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EFFECT OF SUBSTRATE-TWIN-INDUCED MICROSTRUCTURE ON TRANSPORT PROPERTIES OF EPITAXIAL $Tl_2Ba_2CaCu_2O_x$ FILMS IN A MAGNETIC FIELD

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Abstract--The role of substrate-induced microstructure on transport properties in *c*-axis oriented epitaxial $Tl_2Ba_2CaCu_2O_x$ films grown on $LaAlO_3$ and $SrTiO_3$ has been studied. For a magnetic field parallel to the Cu-O planes, resistivity and the critical current density, J_c, have been measured as a function of angle θ between the applied field and the direction of transport current. Resistivity dips and enhancement of the critical current density, J_c, were observed for magnetic fields applied parallel to the substrate twins in $LaAlO_3$ in high fields (≥ 2 T). Meanwhile for films on $SrTiO_3$, resistivity and J_c were Lorentz-force independent.

I. INTRODUCTION

The most widely used substrates in the epitaxial growth of thin films of the high-temperature superconductors(HTS) are $LaAlO_3$ and $SrTiO_3$. The crystal structure of $SrTiO_3$ is cubic, but that of $LaAlO_3$ is cubic only at elevated temperature, above 350° to 512°C. At room temperature, the primitive cell of $LaAlO_3$ is rhombohedral with a rhombohedral angle of 60.1° or face centered rhombohedral with a 90.08° angle. As temperature decreases, the La^{3+} ion shifts gradually along the body diagonal of the cubic unit cell resulting in a structural change to rhombohedral [1]. The twins result because the direction of the initial shift of the La^{3+} at various parts of the crystal can be different, although always along the cube body diagonals. For HTS film preparation, heating the substrate to the temperature range higher than 500°C is necessary for epitaxial growth in either in situ or post annealing process. Since structural changes occur in the cooling process, the films on $LaAlO_3$ will be subject to an additional strain. Thus microstructural changes in the film texture are possible during the cooling process. This change may result in a modification of the flux pinning properties which can be observed by measuring the transport properties. For films on $SrTiO_3$, similar large structural changes are absent.

The effect of the substrate twins on the transport properties of epitaxial $YBa_2Cu_3O_x$ films has been studied [2]. For a magnetic field (H) rotated within the Cu-O planes, resistivity dips were observed when the applied field was aligned with the substrate twins for samples on $LaAlO_3$. For thin films on $SrTiO_3$ in which substrate twins are absent, resistivity dips corresponding to the direction of twin boundaries in the *film* and, perhaps, interfacial dislocations were observed. It was suggested that such a resistivity drop for samples on $LaAlO_3$ may be due to flux pinning by a certain microstructural feature in the films induced by strain associated with the substrate twins. Contrary to the films on $SrTiO_3$, the twins were not observed in the *films* on $LaAlO_3$. This is probably because the formation of the substrate twins removes some strain in the film. However, any features related to the resistivity drops were not clearly identified. In this paper, transport properties of the epitaxial $Tl_2Ba_2CaCu_2O_x$ films grown on $LaAlO_3$ and $SrTiO_3$ have been studied as a continuation of the previous work. For a magnetic field rotated within the Cu-O planes, resistivity dips were observed when applied field is directed along the substrate twins for samples on $LaAlO_3$, but only for magnetic fields higher than 2 T. For films on $SrTiO_3$, no such resistivity dips were observed, and Lorentz-force independent resistivity and J_c were observed.

Precursors of $Tl_2Ba_2CaCu_2O_x$ films were prepared by laser ablation and multi-target dc magnetron sputtering. After annealing, these films exhibited a T_c of ~ 100 K and were confirmed by electron channeling to be epitaxial. All films were found to be highly *c*-axis oriented by x-ray diffraction. Films were patterned or scribed to 50 - 200 μm wide strips with voltage electrodes ~ 0.1 -0.3 cm apart. A typical thickness of the films is 0.8 μm. Among the results of several films studied, those of two on $LaAlO_3$ and one on $SrTiO_3$ will be described.

