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Effect of substrate surface modification using Ag nano-dots on the improvement of J_c and microstructures in $YBa_2Cu_3O_7$ thin films grown on $LaAlO_3$ (100) by pulsed laser deposition

A. H. Li · X. L. Wang · H. K. Liu · M. Ionescu · S. X. Dou · P. Xuan · E. W. Collings

Abstract $YBa_2Cu_3O_7$ (Y123) thin films were grown by pulsed laser deposition (PLD) on $LaAlO_3$ (100) substrates whose surfaces were modified by a discontinuously layer of Ag nano-dots. The Y123 films were characterised by atomic force microscope, X-ray diffraction, scanning electron microscope, and DC magnetization measurements. Effect of substrate surface modification using various densities of the Ag nano-dots on the improvements of critical current density J_c and microstructures in the Y123 films has been studied systematically. The results showed that at fixed physical deposition conditions J_c increased with the number of Ag shots, n. Zero field J_c at 77 K increased from 10^6 to 3.3×10^6 A/cm^2, and from 1.5×10^7up to 4×10^7A/cm^2 for 5 K as the number of Ag shots increased from zero to 150. However, a fluctuation of J_c was observed for $n < 60$ at 77 and 40 K in both low and high fields. Detailed microstructure analysis revealed that *ab* misalignment was gradually improved as Ag nano-dots density gradually increased and believed to be responsible for the J_c enhancement.

Keywords YBCO · Thin films · Substrate · Microstructure · Nano-dots · Flux pinning · Critical current density

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1 Introduction

It is well known that $YBa_2Cu_3O_7$ (Y123) thin films and Y123 coated conductors have great applications in both superconducting microelectronics and large scale high current carrying conductors. However, high critical current density J_c at both low and high fields is required for the Y123 grown on different substrates. In order to improve J_c, effective pinning centers, such as random defects, small tilt grain boundaries, dislocations, twin boundaries, and oxygen vacancies have been extensively studied. Incorporation of nano-size oxide particles [1, 2] or metal particles [3, 4] has proved to be most economic approach to enhance critical current density J_c 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 CeO_2 [5] nano-dots *in-situ* on the substrates prior to the deposition of superconducting films, the J_c was greatly enhanced for (Tl,Cu)$BaSrCa_2Cu_3O_y$ (TBCO) and Y123 thin films [3, 4]. The nano-dots could introduce extended defects into the TBCO and Y123 films resulting in strong pinning centers. However, it is still not clear how the metallic Ag nano-dots play a role in the improvement of J_c in Y123 films if the Ag nano-dots were deposited on substrates prior to the deposition of Y123 thin films.

In our recent work, we have found that the J_c of Y123 films fabricated by pulsed laser deposition on $SrTiO_3$ (100) single crystal substrates whose surfaces were modified by introduction of Ag nano-dots increased with the number of Ag shots [6]. Self-field J_c at 77 K increased from 8×10^5up to 3.5×10^6 A/cm^2 as the number of Ag shots increased from 0 to over 150 times. The nano-Ag dots also have positive effect in enhancing the J_c for films grown on Y-ZrO2 (YSZ) single crystal substrates [7, 8]. Microstructure investigations

