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# Bicrystalline Grain Boundary Junctions of Co-doped and P-doped Ba-122 Thin Films

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**Abstract.** We prepared GB junctions of  $\text{Ba}(\text{Fe}_{0.9}\text{Co}_{0.1})_2\text{As}_2$  thin films on bicrystalline [001]-tilt  $\text{SrTiO}_3$  substrates. The junctions show clear Josephson effects. Electrical characterization shows asymmetric I-V characteristics which can be described within the resistively shunted junction (RSJ) model. A large excess current is observed. Their formal  $I_{\text{C}}R_{\text{N}}$  product is  $20.2 \mu\text{V}$  at 4.2 K, which is decreased to  $6.5 \mu\text{V}$  when taking  $I_{\text{ex}}$  into account. Fabrication methods to increase this value are discussed. Additionally, measurements on GB junctions of  $\text{BaFe}_2(\text{As}_{0.66}\text{P}_{0.34})_2$  thin films on LSAT bicrystalline substrates are shown. Their symmetric RSJ/flux flow-behavior exhibits a formal  $I_{\text{C}}R_{\text{N}}$  product of  $45 \mu\text{V}$ , whereas the excess corrected value is  $11 \mu\text{V}$ .

## 1. Thin Film preparation

### 1.1. Co-doped Ba-122

Grain boundary junctions play an important role regarding applications from superconducting wires to superconducting quantum interference devices (SQUID). Pnictide superconductors in particular have advantageous properties compared to cuprates at large grain boundary (GB) angles [1]. In systematic investigations of suitable substrate materials by Iida et al. [2] and Thersleff et al. [3] STO substrates proved beneficial for the superconducting properties of Co-doped Ba-122. By introducing a buffer layer of crystalline iron the critical current density  $j_{\text{C}}$  and the transition temperature  $T_{\text{C}}$  can be increased compared to other substrate materials or STO substrates without additional iron buffer layer.

To improve these results further a buffer layer of  $\text{MgAl}_2\text{O}_4$  with a thickness of 10 nm is deposited by Pulsed Laser Deposition (PLD) between the STO [001]-tilted bicrystal substrate with an artificial grain boundary (GB) angle of  $30^\circ$  and the Fe buffer (20 nm). The crystal orientation is rotated by  $45^\circ$  between the substrate and the Fe buffer due to the different lattice constants. The (001) surface plane of crystalline iron (bcc structure) and the distance between two iron atoms within the unit cell of Co-doped Ba-122 deviate by just 2% [3]. By depositing superconducting  $\text{BaFe}_{1.84}\text{Co}_{0.16}\text{As}_2$  on the buffer stack the crystal orientation is rotated by another  $45^\circ$  while the artificial lattice mismatch of the

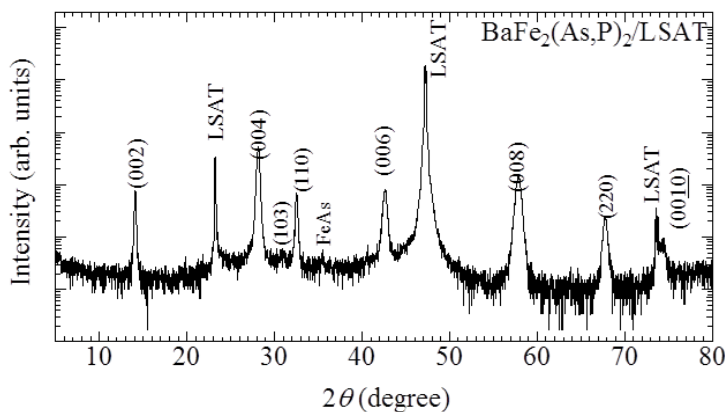


substrate is perfectly passed on to the pnictide thin film [4]. The superconducting transition starts at 24 K and reaches zero resistance at 23 K.

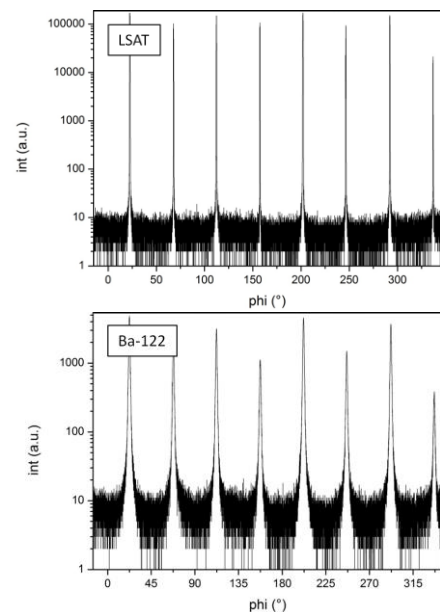
### 1.2. P-doped Ba-122

In contrast to the PLD growth of Co-doped Ba-122 on STO substrates we used Molecular Beam Epitaxy (MBE) to deposit P-doped Ba-122 (figure 1). By using MBE epitaxial growth of optimally doped (34 % phosphorous on the arsenic site) Ba-122 could be achieved without any buffer layers, see figure 2. We want to note that the optimal doping level of P-doped Ba-122 depends on the substrate material.

