Temporal and spatial evolution of laser ablated zirconium plume in different ambient and two-dimensional mapping of zirconium atoms using laser induced fluorescence technique.

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Zirconium ablation in different nitrogen ambient pressure (0.1, 1.0 and 10 mbars) has been carried out by neodymium-doped yttrium aluminium garnet (Nd: YAG) ns laser radiation at varying fluence (14, 20, 28 and 35 J/cm^2). The induced plasma plume is investigated by time- and space-resolved emission and imaging spectroscopy. The temporal and spatial gas-phase reactions due to the chemical reaction at the plume periphery showing the existence of zirconium nitride at pressure of 1.0 mbar resulting with the formation of zirconium nano-clusters begin ~1.6 μs time delay with respect to ablating pulse is seen. The formation of clusters is optimized by the control parameters i.e. laser energy and ambient pressure, and is found to be independent of the ambient gas. Plasma emission spectroscopy confirms the existence of clusters in the plasma plume. The laser induced fluorescence studies of zirconium plasma are performed by probing the expanding plasma using a dye laser tuned at Zr I transition a^3F_2 - z^3F_3 (λ=593.52nm). The LIF spectrum and imaging of fluorescence z^3F_3 - a^3F_3 (λ= 614.32nm) is recorded using a spectrograph attached to the ICCD camera. The LIF investigations are used to measure the atomic beam velocity of zirconium atoms produced at low laser fluence. Laser induced fluorescence technique is also used to analyze the two dimensional number density mapping of zirconium atoms in the plume of a low temperature laser-ablated zirconium plasma at low pressure ~10^{-2} mbar The maximum density is found to be $2.5 \times 10^{22}\, m^{-3}$.

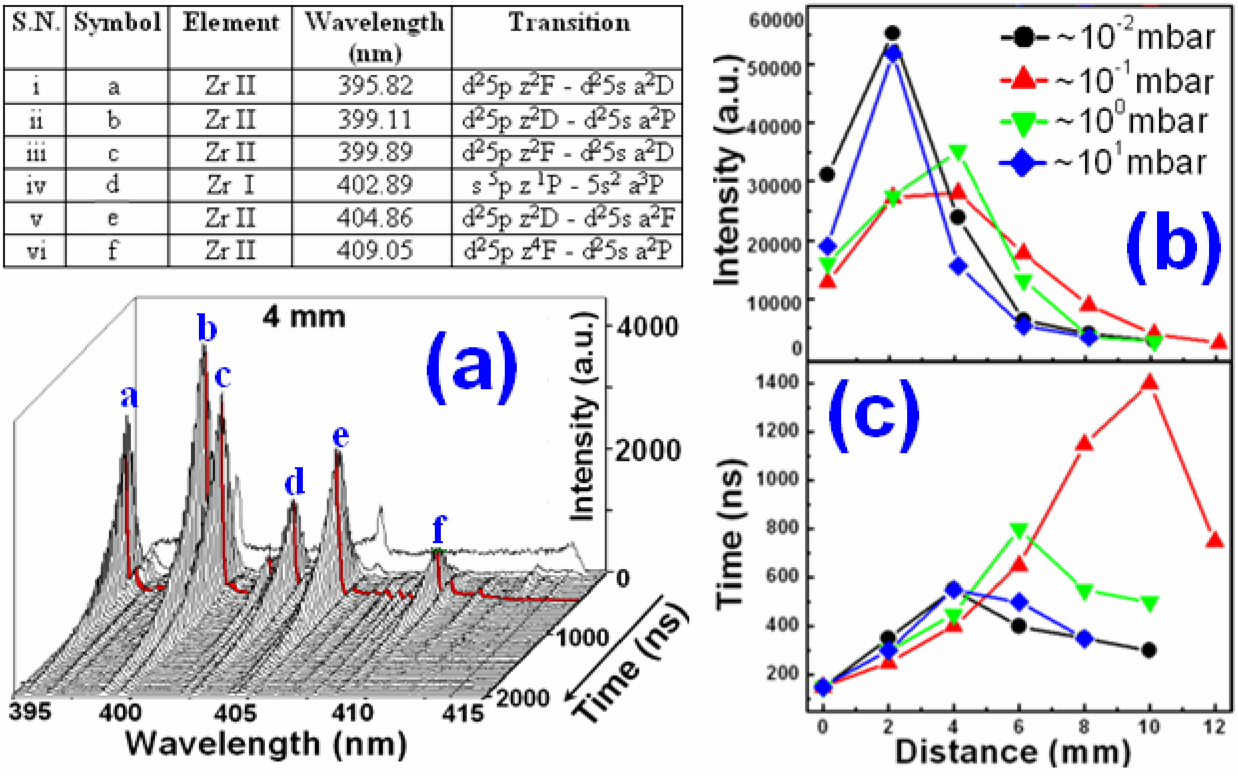
INTRODUCTION

The expansion of a laser ablated plasma into a low pressure background gas leads to complicated interactions. A number of important applications of laser ablation such as PLD are based on there interactions to know the dynamics of a plume of ablated material in a background gas [1]. To understand plume evolution is crucial because the properties of the thin films grown using PLD is related to plume characteristics [2, 3]. To understand the effect of the ambient gas pressure plume dynamics was investigated at three different pressures of the ambient gas, which are (i) low pressure, (ii) intermediate pressure and (iii) high pressure.

