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Abstract
We studied the grain boundaries within mechanically formed polycrystalline Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ micro-bridges. By tunneling current across grain boundaries, current–voltage characteristics (IVCs) demonstrated the typical Josephson weak links behavior in micro-bridges. Shapiro steps were observed for the junctions under the microwave radiation at 10 GHz. The temperature dependence of the critical current $I_c$ was observed as a shoulder, corresponding to a multi-gap symmetry but not a single-gap s-wave or d-wave.

Keywords: grain boundary effect, Josephson junction, multi-gap symmetry, iron-based superconductors

(Some figures may appear in colour only in the online journal)

1. Introduction
The superconducting (SC) mechanism for the newly discovered Fe-based superconductors (FeSCs) is still an open question [1, 2] in regards to high critical temperature ($T_c$) cuprate superconductors. To elucidate the pairing mechanism, it is crucial to probe the symmetry of the SC phase and then present the Cooper pairing dependent energy and momentum [2]. Theoretical scientists have proposed several possible models, including the single-gap s- and d-state, and the multi-gap $s_{\pm}$ wave [3, 4], $s_{++}$ wave [5–7], and $s + id$-wave [8]. Singlet symmetry, such as the $s$- and $d$-state, was determined by NMR experiments from Knight shift measurements [9–11] in several systems including the 1111-, 122- and 11-systems. However, the multi-gap s-wave ($s_{\pm}$ or $s_{++}$) is generally accepted. Both states represent the same Fermi pockets, and have opposite signs for the electron pockets; namely, the $s_{\pm}$ wave is a sign-reversal s-wave model, and a non-sign-reversal for the $s_{++}$ state. Multi-gap symmetry was supported by Josephson tunneling [12, 13], angle-resolved photoemission spectroscopy (ARPES) [14], London penetration [15], and impurity doping [16] experiments. Among these methods, the Josephson tunneling effect is powerful owing to phase- and amplitude-sensitivity, making it possible to distinguish the multi-gap from the single-gap.
Until now, few Josephson effect experiments on FeSCs have been reported. Zhang et al [12] fabricated a hysteretic junction between Pb and (Ba,K)Fe2As2. The observation of robust Josephson coupling and current–voltage characteristics (IVCs) suggested the exclusion of predominant p-wave symmetry, but can hardly identify the multi-gap s-wave from the d-wave. Integer and half-integer flux quantum in polycrystalline NdFeAsO1−xF0.12 [13] also supported s± or d-wave pairing, but ruled out p-wave pairing. On the other hand, a thin-film bicrystal technique with grain boundary was widely used to study the Josephson tunneling effect. Bicrystals can be easily fabricated and therefore have been applied in various SC devices for several decades, including cuprates [17, 18] and MgB2-based [19] superconductors. Although Fe-based thin films have been fabricated successfully in several systems, including Fe(Se,Te) [20], Ba(Fe,Co)2As2 [21–23], and LaFeAsO1−xF [24], there are still some technical difficulties for bicrystal study in FeSCs. Alternatively, polycrystals is another promising way to study the grain boundary because a great number of single crystalline grains connect each other to natural grain boundaries [13].

In the present work, we will discuss the IVCs and temperature dependence of the critical current $I_c$ for grain boundary junctions in polycrystalline Ba0.6K0.4Fe2As2 microbridges. Microwave radiation at 10 GHz was applied to the junctions to confirm Josephson coupling. In particular, the $I_c$–$T$ relation was studied to understand the influence of the order parameter symmetry on the behavior of the Josephson tunneling junction. The experimental data also will also be compared with the theoretical prediction for the simple s- and d-wave, and the multi-gap s-wave.

