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S. S. Harilal, P. Radhakrishnan, V. P. N. Nampoori, and C. P. G. Vallabhan

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Temporal and spatial evolution of laser ablated plasma from $YBa_2Cu_3O_7$

S. S. Harilal, P. Radhakrishnan, V. P. N. Nampoori, and C. P. G. Vallabhan
Laser Division, Department of Physics, Cochin University of Science and Technology, Cochin-682 022, India

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$YBa_2Cu_3O_7$ target was laser ablated, and the time-of-flight (TOF) distributions of Y, Y^+, and YO in the resultant plasma were investigated as functions of distance from the target and laser energy density using emission spectroscopy. Up to a short distance from the target ($\simeq$1.5 cm), TOF distributions show twin peaks for Y and YO, while only single-peak distribution is observed for Y^+. At greater distances (>1.5 cm) all of them exhibit single-peak distribution. The twin peaks are assigned to species corresponding to those generated directly/in the vicinity of target surface and to those generated from collisional/recombination process.

Since the successful deposition of ceramic materials like high T_c superconducting films using pulsed laser ablation,[1,2] there has been intensive interest in the studies on the composition and transport properties of laser-induced plasma (LIP) plume.[3–6] It is of fundamental importance to identify the different atomic, ionic, and molecular species generated from laser-induced plasma and to relate the quality and rate of thin film growth with species present and width various transport processes occurring in plasma plume. Some of the techniques reported by various authors for characterizing LIP from different targets include optical emission spectroscopy,[3–5] optical absorption spectroscopy,[6–8] and laser-induced fluorescence.[9] In spite of the large number of reports available in the literature, studies on LIP from solid targets have not yet yielded conclusive results and this is essentially due to the complexity of the phenomena involved. In this letter, we report some new results of time and space resolved analysis of the spectral emission due to Y, Y^+, and YO species in the LIP from high T_c superconductor $YBa_2Cu_3O_7$ (T_c=90 K, here after YBCO) using 1.06 μm radiation from a Nd:YAG laser.

The schematic diagram of the experimental setup is given elsewhere.[10] The YBCO target having a diameter of 15 mm and thickness of 3 mm was mounted in a stainless steel vacuum chamber ($\sim 10^{-3}$ Torr) equipped with quartz windows such that the target surface could be irradiated at normal incidence using 1.06 μm laser beam from a Q-switched Nd:YAG laser (Quanta ray DCR 11) having a pulse width of 9 ns and repetition rate of 10 Hz. To avoid errors due to local heating and drilling, the sample was rotated about an axis parallel to the laser beam. Laser fluence was varied by attenuating the laser beam. The plasma emission spectrum for the YBCO sample was viewed normal to its expansion direction and imaged using appropriate focusing lenses and apertures onto the slit of a monochromator (Jarrell–Ash 0.5 m) which is coupled to a photomultiplier tube (PMT) (S20 cathode). For spatially resolved studies, the plasma plume from different regions was focused on to the monochromator slit.[3] The characteristic lines were selected by the monochromator and the PMT output was fed to a 200 MHz digital storage oscilloscope (Iwatsu model DS 8621) with 50 Ω termination to record the emission pulse shapes. This setup essentially provides delay as well as decay times for emission from constituent species at a specific point within the plasma and these are extremely important parameters related to the evolution of laser ablated materials in a direction normal to the target surface.

The time-resolved studies of emission lines from various species were made from the oscilloscope traces which showed definite time delays for emission with respect to the laser pulse. Typical time-of-flight (TOF) distributions of Y (558.1 nm), Y^+ (508.47 nm), and YO (601.9 nm) at a distance 1 cm from the target are given in Fig. 1. The initial spike in the figures is due to scattering caused by the laser beam, and can be used as a time marker.

