Characterization of a laser plasma produced from a graphite target

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Abstract. In order to improve the understanding of pulsed laser deposition (PLD) of diamond-like carbon (DLC) films, we have started a detailed study of the plasma dynamics of laser produced carbon plasmas. The carbon plasma is produced by focusing a Nd:YAG laser pulse, 380 mJ, 4 ns at 1.06 μm, onto a graphite target, at a background pressure of 0.3 mTorr. Time resolved optical emission spectroscopic (OES) observations of the carbon plasma plume are obtained, with time and space resolution, using a SpectraPro 275 spectrograph, with a 15 ns MCP gated OMA. Line emission from CII to CIV carbon ions is identified at different stages of the plasma evolution. Line intensity ratios of successive ionization stages, CIII/CIV, was used to estimate the electron temperature throughout the Saha-Boltzmann equation, under the assumption of local thermodynamic equilibrium (LTE), and Stark broadening of CII lines was used to obtain measurements of the electron density. Characteristic plasma parameters, short after plasma formation, are 3.0 eV and $2 \times 10^{17}$ cm$^{-3}$, which after 60 ns of plasma expansion decay to 2.7 eV and $5 \times 10^{16}$ cm$^{-3}$, respectively.

1. Introduction
Pulsed laser deposition (PLD) has become a well established technique for thin film deposition [1, 2]. A short laser pulse is focused onto a solid target, resulting in a laser produced plasma plume that expands away from the target surface. The expansion can take place in vacuum or in low pressure gas background. The effect of several global process parameters on the resulting thin film properties have been studied in detail. These include laser wavelength, pulse energy and pulse duration, background media and substrate conditions, amongst others. Less attention has been given to investigate the laser plasma properties, with time and space resolution, in correlation with the resulting thin film properties. Carbon plasmas produced by pulsed laser ablation of a graphite target in an ambient gas are been used to grow diamond-like carbon (DLC) films and to synthesize carbon nano compounds [3, 4, 5]. We have recently started investigations of DLC film growth using PLD. In order to improve on the understanding of PLD deposition of DLC films, we have studied in detail the plasma dynamics of laser produced carbon plasmas. By using time and space resolved OES, we have characterized the properties of laser produced carbon plasma plumes, including measurements of the electron density and temperature.
2. Experimental Details
The carbon plasma is produced by focusing a Nd:YAG laser pulse, 380 mJ, 4 ns at 1.06 μm, at 10 Hz repetition rate, onto a rotating graphite target (POCO graphite), using a 12.5 cm focal length lens. Base pressure is kept at 0.3 mTorr. Time resolved OES observations of the carbon plasma plume are obtained with a SpectraPro 275 spectrograph (1200 grooves/mm, maximum resolution 0.05 nm), with a 15 ns MCP gated OMA. Light emission from the plasma plume is collected transversal to the plasma plume, with 0.6 mm space resolution, in the range from 1.6 to 3.4 mm from the graphite target surface.

3. Results and Discussion
Figures 1 and 2 show time resolved spectra of the laser plasma plume, at 2.2 mm from the target surface. Emission lines corresponding to CII to CIV transitions are identified. In order to highlight dynamic features in the plasma evolution, different time windows have been selected for both spectral bands. A particular feature in figure 1 is the broadened line profile of a singly charged ion (CII at 426.7 nm), and a general rapid decrease in the intensities of the carbon lines. Figure 2 shows the temporal evolution of the CIV (580.1 nm) line. After the 60 ns the CIV line declines completely, whereas the CII (589.1 nm) grows over the same time interval. This growth can be ascribed to recombination of higher ionization stages, as inferred from overall line intensity evolution.

Stark broadening of spectral lines is a well established technique to obtain measurements of the electron density in the range of $10^{14} - 10^{18}$ cm$^{-3}$ [6]. The FWHM of the Stark broadened lines $\Delta \lambda_{1/2}$ depends on the electron density $n_e$ through the relation

$$\Delta \lambda_{1/2} = 2W \left( \frac{n_e}{10^{16}} \right) [\text{Å}]$$

where ion correction factors have been neglected, $n_e$ is in cm$^{-3}$ and $W$ is the electron impact parameter [6]. We have used Stark broadening of the CII line at 426.7 nm to measure, with time and space resolution, the electron density of the carbon plasma. A Lorentzian profile has been fitted to the spectral line to obtain the FWHM. Time and space resolved values of the plasma density are shown in figure 3, for distances between 1.6 and 3.4 mm from the target. The axial displacement of the plasma plume can be appreciated in the time evolution of the axial electron density distribution. As the plasma emission collecting point moves away from the target surface, the time to reach maximum electron density is delayed, reaching lower characteristic values. These features are consistent with and plasma plume that expands in volume, thus decreasing the characteristic electron density as the plume front moves away from the target surface. Relative line intensities of subsequent ionization stages of the same element are very sensitive to temperature changes. In local thermodynamic equilibrium (LTE), the ratio of line intensities is given by [6]

$$\frac{I'}{I} = \frac{f'f\lambda^3}{f'g\lambda^2} \left( 4\pi^{3/2}a_0^3n_e \right)^{-1} \left( \frac{k_BT_e}{E_H} \right)^{3/2} \times \exp \left( - \frac{E' + E_\infty - E - \Delta E_\infty}{k_BT_e} \right)$$

where the primed symbols represent the line of the atom with higher ionization stage, $f$ is the oscillator strength, $g$ is the statistical weight, $\lambda$ is the associated wavelength, $a_0$ is the Bohr radius, $E_H$ is the ionization energy of the hydrogen atom, $E$ the excitation energy, $\Delta E_\infty$ is the correction to the ionization energy $E_\infty$ of the lower ionization stage due to plasma interactions, and $n_e$ is the electron density. Here we take advantage from the fact that the electron density has been determined independently for the same plasma conditions. In this particular case we have chosen as higher ionization stage the CIV line at 580.1 nm, and as lower ionization stage the CII line at 569.6 nm. The required data to be fitted in equation 2 for both transitions has been obtained from NIST Atomic Spectra devolution of the electron temperature is again
consistent with cooling expanding plasma. The characteristic time span for plasma decay is of the order of 120 ns.

Our results are similar to those reported previously [8] for a laser produced carbon plasma, in which, under equivalent laser irradiation conditions, the plasma temperature was found to be in the 1.5–2.5 eV range, and the electron temperature in the $1.1–2.1 \times 10^{17} \text{ cm}^{-3}$ range. The main difference here is the fact that single shot data allowed both, the electron density and temperature evolution, to be followed in space and time, thus providing valuable data to extrapolate to thin film deposition conditions.

4. Conclusions
We have characterized the time and space evolution of a laser produced carbon plasma, close to the target surface. This laser carbon plasma characterization will be useful in understanding...
the properties of nanostructured diamond like carbon (DLC) films produced under identical experimental conditions, which will be reported elsewhere. Additional experiments to characterize a carbon laser plasma produced in an Argon background are also in progress.

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