

OPTICAL DETECTION OF SLOW EXCITED NEUTRALS IN PLASMA-ASSISTED EXCIMER LASER ABLATION

P. MUKHERJEE, P. SAKTHIVEL AND S. WITANACHCHI
Department of Physics, University of South Florida, Tampa, FL 33620,
prritish@chuma.cas.usf.edu

ABSTRACT

The observation of a slow optically excited component in the excimer laser-ablated YBCO plume due to the presence of a biased ring electrode is reported. The temporal dynamics of the plume were investigated by using time-of-flight (TOF) optical emission spectroscopy (OES). Time-resolved emission signals reveal excitation and resultant fluorescence from slow-moving plume species in the presence of the discharge.

INTRODUCTION

The possibility of lowering substrate temperatures in the growth of YBCO films using pulsed laser deposition (PLD) due to the presence of an *in-situ* biased ring electrode has been demonstrated before [1,2,3]. Ion-probe investigations of the dynamics of the plume in the presence of the biased ring have shown enhanced ionization as well as selective enhancement of the slow moving ions in a temporally bifurcated plume [4]. Ion bombardment during film growth has been shown to enhance film crystallinity by increasing surface adatom mobility and surface chemical reactions [5,6].

Ionization of the plume can be enhanced by increasing the laser fluence [7] but this also increases the particulate density on the surface of the film [8]. However, the generation of a gas discharge between the laser and substrate collisionally excites and ionizes plume species, while not increasing particulate density. As a result of collisional excitation and ionization, gas-phase reaction as well as reactions at the substrate are enhanced, thereby leading to the stoichiometric deposition required for *in-situ* superconducting YBCO. In this paper, we present a comparison of the results of optical fluorescence measurements on the KrF laser ablated plume in the presence of a pulsed discharge to that without the discharge.

EXPERIMENT

A schematic diagram of the experimental set-up is shown in Fig. 1. A 30ns, 100mJ excimer laser (KrF) pulse was used for ablation. The laser generated plume from a rotating YBCO target passed through a 2.5 cm diameter ring electrode, located 2.5 cm from the target, and centered about the axis of the plume. A grounded shield with the same diameter was placed about 2 cm from the ring electrode, concentric with the axis of the ring electrode. This shield acted as a terminating electrode for the discharge and also improved the reproducibility of the discharge. A biased ion probe was located along the plume axis, 7.5 cm from the target. The ion probe is a 3mm diameter single shielded electrode that is biased at -70V to -100V and can collect ions in time-integrated or time-resolved modes. The small area of the probe minimizes plume perturbation by the probe bias voltage.

An EG & G plasma monitoring system was used to view the plume from the side for OES. It included an optical multichannel analyzer (OMA) capable of a wavelength resolution better than 1Å and a temporal resolution of 100ns. The light from the plume was reflected and focused into the viewing area. A fiber optic cable, with an aperture of diameter 1.2mm was translated in the image plane to sample different portions of the plume.

Temporal synchronization of the data acquisition system to the excimer laser pulse was achieved electronically. The time-of-arrival of the laser pulse on the target was confirmed by placing the fiber at the point of intersection of the laser and target (on the image plane) and by varying the delay at which the OMA was triggered at the minimum gate-width (100ns) until a signal was received. Once the time origin was established, the detection delay could be suitably adjusted as needed, to provide temporal windowing of the plume signals. A gate-width of 400ns

