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TUNING POROSITY OF $YBa_2Cu_3O_{7-\delta}$ VICINAL FILMS BY INSERTION OF Y_2BaCuO_5 NANOPARTICLES (POSTPRINT)

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14. ABSTRACT

High critical current density J_c is the most critical specification for high-temperature-superconductor-coated conductors as required by numerous electric power-related applications. This has motivated an intensive research effort on the effects of microstructure on J_c. By growing $YBa_2Cu_3O_{7-\delta}$ YBCO films at a small vicinal angle, we have recently obtained a highly porous structure in these films accompanied with a significantly enhanced J_c. This result raises a challenging question on whether the porosity can be tailored in YBCO films to allow a higher J_c. In this study, we have explored the insertion of Y_2BaCuO_5 (211) nanoparticles in vicinal YBCO thick films to alter the strain at the nanometer scale; a nearly doubled pore density was obtained. A further improved J_c as the consequence of the enhanced pore density in these films suggests a direct correlation between microstructure and J_c and projects an even higher J_c in YBCO films with microstructure engineered optimally at a nanometer scale.

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Tuning porosity of $YBa_2Cu_3O_{7-\delta}$ vicinal films by insertion of Y_2BaCuO_5 nanoparticles

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High critical current density (J_c) is the most critical specification for high-temperature-superconductor-coated conductors as required by numerous electric power-related applications. This has motivated an intensive research effort on the effects of microstructure on J_c. By growing $YBa_2Cu_3O_{7-\delta}$ (YBCO) films at a small vicinal angle [R. L. S. Emergo, J. Z. Wu, T. Aytug, and D. K. Christen, Appl. Phys. Lett. **85**, 618 (2004)] we have recently obtained a highly porous structure in these films accompanied with a significantly enhanced J_c. This result raises a challenging question on whether the porosity can be tailored in YBCO films to allow a higher J_c. In this study, we have explored the insertion of Y_2BaCuO_5 (211) nanoparticles in vicinal YBCO thick films to alter the strain at the nanometer scale; a nearly doubled pore density was obtained. A further improved J_c as the consequence of the enhanced pore density in these films suggests a direct correlation between microstructure and J_c and projects an even higher J_c in YBCO films with microstructure engineered optimally at a nanometer scale. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2140467]

High temperature superconductor coated conductors (HTScc) has been the research focus of many groups recently because of the potential of these so-called second generation superconductor wires in electric power-related applications.[1–3] Among other specifications, the critical current (I_c) is the most critical one for most HTScc applications that include high-field magnets, electrical motors, generators and large-capacity power transmission lines.[4]

However, the J_c of YBCO films deposited on both single crystal and biaxially textured metal substrates experienced a monotonic decrease with increasing film thickness. This J_c-thickness behavior has motivated an extensive effort during the past few years to investigate the related mechanism.[5–12] In a recent experiment to probe the correlation between J_c and YBCO film thickness (0.2–3.0 μm range) with respect to microstructure, we have found higher J_c values at 77 K and SF in vicinal YBCO films of small miscut angle of 5°–10° as compared to their nonvicinal (to be regarded as "flat" in the rest of this paper) counterparts.[5] Furthermore, a slower decrease of J_c with film thickness was observed in the vicinal samples. One distinctive difference between the vicinal and flat YBCO films is in their microstructures. Flat samples are dense with an increasing volume fraction of misoriented (or non-*c*-axis oriented) grains at large thickness above ~0.5–1.0 μm. Vicinal films, on the other hand, are highly porous at thicknesses $\geqslant$0.2 μm while negligible misoriented grains were observed at thickness up to 3.0 μm. This leads one to speculate that the improvement of the J_c may correlate intimately with the porous microstructure of the vicinal YBCO films. In fact, pore surfaces may provide additional pinning on magnetic vortices and thus improve J_c. The downside of including pores in HTS films is the reduction of cross-sectional area for the current flow, which can result in reduced J_c. To minimize reduction of the current cross-sectional area so as to obtain a net J_c enhancement, the dimension of the pores need to be as small as possible, preferably on the order of a few times the coherence length ξ. For YBCO, ξ is anisotropic and typically in the range of 0.3–1 nm. Indeed, improved J_c values were observed in HTS bulks when the average dimension of the pores was reduced to sub-μm range.[13]

