Superconducting $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ Thin Films and Heterostructures on Sapphire

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Abstract—**Superconducting $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ (NCCO) thin films have been made on Yttria-Stabilized Zirconia(YSZ) buffered sapphire. The films are epitaxially grown and highly in-plane oriented. X-ray diffraction shows the c-axis of the film normal to the surface of the substrate. The width of rocking curve is 0.2^o and the RBS channeling yield is 9%, indicating high crystallinity of the film. Cross-sectional transmission electron microscopy images reveal a sharp interface between NCCO and YSZ. The microwave surface resistance of NCCO films on YSZ buffered sapphire at 9.6 GHz was measured and a value of 80 $\mu\Omega$ (at 4.2 K in zero dc magnetic field) was obtained, which is comparable to $Y_1Ba_2Cu_3O_{7-y}$ (YBCO) films at the same reduced temperature. A trilayer structure of YBCO/$SrTiO_3$/NCCO on YSZ buffered sapphire has been fabricated with all layers oriented, in which both the YBCO and NCCO layers are superconducting.**

I. INTRODUCTION

Electron-doped $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ (NCCO) superconducting thin films may be very useful for the potential application of superconductive electronics due to its chemical stability (it is not sensitive to the environment) and long in-plane coherence length (~ 80 Å). There have been extensive studies on the fabrication of high quality NCCO films on various substrates recently [1-3]. Among the commonly used substrates, sapphire is the most favorable for technological applications in microwave devices because of its low dielectric constant and low loss tangent [4]. On the other hand, a suitable buffer layer is needed on the substrate to avoid interactions between the sapphire and the cuprate thin films, as has been extensively explored with *p*-type $Y_1Ba_2Cu_3O_{7-y}$ (YBCO) thin films [4-7]. However, few studies have been carried out in the NCCO system. Recently, we have found that the results from YBCO cannot be directly applied to NCCO films and the fluorite type substrate of YSZ turned out to give the best result [8]. In this paper we report the detailed preparation and characterization of high quality NCCO films on sapphire and the latest success of the fabrication of a YBCO/STO/NCCO trilayer heterostructure on sapphire substrates which could be used for superconducting microwave electric effect devices (FET) [9].

II. EXPERIMENT

Pulsed laser deposition (KrF excimer laser with a wavelength of 2480 Å) was employed for the growth of NCCO on sapphire. A multitarget carousel was used to deposit the buffer layer and the multilayers *in situ*. The laser beam energy density is 1.5 J/cm^2 as it hits a target of NCCO or YSZ. It was shown that N_2O reactive gas yields much better NCCO films [1,2] and in this work all the deposition was done in 200 mTorr N_2O. The substrates were held at different temperatures from 720 - 820 ^oC. Different buffers layer were tested before the deposition of the NCCO layer. After the deposition of NCCO, the samples were cooled down after evacuating the chamber to 10^{-5} Torr. It took one hour to reach room temperature. The films were characterized for superconducting properties using ac susceptibility and dc resistance methods. The surface resistance R_s was measured at 9.6 GHz in a superconducting Nb cavity with a variable-temperature sample stage. The films were analyzed systematically by a Siemens 5000 four-circle X-ray diffractometer. θ–2θ scan, ϕ-scan, and ω-scan were carried out to fully characterize the films. Rutherford backscattering (RBS) channeling analysis, cross-sectional transmission electron microscopy (TEM), and Atomic Force Microscopy were also used for structural characterization of the samples.

III. RESULTS AND DISCUSSION

A. Choice of Buffer Layers

Good buffer layer materials for YBCO growth can not be directly applied to NCCO films, as we mentioned early, for the following reasons: 1) severe chemical reactions occur between NCCO and CeO_2 and MgO; 2) a considerable amount of a-axis NCCO growth occurs on $Pr_1Ba_2Cu_3O_7$ or $BaTiO_3$ buffer layers; 3) the superconducting properties of

