AN ION PROBE STUDY OF PLASMA-ASSISTED LASER DEPOSITION

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ABSTRACT

The ionic content of an excimer laser ablated $YBa_2Cu_3O_{7-\delta}$ (YBCO) plume has been investigated by using an ion probe detection system. The time-of-flight ion signals indicate a bifurcation of the plume, where the degree of bifurcation is dependent on both the oxygen pressure and the laser spot size at the target. A significant enhancement in the ionic content is observed when the plume is passed through a high voltage discharging ring. The results describing the effect of the pressure, laser spot size and the discharge parameters on the propagating laser plume are presented in this paper.

INTRODUCTION

Ionic collisions at the substrate during vapor deposition of thin films have been shown to favorably influence the crystallinity by enhancing the surface adatom mobility [1,2]. In most of the ion-assisted deposition processes ion sources are incorporated to provide the necessary surface activation for crystalline film growth. Low energy (<100eV) ion bombardment during the film growth alters the growth kinetics and leads to film densification, preferred grain orientation, low temperature epitaxy and enhanced surface chemical reactions. In the laser-ablation-deposition of thin films, the laser-target interaction gives rise to a significant ion density in the evaporated material that aids the substrate surface activation [3,4]. In addition, the high photon energy of the excimer laser leads to a high degree of electronic excitation in the evaporated species. The high reactivity of the excited species promotes gas phase reaction with the ambient environment. For these reasons, the laser ablation method has been extremely successful in the growth of films, such as YBCO, that require gas phase reaction for incorporation into the structure [5-8].

For low temperature film growth, it is of interest to increase the ionic content of the laser plume, that would lead to enhanced surface activation in a biased substrate deposition. The generation of a gas discharge between the target and the substrate during the propagation of the laser plume has been shown to facilitate the growth of YBCO films at a lower substrate temperature [9,10]. This has been attributed to increased substrate activation and enhanced gas phase reaction.

Previous studies designed to yield information about the laser ablated plume in flight have primarily included in-situ optical diagnostics via the radiative component of the plume (using fluorescence spectroscopy [11] or ICCD imaging [12]) and ex-situ mass spectroscopic assessments of the plume content [13]. Since observation of the fluorescence restricts the analysis to a small subset of the entire plume and may not be truly representative of the overall plume dynamics, experiments have also investigated the non-emitting neutral species using absorption spectroscopy [14] and post-ablation laser-induced fluorescence [15]. Dyer et. al. have performed preliminary studies of the ionic species using ion-probe measurements [16]. Since the ionic content of the plume is crucial for in-situ deposition of superconducting thin films and in order to preserve the in-situ capability of the plume diagnostics, we have employed an ion-probe technique to measure both the total content and the temporal dynamics of the ions in the plume. In this paper, we present the results of an experimental study conducted to investigate the effect of a discharge on the propagating laser plume. Our results obtained with and without the inclusion of a pulsed plasma discharge during the deposition enable, for the first time, a comparative analysis of the role of the pulsed discharge on the ion dynamics.

EXPERIMENT

The experimental set-up is shown in Fig. 1. A rotating superconducting target placed in a vacuum chamber was ablated using focused excimer (KrF) laser pulses of 15 ns duration and 100 mJ energy. A 30 cm focal length lens was placed on a translation stage to allow variation of the laser spot size at the target. The laser generated plume was passed through a 2.5 cm diameter ring electrode located 2.5 cm from the target. A grounded shield with a 2.5 cm hole was placed about 2 cm from the ring, concentric with the axis passing through the center of the ring electrode and the point of incidence of the laser on the target. This shield acted as a terminating electrode for the discharge while eliminating probe noise due to space charge effects. The presence of the shield also improved the reproducibility of the discharge. The ion probe located along the same axis, 6.25 cm from the target, was a shielded 1 mm diameter single electrode that was biased at -100V. The small area of the probe minimized plume perturbation by the probe bias voltage.

