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Critical Current Densities of Submillimeter Single-Grain Tl-2223 Superconducting Thin Film

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Large single-crystalline superconducting grains of $Tl_2Ba_2Ca_2Cu_3O_{10+x}$ (Tl-2223) with sizes up to 150 μm×150 μm×1 μm are obtained on both MgO(100) and $LaAlO_3$ (100) substrates by a single-target dc-sputtering process followed by appropriate postannealing at temperatures around 910°C. Such superconducting grains were isolated and patterned into 10-μm-wide bridges by reactive ion etching. The etching process was found to only minimally affect the transition temperature, T_c, typically around 110 K, of the films. Although there was evidence that these films were indeed two-dimensional in nature, the critical current densities, J_c, were greater than 10^6 A/cm^2 at temperatures below 90 K. At lower temperatures, *e.g.*, $T = 30$ K, the J_c value of these films remains well above 10^6 A/cm^2 even with magnetic fields as large as 7 T applied parallel to the *c*-axis of the grains. The results are in sharp contrast to the low J_c values expected from the low-dimensionality nature of this system.

KEYWORDS: high-T_c cuprate films, Tl-based compounds, reactive ion etching, critical current densities, vortex pinning

1. Introduction

Although *in situ* fabrication is generally difficult, $Tl_2Ba_2Ca_2Cu_3O_{10+x}$ (Tl-2223) superconducting thin films have displayed great potential in various superconducting electronics applications. In addition to having higher T_c than their counterparts, it has been demonstrated that the Tl-2223 films can have high critical current densities, J_c,[1] and extremely low surface resistivities.[2,3] However, due to the complex chemistry and high volatility of the Tl oxides (primarily Tl_2O and Tl_2O_3)[4,5] involved in the synthesis of these films, the films obtained through various processes (mostly *ex situ*) not only are polycrystalline in nature, but also usually contain appreciable amounts of secondary impurity phases. As a result, the interpretation of most of the experimental results is inevitably complicated by the grain structure of individual films. In particular, when compared with the results for single-crystalline samples, significant inconsistencies were usually encountered. For instance, it has been shown recently by Brandstätter *et al.*,[6] for unirradiated Tl-2223 single crystals, that J_c obtained from magnetization measurements disappeared at temperatures above 40 K, in sharp contrast to the results reported earlier for thin films.[1–3]

In order to clarify the apparent discrepancies we recently made submillimeter superconducting Tl-2223 thin film grains and fabricated bridges on such individual grains by reactive ion etching (RIE).[7] In this paper, we report on the temperature and field dependences of the critical current densities, $J_c(T, H)$, obtained on such samples. The results, in combination with the two-dimensional characteristics manifested in conductivity fluctuation and Kosterlitz-Thouless transition of vortex pair dissociations,[8] suggest that the limiting factors of J_c for these thin film single-crystalline grains are far more complex than originally expected.

2. Experimental

The detailed procedures of making Tl-2223 superconducting thin films with distinct submillimeter grain structure have been described previously.[7] Briefly, precursor films with thickness of approximately 1 μm were deposited on both MgO(100) and $LaAlO_3$(100) single-crystalline substrates by dc sputtering from a presintered target of Tl:Ba:Ca:Cu with 2:2:2:3 stoichiometry. Since the substrates were not intentionally heated, at the estimated temperature of $\approx$ 130°C during deposition, the as-deposited precursor films were amorphous. As revealed by energy dispersive X-ray spectroscopy (EDS), the nominal composition of the precursor films preserves the same stoichiometry of the target. The as-deposited precursor films were then wrapped with a bulk Tl-2223 pellet using gold foil and sealed with about 1 atmosphere of pure oxygen in a quartz tube. It is noted that the bulk pellet used was obtained from the same batch of target material which showed superconducting transition around 120 K. In order to facilitate grain growth, the postannealing temperature of the present process was raised to as high as 910°C, which was about 20–30°C higher than in the usual processes.[5] As will be shown below, large Tl-2223 thin film grains on both types of substrates were obtained.

