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Influence of laser irradiance and helium ambient on the expansion of laser produced carbon plasma

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ABSTRACT

The emission features of laser ablated carbon plasma plume generated in helium ambient have been investigated with time resolved plasma diagnostic technique. At lower irradiance levels only a slowly propagating C_2 component is seen. At higher irradiance levels, emission from C_2 shows a twin peak distribution in time. The present results also show that the helium ambient pressure and laser irradiance has opposite effects on the time of flight (TOF) profiles of C_2 species. A simple adiabatic expansion model appears to provide a good description of the plume range and this may prove useful for scaling deposition experiments in terms of pressure and laser irradiance.

Keywords: laser produced plasma, time resolved measurements, carbon plasma, carbon clusters, DLC films

1. INTRODUCTION

Carbon clusters like C_{60} and higher fullerenes are well known to be formed as a product of the laser ablation of carbon in an ambient helium atmosphere [1]. More over, laser ablation has become an attractive method for depositing carbon containing thin films like Diamond-like carbon (DLC) and carbon nitride thin films [2]. To understand the process of deposition of DLC films as well as the formation of stable carbon clusters, it is essential to investigate the mechanisms of material ejection by laser ablation of carbon. Vapourization, plume development and film deposition are quite complex sequence of processes likely to depend critically on a number of independent mechanisms.

The dynamic behaviour of species in laser ablated plasma plume affects the characteristics of deposited film. There are several studies aimed at the understanding of the pulsed laser deposition process and the correlation between deposited film properties and plasma parameters [3-5]. Among the various diagnostic techniques, which are convenient tools to detect different transient species, the optical emission spectroscopy has definite advantages pertaining to high spatial and temporal resolution without perturbing plasma. Using this technique, different plasma parameters like electron density, temperature, vibrational temperature of molecules and kinetic energy of the ablated species can be estimated [6-10].

In this paper we report emission features of laser ablated carbon plume generated in a helium ambient atmosphere with time resolved measurements. We studied the effect of laser irradiance and helium ambient pressure on the expansion of electronically excited C_2 species in the laser ablated carbon plasma.

2. EXPERIMENTAL SET UP

The experimental set-up used for the present study is similar to the one described elsewhere [11]. The carbon plasma is generated by laser ablation of the high purity carbon sample using 1064 nm radiation pulses from a Q-switched Nd: YAG laser with repetition rate 10 Hz. The target in the form of a disc is placed in an evacuated chamber provided with optical windows for laser irradiation and spectroscopic observation of the plasma produced from the target. The target is rotated about an axis parallel to the laser beam to avoid nonuniform pitting of the target surface. The bright plasma emission is viewed through a side window at right angles to the plasma expansion direction. The section of the plasma is imaged onto the slit of a 1-meter Spex monochromator using appropriate collimating and focusing lenses so as to have one to one correspondences with the sampled area of the plasma and the image. The recording is done by using a thermoelectrically cooled photomultiplier tube (PMT), which is coupled to a boxcar averager/gated integrator. For time resolved spectroscopic studies the output from the PMT is directly coupled to a 200MHz digital storage oscilloscope with 50 Ω termination.

3. RESULTS AND DISCUSSION

The time evolution of emission from the constituent species of the plasma determines the subsequent expansion and should provide the best observations for shaping and understanding of the plasma dynamics. Typical TOF distributions of C_2

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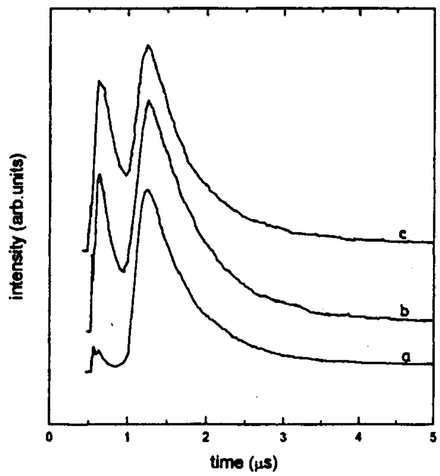

Fig.1. Intensity variation of spectral emission with time for C_2 species (516.5 nm) recorded at a distance 5 mm from the target for different irradiance levels. (a) 74.3 GW cm^{-2}, (b) 77.8 GW cm^{-2} and (c) 81.4 GW cm^{-2}. For these measurements the helium pressure is kept at 0.05 mbar.

