Plasma species dynamics in a laser produced carbon plasma expanding in low pressure neutral gas background

H M Ruiz, F Guzmán, M Favre, H Bhuyan, H Chuaqui and E Wyndham
Departamento de Física, Pontificia Universidad Católica de Chile, Av. Vicuña Mackenna 4860, Santiago, Chile
E-mail: hmruiz@uc.cl

Abstract. We present time and space resolved spectroscopic observations of a laser produced carbon plasma, in an argon background. An Nd:YAG laser pulse, 370 mJ, 3.5 ns, at 1.06 μm, with a fluence of 6.8 J/cm², is used to produce a plasma from a solid graphite target, at a base pressure of 0.5 mTorr, and with 80 mTorr Argon background. The spectral emission in the visible is recorded with 15 ns time resolution. 20 ns time resolution plasma imagining, filtered at characteristic carbon species emission wavelengths, is used to study the dynamics of the expanding plasma. Two different fronts with ionic or molecular compositions are seen to detach from the laser target plasma.

1. Introduction
Several investigations have been reported regarding fundamental processes of carbon plasma dynamics and plasma chemistry in laser produced carbon plasmas [1,2]. Carbon and diamond like (DLC) thin film deposition using graphite targets has been investigated, using different parameter regimes [3]. In this context we have investigated carbon film deposition using a graphite target laser plasma in an argon background. Preliminary analysis of the resulting films indicates a correlation between films properties, as inferred from AFM and other standard materials science diagnostics, with the pressure of the argon background. In order to establish a relationship between the carbon plasma and the resulting films properties, we have studied the time and space evolution of the laser produced carbon plasma. Here, we present time and space resolved spectroscopic observations of the laser plasma.

2. Experimental set-up
The experiments were performed in a vacuum chamber at a base pressure of 0.5 mTorr. An Nd:YAG laser pulse, 370 mJ, 3.5 ns, at 1.06 μm, operating at 10 Hz was applied to a rotating graphite target, at approximately 45° to the normal. The focal spot was 0.0535 cm², thus resulting in a fluence of 6.87 J/cm². Time resolved spectral observations of the carbon plasma plume were performed, at base pressure and at 80 mTorr argon background. The visible spectra were obtained with a Spectra Pro 275 (1200 g/mm) spectrometer, with a gated avalanche array with a time window of 15 ns. Light emission from the carbon plasma plume was collected with a f-f fiber optic arrangement, focusing at 2.5, and 4.5 mm, from the target surface. Plasma images, filtered at characteristic carbon species emission wavelengths, were recorded, with 20 ns time resolution.
3. Experimental results

Figure 1 shows a time integrated spectrum obtained from a point 2.5 mm from the target surface, with 80 mTorr argon background. The spectrum shows emission lines associated with single ionized Carbon, single ionized Argon, and C₂ molecular bands, corresponding to vibrational states of the \( \text{d}^3\Pi_g \rightarrow \text{a}^3\Pi_u \) Swan band transition [4]. The emission band around 405 nm has been assigned previously to transitions \( \text{A}_1\Pi_u \rightarrow \text{X}_1\Sigma_g^+ \), Swing band, of the neutral \( \text{C}_3 \) molecule [5]. A residual second harmonic laser emission line can also be identified, which is due to the optical configuration of the Nd:YAG laser cavity.

Figure 2 shows the time evolution of light emission from a point 4.5 mm from the target surface, with 80 mTorr Argon pressure. The spectra correspond to emission from a C₂ molecular band (\( \Delta \nu = 1 \)), centered at 467.5 nm, and CII ions at 426.78 nm. At early times, ArII emission lines are observed, which disappear when the CII and C₂ emission become prominent.

