

# The role of Ag in the pulsed laser growth of YBCO thin films

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We have studied systematically the role of silver in improving microstructure and properties of  $Y_1Ba_2Cu_3O_{7-\delta}$  (YBCO) thin films. We have more than doubled the grain size to nearly  $1.8 \mu\text{m}$  and reduced processing temperatures by incorporating Ag in the YBCO films, which is accomplished by using a composite target containing 15% by weight of Ag. These films show approximately four times higher  $J_c$  than the best films obtained on MgO(001) substrates deposited from stoichiometric  $Y_1Ba_2Cu_3O_{7-\delta}$  targets. Study of the silver content in the film as a function of the deposition temperature shows clearly a decreasing concentration with increasing temperature and a segregation of the Ag to the surface. The increased oxygen content in the films is also observed at lower processing temperatures, providing strong support for the efficient oxygenation of YBCO via the presence of silver. A qualitative model suggests that the formation of silver oxide, rapid surface diffusion of Ag on MgO surfaces, and the nonreactivity of Ag with YBCO are the key aspects to the improvement in microstructure. The possibility of extending these ideas to the growth of oxides is also discussed, along with the fabrication of *in-situ* superconducting-metal junctions with 3D geometries. © 1999 American Institute of Physics. [S0021-8979(99)00809-9]

## I. INTRODUCTION

Initial reports on the mixing of  $YBa_2Cu_3O_{7-\delta}$  (YBCO) with silver (Ag) addressed improvements in the mechanical properties of the brittle YBCO.<sup>1-3</sup> The composite formed between Ag (or its oxide) and YBCO was found to increase the flexural strength<sup>4</sup> as compared to the pure YBCO phase. Interestingly, it was also observed that small amounts of silver added to bulk YBCO increased the critical current density ( $J_c$ ) of the bulk. This increase in  $J_c$  was attributed partly to the effect of increased alignment of the YBCO grains along the  $c$  axis and partly to increased grain sizes.<sup>5</sup> While the continued addition of Ag increased the flexural strength of the composite, the current density was found to decrease. This decrease in  $J_c$  for large Ag concentrations was viewed to occur because of decoupling of the grains. In essence the Ag (or Ag-O) resides at the grain boundaries and gives rise to weakly coupled SNS or SIS junctions. This reduces the current density, but at the same time provides a possible means to fabricate the junctions. There are a number of important conclusions which can be drawn from the previous studies on Ag+YBCO bulk composites. It was verified that there was no chemical interaction between Ag and YBCO. Second, Ag was not incorporated into the YBCO lattice, an indirect consequence of the lack of chemical reactivity. While these features do not rule out the possible substitution of Ag into lattice sites by nonequilibrium processes like ion implantation, it clearly indicated that the superconducting properties of the composite were not being influenced by interaction of the electronic states of YBCO and Ag. This also indicates that Ag would have to reside at intergranular

boundaries and porosities in the bulk material; regions of lower density required to accommodate relatively large Ag atoms.

The bulk Ag+YBCO composites were subsequently used as targets to grow thin films of YBCO via pulsed laser deposition (PLD). While the mechanical strength of the YBCO thin films was not the motivating factor, the primary goal was to obtain an increase in the critical current densities ( $J_c$ ) of these films. A number of researchers<sup>6-10</sup> have confirmed that the films grown by PLD from these composite targets show enhancement in  $J_c$ . The existing literature emphasizes the role of Ag in improving the growth characteristics of the films and in providing extra oxygen to the growing film.<sup>11-13</sup> In this work, while we provide experimental evidence to support some aspects of the model,<sup>14</sup> an interesting and important feature related to the use of Ag, not observed previously, has been provided. Further, the fact that Ag has not been observed to be distributed in the matrix of the films<sup>15,16</sup> has made it difficult to compare or even provide the analogy of its role in the bulk to explain the effects seen in the thin films of YBCO. The nonequilibrium processing via PLD and the complex processes of the various species and ambient gases within the laser plume make it interesting to study and compare the role of Ag in relation to the bulk or equilibrium processing cases. The improved microstructure at lowered processing temperatures provides reduced thermal budgets for higher current density YBCO films or MgO(001) and similar microstructural improvements can be expected in other oxides. An interesting aspect of the behavior of Ag in YBCO observed in this work also suggests its possible use in the *in-situ* fabrication of films having superconductor-normal (S-N) junctions with different orientations leading to a 3D architecture of Josephson junction devices suggesting a miniaturization analogous to existing semiconductor technology.

