Sr_2AlTaO_6 / $YBa_2Cu_3O_7$ HETEROSTRUCTURES FOR SUPERCONDUCTING DEVICE APPLICATIONS

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Abstract--Using pulsed laser deposition, we have prepared epitaxial heterostructures of a new insulating perovskite Sr_2AlTaO_6 (SAT) and the high temperature superconductor $YBa_2Cu_3O_7$ (YBCO) on $LaAlO_3$ substrates. X-ray rocking curve and Rutherford backscattering channelling measurements on [001] SAT films on [001] $LaAlO_3$ substrates yield full width at half-maximum of < 0.3° and minimum backscattering yield χ_{min} of 5 %, respectively, indicating good crystallinity. From capacitance measurements on SAT films, the real part of the relative dielectric constant ε_r is found to be ~ 23 - 30, with ~ $6x10^7$ V/m static breakdown electric field. A 100 nm x 10 μm x 50 μm YBCO film on SAT shows zero-field critical current density of ~ $1.3x10^6$ A/cm^2 at 77 K. Using a dielectric resonator at 24.5 GHz, we have measured the resistive energy dissipation at the surface of a YBCO/SAT/YBCO trilayer, yielding effective surface resistance value of ~ 12 mΩ at 77 K, which is within a factor of four of the value obtained for a single layer YBCO film.

I. INTRODUCTION

High quality epitaxial insulating layers are necessary for various electronic applications of high temperature superconductors (HTS), such as multiple level interconnects, tunnel barriers and dielectric spacers [1,2]. In general, the complex chemistry and perovskite crystal structure of HTS materials impose severe restrictions on the multilevel film growth conditions and also limit the number of compatible insulating materials [3]. First, lattice constant and thermal expansion coefficient match, and minimal chemical reaction at the HTS-insulator interface are essential for high quality epitaxial growth. Second, application specific considerations preclude many of the already few chemically and structurally compatible insulators, e.g., low-loss and low-dielectric constant materials are desirable in high-frequency, high-speed device applications. Also, from a practical point of view, isotropic insulators without phase transitions in the temperature range of interest are needed so that device fabrication and performance modeling can be accomplished in a simple and reliable fashion.

In this paper, we report the results of extended experiments on the growth and characterization of Sr_2AlTaO_6 (SAT)/$YBa_2Cu_3O_7$ (YBCO) heterostructures [3]. Since the dielectric properties of SAT single crystals are not very wellknown, we have investigated the dielectric properties of SAT films as well as the influence of SAT on the structural and electrical properties of YBCO.

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II. BACKGROUND

To our knowledge, SAT was first prepared by Brandle and Fratello [4] in a study of potential substrate materials for the growth of HTS films. The growth of bulk single crystals has not been reported, but workers at Pennsylvania State University have recently grown single-crystal fibers by a laser heated pedestal technique [5]. On the basis of measurements at 10 kHz on ceramic samples, these workers have reported promising dielectric properties for SAT, with a dielectric constant of ~ 11.8 at both room temperature and 100 K, and dielectric loss values of 1.68 x 10^{-3} and 4.24 x 10^{-4} at room temperature and 100 K, respectively [6].

The structural properties of SAT are closely matched to those of YBCO. Like $SrTiO_3$, from which it can formally be derived by replacing each pair of Ti^{+4} ions by one Al^{+3} and one Ta^{+5} ion, SAT has the cubic perovskite structure. For SAT samples prepared by ceramic techniques or by slow crystallization from the melt, the Al^{+3} and Ta^{+5} ions are ordered, doubling the unit cell. At room temperature, the measured lattice constant a_o is 7.795 Å [4]. Thus SAT, with $a_o/2$ = 3.898 Å, is even better matched to the a-b plane of YBCO than $SrTiO_3$, for which a_o = 3.905 Å. (For single-crystal SAT fibers, which are solidified so rapidly that the Al^{+3} and Ta^{+5} ions do not become ordered, the measured value of a_o is 3.895 Å [5,6].) The thermal expansion coefficient of SAT, as measured by dilatometry on ceramic samples [6], is ~ 9 x 10^{-6} °C^{-1}, very close to the value for YBCO. The dilatometric measurements give no evidence of a structural phase transition between room temperature and 750 °C, nor is any such transition expected at higher temperatures.