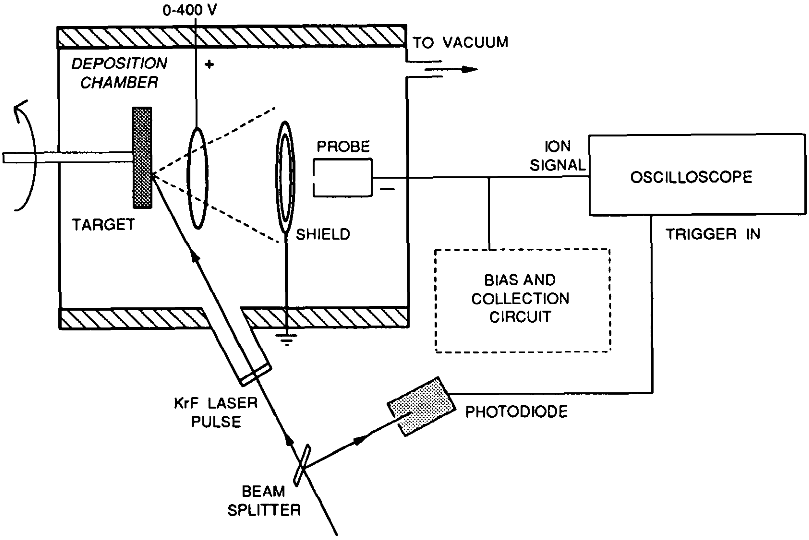


FIG. 1. Schematic diagram of the excimer laser ablation and ion detection system.

RESULTS AND DISCUSSION

The dynamic behavior of the ion content of the laser-ablated YBCO plume was assessed by recording the on-axis time-resolved ion signal. Fig.2 shows the temporal ion profile at a 20 mtorr ambient oxygen pressure and a laser spot size of 3.3 mm^2 at the target, corresponding to an incident laser fluence of 3.1 J/cm^2. Fig. 2(a), obtained in the absence of a voltage on the ring electrode exhibits a single peak with a modal velocity of 1.25 X 10^6 cm/s, consistent with previous optical measurements of the forward directed component of the laser-ablated plume [12]. The application of a 300V bias on the ring electrode shows the formation of a slower ionic component delayed by ~11 μs with respect to the initial peak (Fig. 2(b)). With increasing ring voltage, a further enhancement of the secondary peak is indicated in Fig. 2(c), obtained at a ring voltage of 450V. Quite clearly, the effect of the pulsed discharge caused by the ring voltage during the deposition is a selective enhancement of primarily the slower secondary forward-directed component of the plume.

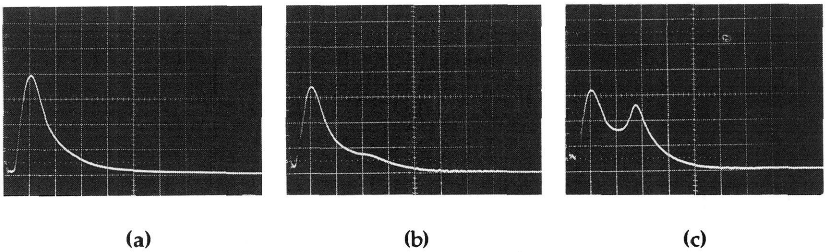
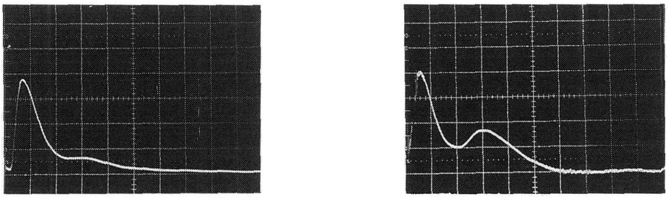

(a) (b) (c)

FIG. 2. Time-of-flight ion signals obtained at an ambient oxygen pressure of 20 mtorr for (a) 0V, (b) 300V and (c) 450V ring bias. The target-probe distance was 6.25 cm. Vertical scale : 0.5V/div.; Horizontal scale : 5μs/div..

The effect on the ion profile as a result of increasing the ambient oxygen pressure to 30 mtorr is recorded in Fig.3. In contrast to Fig. 2(a), its higher pressure counterpart (Fig. 3(a)) exhibits a slow component even in the absence of a ring voltage. As indicated in our previous publication [17], this observation is consistent with the interpretation of the slow component corresponding to a high collision regime more readily evident at higher pressures. As observed for the 20 mtorr data, the effect of the ring bias at the higher (30 mtorr) pressure is again an enhancement of the slower component.