To investigate the dynamic features of the different carbon species in the expanding laser plasma we obtained plasma images using 10 nm band-pass filters centered at 470 nm, which correspond to the C₂, \( \Delta \nu = 1 \), Swan band. Figure 3 shows the dynamic evolution of the plasma emission from, (a) C₂ carbon molecules at the base pressure of 0.5 mTorr and, (b) C₂ carbon molecules at 80 mTorr Argon.
background. At the base pressure, the $C_2$ component is seen to remain nearly stationary. The dynamics is much more complex at 80mTorr Argon. In fact, two distinctive, expanding plasma fronts are seen to detach from the near target semi stationary plasma. The first one, seen in the 66 to 330 ns time interval, is a rather homogeneous front, but the second one, from 750 ns onwards, exhibits a polar dependence with respect to the normal to the target, with a noticeable decrease in brightness close to the cloud axis.

**Figure 2.** Time evolution of light emission from a point 4.5 mm from the target surface, with 80 mTorr Argon pressure. (a) $C_2$ molecular band ($\Delta\nu = 1$), centered at 467.5 nm, and (b) CII ions at 426.78 nm.

**Figure 3.** 20 ns exposure, spectrally resolved laser plasma images: (a) 0.5 mTorr base pressure, (b) 80 mTorr Argon.
4. Discussion
Several results have been reported on laser produced Carbon plasmas, using different laser fluences and gas backgrounds. At a high fluence of 50 J/cm², with Helium as background gas, the formation of C₂ molecules has been studied, using both, OES and plasma imaging [2]. It has been found that in vacuum C₂ formation is only seen near the target surface, but with Helium background, an expanding front of C₂ molecules is observed. In another experiment, at a higher fluence of 955 J/cm², two distinctive fronts, a fast and a slow one, have been identified in the temporal evolution of the C₂ rich laser Carbon plasma [1]. Ionization of the background neutral gas has been reported with a laser produced Tin plasma, in 100 mTorr Argon background [6]. Single ionized Argon background emission lines were found before Tin emission lines, due to laser plasma expansion, were observed. This might be caused by fast electrons produced at the early stages of the laser plasma, which propagate ahead of the laser plasma front.

Our time integrated spectrum in figure 1 shows single ionized Argon emission, from a point 4.5 mm from the target surface. Further time resolved observations indicate that the ionized Argon emission occurs before the Carbon molecules front is formed, as shown in figure 2(b). When the time resolved spectral emission seen in figure 2 is correlated with the time resolved images shown in figure 3, it can be concluded that the presence of ArII and CII ions coincide in space and time with the first plasma front, seen in figure 3(b).

5. Conclusions
We have investigated the role placed by Argon background in molecule formation in laser produced carbon plasmas. At the base pressure of 0.5 mTorr C₂ molecules are only observed near the target surface. At 80 mTorr Argon, two successive plasma fronts are observed. The first one is dominated by CII ions and propagates in a weakly ionized Argon background. The second one is dominated by the presence of C₂ molecules. It is evident that the Argon background favors Carbon molecule formation through three-body recombination processes. These observations contribute to understand morphological properties of thin Carbon films obtained by pulsed laser deposition, under identical experimental conditions, reported elsewhere.

Acknowledgements
This work has been funded by FONDECYT project 1110380. H. M. Ruiz and F. Guzmán acknowledge doctoral studies scholarships from MECESUP and CONICYT, respectively.

References
[1] Harilal S S, Issac R C, Bindhu C V, Nampoori V P N and Vallabhan C P G 1997 J. Appl. Phys. 82 2140
[2] Al-Shboul K F, Harilal S S, Hassanein A and Polek M J 2011 J. Appl. Phys. 110 053301
[3] Qian F, Singh R K, Dutta S K and Pronko P P 1995 Appl. Phys. Lett. 67 3120
[4] A Kushwaha and R K Thareja 2008 Applied Optics 47 G65
[5] Nemes L, Keszler A M, Parigger C G, Hornkohl J O, Michelsen H A and Stakhursky V 2007 Applied Optics 46 4032
[6] Harilal S S, O’Shay B, Tao Y and Tillack M S 2006 Journal of Applied Physics 99 083303