CeO₂ as insulation layer in high T_c superconducting multilayer and crossover structures

M. A. A. M. van Wijck,^{a)} M. A. J. Verhoeven, E. M. C. M. Reuvekamp, G. J. Gerritsma, D. H. A. Blank, and H. Rogalla

University of Twente, Department of Applied Physics, Low Temperature Division, Enschede, The Netherlands

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We present a study of the electrical properties of insulating CeO₂ layers in combination with superconducting (Y/Dy) Ba₂Cu₃O_{7- δ} (RBCO) films over ramps and in crossover structures. CeO₂ is frequently used as a buffer layer, or template layer for biepitaxial grain boundary junctions, but can also be used as an insulating layer in ramp-type junctions and other multilayer structures. Epitaxial thin films of CeO₂ were deposited by pulsed laser ablation using SrTiO₃ substrates. We characterized the insulating performance of CeO₂ thin films in terms of breakdown field E_{bd} and the relative dielectric constant ϵ_r . For 80 nm thick CeO₂ at 77 K we found $E_{bd}=1 \times 10^6$ V/cm, using a 1 nA/100 μm^2 breakdown criterion, which gives us a specific resistivity of $\rho \geq 10^9$ Ω cm up to breakdown. From capacitance measurements on planar RBCO/CeO₂/RBCO structures we obtained for the dielectric constant: $\epsilon_r \approx 15$. The texture of CeO₂ in combination with RBCO on ramped surfaces, simulated by SrTiO₃ (STO) (103) substrates having their normal tilted by 18° away from the STO[001] direction, has been studied by x-ray diffraction. © 1996 American Institute of Physics. [S0003-6951(96)02604-6]

High-quality heteroepitaxial multilayers of superconductors and insulators are required for high T_c superconducting electronic devices, like ramp-type junctions and crossover structures in multiturn coils. For the use in high-frequency applications, an insulator having a low dielectric constant is preferred to reduce capacitive shunting. Materials often used as insulators in high T_c superconducting devices are PrBa₂Cu₃O₇ (PBCO)¹ and SrTiO₃ (STO).² The use of PBCO has some disadvantages for its relatively low resistivity at 77 K and the anisotropic growth. The last property enhances the formation of granular films, and leaks are likely to occur at boundaries between a - and c -axis oriented grains. The main drawback of STO is its large dielectric constant [$\epsilon_r \approx 300$ for thin films (Ref. 2)], which severely limits its applicability in high-frequency devices. CeO₂ has been shown to be useful as a planar insulator.³ It has a cubic crystal structure with lattice constant $a_0=5.411$ Å, and CeO₂[100] is commensurate with YBCO[110], corresponding to a relative rotation of 45° of the lattices. The very simple crystal structure, containing only one metal ion and oxygen, makes CeO₂ an easy material to grow by pulsed laser deposition, as no problems with the stoichiometry are to be expected. The use of CeO₂ in ramp-type junctions and crossover structures requires a nearly perfect heteroepitaxy with RBCO, both on top of flat surfaces and on artificially created ramped surfaces. The rotation of CeO₂ relative to RBCO, however, might induce grain boundaries or cracks when growing on a ramped surface. Reuvekamp *et al.*⁴ showed that the preferential orientation of RBCO grown on YSZ is with the c -axis perpendicular to the local YSZ substrate surface. The difference in orientation in the upper

YBCO layer on the ramp and on flat surfaces then caused cracks and grain boundaries at the inclination points. To simulate this ramped surface, STO(103) substrates were used. Here, the normal of the substrate is 18° tilted away from its [001] direction. This allows us to use x-ray diffraction to investigate the texture of CeO₂ and YBCO grown on tilted surfaces. In Ref. 5 it was shown that the c axis of YBCO grown on STO(305) substrates is parallel to the STO[001] direction, so in the same crystallographic orientation as it is on STO(001) substrates. If the crystallographic orientation of RBCO on CeO₂ grown on tilted substrates is the same as it is on STO(001), growth over a ramp will not necessarily lead to formation of cracks or grain boundaries at the inclination points.