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## II. EXPERIMENTAL ASPECTS

The films grown from a stoichiometric YBCO target were compared with films deposited from targets containing various weight fractions of Ag under chosen experimental conditions. Using Auger spectroscopy (AES), transmission electron microscopy (TEM), x-ray diffraction (XRD), and electrical measurements for transition temperature ( $T_c$ ) and  $J_c$ , important results were obtained on the role of Ag. Combined with our earlier report on the optical emission spectroscopy study of the laser plume,<sup>13</sup> an understanding of efficient use of processing parameters was realized to process the high quality films on bare MgO(001) substrates. Targets of Ag+YBCO composites were prepared by mixing Ag<sub>2</sub>O and YBCO in appropriate quantities to give pellets containing 5, 15, and 25 wt % of Ag. These pellets were sintered at 900 °C in flowing oxygen for 12 h and were then oxygenated at 700 °C for 24 h. Pulsed laser deposition (PLD) was used to deposit films of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$</sub>  (YBCO) on MgO(001) substrates. The mechanically polished MgO substrates (obtained from Commercial Crystal Labs. Inc.) with optical finish were heat treated at 1100 °C for 12 h in flowing O<sub>2</sub> (100 cc/min) to remove the plastic damage due to mechanical polishing.<sup>17</sup> In some cases Si(001) substrates obtained from Virginia semiconductors were also used. Deposition on Si(001) surfaces included an *in-situ* deposited layer of yttria-stabilized zirconia (10 wt % Y<sub>2</sub>O<sub>3</sub>: 90 wt % ZrO<sub>2</sub>-YSZ) which acts as a buffer layer to prevent the degradation of the YBCO films due to interdiffusion of Si.<sup>18</sup> The films were deposited using a 248 nm KrF Lambda Physik excimer laser with a pulse width of 20 ns. The YBCO and YSZ films were grown from rotating targets (20 rpm) at energy densities of  $\sim 2$  J/cm<sup>2</sup>. The YSZ films were grown at substrate temperatures of 760 °C in oxygen partial pressures (PO<sub>2</sub>) of 0.05 mTorr while the YBCO films were grown at temperatures between 550–720 °C in a PO<sub>2</sub> of 200 mTorr. The films were cooled in 1 atm of O<sub>2</sub> at rates of 10 °C/min. The deposition rates of the films usually lie between 0.1–0.3 Å/pulse at a laser repetition rate of 5 Hz. Thickness of the order of 1000 Å were used for the YSZ buffer layers while the YBCO films were 1500–2000 Å thick (on MgO and Si). Current density measurements (using 50  $\mu$ V/cm criterion) were carried out on laser patterned bridges of width  $\sim 125$   $\mu$ m and length of 750  $\mu$ m. The energy density used for laser patterning in air was  $\sim 2$  J/cm<sup>2</sup>.

Conventional x-ray diffraction (XRD) using a Rigaku rotating anode x-ray generator with a Cu K $\alpha$  source was used for  $\theta$ -2 $\theta$  scan of the targets and films. Rutherford backscattering spectroscopy (RBS) measurements were made at Oak Ridge National Labs (ORNL) using 1.85 MeV <sup>4</sup>He<sup>+</sup> ions. The details of the setup for the measurement of optical spectra of the laser plume have been described elsewhere.<sup>13</sup> Auger electron spectroscopy was carried out using a JAMP model 30 spectrometer with an accelerating voltage of 5 kV. A TOPCON 002B 200 keV transmission electron microscope (TEM) was used for the microstructural characterization. TEM plan view samples were prepared by mechanical polishing and dimpling to about 20  $\mu$ m and subsequently ion milled at liquid N<sub>2</sub> temperatures in a GATAN Duo Ion Mill