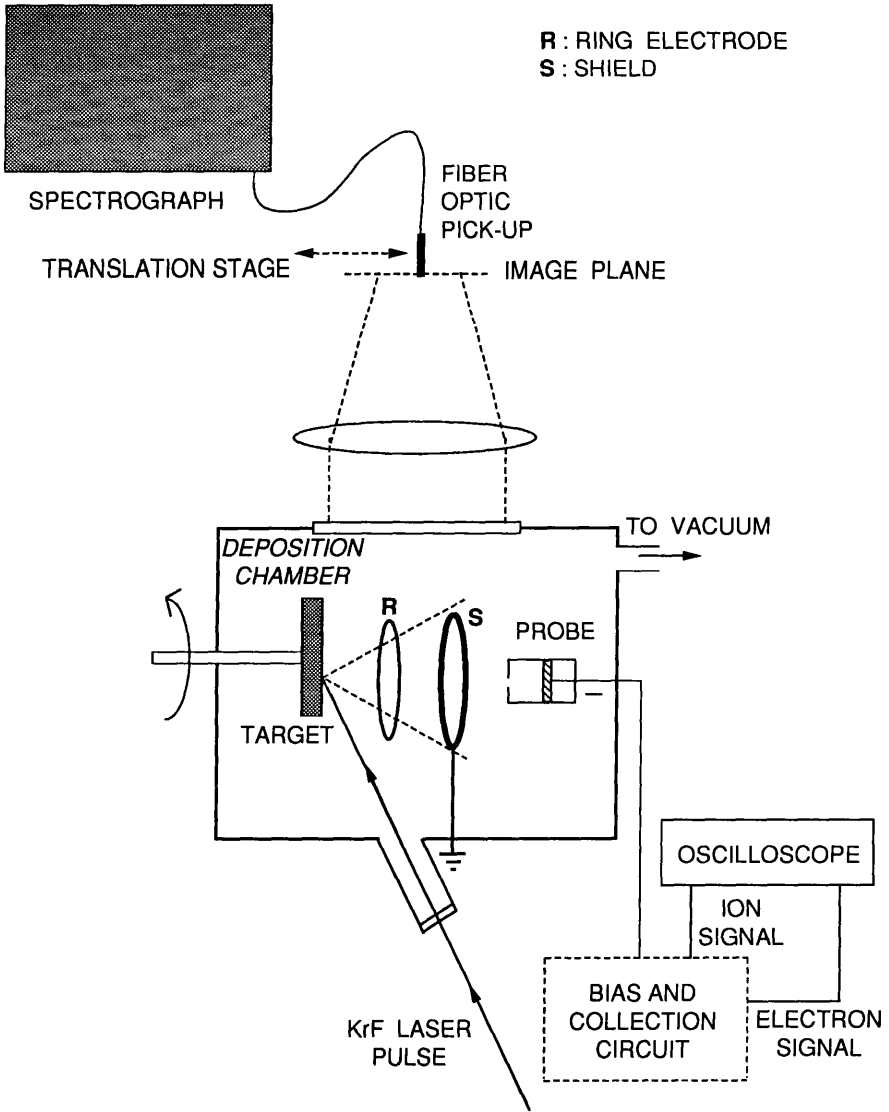


FIGURE 1. SCHEMATIC OF LASER ABLATION SETUP SHOWING RING ELECTRODE AND DIAGNOSTIC CAPABILITY

was used in our experiment. For each data point, 100 spectra were obtained and averaged to enhance the signal-to-noise ratio.

RESULTS AND DISCUSSION

Previous measurements of optical fluorescence from laser ablated plumes have not shown the bimodal velocity distribution observed in time-of-flight(TOF) absorption profiles[7] and laser induced fluorescence [9]. In the present experiments, we investigated the temporal dynamics of the laser ablated plume in the presence of a plasma discharge through TOF emission profiles at different distances from the target along the plume-axis.

As shown in figure 2, TOF profiles obtained for a Ba transition(553.55nm) at 2cm from the target in the presence of the plasma discharge show a temporal bifurcation of the Ba atomic emission into a 'fast' component and a 'slow' component. The slow component has a velocity that is approximately a factor of four less than that of the fast component. In the absence of the discharge, however, it can be seen that there is no fluorescent slow component. The fast component is unaffected by the discharge.

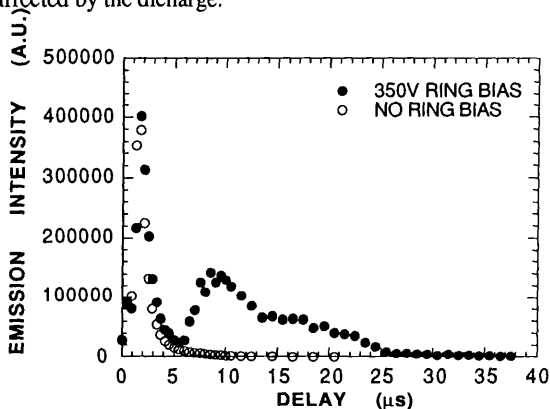


FIGURE 2. TOF EMISSION SIGNALS AT 553.55nm FROM NEUTRAL Ba ATOMS 2cm FROM YBCO TARGET. The ambient oxygen pressure was 40mt.

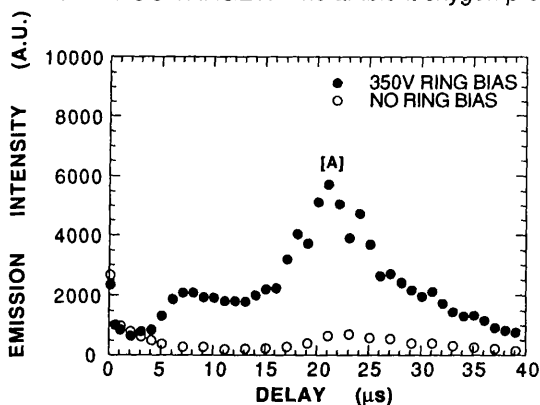


FIGURE 3. TOF EMISSION SIGNALS at 553.55nm FROM NEUTRAL Ba ATOMS 7cm FROM YBCO TARGET. The ambient oxygen pressure was 40mt.