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NCCO films on $LaAlO_3$, $NdGaO_3$ (NGO), $SrTiO_3$(STO), and YSZ are different [10]. So two different types of buffer layer were used to investigate the influence of lattice mismatch and interface chemical stability on the transport and structural properties of the NCCO films on sapphire. They are perovskite materials such as (100) LAO, (100) NGO, (100) STO, and the fluorite material (100) YSZ. Generally the superconducting transition temperatures for thick films (>2000Å) grown under optimized conditions are the same for different substrate materials and only show slightly different transition widths. The normal state resistivity also varies slightly. However, the superconducting transition temperature (T_{co}) of the films decreases significantly as the thickness of the films is reduced below 1000Å [8]. On the other hand, the same thickness films grown on YSZ show the highest T_{co}. In other words, films deposited on fluorite YSZ substrate showed much better superconducting properties than the films grown on perovskite substrates. The major differences between perovskite (say LAO) and YSZ substrates can be attributed to, in addition to the lattice mismatch, different atomic structure at the film-substrate interface. In the case of a fluorite type structure (YSZ), an oxygen sublattice is continuous across the NCCO-substrate interface thus the interfacial electrostatic energy is reduced and less strain retains in the films [10,11]. The YSZ material was thus identified as the best buffer layer and hence was chosen to grow epitaxial NCCO films on $(1\bar{1}02)$sapphire substrates in this work.

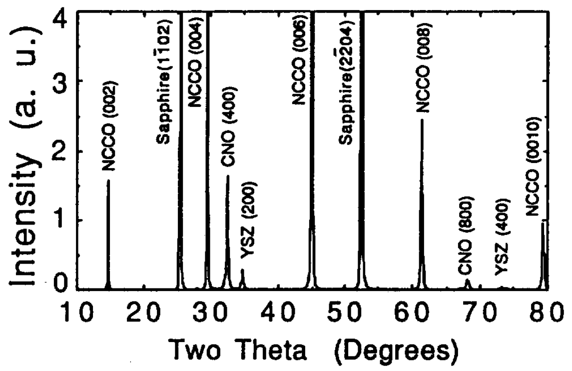

Fig. 1. θ–2θ X-ray diffraction spectrum of 2500Å-thick NCCO on YSZ buffered sapphire.

B. Relationship of Epitaxy

A typical X-ray θ–2θ spectrum of NCCO films on a YSZ buffered sapphire substrate is shown in Fig 1, which is very similar to that of NCCO on bare YSZ substrates [10]. The film is aligned with the c-axis perpendicular to the surface of $(1\bar{1}02)$ sapphire, with a small variation of ~ 0.2° as measured by a rocking curve scan. An X-ray φ scan (Fig. 2) shows the in-plane epitaxial relationship to be (110)NCCO//(001)YSZ, i.e., the NCCO film is rotated by 45° with respect to YSZ. Four clean peaks in the X-ray φ-scan spectrum indicate that the NCCO film on YSZ buffer is of single domain in contrast to the case of YBCO on YSZ, where multiple-in-plane orientations exist [12]. High resolution cross-sectional TEM further confirmed the in-plane orientation [11]. Also TEM shows that the NCCO films do not contain any stacking faults and the interface between NCCO and YSZ is quite sharp.

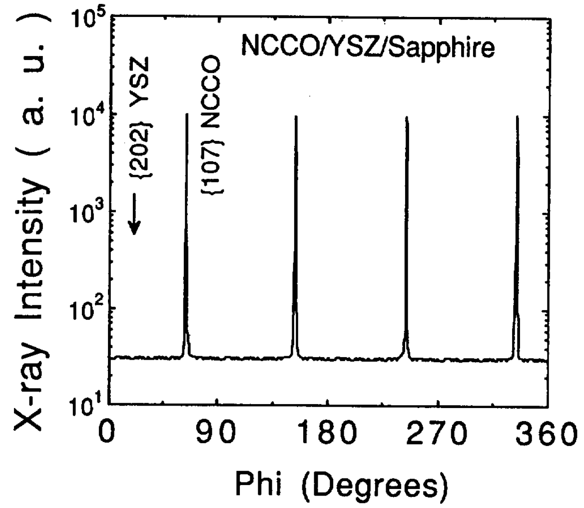

Fig. 2. X-ray diffraction φ scans of NCCO{101} family of reflections for a film grown on YSZ buffered sapphire. Four clean peaks indicate single domain structure. One of the four peak positions from the buffer layer is shown by arrow for clarity.

