Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of the European Ceramic Society 24 (2004) 3623–3634
www.elsevier.com/locate/jeurceramsoc

Influence of irradiated target modification on the quality of pulsed laser deposited $YBa_2Cu_3O_{7-x}$ thin films

C. Andreouli [a,*], T. Efthimiopoulos [b], S. Christoulakis [b], A. Tsetsekou [c], C. Panagopoulos [d]

[a] *CERECO S.A, P.O. Box 146, Chalkida 34100, Greece*
[b] *Department of Physics, University of Crete, P.O. Box 2208, Heraclion 71003, Greece*
[c] *Department of Mining Resources Engineering, Technical University of Crete, Chania 73100, Greece*
[d] *Department of Materials Science, National Technical University of Athens, 15780 Zografou, Athens, Greece*

Received 3 September 2003; received in revised form 29 November 2003; accepted 7 December 2003

Available online 14 April 2004

Abstract

$YBa_2Cu_3O_{7-x}$ thin films were deposited by pulsed laser deposition (PLD) using differently preablated targets. The effect of the total number of laser pulses on the morphology and composition of the $YBa_2Cu_3O_{7-x}$ target was investigated and the induced modification of the irradiated surface was found to affect the characteristics of the produced films. It is shown that both the particle density and the particle size in the prepared *c*-axis oriented films have been greatly reduced using a target preablated with a low number of pulses. The optimum conditions for the preparation of high quality $YBa_2Cu_3O_{7-x}$ thin films with reduced particles and high critical current density are established.

Keywords: Films; Surfaces; Oxide superconductors; $YBa_2Cu_3O_7$; Superconductors; Pulsed laser deposition

1. Introduction

Pulsed laser deposition technique (PLD) has emerged as a very powerful method for the production of high quality superconducting $YBa_2Cu_3O_{7-x}$ thin films.[1–3] However, one of the main disadvantages of PLD technique, the appearance of particles on their surface that limits its applicability in microelectronics and multilayer technology,[4] is still a challenging problem. The most common particles are the "droplets" with typical sizes of 1 μm, which are detrimental to the film surface smoothness. The reduction of droplet density on the surface of films grown by the pulsed laser deposition PLD method is a goal actively aimed by many research groups. Several attempts have been made to reduce the density of droplets by optimising the deposition conditions: changing the deposition geometry,[5] using different laser wavelength beams.[6] Concerning the role of the $YBa_2Cu_3O_{7-x}$ target characteristics on the film quality the use of a freshly polished target,[7,8] and recently, the use of targets fabricated by a modified melt-textured growth method instead of solid-state sintering,[9] as well as the effect of target preablation specifically on the chemical composition of the produced PLD films have been reported.[10] For a Si and FeSiGaRu alloy, it has been shown that the droplet emission correlates closely with the surface roughness.[11] It was also reported that, in addition to the deposition parameters, the interaction between the laser beam and the target induces gradual modification of the target, which affects the plasma plume and thereby the film characteristics.[12] It is still questionable whether the droplets originate from the target or they are formed by condensation of the vapour.[13] However, the target surface roughness depends on the laser fluence,[14,15] the laser beam wavelength and also on the way by which the laser beam moves relatively to the target during ablation.[16]

The goal of this paper is to systematically study the cumulative number of pulses effect on the morphology and composition of the irradiated surface and its subsequent effect on the film characteristics. All reported work so far were focused on the target properties and surface roughness of the film, whereas, the present study, further addresses critical issues, namely the ablation rate, the films' particles density and the films' critical current density and correlates them to the preablation stage.

The stoichiometric transfer of the target material onto the substrate is one of the major attractions of the laser ablation

* Corresponding author. Tel.: +30-2620-71226; fax: +30-2620-71461.
E-mail address: dimdia@hol.gr (C. Andreouli).

0955-2219/$ – see front matter © 2004 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jeurceramsoc.2003.12.010

technique, understanding the evolution of composition of the target surface is very important in order to control the film quality.

2. Experimental

Fig. 1 shows a schematic diagram of the pulsed laser deposition apparatus used in the experiment. For film deposition, a XeCl excimer-laser ($\lambda = 308$ nm, $E_{max,output} = 600$ mJ, $T_L = 30$ ns) was employed. The apparatus includes a cylindrical stainless steel vacuum chamber, a substrate holder with precise temperature control and the target material holder. The laser beam was passed through a circular aperture in order to select the uniform part of the beam. The aperture was inclined by 40° with respect to the optical axis and imaged onto the target surface via a single plano-convex lens ($f = 150$ mm) and a uniform circular laser spot was obtained on the target. The $YBa_2Cu_3O_{7-x}$ targets were prepared by Cereco S.A. using the cold press-sintering process.[17,18] The cylindrical target was mounted on the rotating holder and its dimensions were: 15 mm diameter and 3 mm thickness. Before each deposition, the laser pulse energy in front of the target was measured with a pyroelectric detector.

For the substrate heating, a small resistive heater was fabricated using an insulated Inconel heating wire (Thermocoax Inc-10, 1 mm diameter). The heater consisted of two layers of the heating wire, packed between two stainless steel plates 40 mm in diameter. On the rear side of the heater, between the heating wire and the plate, a thin Al_2O_3 plate was placed in order to reduce the heat losses. The substrate was placed at the front side of the heater, facing the target and was attached using silver paste. The temperature was measured by a thermocouple placed in a borehole of the heater plate and a temperature control unit (Shimaden FP21) was regulating the temperature. Before deposition the substrates were cleaned in an ultrasonic bath using initially trichloroethanol, then acetone and finally isopropyl alcohol. Subsequently the substrates were dried with N_2 gas. During the deposition the substrate temperature was kept constant at the selected value. The oxygen background pressure was 0.7 mbar. Immediately after the deposition, the chamber was filled with 900 mbar of oxygen and the film was annealed at 450 °C for 1 h.

The cumulative number of pulses effect was studied for a stationary target and for a rotating one. For a rotating target, which is the case for the actual film deposition, the effect of number of pulses on the target roughening and on the deposited thin film characteristics, have been investigated through the preablation stage. Preablation is carried out before the film deposition and is related to the radiation of target with a certain number of pulses for the target surface cleaning and conditioning. The stationary targets were ablated with a variable total number of pulses up to 5000 using an energy density $F = 1.8\ \mathrm{J\,cm^{-2}}$, which is above the threshold fluence of $1.0\ \mathrm{J\,cm^{-2}}$ for stoichiometric ablation.[15]

With respect to the rotating target, the effect of multiple pulses on the target roughening and consequently on the films quality was studied for the preablation stage. For this purpose two cases were investigated: (i) a low number of 25 shots/site and (ii) a much higher number of 185 shots/site. In both cases, during the subsequent film deposition, the rotated targets were irradiated with an equal total number of pulses, $N = 4000$ (160 shots/site). The thin films were deposited on (1 0 0) MgO or $LaAlO_3$ substrates. The deposition parameters were: pulse energy on the target $E = 70$ mJ, energy density $F = 3.5\ \mathrm{J\,cm^{-2}}$, substrate temperature $T_S = 750$ or 730 °C, laser spot size $A_S = 2\ \mathrm{mm^{-2}}$, target–substrate distance $d_{T-S} = 51$ mm, oxygen pressure in the chamber during the deposition $P_{O_2} = 0.7$ mbar and laser pulse repetition rate 10 Hz.