At low pressures, the plume expands freely with a constant average velocity without any external viscous force. At intermediate pressures intermingling between plume and ambient gas takes place. This is the transition region where expansion dynamics changes from free expansion to collisional. At high pressure, on the other hand, the plume begins to confine and due to the development of contact boundary between the plume and the gas, the expansion regime is transformed into a hydrodynamical regime.

RESULTS AND DISCUSSION:

In the presence of ambient gas (nitrogen) at later time (>200ns) emission of the zirconium plasma may be attributed to the cluster formation [4] of the zirconium atoms/ions at low temperature region as shown in Fig.1. Emission of the zirconium plasma depends on the ambient pressure. Inset of Fig. 1 shows the transitions involved in the emission.



S.N.	Symbol	Element	Wavelength (nm)	Transition
i	a	Zr II	395.82	$d^25p\ z^2F$ - $d^25s\ a^2D$
ii	b	Zr II	399.11	$d^25p\ z^2D$ - $d^25s\ a^2P$
iii	c	Zr II	399.89	$d^25p\ z^2F$ - $d^25s\ a^2D$
iv	d	Zr I	402.89	$s\ ^5p\ z\ ^1P$ - $5s^2\ a^3P$
v	e	Zr II	404.86	$d^25p\ z^2D$ - $d^25s\ a^2F$
vi	f	Zr II	409.05	$d^25p\ z^4F$ - $d^25s\ a^2P$

FIG. 1(a) Temporal profile of zirconium plasma, (b) and (c) Depicts effect on ambient pressure on the emission profile. Inset shows the table of spectroscopic emission transitions.

At 10 and 0.01mbars of the ambient pressure the emission intensity is optimum and close to the target (2mm) while at moderate pressures (0.1 and 1 mbar) it is away from the target (4mm) as shown in Fig. 1(b). Spatiotemporal dependence of emission peak is shown in Fig 1(c), at pressure 0.1 mbar optimum value of emission peak is evolving later in space and time both while at pressures 0.01, 1 and10 mbar occur earlier times and close to the target.

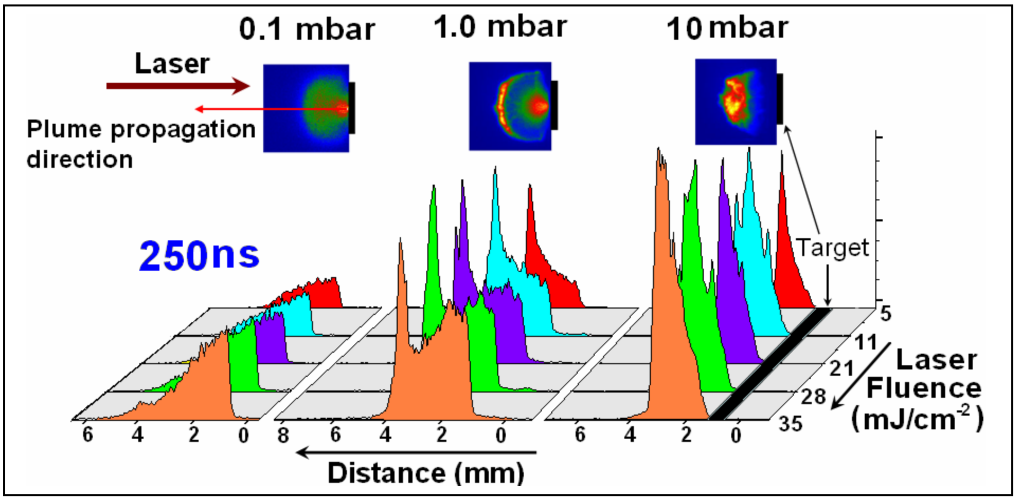


FIG. 2 Emission intensity of the Zirconium plasma along the pixel parallel to the plume propagation direction.

Figure 2 shows the emission intensity of the zirconium plasma along the pixel parallel to the plume propagation direction at different laser fluence in different ambient pressures. It was observed that at 1.0 mbar pressure there is a peculiar chemical reactions at the plume periphery (inset of fig 2.). At higher pressure 10mbar, there is a abrupt change in the intensity

distribution of the plume even at lower laser fluence, which may be because of more feasible chemical reactions of the plasma species with ambient nitrogen.

