MULTILAYER STUDIES AND APPLICATIONS IN TEMPLATE BI-EPITAXIAL DC SQUIDS.

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Abstract—Multilayer deposition for the creation of a well defined grain boundary, based on different in-plane orientations of c-axis oriented thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ layers on a single crystalline substrate, has been performed on three different kinds of substrates: (1102) oriented Al_2O_3 , (100) SrTiO_3 and (100) MgO . The multilayers consist of combinations of SrTiO_3 , MgO , CeO_2 and $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$. The $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ top layers on (1102) Al_2O_3 and (100) SrTiO_3 were polycrystalline. Best epitaxy has been achieved on the MgO substrates. An in-plane rotation of 18° was obtained between $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}/\text{CeO}_2$ and $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ on MgO using Ar ion structuring.

Josephson junctions and dc SQUIDS have been structured in the layers on MgO . Shapiro steps were observed. The $j_c R_n$ -product of the junctions at 4.2 K is of the order of 1 mV. The critical current decreases in good approximation linearly with increasing temperature; whereas the normal state resistance is nearly temperature independent. A maximum modulation voltage of 90 μV was observed at 4.2 K with a flux-to-voltage transfer $(\partial V/\partial \Phi)_{\text{max}} = 310 \mu\text{V}/\Phi_0$. At 77 K a maximum modulation voltage of 2 μV was measured.

I. INTRODUCTION.

In literature several ways to create Josephson junctions and dc SQUIDS using grain boundaries are described. Bicrystals of SrTiO_3 and Yttrium Stabilized ZrO_2 have been used [1,2] giving the possibility to vary the induced weak link angle between 0 and 45 degrees. The dependence of the critical current density on this angle is reported [1,2]. A second way for the creation of bi-epitaxial Josephson junctions makes use of different in-plane orientations of $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ on top of structured multilayers [3,4]. The first reported bi-epitaxial template grain boundaries were obtained using (1102) Al_2O_3 substrates, an MgO template layer and a SrTiO_3 buffer layer [3]. An improvement of this technique was achieved using SrTiO_3 substrates, an MgO template layer and a CeO_2 buffer layer [4]. All reported grain boundaries achieved in these ways have an in-plane rotation of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ of 45 degrees between both sides of the grain boundary.

For practical applications, like the dc SQUID, it is favourable to have the opportunity to place the Josephson junctions at any desired place on the substrate. Therefore epitaxial growth over the complete substrate is needed. In this report we discuss the growth and orientation of MgO and SrTiO_3 on (1102) Al_2O_3 substrates, MgO and CeO_2 on (100) SrTiO_3 substrates, and CeO_2 and $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ on (100) MgO . Electrical measurements

on Josephson junctions and dc SQUIDS structured in these bi-epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ layers on MgO will be presented.

II. EXPERIMENTAL

All layers have been deposited by laser ablation. The laser ablation equipment has been described before [5]. Deposition of the SrTiO_3 took place using a single crystal SrTiO_3 target. Deposition of MgO was done using a metallic Mg target. The CeO_2 was deposited using an amorphous/polycrystalline CeO_2 target. For the deposition of the different layers the deposition temperature T_{dep} , the oxygen pressure P_{O_2} , the energy fluence I , and the target-substrate distance d have been varied to obtain the desired growth orientations. Deposition of $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ took place at a deposition temperature of 750°C and an oxygen pressure of 25 Pa. Powder X-Ray Diffraction (PXRD) and single crystal X-Ray Diffraction have been used to check the (in-plane) orientations of the different layers. The deposited thin films on MgO showed best epitaxy and therefore these substrates have been used to produce bi-epitaxial Josephson junctions and dc SQUIDS. Structuring of the template layer and the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ layer was performed using Ar ion beam etching under an angle.