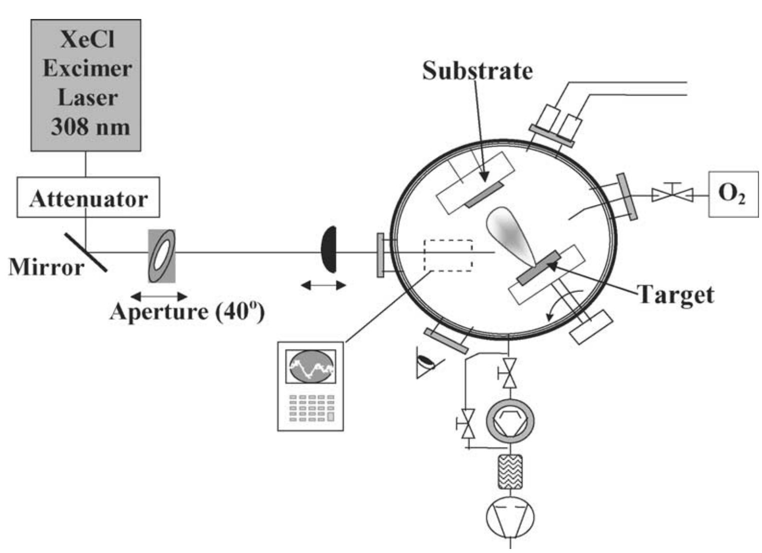


Fig. 1. Experimental apparatus of pulsed laser deposition.

The surface morphology of the targets and films was investigated using a scanning electron microscope (SEM) while energy dispersive X-ray microanalysis (EDX) was employed to determine the chemical composition of the targets, films and particulates. Because the penetration depth of electrons at the SEM microscope was approximately 1 μm, particulates of this size could not be measured accurately and only qualitative reports of their chemical composition were made. The structure and orientation of films was investigated with X-ray diffraction analysis (XRD) and the transition temperature to zero resistance of the superconducting films (T_c), was measured using the ac susceptibility method (magnetic field: $H_{ac} = 1\,G$; frequency: $\nu = 1024\,Hz$). The critical current density J_c as a function of the applied dc magnetic field H was also measured for the deposited thin films. The values of J_c were calculated from hysteresis loops of $YBa_2Cu_3O_{7-x}$ thin films at $T = 5\,K$ using a SQUID magnetometer (Quantum Design MPMS2) and a model reported by Moraitakis et al.[19] The film thickness (d_F) was measured with a mechanical stylus profilometer. The number of droplets on the films was estimated using an image analysis programme (PC-Image, version 2.2.05, Foster Findlay Associates).

(a)

(b)

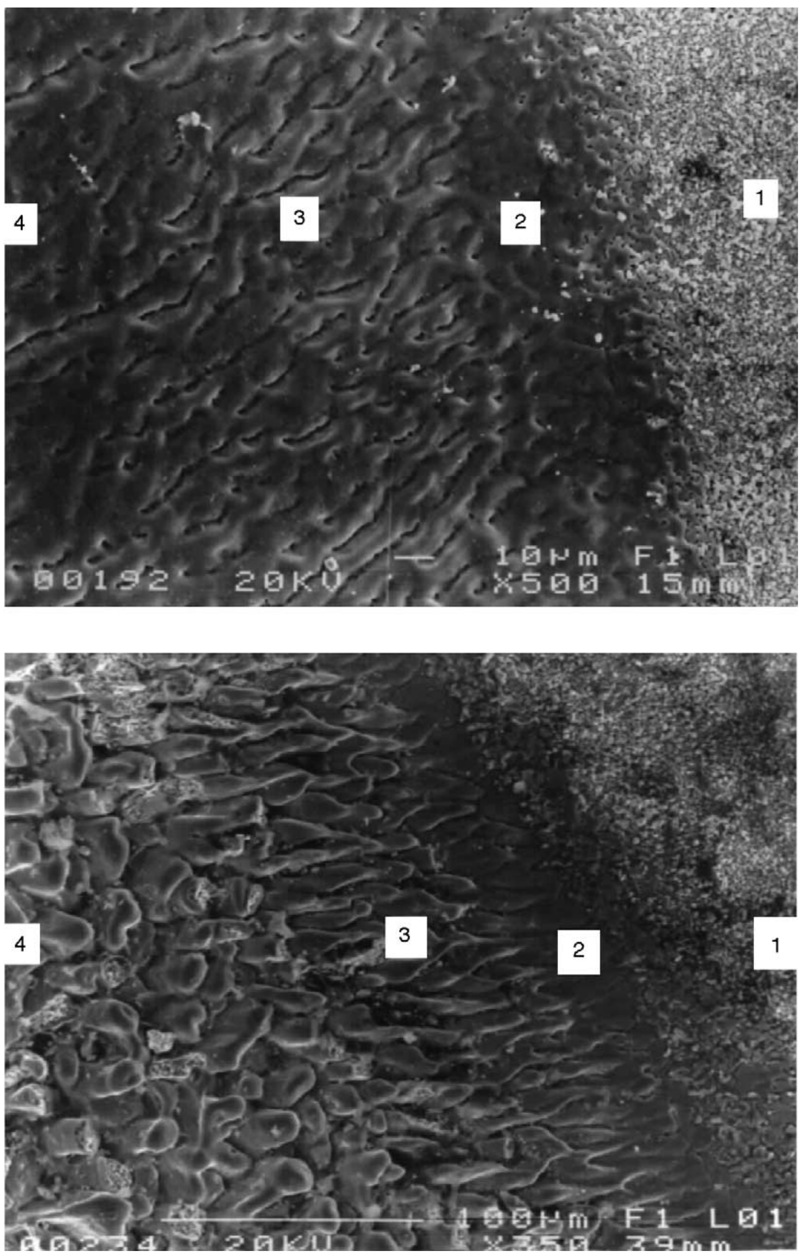


Fig. 2. SEM micrographs of the various regions (1, 2, 3 and 4) appeared on a stationary $YBa_2Cu_3O_{7-x}$ target, after irradiation with $F = 1.8\,J\,cm^{-2}$: (a) 25 pulses and (b) 300 pulses.

3. Results and discussion

In this section the laser–target interaction for a stationary target and the interaction for a rotating target, which is the case of the actual experiments used for the film preparation, will be investigated. Then the film preparation and evaluation will be considered and finally the correlation between the target morphology and the film characteristics will be examined.

3.1. The multiple pulses effect on a stationary $YBa_2Cu_3O_{7-x}$ target

Although the use of a stationary target is not the case for actual film deposition, the number of consecutive laser pulses on the same spot area is used for a thoroughly investigation of the morphological and compositional changes which take place on the irradiated target surface.