The [001]-tilted LSAT bicrystalline substrate with a mismatch angle of 45° is heated up to 780 °C in the MBE chamber, where the bulk materials Fe, Ba, As, and GaP are used as precursors. The solids are heated up in their effusion cells to 1065 °C (Fe), 537 °C (Ba), 186 °C (As), and 702 °C (GaP), respectively. The resulting gases Fe, Ba, As<sub>4</sub> and P<sub>2</sub> are deposited stoichiometrically on the LSAT substrate at a very slow growth rate of 100 nm per hour. The superconducting transition of the resulting BaFe<sub>2</sub>As<sub>1.32</sub>P<sub>0.68</sub> thin films occurs between 34.5 K (T<sub>C,90</sub>) and 30.5 K (T<sub>C,10</sub>).



**Figure 1.** XRD scan of a whole P-doped Ba-122 thin film on LSAT without any additional buffer layers.

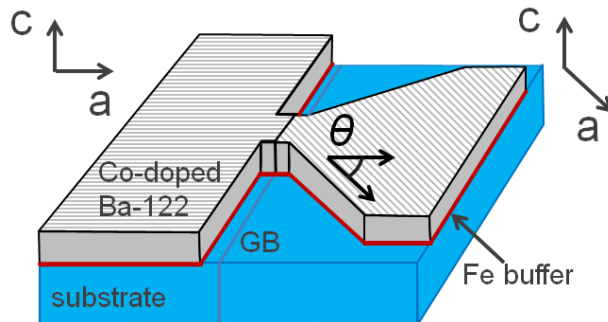


**Figure 2.** Phi-scans of LSAT (220) and P-doped Ba-122 (103). No signs of twinning or imperfect epitaxy between substrate and thin film are visible.

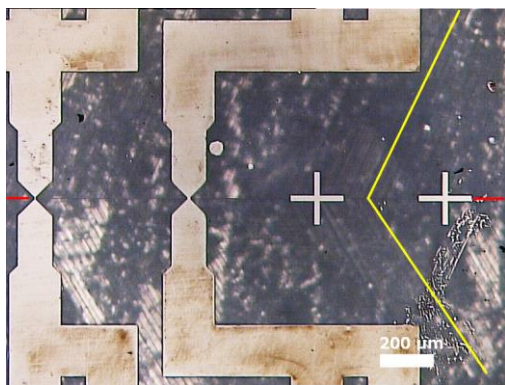
## 2. Junction preparation

The pnictide thin films are patterned by Ion Beam Etching (IBE) with a beam voltage of 500 V according to a photomask. This photolithographic layout forms microbridges of Ba-122 over the artificial grain boundary of both the substrate and the pnictide thin film (see figure 3). The bridge widths vary between 2 μm (see figure 4 left) and 10 μm (see figure 4 right).

Prior to the etching process the Co-doped thin film was covered with a gold layer to prevent surface degradation due to the exposure to air and chemicals used in the photolithographic process. Thus, the Co-doped GB junctions are shunted resistively by both the Au cap layer and the Fe buffer layer. The P-doped Ba-122 grown by MBE resulted to be more stable and do not need any cap layer. Not needing buffer layers, the P-doped thin films are not shunted extrinsically.



**Figure 3.** (left) Sketch of a microbridge forming a GB junction on a bicrystalline substrate (blue). The red layer underneath the superconducting thin film (white) denotes the Fe buffer, which is just present at the Co-doped Ba-122 films. The surface of the Co-doped material is capped with an Au layer, additionally.

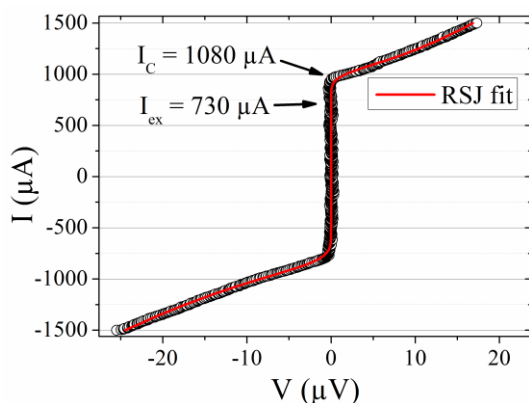


**Figure 4.** (left) Two microbridges of P-doped Ba-122 over the GB area with widths of 2  $\mu\text{m}$  (left) and 10  $\mu\text{m}$  (right), respectively. The GB area is marked with red lines and is visible as a thin line in the middle, oriented horizontally. The LSAT substrate orientation and its mismatch of 45° are marked by the yellow lines. The crosses are markers for adjusting the photomask more precisely.