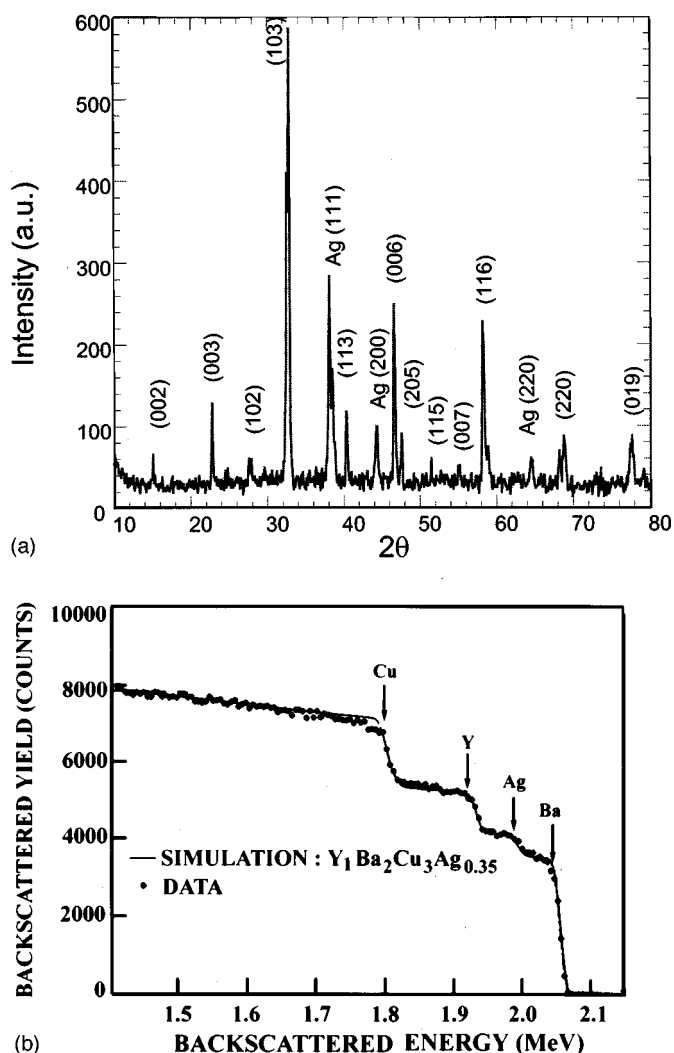


FIG. 1. (a) X-ray  $\theta$ -2 $\theta$  plot of 5 wt % Ag+YBCO composite target showing reflections corresponding to Y<sub>1</sub>Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$</sub>  phase of YBCO and metallic Ag, (b) RBS yield of 5 wt % Ag+YBCO target. From the simulation, the Ag concentration was determined to be 5.6 wt %.

using 5.5 keV Ar<sup>+</sup> ions at an angle of 16°. In this article we have variously used the terms composite target, composite film, etc., to describe films grown from a target of Ag+YBCO as compared to pure, undoped, etc., to mean film grown from a stoichiometric target of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$</sub> .

## III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show an x-ray ( $\theta$ -2 $\theta$ ) plot and an RBS spectrum, respectively, of a 5 wt % Ag composite target. All the visible XRD peaks correspond to the superconducting YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$</sub>  phase. The peaks corresponding to metallic silver are also visible. Visual inspection of the composite targets clearly showed some Ag segregation at the surface,<sup>5</sup> evidence of the difficulty in forming a solution between YBCO and Ag. The RBS spectrum in Fig. 1(b) was used to characterize the Ag content in the target and the value of 5.6% agrees quite well with the starting composition of 5 wt % Ag. A value of 14.4% was measured for the target initially mixed with 15% Ag. As seen later, the large vapor

pressure of Ag can also result in substantial reevaporation, leading to a lowered concentration of Ag in the target due to the annealing steps involved in the preparation of the targets. In this work we mainly present results on the transport properties and microstructure of films grown from the 15 wt% Ag composite target as this target was seen to give the best results. Useful information on the role of Ag was obtained by the other targets though, and wherever necessary those results have been used. Our results of the optical spectroscopy measurements of the laser plume during ablation have been reported earlier.<sup>13</sup> An important observation via optical spectroscopy of the plasma plume due to interaction of the laser pulse with the composite target is the presence of silver oxide species in the plume. We have also observed that the intensity of spectral emission from AgO lines decreases with decreasing oxygen pressure. A comparison between the bond strengths of AgO and O<sub>2</sub> reveals that AgO at room temperature has a strength of  $\sim 220$  kJ/mole while for O<sub>2</sub> it is  $\sim 498$  kJ/mole.<sup>19</sup> This suggests that the release of atomic "O" by dissociation of AgO is more likely than from O<sub>2</sub>.

The presence of reactive oxygen in the form of atomic "O" and the large oxygen requirement for YBCO results in a strong correlation between the oxygen source and film growth. This effect is confirmed by the fact that films from the pure and composite targets deposited at a relatively low substrate temperature of 585 °C on YSZ/Si(001) show widely different electrical behaviors. Figure 2(a) compares the temperature versus resistance plot for the films grown from a pure and 15 wt% composite target. Clearly the film from the pure target is insulating while the film from the composite target has a superconducting transition at  $\sim 64$  K. The  $\theta$ - $2\theta$  x-ray data [Fig. 2(b)] confirms that this difference is due to the fact that the film from the pure target is in the tetragonal nonsuperconducting phase, with an oxygen content  $\delta = 1.0$ , i.e., YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6</sub>, while the film from the composite target is orthorhombic with oxygen content  $\delta = 0.26$ , i.e., YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.84</sub>. The  $\delta$  values were calculated using the average  $c$  spacings obtained by the (001)-type peak positions in the diffraction pattern and substituted in the empirical formula proposed by Jorgensen *et al.*<sup>20</sup> This observation of enhanced oxygenation at lower growth temperatures is the first report of its kind. Earlier observations for increased oxygenation were based upon quenching experiments<sup>11</sup> where it was not possible to separate out the effects of incorporation via the lattice from suppression of oxygen outdiffusion via the grain boundaries. In our view, the observation on growth at lowered temperatures rules out the strong contribution related to the suppression of oxygen loss through grain boundaries. The fact that high background oxygen pressures ( $\sim 200$  mTorr) at substrate temperatures near 700 °C and subsequent slow cooling ( $\sim 10$  °C/min) in an atmosphere ( $\sim 760$  Torr) of oxygen can give films with  $T_c$  near 90 K, suggests that oxygen outdiffusion is not a problem under these annealing conditions. Thus, any "extra" oxygen is a result of AgO contribution of oxygen during growth.

