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Effect of ambient nitrogen pressure on the formation and spatio-temporal behaviour of C_2 and CN

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Abstract. We report the effect of ambient gas on the formation as well as propagation behaviour of ablated species C_2 and CN within the carbon plasma created by focussing a high-power Nd:YAG ($\lambda = 1064$ nm) laser onto the rotating graphite target in the nitrogen ambient. The formation of C_2 takes place earlier as well as nearer the target compared to that of CN which forms later and far from the target, in 1.2 mbar pressure of N_2 gas. Peak arrival time vs. nitrogen gas pressure plot shows a shock wave-like dependence $t \propto p^n$ in the pressure range 1.2–120 mbar (collisional regime) which indicates plume confinement with increases in ambient pressure. At higher pressure, thermalization takes place.

Keywords. Graphite; laser produced plasma; laser spectroscopy.

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1. Introduction

Laser ablated carbon plasma is widely used for the preparation of carbon nitride (CN) thin films [1] by pulsed laser deposition (PLD) technique. To prepare good-quality CN thin films, the role of ambient nitrogen atmosphere is very important. In an ambient gas, the expansion behaviour of the plasma plume can be more complex. It may depend on atomic mass of the plume and atoms of the ambient gas, initial plume energy and density, ambient gas pressure etc. When carbon plasma propagates in ambient nitrogen, the ablated species reacts with ambient gas and gives carbon nitride and by placing suitable substrate in front of the plasma plume, the desirable CN thin film can be obtained. The quality of thin film generally depends on the distribution of kinetic as well as internal energy of ions, atoms, molecules and clusters in the plume. Thus, the dynamics of the emitted species and their evolution within the expanding plume should be thoroughly studied. Among various optical diagnostics techniques, optical emission spectroscopy (OES) [2,3] is preferred to study kinetics and dynamics of the emitted species. In the present work, the effect of ambient nitrogen pressure on the formation of C_2 and CN molecules in laser-induced carbon plasma and its spatio-temporal characteristics are studied.

2. Experimental details

Carbon plasma was formed by focussing a Q-switched Nd:YAG (DCR-4G, Spectra Physics) laser with a pulse width of 8 ns at full-width half-maximum (FWHM) at a repetition rate of 10 Hz which delivers ∼1 J laser energy per pulse operating in the fundamental mode ($\lambda = 1.064$ μm) Nd:YAG laser ($\lambda = 1064$ μm) on rotating graphite target in ambient nitrogen in a stainless steel vacuum chamber. The laser has a Gaussian-limited mode structure with beam divergence less than 0.5 mrad and a beam diameter of 8 mm. Laser energy was monitored by a power meter (Ophir Model 30A) which was placed in the path of the main beam. The chamber was evacuated using a rotary pump and a high-speed turbomolecular pump (TMP). The vacuum chamber was evacuated to a pressure of 10^{-5} mbar and was purged with the N_2 gas several times before introducing gas in a controlled manner. The emission of the expanding plasma was collected by an optical fibre bundle through a collecting lens of 10 cm focal length and then coupled with an intensified charged coupled device (ICCD, DH 720, Andor Technology, USA) via a monochromator. ICCD was interfaced with a personal computer. The triggering (timing for laser pulse and the ICCD gate-opening) was monitored using a delay generator (8082A, Hewlett-Packard) and an oscilloscope (Agilent 54615B).

3. Results and discussion

The time- and spatial-resolved emission spectra of laser-induced carbon plasma in various ambient nitrogen gas pressures (1.2 mbar, 12 mbar and 120 mbar) are recorded by optical emission spectroscopy (OES). In the early stage of plasma expansion, the spectra are predominately continuum emission, whereas, at later times of expansion the ionic and atomic emissions dominate. At much later times, bimolecules (C_2 and CN) and clusters (C_3) are observed. At a laser fluence of 32 J cm^{-2}, C_2 Swan bands ($d^3\Pi_g - a^3\Pi_u$) of the sequence $\Delta\upsilon = 0$ at 516.5 nm $(0,0)$, 512.9 nm $(1,1)$, 509.7 nm $(2,2)$ and CN violet bands ($B^2\Sigma^+ - X^2\Sigma^+$) of sequence $\Delta\upsilon = +1$ at 421.6 nm $(0,1)$, 419.7 nm $(1,2)$, 418.1 nm $(2,3)$, 416.8 nm $(3,4)$, 415.8 nm $(4,5)$, 415.2 nm $(5,6)$ are observed. The spectral variations of C_2 at various delays at a distance of 3 mm in 1.2 mbar nitrogen gas pressures and its spatio-temporal behaviour are shown in figure 1a. It is observed that the formation of C_2 is optimum at 350 ns at 3 mm from the target surface and that of CN is 850 ns at 4 mm. Generally, C_2 forms by chemical reactions of ions and atoms of carbon with the ambient nitrogen gas, but some direct ejected C_2 molecules are there which are responsible for the initial rise which can be clearly seen in figure 1b. After the formation of C_2, CN will be formed by gas phase reaction given by $C_2 + N_2 \rightarrow CN$. In expanding plume, formation of C_2 occurs at early delay (350 ns) and at 3 mm and then C_2 molecules act as seed molecules for the CN molecules which are formed at later delay (850 ns) and at comparatively large distance (4 mm) [4]. The formation of CN molecules depends on the density of C_2 species and ambient pressure.

