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Magnetic Flux Pinning in Epitaxial $YBa_2Cu_3O_{7-\delta}$ Thin Films

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The influence of microstructure on the critical current density of laser ablated $YBa_2Cu_3O_{7-\delta}$ thin films has been examined. Scanning tunneling microscopy was used to examine the morphologies of $YBa_2Cu_3O_{7-\delta}$ films and the morphology data were then correlated with measurements of the critical current density. The films were found to grow by an island nucleation and growth mechanism. The critical current densities of the films are similar to those of films with screw dislocation growth, indicating that screw dislocation growth is not necessary for good pinning. The data suggest that the critical current density in applied magnetic field may be higher in films with higher densities of growth features.

Key words: Dislocations, flux pinning, microstructure, nucleation mechanism

INTRODUCTION

Thin films of $YBa_2Cu_3O_{7-\delta}$ deposited from the vapor have transport critical current densities from 10^6 to 10^8 A/cm^2 in self-field at 77K and the critical current densities remain high in applied fields (>10^5 A/cm^2 in fields up to 6 T).[1–4] These critical current densities are several orders of magnitude higher than those found in single crystal and tape materials (~10^4 A/cm^2 in self-field at 77K).[5–8] However, the defects responsible for the superior magnetic flux pinning, and thus the high critical currents, of the thin films have not been identified. Due to the rapid rate and lower temperature at which the thin films are grown, it is likely that they contain greater numbers of atomic and line defects than single crystals which are slowly grown from the melt or than tape materials which receive numerous high temperature heat treatments. Such defects may be responsible for the enhanced pinning found in YBCO thin films.

Early scanning tunneling microscopy (STM) images of YBCO films deposited by sputtering revealed that the films grew by a spiral growth mechanism.[1,9,10] Shortly after this discovery, it was postulated that screw dislocations at the centers of these features were responsible for the enhanced flux pinning observed in these materials. A study of sputter deposited films with a range of screw dislocation densities showed a correlation between J_c and the screw dislocation density.[11] Films with higher densities of screw dislocation growth features had higher critical current densities in self-field and the critical current densities remained higher in applied field.

Subsequent imaging of films grown by laser ablation showed that YBCO films could also grow by island nucleation and growth rather than spiral

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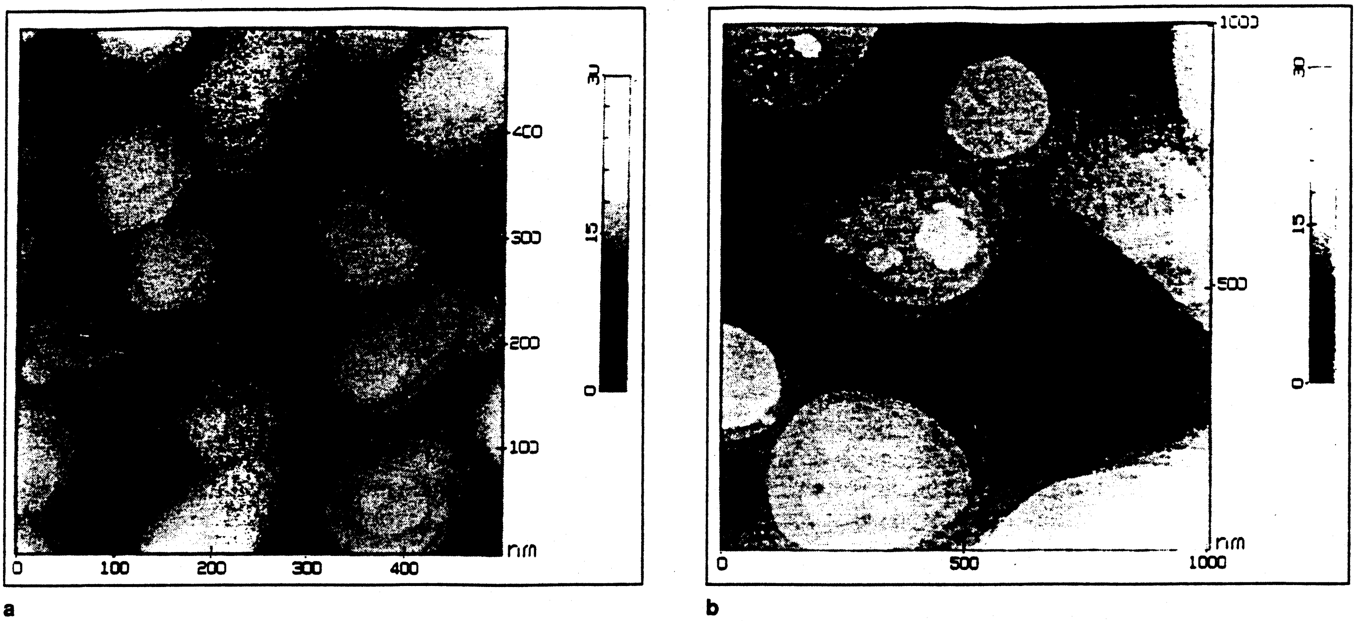

Fig. 1. Scanning tunneling micrographs of YBCO films laser ablated onto LAO at 60 nm/min at (a) 730, and (b) 850°C. Image (a) is of an area 0.5 × 0.5 μm²; image (b) is of an area 1.0 × 1.0 μm². The gray scale for both images corresponds to a z-range of 30 nm.