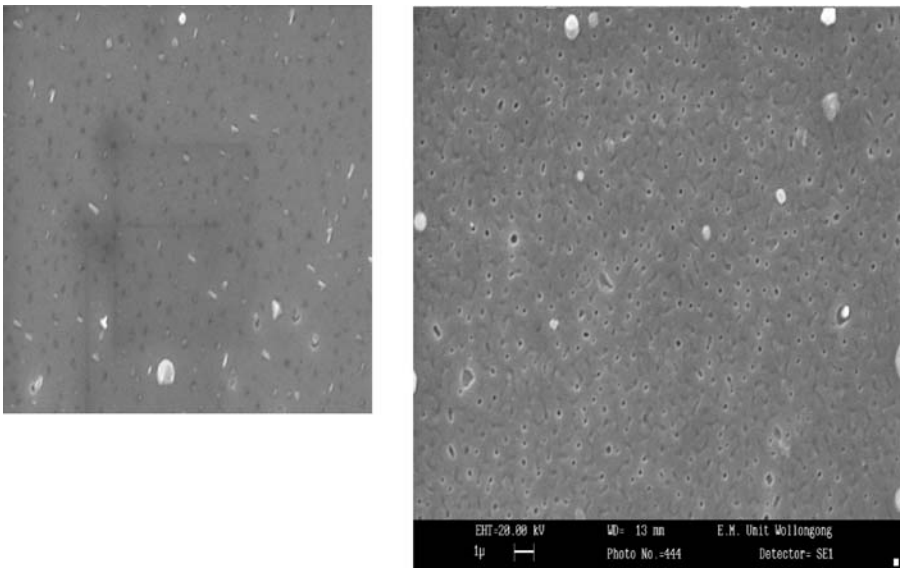

Fig. 1 SEM images for films with 30 (left) and 150 (right) Ag shots

indicated that the crystallinity and the *ab* alignment gradually improved as the number of Ag nano-dots increased for both STO and YSZ substrates. In this paper, we report the effect of the amount of nano-Ag dots on the J_c performance of Y123 grown on $LaAlO_3$ (100) substrates. We found that, under fixed physical deposition conditions, such as oxygen pressure, substrate temperature, etc., the J_c of Y123 films deposited on such surface modified substrates is greatly enhanced with much improved *c*-axis alignment in comparison to the films grown on substrates without Ag nano-dot modification. The underlying mechanisms relating to the improvement of microstructures under the influence of Ag-nano-dots are presented.

2 Experimental

YBCO thin films were deposited by means of pulsed laser dopsition (PLD) suing a Kr-F excimer laser (wave length is 248 nm). Single crystal $LaAlO_3$ (100) was used as substrate. The PLD was carried out under the following deposition conditions. The substrate temperature was between 720–800°C, the oxygen partial pressure was held at 400 mtorr, and the substrate-target distance was 50 cm. The thickness of the Y123 films were about 400 nm. The phase purity and orientations of YBCO films were checked by X-ray diffraction (XRD).

A thin layer of Ag nano-dots was PLD deposited on the LAO substrate, 3×3 mm^2 by ablating a pure Ag target prior to the deposition of Y123 films. Four samples of Y123 films used in this study were grown on LAO substrates whose surfaces were modified by different amounts of Ag nano-dots that were controlled by the number of shots ablated from the Ag target (the number of Ag shots, $n = 0$, 15, 30, 60, 130, and 150).

The surface morphologies of the Y123 films were investigated by atomic force microscope (AFM) and scanning electron microscopy (SEM). The critical current density, J_c for all films was estimated from DC magnetisation hysteresis loop measured over a wide temperature range from 5, 40, and 77 K in external DC fields of up to 5 T parallel to the c-axis of the films using commercial MPMS.

3 Results and discussion

We found that for 0, 15, 30, and 60 Ag shots, the films surface morphologies were indistinguishable under SEM and AFM. Typical SEM images for films with $n = 30$ and 150 are shown in Fig.1. The surface morphologies for these two images look similar. However, films with $n = 150$ samples exhibited a clear feature of nano-holes (Fig.1 (right)). Typical corresponding AFM surface images of these two Y123 films with 30 and 150 Ag shots are shown in Fig. 2. Characteristic of AFM for both films is the island structure, with an island diameter of typically 100–500 nm, separated by deep trenches consisting of nano-size 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 and nano-holes than all the other films.

