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Possible new single-buffer layers for $YBa_2Cu_3O_{7-y}$ coated conductors prepared by chemical solution deposition

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New single-buffer layers of $YBiO_3$ and $SmBiO_3$ have been proposed for $YBa_2Cu_3O_{7-y}$ (YBCO) and $SmBa_2Cu_3O_{7-y}$ coated conductors. Highly *c*-axis oriented $YBiO_3$ and $SmBiO_3$ buffer layers have been deposited on single-crystal $LaAlO_3$ and $SrTiO_3$, respectively, by a low-cost chemical solution deposition method in a temperature range as low as 730 to 800 °C in air. Precursor solution of yttrium nitrate, samarium nitrate, and bismuth nitrate has been deposited using spin coating and heat treated in air in a single stage to yield textured $YBiO_3$ and $SmBiO_3$ buffers. A very dense, smooth, pinhole-free, and crack-free morphology has been observed for both buffers. Dense, homogeneous, and epitaxially grown YBCO film with thickness about 300 nm has been obtained on $YBiO_3$ buffer with onset critical temperature 90 K and J_c (77 K, self-field) over 3 MA/cm^2. These results offer an effective alternative to prepare desirable buffer layer(s) for YBCO-coated conductors.

I. INTRODUCTION

In recent years, the development of second-generation high-temperature superconducting tapes, namely coated conductors, has attracted much attention in the area of the applications of high-temperature superconductivity (HTS).[1–4] One of the key issues is the preparation of biaxially textured buffer layer(s), which act(s) as a template for the epitaxial deposition of the $YBa_2Cu_3O_{7-y}$ (YBCO) superconductor. In addition to being a diffusion barrier,[5] the buffer layer provides sufficient crystalline texture to the HTS layer to alleviate the problems related to weak-linked, high-angle grain boundaries.[6]

For YBCO-based coated conductors, many potential buffer layer materials have been identified, including $SrTiO_3$, $La_2Zr_2O_7$, CeO_2, YSZ, and $SrRuO_3$, among which an excellent result has been obtained via a three-layered architecture such as CeO_2/YSZ/CeO_2.[7] However, deposition of such a complete buffer layer sequence would involve a combination of both in situ and ex situ deposition techniques, which could adversely affect the structural and chemical integrity of the individual buffer layers and add to the complexity as well as to the total cost of the overall process, therefore hampering the scale-up of the process. To circumvent these difficulties, it would be of great importance to develop a single-buffer layer deposition technique, which could significantly simplify the preparation and make the fabrication process cost effective and thus facilitate its scale-up to long lengths.

Chemical solution deposition (CSD) such as metal-organic deposition (MOD) and sol-gel process has been demonstrated to offer the desirable aspects of being more easily scalable, inexpensive, and simple. An all-solution approach has been attempted to deposit YBCO coated conductors, which yielded critical current density $J_c > 1$ MA/cm^2.[8–11] However, the results obtained by CSD methods are still not as good as that produced by the vacuum processes [pulsed layer deposition (PLD), sputtering, metalorganic chemical vapor deposition (MOCVD), etc.]. One of the main reasons is the relatively poor quality of buffer layer(s), which has high melting point relative to $REBa_2Cu_3O_{7-y}$ (denoted as REBCO, here RE represents rare earth elements except

