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Large area deposition of $YBa_2Cu_3O_7$ films by thermal co-evaporation

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Abstract

Reactive evaporation techniques cannot be simply scaled up for larger YBCO films because the zone of enhanced O_2 pressure in front of the substrate will also increase in height and is impenetrable for the metal vapours. We have circumvented this problem by separating the evaporation and oxidation zones. Smooth films without any precipitates on a 10x10 μm^2 scale could be fabricated on 30x30 mm^2 MgO substrates. The homogeneity of the stoichiometry and of the thickness was 2%. Good normal state electrical properties have been achieved: $\rho(300\ K)/\rho(100\ K) = 3.0$ and $\rho(100\ K) = 40\ \mu\Omega cm$. The superconducting transition temperature was typically 87 K with a variation of $\Delta T_c \leq 0.5$ K over the sample area while J_c at 77 K varied between 1 and $6 \cdot 10^6$ A/cm^2. The microwave surface resistance at 77 K was found to be $R_{eff} = 60\ m\Omega$ at 87 GHz. In addition, we deposited YBCO films on 3 inch $LaAlO_3$ wafers with $T_c = 86\ K \pm 1\ K$ and tested the feasibility of deposition of YBCO films on 4 inch silicon wafers with a YSZ buffer.

1. Introduction

YBCO thin films on single crystal substrates have shown the highest potential for practical applications. For most electronic devices 10 x 10 mm^2 substrates have been used until now. However, for many microwave applications such as antennas, delay lines or filters larger film areas are desirable. We report here on the fabrication of large area films by thermal evaporation.

In previous work [1,2] we have shown that thermal reactive co–evaporation is a quick and versatile method for growing high quality YBCO thin films without any precipitates at rather low substrate temperatures. As the background pressure of about 5×10^{-5} mbar is far below the thermal stability limit of $YBa_2Cu_3O_x$ [3,4] the O_2 pressure was locally enhanced in front of the substrate. This technique cannot be simply scaled up to larger substrates because the zone of high pressure will increase in height and is impenetrable for the metal vapours. We have circumvented this problem by separating the evaporation and oxidation zones.

2. Large area film deposition

A schematic diagram of our technique is depicted in Fig. 1. The samples were mounted on a rotating disk. A fixed oxidation cup was closely placed under one half of the disk while the other half was freely accessible to the metal vapours. The gap between the disk and the oxidation chamber was about 0.5 mm. This was achieved by embedding the sample flush with the disk surface.

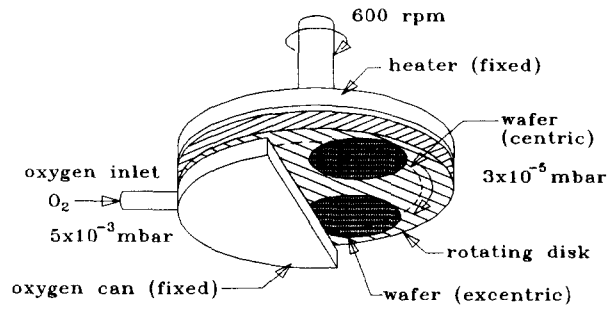

Figure 1. Rotating disk substrate holder: film deposition and oxidation are separated spatially.

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The controlled flow resistance between the brim and the disk allowed an O_2—pressure drop from 5×10^{-3} mbar inside the cup to 3×10^{-5} mbar outside. (We measured the oxygen pressure inside the cup by an oxygen sensor consisting of a sputtered $SrTiO_3$ film doped with Nb [5]. At constant temperature the electrical conductivity of the film rises with decreasing oxygen partial pressure because oxygen vacancies are formed.).We used 30 x 30 mm^2 MgO substrates which were mounted excentrically on the disk (see Fig. 1), and 3 inch $LaAlO_3$ wafers which were positioned centrically on the substrate holder. Films of up to 9 cm diameter can be deposited in this way. The rotating disk and the oxygen cup were were made of stainless steel. They were radiatively heated by miniature coaxial heating cables. The whole assembly was shielded by nichrome radiation shields to minimize the heater input power which was 800 W at 650 °C substrate temperature. We measured the substrate temperature by a pyrometer.

Two turbo molecular pumps (pumping speed 1800 l/s) maintained a background pressure of 3×10^{-5} mbar in the evaporator. We used tantalum boats for the evaporation of Y and Ba, and a tungsten boat for Cu. The rates were controlled for each boat separately by three quartz crystal monitors. After evaporation the films were cooled down to room temperature within 90 min in an O_2 atmosphere of 200 mbar.

As the metal atoms condense on the substrate before they are oxidized in the oxidation cup, it is interesting to study the film properties as a function of the rotation frequency. As shown in Fig. 2, the superconducting transition temperature drops markedly below 1 Hz. In addition, the film surface is getting rough below this frequency. This is caused by formation of different phases as revealed by SEM micrographs. For an average evaporation rate of 0.1 nm/s and a frequency of 1 Hz each metal atom is exposed to oxygen before the next layer is formed. Optimum film quality has been obtained for a rotation frequency of 10 Hz and an average evaporation rate of 0.2 nm/s.

