Bull. Mater. Sci., Vol. 14, No. 3, June 1991, pp. 545–549. © Printed in India.

Characteristics of laser-induced plasma from high T_c superconductor

G PADMAJA, A V RAVI KUMAR, V VIDYALAL, P RADHAKRISHNAN, V P N NAMPOORI and C P G VALLABHAN

Department of Physics, Cochin University of Science and Technology, Cochin 682 022, India

Abstract. The spectroscopic analysis of the emission from the plasma produced by irradiating a high T_c superconducting $GdBa_2Cu_3O_7$ target with a high power Nd:YAG laser beam shows the existence of the bands from different oxides in addition to the lines from neutrals and ions of the constituent elements. The spectral emissions by oxide species in laser-induced plasma show considerable time delays as compared to those from neutral and ionic species. Recombination processes taking place during the cooling of the hot plasma, rather than the plasma expansion velocities, have been found to be responsible for the observed time delays in this case. The decays of emission intensities from various species are found to be non-exponential.

Keywords. Laser-induced plasma; delayed spectral emission.

1. Introduction

Preparation of high T_c superconducting thin films using laser ablation method is vigorously being pursued in many laboratories (Dijkkamp *et al* 1987; Moorjani *et al* 1987; Lynds *et al* 1988; Fogarassy *et al* 1988; Neifeld *et al* 1988). In order to deposit good-quality films which are superconducting, one has to control rather promptly the parameters of the laser-generated plasma. Emission studies from laser-induced plasma are a very convenient method to obtain information on the neutral, ionic and molecular components present in the same. The presence of oxides and ions of the constituent elements of the target material in the plasma generated is very crucial to obtain as-grown superconducting thin films (Fogarassy *et al* 1988). We have recently reported the presence of oxides and ionic species in laser-induced plasma generated by Nd:YAG laser beam from $GdBa_2Cu_3O_7$ and $YBaCu_3O_7$ samples (Padmaja *et al* 1989). In order to understand the detailed aspects of laser beam interaction with target material and the recombination processes following laser ablation, time-resolved studies of the spectral emission from plasma offer the most convenient approach. In this note, we report some of the important results of the dynamical measurements made on spectral emission from individual species present in the plasma generated by a high-energy laser pulse from a typical rare earth-based superconductor viz $GdBa_2Cu_3O_7$ ($T_c = 93$ K).

2. Experimental details

The schematic diagram of the experimental set-up is shown in figure 1. Plasma was produced using 1·06 μm radiation (10 ns pulses) from a Q-switched Nd:YAG laser with an average pulse energy ~ 20 mJ. The sample ($GdBa_2Cu_3O_7$ pellet) was mounted in a stainless steel vacuum chamber with quartz windows suitably located for target

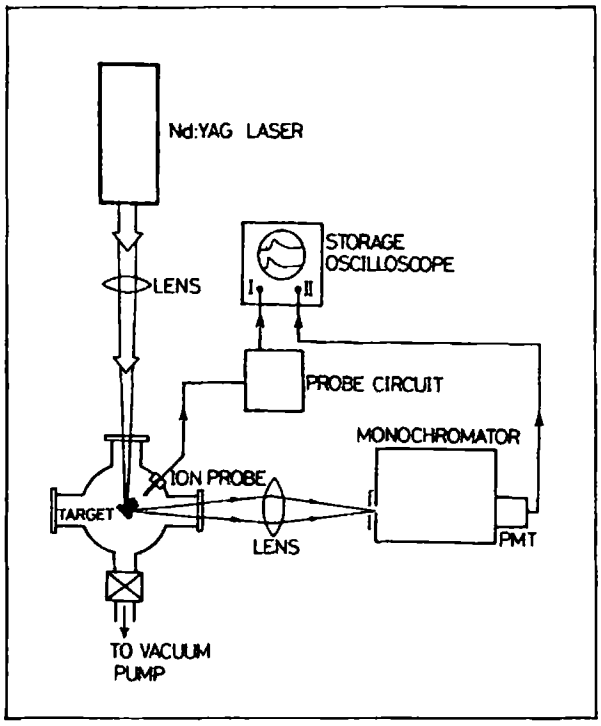

Figure 1. Schematic diagram of the experimental set-up.

irradiation and plasma detection. Details of the method used for spectroscopic analysis of the plasma were reported earlier (Padmaja *et al* 1989).