III. PULSED LASER DEPOSITION

Single layer films of SAT and multilayers of SAT/YBCO were grown on [001] $LaAlO_3$ and [001] MgO substrates by a pulsed laser deposition (PLD) process [7]. Briefly, PLD involved firing of highly energetic 248-nm laser pulses (2 J/cm^2, 20 ns) at a stoichiometric target placed face to face with a heated substrate (700-800 °C) in the presence of ~ 100 mTorr flowing oxygen. The SAT target was a disc cut from a polycrystalline boule that had been directionally solidified from the melt by the vertical gradient-freeze technique. The deposition rate was ~ 0.5 Å/pulse for SAT and ~ 1 Å/pulse for YBCO. Upon

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completion of film deposition, the samples were cooled to room temperature in ~ 1/2 h in ~ 200 Torr oxygen. These conditions were close to the optimum conditions for obtaining high-quality YBCO films on $LaAlO_3$.

III. SAT DEPOSITION

The dependence of the cation stoichiometry on the deposition parameters was examined by Rutherford backscattering spectrometry (RBS) performed on a series of ~100-nm-thick SAT films on MgO. The cation composition of the films was found to be within ± 20% of the stoichiometric target pellet as long as the laser energy density on the pellet was ≥ 1 J/cm^2 and the oxygen pressure was maintained above 25 mTorr. The (Al / Sr and Ta / Sr) ratios in the films increased monotonically from (0.40 and 0.53) to (0.53 and 0.64), respectively, with increasing oxygen pressure from 25 to 250 mTorr. Thus, the Al / Ta ratio remained roughly constant at ~ 0.8.

After determining the PLD system parameters for obtaining near-stoichiometric ($Sr_2AlTa_{1.2}O_x$) SAT films, the substrate temperature was varied to maximize the film crystallinity. With an oxygen pressure of 125 mTorr, films were deposited on [001] $LaAlO_3$ substrates at four different temperatures in a window of ~ 50 °C, including the optimum temperature of 785 °C used for high-quality YBCO growth. All four SAT films were transparent, with smooth mirror-like surfaces and optical interference related color patterns. X-ray diffraction (XRD) showed epitaxial growth of SAT on $LaAlO_3$ with the *c* axis oriented normal to the substrate plane (Fig. 1). (The x-ray diffractometer pattern in Fig. 1 is consistent with either ordered or random distribution of Al^{+3} and Ta^{+5} ions in the SAT film. The [002] and [004] lines in the pattern, which are indexed for the ordered structure, would be indexed as [001] and [002], respectively, for the disordered structure.)

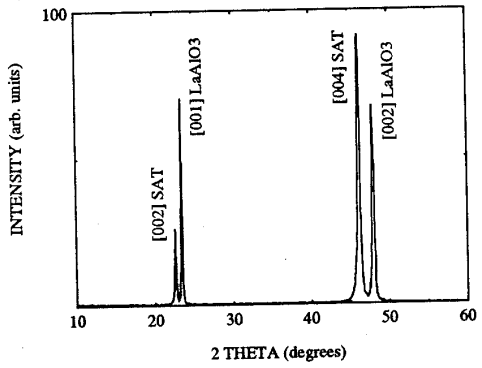

Figure 1. θ-2θ x-ray diffractometer pattern for a 100-nm-thick SAT film on [001] $LaAlO_3$.

Rocking curve measurements on the [002] peak for this set of samples yielded < 0.5° full width at half-maximum (FWHM). The minimum value of 0.25° was obtained for the sample deposited at 785 °C. The FWHM of the substrate [001] peak was measured to be > 0.1° for the same set of samples. The crystallinity of the SAT films was also investigated by RBS/channeling along the [001] substrate normal direction. As seen in Fig. 2, the minimum backscattering yield χ_{min} is 5 % for the sample with the minimum XRD FWHM.