The detection of Ag in the films has been a difficult task. The reason for this can be seen from the results of Auger spectroscopy. Films were grown at different substrate temperatures and the Ag signal from these films were measured

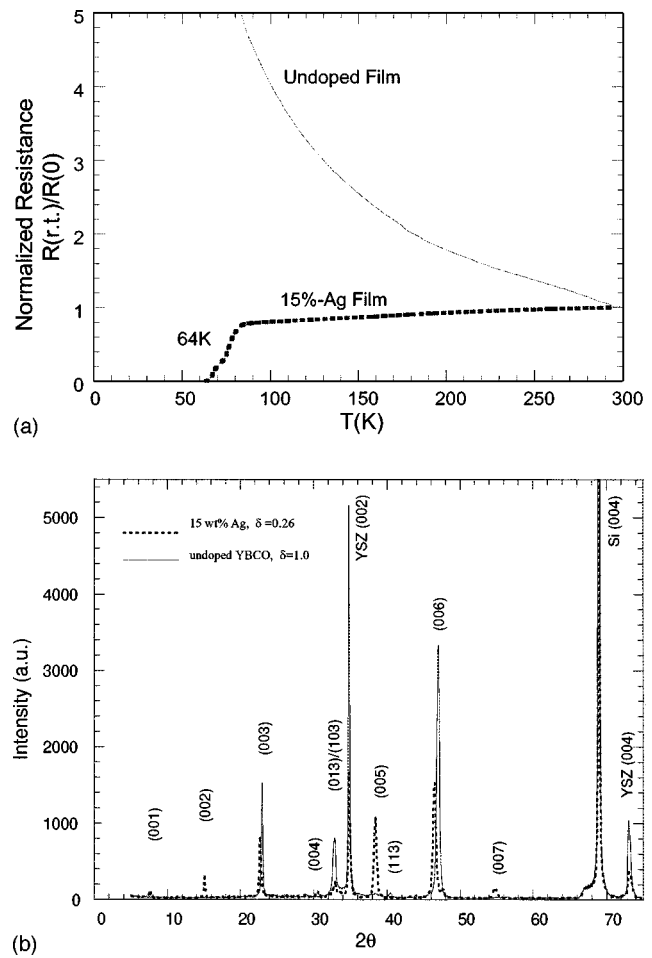


FIG. 2. (a) Comparative  $T_c$  plot of YBCO films on YSZ/Si(001) deposited at 585 °C. The top curve corresponds to the film from the pure target and is insulating, while the bottom corresponds to the film from 15 wt% Ag+YBCO target, and it has a  $T_c$  at 64 K (b) x-ray  $\theta$ - $2\theta$  plot comparing the above films clearly shows difference in phase (i.e., tetragonal vs orthorhombic) due to oxygen content difference.

by AES. Figure 3(a) compares the Auger spectra of films grown from a 25 wt% composite target as a function of the deposition temperature. As clearly seen, the intensity of the Ag peak at  $\sim 352$  eV decreases with increasing deposition temperature and at 675 °C it is below the detectable limits of AES. Sputter depth profiling of the film grown at 550 °C shows [Fig. 3(b)] that the Ag segregates to the surface of the film. This result is in agreement with previous results of Auger spectroscopy on YBCO films doped with Ag.<sup>15</sup> The diffusion constant of Ag in bulk YBCO has been estimated to be  $D = 0.01 \exp(-1.1/kT) \text{ cm}^2/\text{s}$ .<sup>21</sup> At a temperature of 650 °C the diffusion length ( $\sqrt{Dt}$ ) in 1 s is  $\sim 0.9 \mu\text{m}$ . Also, the vapor pressure of Ag at 650 °C is  $\sim 5$  mTorr and is even larger ( $\sim 30$  mTorr) at 700 °C. Further, it is now accepted that Ag is segregated to grain boundaries and other inhomogeneities in YBCO. Also, we have shown recently that majority of the grain boundaries in PLD films of YBCO/MgO(001) are pure [001]-tilt boundaries,<sup>22</sup> i.e., the boundary faces are almost normal to the substrate surface (i.e., MgO(001)). Thus, the large diffusion length of Ag will lead to segregation near the surface and film-substrate interface. Further, the large vapor pressure will cause reevaporation

