe_2As_2$. Significant differences in the spectra are observed when measured within the *ab* planes and in the *c* direction. Within the planes two superconducting gaps are detected with $2\Delta_1/kT_c = 2.5-4$ and $2\Delta_2/kT_c = 9-10$. Within the *c* direction only a reduced conductance could be found persisting well above T_c, possibly connected with the reduced DOS in a mesoscopic antiferromagnetic region spatially separated from the superconducting phase.

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