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# Improvement of critical current density and thermally assisted individual vortex depinning in pulsed-laser-deposited $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films on $\text{SrTiO}_3$ (100) substrate with surface modification by Ag nanodots

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$\text{YBa}_2\text{Cu}_3\text{O}_7$  films were fabricated by pulsed laser deposition on  $\text{SrTiO}_3$  (100) single-crystal substrates whose surfaces were modified by the introduction of Ag nanodots. The critical current density ( $J_c$ ) was found to increase with the number of Ag shots. Zero-field magnetic  $J_{c0}$  at 77 K increased from  $8 \times 10^5$  up to  $3.5 \times 10^6$  A/cm<sup>2</sup> as the number of Ag shots increased from 0 to over 150 times. Microstructure investigations indicated that the crystallinity and the *ab* alignment gradually improved as the number of Ag nanodots increased. Thermally activated depinning of individual vortices is suggested responsible for a field-independent  $J_c$  plateau. © 2005 American Institute of Physics. [DOI: 10.1063/1.1851877]

## INTRODUCTION

Due to numerous potential applications for superconducting tapes based on  $\text{YBa}_2\text{Cu}_3\text{O}_y$  (Y123) film, extensive studies are currently being carried out worldwide on Y123 films grown on different single-crystal or metal-based substrates with or without buffer layers.

In addition to neutron- or heavy-ion irradiation,<sup>1</sup> chemical incorporation of nanosize oxide particles,<sup>2,3</sup> or metal particles<sup>4,5</sup> has proven to be an alternative economic approach to enhancing critical current density in either low or high magnetic fields for Y123. Y123 films with nano-Ag particle inclusions exhibited higher  $J_c$  values compared to pure Y123 films. It has been reported that, by creating an array of Ag or  $\text{CeO}_2$  (Ref. 6) nanodots *in situ* on the substrates prior to the deposition of superconducting films, the  $J_c$  was greatly enhanced for  $(\text{Tl,Cu})\text{BaSr}_2\text{Ca}_2\text{Cu}_3\text{O}_y$  and Y123 thin films.<sup>4,5</sup> It is believed that the nanodots can introduce extended defects, resulting in strong pinning centers. However, it is still not clear how the metallic Ag nanodots play a role in the improvement of  $J_c$  in Y123 films with nanoinclusions. In this paper, we introduced the Ag nanodots onto the surface of  $\text{SrTiO}_3$  single-crystal substrates, or the substrate surfaces were modified by Ag nanodots, prior to the deposition of Y123 film. The effect of the amount of nano-Ag dots on the film performance is systematically investigated. It was found that, under fixed physical deposition conditions such as oxygen pressure, substrate temperature, etc., the  $J_c$  of Y123 deposited on such surface-modified substrates is greatly enhanced with much improved *c*-axis align-

ment in comparison to the films grown on substrates without Ag nanodot modification.

## EXPERIMENT

The fabrication of nano-Ag dots and Y123 thin films used in this study was carried out using a standard pulsed laser deposition (PLD) system, comprising a deposition chamber with a base pressure of  $10^{-7}$  Torr, fitted with a resistive sample heater, and a six-target manipulator. A KrF excimer laser, with a wavelength of 248 nm, was used as the ablating power source. The laser beam was focused onto the rotating target by a fixed-beam optical train.

A thin layer of Ag nanodots was PLD deposited on (100)  $\text{SrTiO}_3$  (STO) substrate,  $3 \times 3$  mm<sup>2</sup> by ablating a pure Ag target prior to the deposition of Y123 films. Four samples of Y123 films (with 0, 15, 60, and 150 Ag shots) used in this study were grown on STO substrates whose surfaces were modified by different amounts of Ag nanodots that were controlled by the number of shots ablated from the Ag target. The Y123 film deposition was carried out at 780 °C, at a pressure of 400 mTorr of high-purity oxygen, using a laser repetition rate of 6 Hz, and a laser energy of 300 mJ/pulse. After the deposition, the Y123 film was annealed for 1 h at 550 °C at a pressure of 760 Torr of oxygen, followed by a slow cooling to 400 °C in 30 min, and then free cooled to room temperature. The Y123 film produced was approximately 400 nm thick. The surface topographies of the Ag dots on the substrates and the Y123 films were investigated by atomic force microscopy (AFM) and scanning electron microscopy (SEM). Y123 phase formation was assessed by x-ray diffraction (XRD). The superconducting transition temperature,  $T_c$ , was measured by ac susceptibility, and the critical current density,  $J_c$  was estimated from dc magnetization