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for Pr, Ce, and Tb), making it difficult to obtain a sufficiently dense buffer layer for the subsequent deposition of superconducting layer. Another drawback of the current CSD process focuses on the great popularity of the expensive metal alkoxides such as butoxides,[8,9] propoxides,[10–12] methoxyethoxide,[13] and metal carboxylates with large hydrocarbon chain such as pentanedionates[12,14] and naphthenates.[15] Moreover, relatively high annealing temperature as well as reducing atmosphere, which hamper the scale-up of the process, have to be used to obtain well-textured, smooth buffer layer(s). Siegal et al. have reported[8] a Nb-doped $SrTiO_3$ buffer layer prepared up to 900 °C in low $P(O_2)$ ambient. Okuyucu et al. have reported[12] the preparation of RE_2O_3, which was annealed at 1150 °C in Ar/H_2 4% gas for Ni or Ni–W substrates. Highly textured, smooth, and crack-free $La_2Zr_2O_7$ layer has been obtained by Engel et al.;[16] the annealing has been done at 900 in $5\%H_2$–Ar. Akin et al. have succeeded[17] in preparing $CeO_2/YSZ/CeO_2$ architecture via MOD process, while the annealing has been done in $4\%H_2$–Ar at up to 950 °C for CeO_2 and 1050 °C for YSZ, respectively. If a new buffer layer material that can be annealed under 800 °C in air or even lower temperature in a reducing atmosphere can be found, it would certainly add to the scalability of the overall process.

In this paper, we propose the new buffer layer materials $YBiO_3$ for YBCO and $SmBiO_3$ for SmBCO-coated conductors. $REBiO_3$ compounds are chosen as the candidate for our study because: (i) the pseudocubic cell of $REBiO_3$ matches well with the REBCO compounds; (ii) Bi cannot be doped into YBCO lattice,[18] and thus will not suppress its superconductivity; (iii) the melting point of $REBiO_3$ should be around the center between that of RE_2O_3 (about 2000 °C) and Bi_2O_3 (825 °C). The relatively low melting point of $REBiO_3$ with respect to those of other buffer materials used currently may lead to a partial melting process for formation of dense textured layer of $REBiO_3$ at a low processing temperature. Single bismuth yttrium oxide $YBiO_3$ and bismuth samarium oxide $SmBiO_3$ buffer layers have been deposited, respectively, via chemical solution deposition process for YBCO-coated conductors. Precursor solution of yttrium nitrate and bismuth nitrate has been spin coated on $LaAlO_3$, while samarium nitrate and bismuth nitrate are spin coated on $SrTiO_3$. The coated samples were then subjected to heat treatment in air in a temperature range 730 to 800 °C in air. The grown buffer layers yielded a *c*-axis oriented texture. Very dense, smooth, pinhole-free, and crack-free morphology has been observed for both buffers. Dense, homogeneous, and epitaxially grown YBCO film has been deposited on $YBiO_3$ buffer via a fluorine-free method. Superconducting transition at 90 K and J_c (77 K, self-field) over 3 MA/cm^2 have been achieved.

II. EXPERIMENTAL

Single crystals $LaAlO_3$ (100) and $SrTiO_3$ (100), with a dimension of 10×10 mm^2, have been used as substrate. The precursor solution was prepared in ambient atmosphere. The reagents yttrium nitrate (99.9%, Kelong, Beijing, China), samarium nitrate (99.9% Kelong), and bismuth nitrate (99.9%, Kelong) were dissolved in deionized water with continuous stirring. A proper amount of carbamide was then added, which helps to adjust the pH value of the solution. Polyvinyl alcohol (PVA) (homemade) was finally added and stirred for 6 h to adjust the solution viscosity. The final concentration of the solution was 0.25 M. Two solutions were then spin coated on $LaAlO_3$ and $SrTiO_3$ single-crystal substrate, respectively, followed by heat treatment in a furnace at 730 to 800 °C for 15 min in air.

YBCO was deposited on the $YBiO_3$-buffered $LaAlO_3$ via a fluorine-free approach developed in our laboratory. During processing, yttrium acetate (99.9%, Kelong), barium acetate (99.9%, Kelong), and cupric acetate (99.9%, Kelong) were dissolved in propionic acid with the cationic ratio Y:Ba:Cu = 1:2:3.2. The solution was then spin coated on the YBO-buffered LAO, and the coated sample was dried at 100 to 200 °C. Subsequently, the sample was subjected to 400 to 500 °C to decompose the organic salts. The furnace temperature was then increased to 770 °C and maintained for 3 h for the formation of YBCO phase in Ar. At the end of 3h, the Ar atmosphere was replaced by O_2 with oxygenation at 400 to 450 °C for 0.5 to 4 h.