The ambient nitrogen gas plays an important role in the formation and dynamics of C_2 and CN. The temporal variation of C_2 in 1.2, 12 and 120 mbar pressures

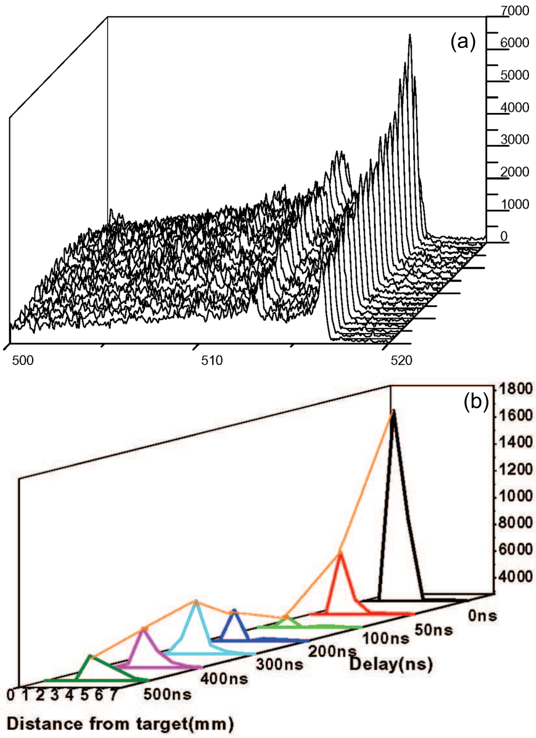

Figure 1. (**a**) Spectral variation of C_2-516.5 nm at various delays at a distance of 3 mm in 1.2 mbar nitrogen pressure. (**b**) Spatio-temporal variation of C_2.

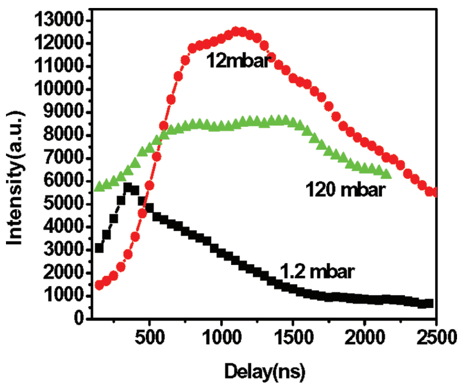

Figure 2. Temporal variation of C_2 at 3 mm for various pressures.

at 3 mm from the target surface is shown in figure 2. At low pressure/vacuum, plume expands freely. On increasing pressure, ablated species (at plume front) starts colliding with ambient gas atoms. In this collisional regime, plume–ambient gas interaction results in shock wave formation, plume splitting etc. Peak arrival

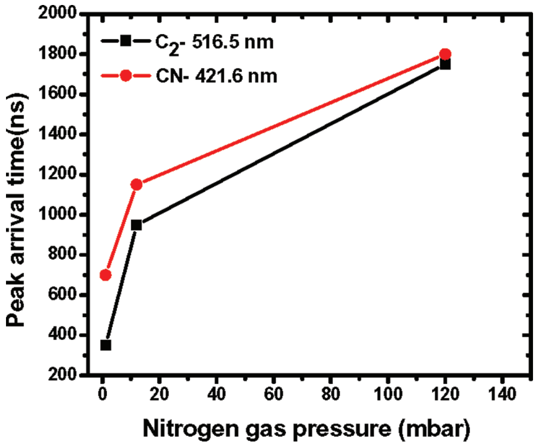

Figure 3. Delay of peak intensity of C_2 and CN vs. N_2 pressure at 3 mm.

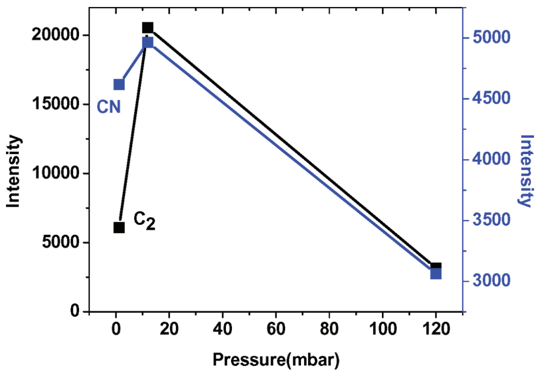

Figure 4. Pressure vs. intensity for C_2 and CN at 3 mm from the target.

times of C_2 and CN are also plotted with nitrogen gas pressure (figure 3) which shows a shock wave-like dependence ($t \propto p^n$) in the pressure range 1.2–120 mbar. Formation of shock wave-like expansion shows plume confinement with increase in ambient pressure [5]. At/above 120 mbar pressure, thermalization will take place. It is also found that formation of C_2 increases with time and after attaining an optimum, it goes down. C_2 is maximum in 12 mbar nitrogen gas pressure at a delay of 1100 ns. Formation of C_2 as well as CN increases from 1.2 mbar to 12 mbar and it decreases at 120 mbar pressure which is shown in figure 4.

4. Conclusion

In this study we focussed on the effect of the ambient nitrogen gas pressure on the formation and propagation behaviour of C_2 and CN within the laser-induced carbon plasma. It is observed that formation of C_2 occurs at early delay (350 ns) and at 3 mm from the target surface and then it acts as seed molecules for the

CN molecules which are formed at later delay (850 ns) and at comparatively large distance (4 mm). The formation of C_2 is maximum in 12 mbar nitrogen gas pressure at a delay of 1100 ns. C_2 as well as CN intensities increase with ambient nitrogen pressure and then decrease. The plot between peak arrival time of C_2 and CN; and nitrogen gas pressure (figure 3) shows a shock wave-like dependence ($t \propto p^n$) in the pressure range 1.2–120 mbar. Formation of shock wave-like expansion shows plume confinement with increase in ambient pressure.

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