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Optimization of the Superconducting Properties of Laser Ablated $YBa_2Cu_3O_{7-\delta}$ Films on CeO_2-Buffered Sapphire

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We use pulsed laser deposition to grow $YBa_2Cu_3O_{7-\delta}$ (YBCO) superconducting films for microwave applications. The films are grown on R-cut sapphire substrates, with CeO_2 buffer layers, which are re-crystallized at high temperature prior to YBCO growth. Using the atomic force microscopy (AFM) and X-ray diffractometry we determine the optimal temperature for recrystallization (1000°C) and the optimal buffer layer thickness (30 nm). The properties of YBCO films of various thickness, grown on the optimized CeO_2 buffer layers, are studied using several methods, including AFM, magnetooptical imaging, and transport experiments. The YBCO film roughness is found to increase with the increasing film thickness, but the magnetic flux penetration in the superconducting state remains homogeneous. The superconducting parameters (the critical temperature and the critical current density) are somewhat lower than the similar parameters for YBCO films deposited on mono-crystalline substrates.

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1. Introduction

In recent years, the superconducting microwave filters based on the high-temperature superconducting films, mainly YBCO, are intensively developed [1, 2]. To avoid high microwave losses in such application, it is necessary to grow films on sapphire substrates, with suitable buffer layer to improve matching of the lattice parameters and to prevent chemical reaction between the superconductor and the substrate. It has been found that CeO_2 is the excellent material for the buffer between the sapphire and the YBCO [3, 4]. It is chemically very stable, it has high melting temperature (2400°C) and it has fluorite structure with a lattice constant 5.411 Å [5]. YBCO, rotated 45° in the CeO_2 basal plane, has small lattice mismatch with this buffer layer, equal to 1.16% and 1.7%, along the a and b axes of YBCO, respectively. However, the properties of YBCO films grown on CeO_2 buffer layers vary a lot, depending strongly on the growth method and on the growth parameters. The origin of this variability is still a subject of intensive scrutiny. In the present paper we investigate the application of the pulsed laser deposition (PLD) method to the growth of YBCO films on sapphire substrates, suitable for microwave applications, i.e., with high superconducting state parameters: the superconducting transition temperature T_c and the critical current density j_c.

2. Film preparation and measurement details

The Al_2O_3 substrates are annealed at 1000°C prior to CeO_2 deposition in order to smooth their surface. CeO_2 thin films are deposited by laser ablation on the 5×5 mm^2 (1102) Al_2O_3 (R-cut sapphire) substrates. Ablation is performed by Nd:YAG laser (Quanta-Ray Pro 350-10, Spectra Physics, USA) with 4th harmonic generation (laser wavelength 266 nm), with pulse duration 9 ns, repetition rate 2 Hz, and the energy density 1.5 J/cm^2. During deposition the substrate temperature is held at 785°C and the oxygen pressure in the chamber is 400 mTorr. After deposition, the film is cooled down to room temperature with the rate of 20 K/min at $P_{O_2} = 300$ Torr. Subsequently, the substrates with CeO_2 buffer layers are removed from the deposition chamber, and CeO_2 is re-crystallized inside a tube furnace at high temperatures (800–1200°C) in oxygen flow for two hours. After recrystallization, the YBCO films are deposited on the top of CeO_2 buffer layers using the same PLD parameters as for the buffer layers. We have tested the quality of the CeO_2 and YBCO films with thicknesses in the range between 30 and 200 nm, and between 100 and 300 nm, respectively. For comparison, we have also prepared YBCO films on substrates of SAT–CAT–La $[(SrAl_{0.5}Ta_{0.5}O_3)_{0.7}(CaAl_{0.5}Ta_{0.5}O_3)_{0.1}(LaAlO_3)_{0.2}]$, with lattice parameters well matched to YBCO.