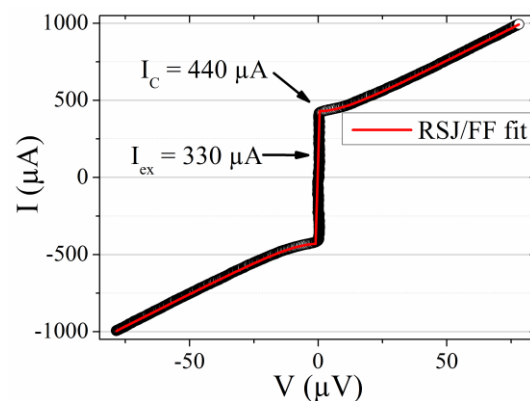
### 3. Electrical measurements on Co-doped Ba-122

Both thin film materials show Josephson-like behaviour when measuring over the GB.

In the case of Co-doped Ba-122, the I-V characteristics can be modelled within an asymmetric RSJ model at low temperatures. A large excess current of 645  $\mu\text{A}$  lowers the formal  $I_C$  from 950  $\mu\text{A}$  to 305  $\mu\text{A}$  (see figure 5) which decreases the formal  $I_C R_N$  value of 20.2  $\mu\text{V}$  to a corrected value of 6.5  $\mu\text{V}$  [4]. The critical current density across the microbridge is up to  $1.4 \cdot 10^5 \text{ A cm}^{-2}$  (formal value), which is also reduced to  $1.2 \cdot 10^4 \text{ A cm}^{-2}$  (corrected value). Instead the P-doped Ba-122 microbridges exhibit symmetric I-V characteristics, which can be fitted by a flux-flow extension of the RSJ model as shown by Saitoh et al. [5]. A formal  $I_C$  of 440  $\mu\text{A}$  as visible in figure 6 leads to a formal  $I_C R_N$  product 45  $\mu\text{V}$  which has to be corrected to a value as low as 11  $\mu\text{V}$  when taking  $I_{ex}$  into account.

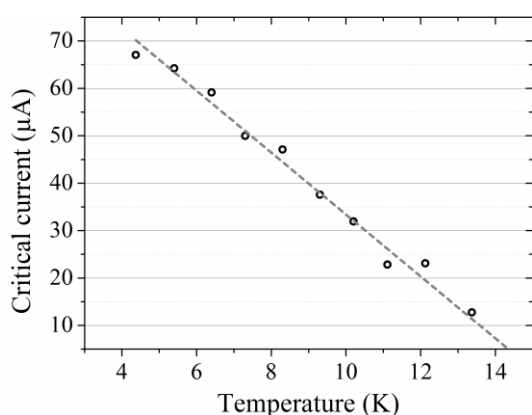


**Figure 5.** I-V characteristics of a microbridge ( $d = 7 \mu\text{m}$ ) of Co-doped Ba-122 on a 30° STO bicrystal substrate. The red line shows a classical RSJ fit.

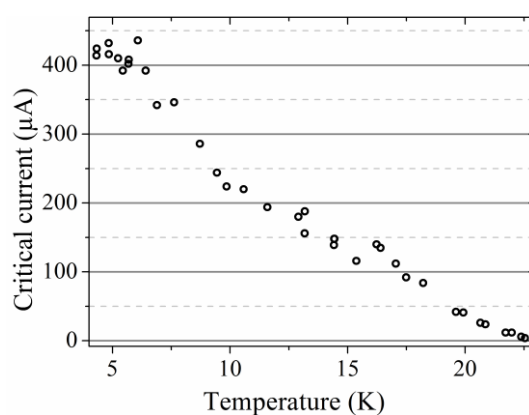


**Figure 6.** I-V characteristics of a microbridge ( $d = 10 \mu\text{m}$ ) of P-doped Ba-122 on a 45° LSAT bicrystal substrate. The red line shows a combined RSJ and flux-flow fit as in ref. [5].

The critical currents over the microbridges decay with increasing temperature differently for both materials. While the Co-doped junctions follow a rather linear decline (see figure 7), the P-doped ones behave nonlinear at both low temperatures and near  $T_C$  (see figure 8), which may be explained by a more SINIS-like behaviour of the GB [6], but further investigation would be required to verify this assumption. The low critical current of the Co-doped bridges in comparison to the results shown in figure 5 are caused by disturbances of the GB area during an ion beam etching process to remove the resistive shunt of the Au cap layer mentioned in section 2. The general temperature dependence before the cap layer removal was linear as well. During this process not only the critical current drastically decreased, but the normal state resistance increased simultaneously leaving the  $I_C R_N$  product unchanged. With a more sensitive way to remove the Au cap layer, the normal state resistivity should be increased by leaving  $I_C$  more or less constant. In this way we could tune, i.e. increase the  $I_C R_N$  product.



**Figure 7.** Critical current versus temperature of a Co-doped Ba-122 microbridge. The dashed grey line is a guide for the eye.



**Figure 8.** Critical current versus temperature of a P-doped Ba-122 microbridge.

Measurements of the Co-doped GB junctions under microwave irradiation and under influence of an external magnetic field are shown in ref. [4] and suggest that the Fe-buffer shields microwaves, effectively. Additionally, the buffer stack disturbs magnetic field measurements by trapping magnetic flux and making the critical current over magnetic field dependence highly asymmetric. First measurements on P-doped and thus non-buffered Ba-122 GB junctions show clear microwave dependence and a higher stability in magnetic fields instead.

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