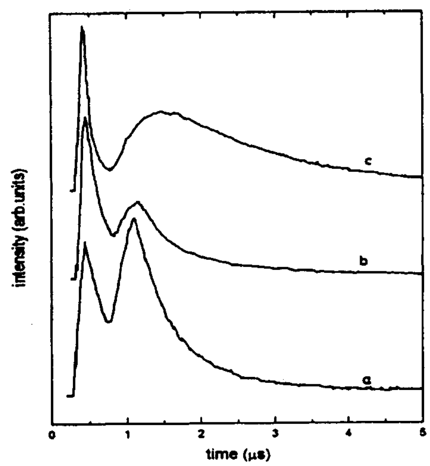

Fig. 2. Temporal profiles of C_2 species at different ambient helium pressures. (a) 0.065 mbar, (b) 1 mbar and (c) 10 mbar. These measurements are made at a distance of 8 mm from the target surface and at a laser irradiance of 81.34 GW cm^{-2}.

species obtained by monitoring the spectral emission from C_2 in electronic excited state (at λ = 516.5 nm corresponding to the (0, 0) transition $d^3\Pi_g \rightarrow a^3\Pi_u$ of C_2 Swan system) recorded at a distance 5 mm from the target surface for different laser irradiances are given in figure 1. The time evolution of the spectral emission obtained in the present work clearly reveals that the C_2 species ejected from graphite target has a twin peak distribution. This interesting feature in TOF pattern of C_2 species becomes prominent only beyond a threshold laser irradiance. Below this threshold laser irradiance only single peak distribution is observed. The above observations indicate that the plasma apparently develops fast and slow components above threshold laser irradiance.

There are two mechanisms that could exist for the particle formation during the laser ablation of carbon, viz, dissociative and recombinational. Most of the models of fullerene formation are based on the recombination mechanism, i.e., and the formation by nucleation from carbon vapour consisting of carbon atoms and very small carbon molecules. It is well known that graphite exhibits a large difference between the inter-layer and the intra-layer bond strengths. It is expected that at low laser irradiance levels, graphite will be ablated layer by layer producing large particles which in turn get dissociated to form C_2 species [12]. The dissociate mechanism can further be supported by the observation of long duration of Swan band emission at low irradiants.

The dominant mechanism for the production of C_2 Swan band emission at low irradiances is the electron collision with C_n cations and neutrals (n > 2) followed by dissociation where one of the fragments is an ejected C_2 molecule [10]. As the laser irradiance is increased, clusters with lower values of n will be ejected directly from the target. Above a threshold laser irradiance, temperature of the plasma becomes so large so as to dissociate C_n to neutral and ionized carbon atoms just outside the target. At higher irradiance levels Swan band formation is mainly due to electron-ion and ion-ion recombination. It is observed here that at high laser irradiances, after a threshold, an emission peak showing a faster component with higher kinetic energy for C_2 molecules begins to appear [9,10]. With increasing laser irradiance, we also observed a broadening and narrowing phenomena for slower and faster peaks respectively.

The variation of time delay for the faster (Pk1) and slower (Pk2) peaks also show different peculiarities. It is noted that the time delay for Pk1 increases with respect to increase in laser irradiance contrary to the normal observation. By considering the velocity distribution, the anomaly can be explained only if one can have some type of a "negative diffusion" or anomalous diffusion of C_2 towards the target [10]. Contrary to the case of Pk1, delay of Pk2 remains almost constant up to a threshold laser irradiance and thereafter it decreases [10].

The pressure of the ambient atmosphere is one of the controlling parameters of the plasma characteristics, as well as factors related to the laser energy absorption. Plume dynamics changes substantially when the pulsed laser evaporation takes place in an ambient atmosphere. We also observed that the emission characteristics of carbon plasma produced in Ar, He and air atmosphere depend strongly on nature and pressure of the ambient gas [6]. As the pressure is increased spectroscopic data showed increase in light emission from C_2 swan bands [13]. The presence of helium gas during the ablation process has dramatic consequences on the expansion dynamics. The TOF profile is found to be very sensitive to the pressure of the background gas. With increasing pressure of the helium gas the temporal profile of Pk1 becomes more and more narrow, while for Pk2, it broadens (fig. 2). The helium serves to cool the hot electrons by collisions leading to a more efficient electron impact excitation and plasma recombination, leading also to the plasma confinement near to the target which enhances the emission from these species. We also noticed that delay for the first peak decreases and for the second peak it is increased with increasing helium pressure.