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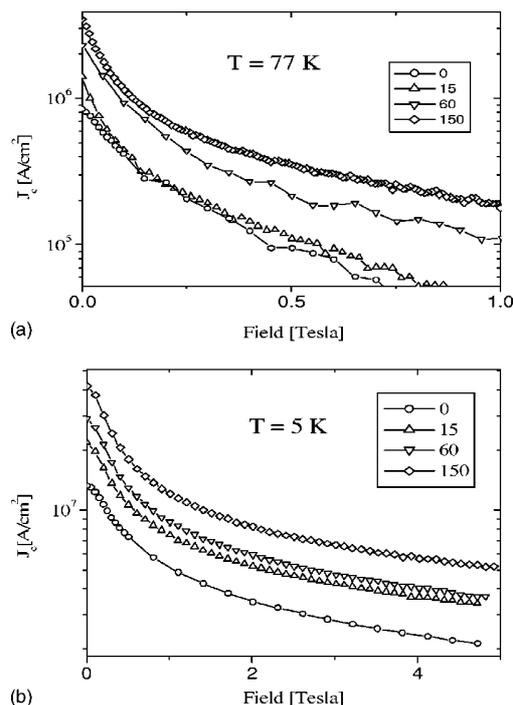


FIG. 1.  $J_c$  field dependence at 77 K (a) and 5 K (b) for films with different numbers of Ag shots.

hysteresis loops measured using a commercial magnetic property measurement system (MPMS) and vibrating-sample magnetometry (VSM) at 4.2, 5, 10, 20, 30, 40, 50, 60, 70, and 77 K in external dc fields of up to 1.75 T or 5 T parallel to the  $c$  axis of the films.

## RESULTS AND DISCUSSION

The study of the shape and distribution of the Ag nanodots investigated by AFM showed that regardless of the number of Ag shots, Ag formed as nanodots on the substrate surfaces. The average density of these dots is approximately  $2\text{--}10/\mu\text{m}^2$ , which increases as the number of Ag shots increases. The height of the Ag dots is between 0.2 and 2 nm with diameters of 10–20 nm.

The  $J_c$  was calculated from magnetic hysteresis loops measured at different temperatures using the Bean model,  $J_c = 20 \Delta M / [a(1 - a/3b)]$ , with  $a < b$ , where  $a$  and  $b$  are the sample dimensions. The field dependence of  $J_c$  at 5 and 77 K is shown in Fig. 1. It can be clearly seen that the  $J_c$  increases as the Ag shots increase from 0 to 150, both at low and high fields. At 77 K and zero field, the  $J_c$  increased from  $8 \times 10^5$  to  $3.5 \times 10^6$  A/cm<sup>2</sup> and increased almost one order of magnitude higher at 1 T. At 5 K, the  $J_c$  increased from  $1.2 \times 10^7$  to  $4.2 \times 10^7$  A/cm<sup>2</sup>. The factors of  $J_c$  enhancement are almost the same for the low and high fields. Figure 2 shows the zero field  $J_c$  at 77 K as a function of the numbers of silver shots. This result clearly showed that the Ag nanodots greatly improve the  $J_c$  values compared to the films grown on the STO substrate without Ag nanodot modification.