Fig. 2 shows a SEM micrograph of the target irradiated by either 25 consecutive pulses (a) or 300 pulses (b). The numbers on the SEM micrographs correspond to the region: just outside the ablated area (region 1), at the edge of the ablated surface (region 2), at the inside section (region 3) and at the center of the irradiated area (region 4). It is clear from these micrographs that the target surface modification differs across the irradiated area as the number of pulses increases. For the low number of 25 laser pulses using the pulse energy density of $1.8\,J\,cm^{-2}$ the central ablated area has a melted appearance, the area near the edge looks rougher while the not ablated area is covered with a

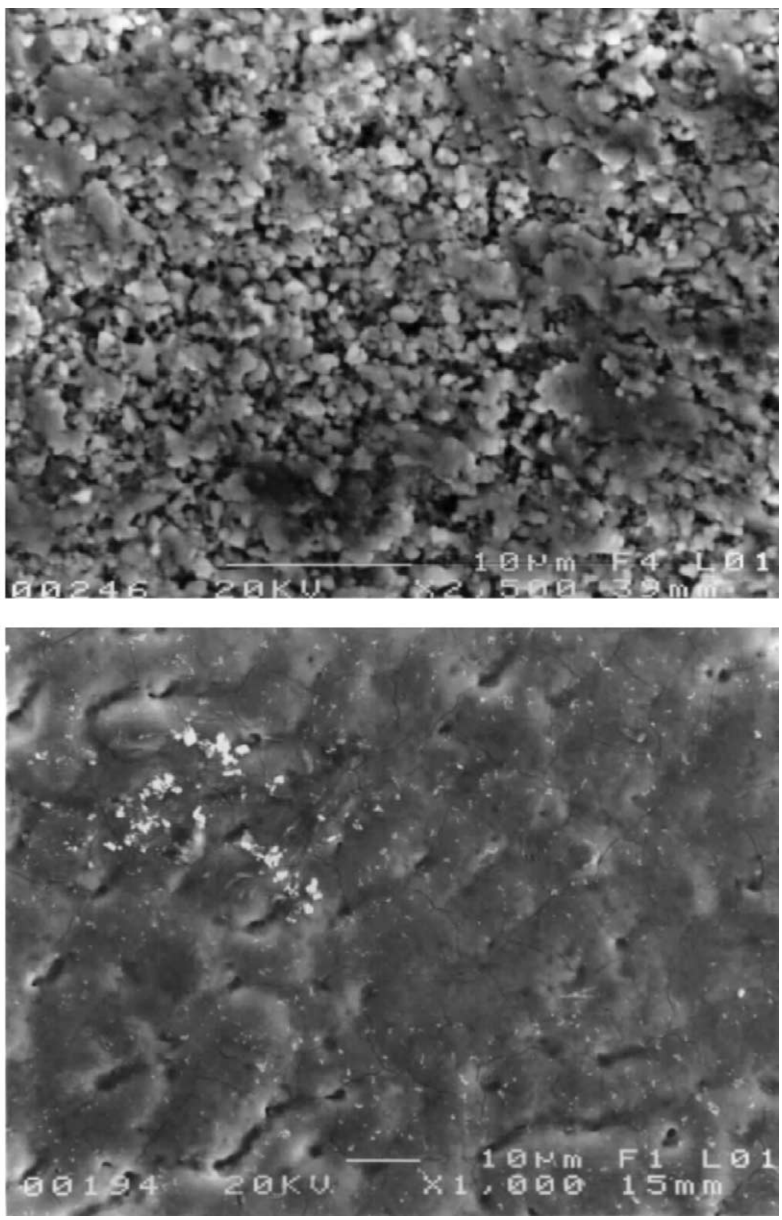
(a)
(b)


Fig. 3. SEM micrographs of a stationary $YBa_2Cu_3O_{7-x}$ target after irradiation with $F = 1.8\,J\,cm^{-2}$ ($\nu = 10\,Hz$) and with: (a) 0 pulses, (b) 25 pulses, (c) 75 pulses, (d) 300 pulses, (e) 3000 pulses and (f) 5000 pulses.

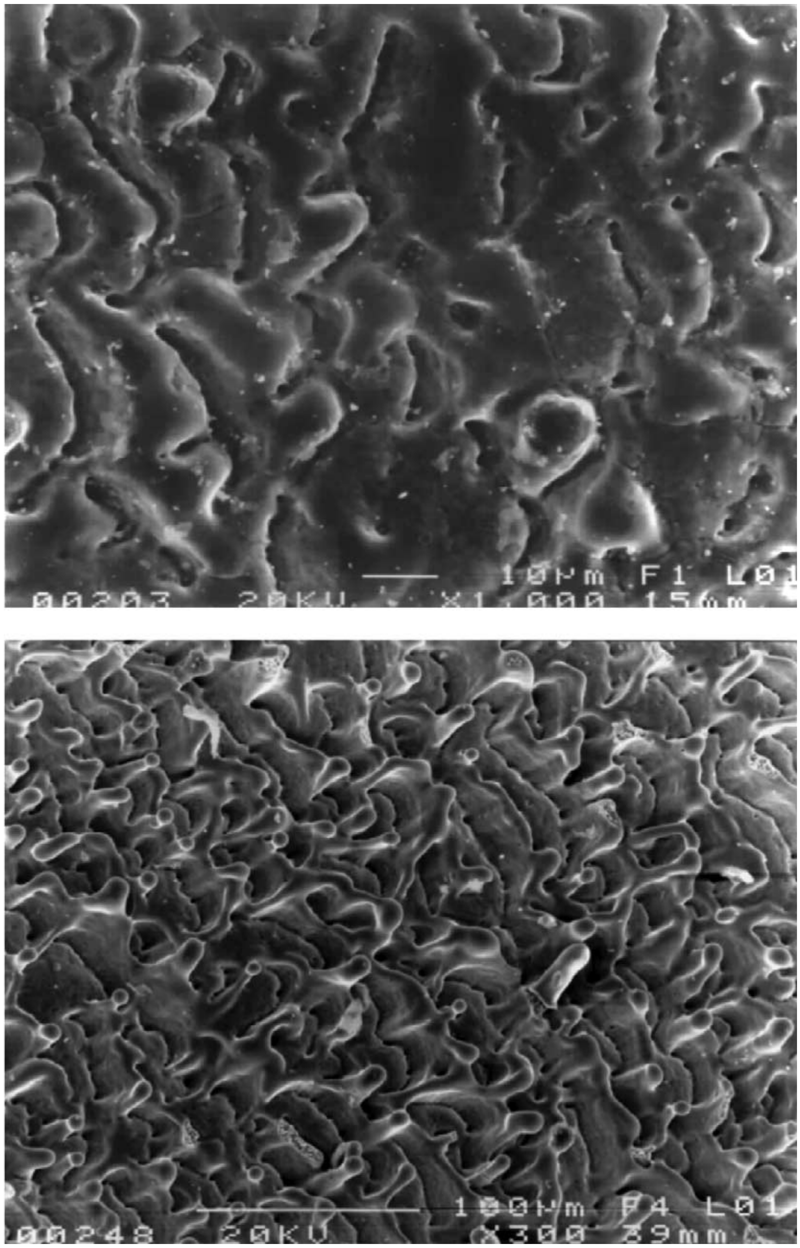
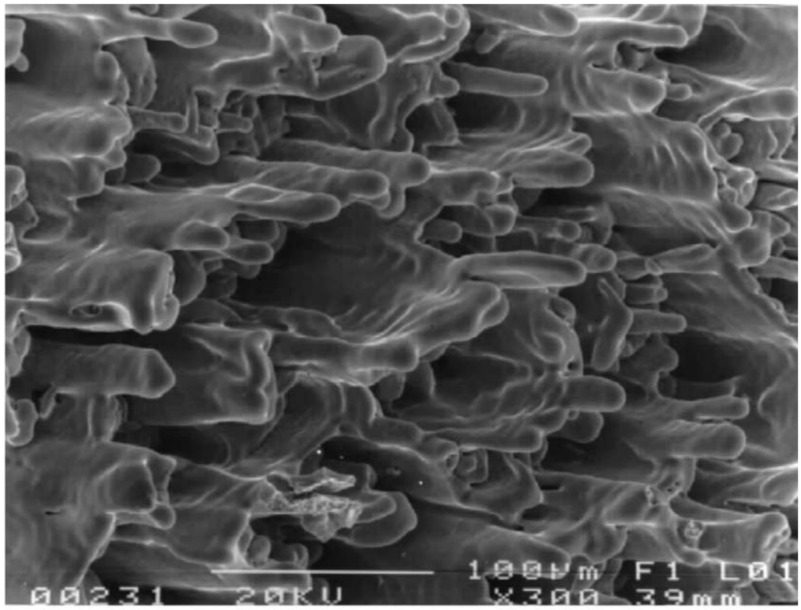
(c)
(d)


