BaFe$_2$As$_2$/Fe bilayers with [001]-tilt grain boundary on MgO and SrTiO$_3$ bicrystal substrates

Kazumasa Iida$^a$*, Silvia Haindl$^b$, Fritz Kurth$^a$, Jens Hänisch$^a$, Ludwig Schultz$^a$, Bernhard Holzapfel$^a$

$^a$Institute for Metallic Materials, IFW Dresden. 01171 Dresden, Germany
$^b$Institute for Solid State Research, IFW Dresden. 01171 Dresden, Germany

Abstract

Co-doped BaFe$_2$As$_2$ (Ba-122) can be realized on both MgO and SrTiO$_3$ bicrystal substrates with [001]-tilt grain boundary by employing Fe buffer layers. However, an additional spinel (i.e. MgAl$_2$O$_4$) buffer between Fe and SrTiO$_3$ is necessary since an epitaxial, smooth surface of Fe layer can not be grown on bare SrTiO$_3$. Both types of bicrystal films show good crystalline quality.

Keywords: Pnictide, Grain boundary, Thin film, Bicrystal, Buffer layer

1. Introduction

In high-$T_c$ cuprates, [001]-tilt grain boundaries (GBs) in MgO and SrTiO$_3$ bicrystal substrates have been commonly used for realizing Josephson junctions as well as investigating transport properties of GBs. After the discovery of Fe-based pnictide superconductors,[1] the community became immediately interested in exploring grain boundaries in this new class of superconductors.

Despite the recent success of realizing Co-doped BaFe$_2$As$_2$ (Ba-122) on [001]-tilt GB MgO and SrTiO$_3$ bicrystal substrates,[2, 3] these substrates may not be suitable for several reasons. Firstly, a large misfit between Ba-122 and MgO of around 6% leads to low crystalline quality or even non-epitaxial growth under our deposition condition.[4] Secondly, the SrTiO$_3$ substrate becomes electrically conductive under ultra-high vacuum (UHV) conditions, which may compromise transport measurements particularly near the normal-superconducting transition. However, implementation of Fe for MgO and Fe+MgAl$_2$O$_4$ for SrTiO$_3$ as buffer layers can solve those problems. An Fe layer itself is electrically conductive, however, the recalculation of the superconducting transition temperature ($T_c$) and the critical current density ($J_c$) is possible due to its thin layer thickness (20 nm).[5] In this report, the fabrication of Co-doped Ba-122 thin films on both [001]-tilt bicrystal MgO and SrTiO$_3$ substrates and their transport properties are presented in detail. For the aim of bicrystal Josephson junctions, substrates with a large [001]-tilt angle of over 20° have been used in this investigation.

2. Experimental procedure

2.1. Ba-122 on [001]-tilt MgO bicrystal

A 20 nm thick Fe buffer layer was deposited by means of pulse laser deposition (PLD) on a [001]-tilt MgO bicrystal substrate with misorientation angle of $\theta_{GB}=36.8^\circ$ (nominal value) at room temperature, followed by a high-temperature annealing at 700°C. This process is very effective for obtaining epitaxial Fe layers with flat surface. After the Fe buffer preparation, Co-doped Ba-122 (BaFe$_{1.8}$Co$_{0.2}$As$_2$) with 100 nm thickness was deposited at 700°C. All deposition processes have been conducted under UHV condition. A detailed description of both Fe buffer and Co-doped Ba-122 deposition procedure can be found in Ref.[6]. After the structural characterization described in sub-section 2.3, Au layers were deposited on the films by PLD at room temperature followed by ion beam etching to fabricate bridges of 0.5 mm width and 1 mm length for transport measurements.

*Corresponding author: Kazumasa Iida; Tel.: +49-351-4659-608; Fax: +49-351-4659-541.
Email address: k.iida@ifw-dresden.de (Kazumasa Iida)
2.2. Ba-122 on [001]-tilt SrTiO$_3$ bicrystal

A MgAl$_2$O$_4$ buffer was deposited on a [001]-tilt GB SrTiO$_3$ bicrystal substrate with $\theta_{GB}$=30° at 650 °C by PLD in UHV condition (pulse number 4000 at 40 mJ). In a pilot experiment, we have confirmed that MgAl$_2$O$_4$ can be grown epitaxially on SrTiO$_3$ (100) substrate. After the primary buffer deposition, the MgAl$_2$O$_4$-buffered SrTiO$_3$ substrate was cooled to room temperature for the secondary, Fe buffer deposition. A 20 nm thick Fe buffer was prepared by the same procedure as described in sub-section 2.1 except for the annealing temperature. Here, a slightly higher temperature of 750 °C was employed. After the Fe buffer preparation, a 100 nm thick Co-doped Ba-122 (BaFe$_{12}$Co$_{0.16}$As$_2$) layer was deposited. In-situ gold layer deposition at room temperature was conducted after the Ba-122 deposition for the aim of protection from possible damage during the structuring process.

