Effect of noble metal buffer layers on superconducting $YBa_2Cu_3O_7$ thin films

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Superconducting $YBa_2Cu_3O_7$ thin films have been prepared by using a magnetron sputtering system in the single-source mode. Samples deposited on [100] single-crystal MgO with and without a Au buffer layer all show high transition temperatures (82–87 K). The use of a Au buffer layer significantly improves the superconducting properties, particularly the Meissner effect and critical current density (3.3×10^6 A/cm² at $T = 2$ K and 3.5×10^4 A/cm² at $T = 77$ K). The Au films remain metallic after high-temperature annealing in an oxygen atmosphere. We propose to use Au buffer layers as current shunts to protect superconducting films and devices.

The recent discovery of high T_c cuprate superconductors has generated unprecedented research interest. For many applications as well as fundamental studies, it is essential to prepare these materials in the form of thin films on suitable substrates.[1-4] One of the major difficulties in the fabrication of Y-Ba-Cu-O superconducting films is the selection of substrates that can survive the stringent annealing conditions (e.g., 900 °C in O_2 atmosphere). To complicate the situation further, the constituents of the superconducting phase (Y, Ba) readily react with many rather inert substrates, thus degrading the superconducting properties. At present, single-crystal $SrTiO_3$ has proven to be the best substrate.[1-4] However, its high cost and very large dielectric constant are unattractive.

To circumvent these problems, it is desirable to find some other substrates and buffer materials which, when deposited between the substrate and the thin film, would serve as a diffusion barrier. In this work, we present the properties of $YBa_2Cu_3O_7$ thin films deposited on [100] MgO substrates. We explore the possibility of using noble metal films as barriers to prevent the interface diffusion between the superconducting film and the underlying substrate. It has been found that a Au buffer layer improves the quality of the films, particularly the Meissner effect and the critical current density (j_c).

The thin-film samples were deposited by magnetron sputtering. In this work, we describe only the results from samples deposited with a single sputtering source onto a rotating substrate platform. Deposition of the $YBa_2Cu_3O_7$ film was carried out at a rf power level of 50 W. Targets having the stoichiometry of $YBa_2Cu_3O_7$ were ground, pressed, and sintered at 950 °C a few times. Gold was chosen as the buffer layer because it remains metallic under high-temperature oxygen annealing. Furthermore, T_c is not significantly reduced when substantial amounts of gold (e.g., 30% volume fraction) are intentionally sintered with $YBa_2Cu_3O_7$ powders. This indicates that the presence of Au has little effect on the properties of the oxide superconductors. The thin Au buffer layer and the superconducting thin films were deposited under an argon pressure of 4.5 mTorr with the substrate rotating at about 6 rpm to assure uniform coverage. The substrate platform was neither intentionally heated nor cooled; its temperature was about 100 °C due to plasma heating.

The as-sputtered films were always amorphous and insulating. They became superconducting only after they were annealed in O_2 at 920 °C for 1–3 h, and then cooled to room temperature at a rate of 2 °C/min. The low cooling rate is crucial in providing the necessary oxygen content.

The thicknesses of the Au buffer layers and the Y-Ba-Cu-O films were determined by using a Dektak profilometer. We found that the thickness of the Y-Ba-Cu-O films before and after annealing was essentially identical. This implies that the oxygen content of the as-sputtered film is already quite high. Furthermore, the density of the as-sputtered films is close to that of the superconducting phase. Annealing at high temperature in O_2 serves mainly to crystallize the sample into the orthorhombic structure and make minor adjustments to the oxygen content.

The structures of the thin films have been examined by using x-ray diffraction in θ-2θ geometry. The scans of the samples on [100] MgO with and without Au buffer layers