II. RESULTS

The microbridges were patterned perpendicular to the substrate twins in Sample A and ~ 45° in Sample B, both on

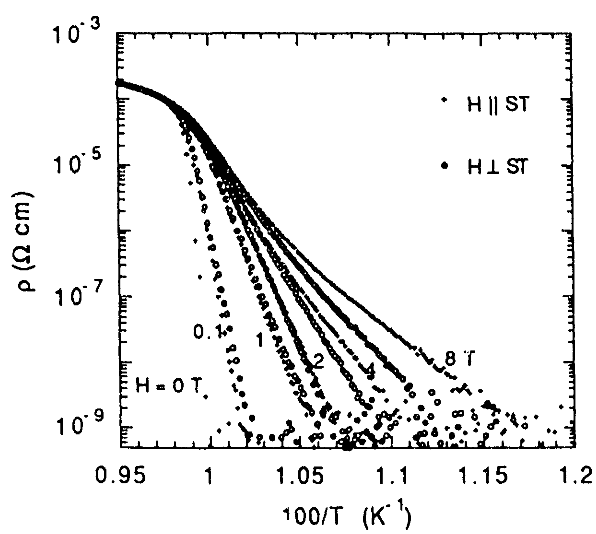

Fig. 1 The resistive transitions of epitaxial $Tl_2Ba_2CaCu_2O_x$ film (Sample A) grown on $LaAlO_3$ for magnetic field (H) parallel and perpendicular to the substrate twins (ST).

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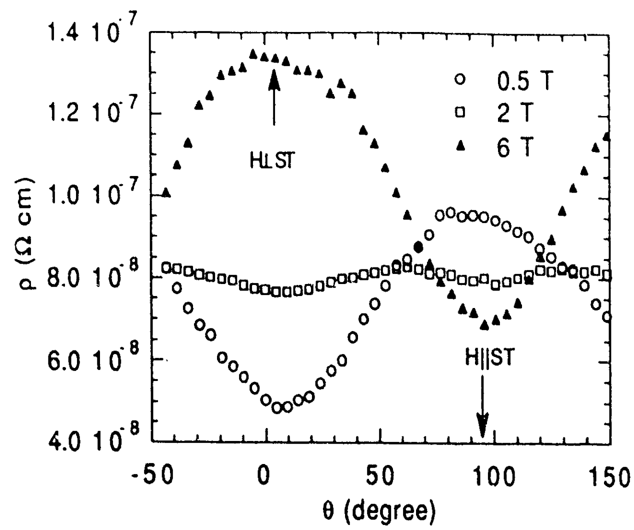

Fig. 2 Angular dependence of the resistivity of Sample A at various fields. The effect of the substrate twins are observed only at high fields

$LaAlO_3$. Figure 1 shows the resistive transitions of Sample A for magnetic fields parallel and perpendicular to the substrate twins (H||ST and H⊥ST). Note that Sample A has a single twin domain. The transport current always flows across the substrate twins in Sample A. Therefore, H⊥ST configuration corresponds to field parallel to the transport current (H||J), i.e., zero macroscopic Lorentz-force configuration, and H||ST corresponds to H⊥J, the maximum Lorentz force configuration. At low magnetic fields (≤ 1 T), dissipation is higher for H||ST, however at higher fields (> 2 T), dissipation for H⊥ST is higher. This crossover can be clearly seen in Fig. 2 where the angular dependence of the resistivity at fixed temperatures is shown. Temperatures are chosen in such a way that resistivities at different fields are almost equal. An angular position of 0° corresponds to the H⊥ST (H||J) configuration and 90° to the H||ST (H⊥J). At 0.5 T, the overall angular dependence follows a ~ $\sin^2\theta$ behavior, and the resistivity minimum is near H⊥ST (H||J). As field increases, angular dependence disappears around 2 T and eventually the resistivity minimum occurs at H||ST (H⊥J) at higher fields.

