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Growth of epitaxial $\text{YbBa}_2\text{Cu}_3\text{O}_7$ superconductor by liquid-gas-solidification processing

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Superconductive films of $\text{YbBa}_2\text{Cu}_3\text{O}_7$ have been epitaxially grown on SrTiO_3 substrate by a novel liquid-gas-solidification process. A layer of metallic YbBa_2Cu_3 melt was coated on the substrate and *in situ* oxidized. The oxide films grown on the SrTiO_3 (100) substrate are epitaxially oriented in structure with the *c* axis normal to the plane film. Values of T_c ($R = 0$) of 82 K with a transition width of 1 K have been achieved. The critical current density J_c is typically 10^5 A/cm² at 50 K and 10^4 A/cm² at 77 K.

The discovery of the possible existence of superconductivity in La-Ba-Cu-O above 30 K by Bednorz and Müller¹ in 1986 stimulated worldwide research activity. More recently, the discoveries of bulk superconductivity above 77 K, the boiling temperature of liquid N_2 , in a large family of compounds of prototype $\text{YBa}_2\text{Cu}_3\text{O}_7$,² Bi-Sr-Ca-Cu-O,³ and Ti-Ba-Ca-Cu-O system⁴ have opened the door to wider application of superconductors, because liquid nitrogen is considered to be one of the most advantageous cryogenic refrigerants.

There are two general approaches for forming superconductive oxide bodies. Thin films are formed by deposition of material on a substrate, either by sputtering,⁵ electron beam and thermal co-evaporation,⁶ molecular beam epitaxy,⁷ pulse laser evaporation,⁸ or decomposition from solution.⁹ On the other hand, bulk bodies and thick films are generally produced by synthesizing of a powder of the appropriate composition, and forming the powder into the desired shape by hot pressing,¹⁰ drawing,¹¹ extrusion,¹² or a powder-in-binder method.¹³ All these methods require post-heat treatment at elevated temperature to produce the appropriate structure. An alternate method consists of the melting of an oxide powder and forming bulk bodies by solidification of the oxide melt.¹⁴

Work to date has shown that thin films of $\text{YBa}_2\text{Cu}_3\text{O}_7$ can have high J_c of the order of 10^6 A/cm² at 77 K.^{6,15} Indirect measurements of J_c from magnetization loop measurements for single crystals¹⁶ and crystalline grains¹⁷ suggest an intrinsic J_c ($T = 60$ K, $H = 0$) $\sim 10^5$ A/cm² of individual grains. However, the critical current density of bulk bodies produced by sintering of the particles is relatively small of the order of 10^3 A/cm². The huge difference in J_c between a single particle and an assembly of particles is generally attributed to the presence of weak links between adjacent particles, frequently associated with the surface inhomogeneity of a particle and with the contact interface between particles. Indeed, melt-grown polycrystalline $\text{YBa}_2\text{Cu}_3\text{O}_7$ using directional solidification, which is essentially 100% dense and exhibits a highly textured microstructure, shows an improved $J_c = 1.7 \times 10^4$ A/cm² at 77 K in zero field.¹⁴

We have developed a novel, simple, scalable processing method, i.e., liquid-gas-solidification process (LGS), which differs fundamentally from all relevant processes mentioned

above. This process consists of (1) forming a melt (the precursor melt, e.g., YbBa_2Cu_3) composed of the metallic components of the compound (e.g., the cuprate oxide) to be formed, and (2) introducing the missing element, oxygen, into the precursor melt to form the oxide. This process produces single phase materials with uniform composition. As an example, we present the fabrication of epitaxially grown thick films ($3 \mu\text{m}$ thick) of $\text{YbBa}_2\text{Cu}_3\text{O}_7$ on a SrTiO_3 (100) substrate. Materials produced by this process have T_c ($R = 0$) = 82 K with a sharp transition width of 1 K. The critical current density J_c is typically of 3×10^4 A/cm² at 70 K which is comparable to that of bulk single-crystal samples.