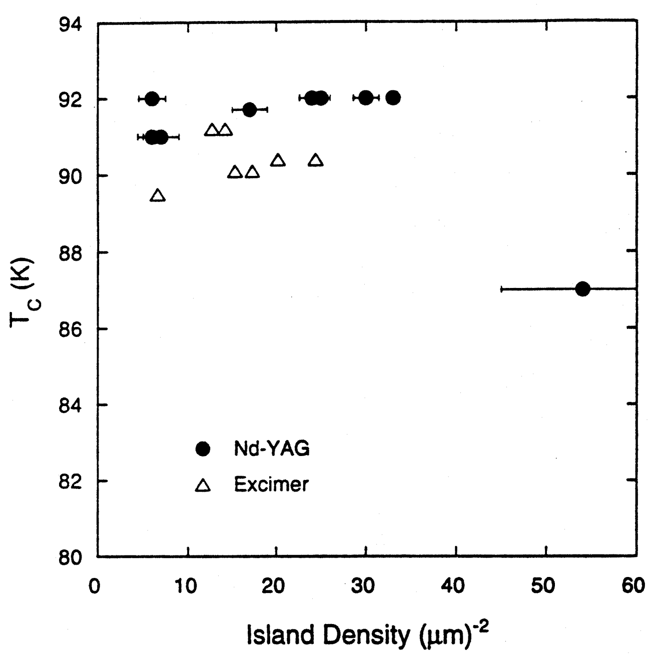

Fig. 2. Resistive transition temperatures of: ●YAG grown, Δ excimer grown YBCO films as a function of the density of growth features.

growth.[12] Films with island growth features have high critical current densities in self and applied fields similar to those of films with growth spirals.[13] Therefore, screw dislocations at the core of growth features are not necessary for good pinning in YBCO films.

EXPERIMENTAL

Thin epitaxial films of YBCO were laser ablated onto (001) LAO substrates. Both a frequency tripled Nd-YAG laser (355 nm) and a KrF excimer laser (248 nm) were used to deposit the films. The oxygen pressure during the depositions was held at 26.7 Pa (200 mTorr), the substrate temperature was varied from 730 to 875°C, and the deposition rate was 60 nm/min. Each laser was used to grow a series of films; for each series a single YBCO target was used. Films were grown to a nominal thickness of 200 nm.

The morphologies of the films were characterized by STM. The STM images were taken under ambient conditions with 80% Pt-20% Ir tips. Tunneling currents of 0.3 to 0.5 nA and bias voltages of 700 to 900 mV were used. The resistive transition temperature of each film, prior to patterning, was measured using a four probe ac technique. The transition temperature, T_c, was taken to be the temperature at which the resistance dropped below 1 mΩ. The critical current densities, J_c, were measured using both transport and magnetization techniques. For the transport measurements, films were patterned into strips 100 μm wide with a voltage tap spacing of 3 mm; an electric field criterion of 1 μV/cm was used. For the magnetic measurements 5 mm × 5 mm squares were used. The critical current was calculated using the Bean model corrected for the shape of the films. In both cases, the magnetic field was applied parallel to the c axis of the film and perpendicular to the measured current flow. Measurements were made at 76K.

RESULTS AND DISCUSSION

From the STM images, it was found that all of the films have an island nucleation and growth mechanism. As shown in Fig. 1, the density of the islands is lower in films grown at higher substrate temperatures. The temperature dependence of the island density is consistent with nucleation theory and will be described elsewhere.[14] The films used for this study had a range of island densities from 6 to 54 /μm².

The resistive transition temperatures of the films, as a function of the density of growth features, are given in Fig. 2. Data for films deposited with the YAG laser are shown with solid circles; data for films grown with the excimer laser are shown with open triangles. Except for the film with the highest density of growth features, all of the films have T_cs of 89K or higher.

The critical current densities of the films are shown in Fig. 3. The data shown by solid circles are for films deposited with the YAG laser and measured by transport at 76K in a field of 0.5 T. The data shown by open triangles are for films grown with the excimer laser and measured by magnetization at 76K in a field of 0.2 T. The error in measuring the transport J_c is dominated by uncertainty in the thickness of the film, which could be as large as ±5%. By comparison, the scatter in J_c for nominally identical films is much larger, as seen by the spread in J_c for the three films with island density near 5 /μm². We, therefore, estimate that the reproducibility of J_c may be only ±25% for films made under the same conditions but in separate depositions.