2. Experimental methods

The synthesis of Ba0.6K0.4Fe2As2 polycrystals was described elsewhere [25]. Previous microscopic analysis demonstrated a denser and uniform microstructure for the Ba0.6K0.4Fe2As2 polycrystals due to the rolling process under high pressure. Figure 1(b) shows a scanning electron microscopic image for the enlarged view of the polycrystal. The fine single crystalline grains were randomly oriented within the bulk crystal and connected to each other with a misaligned angle, resulting in abundant grain boundaries. Grains were observed having a width of 1–5 μm. We selected a bulk polycrystal and polished it to a thickness of 100–200 μm, and then mounted it onto a MgO substrate using epoxy (EPOTEK, 353ND04), and heat-treated it at 110 °C for 2–3 h. Finally, the thin region was then cut into a microbridge using a sharp knife, as shown in the inset of figure 1(a). Figure 1(a) shows the schematic view of the microbridge and electrodes. We emphasize that the cross-sectional area of the microbridge was gradually shrunk until the two-terminal resistance reached a few ohm. The microbridge was connected to Au wires using silver paste for electrodes. The transport properties of the junctions were measured in a Janis helium flow cryostat.

![Figure 1](image)

3. Results and discussion

The temperature dependence of resistance $R$ for a microbridge is given in figure 2. The onset $T_c$ is about 35.1 K, being similar in behavior as that of the bulk crystal [25]. Since the microbridge cross-section is considerably less than that of the electrodes, the total resistance can be attributed to the microbridge. According to our previous measurements on Ba0.6K0.4Fe2As2 polycrystals [25], the normal state resistivity at 38 K is around 100 μΩ · cm. Thus, we can roughly estimate the cross-sectional area of the microbridge as 28.6 μm².

Figure 3 shows the IVCs of the microbridge with or without microwave irradiation at 10 GHz. The power ranges from 7 to 23 dBm. Without microwave irradiation, the IVC was recorded by sweeping the current up and down through the microbridge from −30 to 30 mA. The critical current density $J_c$ was estimated as 62.9 kA cm⁻² for the microbridge at 10 K, which was considerably less than the Ginzburg–Landau limit of supercurrent capacities for Ba0.6K0.4Fe2As2 superconductors, regardless of the ab-plane or c-axis. The ab-plane depairing current density of the Ba0.6K0.4Fe2As2 patterned microbridge was as high as 10.8 MA cm⁻² at 4 K [26]. Additionally, the present Ba0.6K0.4Fe2As2 microbridge demonstrated no obvious flux-flow behavior from $R$ – $T$ and IVC curves. Furthermore, the flux motion dominated $J_c$ was in a magnitude of MA/cm² in a
pure crystal [27], about two orders of magnitude higher than the present $J_c$. Therefore, we ascribe the considerably less $J_c$ to the grain boundaries rather than to flux motion or depairing.

When the current was reduced, the curve returned to the resistive branch without hysteresis, indicating a resistively shunted junction [17, 28]. The profile of the present IVCs resembles that of the YBa$_2$Cu$_3$O$_{7-\delta}$(YBCO) grain boundary junctions, but with remarkable excess current. At 10 K, $I_c/\text{R}_n$ of the whole microbridge is about 70 mV ($\text{R}_n$ is the normal resistance), much larger than the value estimated at the superconducting gap (as an order of micro-voltage for the 122-type [28]), suggesting that a great number of grain boundary junctions exist in series in this microbridge. Although we cannot confirm the grain boundary geometry from the scanning electron microscopic image, grain connections should be intense due to the extremely low coherence length of the high-$T_c$ Fe-based superconductors, which is normally below 2 nm at temperatures far below the $T_c$ [26].

When the junctions were irradiated with microwaves at 10 GHz, the IVCs demonstrated a series of pronounced Shapiro steps. The Shapiro steps were observed with voltage interval $D_V$ of around 21 $\mu$V. The interval voltage $D_V$ satisfied Josephson relation

$$\Delta V = f \cdot \Phi_0,$$

where $f$ is the microwave frequency and $\Phi_0$ is the flux quantum [17].