Following are the new significant observations made from the present studies. (1) The time evolution of the spectral emission profiles obtained in the present work clearly reveals that the species ejected by the YBCO target exhibit twin peaks for the TOF distribution. (2) The twin-peak distribution exists only for neutral atoms (like Y) and molecular species (like YO) while this feature is absent in the case of ionized species (like Y^+). (3) The twin-peak distribution was observed only up to a certain distance from the target (up to $\simeq$1.5 cm). At farther distances only the component corresponding to the first peaks occurs. (4) The intensity of the

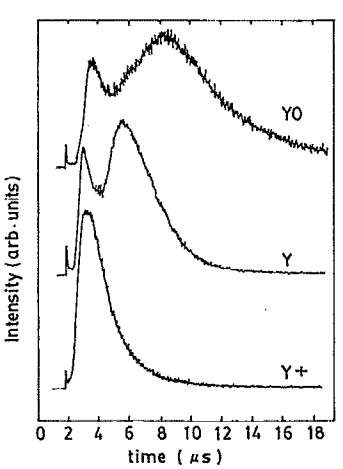

FIG. 1. Intensity variation of spectral emission with time for Y^+ (508.5 nm), Y (558.1 nm), and YO (601.9 nm) observed at the 1 cm away from the target at a laser fluence of 12 J/cm^2.

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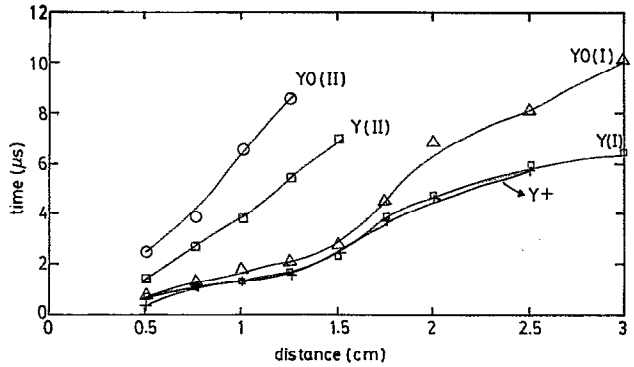

FIG. 2. Variation of time delay vs distance for the intensity peaks I and II in the case of Y^+, Y, and YO at constant laser fluence of 12 J/cm^2.

ionized species decreases drastically at distances greater than 1.5 cm from the target. (5) The time delay corresponding to the first peak decreases with respect to fluence. However, the time delay corresponding to the second peak is constant at lower fluences and decreases with fluence above a threshold.

Similar reports available in the literature describe twin-peak TOF distribution in laser generated plasma from YBCO using optical absorption spectroscopy[8] and fluorescence technique.[9] However, using plasma emission studies, this is the first observation of a twin-peak distribution.

It is seen that the intensity of Y^+ emission is very high near the target and decreases drastically beyond 1.5 cm and goes below the detection limit at distances greater than 1.75 cm. With reference to the twin peaks shown in Fig. 1, the peak I corresponding to Y and YO grows in intensity up to an optimal distances ($\simeq$1 cm) and then diminishes. The delayed peaks (peak II) of Y and YO go down in intensity throughout the plasma region and its range is limited up to 1.5 cm. It may be noted that the time delay for peak I of Y is almost the same as that of Y^+. The variation of time delays as a function of distance and fluence is shown in Figs. 2 and 3, respectively. The variation of time delays as a function of distance is linear for the second peak of Y and YO, while that for the first peaks of the Y and YO and for the single peak of Y^+ are slightly nonlinear showing a saturation effect farther from the target surface. A plot of delay time as a function of position (Fig. 2) shows that the first peak of Y and YO gives