For both sets of samples, the data suggest an increase in J_c with increased island density. However, the increase is not large; for an order of magnitude increase in island density (from 6 to 55 /μm²), the J_c increases by only a factor of three. In addition, the significant scatter in J_c makes drawing conclusions difficult. Similar results were obtained for films with screw dislocation growth features, by Mannhart et al.[11] Their data also show a large degree of scatter, and the observed increase in J_c with the density of growth features is small.

Assuming that the increase in J_c is real, it is interesting to contemplate the source of the improved pinning. As described above, since films which grow by an island nucleation and growth mechanism have J_cs as high as films with screw dislocation growth, clearly the dislocations at the core of the growth spirals are not solely responsible for the good pinning in these material. The data presented here suggest that J_c increases as the square root of the density of islands. This could be a result of defects in the inter-island interface contributing to pinning. Or, since the variation in island density was achieved by changing the substrate temperature, the increase in J_c could reflect an increase in the total defect density throughout the film due to a lower deposition temperature.

There are several possible defects that may be associated with the inter-island regions and may contribute to pinning. Line defects such as edge and screw dislocations may be trapped in these regions to accommodate slight misorientations and displacements of the islands (or, in other films, growth spirals) as they grow together. Likewise a large number of point defects may be incorporated as the islands coalesce. In addition, the film surface between the islands is slightly depressed and this surface roughness may also contribute to pinning.

Twin boundaries are not included in this list because measurements on both films and single crystals with varying densities of twins have shown no correlation between twins and pinning.[15,16] Likewise, nanoscale precipitates of both Y_2O_3 and BaO have been observed in YBCO thin films.[17–19] However, no correlation has been found between the density of these precipitates and the film growth temperature.[18]

Distinguishing between the contributions of the various possible defects is difficult. High densities of dislocations in YBCO thin films on MgO substrates have been observed by transmission electron microscopy (TEM).[20,21] Cross-sectional imaging has shown that the dislocations "thread" from the film-substrate interface to the film surface.[20] The density of these defects, from 10^{10} to 10^{11}/cm² (10^2 to 10^3 /μm²), is at least an order of magnitude higher than the density of growth features typically found in YBCO films. Therefore, these dislocations are not all located at the cores of spiral growth features. In fact, a TEM study of very thin, <10 nm, YBCO films revealed that dislocations frequently occur in regions of island coalescence.[21]

These dislocations would be expected to occur in films which grow by either island nucleation or screw dislocation mechanisms and their densities are high enough to account for significant pinning.

Point defects may also be present in notable concentrations. While oxygen diffuses relatively rapidly in YBCO, $\sim 10^{-9}$ cm²/s at 700°C,[22] cation diffusion is orders of magnitude slower.[23] The slowest diffusing species is Y; the next slowest species is Ba. Extrapolating from data taken between 900 and 975°C, it is found that Ba has a diffusion coefficient at 700°C of $\sim 10^{-18}$ cm²/s.[23] This means that during a growth run lasting 3 min, a Ba ion, trapped in the "bulk" of the film, will diffuse approximately 0.1 nm. Therefore, any point defects trapped in the cation sublattice are likely to remain throughout the film growth and even after extended anneals.

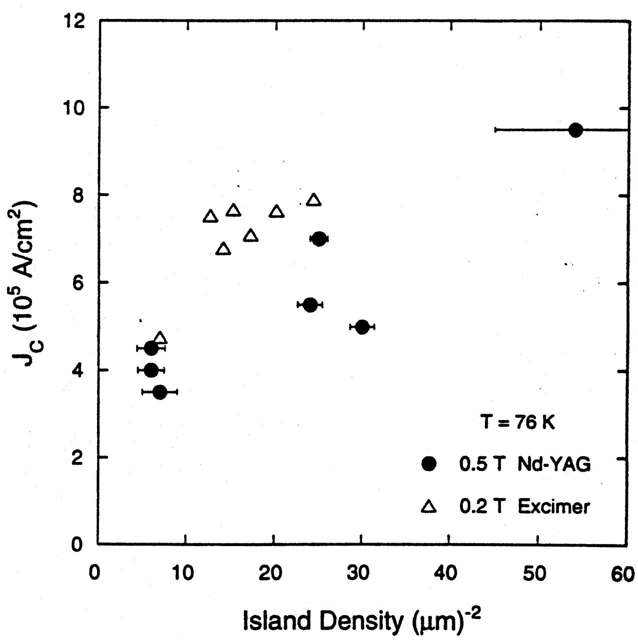

Fig. 3. Critical current densities as a function of the density of growth features for: ● films on LAO at 77K in a field of 0.5 T measured by transport, Δ films on LAO at 77K in a field of 0.2 T measured by magnetization.

SUMMARY

The critical current densities of laser ablated films with island nucleation and growth morphologies have been examined as a function of the density of growth features in the films. The critical current density in field was as high as that in films with screw dislocation growth mechanisms. The results suggest that J_c in field is higher in films with higher densities of growth features. However, the large degree of scatter in the data preclude stronger conclusions.

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