It should be emphasized that all the Y123 films were laser deposited under the same ablation conditions, such as the same oxygen pressure, substrate temperature, laser en-

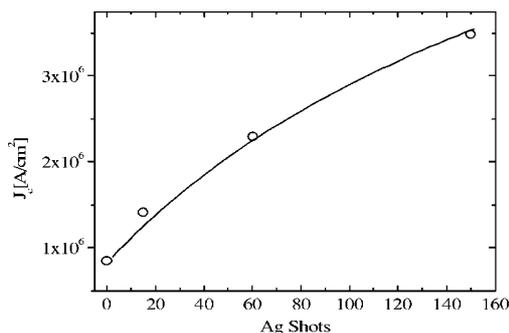


FIG. 2. Zero field  $J_c$  at 77 K vs number of Ag shots. The solid line is only a guide to the eyes.

ergy, etc. The only difference is the amount of Ag dots deposited on the substrates prior to the deposition of Y123 films. It was worth noting that the  $J_c$  values at 77 and 5 K for the reference sample (with zero Ag shots) are lower compared to the pure Y123 films fabricated under optimized PLD conditions.<sup>7</sup> This implies that the ablation conditions used for our samples are not optimum. The results of the  $J_c$  enhancement by the Ag nanodots for our samples indicated that surface modification with Ag nanodots is another factor controlling the performance of the Y123 films in addition to other physical deposition conditions.

To clarify why the Ag nanodots play a role in the  $J_c$  enhancement, we checked for any differences in the surface morphologies of our films. We found that for 0, 30, and 60 Ag shots, the films' surface morphologies were indistinguishable under SEM and AFM. The surface morphologies for a film with 150 Ag shots look similar to all the others; however, this sample exhibited clear growth islands and nano-holes in the trenches between the growth islands. Typical AFM surface images of two Y123 films grown on STO with 30 and 150 Ag shots are shown in Fig. 3. Characteristic of

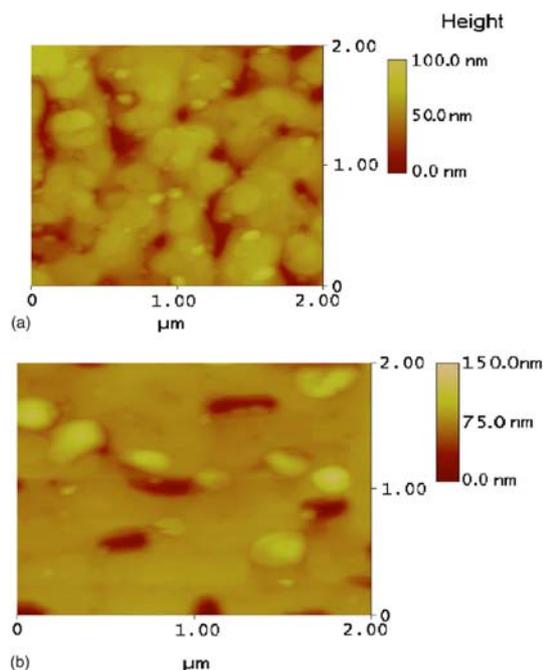


FIG. 3. AFM images for films with 150 (a) and 30 (b) Ag shots.

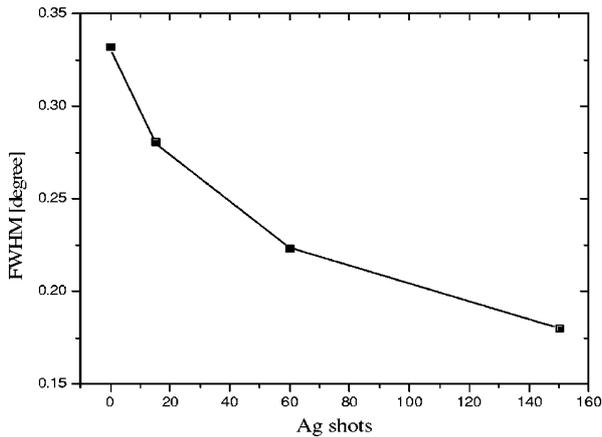


FIG. 4. Full width at half maximum for (005) peak of films with different Ag shots.

AFM for all films is the island structure, with an island diameter of typically 100–500 nm and the islands separated trenches consisting of nanosize holes, which can be clearly visualized under high magnification SEM as well. For 150 shots, the AFM showed a higher density of growth islands with smaller diameters than all the other films. The higher density of growth islands implies that the 150 Ag shot films should have extra linear defects acting as pinning centers for flux pinning compared with other samples, as it has been reported that each growth island provides one linear defect in PLD Y123 film on STO substrates.<sup>7,8</sup> However, as the  $J_c$  increases monotonically with Ag shots in our samples, it seems that the surface morphology has no direct relationship to the  $J_c$  enhancement. Therefore, we have to look at the phases, orientations, and crystallinity of our films.