The dc SQUIDS were structured in a square washer design with a self-inductance of about 94 pH. The junctions were positioned at the outer edge of the washer. The washer was placed on the part of the substrate where the template CeO_2 was etched away. All the electrical measurements (IV and $V(\Phi)$) were 4 point measurements using 25 μm Al-wire bonded on Ag-contacts on the sample.

III. RESULTS ON MULTILAYERS.

a. (1102) Al_2O_3 substrates.

The bi-epitaxial grain boundaries on (1102) Al_2O_3 (r-plane sapphire) are based on different in-plane orientations of SrTiO_3 deposited directly on the Al_2O_3 and SrTiO_3 deposited on an MgO layer on Al_2O_3 [3,6]. MgO layers grown on Al_2O_3 with T_{dep} between 400 and 650°C , $P_{\text{O}_2} = 15$ Pa, $I = 1.2 \text{ J/cm}^2$, $d = 30 \text{ mm}$, and a constant number of laser pulses only show (h00) reflections in PXRD measurements. At $T_{\text{dep}} = 650^\circ\text{C}$ maximum intensity of the (h00) reflections is found. Raising T_{dep} only 50°C results in a disappearance of the (h00) reflections. Raising the temperature to 750°C for 20 minutes after depositing the MgO layer at 650°C does not change the PXRD pattern.

The growth of SrTiO_3 on Al_2O_3 has been studied varying T_{dep} , P_{O_2} , d , and I . T_{dep} has been varied between 650 and 770°C at constant P_{O_2} (15 Pa), I (1.2 J/cm^2), and d (30 mm). SrTiO_3 layers deposited at $T_{\text{dep}} > 700^\circ\text{C}$ show a mixture of (h00), (hh0) and (hhh) orientations, which has also been

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reported in literature [7]. If $T_{\text{dep}} < 700^\circ\text{C}$ the (hhh) orientation disappears. However, also the intensities of the (h00) and (hh0) reflections decrease, indicating a less crystalline layer. The relative amount of (hh0) material increases for decreasing T_{dep} . For $T_{\text{dep}} = 650^\circ\text{C}$, 88% of the crystalline material is (hh0) oriented. By varying P_{O_2} between 15 and 100 Pa at $T_{\text{dep}} = 680^\circ\text{C}$ the relative amount of (h00) oriented material can also be changed. For $P_{\text{O}_2} = 70$ Pa the maximum amount of (h00) oriented material is found: 70%. The rest of the crystalline material has the (hh0) orientation.

Changing the deposition rate by varying d between 25 and 40 mm, and I between 1.2 and 3.0 J/cm^2 did not influence the relative amounts of the different orientations significantly, when deposited at $T_{\text{dep}} = 680^\circ\text{C}$ and $P_{\text{O}_2} = 25$ Pa.

Because of the polycrystalline SrTiO_3 on (1102) Al_2O_3 , the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ on top of it will also be polycrystalline. As a result no reproducible Josephson junctions and dc SQUIDS can be structured all over the substrate.

The in situ deposition of SrTiO_3 on a MgO layer on Al_2O_3 resulted in (h00) oriented SrTiO_3 for $T_{\text{dep, MgO}} = 630^\circ\text{C}$ and $T_{\text{dep, SrTiO}_3} = 730^\circ\text{C}$. Both layers were deposited at $P_{\text{O}_2} = 15$ Pa, $I = 1.2 \text{ J/cm}^2$, and $d = 32$ mm.

b. (100) SrTiO_3 substrates.

CeO_2 grown directly on (100) SrTiO_3 and CeO_2 grown on a MgO layer on (100) SrTiO_3 show different in-plane orientations [4,8], creating a grain boundary in the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ top layer. MgO deposition took place at T_{dep} between 560 and 630°C , $P_{\text{O}_2} = 15$ Pa, $I = 1.2 \text{ J/cm}^2$, and $d = 35$ mm. PXRD patterns only show (h00) oriented MgO for all layers.