To investigate the influence of the order parameters on the behavior of the Josephson junction, it is essential to know the temperature dependence of the critical current. In figure 4, the experimental data of the normalized $I_c/I_{c0}$ is plotted as a function of normalized temperature $T/T_c$, where, $I_{c0}$ is the critical current at 0 K. To study $(I_c/I_{c0}) - (T/T_c)$ curves within various gap symmetry models, we applied the single-gap model of the s-wave and d-wave and an effective two-gap superconductor model, similar to those in MgB$_2$ calculations. To simplify beginning calculations, we considered the weak coupling regime for order parameter calculations in the two-gap model. We used the standard self-consistent two-gap equations [29] for a clean superconductor in zero field to...
calculate the components of the order parameters.

\[
\Delta_n = \sum_{n'} \lambda_{nn'} \Delta_{n'} \int_0^{\omega_D} \frac{de}{\sqrt{\varepsilon^2 + \Delta_{n'}^2}} \tan \left( \frac{\sqrt{\varepsilon^2 + \Delta_{n'}^2}}{2T} \right),
\]

where \( \Delta_n \) is the components, \( \lambda_{nn'} = g_{nn'} N_{n'} \) (where \( N_{n'} \) is the density of states in the \( n' \) gap) is the dimensionless coupling constant (between \( n \) and \( n' \) components), and \( \omega_D \) is the Debye frequency.

\[
\omega_D = T_c e^{-\gamma \pi / 2},
\]

where \( \gamma \) is the highest eigenvalue of the \( \lambda_{nn'} \) matrix, \( T_c \) is the critical temperature, and \( C \) is the Euler constant. Here we only consider a two-gap model case, namely, two components, \( \Delta_1 \) and \( \Delta_2 \) for the larger and the smaller gap, respectively.

\[
\Delta_1 = \frac{2}{\lambda} \sum_{n'=-1}^{1} g_{1n'} \Delta_{n'} \int_0^{\omega_D} \frac{de}{\sqrt{\varepsilon^2 + \Delta_{n'}^2}} \tan \left( \frac{\sqrt{\varepsilon^2 + \Delta_{n'}^2}}{2T} \right),
\]

\[
\Delta_2 = \frac{2}{\lambda} \sum_{n'=-1}^{1} g_{2n'} \Delta_{n'} \int_0^{\omega_D} \frac{de}{\sqrt{\varepsilon^2 + \Delta_{n'}^2}} \tan \left( \frac{\sqrt{\varepsilon^2 + \Delta_{n'}^2}}{2T} \right),
\]

and \( g_1 = g_1, g_{12} = g_{31} = g_3 \), and \( g_{22} = g_2 \), where \( g_1 \) and \( g_2 \) are the pair interaction constants of the first and second gaps, respectively, \( g_3 \) is the inter-band scattering between Copper pairs of the two gaps. For simplification, we considered that \( \lambda_2 = \lambda_{31} \).

We then applied the previously calculated gaps to the Ambegaokar-Baratoff [17] equation for the different tunneling channels [30, 31]

\[
I \cdot R_n = \sum_i \frac{C_i \pi}{2} e^{-\Delta_i(T)} \tanh \left( \frac{\Delta_i(T)}{2k_B T} \right),
\]

where \( e \) is the elementary charge, \( k_B \) is the Boltzmann constant, \( \Delta_i(T) \) means the \( i \) gap of order parameters, and \( C_i \) is the ratio coefficient for the \( i \) channel (we note that \( \sum C_i = 1 \)). The \( C_i \) depends on the curvature near \( T_c \) of the \( I \cdot R_n \) experimental curve, which is generally narrow and corresponds to a smaller value than \( C_2 \), i.e., the probability of tunneling though the second channel is much higher than that of the first channel. Therefore, we must only consider the value of the second gap \( \Delta_2 \) in the calculation of equations (4) and (5).

