S/N-Heterostructure of BiSrCaCuO/BiSrCuO Grown by Halide CVD

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Thin film S/N heterostructures of superconducting BiSrCaCuO and metallic BiSrCuO with a good surface morphology, crystallinity, and superconducting properties were formed on (100) MgO substrates by halide CVD. Critical temperatures (zero resistance at Tc) and critical current densities (Jc) of the BiSrCaCuO films on MgO substrate were Tc =75 to 80 K and Jc = $1 \times 10^{6} \text{ A/cm}^{2}$ at 60 K. Electrical resistance of the metallic BiSrCuO films was 0.7 to 1.5 x10⁻³ ohm-cm from 10 to 300 K. High resolution transmission electron microscopy (HRTEM) images showed the interface between the BiSrCaCuO and BiSrCuO films to be atomically very abrupt. Energy-dispersive X-ray spectroscopy (EDXS) depth profiles were also evaluated using TEM samples. No Ca composition interdiffusion was detected at the BiSrCaCuO and BiSrCuO film interface. The critical temperatures of S/N-multilayer samples was about 75 K. In this work, we fabricated S/N-heterostructures of bismuth-compound crystals grown by halide CVD and studied the film quality and superconducting properties.

1. INTRODUCTION

To obtain superconducting electronic devices such as SIS or SNS tunnel junctions and three terminal devices, we must produce heterostructure thin films of both superconducting and nonsuperconducting materials. Several researchers have attempted to produce YBaCuO/DyBaCuO [1], YBaCuO/PrBaCuO [2], BiSrCaCuO/ BiSrCuO and [3] heterostructure materials using physical vapor deposition (PVD), because these materials exhibit similar crystal structures and lattice constants. It has been difficalt, however, to obtain high-quality heterostructures with good crystallinity and electrical properties, i.e., good surface morphology, low defect density, stoichiometric composition, higt Tc and high Jc, which depends on deposition temperature for films, using usual growth techniqued.

We developed a chemical vapor deposition (CVD) technique for growing high-quality heterostructures of metallic BiSrCuO thin film on super-conducting BiSrCaCuO on MgO substrates using OT-CVD with a higher growth temperature for the first layer and LP-CVD and a lower growth temperature for the 2nd layer. We evaluated the heterostructure materials using polarizing microscopy, SEM, XRD, high resolution TEM, EDXS, and the electrical properties of films.

2. EXPERIMENTS

Superconducting BiSrCaCuO films were grown on MgO substrates using BiCl₃-SrI₂-CaI₂-CuBr₂-O₂-He and an open-tube CVD technique [4]. MgO crystal surfaces were oriented at $\pm 1^{\circ}$ along (100) and 6° off (100) toward <010>. The area was usually 30 mm x 30 mm. Growth conditions for the films were $TB_iC_{13} = 150$ to $170^{\circ}C$, $TC_uBr_2 = 370$ to $390^{\circ}C$, $TS_{rI_2} = 770$ to $790^{\circ}C$, and $TC_{aI_2} = 760$ to $780^{\circ}C$. The oxygen concentration in the He carrier gas was about 5% and the H₂O concentration was about 500 ppm. Films were grown at 0.8 to 1.2 nm/min at 775°C. Typical critical temperatures and critical current densities for 0.2-µm-thick BiSrCaCuO films grown at 750 to 850°C were Tc = 75 to 98 K and Jc = 0.04 to 7 x 10⁶ A/cm² at 10 K.

Metallic BiSrCuO and superconducting BiSrCaCuO films were grown using a low pressure halide CVD system [5]. Growth conditions for the films were $T_{BiC13} = 160$ to 180°C, $T_{CuI} = 470$ to 490°C, $T_{SrI2} = 890$ to 910°C. The total flow rate of He carrier gas into the source cells was 1300 sccm. The oxygen concentration was about 8%. The back pressure at the deposition chamber was 0.1 to 1 torr. Films were grown at 0.2 to 0.4 nm/min at 580°C. Typical critical temperatures and critical densities of 70-nm-thick a current BiSrCaCuO films grown at 580 to 680°C were Tc = 65 to 70 K and Jc = 1.5 to 5.0 x 106 A/cm² at 10 K.