The J_c was calculated from magnetic hysteresis loops measured at different temperatures using, $J_c = 20\ \Delta M/[a(1-a/3b)]$, where a and b are the sample dimensions, with a < b. The field dependence of J_c at 5K is shown in Fig. 3. It can be seen that the J_c at zero field increases as the Ag shots increase from 0 to 150. However, J_c in the field does not appear to increase monotonically with the number of Ag shots under magnetic fields as that observed at zero field. The J_c vs the number of Ag dots for 5, 40 and 77 K at both zero

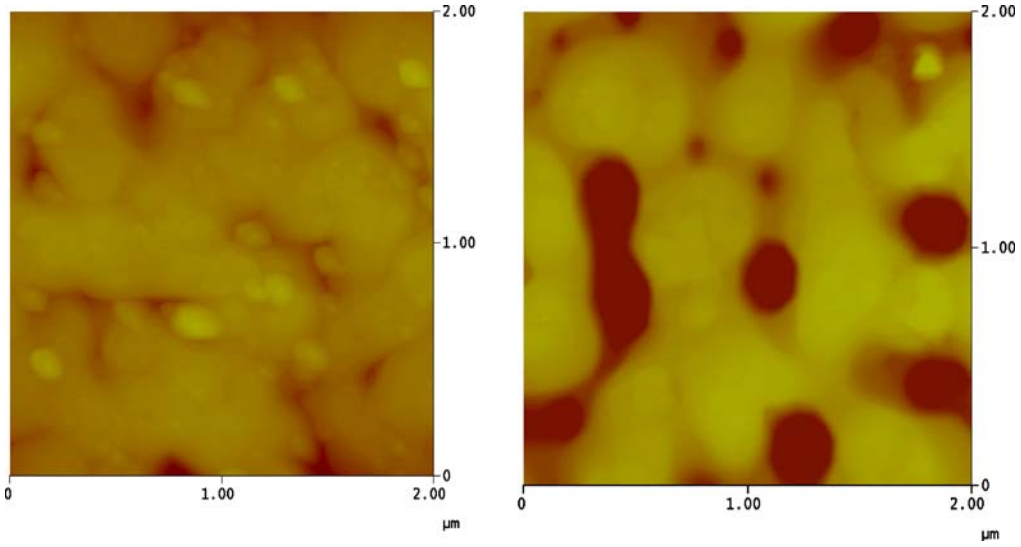

Fig. 2 AFM images for films with 30 (left) and 150 (right) Ag shots

and 4 Tesla was shown in Fig. 4. The general trend of J_c with the number of Ag shots is that J_c increases with n. Zero field J_c at 77 K increased from 10^6 to 3.3×10^6 A/cm^2, and from 1.5×10^7up to 4×10^7A/cm^2 for 5 K and zero field as n increased from zero to 150. However, at 77 and 40 K the J_c fluctuates with the number of Ag dots with $n < 60$ for both zero and 4 T. The same fluctuation of J_c is also seen for 5 K and 5 T. The J_c at 5 K and zero field, however, increases monotonically with the numbers of Ag shots. The reasons for these J_c fluctuations are not clear at this moment. It might be possible that the Ag-nano-dots may have different ways to settle onto the surface of the LAO substrate, most likely due to the Ag dots having different chemical activity compared to the dots deposited on both YSZ and STO substrates. This might affect the distribution or diameter of Ag-dots when the number of Ag shots is less than 60. A detailed study is needed to clarify what causes the fluctuation in J_c.

It seems that the surface morphology has no direct relationship with the J_c enhancement, as the surface morphologies are indistinguishable for all the films with $n < 130$. Therefore, we have to check the phases, orientations and crystallinity of our films.