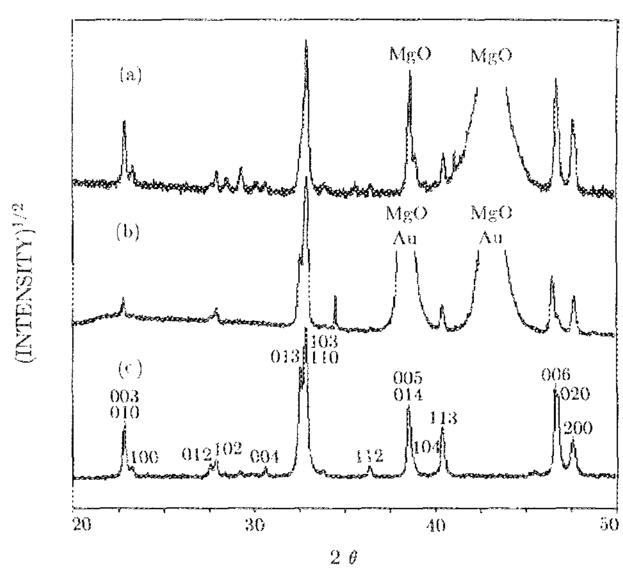

FIG. 1. θ-2θ x-ray diffraction patterns of (a) $YBa_2Cu_3O_7$ thin film on [100] MgO substrate, (b) thin film on MgO with a Au buffer layer, and (c) bulk sintered $YBa_2Cu_3O_7$ powders.

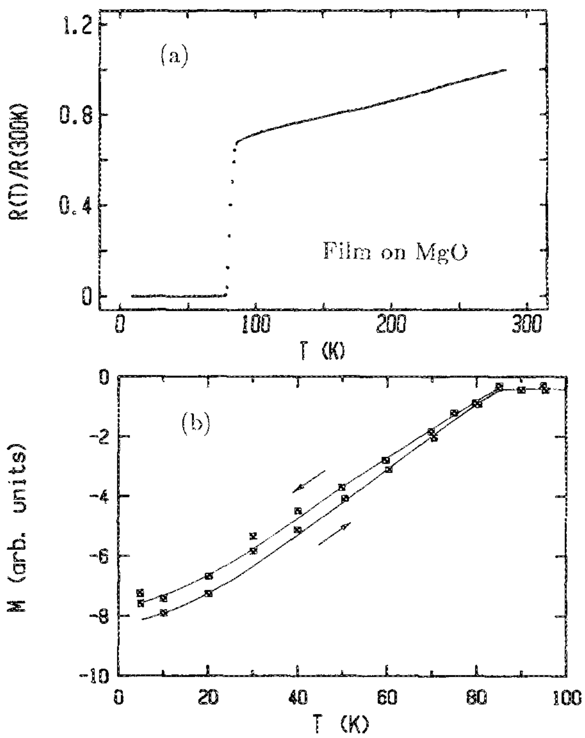

FIG. 2. (a) Temperature dependence of normalized resistance, and (b) zero-field-cooled and field-cooled magnetizations under an external field of 50 Oe for the $YBa_2Cu_3O_7$ superconducting film on MgO substrate.

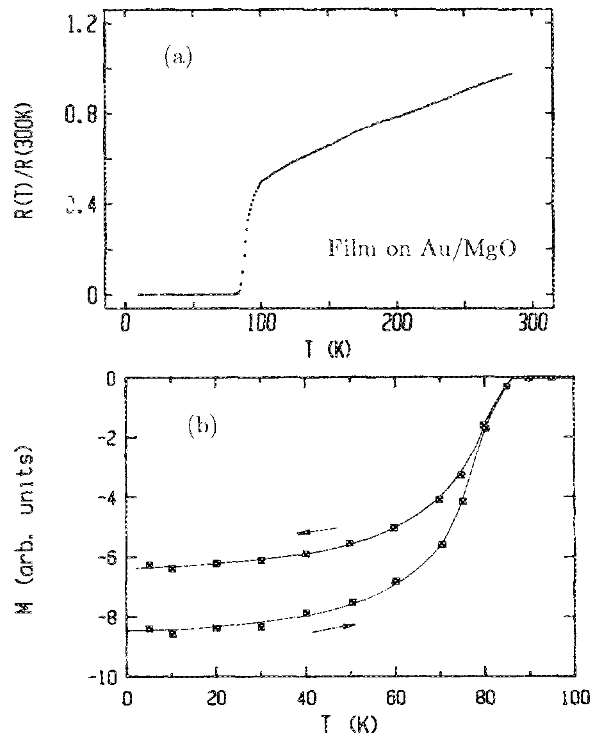

FIG. 3. (a) Temperature dependence of normalized resistance, and (b) zero-field-cooled and field-cooled magnetizations under an external field of 50 Oe for the $YBa_2Cu_3O_7$ superconducting film on MgO with a Au buffer layer.

are shown in Fig. 1. Both samples are primarily single phase with only minor differences upon comparison with a bulk $YBa_2Cu_3O_7$ sample. Grain orientation is generally random, although the [200] peaks are slightly enhanced indicating that a small excess of the grains is oriented with the a axis perpendicular to the film plane. The lattice parameters for the film with a Au layer are $a = 3.814(3)$ Å, $b = 3.885(3)$ Å, $c = 11.72(1)$ Å, while those for film without the Au layer are $a = 3.817(2)$ Å, $b = 3.882(2)$ Å, $c = 11.668(5)$ Å.