To pattern bridges, typically 10 μm in width and 100 μm in length, on selected areas of a film for subsequent transport measurements, the RIE process with complementary wet etching was employed. The details of the RIE process suitable for patterning Tl-based compounds and $YBa_2Cu_3O_7$ will be reported elsewhere.[9] Briefly, for etching Tl-based compounds, a complementary wet etching process for making large-feature-size bridges is required to achieve better results. This is mainly due to the relatively large thickness of the films used in this study. For the RIE process, a gas mixture of $SiCl_4$ and Ar (about 1:1) with a total pressure of 70 mTorr was used. An AZ6112 photoresist about 1 μm thick was applied uniformly on the film surface and then exposed to UV light via a metal mask to form the desired etching mask. A total RF power of 55 watts was employed to induce the RIE process. It is noted that after the above patterning process, both T_{c0} (as high as 113 K) and the transition width of the films remained virtually unchanged. More-over, due to the elimination of grain boundary effects and possible minor secondary phases, the characteristics, in most cases, even showed improvements.[7)]

For transport measurements, the conventional four-probe configuration was used. The temperature was monitored and controlled by two separated thermometers located near the samples. The magnetic fields were applied perpendicular to the film surface using both an electromagnet (for fields up to 0.5 T) and a superconducting magnet capable of providing 9 T field. The critical current density is defined by an 1 μV criterion.

3. Results and Discussion

The typical film microstructure obtained by the process described above is shown in Fig. 1(a). It is noted that the result shown in the figure is for films grown on $LaAlO_3$(100) substrates. For films grown on MgO(100) substrates, though similar grain morphology was obtained, the in-plane orientation of the individual grains appeared to be more random than that seen in Fig. 1(a). As has been discussed previously,[7,10)] this is due mostly to the differences in the degree of film/substrate lattice mismatches. As is evident from Fig. 1(a), distinct individual square Tl-2223 superconducting grains with sizes up to 150 μm in each planar dimension are obtained. The areas with somewhat irregular structure were identified to be the Tl-2212 phases. Also shown schematically in Fig. 1(a) is the selected region wherein the bridges were patterned. As shown in Fig. 1(b), the actual bridges used for transport measurements prepared by the RIE process is entirely within one single grain. The detailed X-ray diffraction analyses on films grown on various substrates and mechanisms of phase formation are described elsewhere.[7)] It is noted that by physically removing the possible complications originating from the film microstructures, "*intrinsic*" properties of this material derived from single crystals in both bulk and thin film forms can be compared for the first time to our knowledge.

Figure 2 shows the typical zero-field temperature-dependent critical current densities, $J_c(T)$, obtained on bridges formed within a single grain for films grown on MgO and $LaAlO_3$ substrates, respectively. We discuss the significance and implication of the present results below. First of all, due to the large spacing between the superconducting CuO layers, systems such as Tl-2223 and Tl-2212 are usually considered to be more 2D-like as compared to their counterparts such as Tl-1223 and YBCO, leading to relatively weaker interlayer coupling and more severe thermal fluctuations.[3,6,11,12)] Indeed, evidence of 2D conductivity fluctuation and vortex-pair dissociation related Kosterlitz-Thouless (K-T) transition was observed in these Tl-2223 single-crystalline grains.[8)] However, depending on the substrate used, subtle differences were observed. For instance, it was found[8)] that the conductivities between T_{c0} to $2T_{c0}$, although fit well to the Aslamazov-Larkin expression:[13)]

$$\Delta\sigma_{AL} = (e^2/16\hbar d)[(T - T_{c0})/T_{c0}]^{-1} \quad (1)$$

for 2D superconductors with correct slope of -1, resulted in different values of effective thickness, d: $d =$