The structural properties of the films are studied using Philips XPert Pro Alpha-1 MPD diffractometer and the surface microstructure is investigated by atomic force microscope MultiMode Nanoscope IIIA (Digital Instruments). The YBCO films are patterned by photolithography into 50 μm strips for transport measurements (resistance $R(T)$ and I-V characteristics), which are carried out in the 4–300 K temperature range using standard DC four-probe method. The T_c is defined at $R/R_N = 0.1$,

where R_N is the normal-state resistance just above the superconducting transition, and j_c is defined by the criterion of voltage exceeding $\approx 1\times10^{-6}V$ over 2 mm between the voltage leads.

3. Experimental results and discussion

3.1. CeO_2 buffer films

In the first step we have evaluated the influence of recrystallization temperature (T_r) on the structural parameters of CeO_2 films. Figure 1 shows the full width at half maximum (FWHM) for (400) reflection, extracted from the θ–2θ-scans. We see that the FWHM decreases with increasing T_r, indicating that the structural order is markedly improved for larger T_r. However, the study of CeO_2 film surface by AFM shows that this improved structural quality is accompanied by the appearance of the porosity in the films. Both the density and the size of the pores increase with increasing T_r, until above T_r=1050°C cracks appear in the films. To avoid cracking we have chosen the temperatures below T_r=1000°C as the temperatures for recrystallization.

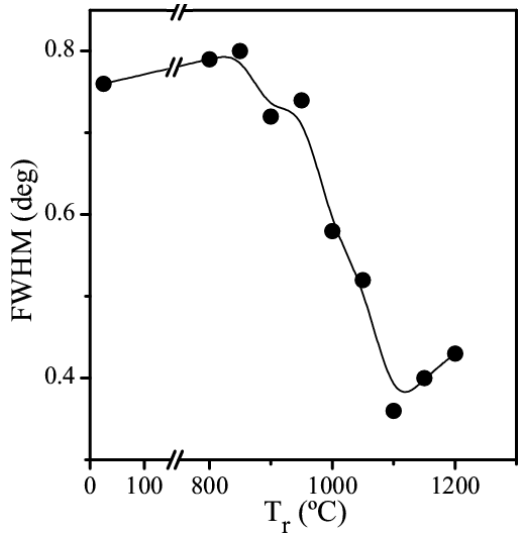

Fig. 1. Plot of the FWHM of the (400) θ-2θ-scan for CeO_2 films with respect to re-crystallization temperature.

Further optimization of the buffer layer has been carried out by varying the buffer layer thickness. Figure 2 shows a series of AFM images for films with the thicknesses equal to 30 nm (top raw), 60 nm (middle raw), and 90 nm (bottom raw), recrystallized at three different temperatures. The numbers in the bottom right corners indicate the root mean square roughness (RMS), calculated for the film area 5 μm by 5 μm. We observe that in thinnest films (30 nm) the RMS is reduced with increasing T_r, reaching small value of 1.26 nm for the film recrystallized at T_r=1000°C. This smooth surface is likely to result in good quality YBCO films deposited on the top. In case of thicker buffer layers recrystallization does not produce such a well-defined improvement of the surface, leading instead to increased porosity and large values of RMS. We conclude that the most promising is the use of thin buffer layer, recrystallized at T_r=1000°C.

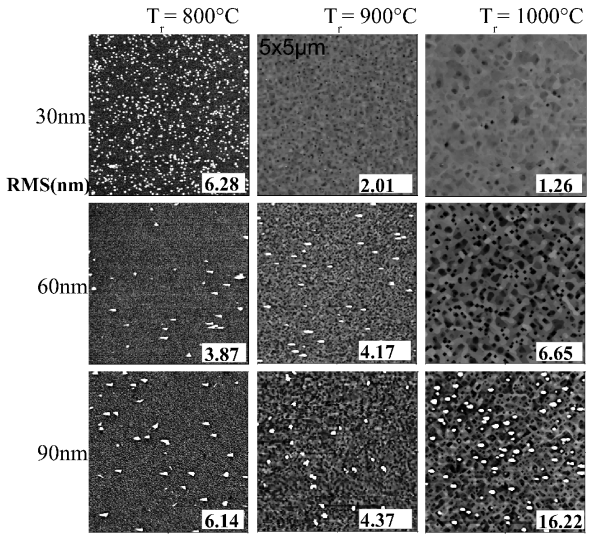

Fig. 2. AFM images of CeO_2 films of different thicknesses.