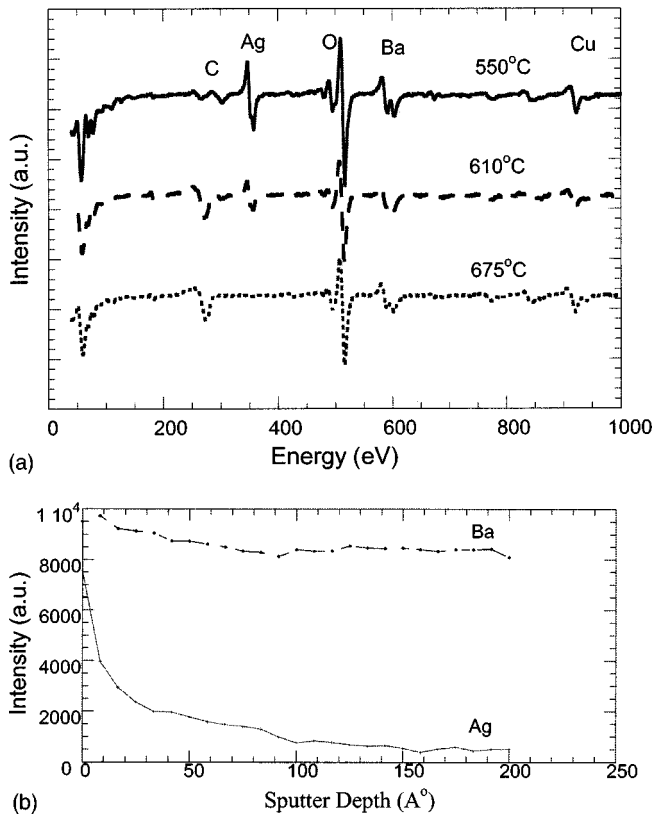


FIG. 3. (a) Auger electron spectra (AES) from YBCO films deposited at various temperatures showing decrease in the intensity of the Ag signal at  $\sim 352$  eV as growth temperature increases. (b) Segregation of Ag to surface is clearly seen in the sputter depth profile of Ag in the YBCO film deposited at  $550^\circ\text{C}$  from a 25 wt % Ag+YBCO composite target.

resulting in an enhanced gradient towards the surface. The evidence for Ag accumulation near the YBCO/MgO interface has been shown for YBCO films coevaporated with Ag grown by the DC sputtering technique.<sup>16</sup> Interestingly, this suggests that it is possible to control the Ag content in the

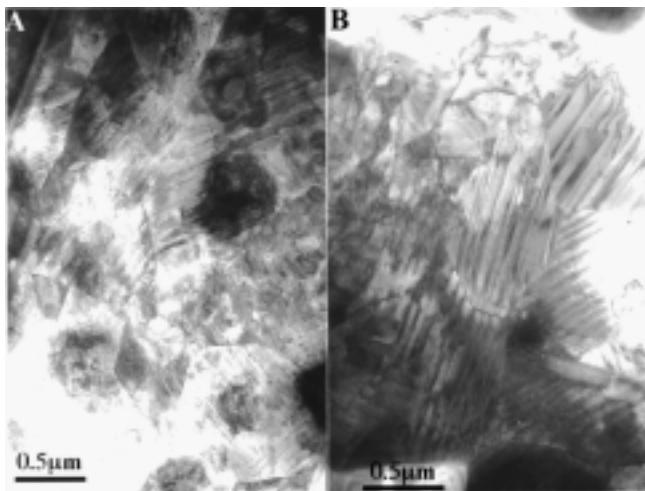


FIG. 4. Plan-view TEM from YBCO/MgO(001) films deposited at  $720^\circ\text{C}$ . (a) Film deposited from a pure YBCO target has average grain size of  $\sim 0.73 \mu\text{m}$  and  $J_c$  of  $1.02 \times 10^6$  amps/cm<sup>2</sup> at 77 K. (b) Film deposited from a 15 wt % composite target has average grain size of  $\sim 0.97 \mu\text{m}$  and  $J_c \sim 1.6 \times 10^6$  amps/cm<sup>2</sup>.

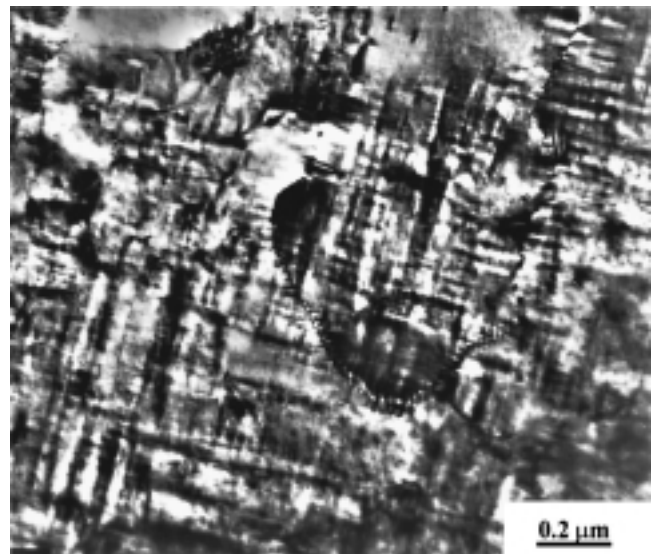


FIG. 5. TEM image of YBCO/MgO(001) grown at  $650^\circ\text{C}$  from composite target. The average grain size measured for this film was  $\sim 1.8 \mu\text{m}$  and the  $J_c$  was  $3.8 \times 10^6$  amps/cm<sup>2</sup> at 77 K.

grain boundaries as a function of growth temperature, film thickness, and growth rate. This possibility will be discussed later in this article.