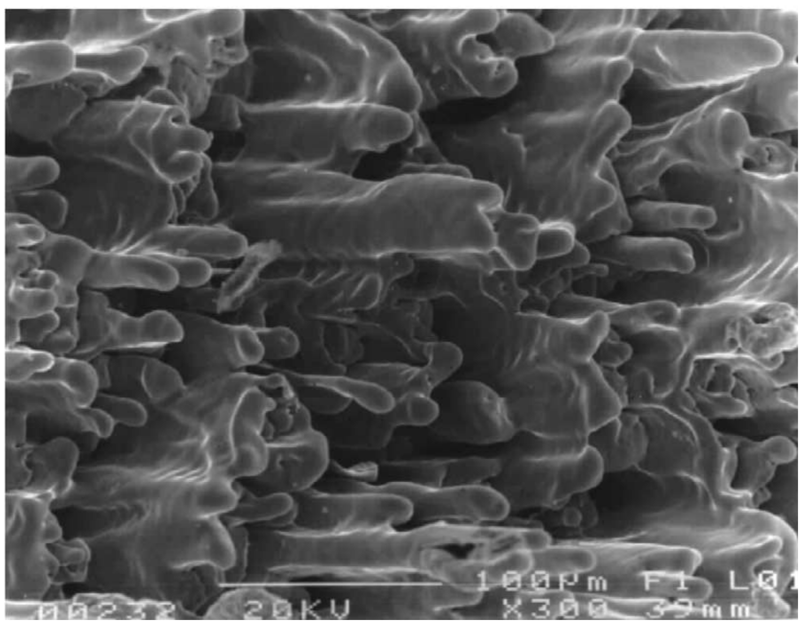
Fig. 3. (*Continued*)

powder like layer. As the total number of pulses increases, the roughness of the irradiated area of the target increases and different kind of columns and craters appear on the surface.

Selected characteristic SEM micrographs of the central target area (region 4), when different total number of consecutive pulses were employed, are shown in Fig. 3, indicating the presence of several irregularities. With 25 pulses fallen on the initial rough surface shown in Fig. 3a, a smooth, "melt-like" surface is observed as shown in Fig. 3b. As the total number of pulses increases to 75 small hillocks (5 μm height) start to form within the ledges (Fig. 3c) and for a total number of 300 pulses hillocks (13–20 μm height) have formed with some of their flat parts cleaved away (Fig. 3d). After a prolonged ablation with several thousand of pulses, the target surface is transformed into a closely packed and broad columnar structure aligned in the direction of the laser beam (Fig. 3e and f). The average height of these columnar irregularities formed on the target surface as a function of the total number of pulses, is presented in Fig. 4. The existence of the columnar structure modifies the effective illumination of the surface, which seems to be position and energy dependent. Fig. 5 shows the stoichiometry of the different regions 1–3 (shown in Fig. 2), concerning the Y, Cu and Ba elements and Fig. 6 shows the stoichiometry of the presented morphological irregularities located in the central area of the target (region 4) (shown in Fig. 2). It can be clearly seen that during the first few hundred pulses



(e)



(f)

Fig. 3. (*Continued*).

there is a strong change in stoichiometry. Copper from the plasma plume is deposited on the cold surface outside the ablated area causing the change in stoichiometry of the region. The diagrams concerning the regions 2 and 3 suggest that in the first 250 pulses approximately, Cu is leaving faster than the other elements. Above 250 pulses the Ba concentration drops slightly, while the Y concentration drops considerably. This means that there is an excess removal of Y preferentially. With respect to the EDX study of the formed irregularities (of region 4), the results are presented in Fig. 6, more specifically, (i) the tip of the hillocks or columns (Fig. 6a), (ii) the body of the columns (Fig. 6b) and (iii) the flat areas between the columns (Fig. 6c). The target was irradiated with $F = 1.8\,\mathrm{J\,cm^{-2}}$ for various number of pulses. It can be clearly seen that: (a) the hillocks to be Y-rich and Cu-deficient, (b) the flat melted surface to have an almost stoichiometric $YBa_2Cu_3O_{7-x}$ composition and (c) for the observed columns at 1000 pulses, the tip of the column has a smaller Y enrichment than the hillocks and their body side is Y-poor and Cu-rich. The SEM micrograph of the column main body taken from some scraped columns reveals a polycrystalline interior with the initial $YBa_2Cu_3O_{7-x}$ stoichiometry.

Therefore, as the number of pulses increases, leading to increased surface roughening, the effective irradiated area increases and the absorbed laser energy flux begins to drop below the ablation threshold flux leading to preferential evaporation. According to some reported works, the formed

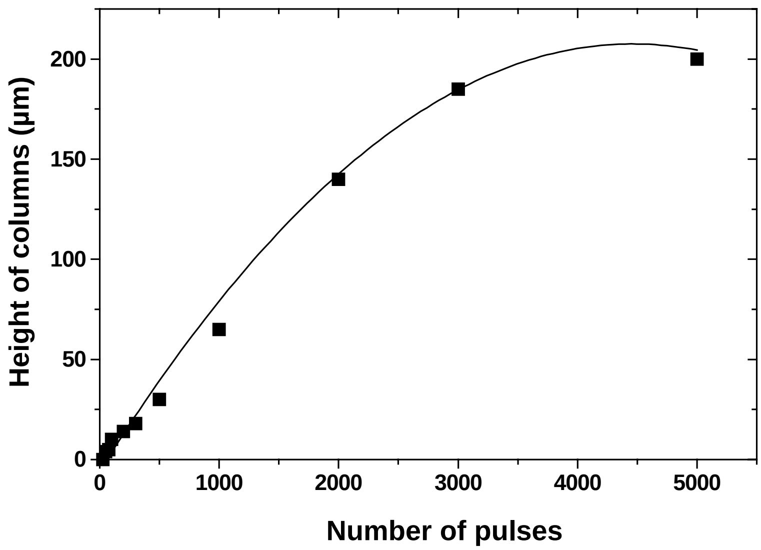


Fig. 4. Average height of columnar irregularities formed after irradiation with $F = 1.8\,\mathrm{J\,cm^{-2}}$, as a function of total number of pulses.

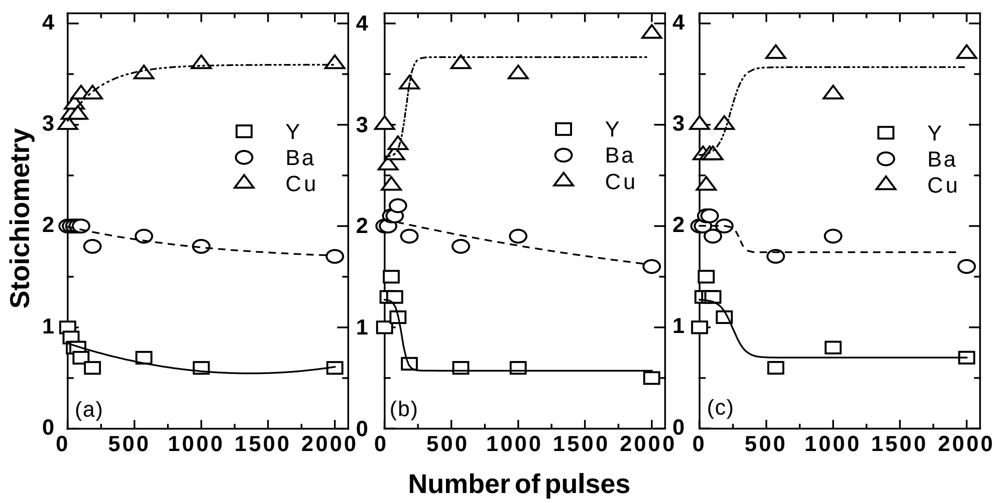


Fig. 5. Stoichiometry of: (a) region (1), (b) region (2) and (c) region (3) of a target irradiated with $F = 1.8\,\mathrm{J\,cm^{-2}}$, as a function of the total number of pulses (regions 1, 2 and 3 are presented in Fig. 2).