The role of the pulsed discharge may be systematically analyzed by considering the percentage enhancement of the total ion content as a function of ring voltage. We obtained this enhancement by a comparative analysis of the integrated ion voltage with and without the pulsed discharge at various ring biases. The results are shown in Fig.4, at 10mtorr and 40 mtorr ambient pressures. The ionic enhancement as a function of pressure and ring bias are consistent with the increases observed in the time-resolved study. The possibility of significant enhancement (above 100%) using a suitable choice of pressure and ring bias is evident.



(a) (b)

FIG. 3. Time-of-flight ion signals obtained at an ambient oxygen pressure of 30 mtorr for (a) 0V and (b) 450V ring bias. Vertical scale : 0.2V/div.; Horizontal scale : 5 μs/div.. The target-probe distance was 6.25 cm.

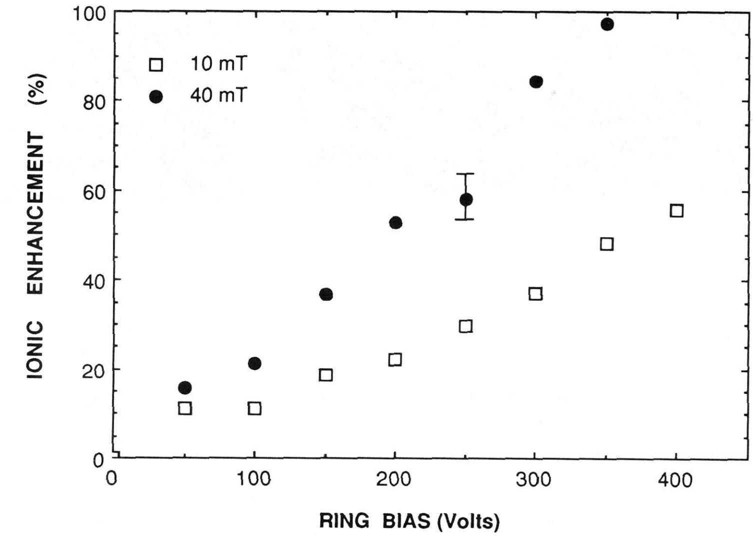


FIG. 4. Ionic enhancement as a function of ring bias at ambient oxygen pressures of 10 mtorr and 40 mtorr respectively. The target-probe distance was 7.5 cm.

An interesting observation, reported by us elsewhere [18], revealed the effect of modifying the laser spot size on the target as a means of affecting the dynamic behavior of the propagating plume. An increase in the laser spot size contributes to augmentation of the secondary ionic peak due to an intra-plume pressure gradient driven expansion process. This effect may be used to cause a further increase in the secondary ions in the presence of a pulsed plasma. This is shown in Fig.5, obtained at a defocused laser spot size of 4.4 mm^2 on the target at an ambient pressure of 30 mtorr. Two observations are of interest. Firstly, Fig.

5(a), on comparison with its focused counterpart (Fig. 3(a)), shows the increased relative contribution of the secondary ion peak to the total ion content in the plume for the larger laser spot size, even in the absence of the plasma . Secondly, as anticipated, the presence of the pulsed discharge (shown in Fig. 5(b) for a 450V ring bias) causes a significant increase in the secondary peak. It is noteworthy that the secondary ion peak for the defocused case is larger than the primary peak in the presence of a 450V ring bias. This is in contrast to the behavior exhibited by the focused beam as revealed by a comparison of Figs. 3(b) and 5(b). It is therefore possible to combine the variation in laser spot size with changes in the ring bias to selectively enhance the secondary peak while leaving the primary peak essentially unaltered.

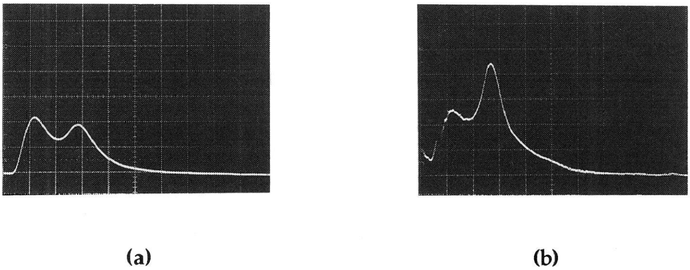


FIG. 5. Time-of-flight ion signals obtained at an ambient oxygen pressure of 30 mtorr using a defocused laser spot size of 4.4 mm^2 on the YBCO target for (a) 0V, and (b) 450V ring bias. Vertical scale : 0.2V/div.; Horizontal scale : 5 μs/div..