Thin films of DyBCO were deposited onto STO(001) substrates, using off-axis rf-magnetron sputtering from a single stoichiometric target, as was described in Ref. 6. The deposition of the CeO₂ was done by pulsed laser ablation, using an eximer laser (XeCl mode; $\lambda=308$ nm), 75 mJ pulse, pulse rate 1 Hz, spot size 6.4 mm², from a 99.9% pure CeO₂ target. This process was carefully optimized by varying the oxygen pressure and the substrate temperature (ranging from 700 to 830 °C). At low oxygen pressures we saw large precipitates (~200 nm) on the surface, but they disappeared for high enough oxygen pressures (≥ 10 Pa). Here, the growth rate was about 1 Å/s. Films deposited at lower temperatures appeared smoother through the optical microscope, but for temperatures below 740 °C, submicron sized pinholes in the CeO₂ layer could be observed by scanning electron microscopy. X-ray diffraction (XRD) analysis, using a Philips MRD diffractometer, showed that CeO₂(001) grows on RBCO(001) with [100] CeO₂ parallel to [110]RBCO. The mosaic spread was smaller than the detection limit, full width at half-maximum (FWHM) $<0.5^\circ$, within the whole

^{a)}Electronic mail: w.a.m.aarnink@tn.utwente.nl

FIG. 1. (a) DBCO strip line, meandering over a ramp structured in CeO_2 (b) linear array of crossover structures.

range of temperatures used in the optimization. We found for the optimum conditions for laser ablation of CeO_2 a substrate temperature of 740°C at $P(\text{O}_2)=10\text{ Pa}$. On top of the CeO_2 a thin film of YBCO was deposited by pulsed laser ablation. The same conditions were used for the deposition on $\text{STO}(103)$ substrates.

To determine the critical current density of the top electrode grown over ramps, we used two types of test structures. The first structure [see Fig. 1(a)] consists of a (RBCO) superconductor strip line, meandering over a ramp in a CeO_2 layer, crossing it six times. The second one [Fig. 1(b)] is a linear array of five crossover structures, where the top electrode has to surpass lines in a bottom electrode, insulated by a layer of CeO_2 . For this, a thickness of 80 nm was used for the CeO_2 as well as the DBCO films. Ramps were structured using standard photolithography by Ar-ion beam milling with an incidence angle of about 45° . After stripping photoresist in acetone, the ramps were cleaned by Ar-ion beam milling as is described in Ref. 4 to remove residues of photoresist and surface contamination due to the *ex situ* process. The etched ramps have an angle of about 20° .

The electrical measurements were performed in a flow cryostat. We used $1\text{ }\mu\text{V}$ as the critical current criterion, and a $1\text{ nA}/100\text{ }\mu\text{m}^2$ breakdown field criterion which is in the sensitivity range of our measurement setup. For the determination of the relative dielectric constant of the CeO_2 film, a multilayer of YBCO (100 nm)/ CeO_2 (80 nm)/YBCO (100 nm) was deposited. This multilayer was structured into squares of $1000\times1000\text{ }\mu\text{m}^2$ and $500\times500\text{ }\mu\text{m}^2$, and their capacitance was measured using a Philips HP4194A Gain Phase Analyzer.

The superconducting layers on top and under CeO_2 thin films had a critical temperature of about 89 K , which is about the same as for the layers fabricated directly on STO . Furthermore, XRD measurements revealed that the *c*-axis length of RBCO was not changed by the CeO_2 deposition, indicating sufficient oxygen loading. Therefore, we conclude that

FIG. 2. Pole plots (Ψ, ϕ) showing reflections from the (111) planes of CeO_2 . The numbers between brackets, placed near the reflections, then give the expected orientations of CeO_2 in the sample; (a) CeO_2 on $\text{RBCO}(001)$; (b) CeO_2 on $\text{STO}(001)$; and (c) CeO_2 on $\text{STO}(103)$.

the use of CeO_2 is not affecting the quality of superconducting layers.