Although the porosity in the bulks can be altered using secondary phase doping and processing control, tuning porosity in *in situ* grown HTS films remains challenging, especially at the nanometer scale. If the pores are due to the strain developed when the a-b planes are tilted, therefore in order to reduce the dimension of the pores in the vicinal YBCO films, one must be able to locally control the strain. One way to achieve this is to insert nanoparticles (NPs) that deform the YBCO lattice within a short range comparable to the dimension of the NP. It is anticipated that by introducing this localized strain, the development of the large-scale strain can be impeded, leading to smaller and denser pores on the YBCO matrix. In this work, we have studied the effect of inserting Y_2BaCuO_5 (211) NPs on the microstructure of vicinal YBCO films using an *in situ* multilayer deposition scheme.[14,15] Hereafter, YBCO fabricated with 211 NPs will be called YBCO/211 films in contrast to the ordinary YBCO films. We report the experimental results in this letter.

The YBCO and YBCO/211 films were fabricated on flat and 10° miscut STO substrates using pulsed laser deposition (PLD). The processing conditions were optimized first on YBCO films and the same were applied for fabricating the YBCO and YBCO/211 films. Details of fabrication are discussed in our previous papers.[14,15] To avoid run-to-run variations, the YBCO/211 (or YBCO) films of the same thickness on the two types of substrates, flat and vicinal, were made in the same run.

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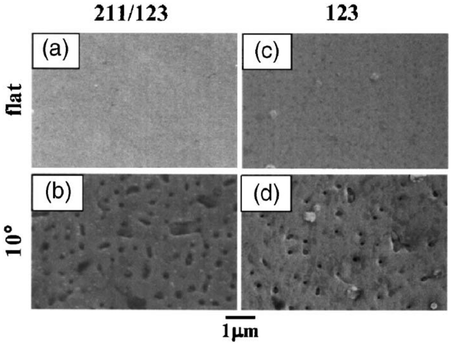

FIG. 1. SEM micrographs of 0.2 μm thick YBCO/211 and YBCO films. Left column: YBCO/211 films on (a) flat and (b) 10° miscut STO substrates. Right column: YBCO films on (c) flat and (d) 10° miscut STO substrates.

The surface morphology of the samples was analyzed using scanning electron microscopy (SEM) and the results are illustrated in Fig. 1 on four films: YBCO/211 on flat [Fig. 1(a)] and 10° miscut [Fig. 1(b)] STO, respectively; and YBCO on flat [Fig. 1(c)] and 10° miscut [Fig. 1(d)] STO, respectively. On the flat STO substrates, the YBCO films are dense and nearly featureless, particularly for the YBCO/211 film, suggesting that 211 NPs alone will not generate porosity for these deposition conditions. On the miscut STO substrates, on the other hand, both YBCO and YBCO/211 films are highly porous but differences can be observed. The majority of pores in both types of vicinal films have a circular shape with diameters ranging from 70 nm to 300 nm. With 211 NPs, some oblong-shaped and boomerang shaped pores are also observable [Fig. 1(b)], resulting possibly from two nearby pores coalescing. Interestingly, a higher pore density was observed on the YBCO/211 vicinal films as compared to that in the YBCO vicinal films. At 0.2 μm thickness, the pore density for the vicinal YBCO/211 film is approximately 2.48×10^8 pores/cm^2 while that for the vicinal YBCO film, 1.36×10^8 pores/cm^2. This means that the pore density was almost doubled when the 211 NPs were inserted in the vicinal YBCO films as expected from local perturbation of the strain in the vicinal films with NP insertion. This may explain the appearance of more noncircular shaped pores in the vicinal YBCO/211 films as a consequence of the circular pores coalescing more often at the higher density.

The evolution of the film microstructure with respect to film thickness differs in the cases of with and without 211 NPs. Figure 2 illustrates the SEM pictures of YBCO/211 and YBCO films with 2.0 μm thickness on flat (top row) and vicinal (bottom row) STO substrates, respectively. On flat YBCO films [Fig. 2(c)], many misoriented grains were visible as reported earlier.[5] With 211 NP insertion, the misoriented grains were replaced by a large number of microcracks [Fig. 2(a)], which may be due to the increase of the strain in the film with increasing thickness. None of the abovementioned features, however, were observed on the vicinal samples. The pores remained through the thickness while their shapes were highly deformed. In the vicinal YBCO/211 film, the pore size is much smaller at larger thickness, except for a few large pores. Similarly, the vicinal YBCO film without 211 NPs also has deformed pores but the pore size is bigger and the pore density is smaller than in the vicinal YBCO/211 film.