2.3. Structural characterization and transport measurement

Phase purity and out-of-plane texture were investigated by means of X-ray diffraction in Bragg-Brentano geometry with Co-K$_\alpha$ radiation. In-plane orientation of both Fe and Co-doped Ba-122 were investigated by using the 110 and 103 poles, respectively, in a texture goniometer operating with Cu-K$_\alpha$ radiation. The respective measured reflection of the MgO and SrTiO$_3$ bicrystal substrates for in-plane texture measurements are 220 and 110.

Superconducting properties were measured in a Physical Property Measurement System (PPMS, Quantum Design) by a standard four-probe method with a criterion of 1 $\mu$Vcm$^{-1}$ for evaluating $J_c$.

3. Results and discussion

XRD patterns in logarithmic scale (fig. 1(a)) clearly show that the film has been grown with high phase purity and c-axis texture, i.e. with [001] perpendicular to the substrate surface. Fig. 1(b) shows the $\phi$ scans of the 103 Ba-122, the 110 Fe and the 220 MgO. It is evident from fig. 1(b) that two grains with $\theta_{GB}$ ∼ 31°, which is slightly larger than the nominal value of 36.8°, are grown due to the perfect transfer of the grain orientation from the substrate via Fe buffer to the Ba-122 layer. Here the respective average $\Delta \phi_{Ba-122}$ and $\Delta \phi_{Fe}$ are 0.84° and 0.80° (not corrected for instrumental broadening), indicative of good in-plane alignment for both layers. The GB is symmetric, i.e. it is situated with $\theta_{GB}$/2 to (100) in both grains.

$R$ vs $T$ curves in fig. 2(a) show that both the inter- and intra-grain bridge have the same onset $T_c$ as well as zero resistance, indicating the high quality of the bicrystal films. The field dependence of $J_c$ ($B \parallel c$) for both the inter- and intra-grain bridge at various temperatures is displayed in fig. 2(b). Clearly, the intra-grain $J_c$ ($J_c^{\text{intra}}$) is higher than the inter-grain $J_c$ ($J_c^{\text{inter}}$) at low magnetic fields regime due to the large $\theta_{GB}$. The difference between $J_c^{\text{intra}}$ and $J_c^{\text{inter}}$ is getting smaller with increasing applied field and finally both curves overlap at the irreversibility field for $\theta_{GB}$ ∼ 37°.

On the assumption that the weak-link behavior is empirically described by $J_c^{\text{inter}} = J_c^{\text{intra}} \exp(-\theta_{GB}/\theta_0)$ in low field.

![Figure 1](image1.png)

**Figure 1**: (a) The $\theta$/2$\theta$ scan of Ba-122/Fe bilayer on [001]-tilt GB MgO bicrystal substrate in Bragg-Brentano geometry using Co-K$_\alpha$ radiation. (b) The $\phi$ scans of the 103 Ba-122, the 110 Fe and the 220 MgO. Average misorientation angles are 37.1°, 37.0°, and 37.1° for Ba-122, Fe and MgO, respectively.
Figure 2: (a) $R - T$ curves of inter- and intra-grains. Both curves show the same $T_c$ of around 20 K. (b) $J_c - B$ performances ($B \parallel c$) of the corresponding grains at different temperatures. Open symbols represent the intra-grain and the solid symbols show the inter-grain curves. (c) Normalized critical current density ($J_{\text{inter}}/J_{\text{intra}}$) as a function of $\theta_{\text{GB}}$ at 10 K.