In Fig. 2(a) we show the resistive transition of the $YBa_2Cu_3O_7$ film on the [100] single-crystal MgO substrate. A rather sharp transition free of any resistive tail is obtained, with T_c (mid-transition) = 82 K, T_c(onset) = 87 K, and $T_c(R = 0) = 78$ K. Above T_c the temperature coefficient (TCR) is positive, typical of metallic samples. We have found a positive TCR in the normal state only in films deposited on MgO and $SrTiO_3$. The films deposited on polycrystalline Al_2O_3 and SiO_2 substrates exhibit much lower T_c and a negative TCR. Thus MgO is an attractive substrate, second perhaps only to $SrTiO_3$.

Another necessary measurement is the magnetization, which is sensitive to the homogeneity of the sample. In Fig. 2(b) we present the magnetization of the sample as a function of temperature obtained from a superconducting quantum interference device (SQUID) magnetometer under an external field of 50 Oe. Both zero-field-cooled (ZFC) (flux-exclusion-effect) and field-cooled (FC) (Meissner effect) measurements were performed. T_c is 87.5 K, in good agreement with the resistive measurement. However, the superconducting transition is by no means a sharp one. This example illustrates that, while obtaining a good resistive transition in superconducting films may be nontrivial, a sharp resistive transition cannot assure good superconducting properties.

In Fig. 3 we show the results of a Y-Ba-Cu-O film on a [100] MgO substrate coated with 2000 Å of Au buffer layer. The resistance measurement shows T_c(mid-transition) = 88 K. The improvement of the superconducting properties due to the Au buffer layer is best revealed from the low field magnetization measurement as shown in Fig. 3(b). A much sharper transition appears at $T_c = 86$ K. The Meissner effect reaches 75% of the flux-exclusion effect, a value much larger than that of a bulk sintered $YBa_2Cu_3O_7$ sample, indicating that the thin film has good uniformity and high density.

We have determined $j_c(H)$ from magnetic hysteresis between 0 and 50 kOe at $T = 2$ and 77 K. For a flat sample with the sample plane parallel to the magnetic field, the value of j_c can be determined from the magnetizations[5] under increasing (M^+) and decreasing (M^-) fields according to

$$j_c = 20(M^+ - M^-)/d, \quad (1)$$

where j_c is in units of A/cm^2, M in units of emu/cm^3, and d in units of cm. The magnetizations in increasing and decreasing fields are shown in Fig. 4. The shapes of the curves are largely due to the magnetizations of the substrates; j_c is only related to the difference between M^+ and M^-. Both samples are rectangular in shape, approximately 0.62×0.42 cm. The thicknesses of the films with and without a Au layer

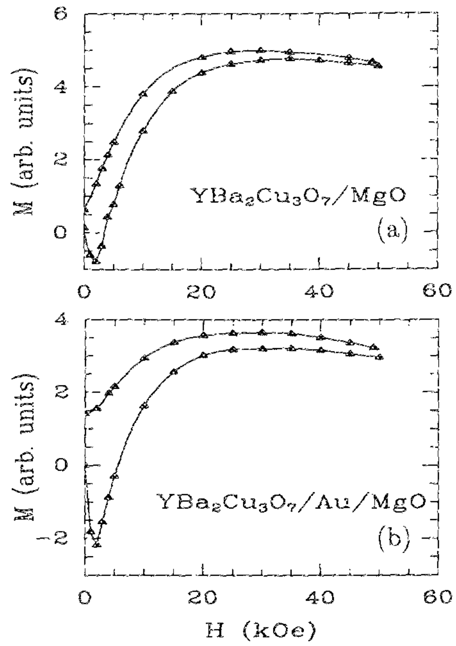

FIG. 4. Magnetic hysteresis curves at $T=2$ K for (a) $YBa_2Cu_3O_7$ thin film on MgO and (b) thin film on MgO with a Au buffer layer.