At 7cm from the target, which is the position of the substrate, there is again a fast and a slow component, in the presence of the plasma discharge. A greater spread in the TOF distributions is observed, compared to the profiles at 2cm, as seen in figure 3. Unlike data obtained previously using the ion probe in the absence of the plasma discharge[8], however, there is an increase in velocity of the slow component as a function of distance from the target. This may be due to intra-plume collisions with fast moving ionic species in the plume that are accelerated by the electric field. This enhances the forward-directed motion for all plume species including the atomic species. Spectra obtained at point 'A' (at a delay of $\sim 22\mu\text{s}$) indicated in figure 3 in the presence and absence of the biased ring electrode are shown in figure 4. A clear enhancement in the excitation of atomic and ionic species in the plume in the presence of the ring bias is evident.

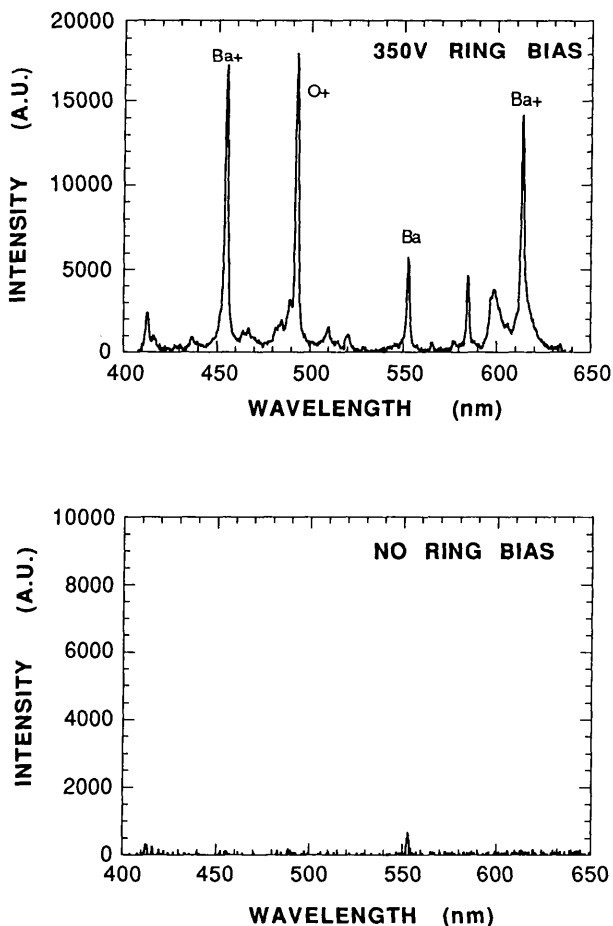


FIGURE 4. OPTICAL EMISSION SPECTRA FROM YBCO PLUME 7cm FROM TARGET SHOWING THE EFFECT OF THE BIASED RING.

A comparison of TOF profiles of Y and Cu transitions showed identical behavior. A similar enhancement in the optical excitation of O_2^+ was also obtained in the presence of the ring bias. This supports enhanced ionization of oxygen in the plume, thereby resolving a controversy in previously published work [2,10].

The observed behavior is also very similar to the non-species resolved ion probe TOF profiles, an example of which is shown in figure 5 in the presence of the plasma discharge. The fast and the slow component of the ion probe profile in the presence of the discharge have peaks that are slightly earlier than the peaks in the optical emission data for neutrals. This suggests a recombination of the ions leading to successive fluorescence of the atomic species.

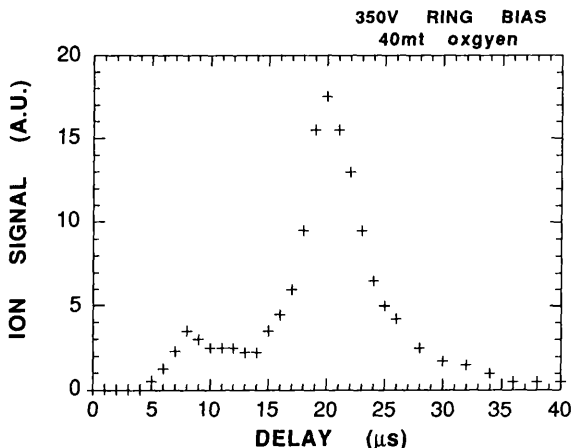


FIGURE 5. TOF ION SIGNAL AT 6cm FROM YBCO TARGET IN THE PRESENCE OF BIASED RING.

CONCLUSIONS

The observation of slow excited neutrals is possible due to their production by recombining ions in the discharge. TOF emission profiles show a temporal bifurcation. Unlike previous ion probe measurements, the slower bifurcated component exhibits acceleration as it propagates. The velocity of the fast component is unaffected. An increase in plasma temperature beyond the grounding electrode in the presence of the discharge has been previously observed[11] and was confirmed. The lowering of substrate temperatures for in-situ superconducting film deposition in the presence of a pulsed discharge during the deposition is attributed to both enhanced ionic excitation and acceleration of the slower neutrals in the plume. The possibility of using an in-situ low-voltage discharge to excite previously non-radiating neutrals thus permitting their detection using standard emission techniques is currently being explored.

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