The as-grown films were characterized using x-ray diffraction (XRD) for phase purity and texture, scanning electron microscopy (SEM) for homogeneity and microstructure, and atomic force microscope (AFM) for crystalline quality and density, as well as roughness. A Philips X'Pert MRD diffractometer (PANalytical, Southwest Jiaotong University, Chengdu, People's Republic of China) with Cu K_α radiation was used to record the θ–2θ XRD patterns. The microstructure, the crystalline quality, and the roughness samples were analyzed using an SPI3800N AFM (Seiko Instruments Inc., Southwest Jiaotong University, Chengdu, People's Republic of China) and an environmental scanning electron microscope (ESEM) equipped with energy dispersive spectroscopy (EDS). Superconducting transition and the hysteresis loops of $M(H)$ were measured by using Quantum Design (Southwest Jiaotong University, Chengdu, People's Republic of China) SQUID XL (7T).

III. RESULTS AND DISCUSSION

Bismuth yttrium oxide, $YBiO_3$, has a cubic structure as shown in Fig. 1, with lattice parameter $a = 5.428$ Å and pseudocubic cell with $a' = 3.838$ Å, which is closely matched to that of the orthorhombic YBCO (1% mismatch with a_{YBCO} and 0.5% mismatch with b_{YBCO}).

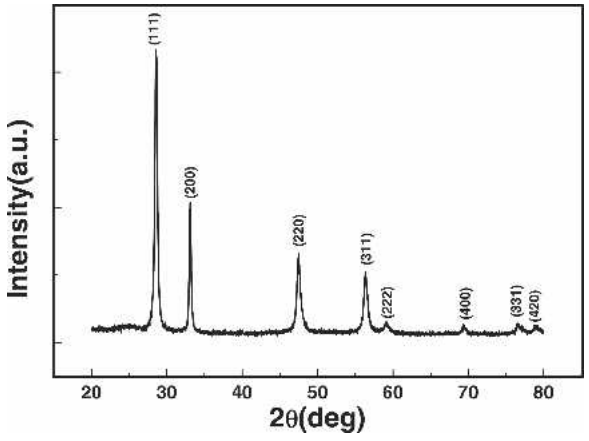

FIG. 1. Typical powder θ–2θ XRD pattern of $YBiO_3$.

$SmBiO_3$ has an identical cubic structure, except that the lattice parameter $a = 5.536$ Å and pseudocubic cell with $a' = 3.914$ Å (0.07% mismatch with a_{SmBCO} and 1.5% mismatch with b_{SmBCO}). No phase transition has been observed from room temperature up to 1050 °C through the XRD powder diffraction for the two buffers.

A typical XRD θ–2θ scan for the YBO layer deposited on $LaAlO_3$ single-crystal substrate is given in Fig. 2(a), which reveals the presence of a sharp out-of-plane texture for the $YBiO_3$ buffer layer. It is obvious that the intensities of the (111) peak of $YBiO_3$ buffer layer is negligible compared with the intensity of its (200) peak. Figure 2(b) displays the phase and texture information for $SmBiO_3$ buffer on $SrTiO_3$. The $SmBiO_3$ film exhibits an (n00) texture except for a tiny trace of undesirable (111) peak. This suggests that these buffer layers have a strong tendency toward preferred orientation. As a matter of fact, one of the starting materials, Bi_2O_3 melts at 817 °C under ambient atmosphere as we used in our experiment. Therefore, upon heating after decomposition, the mixtures of Y_2O_3 and Bi_2O_3 (or Sm_2O_3 and