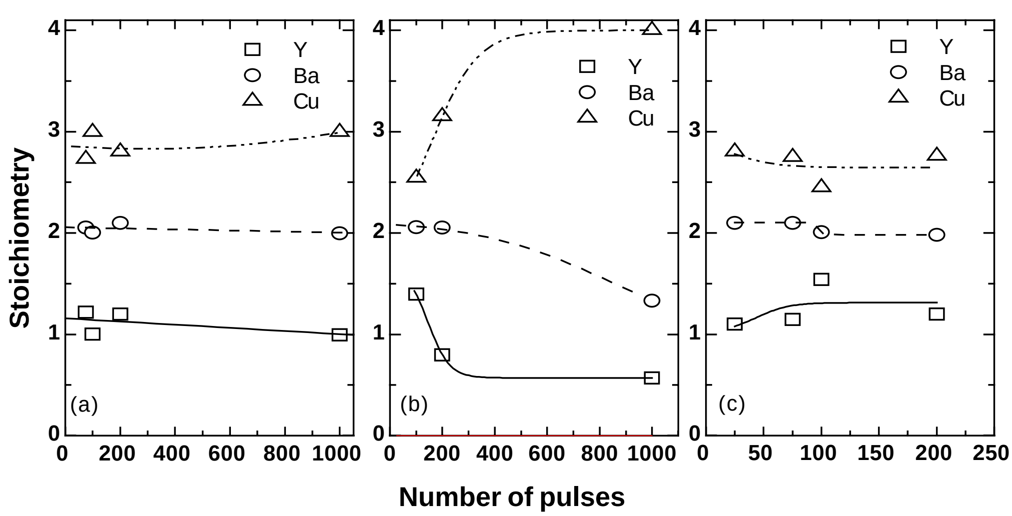


Fig. 6. Stoichiometry of: (a) the tip of the columns, (b) the body of the columns and (c) the flat areas between the columns, for targets irradiated with various total number of pulses.

columns exhibit phase segregation and the Y-rich hillocks, which are formed on an initially smooth surface, act as a shield for the target, due to their greater resistance to laser vaporisation.[12,14]

In conclusion, the laser irradiation of an $YBa_2Cu_3O_{7-x}$ target, with several total numbers of pulses on the same stationary area, produces various morphological irregularities of different shapes and stoichiometry. The stoichiometry of the irregularities as a function of the pulses reveals that: (i) Cu leaves first and part of the evaporated Cu is redeposited in the region around the irradiated area changing the local stoichiometry and (ii) the change of the Cu concentration equals the reduction of Ba and Y, for the large total number of pulses.

3.2. The multiple pulses effect on a rotating $YBa_2Cu_3O_{7-x}$ target and the deposited thin films

The above results show that as the total number of pulses increases the irradiated target surface roughness increases along with higher stoichiometry deviations. For this reason the target has to be rotated, limiting the number of laser shots per site and keeping the atomic composition ratio close to Y:Ba:Cu = 1:2:3. As it has been recently reported, in order to prepare high quality $YBa_2Cu_3O_{7-x}$ epitaxial films, the irradiated target should have exactly the stoichiometric ratio 1:2:3, as the films deposited from non-stoichiometric targets exhibited a large number of outgrowths in addition to droplets and a lower T_c.[20]

SEM observations of freshly polished targets, which were initially irradiated with $F = 3.5\,\mathrm{J\,cm^{-2}}$ receiving 25 shots/site at 10 Hz repetition rate, revealed that the target surface is smooth and “melt-like” with the correct $YBa_2Cu_3O_{7-x}$ stoichiometry (Fig. 7a). In the case of a higher number of 185 shots/site some morphological irregularities appeared at the target edges (Fig. 7b). These targets were subsequently used for the deposition of thin films. During the deposition stage, the targets were irradiated with the same energy density of $F = 3.5\,\mathrm{J\,cm^{-2}}$ and were loaded with an additional 160 shots/site. The films were deposited on MgO substrates heated at $T_S = 750\,^{\circ}\mathrm{C}$.

Thus, at the end of the deposition, the first target was irradiated with a total number of 185 shots/site (25 shots/site initially during the preablation period and 160 shots/site during the deposition) and the second one was irradiated with a total number of 345 shots/site (185 shots/site initially during the preablation and 160 shots/site during the deposition). The irradiated groove area of the first target (SEM micrograph in Fig. 7b) exhibits some morphological irregularities at the edges only. The second target, irradiated with the higher total number of pulses, shows morphological irregularities, which are more intense, covering a wider area of the groove (Fig. 7c).

In order to investigate the effect of laser induced target surface modification on the film surface morphology, SEM micrographs were taken for the films deposited using the above described targets, shown in Fig. 8a and b, respectively. It was observed that the film corresponding to the lower number of 25 shots/site during the preablation (therefore with the lower total number of 185 shots/site after the end of