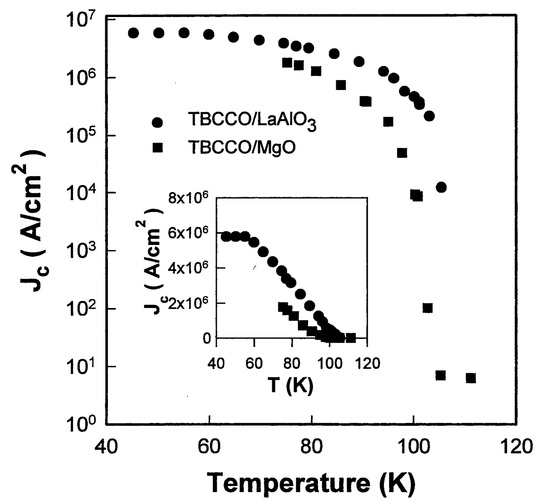

Fig. 2. The $J_c(T)$ results for single-grain Tl-2223 bridges grown on two different substrates. The inset shows the same results plotted in a linear scale to demonstrate the linear $J_c(T)$ behavior over a wide range of temperatures.

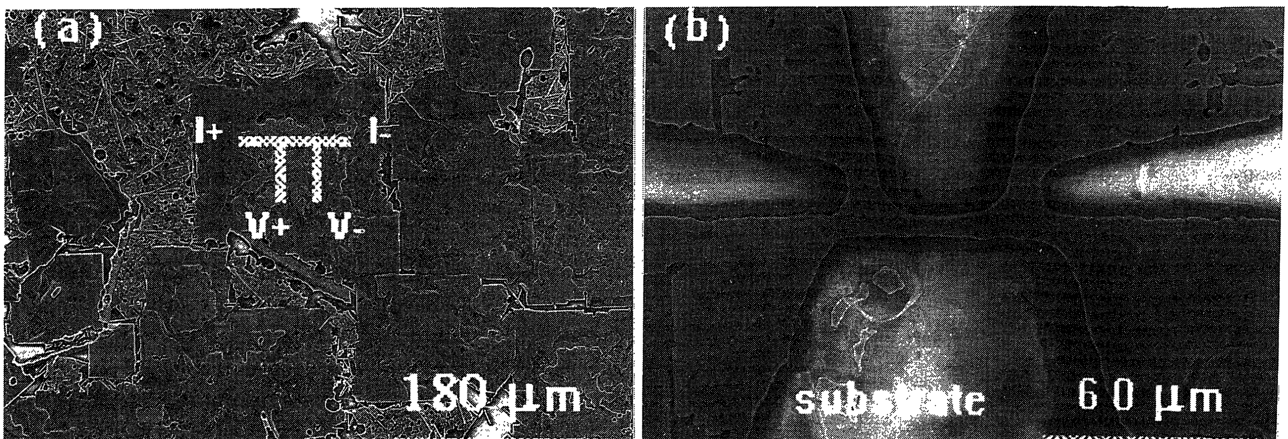

Fig. 1. (a) The typical grain structure of Tl-2223 superconducting thin films grown on $LaAlO_3$(100) substrates as revealed by SEM. The schematic pattern shows the configuration and areas selected to make microbridges for the present studies. (b) The typical bridges within a single thin film Tl-2223 grain made by reactive ion etching.

1.75 nm and $d = 3.5$ nm for Tl-2223/MgO and Tl-2223/$LaAlO_3$, respectively. It is noted that 1.75 nm is approximately the distance between the two trilayered CuO_2 planes and 3.5 nm is exactly the *c*-axis lattice constant of the Tl-2223 system, respectively. This implies that in Tl-2223/MgO the conductivity fluctuations take place essentially within the three layers of CuO_2 planes and coupling between groups of the trilayers is negligible, whereas for Tl-2223/$LaAlO_3$, the average coupling strength between the groups of trilayers is stronger. In addition to the interesting behavior of quasi-particle conduction above T_c described above, it has been found that the correlations between the vortices, as reflected by the vortex-pair dissociation effects below T_{c0}, are also stronger for grains grown on $LaAlO_3$ substrates.[8] These, in fact, are consistent with the present J_c (T) results where J_c of Tl-2223/$LaAlO_3$ is higher than that of Tl-2223/MgO. Considering the fact that they are essentially the same Tl-2223 superconducting thin film grains, the question as to what causes the differences remains.