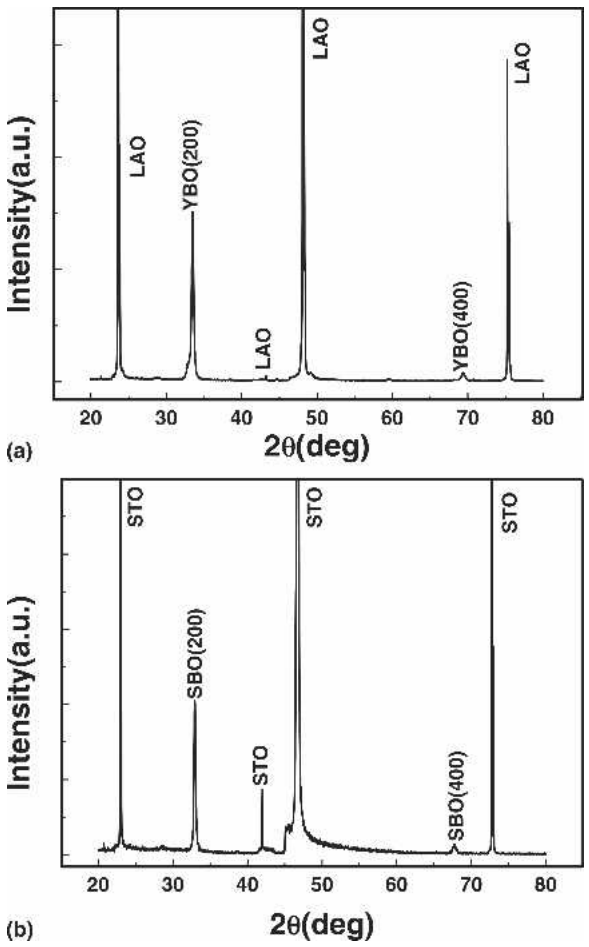

FIG. 2. Typical θ–2θ XRD pattern of: (a) $YBiO_3/LaAlO_3$, and (b) $SmBiO_3/SrTiO_3$, showing a good c-axis texture of $YBiO_3$ and $SmBiO_3$.