All the XRD results revealed that the out-of-plane orientation of all the Y123 films is strong, with only the (001) reflections being present. Therefore, it seems that there is no difference in the orientation of the samples. However, if we check the full width at half maximum (FWHM) for the (005) reflection peak for different samples, it is found that the values of the FWHM decrease as the Ag shots increase, i.e., the (005) peak becomes sharper with more Ag shots, indicating improvement of crystallinity in the films or improvement of the (001) orientation of crystal grains (Fig. 4). Therefore, the increase in  $J_c$  with Ag nanoshots is believed to be due to the

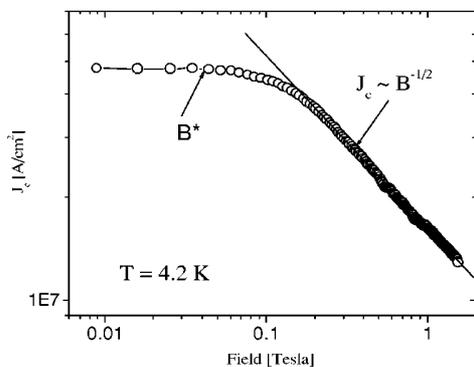


FIG. 5. Log  $J_c$  vs Log  $B$  at 4.2 K for film with 150 Ag shots. The solid line is a linear fitting.

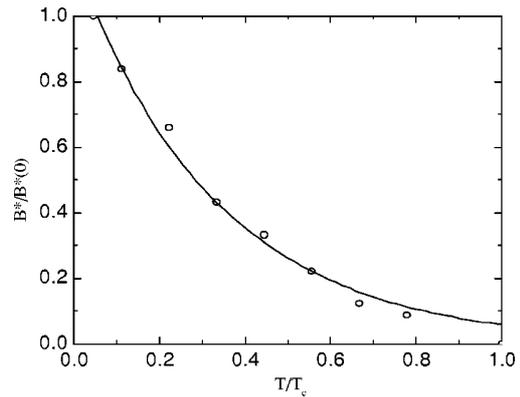


FIG. 6. Normalized characteristic field  $B^*$  (shown in Fig. 5) as a function of reduced temperature for film with 150-Ag shots. The solid line is an exponential fitting.

improvement of the out-of-plane alignment caused by Ag nanodot modification of substrate surfaces. The enhancement of the  $c$  alignment and in turn the enhancement of  $J_c$  observed in our Y123 thin films grown by PLD on STO substrates whose surfaces were modified by nano-Ag dots are in agreement with what has been reported for Ag nanodots including Y123 thin films made from a Y123 target mixed with Ag particles.<sup>4</sup> It might be possible that the Ag nanodots have a surfactant effect on the growth of the Y123 films. The surfactant effect of Ag could enhance the chemical activity between Y123 and substrate and therefore enhance the  $c$ -alignment growth. A further study on the surfactant effect of Ag on the Y123 film growth on various substrates would be very useful and interesting.

For all the films with different Ag shots,  $J_c$  exhibited a plateau for magnetic fields below a characterization field  $B < B^*$ . Above  $B^*$ , the  $J_c$  scales as  $B^\alpha$  (with  $\alpha = -0.5$  at 4.2 K) indicating plastic pinning where the vortices are pinned via shear interaction with strongly pinned vortex lattice (Fig. 5).<sup>7,8</sup> It has been reported that the value of  $J_c$  at  $B < B^*$  is solely determined by the pinning of single vortices along extended defects in the Y123 films grown on STO by PLD. We plotted  $B^*$  for films with 150 nano shots as a function of reduced temperature  $t(T/T_c)$ . As shown in Fig. 6, it was found that  $B^*$  vs  $T/T_c$  can be fitted by  $B^*(t) = B^*(0)\exp(-qt)$  where  $q = 2.5$ . The exponential  $B^*(t)$  dependence of our samples implies that the position of  $B^*$  could arise from the onset of thermally activated depinning of individually pinned vortices as expected due to the linear defects that are commonly present in PLD Y123 thin films.<sup>7-9</sup>

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