Characteristic lines were selected using 0·5 m Jarrel-Ash monochromator (resolution, $\Delta\lambda = 1$ Å) coupled to a PMT (50 ohm load) and a 100 MHz storage oscilloscope. This method offers precise and unambiguous line selection in contrast to the use of narrow band ($\Delta\lambda = 10$ nm) filters (Wu *et al* 1989). Time evolution of the emission from each species was observed by setting the monochromator to the respective emission lines and recording the pulse shape on the storage oscilloscope. A typical pulse shape recorded on the CRO due to the $\lambda = 455{\cdot}5$ nm line from the emission of copper ions is shown in figure 2.

3. Results

It is seen that the characteristic emissions from various species exhibit widely different time delays. The time delays measured for emission from different atomic, ionic and molecular species are displayed in figure 3. The most significant feature of the result is that the emissions from oxides are found to occur after a considerable delay as compared to that from ionic and atomic species. This evidently is due to the fact that the initial plasma temperature is so large that formation of oxides is not possible. As the plasma cools down, recombination processes help to form the oxides. A closer look at the pulse shape shows a non-exponential decay of the emission. This becomes

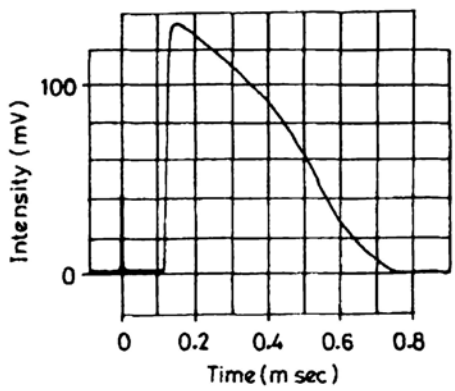

Figure 2. Typical CRO trace of the PMT response due to the emission line ($\lambda = 455{\cdot}5$ nm) of copper ions.

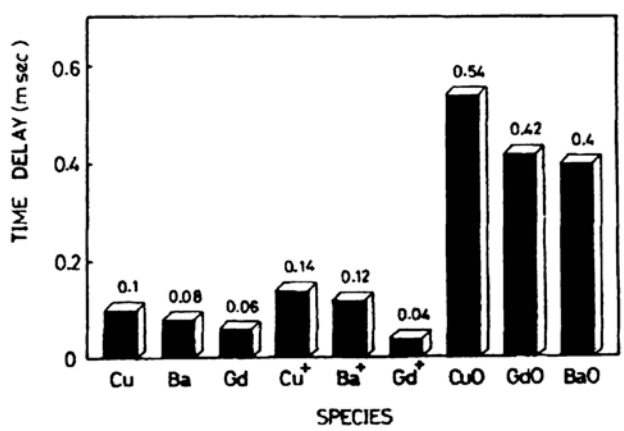

Figure 3. Time delays for the spectral emissions from various species in the plasma. The wavelength monitored for each species is given below. (i) Cu ($\lambda = 612{\cdot}7$ nm); (ii) Ba ($\lambda = 413{\cdot}2$ nm); (iii) Gd ($\lambda = 493{\cdot}4$ nm); (iv) Cu^+ ($\lambda = 455{\cdot}5$ nm); (v) Ba^+ ($\lambda = 416{\cdot}7$ nm); (vi) Gd^+ ($\lambda = 509{\cdot}8$ nm); (vii) CuO ($\lambda = 616{\cdot}3$ nm); (viii) BaO ($\lambda = 586{\cdot}0$ nm); (ix) GdO ($\lambda = 566{\cdot}4$ nm).

clearer from the $\log t$ vs $\log I$ graph shown in figure 4. The temporal decay of the emission due to plasma cooling can be represented as,

$$I(t) \sim H(t_1 - t)t^{-b_1} + H(t - t_1)t^{-b_2}$$

where $H(\tau)$ is Heaviside function so that $H(\tau) = 1$ for $\tau > 0, 0$ for $\tau < 0$. b_1 and b_2 are "decay constants" for respective species in the "hot" and "cold" phases of the plasma. For $t < t_1$, it is observed that $b_1 < 1$ for all species while $b_2 \geqslant 1$ except for BaO and Ba (figure 5). Due to collisional excitations and recombinations, initially the plasma is in a highly non-equilibrium state and for $t > t_1$, it tends to thermal equilibrium with a faster cooling rate.