Table 1: The crystal structure, lattice parameter and the lattice mismatch to Fe of MgO and MgAl$_2$O$_4$.

| Materials   | Crystal structure | Lattice parameter (nm) | Lattice mismatch (%) |
|-------------|-------------------|------------------------|----------------------|
| MgO         | Rock salt         | 0.421                  | 3.7                  |
| MgAl$_2$O$_4$| Spinel            | 0.8083 ($a/2=0.4042$) | -0.4                 |

regime, the characteristic angle ($\theta_0$) is estimated to around 7.9° for our Co-doped Ba-122 bicrystal films (fig. 2(c)), which is almost twice as large as that of YBa$_2$Cu$_3$O$_7$.[7]

The growth of epitaxial Fe with smooth surface on SrTiO$_3$ substrate is not straightforward, which is presumably due to the difference in crystal structure rather than to the lattice mismatch. As a result, epitaxial growth of Ba-122 is hardly achieved on Fe-buffered SrTiO$_3$. Indeed, fiber textured Fe is observed (fig. 3(a)), when it is prepared directly on SrTiO$_3$ substrate with the same procedure as on MgO substrates. In order to avoid this problem, an additional buffer layer of MgAl$_2$O$_4$ between Fe and SrTiO$_3$ substrate has been used. MgAl$_2$O$_4$ fits better to accommodate the lattice mismatch to Fe rather than MgO as shown in Table 1. Here, the lattice mismatch is defined as $(a_f - a_s)/a_f$, where $a_f$ and $a_s$ are the lattice parameters of Fe and substrates, respectively. Additionally, a small lattice mismatch of 3.4% between MgAl$_2$O$_4$ and SrTiO$_3$ may lead to good epitaxial growth of the MgAl$_2$O$_4$ buffer.

Fig. 3(b) shows the 110 pole figure of Fe on MgAl$_2$O$_4$-buffered SrTiO$_3$ fabricated by the same procedure described in sub-section 2.1. It is clear from fig. 3(b) that a perfect textured growth of Fe is realized (average $\Delta \phi_{\text{Fe}} = 0.61°$). During heating of Fe, evolution of the Fe-layer texture was observed through reflection high energy electron diffraction.

Figure 3: (a) The 110 pole figure measurement of Fe on bare SrTiO$_3$ and (b) MgAl$_2$O$_4$-buffered SrTiO$_3$. 


(RHEED), which is a similar observation of Fe on single crystalline MgO substrates. The RHEED images of the Fe layer on MgAl$_2$O$_4$-buffered SrTiO$_3$ after the high temperature annealing showed only streak patterns, indicative of surface smoothing. This architecture also prevents the current-shunting effect between SrTiO$_3$ and Co-doped Ba-122. This buffer architecture offers the opportunity to grow smooth epitaxial Fe layers on various perovskite substrate such as (La,Sr)(Al,Ta)O$_3$ which is almost twice as large as that of YBa$_2$Cu$_3$O$_7$.

Structural characterization of Ba-122/Fe/MgAl$_2$O$_4$ on [001]-tilt GB SrTiO$_3$ substrate by means of X-ray diffraction is summarized in figs. As stated earlier, in-situ Au layer deposition was conducted after the Ba-122 deposition, which explains the 111 reflection of Au in fig. (a). It is further clear from fig. (a) that only 00l reflections of Ba-122 with a small amount of the 110 reflection are observed. Fig. (b) displays the $\phi$ scans of the 103 Ba-122, the 110 Fe, and the 110 SrTiO$_3$. Clear 8 peaks from two grains with a rotational angle of 30$^\circ$ are apparent. Accordingly, an epitaxial Ba-122 film is formed on [001]-tilt SrTiO$_3$ bicrystal substrate. The onset $T_c=24$ K of the film is almost identical to that of Ba-122/Fe bilayers on single crystal substrates (fig. (c)). This value is higher than the intra-grain $T_c$ deposited on MgO bicrystal presented in fig. (a) due to a higher deposition temperature as well as different PLD targets (i.e. different Co concentration). The resultant bicrystal junctions show clear Josephson effects and the detailed studies can be found in Ref. 9.

4. Summary

High-quality Co-doped Ba-122 bicrystal films can be realized on both [001]-tilt MgO and SrTiO$_3$ substrates by employing Fe buffer layers via pulsed laser deposition. The additional MgAl$_2$O$_4$ buffer between Fe and SrTiO$_3$ is necessary for realizing epitaxial Ba-122. The characteristic angle of Co-doped Ba-122 in low field regime is about 8$^\circ$, which is almost twice as large as that of YBa$_2$Cu$_3$O$_7$.

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