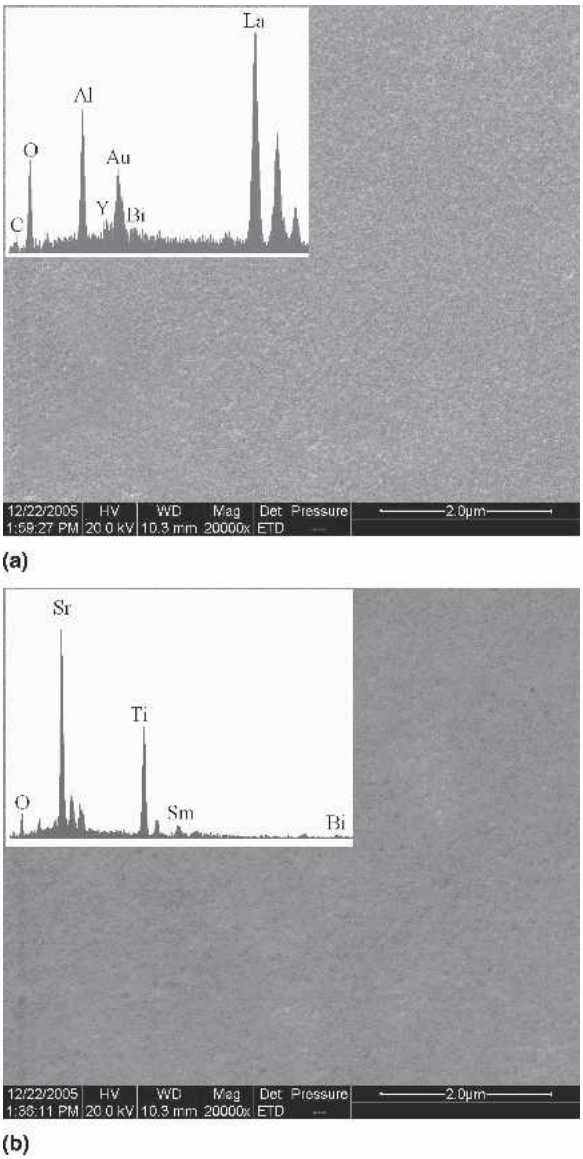

FIG. 3. SEM micrograph of buffer layers: (a) $YBiO_3$ deposited on $LaAlO_3$ single-crystal substrate, and (b) $SmBiO_3$ buffer layer deposited on $SrTiO_3$ single-crystal substrate. Insets show the results of EDS analyses for the Y, Sm, and Bi concentrations.

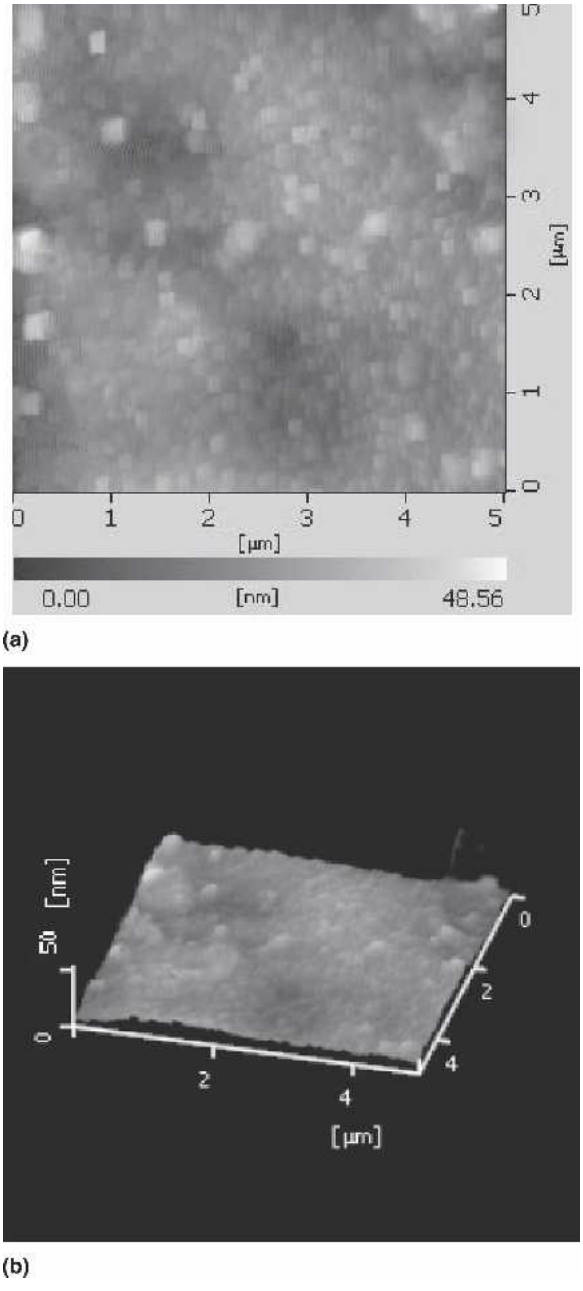

FIG. 4. Surface cut-out (5 μm × 5 μm) of $YBiO_3$ buffer layer on $LaAlO_3$ measured by AFM.