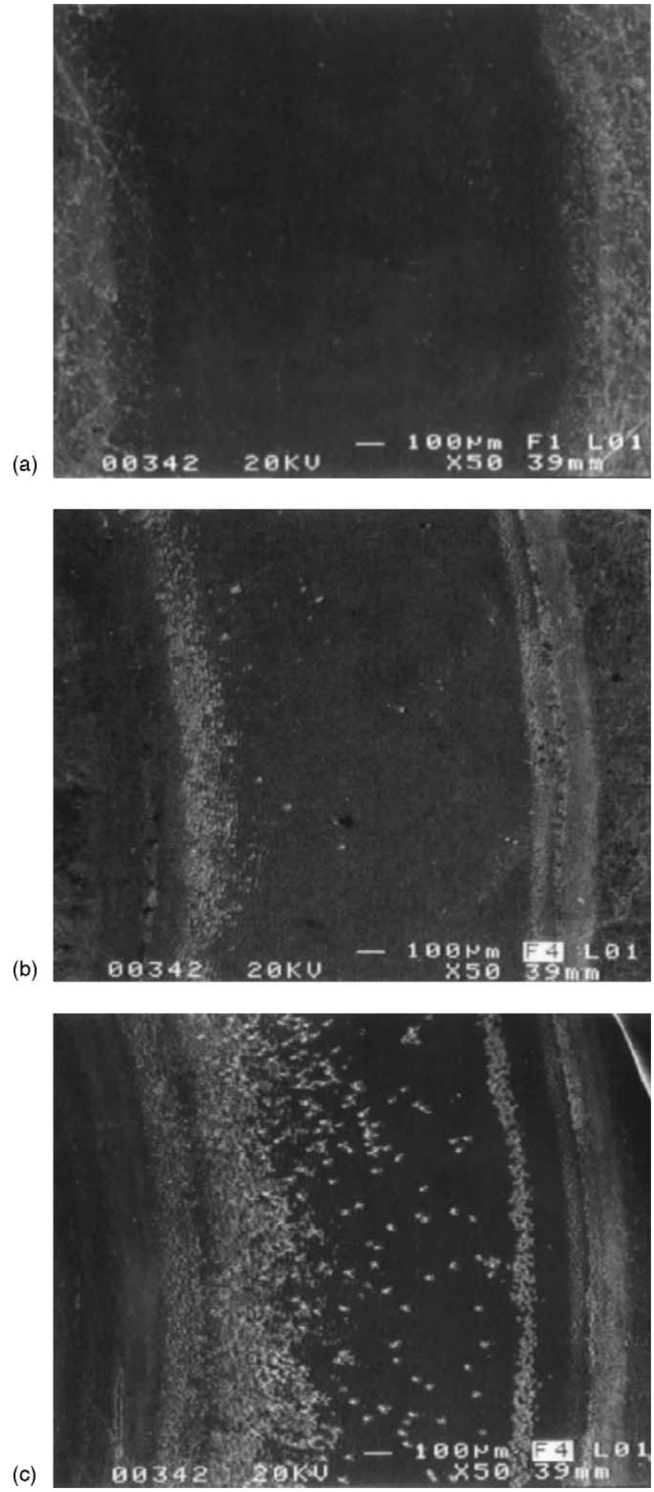


Fig. 7. SEM micrographs of the groove of an $YBa_2Cu_3O_{7-x}$ rotating target irradiated with energy density $F = 3.5\,\mathrm{J\,cm^{-2}}$ using: (a) 25 shots/site, (b) 185 shots/site and (c) 345 shots/site.

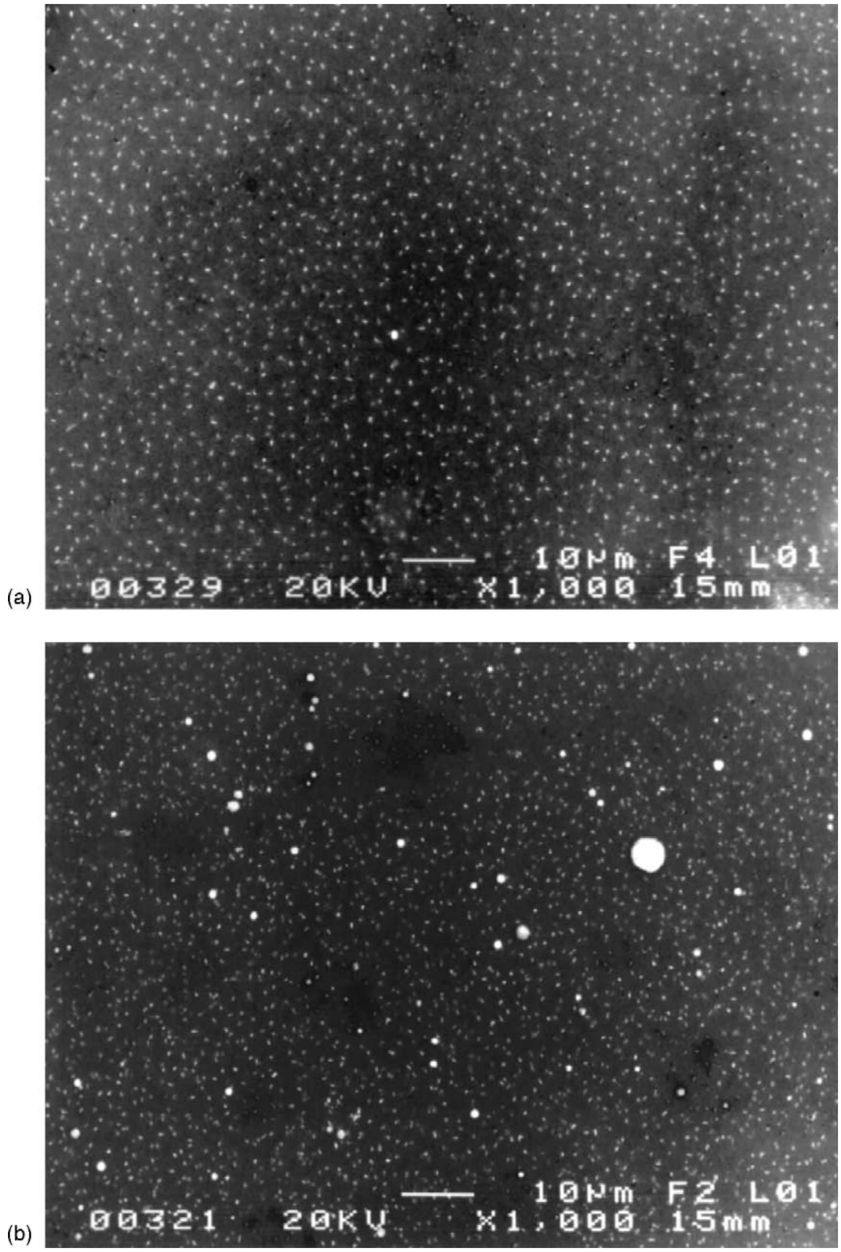


Fig. 8. SEM micrographs of films deposited on MgO substrates with $T_S = 750\,^\circ C$ and $F = 3.5\,J\,cm^{-2}$, using $YBa_2Cu_3O_{7-x}$ rotating targets irradiated with 160 shots/site during the film deposition process and having a preablation with: (a) 25 shots/site and (b) 185 shots/site.

deposition), exhibited insignificant number of droplets on its surface. Image analysis measurements showed that the number density of droplets is about $n_P \approx 10^5\,cm^{-2}$ (Fig. 8a). The fraction of the total area covered by these droplets is approximately 0.05%. On the contrary, the film resulted from the target that was irradiated with the higher number of 185 shots/site during the preablation (total number after deposition 345 shots/site), exhibited quite a few droplets on its surface and approximately the same number of submicron particles (Fig. 8b). The droplets number density in that case is about $n_P \approx 6 \times 10^5\,cm^{-2}$ and the fraction of the total area covered by these droplets is approximately 0.4%. The above described experiments were repeated for depositions with a lower substrate temperature of $T_S = 730\,^\circ C$ in order to test its effect on the number of droplets and similar results were obtained as shown in Fig. 9. The fraction of the total film area covered by the droplets is by a factor of six larger, in the case of targets that have undergone preablation conditioning with the higher number of pulses. Thus, the appearance of droplets seems to correlate with the target roughness and consequently with the irradiation conditions that cause this roughness.

Figs. 8 and 9 show also many submicron particles of the order of 0.2 µm. These particles are present for all the