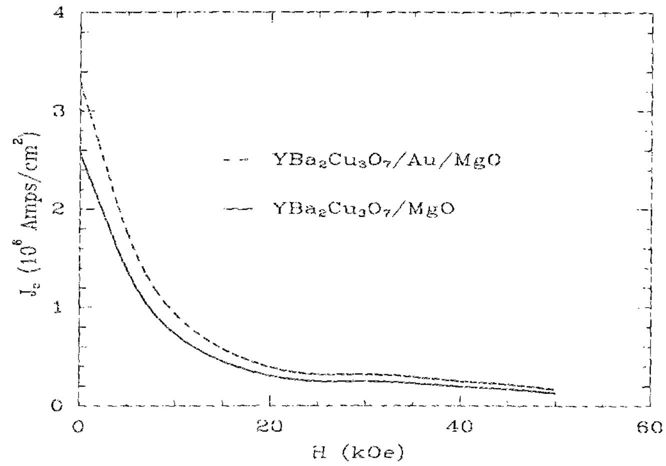

FIG. 5. Critical current densities as a function of magnetic field at $T=2$ K for $YBa_2Cu_3O_7$ thin film on MgO with and without a Au buffer layer.

are 3.3 and 3.7 μm, respectively. The j_c deduced from the magnetization curves are shown in Fig. 5 as a function of external field up to 50 kOe. At zero field, $j_c = 3.3 \times 10^6$ A/cm^2, and 2.6×10^6 A/cm^2 are obtained for the samples with and without the Au layer. At $T = 77$ K, the enhancement of j_c is much more obvious. For the sample with a Au layer, $j_c = 3.5 \times 10^4$ A/cm^2, while for the sample without a Au layer, $j_c = 2.1 \times 10^3$ A/cm^2. Overall, the Au buffer layer significantly improves the quality of superconducting films deposited on MgO.

The advantages of using a Au buffer layer are twofold. Firstly, Au does not readily form oxides under high-temperature oxygen annealing, and it prevents diffusion between the thin film and the substrate. The Au layer can be visually inspected from the back side of the substrate after annealing. It retains its original luster. At the interface between the Au buffer layer and the superconducting film, diffusion may be difficult to avoid. However, the presence of Au has a negligible effect on the superconducting properties of the high T_c superconductors. Secondly, the room-temperature resistivity[2] in the new oxide superconductors is 500–1000 $\mu\Omega$ cm, 2 to 3 orders of magnitude larger than that of Cu. Thus, the underlying Au buffer layer can serve as an excellent current shunt. In the normal state of the superconductor, large currents will pass through the Au film, protecting the superconducting materials. Indeed, our samples with Au buffer layers have much lower resistivities than those without the buffer layers. There is one additional point worth mentioning: since the oxide superconducting films are rather vulnerable to environment, a coating of Au after annealing can also prevent the thin films from deteriorating.

In summary, thin films of high T_c $YBa_2Cu_3O_7$ have been prepared by using magnetron sputtering onto MgO substrates with and without Au buffer layers. In all cases, T_c in excess of 80 K and metallic behavior in the normal state have been observed. The samples with a Au buffer layer show excellent Meissner effect and a significantly higher critical current density.

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[1] P. Chaudhari, R. H. Koch, R. B. Laibowitz, T. R. McGuire, and R. J. Gambino, Phys. Rev. Lett. 58, 2684 (1987).
[2] B. Oh, M. Naito, S. Arnason, P. Rosenthal, R. Barton, M. R. Beasley, T. H. Geballe, R. H. Hammond, and A. Kapitulnik, Appl. Phys. Lett. 51, 852 (1987).
[3] J. Kwo, T. C. Hsieh, R. H. Fleming, M. Hong, S. H. Liou, B. A. Davidson, and L. C. Feldman, Phys. Rev. B 36, 4039 (1987).
[4] D. Dijkkamp, T. Venkatesan, X. D. Wu, S. A. Shaheen, N. Jisrawi, Y. H. Min-Lee, M. L. McLean, and M. Croft, Appl. Phys. Lett. 51, 619 (1987).
[5] W. A. Fietz, M. R. Beasley, J. Silcox, and W. W. Webb, Phys. Rev. 136, A335 (1964).