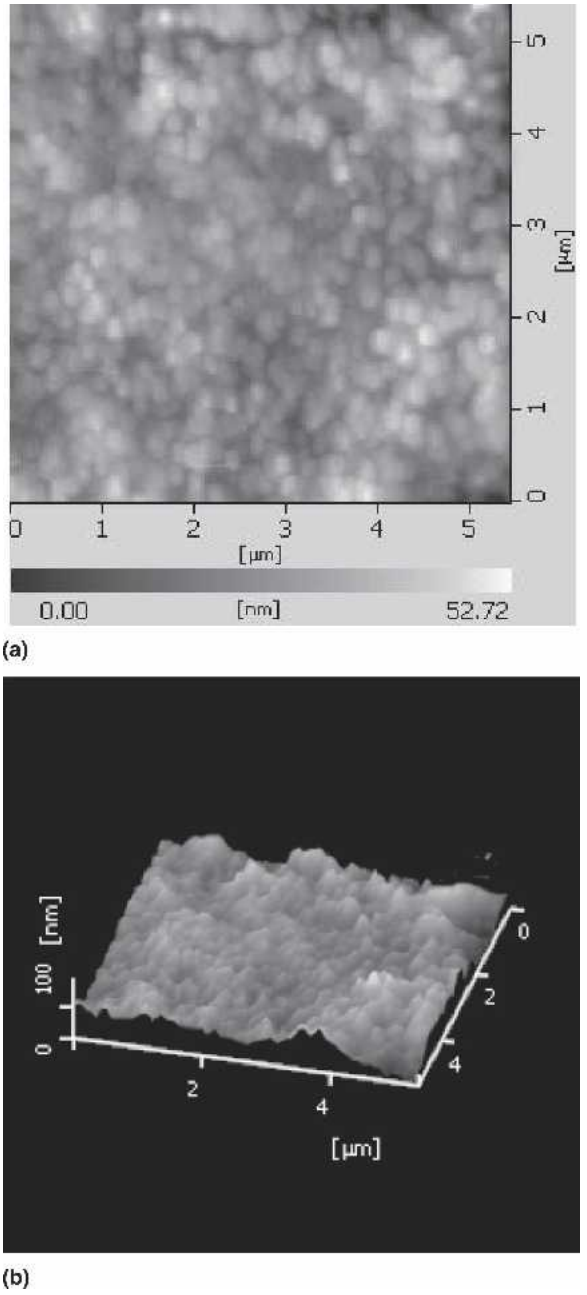

FIG. 5. Surface cut-out (5 μm × 5 μm) of $SmBiO_3$ buffer layer on $SrTiO_3$ measured by AFM.

Bi_2O_3) will undergo a partial melting process. Similar to the melt texture growth (MTG) process used for REBCO, this partial melting process is likely to lead to preferred orientation, with the $LaAlO_3$ ($SrTiO_3$) substrate serving as a big seed layer.

In addition to the excellent crystalline quality, $YBiO_3$ and $SmBiO_3$ layers show a dense, homogeneous, crack-free, and pinhole-free microstructure, as displayed in Figs. 3(a) and 3(b), respectively. A pebble-like morphology, with an approximate grain size of 50 nm has been observed for $YBiO_3$, which can be interpreted as corresponding to good *c*-axis texture, while for $SmBiO_3$ buffer layer, the grains are still finer. Qualitative EDS analysis reveals a very good distribution of the cations with a ratio Y(Sm):Bi ≈ 1:1. AFM investigations of the surface structure of the buffer layers YBO and SBO point toward a homogeneous granular structure, as shown in Figs. 4 and 5. The root-mean-square roughness of $YBiO_3$ buffer layer is about 5 nm and that of $SmBiO_3$ is about 9 nm, which indicates a relatively smooth surface of the buffer systems, which is important for a successful epitaxial growth of *c*-axis aligned YBCO layer. The effect of the densification obtained by SEM images can equally be confirmed.