In term of the process, the only variation between the two cases is that the substrates are different. Since the lattice mismatch is much larger for Tl-2223/MgO (≈9%) than for Tl-2223/$LaAlO_3$ (<1%), it is expected that the film/substrate correlation during the growth of these submillimeter grains would be much less influential in the former. As a result, the strain energy built up during grain growth is much lower in Tl-2223/MgO and the resultant grains become less defective due to the absence of strain relief-induced dislocations. Furthermore, as has been demonstrated previously by Kim *et al.*,[14] there is clear evidence that substrate twinning could also result in high-density dislocations in regions near the film/subtrate interface. Since the crystal structure of $LaAlO_3$ is cubic only at elevated temperatures, above 350–512°C, and becomes rhombohedral at room temperature, the structural transition during cooling is expected to induce a significant degree of twinning, which in turn would induce an appreciable number of dislocations. Thus it is reasonable to expect that the Tl-2223 grains grown on $LaAlO_3$ will be much more defective than those grown on MgO substrates. However, clarification of whether these microstructure effects are the sole origin of the differences observed is left for further detailed investigations.

As shown in the inset of Fig. 2, $J_c(T)$ in both cases displays linear behavior over a wide range of temperatures. As pointed out by Lemberger,[15] the linear zero field $J_c(T)$ and the 1% limit to the theoretical $J_c(0)$ ($\approx 10^9$ A/cm^2) usually obtained in YBCO films fabricated by various deposition methods, are indicative of the high density of pinning centers inherent to thin film processes. If this is true, the present results would suggest a high density of pinning centers in these single-crystalline thin films. This also explains why most Tl-2223 thin films evidently have J_c values already on the order of 10^6 A/cm^2 even at temperatures as high as 90 K, as shown here and previously.[1–3] This, however, is in sharp contrast with bulk single crystals, where the J_c vanished at temperatures around 40 K, as deduced from magnetization measurements.[6] The fact that the thin films, even in single-crystal form, can manifest themselves as a 2D superconductor on the one hand, and yet can sustain such high critical current densities comparable to those of more 3D-like YBCO films at temperatures very close to T_{c0} on the other hand, makes them of interest from both fundamental and practical points of view.

In order to examine how the pinning effects are related to the above paradox, preliminary studies on the macroscopic field-dependent critical current densities, $J_c(T, H)$, of these films were performed. Figure 3 shows the typical $J_c(H)$ behaviors obtained in these single-crystal bridges. It is noted that, although at temperatures very close to T_{c0} J_c's drop markedly with the application of small fields, the behaviors are different from those of typical granularity-induced weak links.[16] More importantly, as shown in the inset of Fig. 3, at low temperatures, J_c is greater than 10^6 A/cm^2 even at an applied field of 7. If we further plot the results in Fig. 3 within the context of the global pinning scheme,[17] scaling of the global pinning force density F_p as a function of the reduced field, $b = H/H_{c2}$, displays very different behavior for different temperature ranges. It is noted that, due to the difficulty of determining the upper critical field, H_{c2}, precisely, H_{c2} used here is defined by direct extrapolation of the $J_c(H)$ curve. As shown in Fig. 4, it is evident for temperatures near the K-T transition temperature, T_{KT} ($T_{KT} \approx 106$ K in this case),[8] that the scaling, though showing the similar bell-shape behavior exhibited by conventional superconductors, cannot be explained by any conventional pinning mechanism. Namely, the functional form of $F_p/F_p(\max) \approx c \times b^{0.3}(1-b)^5$ (dashed line in Fig. 4) and the peak position near $b = 0.1$ are not compatible with any known mechanisms based on either normal core condensation of pin-vortex interaction or magnetic interaction between vortices.[17] As indicated very recently by Pastoriza and Kes,[18] in the vicinity of vortex phase transition, the shear viscosity of vortices, rather than the pin-vortex interactions, becomes the dominant effect in determining vortex dynamics. This may explain the peculiar scaling described above.