Plan view study of YBCO films on MgO(001) substrates was carried out to investigate the relationship between growth variables (substrate temperature and target silver content), microstructure, and observed  $J_c$  values. In general, films grown from the 15% composite target showed larger grain sizes than those deposited from the pure target. Figures 4(a) and 4(b) show low magnification micrographs of the YBCO films on MgO(001) from a pure and 15 wt % composite target deposited at substrate temperatures of  $\sim 720^\circ\text{C}$ . In this case the grain boundary sizes are not vastly different, i.e., are  $0.73$  and  $0.9 \mu\text{m}$  for the pure and composite films, respectively. The  $J_c$  values of these films were also comparable,  $\sim 1.02 \times 10^6$  amps/cm<sup>2</sup> at 77 K (zero field) for the pure films and only slightly higher ( $1.09 \times 10^6$ ) for the composite film. As compared to this, Figs. 5 and 6 show micrographs of YBCO films grown from the 15 wt % composite target at temperatures of  $650^\circ\text{C}$  and  $600^\circ\text{C}$ , respectively. Figure 5 shows very few grain boundaries, and the grain size measured for this film was  $\sim 1.8 \mu\text{m}$ , with a measured  $J_c$  of

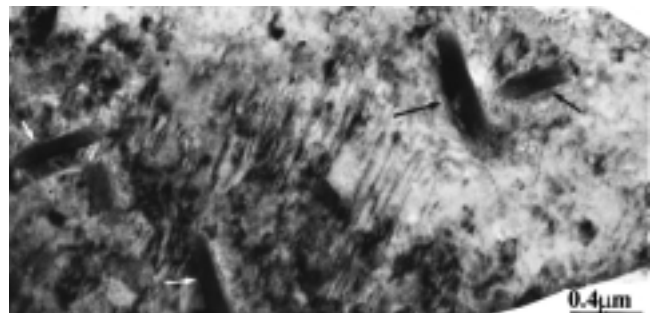


FIG. 6. TEM image of YBCO film deposited at  $600^\circ\text{C}$ . While the average grain size is still relatively large ( $\sim 1.8 \mu\text{m}$ ) the  $a$ -axis oriented grains reduce  $J_c$  ( $9.1 \times 10^4$  amps/cm<sup>2</sup>).

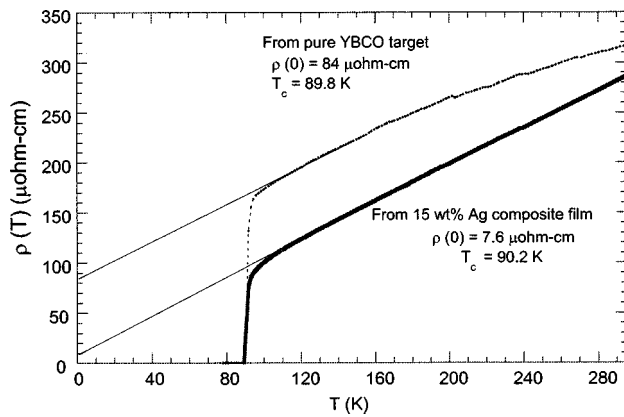


FIG. 7.  $T_c$  for YBCO films grown from pure and composite targets. The lower curve corresponds to the YBCO film grown at 650 °C from the 15 wt % composite target and has a residual resistivity,  $\rho(0)$  of  $\sim 7.6 \Omega \text{ cm}$ , while the top curve corresponds to a YBCO film grown from a pure target at 720 °C and has  $\rho(0)$  of  $\sim 84 \Omega \text{ cm}$ .

$\sim 3.8 \times 10^6 \text{ amps/cm}^2$ , which is better than optimized films obtained from pure targets on MgO(001) substrates, usually at temperatures between 700 °C–720 °C. For the 600 °C film shown in Fig. 6, while the grain size is similar or larger than the previous film, it has a number of  $a$  axis oriented grains in it. The  $J_c$  measured for this film was low,  $\sim 9.1 \times 10^4 \text{ amps/cm}^2$  consistent with the lowered current allowed in the  $c$  direction. The films grown at these lowered temperatures from the pure targets invariably showed lower  $J_c$ s than the films from the composite target and also lowered  $J_c$ s than those from films grown near 720 °C. This corresponded with the observation that the grain boundary size was found to decrease slightly ( $\sim 0.5 \mu\text{m}$ ) for films grown at 650 °C as compared to the case for the films from the 15% Ag target.

The influence on  $T_c$  was also investigated and Fig. 7 shows the resistance versus temperature behavior of the films grown at 650 °C (15% Ag,  $J_c \sim 3.8 \times 10^6 \text{ amps/cm}^2$ ) and 720 °C (pure target,  $J_c \sim 1.02 \times 10^6 \text{ amps/cm}^2$ ). The  $T_c$  values for the two films show no appreciable difference, both have transitions between 88–90 K and  $\Delta T$ 's of  $\sim 1.5 \text{ K}$ . Resistivity,  $\rho(T)$ , measurements over a number of samples gave room temperature values,  $\rho(\text{RT})$ , in the range of 280–510  $\mu\Omega \text{ cm}$ , and no particular correlation between the films grown from the composite and the pure target was found. For the case of Fig. 7 the films have almost similar resistivities, i.e., the composite film has a resistivity of 287  $\mu\Omega \text{ cm}$  and the pure film had  $\rho = 317 \mu\Omega \text{ cm}$  close to room temperature. But, a clear difference was observed in the values of extrapolated  $\rho$  at 0 K, i.e., calculated by extrapolating the normal state  $\rho(T)$ , for  $T > T_c$  to the abscissa. In this work the extrapolated  $\rho(0)$  values were consistently found to be lower for the films grown from the composite target than those from a pure target, as seen in Fig. 7. While  $\rho(\text{RT})$  is an important factor in calculating  $\rho(0)$ , the slope of the normal state resistance is also important. In Fig. 7, the pure film has a smaller rate of decrease in resistivity with temperature as compared to the composite film. Both these factors, i.e.,  $\rho(0)$  and the slope, provide information on the defect density in the films which contribute to charge carrier scattering.<sup>23</sup> The