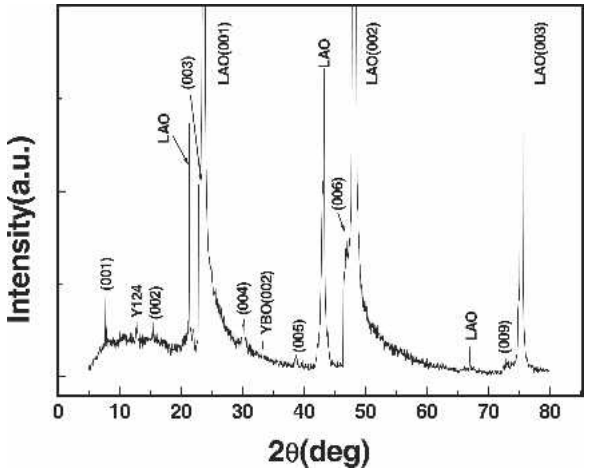

FIG. 6. Typical θ–2θ XRD pattern of $YBCO/YBiO_3/LaAlO_3$ showing the good *c*-axis texture of YBCO.

The morphology observed above results on one hand from the careful control of the thermal process and on the other hand from the partial melting process, the latter due to the relative low melting temperature of Bi_2O_3, facilitating the densification of $YBiO_3$ ($SmBiO_3$). As a matter of fact, it is possible that Bi_2O_3 undergoes a partial melting process, which may be favorable for the solid solution $YBiO_3$ ($SmBiO_3$) to obtain a better density, i.e., a better densification of $YBiO_3$ ($SmBiO_3$. Moreover, the melting point of the final product $YBiO_3$ ($SmBiO_3$) has been proved to be well above 1050 °C through a differential thermal analysis (DTA) process, which means that the partial melting process will exert little influence on the subsequent YBCO layer. As we mentioned previously, the prevailing buffer layer materials including $SrTiO_3$, $La_2Zr_2O_7$, CeO_2, YSZ, $SrRuO_3$, etc., are all heritages from those used in the former physical process. In the in situ vacuum process, densification can be easily ensured because of the layer-by-layer route, whereas in the nonvacuum CSD process, densification seems to have greatly hampered its scale-up and remains a problem. The introduction of a new compound Bi_2O_3, with its relative low melting point, offers a new approach to densify the buffer layer and facilitate the deposition of superconducting layer.

In addition to the densification effect, the introduction of Bi_2O_3 has decreased the process temperature by 150 to 300 °C. $YBiO_3$ and $SmBiO_3$ can be prepared at 730 to 800 °C, which makes the process cost effective and easily scaled up.

The YBCO layer with a thickness of about 300 nm deposited on $YBiO_3$-buffered $LaAlO_3$ shows a highly developed *c*-axis texture as in Fig. 6, though there may exist Y-124, which may due to the relatively low annealing temperature. A dense, homogeneous, crack-free, and pinhole-free microstructure has been observed, as shown in Fig. 7. Figure 8 shows the superconducting transition

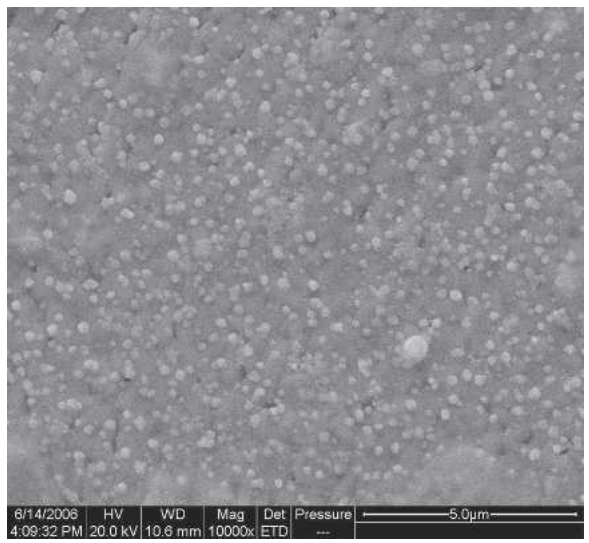

FIG. 7. SEM micrograph of YBCO layer deposited on $YBiO_3$-buffered $LaAlO_3$ single-crystal substrate showing good density and surface morphology.