3.2. YBCO films

To verify the above conclusion we have deposited two series of YBCO films on the top of CeO_2 buffer layers, and we have studied both the structural and the transport properties of YBCO. The first series contains YBCO films of identical thickness of about 170 nm, deposited on the top of buffer layers of various thickness (30 to 90 nm). We have found that the smoothest YBCO films grow on the thinnest CeO_2 buffer (30 nm), as could be expected based on the RMS of the buffer layers. Figure 3a shows the temperature dependence of j_c for some of these YBCO films, together with $j_c(T)$ for YBCO films deposited on well-matched SAT–CAT–La substrates without any buffer layer. We observe that the T_c in the films grown on sapphire is only slightly reduced in comparison with the YBCO grown on well-matched substrate, and remains in the range 73 K to 85 K. However, the j_c is markedly reduced, in some films by an order of magnitude. The best values of j_c are obtained for films deposited on thin (30 nm) buffer layer, confirming the conclusion that the thin buffer is the best choice.

The second series contains YBCO films of various thickness, in the range 100 nm to 300 nm, deposited on thin (30 nm) buffer layer (full points in Fig. 3b). We find that the j_c initially increases with increasing YBCO thickness, but it starts to decrease when the YBCO thickness becomes larger than 250 nm. This is compatible with the results observed previously, attributed to microcracking, which occurs in YBCO films on CeO_2-buffered Al_2O_3 due to thermal strain during cooling when the YBCO film thickness exceeds

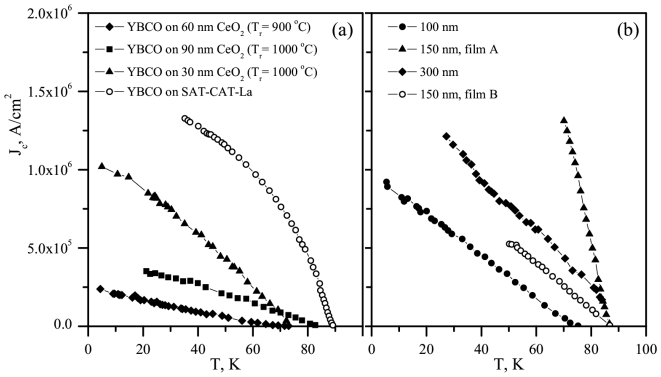

Fig. 3. Temperature dependence of j_c of YBCO films deposited on 30 nm CeO_2 buffer annealed in 1000°C.

a value of $\approx$ 250–300 nm [6]. Interestingly, the results for 150 nm film (film A) indicate that the j_c is quite large, maybe even exceeds, the j_c observed for YBCO on well-matched substrate of SAT–CAT–La. However, we observe some problems with the reproducibility of this result. This is illustrated by the data measured on a second film B with the thickness of 150 nm (open points), which shows considerably lower j_c. Preliminary investigation shows very similar AFM images on films A and B. On the other hand, c-axis lattice parameter is slightly smaller in film A, suggesting better oxygenation of this film. This may affect the behavior of j_c, although it is difficult to understand why the effect would be so large while the difference in T_c of both films is negligible. Further studies are necessary to clarify the origin of j_c variability in these films.

4. Conclusions

We have performed study of CeO_2 film properties grown on sapphire substrates in order to obtain buffer layers suitable for deposition of YBCO films characterized by good superconducting parameters. We have found that 30 nm buffers recrystallized at 1000°C show the best structural properties. YBCO films deposited on such buffered substrates are characterized by good superconducting properties, but further improving of the reproducibility of the critical current density is needed.

Acknowledgements

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