The growth of the CeO_2 directly on the substrate took place at T_{dep} between 700 and 750°C , $P_{\text{O}_2} = 20$ Pa, $I = 2.0 \text{ J/cm}^2$, and $d = 35$ mm. All layers showed mainly (h00) oriented material. Small amounts (0.1%) of (hhh) oriented material were observed.

The deposition of CeO_2 on MgO, deposited at conditions mentioned above, on SrTiO_3 resulted in a polycrystalline CeO_2 layer consisting of 60% (h00), 35% (hhh), and 5% (311) oriented material for $T_{\text{dep}} = 730$ – 750°C , $P_{\text{O}_2} = 20$ Pa, $I = 2.0 \text{ J/cm}^2$ and $d = 35$ mm.

As with the Al_2O_3 substrates the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ on the CeO_2 is polycrystalline, making it impossible to structure reproducible Josephson junctions all over the substrate.

c. (100) MgO substrates.

Using MgO substrates and a CeO_2 template layer it is possible to create an in-plane rotation of $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ on CeO_2 on MgO with respect to $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ deposited directly on MgO. By using an MgO substrate instead of a laser deposited MgO layer on a SrTiO_3 substrate the epitaxy of the CeO_2 layer on top of the MgO is considerably improved: CeO_2 layers deposited on an MgO substrate at the same deposition conditions as CeO_2 on an MgO layer show 95% (h00) oriented material and 5% (hhh) oriented material instead of 60% (h00) oriented CeO_2 as mentioned before. $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ grown on the MgO substrate and on the CeO_2 on MgO both show mainly (00h) reflections for $T_{\text{dep}} = 750^\circ\text{C}$ and $P_{\text{O}_2} = 25$ Pa. The in-plane orientation of the CeO_2 layer with respect to the MgO has been checked using XRD. In figure 1 a phi-scan of the (113) CeO_2 and the (204) MgO reflection is given. We find an angle of 18 degrees instead of the reported 0 degrees between the [100] direction of both MgO and CeO_2 [4]. Using the near coincidence site lattice theory a misfit of 1.5% is found for this orientation, compared to a 25% misfit for the cubic to cubic

Fig. 1 Phi-scan of a CeO_2 layer on an MgO substrate. Shown are the (204) MgO reflection (bottom), and the (113) CeO_2 reflection (top).

Fig. 2 Pole diagram of $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ on Ar ion beam etched MgO. A=(206) $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$, B=(108) $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$, C=(204) MgO.

orientation.

The CeO_2 layer is structured using standard photolithography and Ar ion beam etching with a beam voltage of 500 V and a beam current of 10 mA. By using this structuring technique also the toplayer of the MgO substrate not covered with photoresist is etched. The pole diagram of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ layer on this Ar etched MgO is given in figure 2. The [110]

Fig. 3 Pole diagram of $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ on CeO_2 on MgO .
A=113° CeO_2 , B=108° $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$.

direction of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ is parallel with the [100] direction of the MgO [9]. On CeO_2 the in-plane orientation of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ is [110] $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ // [100] CeO_2 as can be seen in figure 3. The resulting angle between the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ deposited on the CeO_2 on MgO and directly on the etched MgO is 18°.

IV. DC SQUIDS.

The 18° grain boundaries on MgO substrates with a CeO_2 template layer have been used to structure dc SQUIDS. In figure 4-7 electrical measurements on one dc SQUID are presented. The washer type dc SQUID consists of two 8 μm wide junctions separated by a 5 μm wide slit. The thickness of the CeO_2 layer is 50 nm, the $\text{YBa}_2\text{Cu}_3\text{O}_{7-d}$ has a thickness of 200 nm. The CeO_2 layer is etched under an angle of 20 degrees with the substrate surface.

fig. 4 IV curves at 40 K for a dc SQUID without (a) and with (b) microwave irradiation ($\nu=10$ GHz).

Fig. 5 Temperature dependence of the critical current.