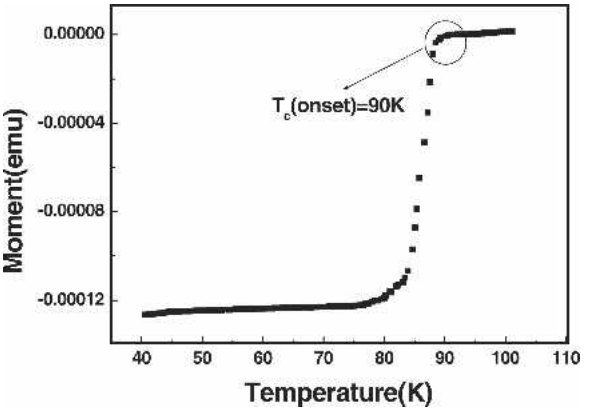

FIG. 8. Temperature dependence of magnetic moment for YBCO/$YBiO_3$/$LaAlO_3$ yielding an onset critical temperature 90 K.

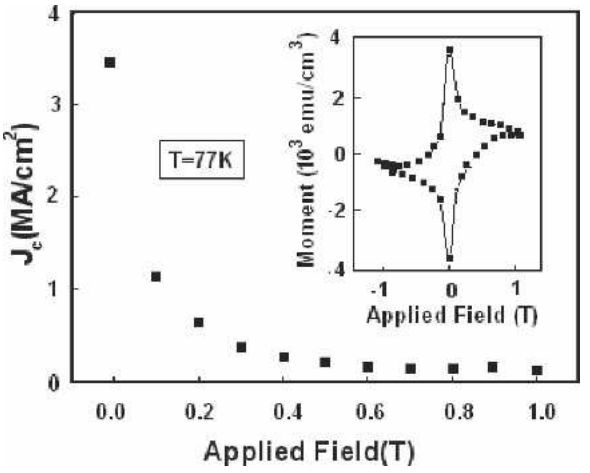

FIG. 9. Field dependence of critical current density at 77 K and self-field for YBCO films deposed on $YBiO_3$ buffered $LaAlO_3$. Inset: Hysteresis loop of magnetization at 77 K.

of the YBCO, which starts at 90 K. The sharp superconductivity transition of the YBCO film demonstrates a high quality and phase uniformity of the film. The critical current density deduced from the hysteresis loops of $M(H)$ curves is more than 3 MA/cm^2 at 77 K in self-field, demonstrating a high performance of the superconducting layer (see Fig. 9).

A $YBiO_3$ buffer has also been deposited on untextured Ni substrates in Ar/H_2 4% gas. Unlike when processing in air, the processing temperature for $YBiO_3$ phase formation is decreased to as low as 700 °C in the reducing ambient. At the same time, no chemical reaction between $YBiO_3$ and Ni substrate was observed, demonstrating that the new buffer materials and related processing are very metal-friendly. This result, together with the formation of a highly textured $YBiO_3$ film on $LaAlO_3$ single-crystal substrate, strongly suggests that $YBiO_3$ should be a good candidate as a buffer material for Ni-substrate-based YBCO coated conductors prepared by an all-solution method.

IV. CONCLUSION

New materials $YBiO_3$ and $SmBiO_3$ have been proposed to be deposited as a single buffer layer for YBCO and SmBCO coated conductors. $YBiO_3$ and $SmBiO_3$ layer have been deposited, respectively, on $LaAlO_3$ and $SrTiO_3$ single-crystal substrates via a chemical solution deposition route. In addition to the excellent *c*-axis texture of these buffer layers, a very dense, homogeneous, crack-free and pinhole-free microstructure has been observed. The subsequently deposited YBCO film with a thickness about 300 nm on $YBiO_3$ shows good *c*-axis texture, excellent surface morphology, and high superconducting performance. The introduction of Bi_2O_3 has been argued to be mainly responsible for the amelioration of the densification as well as the decrease of the process temperature, which is more metal-friendly. This approach offers an alternative for densifying the buffer layer.

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