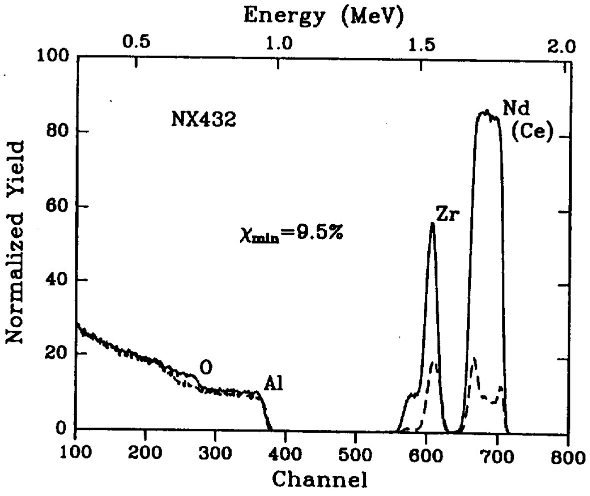

Fig. 3. RBS/Channeling spectra for an NCCO/YSZ/sapphire film.

There still exist grains of insulating $Ce_{0.5}Nd_{0.5}O_{1.75}$(CNO) in films grown on YSZ buffered sapphire. The concentrations of that grains is similar to films grown on LAO [3,10]. The RBS/channeling spectra in Fig. 3 reveal a minimum yield of ~ 9% from the film grown on YSZ/sapphire.

C. Microwave Properties

Though the sharp transition width of 0.2K obtained from the ac susceptibility and the low residual dc resistivity of 60 μΩcm at 25K indicate a high degree of homogeneity and purity, the most stringent evaluation of the quality of superconducting thin films is surface resistance (R_s) [1,8]. Fig. 4 shows the microwave loss at 9.6 GHz measured on a sample of NCCO/YSZ/sapphire using a Nb cavity which has a resolution of ~10 μΩ [13]. The surface resistance of the film at 4.2K in zero external magnetic field is only 80 μΩ, which is close to the best values of YBCO films at the same reduced temperature [14]. It should be added that all the samples used for microwave measurements in this work are as-fabricated, without any surface post-treatment.

It should also be mentioned that in previous work we observed a behavior consistent with s-wave BCS type behavior from the penetration depth (frequency shift) measurement using films of NCCO/LAO with very high residual R_s of 2.5 mΩ [13]. Interestingly, with a NCCO/YSZ/sapphire film in which the residual R_s is two order of magnitude lower than that of a film on $LaAlO_3$ [1,8], we have observed the same exponential behavior in the temperature dependent penetration depth with the same reduced energy gap value of $2\Delta(0)/kT_c=4.1$ [15]. Furthermore, as can be seen in Fig. 4, the temperature dependence of R_S is also consistent with BCS s-wave behavior. The similar activated temperature dependence observed in both NCCO/LAO and NCCO/YSZ/sapphire films with quite different residual R_S's indicates that this behavior is not sensitive to disorder, implying that NCCO is more like a conventional superconductor such as Nb.

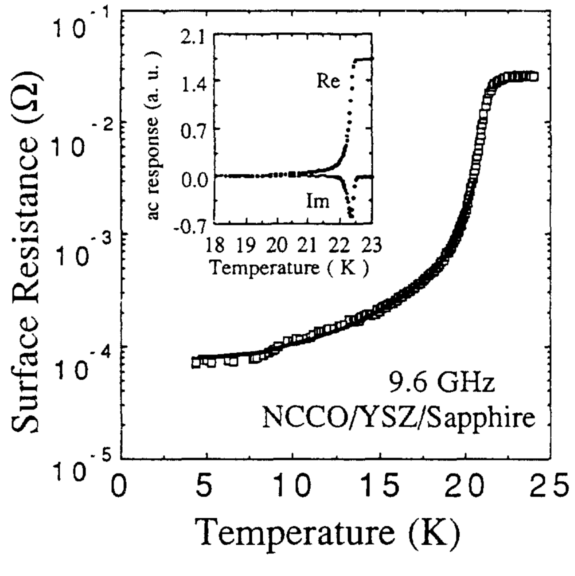

Fig. 4. Microwave surface resistance R_s at 9.6 GHz vs. temperature for a film of 2500Å NCCO on YSZ/sapphire. A residual R_{so} value of ~80 μΩ was observed at T = 4.2 K for this sample, the value comparable to that of YBCO at the same reduced temperature. The solid line is the best fit using the BCS formula $R_s = CT^{-1}\exp\{-\Delta(T)/kT\} + R_{so}$ with $2\Delta(0)/kTc = 4.1$ as described in the text. Inset is the ac susceptibility measurement. The dc resistivity before the superconducting transition is 60 μΩcm for this sample.