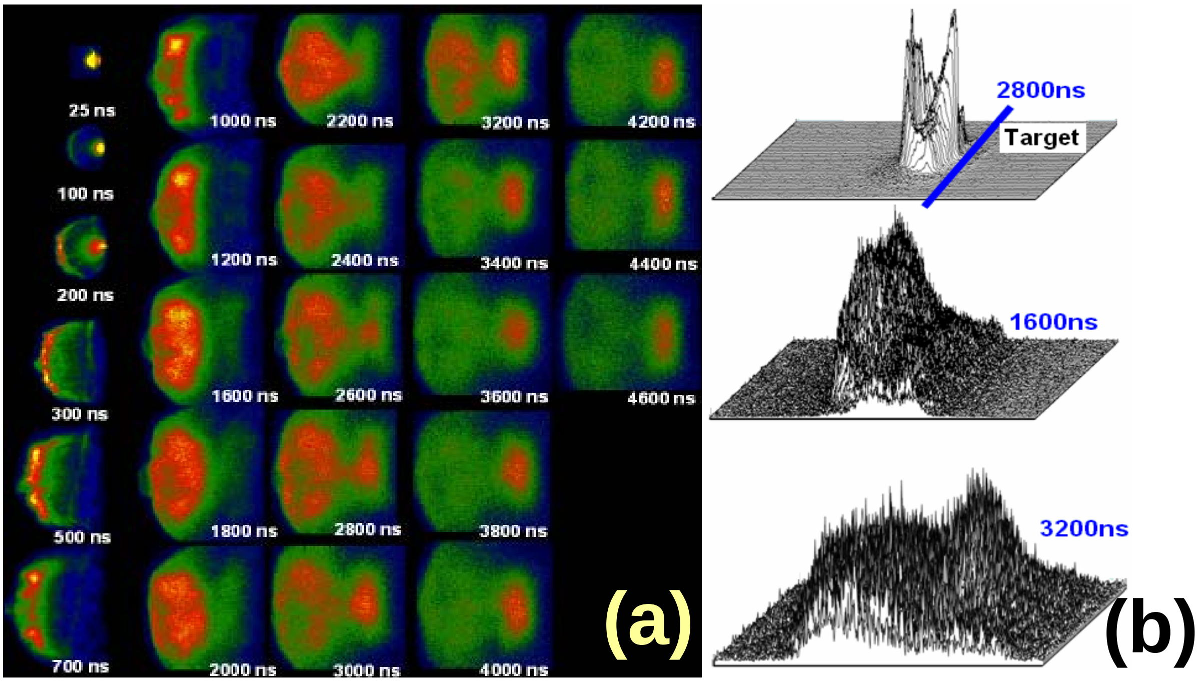


FIG. 3 (a) Emission intensity of the Zirconium plasma at 1.0 mbar of nitrogen ambient. (b) Intensity profile of the emission images.

Formation of zirconium clusters is observed at later time when the temperature of the plasma lowers down, which initiates around 1.6 μs and get maximum at 3.2 μs and survives for longer time even more that 4.2 μs, at ambient pressure 1.0 mbar with laser fluence of $35 J/cm^{-2}$.

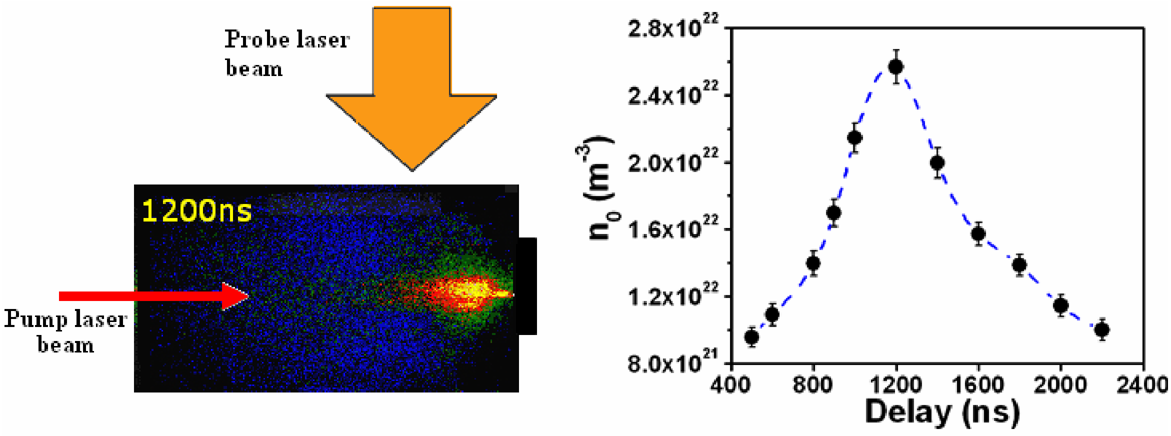


FIG. 4 (a) Laser induced fluorescence imaging and (b) temporal variation of ground state density of zirconium atoms.

The zirconium atom density is mapped using 2-D planner laser induced fluorescence spectroscopy and imaging. LIF spectroscopy [5] was used to estimate the atomic velocity ($5.4x10^5$cm/s) of the zirconium atoms which LIF imaging along with absorption experiment used to calculate the density map as shown in fig.4. Temporal profile of ground state density is shown in fig. 4(b) which varies from $9.5\times10^{21} m^{-3}$ to $2.5\times10^{22} m^{-3}$.

SUMMARY

Spatiotemporal study of zirconium plasma emission performed, which shows the dependency of emission peak on ambient pressure and the laser fluence. A optimum condition for the formation of zirconium cluster was found. Mapping of the neutral zirconium atoms and velocity is performed using LIF imaging and spectroscopy respectively.

ACKNOWLEDGMENTS

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