In figure 4 the IV curves at 40 K for this SQUID with (b) and without (a) microwave ($\nu=10$ GHz) irradiation are given. I_c at 40K is 115 μA , R_n equals 1.07 Ω ; the resulting $j_0\rho_n$ value is 123 μV . In general we found values of $j_0\rho_n$ in the order of 1 mV at 4.2 K and 1-5 μV at 77 K, although relatively large variations (up to 300%) occurred.

Since the normal state resistance was found to be nearly independent of temperature, the temperature dependence of the $j_0\rho_n$ -product follows that of the critical current. In figure 5 it is shown for this dc SQUID that the critical current is nearly linear decreasing with increasing temperature. Such a behavior was also found by others for different types of grain boundary junctions [4,10,11]. So far however it is not clear if the grain boundary junctions should be regarded as SNS, SIS, SSeS or of another type.

Figure 6 shows the flux-to-voltage modulation of the same dc SQUID measured at different values of the bias current at 77 K. The maximum modulation voltage at this temperature was 2 μV . Figure 7 shows the voltage modulation depth as a function of temperature for this dc SQUID.

Fig. 6 $V(\phi)$ dependence of the dc SQUID at 77K

Fig. 7 Modulation depth as a function of T for the dc SQUID of fig. 4.

The highest modulation voltage we have observed was 90 μV at 4.2K, with a flux to voltage transfer $\partial V/\partial \Phi = 310 \mu\text{V}/\Phi_0$. The screening parameter β_L is approximately 8 at this temperature.

Fig. 8: IV-characteristic ($n\Phi_0$ and $(n+1/2)\Phi_0$ branch) of another dc SQUID at 4.2K

Often a crossing of the $n\Phi_0$ and the $(n+1/2)\Phi_0$ branch was observed with increasing bias-current as indicated in figure 8. This also occurs in low T_c dc SQUIDS and it can be modeled by taking a shunt impedance into account [12-15]. If we assume the junctions to be capacitively and resistively shunted, resonances in the $(n+1/2)\Phi_0$ -branch will occur at the resonance voltage:

$$V_{\text{res}} = \Phi_0 / [2\pi(LC/2)^{1/2}]. \quad (1)$$

In our case (figure 8) L is approximately 94 pH, $I_c = 75 \mu\text{A}$ and $R_n = 7.7 \Omega$. The resonance voltage is taken as 175 μV . This results in a calculated capacitance of $7.3 \cdot 10^{-14}$ F or $2.2 \cdot 10^{-2} \text{ Fm}^{-2}$ of the junctions. This capacitance value implies that $\beta_c = 1.0$, which is in agreement with the non-hysteretic behavior of the junctions.

V. CONCLUSIONS.

The creation of a reproducible 45° template grain boundary using (1102) Al_2O_3 substrates, an MgO template layer and a SrTiO_3 buffer layer is not possible all over the substrate because of polycrystalline growth of the SrTiO_3 on the Al_2O_3 substrate. Also the creation of a reproducible grain boundary on SrTiO_3 substrates with an MgO template layer and a CeO_2 buffer layer is not possible with our deposition conditions. Deposition of CeO_2 directly on an MgO substrate and Ar ion beam etching of the deposited CeO_2 leads to a grain boundary with an in plane angle of 18 degrees between the $\text{YBa}_2\text{Cu}_3\text{O}_{7.4}$ on both sides of the grain boundary.

Based on these grain boundary junctions, dc SQUIDS were fabricated. Voltage modulation was observed at temperatures up to 77 K. The critical current shows a nearly linear temperature dependence, whereas the normal state resistance is nearly temperature independent. Values in the order of 1 mV were found for the $j_c \rho_n$ -product at 4.2 K.

From resonances in the IV characteristics an estimation of the capacitance of the junctions is made. We found a value of $2.2 \cdot 10^{-2} \text{ Fm}^{-2}$ for the capacitance per unit area of the junctions.

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