results in this work indicate that the composite films contain lower defect densities than the pure films. One reason can be traced back to the microstructure. The grain sizes of the composite films are invariably larger than those of the pure films grown under identical conditions of substrate temperature, growth rates, etc. Therefore, lowering the number of grain boundaries lowered the resistivity and, as already mentioned, influenced the  $J_c$ . Recent measurements of grain boundary resistance via  $J_c$  by Selvam *et al.*,<sup>24</sup> on YBCO films grown from composite targets show evidence for increased conductivity across the grain boundaries in these films. This will also reduce the grain boundary resistance and hence decrease the residual resistivity.

Earlier work was concerned with the microstructure of the films as a function of the silver concentration in the targets.<sup>5,14,16</sup> Our observation of increase in grain size with reduced processing temperatures is a new and important observation as it provides direct evidence for the improved transport properties of YBCO films. From the known clustering behavior of Ag due to its rapid surface diffusion on various surfaces,<sup>25–27</sup> this is not an unexpected result. Lowering the temperature increases the residence time for Ag on MgO resulting in clusters of larger sizes. The direct effect is a lowering in the density of YBCO grains nucleating on MgO, resulting in grain growth to a larger spatial extent. Since this behavior is dependent only on the wetting properties of Ag and the substrate, the effect is of a general nature and can be made use of in the growth of other complex oxides. While reducing the temperature to 600 °C maintained the large grain size of YBCO, it did not prevent the nucleation of  $a$  axis oriented grains, suggesting that silver does not have a strong influence on the growth anisotropy of YBCO. The earlier reports on improved texturing of the YBCO grains has also been indirectly observed by us because of the fact that larger grain sizes will tend to reduce the amount of in-plane misorientation thus improving texture.

#### IV. CONCLUSION

While the role of silver in improving the quality of YBCO films is well accepted, its influence on the microstructure as a function of the substrate temperature has been relatively unknown. We have shown that the efficient use of Ag is related to a proper choice of the substrate temperature and, in fact, lowering the temperature improves the microstructure of the film by increasing the grain size. We have more than doubled the grain size (from 0.73 to 1.8  $\mu\text{m}$ ) by reducing processing temperatures (from 720 °C to 650 °C) and increased the  $J_c$  almost four times for these films. This is a very interesting result not only from the viewpoint of lowered thermal budgets but also because it provides the possibility of controlling the Ag content in the films. In the introduction, the possibility of 3D architecture of S-N junctions was mentioned. A qualitative approach to an *in-situ* growth of such structures can be visualized by considering the well-known facts of Ag in YBCO, i.e., it exists only at grain boundaries, voids, interfaces, etc. Thus, suitably chosen substrate temperature, film growth rate, and background silver pressure can be combined to segregate Ag to the film–film

interface (i.e., a doped YBCO film deposited on a single crystal YBCO film to form a thin metal interlayer), to remain within a grain boundary normal to the substrate giving an out of plane S-N junction, or to diffuse to the surface to form the contacts to the YBCO films. Our own work combined with the evidence of Moshfegh *et al.*<sup>16</sup> on interfacial silver and the work of Selvan *et al.*<sup>24</sup> on the increased conductivity across grain boundaries provide preliminary experimental evidence that this novel application could become a reality. Also, the use of silver is not restricted to YBCO as any oxide which is nonreactive with Ag can be deposited at lowered temperatures on surfaces which allow Ag to agglomerate rapidly. In fact, we have used this property to grow oxides with giant magnetoresistive properties at lowered temperatures.<sup>28</sup> In conclusion, the enhanced incorporation of oxygen via the formation of silver oxides in the plume and the rapid surface diffusion of Ag on MgO surfaces can be used to explain the observed improvement in properties of YBCO films grown from targets containing silver.

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<sup>1</sup>S. Jin, R. C. Sherwood, T. H. Tiefel, R. B. van Dover, and D. W. Johnson, Jr., *Appl. Phys. Lett.* **51**, 203 (1987).

<sup>2</sup>T. H. Tiefel, S. Jin, R. C. Sherwood, R. B. van Dover, R. S. Fastnacht, M. E. David, D. W. Johnson, Jr., and W. W. Rhodes, *J. Appl. Phys.* **64**, 5896 (1988).

<sup>3</sup>D. T. Shaw and S. Jin, in *Processing and Properties of High-Tc Superconductors*, edited by S. Jin (World Scientific, Singapore, 1993), p. 87.