A ternary alloy of YbBa_2Cu_3 was prepared by induction melting of high-purity metals. The alloy was placed in a high-purity alumina crucible and was sealed together with a SrTiO_3 (100) substrate which was mounted on a translational stage. The assembly was heated in a resistance furnace above the melting temperature T_m ($= 870^\circ\text{C}$) of the alloy. The SrTiO_3 substrate was then immersed into and pulled out of the melt. A melt layer $\sim 2 \mu\text{m}$ thick coated the substrate. Oxidization of the melt coating was conducted *in situ* by introduction of oxygen into the tube. The resultant oxide film was annealed at 500°C for 5 h.

X-ray diffraction measurements were made with a Philips APD 6300 automatic diffractometer with $\text{Cu } K_\alpha$ radiation. As shown in Fig. 1, the film grown on a SrTiO_3 (100)

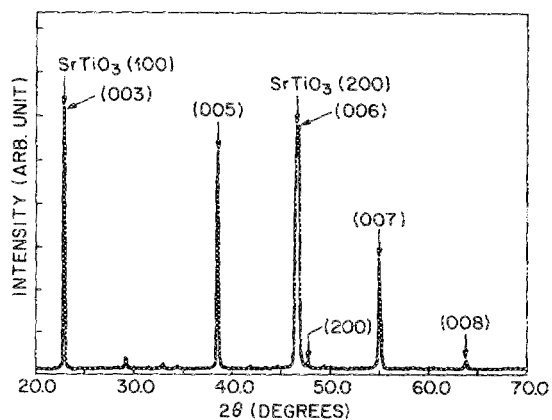


FIG. 1. X-ray diffraction pattern of an epitaxial $\text{YbBa}_2\text{Cu}_3\text{O}_7$ film grown on a SrTiO_3 (100) substrate.

substrate shows an epitaxially oriented structure with the c axis perpendicular to the film plane. The lattice parameters of the orthorhombic $\text{YbBa}_2\text{Cu}_3\text{O}_7$ were determined to be $a = 3.808 \text{ \AA}$, $b = 3.887 \text{ \AA}$, and $c = 11.659 \text{ \AA}$. It is clear in Fig. 1 that the lattice parameters b and $c/3$ of the o phase are nearly perfect matched to the cubic lattice $a_0 = 3.89 \text{ \AA}$ of the cubic SrTiO_3 . One might expect that the film would grow epitaxially with b and c axes in the plane, i.e., a axis normal to the film. However, the growth of $\text{YbBa}_2\text{Cu}_3\text{O}_7$ is highly anisotropic, producing platelets with growth along the c axis limiting the thickness. The mismatch between a and a_0 ($\Delta a/a_0 = -0.02$) is small, however, and the film growth habit is epitaxial with the a and b axes in the plane of the substrate (i.e., c axis normal to the plane).

Figure 2 is a scanning electron micrograph (SEM) of a film grown on (100) SrTiO_3 . The film exhibits rectangular tile patterns with platelet dimensions $\sim 10 \mu\text{m} \times 10 \mu\text{m} \times 2 \mu\text{m}$. The growth rates of the o - $\text{YbBa}_2\text{Cu}_3\text{O}_7$ crystal are highly anisotropic with the fast growth along a and b axes, and it grows as a platelet with c axis normal to the plane. A distinct separation among platelets was frequently seen in the SEM; however, close examination near the film-substrate interface revealed a uniform interconnection between them.¹⁸ Transmission electron microscopy reveals that the film consists of twinned domains of 200 \AA in size.¹⁹

Four-terminal dc transport measurements were made using indium contacts. The film was shaped with a carbide tool to restrict current flow to a narrow path $\sim 150 \mu\text{m}$ in width. The resistive to superconducting transition at a $2.5 \mu\text{A}$ current level is shown in the insert in Fig. 3. The T_c ($R = 0$) for this sample is 82 K with (10%–90%) transition width of 1–2 K. Extrapolation of the high-temperature resistance yields a small resistance ratio $R(T = 0)/R(T = 300 \text{ K}) = 1/5$, which is a characteristic of high quality material. J_c measurements are shown in Fig. 3 for temperatures between T_c and 50 K . The beginnings of the resistive transition were monitored at several levels of applied current. The critical current J_c was defined by a flux flow resistance of $10^{-3} R_N$, where R_N is the normal resistance at T_c , or a