XRD measurements on YBCO films showed that $\text{YBCO}(001)$ grows epitaxially on $\text{CeO}_2(001)$ with $\text{YBCO}[110]$ parallel to $\text{CeO}_2[100]$. The pole plots in Figs. 2(a)–2(c), obtained from XRD measurements on (a) CeO_2 on $\text{YBCO}/\text{STO}(001)$, (b) CeO_2 on $\text{STO}(001)$, and (c) CeO_2 in $\text{YBCO}/\text{CeO}_2/\text{STO}(103)$, show that CeO_2 grows with $\text{CeO}_2[001]$ parallel to $\text{STO}[001]$ for each of the samples. This means that the multilayer structure is tilted by the same amount as the $[001]$ crystal axis of substrate. In Figs. 2(b) and 2(c) there are indications of other signals just above the noise level, possibly from traces of the $\text{CeO}_2(111)$ orientation. Generally, however we see that growth of multilayers of YBCO and CeO_2 is truly (hetero-) epitaxial, both on flat and on ramped surfaces. This strongly implies that CeO_2 can be used without formation of cracks on ramps in RBCO.

The measured critical current density J_c for the YBCO meander structures on the ramps in CeO_2 [Fig. 1(a)] at 77 K is $1\times10^6\text{ A/cm}^2$. For the crossovers surpassing both YBCO and CeO_2 [Fig. 1(b)], it is $J_c=8\times10^4\text{ A/cm}^2$. Due to the finite thickness (80 nm) of the electrodes on the ramps, the current has to flow locally in the *c* direction. In this direction, the critical current density is about ten times smaller than in the *ab* plane.⁷ This explains the reduced critical current densities for the crossover and meander structures with respect to the critical current density of a bridge in a single YBCO film. It also accounts for the difference in J_c between the different test structures: The top electrode has to cross only 80 nm high steps in the meander structures, whereas it has to surmount 160 nm in the crossovers. We think that the reduction of J_c is exclusively caused by the increased step height, as from our XRD analysis, the whole trilayer is expected to be *c*-axis oriented. These evidenced that there is hardly any increase in probability for the formation of additional grain boundaries at the step.

The breakdown characteristics (Fig. 3) of the CeO_2 plate capacitors are symmetric and we find a breakdown field of $E_{\text{bd}}=1\times10^6\text{ V/cm}$. This yields a specific resistance of $\rho\geq10^9\text{ }\Omega\text{ cm}$ up to breakdown at 77 K for the 80 nm thick CeO_2 film. A more conservative breakdown criterion, as was applied by Walkenhorst *et al.*,³ would because of the shape

FIG. 3. Typical IV characteristic at 77 K for a planar RBCO/CeO₂/RBCO trilayer structure ($A=100\ \mu\text{m}^2$), with 80 nm thick CeO₂.

of the breakdown characteristics (Fig. 3) lead to even higher estimates of the specific resistance. Clearly, the specific resistance for CeO₂ is much better than for PBCO thin films, also at low temperatures,¹ and slightly better than the resistance found for STO films.⁸ From capacitance measurements of a planar structure with 80 nm thick CeO₂ between two superconducting RBCO films we obtain $\epsilon_r \approx 15$ (in the frequency range: 10 kHz–100 MHz) for the relative dielectric constant, which is in reasonable agreement with values reported by others (e.g., Ref. 2).

These results show that for application purposes, 80 nm thick CeO₂ layers are well suited. It would be beneficial in the future, to perform a systematic study as a function of layer thickness, to gain more insight in the insulating performance of CeO₂ layers.

In conclusion, our investigations have demonstrated the (hetero-) epitaxial growth of CeO₂ in combination with RBCO thin films on STO substrate from which the [001] direction was tilted from the surface normal by 0° [STO(001) substrates] or 18° [STO(103) substrates]. The CeO₂ layers were always very well textured, having the CeO₂[100] parallel with the STO[110] for both types of substrates used.

The electrical measurements show that the properties of superconducting RBCO layers are unaffected by the use of CeO₂ films. For 80 nm thick CeO₂ we found a breakdown field of $E_{\text{bd}} \approx 1 \times 10^6\ \text{V/cm}$ (using $1\ \text{nA}/100\ \mu\text{m}^2$ as the breakdown criterion) yielding a resistivity $\rho \geq 10^9\ \Omega\ \text{cm}$ at 77 K up to breakdown. From capacitance measurements in the frequency range: 10 kHz–100 MHz, done on a planar structure with 80 nm thick CeO₂ between two superconducting RBCO films, we obtained the relative dielectric constant $\epsilon_r \approx 15$.

The high value breakdown field, the low value of the dielectric constant and the good heteroepitaxy of RBCO and CeO₂ over ramps clearly demonstrates the advantage of using a CeO₂ insulating layer in ramp and crossover structures.

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