D. YBCO/STO/NCCO Heterostructure

Since the charge carriers in YBCO are holes but in NCCO the carriers are electrons, when a voltage is applied to the trilayer structure with YBCO and NCCO as the top and bottom electrodes, the modulation would be enhanced (or depleted) significantly [9,16]. Motivated by this idea, we have made a heterostructure of YBCO/STO/NCCO on sapphire. The deposition parameters are similar to the standard YBCO/NCCO bilayers [17]. X-ray diffraction shows an epitaxial growth from the bottom to the top layer through the structure. Fig. 5 is the normal coupled scanning (θ–2θ scan). All the peaks can be indexed well as shown in the figure. The multilayer structure is c-axis oriented out of the plane. The φ-scans (Fig. 6) reveal the in-plane relationship, which is YBCO(100)//STO(010)//NCCO(100) //YSZ(110). The rocking curve width for the bottom layers(YSZ and NCCO) is 0.3° and for top layers (STO and YBCO) is 0.6° due to the reduced deposition temperatures for YBCO layer [17]. After the deposition of YBCO layer the sample was cooled down in the optimized condition, which is to cool the sample in 200 mTorr N_2O to 350 °C and then

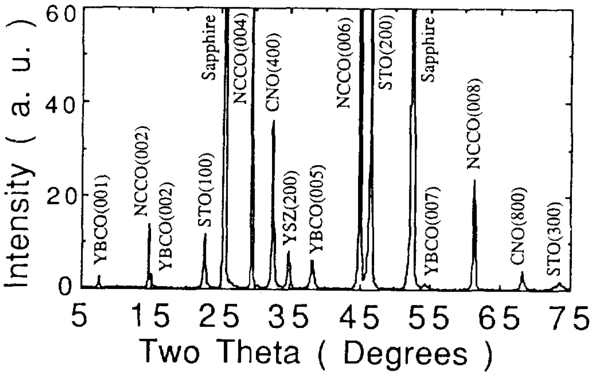

Fig. 5. X-ray diffraction patterns for a multilayer film of (800Å)YBCO/(3000Å)STO/(1100Å)NCCO/(500Å)YSZ/sapphire.

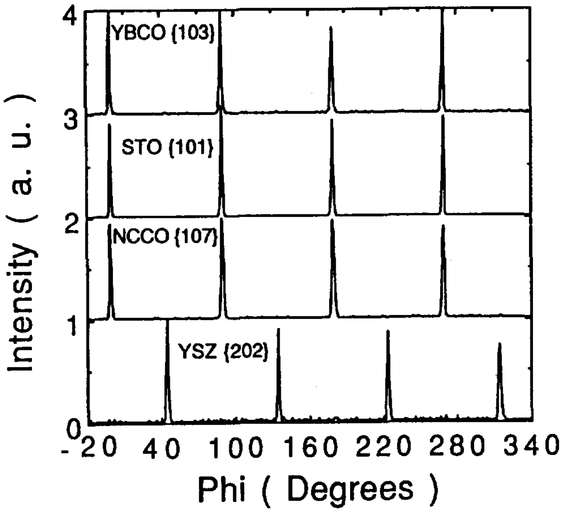

Fig. 6. Φ - scans for the sample showing in Fig. 5. Four clean peaks for each layer indicate that the layer is highly in-plane oriented. Among different layers the epitaxial orientation is also locked with each other. The spectra is normalized and offset for clarity.

anneal in 200 Torr pure O_2 at this temperature for 10 minutes [17]. The heterostructure shows a T_c of 88K for YBCO layer and a T_c of 15K for the bottom NCCO layer by a mutual inductance measurement. A relative dielectric constant of ~ 200 was obtained from the STO layer. Extensive work on the optimization of the dielectric properties of STO and a test of the microwave FET are underway.

IV. CONCLUSIONS

High quality NCCO superconducting thin films with low microwave surface resistance of 80 μΩ at 4.2K have been fabricated on sapphire substrates with a YSZ buffer layer. The fluorite-type buffer layer is favorable for the growth of the NCCO film, giving better structural and superconducting properties. The exponential temperature dependence of surface resistance is observed both in earlier films on LAO and improved films on sapphire, which indicates this thermal activation behavior is not sensitive to the disorder in NCCO system. A heterostructure consisting of dielectric STO, *n*-type superconductor NCCO, and *p*-type YBCO has been prepared in which both YBCO and NCCO layers are superconducting.

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