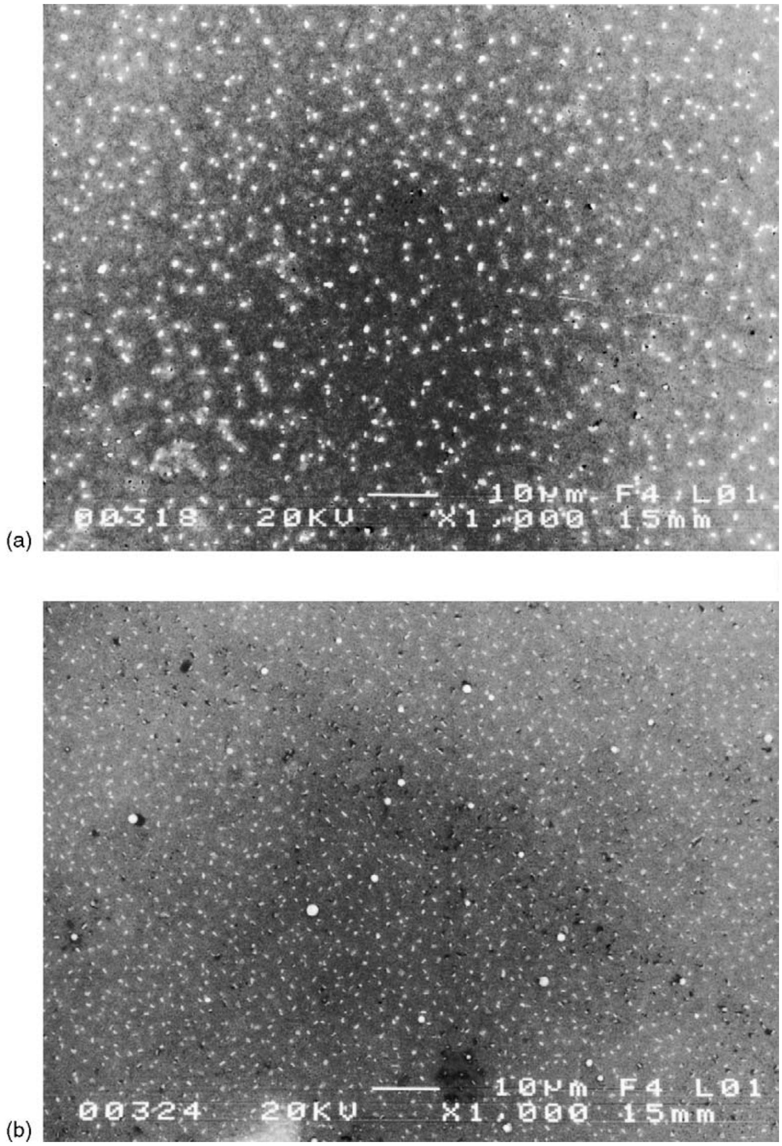


Fig. 9. SEM micrographs of films deposited on MgO substrates with $T_S = 730\,^{\circ}C$ and $F = 3.5\,J\,cm^{-2}$, using $YBa_2Cu_3O_{7-x}$ targets irradiated both with 160 shots/site during the film deposition process and having a preablation with: (a) 25 shots/site and (b) 185 shots/site.

examined experimental conditions with a number density of the order of $10^7\,cm^{-2}$ and it seems that their presence is not related to the morphology of the target. The presence of such submicron particles has been observed by many authors, in pulsed laser deposition of superconducting thin films.[3,7,14,20,21] According to Lee et al.[21] their presence is attributed to the growth mechanism of the film.

The films deposited using targets that were preablated with 25 and 185 shots/site and the same number of pulses for deposition, had a thickness $d_F \approx 320$ and 230 nm, respectively. All the other irradiation and deposition parameters were kept constant and the number of pulses for the film deposition was 4000 (160 shots/site). The measured film thickness corresponds to a deposition rate of $\Delta h_D \approx 0.8$ and 0.6 Å/pulse, respectively. The fact that the targets had been preablated with a different number of pulses, while in

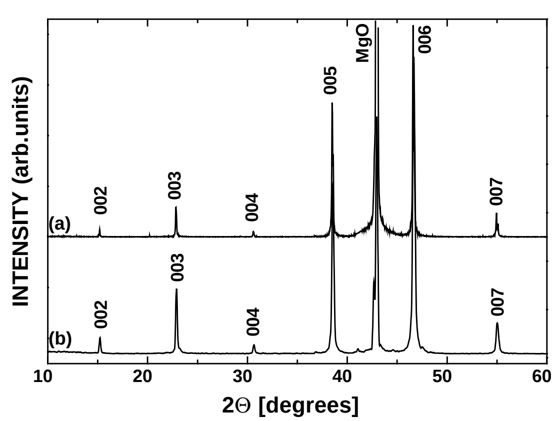


Fig. 10. X-ray diffraction patterns of $YBa_2Cu_3O_{7-x}$ films deposited on (1 0 0) MgO substrates, using targets that were preablated with: (a) 25 shots/site and (b) 185 shots/site.

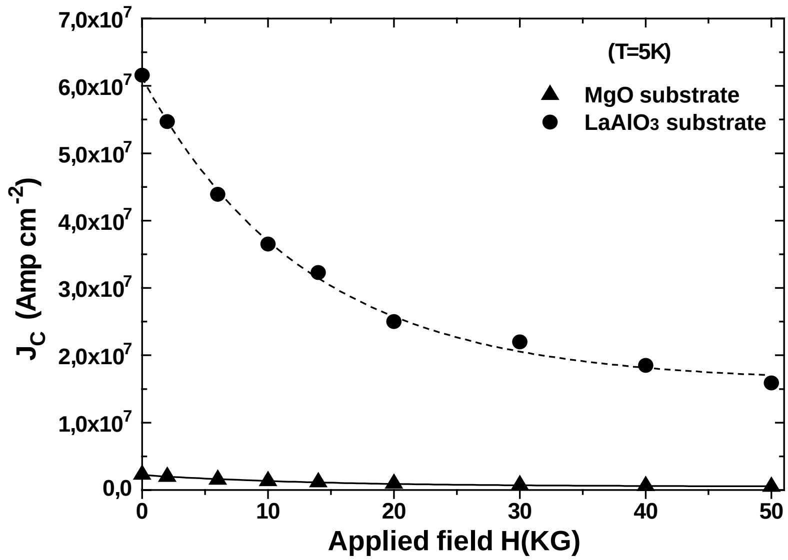


Fig. 11. Critical current density J_c as a function of the applied dc magnetic field H at 5 K, for $YBa_2Cu_3O_{7-x}$ films deposited on (1 0 0) MgO and (1 0 0) $LaAlO_3$ substrates using a target preablated with the low number of 25 shots/site.

both cases the deposition process was identical, shows the importance of target preablation on the film thickness, the ablation rate and the droplet number.

The XRD measurements give XRD patterns composed of only (0 0 1) sharp diffraction peaks for both types of films, as shown in Fig. 10a and b. These results indicate that the $YBa_2Cu_3O_{7-x}$ films are single phase and have c-axis orientation and high crystal homogeneity, suggesting formation of epitaxial films. For the film corresponding to the target of 25 shots/site $c \approx 11.67$ Å and for the film corresponding to the target prepared with 185 shots/site $c \approx 11.65$ Å. Such films with c-axis orientation and reduced surface roughness and droplet number density, are needed in devices where large transport currents of microwave power are required.[22] All the deposited films were stoichiometric, as determined by EDX analysis, while the droplet particles were Y-rich and Cu-deficient. For the smaller, submicron particles EDX analysis was not possible because their diameter of approximately 0.2 μm, was much smaller than the penetration depth of the electrons (∼1 μm) of the SEM microscope.

The observed droplets as well as the submicron particles on the film surface are not expected to affect the film superconducting properties as measured by the ac susceptibility at low frequency. However, a strong influence on the transmission characteristics of the films is expected for the microwave region due to the reduced penetration depth of the traveling wave at higher frequencies. Concerning the films that were deposited using the lower number of 25 shots/site during the preablation and exhibiting the lower number of 1 μm droplets on their surface, their superconducting properties were studied using ac susceptibility measurements and measurements of the critical current density J_c as a function of the applied dc magnetic field H. These films deposited on MgO substrates, exhibit a transition temperature at 91 K and for zero applied field ($H = 0$), the critical current density at 5 K is $J_c(0) = 2.3 \times 10^6\ \mathrm{A\,cm^{-2}}$. This value increases significantly when $LaAlO_3$ substrates are used instead of MgO substrates and the J_c value for zero applied field is as high as $J_c(0) = 6.1 \times 10^7\ \mathrm{A\,cm^{-2}}$ (Fig. 11).