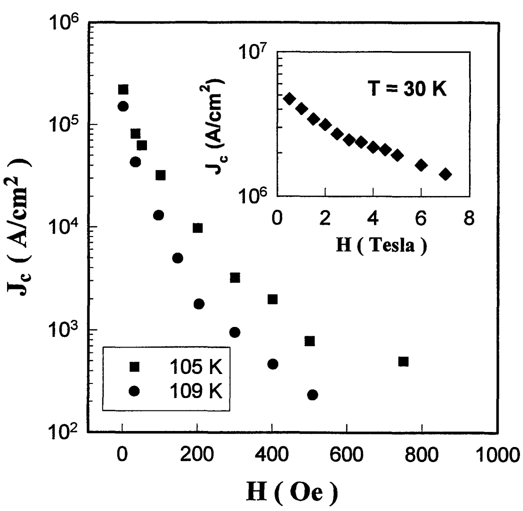

Fig. 3. The $J_c(H)$ results for Tl-2223/MgO(100) at three different temperatures. The temperatures were chosen to demonstrate the different behaviors manifested in the vicinity of T_c and very far from T_c, respectively.

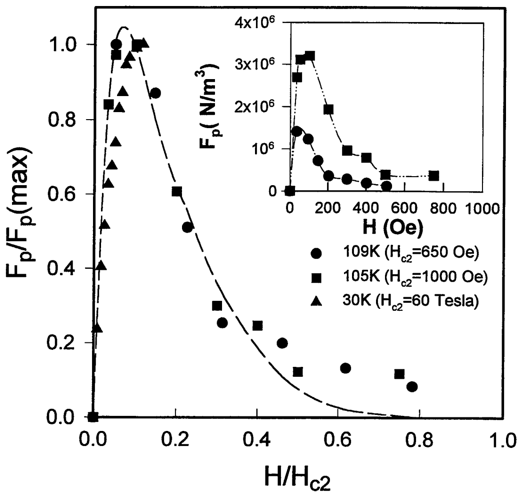

Fig. 4. The scaling behaviors of the pinning force density as a function of the reduced field. The upper critical fields indicated were determined by direct extrapolation of the $J_c(H)$ data.

On the other hand, for lower-temperature results, the scaling behavior is very similar to that obtained in high-critical-current Tl-2223 films reported previously[1] with the peak position of $F_p(b)$ located at around $b = 0.2$, and suggests surface core pinning by normal defects. It is noted that, due to the limitation of our magnetic field facility and a contact problem, the data presented here should only be considered as preliminary ones, and the exact functional form of $F_p(b)$ has not been determined due to the lack of high field data. However, as pointed out by Beasley,[19] the 2D nature of the high-T_c cuprates complicates the characteristics of the vortex and its lattice form significantly. Thus it is not surprising to observe vastly different pinning and associated vortex dynamics in different temperature ranges.

4. Conclusions

The critical current densities of single-grain Tl-2223 superconducting thin films are reported for the first time, to the best of our knowledge. It was found that, although these Tl-2223 grains were indeed two-dimensional in nature, the critical current densities are well above 10^6 A/cm^2 at a temperature of 90 K. The preliminary pinning studies revealed that, depending on the temperature range, the dominant limiting effects in determining the critical current densities can be very different. The results further imply that the substrate twinning-induced defects may have played some important role in determining the ultimate properties of this material.

Acknowledgements

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