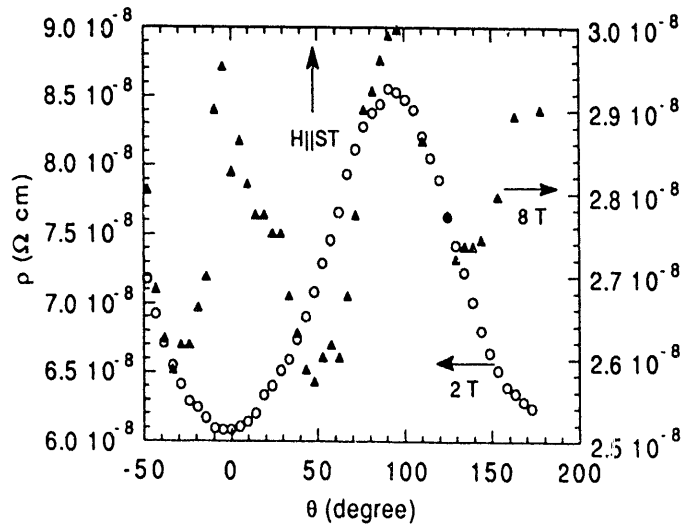

Fig. 3 Angular dependence of the resistivity of Sample B at various fields (circle; 2 T, triangle; 8T).

A similar angular dependence of the resistivity was observed in Sample B as shown in Fig. 3. At a 2 T field, the resistivity shows ~ $\sin^2\theta$ dependence with slight reductions at -40, 50, 140°, field parallel to the substrate twin orientations. Sample B has multi twin domains which are 90° apart. These resistivity dips become more apparent in the high field (8 T) data. However, the relative resistivity drops of Sample B for H||ST are smaller than those of Sample A probably due to the different sample microstructures. Note that the Lorentz-force dependence is absent in the 8 T data within the experimental error. For the sample on $SrTiO_3$, Sample C, only slight Lorentz-force dependence at low fields and no dependence at high fields (> 4T) was observed, similar to the results of Sample B except with substrate twin effects.

The critical current density, J_c, for the two orientations, H||J and H⊥J, for Samples A and C are shown in Fig. 4 as a function of magnetic field near 77 K. For Sample A, J_c are almost the same at low fields, but at high fields, J_c for H⊥J (H||ST) is much larger than those for H||J (H⊥ST). Sample B showed similar behavior with the highest J_c for H||ST for fields ≥ 3T, but the difference was rather small compared to

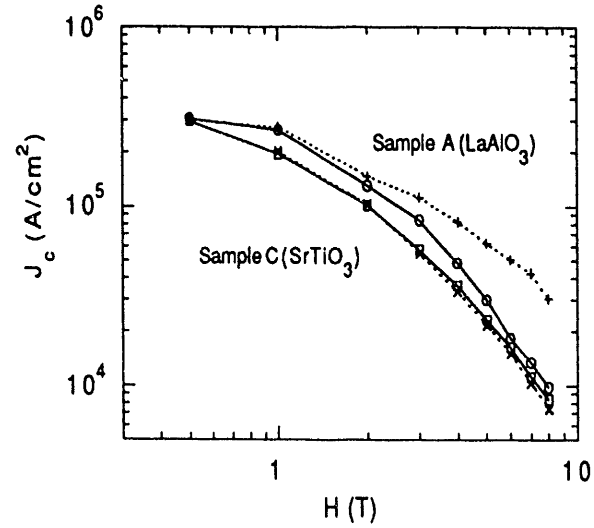

Fig. 4 The J_c vs H at ~ 77 K for Sample A (above) and Sample C (below). Solid lines are for H||J and dotted lines are for H⊥J.

that of Sample A, consistent with the resistivity results. For Sample C, J_c are almost the same over all field values showing no Lorentz-force dependence (Fig. 4). Similar Lorentz-force independence has been observed in the polycrystalline films although J_c is an order of magnitude smaller [3].

III. DISCUSSION

The resistivity drop and J_c enhancement for H||ST can be interpreted as a result of enhanced flux pinning effects due to the microstructural features in the films arising from the underlying substrate twins. Figure 5 is a sketch of the surface profile of a $LaAlO_3$ substrate measured by a thickness

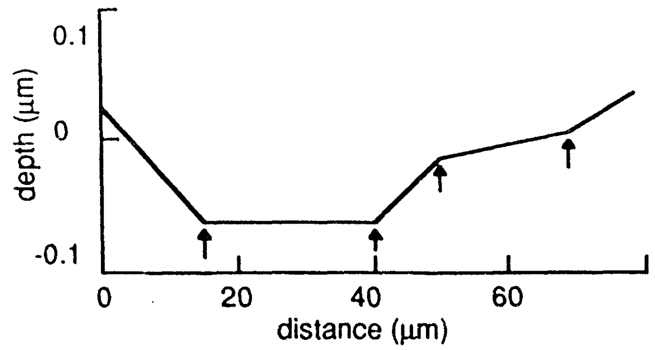

Fig. 5 A sketch of the surface profile of a twined $LaAlO_3$ substrate. The arrows indicate where the twins are located.

profilometer. A step-like structure is observed and it is always associated with the substrate twins which are located along the step edges. The angle of the step edge varies at different locations of the substrate, but it is found to be always smaller than 0.04°. It is interesting to note that this angle is roughly equal to the angular deviation of the face centered rhombohedral from the cubic structure.