CONCLUSION

Recent experiments have pointed out the possibility of misinterpreting ion-probe results due to interference caused by secondary electron emission from the probe as a result of fast neutral bombardment [19,20]. It is important to emphasize that those experiments were conducted in vacuum (without the presence of an ambient) at higher plume species velocities. Also, there was no further ionization of the laser plume by a plasma as is the case in our experiments. The behavior of the slow component and its enhancement that we report in this paper are therefore not affected by the interference envisaged in Refs. 19 and 20. Additionally, the observed dependence of the ionic enhancement on ring voltage and ambient pressure (Fig. 4) are consistently explained only on the basis of a true ionic enhancement of the plume.

We have presented, in this paper, the results of dynamic ion probe measurements on the laser ablated YBCO plume, specifically to assess the effect of a pulsed discharge on the propagating plume. Our results show a significant enhancement in the secondary ion signal as a function of the voltage on the ring electrode as well as the ambient oxygen pressure. Further, the use of a defocused laser spot on the target has been shown to yield a unique capability of selectively enhancing the secondary ions. This should have important implications for the deposition of in-situ superconducting films on a low temperature substrate. The precise deposition characteristics using the ionic enhancement outlined in this paper is currently under investigation.

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REFERENCES

1. I. Takagi, Thin Solid Films, **92**, 1 (1982).
2. J. E. Green, CRC Critical Reviews of Solid State and Materials Science **11**, 47 (1983).
3. H. Sankur, J. DeNatale, W. Gunning, and J. G. Nelson, J. Vac. Sci. Technol. **A5**, 2869 (1987).
4. R. E. Walkup, J. M. Jasiske, R. W. Dreyfus, Appl. Phys. Letts. **48**, 1960 (1986).
5. T. Venkatesan, A. Inam, B. Dutta, R. Ramesh, M. S. Hegde, X. D. Wu, L. Nazar, C. C. Chang, J. B. Barner, D. M. Hwang and C. T. Rogers, Appl. Phys. Lett. **56**, 391 (1990).
6. R. K. Singh, J. Narayan, A. K. Singh and J. Krishnaswamy, Appl. Phys. Lett. **54**, 2271 (1989).
7. P. K. Fork, F. A. Ponce, J. C. Tramontana, N. Newman, J. M. Phillips, and T. H. Gaballe, Appl. Phys. Lett. **58**, 2432 (1991).
8. G. Koren, A. Gupta, E. A. Giess, A. Segmuller and R. B. Laibowitz, Appl. Phys. Lett. **54**, 1054 (1989).
9. S. Witanachchi, H. S. Kwok, X. W. Wang and D. T. Shaw, Appl. Phys. Lett. **53**, 234 (1988).
10. J. P. Zheng, Q. Y. Ying, S. Witanachchi, Z. Q. Huang, and D. T. Shaw, Appl. Phys. Lett. **54**, 954 (1989).
11. W. A. Weimer, Appl. Phys. Lett. **52**, 2171 (1988).
12. D. B. Geohegan, Appl. Phys. Lett. **60**, 2732 (1992).
13. T. Venkatesan, X. D. Wu, A. Inam, Y. Jeon, M. Croft, E. W. Chase, C. C. Chang, J. B. Wachtman, R. W. Odom, F. R. di Brozolo and C. A. Magee, Appl. Phys. Lett. **53**, 1431 (1988).
14. D. B. Geohegan and D. N. Mashburn, Appl. Phys. Lett. **55**, 2345 (1989).
15. T. Okada, N. Shibamaru, Y. Nakayama, and M. Maeda, Appl. Phys. Lett. **60**, 941 (1992).
16. P. E. Dyer, R. D. Greenough, A. Issa and P. H. Key, Appl. Phys. Lett. **53**, 534 (1988).
17. S. Witanachchi, K. Ahmed, P. Sakthivel and P. Mukherjee, to appear in the Proceedings of the Conference on Superconductivity and Applications, NYSIS, 1992.
18. S. Witanachchi, K. Ahmed, P. Sakthivel and P. Mukherjee, submitted for publication.
19. S. H. Brongersma, J. C. S. Kools, T. S. Baller, H. C. W. Beijerinck and J. Dieleman, Appl. Phys. Lett. **59**, 1311 (1991).
20. J. C. S. Kools, S. H. Brongersma, E. van de Riet and J. Dieleman, Applied Physics B, **53**, 125 (1991).