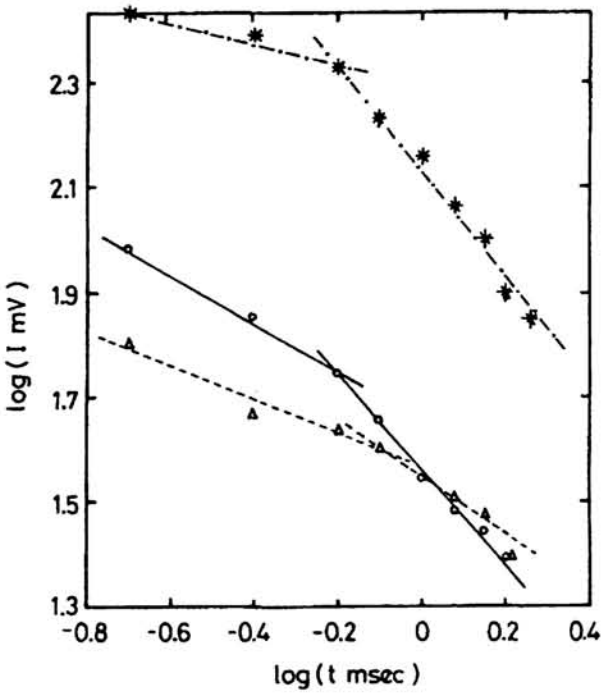

Figure 4. Log t vs I plot for the emissions of Ba, Ba^+ and BaO. (i) o–o–o BaO ($\lambda = 586{\cdot}0$ nm); (ii) Δ–Δ–Δ Ba ($\lambda = 413{\cdot}2$ nm); (iii) •–•–• Ba^+ ($\lambda = 416{\cdot}7$ nm).

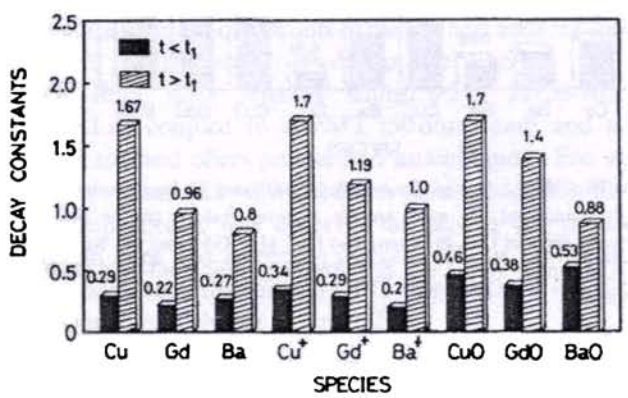

Figure 5. Decay constants b_1 and b_2 observed for the spectral emission from various species in the plasma.

4. Conclusions

It is obvious from the measured time delays in the case of CuO, GdO and BaO that the oxides are not formed directly from the target material but are generated in the recombination processes occurring in the plasma while it cools down.

It may be mentioned that Dyer *et al* (1988) attributed the time lags for the various species to plasma expansion time delays of different components so that the velocities should have $(M)^{-1/2}$ dependance (M = mass of the species). Our results apparently do not support this view (figure 3). Of course, the plasma expansion velocities may also contribute to the delays in certain cases (very small delays), but the observed large time delays (up to 540 μs) can certainly be not due to a time-of-flight phenomenon. Results of our initial measurements of the plasma velocities using a Langmuir probe give values near 10^4 ms^{-1} for positive ion velocities. This observation also fully supports the above conclusion.

Acknowledgements

The authors are grateful to the Department of Science and Technology (New Delhi), the Ministry of Human Resource Development (New Delhi) and the University Grants Commission (New Delhi) for financial assistances.

References

Dijkkamp D, Venkatesan T, Wu X D, Shaheen S A, Jisrawi N, Min-Lee Y H, McLean W L and Croft M 1987 *Appl. Phys. Lett.* **51** 619

Dyer P E, Greenough R D, Issa A and Key P H 1988 *Appl. Phys. Lett.* **53** 534

Fogarassy E, Fuchs C, Siffert P, Perriere J, Wang X Z and Rochet F 1988 *Solid State Commun.* **67** 975

Lynds L, Weinberger B R, Peterson G G and Kransinski H A 1988 *Appl. Phys. Lett.* **52** 320

Moorjani K, Bohandy J, Adrian F J, Kim B F, Shull R D, Chiang C K, Swartzendruber L J and Bennett L H 1987 *Phys. Rev.* **B36** 4036

Neifeld R A, Gunapala S, Liang C, Shaheen S A, Croft M, Price J, Simons D and Hill III W T 1988 *Appl. Phys. Lett.* **53** 703

Padmaja G, Ravi Kumar A V, Vidyalal V, Radhakrishnan P, Nampoori V P N and Vallabhan C P G 1989 *Pramana J. Phys.* **32** L693

Wu X D, Dutta B, Hegde M S, Inam A, Venkatesan T, Chase E W, Chang C C and Howard R 1989 *Appl. Phys. Lett.* **52** 179