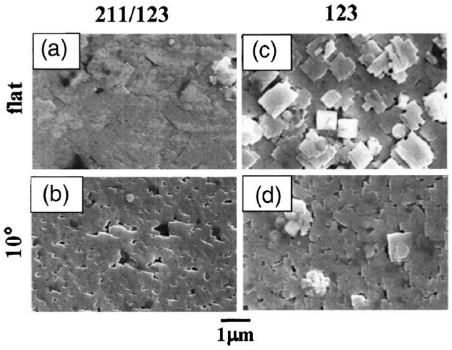

FIG. 2. SEM micrographs of 2.0 μm thick YBCO/211 and YBCO films. Left column: YBCO/211 films on (a) flat and (b) 10° miscut STO substrates. Right column: YBCO films on (c) flat and (d) 10° miscut STO substrates.

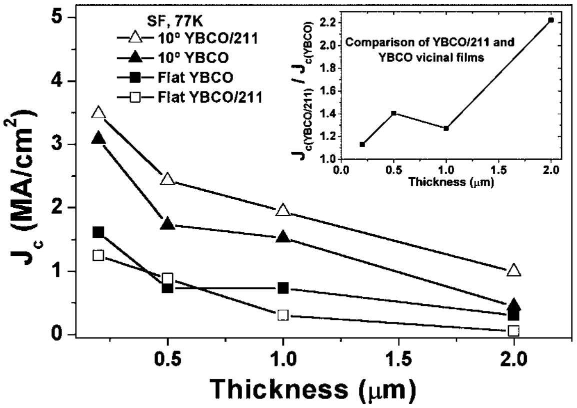

FIG. 3. J_c-thickness plots of YBCO and YBCO/211 films on flat and vicinal STO substrates. Inset shows the ratio of J_c of the YBCO/211 and YBCO vicinal films.

The film microstructure correlates closely with the J_c. Figure 3 compares the J_c-thickness curves measured magnetically at 77 K and SF on flat and vicinal YBCO/211 and YBCO films using a superconducting quantum interference device magnetometer. The Bean model was applied for calculation of the J_c.[16] As previously reported,[5] the vicinal YBCO films carry overall higher J_c's than the flat YBCO films throughout the thickness range of 0.2–2.0 μm. Interestingly, the two types of flat YBCO films, with or without 211 NPs, show similar J_c values and J_c-thickness dependence, suggesting that the incorporation of 211 NPs alone in YBCO films does not improve the J_c at SF (or low field $<0.5-1.0$ T, see discussion of Fig. 4) of YBCO films for the given YBCO/211 structure. In contrast, the vicinal YBCO/211 films have a consistently higher J_c than the vicinal YBCO films without 211 NPs. Since the former has a higher density of the pores, a plausible explanation for its higher J_c is the additional porosity induced by 211 NP insertion including a smaller pore dimension and higher pore density.

The benefit on the J_c by inserting 211 NPs in vicinal YBCO films can be more clearly seen in the inset of Fig. 3. The ratio of the YBCO/211 vicinal film J_c and the YBCO vicinal film J_c increases with the film thickness up to 2.0 μm, suggesting that the insertion of 211 NPs in the vicinal YBCO films further improved the J_c by engineering the porous microstructure in a more favorable fashion.

The in-field J_c is depicted as a function of magnetic field (H) in Fig. 4 for 0.2 μm [Fig. 4(a)] and 2.0 μm [Fig. 4(b)]
2 thick YBCO/211 and YBCO films on flat and vicinal substrates. At the smaller thickness, the J_c-H curves for the vicinal films cross over with those for the flat films in the H