Thus, using a limited number of pulses during the preablation stage, a reduced target roughening is observed resulting in the deposition of higher quality thin films. Using $LaAlO_3$ substrates the deposited films exhibit: (a) the lesser number of 1 μm droplets on their surface and (b) very good superconducting properties. The film surface roughness due to the droplets will have a destructive effect in case of high frequencies applications. It seems that the on-line control of the target surface condition is needed in order to prepare good quality films.

4. Conclusions

In this study, the laser–$YBa_2Cu_3O_{7-x}$ target interaction was systematically investigated in order to study the presence of particles on the films, which is one of the main drawbacks associated with the PLD technique and thereby further optimise the thin film properties. For stationary $YBa_2Cu_3O_{7-x}$ targets, it was found out that for increasing number of pulses, significant morphological changes take place, accompanied by local differences in the chemical composition. Columnar features are developing, the tip of which has Y excess and Cu depletion. This necessitates the use of a limited number of pulses per irradiated area for a rotated target, during the film deposition. When, at the preablation stage, a high number of pulses per irradiated area is applied, the target after the actual ablation–deposition stage, exhibits a plethora of columnar features, whereas on the surface of the deposited films there are spherical particles of 1 μm in size. These particles are undesirable if the films are to be used as a base for microelectonic devices working in the microwave region, since they are Y-rich and Cu-poor.

Therefore, the use of a limited number of pulses during the preablation stage, results in a reduced target roughening enabling a higher ablation rate and the deposition of high quality thin films on the appropriate substrates. Such films exhibit the lesser number of 1 μm droplets on their surface and have very good superconducting properties, nearly the same as the best $YBa_2Cu_3O_{7-x}$ thin films yet fabricated.

Acknowledgements

A part of this work was done in the Materials-Laser Lab of TEI of Heraklion (Greece). We are also grateful to Dr. D. Niarchos of the National Center for Scientific Research “Demokritos”, for the use of his lab facilities, for the thin film electrical measurements, and to Dr. E. Moraitakis for his helpful discussions on the electrical measurements.

References

1. Kühle, A., Skov, J. L., Hjorth, S., Rasmussen, J. and Hansen, J. B., Smooth $YBa_2Cu_3O_{7-x}$ thin films prepared by pulsed laser deposition in O_2/Ar atmosphere. *Appl. Phys. Lett.* 1994, **64**, 3178–3180.
2. Wördenweber, R., Growth of high-T_c thin films. *Supercond. Sci. Technol.* 1999, **12**, R86–R102.
3. Proyer, S., Stangl, E., Borz, M., Hellebrand, B. and Baüerle, D., Particulates on pulsed-laser deposited Y–Ba–Cu–O films. *Physica C* 1996, **257**, 1–15.
4. Wellstood, F. C., Kingston, J. J. and Clark, J., Thin-film multilayer interconnect technology for $YBa_2Cu_3O_{7-x}$. *J. Appl. Phys.* 1994, **75**, 683–702.
5. Holzapfel, B., Roas, B., Schultz, L., Bauer, P. and Saemann-Ischenko, G., Off-axis laser deposition of $YBa_2Cu_3O_{7-x}$ thin films. *Appl. Phys. Lett.* 1992, **61**, 3178–3180.
6. Koren, G., Gupta, A., Baseman, R. J., Lutwyche, M. I. and Laibowitz, R. B., Laser wavelength dependent properties of $YBa_2Cu_3O_{7-x}$ thin films deposition by laser ablation. *Appl. Phys. Lett.* 1989, **55**, 2450–2452.
7. Misra, D. S. and Palmer, B., Laser ablated thin films of $YBa_2Cu_3O_{7-x}$: the nature and origin of the particulates. *Physica C* 1991, **176**, 43–48.
8. Chang, C. C., Wu, X. D., Ramesh, R., Xi, X. X., Ravi, T. S., Vencatesan, T. *et al.*, Origin of surface roughness for *c*-axis oriented Y–Ba–Cu–O superconducting films. *Appl. Phys. Lett.* 1990, **57**, 1814–1816.
9. Kim, C. H., Kim, I. T., Hong, K. S., Hahn, T. S. and Choi, S. S., Effects of target microstructure on pulsed laser deposited $YBa_2Cu_3O_7$-thin films. *Thin Solid Films* 2000, **358**, 223–228.
10. Podgursky, V. and Friesel, M., Congruent laser ablation due to pre-irradiation of target. *Physica C* 2000, **339**, 49–52.
11. Van de Riet, E., Nillesen, C. J. C. M. and Dieleman, J., Reduction of droplet emission and target roughening in laser ablation and deposition of metals. *J. Appl. Phys.* 1993, **74**, 2008–2012.
12. Pinto, R., Pai, S. P., D’Souza, C. P., Gupta, L. C., Vijayaragharan, R., Kumer, D. *et al.*, Optimization of KrF laser ablation parameters for in-situ growth of $YBa_2Cu_3O_{7-x}$. *Physica C* 1992, **196**, 264–270.
13. Strikovsky, M. D., Klyuenkov, E. B., Gaponov, S. V., Schubert, J. and Copetti, C. A., Crossed fluxes technique for pulsed laser deposition of smooth $YBa_2Cu_3O_{7-x}$ films and multilayers. *Appl. Phys. Lett.* 1993, **63**, 1146–1148.
14. Foltyn, S. R., Dye, R. C., Ott, K. C., Peterson, E., Hubbard, K. M., Hutchinson, W. *et al.*, Target modification in the excimer laser deposition of $YBa_2Cu_3O_{7-x}$. *Appl. Phys. Lett.* 1991, **59**, 594–596.
15. Dam, B., Rector, J., Chang, M. F., Kars, S., de Groot, D. G. and Griessen, R., Laser ablation threshold of $YBa_2Cu_3O_{7-x}$. *Appl. Phys. Lett.* 1994, **65**, 1581–1583.
16. Doughty, C., Fidikoglu, A. T. and Vencatesan, T., Steady state pulsed laser deposition target scanning for improved plume stability and reduced particle density. *Appl. Phys. Lett.* 1995, **66**, 1276–1278.
17. Andreouli, C. and Tsetsekou, A., Synthesis of HTSC $Re(Y)Ba_2Cu_3O_x$ powders: the role of ionic radius. *Physica C* 1997, **291**, 274–286.
18. Andreouli, C. and Tsetsekou, A., Processing effect on microstructure and superconducting properties of sintered $ReBa_2Cu_3O_y$ ceramics—the role of ionic radius. *J. Eur. Ceram. Soc.* 2000, **20**, 2101–2114.
19. Moraitakis, E., Pissas, M. and Niarchos, D., Modelling of the hysteresis loop for $YBa_2Cu_3O_7$ thin films. *Physica C* 1995, **241**, 63–70.
20. Kusumori, T. and Muto, H., Influence of target composition on the quality of YBCO films fabricated by Nd:YAG pulsed laser deposition. *Physica C* 2001, **351**, 227–244.
21. Lee, S. G., Hwang, D. S., Park, Y. K. and Park, J. C., Deposition angle-dependent morphology of laser deposited $YBa_2Cu_3O_7$ thin films. *Appl. Phys. Lett.* 1994, **65**, 764–766.
22. Singh, R. and Kumar, D., Pulsed laser deposition and characterization of high-T_c $YBa_2Cu_3O_{7-x}$ superconducting thin films. *Mater. Sci. Eng.* 1998, **R22**, 113–185.