The present results show that laser irradiance and pressure of the ambient helium gas have opposite effects on the plasma expansion processes. As irradiance increases, time delay for Pk1 increases while time delay for Pk2 is more or less constant up to threshold irradiance and then decreases. Similarly increase in helium gas pressure decreases the time delay for Pk1 and in the case of Pk2; time delay is being constant up to a particular pressure and then increases. Pressure and laser irradiance also showed opposite effects on narrowing and broadening of Pk1 and Pk2. With increasing irradiance, a broadening and narrowing phenomena is observed for Pk1 and Pk2 respectively. These interesting results can be explained by assuming a simple adiabatic model. The plume lengths L can be estimated if it is assumed that at the end of the laser pulse the ablation products occupy a volume V_i at high pressure and subsequently expand adiabatically. Assuming adiabatic expansion model, the ablated material pushes the gas species until the plasma and gas pressures equilibrate. Then the length of the plasma is given by [14]

$$L = A\left(\frac{(\gamma-1)E}{P_0}\right)^{1/3\gamma} V_i^{(\gamma-1)/3\gamma} \qquad (1)$$

where A is the geometric factor related to the shape of the laser spot at the target surface, γ is the specific heat ratio (C_p/C_v), E is the laser energy density, P_0 is ambient gas pressure, V_i is the initial volume of the plasma ($V_i = v_0\, \tau_{laser}\, w$, v_0 being the initial species velocity, τ_{laser} is the laser pulse duration and w, the laser spot size at the target surface). From eqn. 1, it is clear that E/P_0 is the parameter controlling the length of the plasma if the experimental geometry remains constant; therefore, E and P_0 have opposite effects as experimentally observed in the present work. According to this model, the length of the plasma L is expected to increase as the laser energy density is increased or ambient gas pressure is decreased. Considering the experimental parameters used in this work, evaluating eqn. 1 with $A = 1.6$ (for $\theta = 35^0$), $v_0 = 4\text{-}8 \times 10^6$ cm s^{-1}, $\tau_{laser} = 9$ ns, gives a straight line as shown in fig. 3 corresponding to the present experimental pressure range. In our experiments, the estimated length of the plasma at a helium pressure of 0.05 mbar is 24 mm. Both the predicted pressure dependence and absolute value of L are seen to be reasonably in good agreement with experiment. It must be noted that the deposition rate and kinetic energy of ablated species will fall into a very low value for substrates located beyond L because of the subsequent cooling which tends to pull the material back to the target.

4. CONCLUSIONS

1064 nm radiation from a Q-switched Nd:YAG laser was focused onto the carbon target where it produced a transient and elongated plasma. TOF studies of various species have differentiated the various mechanisms of the formation of C_2 species in the laser-produced plasma from graphite in a helium gas atmosphere. An oscillatory behaviour is observed in the TOF distribution of C_2 species and this is observed only above a certain threshold value for irradiance. The peak due to low kinetic energy component, which is observed at all levels of irradiance, is formed as a result of dissociation of higher clusters. The departure of single peak velocity distribution at higher laser irradiances is due to processes like recombination of the high energetic particles. The present results also show that the ambient gas pressure and laser irradiance has opposite effects on the TOF profiles of C_2 species. The broadening and narrowing of the multiple peaks with these parameters is expected due to collision and recombination phenomena. A simple adiabatic expansion model appears to provide a good description of the plume range and this may prove useful for scaling deposition experiments in terms of pressure and laser irradiance.

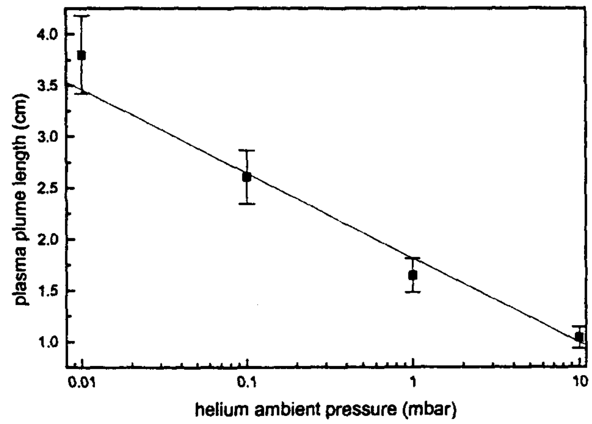

Fig. 3. Estimated length of the plasma plume L as a function of helium pressure

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