All the XRD results revealed that the out-of-plane orientation of the Y123 films in all samples is strong, with only the (00l) reflections being present. A typical XRD pattern of the Y123 films is shown in Fig. 5. Therefore, it seems that there is no difference in the orientation of the samples. However, after checking the full width at half maximum (FWHM) for the (005) reflection peak for different samples, it was found that the values of the FWHM decrease as the Ag shots increase, i.e., the (005) peak becomes sharper with Ag dots (shown in Fig. 6), indicating the improvement of crystallinity in the films or improvement of the (00l) orientation of crystal grains. It is believed that the sharpening of diffraction peak indicate the improvement of crystallinity

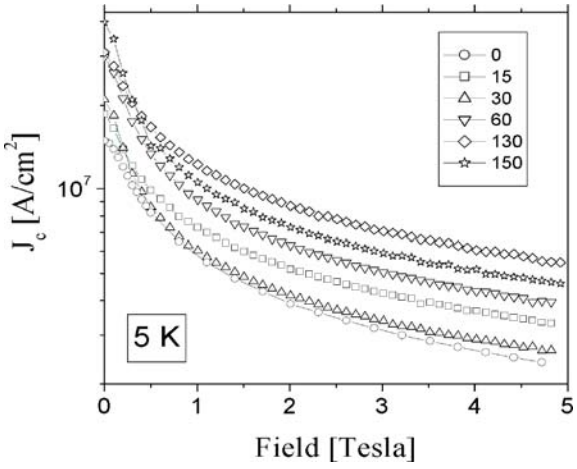

Fig. 3 Field dependence of J_c at 5K for Y123 films with different numbers of Ag shots

or increase in crystal size. It should be noted that the crystallinity refers to two important factors: (1) crystal sizes; (2) crystal defects. The sharpening of diffraction peak is decided by two things: one is crystallinity, the other is orientation of crystal grains. Only (00l) peak are present in our samples. This fact can only indicate that the grains in our film are (00l) or *c*-axis oriented. However, it should be pointed out that even the grain are *c*-oriented, there may be a little *c*-axis misalligent with very small tilt angles for tiny grains. These tiny angles can only be reflected from the peak sharpening in the case all the grains have the same grain sizes. As we have shown in our AFM images studies, there were no changes in grain sizes for all the samples with Ag shots (n) of less than 150. Furthermore, the grain size reduced for $n = 150$ times. These facts strongly indicated that the *c*-axis orientation of crystal grain in our sample were improved as the numbers of Ag shots increased. Therefore, we believe that the increase of J_c with Ag nano-shots should be ascribed to the improve-

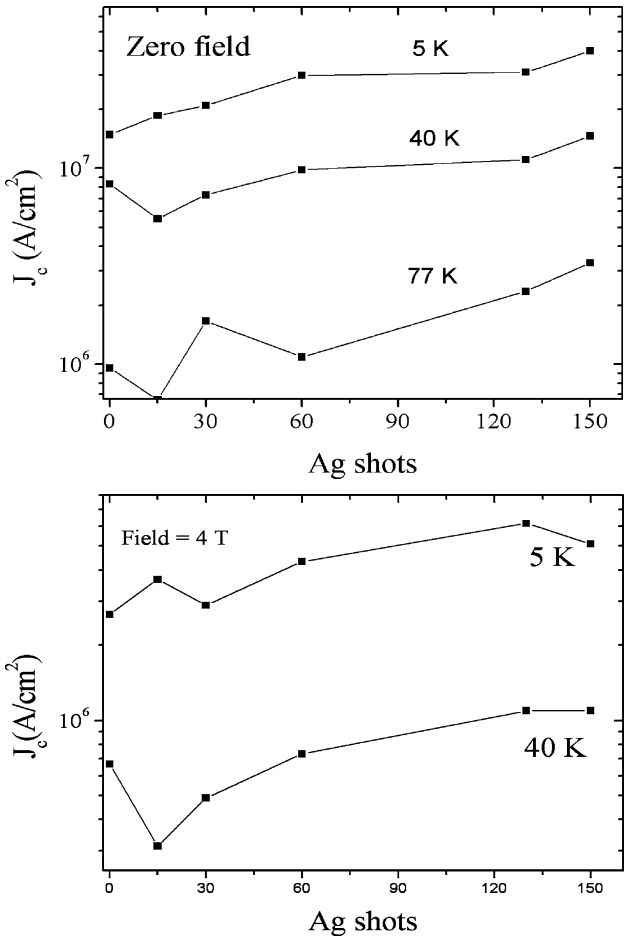

Fig. 4 J_c vs the number of Ag shots for the film grown on LAO in zero (up) and 4 T (down)