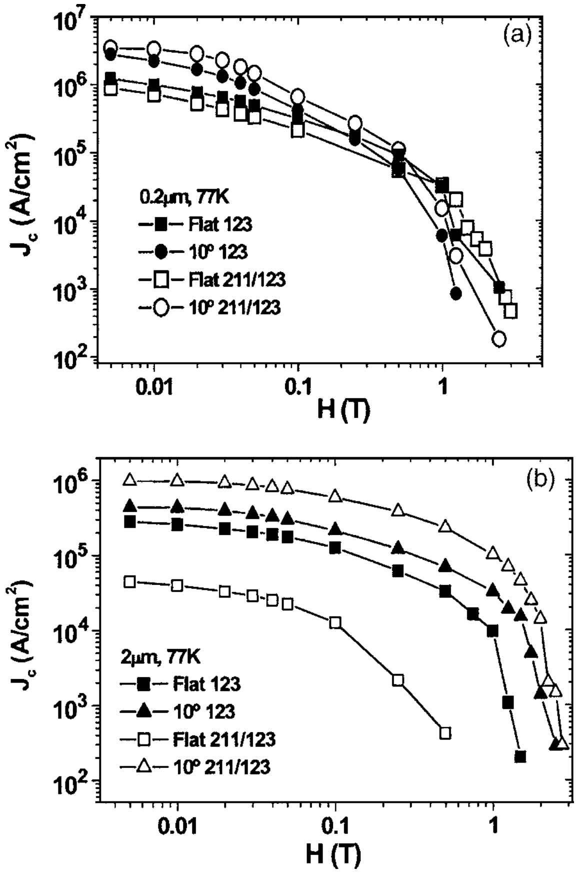

FIG. 4. (a): J_c-H plots of 0.2 μm thick YBCO and YBCO/211 flat and vicinal films and (b) J_c-H plots of 2.0 μm thick YBCO and YBCO/211 flat and vicinal films.

range of 0.5–1.0 Tesla. Below the crossover, the vicinal samples have significantly higher J_cs. Since the flat YBCO/211 film has lower J_c in this regime, it is unlikely that the 211 NPs alone are responsible for the improved J_c in the vicinal YBCO/211 film for this regime. Instead, we argue that the greater porosity is the more probable reason for the improvements to J_c for fields lower than ~0.5–1.0 T based on the fact that the J_c of the vicinal YBCO/211 is even higher than that of the vicinal YBCO sample. Above ~1.0 T, the nanoparticulate additions appear to be the primary cause of the J_c improvement. The occurrence of the crossover indicates that the pores at the current dimension and density benefit pinning of magnetic vortices mostly in the lower field range below ~1.0 Tesla. It may be possible for the crossover point to be pushed to higher H field by reducing the pore dimension and increasing pore density. In fact, no crossover was observed at the larger thickness of 2.0 μm [Fig. 4(b)], which is not surprising considering the reduced pore dimension with increasing film thickness. The vicinal films have overall higher J_c values in the whole H range. Recall that negligible misoriented grains were formed in the vicinal samples, in a sharp contrast to the large fraction of misoriented grains on the flat YBCO films at large thickness, which obstruct J_c. For the vicinal films, the one with 211 NPs (open triangles) has approximately a factor of 2 higher J_c in most H range as compared to the one without (solid triangles). We attribute this enhanced J_c to the combined benefit of more favorably engineered porous structure and 211 NP pinning, although other mechanisms, such as anisotropy of J_c, should not be excluded. Interestingly, the flat YBCO/211 sample (open squares) of 2.0 μm thickness has a significantly lower J_c, even compared to that of the flat YBCO sample (solid squares) and we suspect that the observed microcracks in the former further reduced its J_c. The fact that no misoriented grains and microcracks appeared in the vicinal films with or without 211 NPs suggests that the formation of pores may release the strain accumulated with increasing thickness in a favorable manner. In another word, the combination of pores and NPs may provide a promising scheme for achieving high J_c in thick YBCO films via microstructure engineering.

In summary, we have found that the porosity of vicinal YBCO films can be tuned using 211 NPs insertion in a multilayer growth process. By inserting 1 nm layer of 211 NPs after the growth of each 9 nm thick YBCO layer on 10° miscut STO substrates, the pore density can be nearly doubled and the pore dimension reduced. The investigation of J_c in these porous YBCO/211 films of thickness ranging from 0.2 to 2.0 μm revealed an overall enhanced J_c as compared to reported results, suggesting a correlation between J_c and microstructure of the film. The result suggests that the combination of pores and NPs may provide a promising scheme for achieving high J_c in thick YBCO films and coated conductors via microstructure engineering.

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