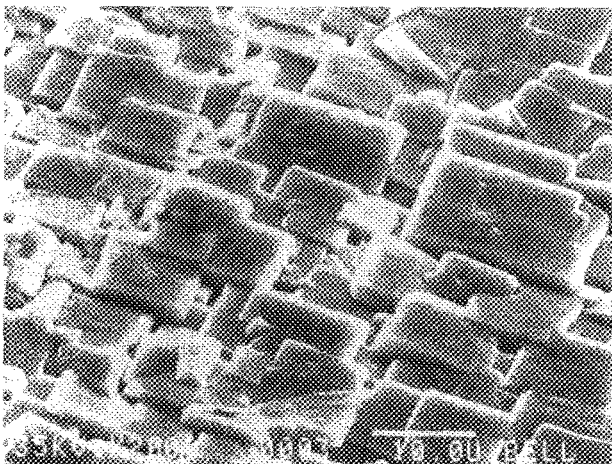


FIG. 2. Scanning electron micrograph (SEM) of the film grown on a $\text{SrTiO}_3(100)$.

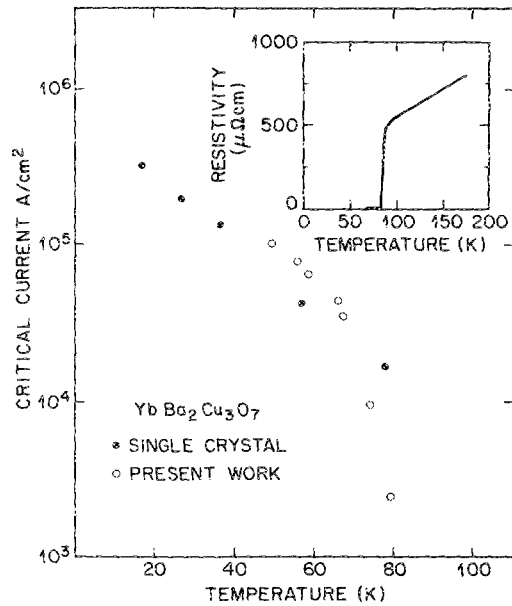


FIG. 3. Temperature dependence of resistivity (insert) and critical current density J_c of the film corresponding to Fig. 2.

measured voltage of $1 \mu\text{V}/\text{cm}$. As shown, J_c increases exponentially near T_c and then more gradually below 70 K . Typically, J_c attains values of $10^4 \text{ A}/\text{cm}^2$ at 77 K and $10^5 \text{ A}/\text{cm}^2$ at 50 K which are comparable to values reported for single-crystal $\text{YbBa}_2\text{Cu}_3\text{O}_7$.¹⁶ The present sample shows T_c ($R = 0$) = 82 K , which is lower than that ($T_c = 90 \text{ K}$) reported for the single crystal. The causes of the lower T_c of the sample are not clear and are under investigation.

The improved J_c of the thick films produced by the present process can be attributed to the significant improvement in compositional and structural uniformity. Epitaxially grown films can, in principle, eliminate the weak links associated with grain boundaries. The improved homogeneity is believed to be due to the fact that a single, well-defined product of the reaction of oxygen with the melt is formed.

The uniform distribution of metallic constituents is a critical factor in producing improved superconducting materials. Superconducting cuprate oxides produced by the oxidation of precursor solid phase alloys have been reported.²⁰ The resultant oxides show a multiphase structure and exhibit low critical current density $J_c \sim 1 \text{ A}/\text{cm}^2$.²¹ Apparently, compositional and structural inhomogeneity arise from the decomposition of the solid phase alloy during the oxidation process. The distinguishing feature and fundamental advantage of the LGS process is that the desired compound formation occurs at an accessible and kinetically favorable liquidus phase boundary rather than the more limited solid-solid phase transformation path.

The LGS method has other advantages. For instance, it is a one-step procedure capable of producing bulk bodies, whereas the established sintering techniques require repeated calcining and comminuting of the starting material. Finally, we expect that the inherent advantages discussed above will give LGS a wide range of application in the processing of ceramic materials, in general.

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