<sup>4</sup>T. P. Sheahen, in *Introduction to High-Temperature Superconductivity* (Plenum, NY, 1994), pp. 203–222.

<sup>5</sup>U. De, S. Natarajan, and E. W. Seibt, *Physica C* **183**, 83 (1991).

<sup>6</sup>B. Oh, K. Char, A. D. Kent, M. Saito, M. R. Beasley, T. H. Geballe, R. H. Hammond, and A. Kapitulnik, *Phys. Rev. B* **37**, 7861 (1988).

<sup>7</sup>T. Kumagai, T. Manabe, T. Tsunoda, W. Kondo, and S. Mizuta, *Jpn. J. Appl. Phys.* **30**, L1268 (1991).

<sup>8</sup>R. K. Singh, D. Bhattacharya, P. Tiwari, J. Narayan, and C. B. Lee, *Appl. Phys. Lett.* **60**, 255 (1992).

<sup>9</sup>T. Clausen, M. Ejmaes, M. Olesen, K. Hilger, J. L. Skov, P. Bodin, A. Kuhle, and I. Chorkendorff, *Appl. Phys. Lett.* **65**, 2350 (1994).

<sup>10</sup>D. Kumar, P. R. Apte, R. Pinto, M. Sharon, and L. C. Gupta, *J. Electrochem. Soc.* **141**, 1611 (1994).

<sup>11</sup>D. Kumar, M. Sharon, P. R. Apte, R. Pinto, S. P. Pai, S. C. Purandare, C. P. D'Souza, L. C. Gupta, and R. Vijayaraghavan, *J. Appl. Phys.* **76**, 1349 (1994).

<sup>12</sup>R. Pinto, P. R. Apte, K. P. Adhi, S. B. Ogale, D. Kumar, and M. S. Hegde, *J. Appl. Phys.* **78**, 5204 (1995).

<sup>13</sup>D. Kumar, S. Oktyabrsky, R. Kalyanaraman, J. Narayan, P. R. Apte, R. Pinto, S. S. Manoharan, M. S. Hegde, S. B. Ogale, and K. P. Adhi, *Mater. Sci. Eng.* **45**, 55 (1997).

<sup>14</sup>D. Kumar, P. R. Apte, and R. Pinto, *J. Appl. Phys.* **77**, 1 (1995).

<sup>15</sup>C. Hart, O. Ares, J. L. Pena, P. Bartolo-Perez, V. Sosa, W. Cauch, G. A. Hirata, L. Cota-Araiza, and M. H. Farias, *Appl. Phys. Lett.* **67**, 2078 (1995).

<sup>16</sup>A. Z. Moshfegh, Y. Q. Wang, Y. Y. Sun, A. Mesarwi, P. H. Hor, and A. Ignatiev, *Physica C* **218**, 396 (1993).

<sup>17</sup>M. G. Norton, S. R. Summerfelt, and C. B. Carter, *Appl. Phys. Lett.* **56**, 2246 (1990).

<sup>18</sup>D. K. Fork, D. B. Fenner, R. W. Barton, J. M. Phillips, G. A. N. Connell, J. B. Boyce, and T. H. Geballe, *Appl. Phys. Lett.* **57**, 1161 (1990).

<sup>19</sup>D. R. Lide, in *CRC Handbook of Chemistry and Physics*, 71st ed. (CRC, Boca Raton, FL, 1990), pp. 9–86.

<sup>20</sup>J. D. Jorgenson, B. W. Veal, A. P. Paulikas, L. J. Nowicki, G. W. Crabtree, H. Claus, and W. K. Kwok, *Phys. Rev. B* **41**, 1863 (1990).

<sup>21</sup>S. F. Gafarov, T. D. Dzhafarov, G. S. Kulikov, R. Sh. Malkovich, E. A. Skorytina, and V. P. Usacheva, *Sov. Tech. Phys. Lett.* **15**, 858 (1989).

<sup>22</sup>S. Oktyabrsky, R. Kalyanaraman, K. Jagannadham, and J. Narayan, *J. Mater. Res.* (to be published).

<sup>23</sup>C. Kittel, in *Introduction to Solid State Physics*, 5th ed. (Wiley Eastern Ltd., India, 1990), p. 171.

<sup>24</sup>P. Selvam, E. W. Seibt, D. Kumar, R. Pinto, and P. R. Apte, *Appl. Phys. Lett.* **71**, 137 (1997).

<sup>25</sup>N. J. Wu, A. Natori, and H. Yasunaga, *Surf. Sci.* **242**, 191 (1991).

<sup>26</sup>U. Kurpick, G. Meister, and A. Goldman, *Appl. Surf. Sci.* **89**, 383 (1995).

<sup>27</sup>R. Kalyanaraman, K. Jagannadham, A. K. Sharma, and J. Narayan, presented at Evolution of Surface Morphology and Thin Film Microstructure, MRS Symposium A, December 1997, Boston, MA.

<sup>28</sup>D. Kumar, R. Kalyanaraman, R. D. Vispute, J. Narayan, D. K. Christen, and C. E. Klabunde, *Mater. Sci. Eng.* **45**, 122 (1997).