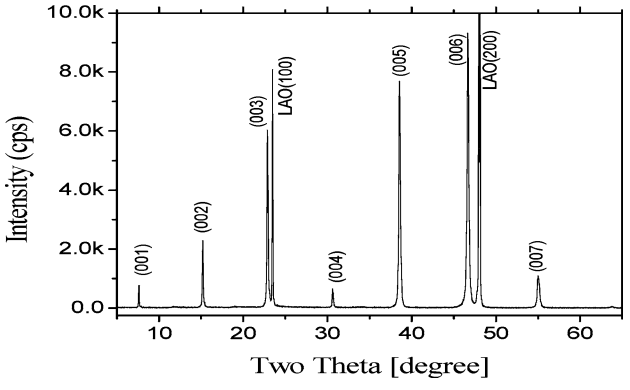

Fig. 5 XRD for Y123 grown on LAO with 60 Ag shots

ment in the out-of-plane alignment caused by Ag nano-dot modification to substrate surfaces.

The results of J_c enhancement with n present in this study show that for Y123 film grown on LAO the introduction of Ag-nano-dots onto the substrates prior to the deposition of Y123 thin films really played a role in enhancing the J_c at both low field and high field and confirmed what we have obtained for both YSZ and STO substrates [6–8].

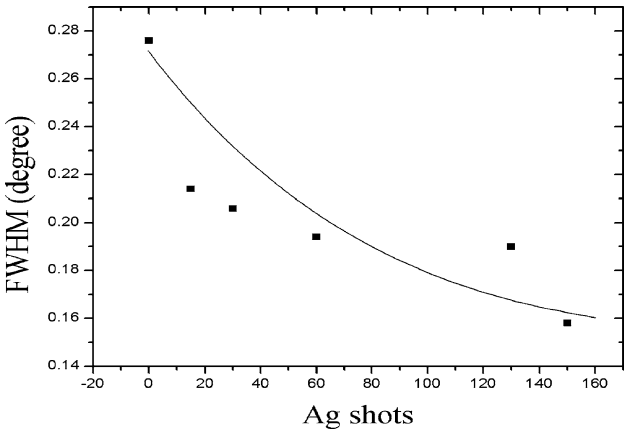

Fig. 6 Full width at half maximum for (005) peak of films with different Ag shots

It should be emphasised that all the Y123 films were laser deposited under the same ablation conditions, such as the same oxygen pressure, substrate temperature, laser energy, etc., The only difference is the amount of Ag dots deposited on the substrates prior to the deposition the Y123 films. It worth pointing out that the J_c values at 77 and 5 K for the reference sample (with zero Ag shots) are relatively low compared to pure Y123 films fabricated under optimised PLD conditions [7]. This implies that the ablation conditions used for our samples are not optimised. The results of the J_c enhancement by the Ag nano-dots for our samples indicated that surface modification with Ag nano-dots is another factor controlling the performance of the Y123 films in addition to other physical deposition conditions. We thought that the Ag should locate at the interface of Y123 and substrate as the Ag nano-dots were produced onto the substrate surface before the Y123 was deposited. This has been confirmed by our TEM observation of distribution of the Ag nano-dots on YSZ substrates prior the deposition of Y123 [8]. After the deposition of Y123, the Ag dots might migrate due to the interaction with Y123 droplets ablated from Y123 target. A detailed TEM is highly desirable for the direct evidence of the whereabout of the Ag and for the detailed crystal defects that should be introduced by the Ag nano-dots inclusion.

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