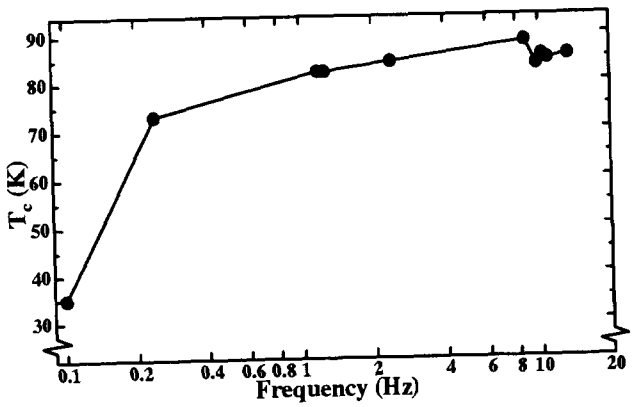

Figure 2. T_C versus revolutions per second of the rotating disk.

3. Film characterization

Fig. 3 shows the variation of thickness of a YBCO film on a 3 inch $LaAlO_3$ wafer as a function of the distance r from the centre. The thickness decreases by about 20 % in an area of 2 mm diameter at the centre. Outside this region it is constant within 2 %. The same thickness variation of 2 % is also found for films on 30 x 30 mm^2 MgO substrates mounted excentrically on the disk. X—ray diffraction patterns revealed c—axis oriented single phase $YBa_2Cu_3O_{7-x}$ with c = 1.169 nm and with a FWHM of 0.32° for the rocking curve of the YBCO (005) peak on MgO substrates. Volume fractions of misaligned c—axis oriented grains were determined by ϕ—scans of the YBCO (103) reflection. The most prominent orientation apart from YBCO (100) $\parallel$ MgO $\{100\}$ defining $\phi = 0$ was one differing by 45°. We found I(45°)/I(0°) = 0.2% in the best case.

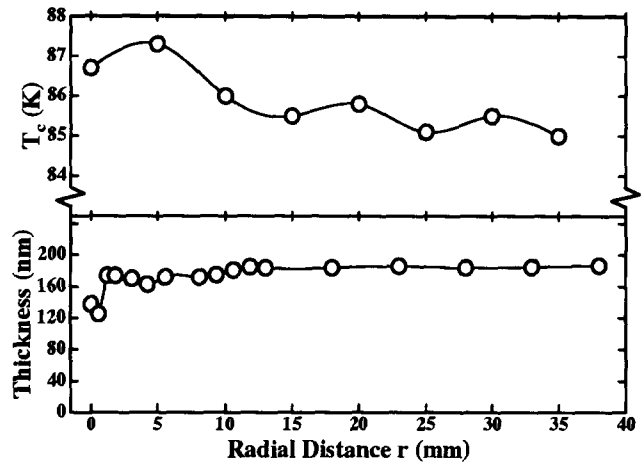

Figure 3. Variation of thickness (bottom) and T_C (top) as a function of radial distance for a film on a 3 inch $LaAlO_3$ wafer.

We have measured the Y, Ba and Cu contents by heavy ion Rutherford backscattering and found a variation of about 2 % across the wafers. The impurity levels due to contamination by the heaters, the oxidation cup or the boats were analyzed by SIMS. They were below 0.1 at% for Ni, Fe, Cr and Ta. By optimizing the stoichiometry, films without any precipitates on a 10x10 μm^2 scale and with good superconducting properties could be made. However, the reproducibility of the stoichiometry is still a problem.

4. Superconducting properties

We achieved good normal state electrical properties: $\rho(300\ K)/\rho(100\ K) = 3$ and $\rho(100\ K) = 40\ \mu\Omega cm$, so that the R(T) curve above T_C extrapolated well to R = 0 at T = 0 K. The critical temperatures $T_C(R = 0)$ were in the best case 89 K and typically 87 K with $\Delta T_C(10\%–90\%) \leq 1$ K. The narrow transition to superconductivity also showed up in inductive measurements which we used to check the homogeneity of T_C over the film area. A variation of T_C smaller than 0.5 K was found for 30x30 mm^2 MgO substrates. On 3 inch $LaAlO_3$ wafers, T_C varied between 85 K and 87 K (see Fig. 3). The critical current density J_C was measured on a 50 x 50 μm^2 microbridge patterned by conventional photolithography and wet etching. Applying a 1 μV voltage criterion we found Jc = 3.4 MA/cm^2 at 77 K for a film on MgO (100). The homogeneity of the critical current was studied with an AC susceptibility method [6] at 77 K which was calibrated by a direct measurement of J_C as described above. As shown in Fig. 4, J_C exceeds 10^6 A/cm^2 on the film but there are still significant variations of J_C probably due to substrate imperfections. In a first measurement the microwave surface resistance at 87 GHz was $R_{eff} = 60\ m\Omega$ at 77 K and 12 $m\Omega$ at 10 K, respectively, for a 180 nm thick YBCO film on a MgO substrate [7].

3	5	6	5	6
4	4	4	1	3
4	4	4	1	4
4	4	4	1	3
3	1	1	2	2

x 10^6 A/cm^2

Figure 4. Spatial distribution of J_C (77 K) on a 30x30 mm^2 film on MgO.

5. Conclusions

We have shown that the problem of insufficient oxidation of high T_C films associated with MBE methods at low oxygen pressure can be circumvented by separating deposition and oxidation zones. YBCO films can be made with the same high quality as with conventional methods. Our technique is well suited for large area deposition as has been demonstrated by our 3 inch films on $LaAlO_3$ wafers. Preliminary results show that epitaxial YSZ films can be grown on 4 inch Si (100) wafers with the same technique. Therefore we can grow both the buffer layer and the YBCO in situ without breaking the vacuum.In addition, it is possible to upscale the present setup for a deposition on 6 inch wafers.

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