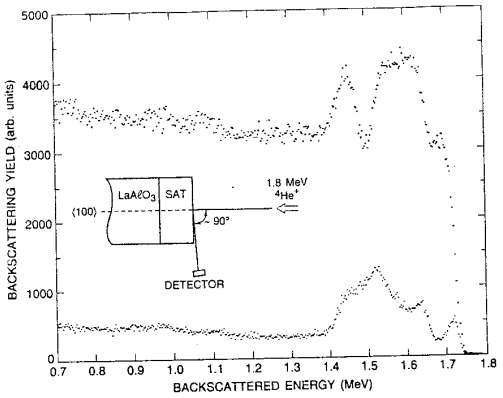

Figure 2. Random and channeling RBS spectra (1.8 MeV He^+) along [001] axis of the substrate with 90° backscattering geometry.

In addition, channeling was performed along the [110] $LaAlO_3$ direction tilted approximately 35° to the substrate normal to investigate the degree of film orientation parallel to the substrate surface. The film χ_{min} is ~ 25% along this direction, which indicates that the film is essentially singly oriented in the plane but that there is considerable disorder along this axis.

IV. YBCO on SAT

In order to investigate the compatibility of SAT films with YBCO, YBCO/SAT bilayer films were deposited in situ on [001] $LaAlO_3$ substrates using an oxygen pressure of 125 mTorr and substrate temperature of 785 °C. XRD studies of YBCO/SAT multilayers showed complete *c* axis orientation of YBCO on SAT. Measurements of ac susceptibility showed Tc_{onset} ~ 89.5 K and ΔTc < 1K for a YBCO film ~ 100 nm thick on an SAT film ~ 100 nm thick . The YBCO film was then patterned in the form of a 10-μm-wide, 50-μm-long line by following our standard photolithography process which involves photoresist line definition, standard bake, dilute nitric acid etch after exposure of the film, photoresist removal by acetone, and methanol rinse. Four-probe dc measurements performed on this patterned film showed Tc_o ~ 89.2 K. The critical current density at zero magnetic field was measured to be ~ 1.3x10^6 A/cm^2 at 77 K using the 1 μV/cm criterion.

V. SAT on YBCO

There is no established value for the bulk dielectric constant of SAT, since bulk single crystals are not yet available. Although the dielectric properties of thin films can be very different from those of bulk crystals [8], we have

investigated the dielectric properties of SAT films. For dielectric measurements, SAT films were deposited on YBCO films on $LaAlO_3$, and ~ 200 nm gold was evaporated through a mask to obtain ~ 1 - 6 mm^2 capacitor plates. The capacitance was measured at 1 kHz using an LCR meter. The relative dielectric constant ε_r of the SAT was then determined from the expression $\varepsilon_r = Cd/\varepsilon_o A$, where C is the net capacitance, d is the thickness of the SAT film obtained by RBS and thickness calibration, ε_o is the permittivity of the free space, and A is the area of the gold pad. The values of ε_r were determined to be 23 ± 3, 26 ± 3 and 30 ± 3 for films that were 100, 225 and 390 nm thick, respectively. The static breakdown voltage for the 100-nm-thick film was ~ 6 V for both polarities, yielding ~ $6x10^7$ V/m for the breakdown electric field. Neither the dielectric constant nor the breakdown field showed any appreciable temperature dependence between 10 and 300 K. The ε_r values are approximately double the value reported [6] for bulk ceramic samples. Factors that might contribute to this discrepancy include the effect on bulk properties of less than theoretical densities or the presence of grain boundaries or secondary phases [9], and the effect on film properties of deviations from perfect cation stoichiometry and strain.

VI. YBCO/SAT/YBCO TRILAYERS

Superconductor/dielectric/superconductor trilayers are building blocks for various superconducting thin film devices. Thus, we have fabricated YBCO/SAT/YBCO trilayers by developing a simple extension of our standard SAT / YBCO bilayer in situ PLD process.

Fig. 3 shows an x-ray diffraction scan of a YBCO/SAT/YBCO trilayer on $LaAlO_3$, with estimated thicknesses of 200 nm and 500 nm for YBCO and SAT films, respectively. (00k) peaks of the scan are consistent with the growth of a well-oriented c axis trilayer.

Then, we have used mutual inductance measurement to investigate the superconductive quality of the YBCO layers. Fig. 4 shows the mutual inductance signal for the same trilayer, indicating a transition around 91 K. This result indicates that at least one YBCO layer of the trilayer expels the magnetic field i.e., goes into the Meissner state, at that temperature.