When a film grows epitaxially, there can be an additional strain in the area directly on the substrate twins due to the step-like structure or different underlying lattice. In order to find any microstructural features associated with the substrate twins, TEM samples were prepared. Figure 6 shows a TEM micrograph of a substrate twin as well as the film on top of it. The twin structure is indicated by an arrow and is directed normal to the figure. There are indeed dislocations in the films directly on the top of the substrate twins. However, the substrate twins are not the sole origin of the dislocations as can be seen in Fig. 6; the neighboring dislocations are not associated with any visible substrate twins. Anyhow we conclude that the substrate twin is at least one of the origins of dislocations in the film structure. The different flux pinning strength between Samples A and B could be due to the

Fig. 6 A TEM micrograph of $Tl_2Ba_2CaCu_2O_x$ on $LaAlO_3$. The arrow indicates a twin in the substrate. Dislocations on top of the twin are visible.

different substrate twin properties, such as the average spacing, the angle of step edges, and whether the twins exist in single or multi domains.

Another interesting point is that the enhanced flux pinning due to the substrate twins is effective only at high fields (> 2 T). The angular dependence of the resistivity follows ideal ~ $\sin^2\theta$ dependence of flux flow at low fields. This implies that the region of dislocations are still good superconductors at low fields, but high magnetic field can weaken the superconductivity in this region to become effective pinning sites. This probable nonuniform suppression of the superconductivity in the dislocations at high fields can disturb the macroscopic current path. The experimental result that the resistivity for H||ST is smaller than that of the minimum macroscopic Lorentz force configuration (H||J) at high fields can be explained by evoking a meandering current path. However, it is important to note that the angular variation of the resistivity is very small compared to overall resistive broadening as can be seen in Fig. 1.

The absence of the Lorentz-force dependence in the resistivity and J_c has been consistently observed in all epitaxial films on $SrTiO_3$ and polycrystalline films [3]. The result of Samples A and B also indicates that the Lorentz-force dependent term is negligible compared to pinning effects especially at high magnetic fields even in the samples grown on $LaAlO_3$. A small Lorentz-force dependence of the flux motion resistance at low fields indicates that this material is not completely "magnetically transparent" for magnetic fields applied parallel to the Cu-O layers, contrary to the suggestion made by Kes et al. [4]. Disappearance of the Lorentz-force dependence at high fields as well as large residual broadening is still to be understood. There have been attempts [3, 5] to explain unusual the Lorentz-force independence, so far no definitive answers have been provided.

IV. SUMMARY

For $Tl_2Ba_2CaCu_2O_x$ on $LaAlO_3$, resistivity dips and enhancement of J_c were observed for fields parallel to the substrate twins only in high fields ≥ 2 T. TEM studies showed dislocations in the films on top of the substrate twins. These defects could act as pinning centers at high fields due to the suppression of the superconductivity in this area. Meanwhile for films on $SrTiO_3$, Lorentz-force independent resistivity and J_c were observed.

REFERENCES

[1] H. Fay and C.D. Brandle, in *Proceedings of an International Conference on Crystal Growth*, Boston, MA, June 1966, pp. 51-55.

[2] D.H. Kim, D.J. Miller, J.C. Smith, R.A. Holoboff, J.H. Kang, and J. Talvacchio, Phys. Rev. B44, 7607, 1991.

[3] D.H. Kim, K.E. Gray, R.T. Kampwirth and D.M. McKay, Phys. Rev. B42, 6249, 1990.

[4] P.H. Kes, J. Aarts, V.M. Vinokur and C.J. van der Beek, Phys. Rev. Lett. 64, 1063, 1990.

[5] S. Chakravarty, B.I. Ivlev, Y.N. Ovchinnikov, Phys. Rev. B42, 2143, 1990.

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