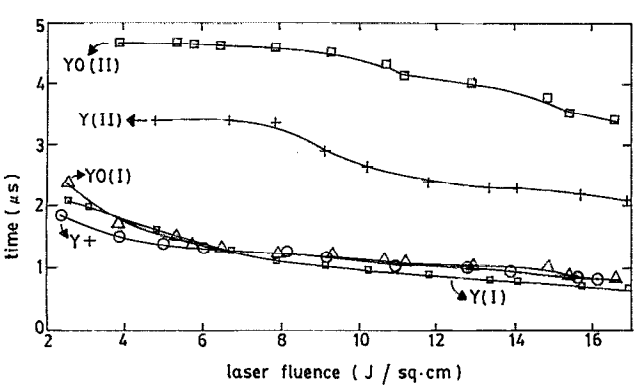

FIG. 3. Variation of time delay of intensity peaks with laser fluence for Y^+, Y, YO at a distance of 1 cm from the target.

a constant velocity ($V_{yo}\simeq 6\times10^5$ m/s, $V_y\simeq 7.2\times10^5$ m/s) until it reaches $\simeq$1.5 cm from the target surface after which they slow down rapidly to a much smaller expansion velocity ($V_{yo}\simeq 3.2\times10^5$ m/s, $V_y\simeq 4.4\times10^5$ m/s).

The possibility of twin-peak structure for Y and YO due to radiation trapping or self-absorption[12] is unlikely due to the fact that the time delays between the peaks vary with respect to distance from the target and they are of the order of microseconds. Okada *et al.*[9] attributed the time delays solely to TOF arising from different velocities of the species. They assigned the observation of twin peak to the presence of faster and slower components of the same species. If it is assumed that the terminal stage of the plume can be modeled as a free expansion into vacuum, the expansion velocity V can be written as[12]

$$V=\frac{2}{\gamma-1}\sqrt{\frac{\gamma kT}{M}}, \tag{1}$$

where M is the mass of the species, γ is the specific heat ratio the value of which varies from 1.2 to 1.3 to account for the degree of freedom associated with ionization and excitation,[13] and k is the Boltzmann's constant. However at a given point, using Eq. (1), since velocities depend only on the mass of the species and plasma temperature, it is not possible to have two different velocity distributions for the same species. The most plausible explanation is as follows. The twin-peak distribution arises as due to different origins for the same species occurring in the plasma. Species like Y and YO are generated directly from the target, while Y^+ is produced mainly just outside the target due to electron impact. In the vicinity of the target surface, the temperature is so high that ionization of Y by electron collision is highly probable.[14] As time evolves plasma temperature drops so that Y^+ generation decreases and collisional recombinations of Y^+ with electrons result in enhanced spectral emission from Y apart from those directly coming from the target. At a slightly later time YO is generated from collisional recombination of Y and O. Thus we have two types of same species in plasma. (i) Those coming directly/generated in the vicinity of the target surface and having larger TOF delays given rise to a second peak. (ii) Those generated away from the target surface due to collisions and recombination process which occur earlier at the point of observation giving rise to a first peak.

It must be noted that there is only one kind of Y^+, viz., that generated in the vicinity of the target surface due to collisional impact in the plasma. According to Eq. (1), since the masses are the same, both Y and Y^+ should have identical time delays which implies that the peak of Y^+ should coincide with peak II of Y. However the peak of Y^+ is observed earlier (coinciding with peak I of Y) due to the fact that the velocity of Y^+ is larger than that observed for Y, because of the ambipolar diffusion process due to space-charge effect produced by ion and electron densities near the target.[15]

From Fig. 3 we can see that the delay time for the first peak of species for Y and YO decreases with respect to increasing laser fluence, while the time delay corresponding to the second peak is constant at lower fluences and decreases

at higher fluences. This implies that the kinetic energy of the species coming directly/from the vicinity of the target surface do not vary much at lower fluence, while above a certain threshold sudden enhancement in kinetic energy takes place due to the initiation of nonlinear process in laser-matter interaction.

In conclusion, the present work has differentiated the type of species directly coming from the target surface and those due to the recombination process. We have observed the twin-peak structure for atomic and molecular species at short distances. Evaporation from the target/expansion of those generated in the vicinity of the target leads to the appearance of the delayed component and those created by collisional or recombination process apparently give rise to first peak.

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