3. RESULTS AND DISCUSSION

We developed a way to determine the grain size and inter-grain orientation nondestructively using a polarizing microscope under the crossed Nicols condition [6]. Large grain patterns from 30 to 300 μ m were observed on a 0.2- μ m - thick BiSrCaCuO film along a ± 1° (100) MgO substrate. Films 6° off (100)-oriented MgO substrates, we could not observe large grain patterns (> 0.5 μ m).

We evaluated the tipical temperature dependence of the critical current density and layers' electrical resistance for superconducting BiSrCaCuO, metallic BiSrCuO films, and BiSrCaCuO/ BiSrCuO S/N-multilayers. Samples were formed along the a and b axis planes on BiSrCaCuO films and they were etched into 10 x 20-µm-stripe-patterned using standerd photolithography. Critical current densities along the a-axis plane of the BiSrCaCuO film 6° off (100)-oriented MgO

substrate were 1.5 to 3 x 10^6 A/cm² at 10 K. Critical current densities along the baxis plane of the film were 1.5 to 2.5 x 10^5 A/cm² at 10 K, revealing a large anisotropy. The critical temperatures of the film were 75 to 80 K.

The temperature dependence of layers' electrical resistivity is shown in Fig. 1. Curve a is a 70-nm-thick metallic BiSrCuO on a MgO substrate. Film resistivity was 0.7 to 1.5 x 10^{-3} ohm-cm at 10 to 300 K. Curve b is a 0.2-µm-thick superconducting BiSrCaCuO film on a MgO substrate. Curve c is a 70-nm-thick metallic BiSrCuO film on a 0.2-µm-thick BiSrCaCuO/MgO substrate. Critical temperatures for S/N multilayer samples were all around 75 K [7].



Fig. 1 Temperature dependence of the electrical resistivity of (a) a 70-nm-thick metallic BiSrCuO film on a MgO substrate, (b) 0.2-µm-thick BiSrCaCuO film on a MgO substrate, and (c) S/N BiSrCuO / BiSrCaCuO / MgO substrate.

Figure 2 shows the cross-sectional HRTEM images of the interface between the BiSrCaCuO and BiSrCuO films was and abrupt atomically very no preferential precipitation or reaction between these materials was detected. Lattice constant for the c-axis plane of the BiSrCuO film was estimated to be 2.5 nm and intergrowth of the BiSrCaCuO film with c = 2.5, 3.0, 3.6, 4.0, and 4.8 nm were very rarely observed. Stacking faults and dislocations were observed the in

superconducting BiSrCaCuO film, but not in metallic-BiSrCuO film.

EDXS depth profiles at the BiSrCaCuO and BiSrCuO film interface is shown in Fig. 3. Analysis was performed in a VG-HB501 HRTEM system, a Kevex-Super 8000 EDX system and formed in STEM mode. No interdiffusion of the Ca composition was observed at the BiSrCaCuO and BiSrCuO film interface. It can be seen that the Ca concentration changes over a distance of less than 2 to 3 unit cells of the crystal. The composition (atm %) of the BiSrCaCuO and BiSrCuO films was estimated to be Bi:Sr:Ca:Cu = 2:2:2:3.5 and Bi:Sr:Cu = 1:1:1.



Fig. 2 TEM images of the BiSrCaCuO and BiSrCuO filminterface.

4. CONCLUSIONS

We developed a technique for growing S/N heterostructures for superconducting BiSrCaCuO and metallic BiSrCuO films using a halide CVD system. Lavers were evaluated by polarizing microscopy, SEM, XRD, EDXS, and HRTEM observe as well as measurement of their electrical properties. Cross-sectional HRTEM image of the interface between the BiSrCaCuO and BiSrCuO films was atomically very abrupt. No interdiffusion of the Ca composition was detected at the BiSrCaCuO and BiSrCuO films interface using EDXS. The critical temperature of the S/N multilayer samples was about 75 K.



Fig. 3 EDXS depth profiles of film compositions at the interface between BiSrCaCuO and BiSrCuO films.

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