Also, we have measured the resistive losses at the surface of the trilayer by using a dielectric resonator technique at 24.5 GHz. In this technique [10], a sapphire disc is placed on the surface of the film inside a copper housing. Under suitable conditions [10], the measured quality factor of the dielectric resonator is inversely proportional to the energy dissipation induced by circulating currents in the film. Fig. 5 shows the quality factor of the resonator with the trilayer and a 2-mm-thick OFHC copper plate as a function of temperature. The rapid increase of the quality factor below ~ 90 K for the trilayer suggests that the top YBCO layer has a transition temperature of ~ 90 K. Furthermore, with the assumption that the bulk OFHC has ideal characteristics, we estimate the effective surface resistance of the trilayer to be ~ 12 mΩ at 77 K. This value is within a factor of four of what we measure for single layer YBCO films, which indicates that the top YBCO layer of the trilayer is of fairly good quality.

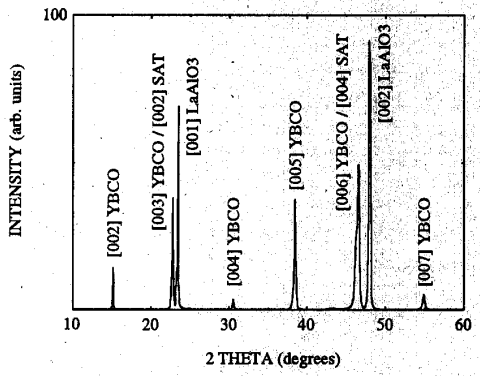

Figure 3. θ–2θ x-ray diffractometer pattern for a YBCO[200 nm]/SAT [500 nm]/YBCO[200 nm] trilayer on $LaAlO_3$.

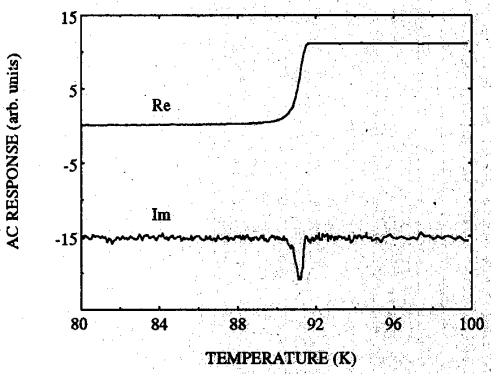

Figure 4. Mutual inductance signal showing onset of the superconducting transition above 91 K. Data is from the same trilayer used for Fig. 3.

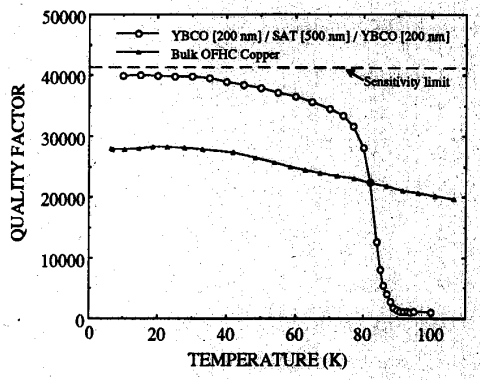

Figure 5. Quality factor versus temperature for a trilayer (same trilayer used for Figs. 3, and 4) and bulk OFHC copper at 24.5 GHz. Dashed line indicates the sensitivity limit of the resonator for determining surface resistance.

VII. SUMMARY

In conclusion, high quality SAT/YBCO heterostructures with *c* axis orientation have been obtained by PLD on [001] $LaAlO_3$ substrates. Like $SrTiO_3$, SAT is a cubic perovskite (thus, there are no twins) that is closely lattice matched to YBCO, but SAT films have a much lower dielectric constant than $SrTiO_3$. Our results indicate that SAT is promising for use with YBCO in bilayer and trilayer structures for superconducting device applications. Moreover, structural and chemical similarity between SAT and $SrTiO_3$ may prove SAT dielectric material very useful in novel device structures such as dielectric-base transistor [11], where one might use quality YBCO/SAT/$SrTiO_3$ heterostructures.

ACKNOWLEDGEMENTS

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