In MgB\(_2\), the superconducting gap with lowest critical temperature contributes mainly to the tunneling current in a Josephson junction along the layers of the material. The reason for this is that the superconducting gap with a low critical temperature is formed on a 3D Fermi surface pocket, and the other gap is created on a 2D Fermi surface pocket along the layers, making it more difficult for the Cooper pairs to tunnel [32, 33]. The simulation results for \( s \)-wave, \( d \)-wave, and two-gap models are given in figure 4. The data is obviously different from the single-gapped \( s \)-wave or \( d \)-wave, but suggest a multi-gap model. Considering the unequal \( T_c \) results for each gap, we obtained a relatively large value of \( \lambda_{12} \) (0.3587) and \( \lambda_{21} \) (0.1771). However, both of should be \( \ll 1 \) for the weak coupling case [34]. Consequently, the gaps should be strongly coupled. Thus, we fixed the \( T_c \) for both gaps as 35.1 K and obtained a fit result as shown in figure 4. Here, the corresponding parameters are as follows: \( \lambda_{11} = 0.9610, \lambda_{12} = 0.7543, \lambda_{22} = 0.3183, \lambda_{31} = 0.1069, \) and \( \lambda_{32} = 0.8931 \). Note that \( \lambda_2 \) is considerably larger than \( \lambda_1 \), indicating that one gap dominates the Josephson tunneling.

Ota and co-workers [30, 31] calculated the \( T \)-dependence of \( I \cdot R_n \) for a heterojunction between the Fe-based superconductors and a single-band BCS type superconductor (lead). They also indicated that the grain boundary junctions show similar behavior to that of the heterojunction. However, the \( (I_c / I_c) \) - \( (T_c / T_c) \) curve has different curvature near \( T_c \) from the experimental one in figure 4, mainly because we considered that all gaps participate in the tunneling in equal form. Additionally we compared the experimental data with calculations made for other types of order parameter symmetries that could be considered as candidates, standard single-gap \( s \)-wave and \( d \)-wave, for which we calculated and plotted the \( (I_c / I_c) \) - \( (T_c / T_c) \) curves (figure 4). Our results indicate that both single-gap models seem unlikely for the present experimental results, and instead, the two-gap model is a highly promising candidate. Nevertheless, we cannot fit the present experimental results by applying the two-gap model directly. This is because the microbridge consists of abundant grain boundary junctions, which are connected to each other via both parallel and series connections. We assume that the junctions are connected in series. Thus, a more accurate estimation should be as follows:

\[
I \cdot R_n = \sum_j I \cdot R_{nj},
\]

where, the normal resistances may vary in different junctions, resulting in differences in total normal resistance. However, the \( (I_c / I_c) \) - \( (T_c / T_c) \) curves of each junction should be synchronous due to the same superconducting gap. As a result, we can obtain the gap symmetry profile from the \( (I_c / I_c) \) - \( (T_c / T_c) \) calculation results, even though we can hardly estimate the magnitude of the gap function. Thus, the consistency between the two-gap model and the characteristics of the experimental \( (I_c / I_c) \) - \( (T_c / T_c) \) curve suggests the presence of two effective gaps with differential magnitude. The present result is in agreement with the presented belief from the scientific community that the family of iron-based superconductors is a multi-gap \( s \)-wave superconductor. Howeverm, our results are inconclusive in regards to the symmetry of the hidden gap, namely, whether it is \( s_{\perp}, s_{++} \) or even \( s+id \)-wave. Based on the ARPES and other measurements [35], the pairing symmetry was considered as pure \( s \)-wave for the BaK\(_{1-x}\)Fe\(_2\)As\(_2\) with \( x = 1 \). For the optimal-doped Ba\(_1-x\)Fe\(_2\)As\(_2\) with \( 0.4 < x < 1 \) however, the symmetry is still under active debate, although most results indicate that it should be a mixture state [8, 36], namely, the \( s+is \)-wave or \( s+id \)-wave.
4. Conclusions

In summary, the Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ microbridge with a width of about 20 μm consists of numerous grains connected with each other and forming grain boundaries. By tunneling current across grain boundaries, the IVCs of the microbridge showed the superconductor/normal-metal/superconductor-type Josephson tunnel junction characteristics. By applying microwave radiation with a frequency of 10 GHz, typical Shapiro steps were observed for the junctions. The temperature dependence of the critical current behaved as a pronounced kink at $T \sim 20$ K, which is clearly different from the result assuming the singlet symmetry of $s$- or $d$-wave. However, this is consistent with the presented model with two effective gaps of different magnitude and $T_c$. Our results support multi-gap symmetry as $s+is$ or $s+id$-wave.

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