Comparative study of pulsed laser ablated plasma plumes from single crystal graphite and amorphous carbon targets. Part I. Optical emission spectroscopy

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A comparative study of ablation plasma plumes originated from single crystal graphite (SCG) and amorphous carbon (a-C) targets during the preparation of diamond-like carbon (DLC) films by KrF excimer pulsed laser deposition (PLD) has been carried out by means of a monochromator equipped with an intensified optical multichannel analyzer. In high vacuum, the emission lines of carbon neutral C and ions of C+, C2+, and C3+ can be observed from both the SCG and a-C plasma plumes. The emission intensity from C atoms increases with laser energy density (EL) increase for both cases. The C2 emission intensity from the SCG plasma plume changes drastically with EL, while that from the a-C plasma plume is almost constant. The C2/C emission intensity ratio for the a-C case decreases with EL increase. As for the SCG case, the C2/C ratio decreases with EL increase up to 3.0 J/cm2, and increases slightly with further EL increase. Nanohardness of the deposited films decreases with the increase of the C2/C emission intensity ratio. It is suggested that for both the SCG and a-C target cases, the C2 molecule in the ablated plasma plume may not play an important role in producing high quality DLC films. It is further proposed that the threshold of laser fluence for the formation of diamond-like character film using KrF excimer PLD is 2.1 J/cm² (0.84 × 10^8 W/cm²) for the a-C target and 3.0 J/cm² (1.2 × 10^9 W/cm²) for the SCG target.

I. INTRODUCTION

Recently, a wide variety of carbon-related materials, such as diamond-like carbon (DLC),1–18 high fullerene carbon molecules of C60, C70, and C84,19–21 carbon nanotubes,17,22,23 and carbon nitride,24,25 has attracted much interest. Hydrogen-free DLC films with attractive properties, such as extreme hardness, chemical inertness, very low electrical conductivity, optical transparency over a wide range, and low electron affinity, have great potential for applications in mechanical and optical coatings, electronic devices, and field emitters. Carbon fullerenes and carbon nanotubes have been extensively studied because of their interesting characteristics, such as structural, electronic, magnetic, and especially nonlinear optical properties.

Pulsed laser deposition (PLD) has many advantages, such as low temperature processing, low levels of contamination, stoichiometry preservation, and reproducibility of film characteristics compared with the other deposition methods. Since the average energy of laser ablated species is much higher than kT and a certain fraction of these species is ionized, it is possible to form metastable phases such as DLC. Consequently, PLD has been regarded as one of the most successful techniques to synthesize novel and metastable materials and structures. Usually in the PLD system, amorphous carbon (a-C) is used as the target to prepare these materials. Single crystal graphite (SCG) has strong in-plane bonds and very weak bonds between planes. Since the in-plane C–C bond length in graphite is 1.43 Å compared to 1.54 Å C–C bond length in diamond, it is envisaged that in-plane bulk modulus of graphite may be greater than that of diamond. Carbon fullerenes and nanotubes are graphite sheets rolled up in spherical and cylindrical shapes, respectively. If the fraction of a graphite sheet is ejected from the target by laser irradiation, it may form spherical and/or cylindrical shaped structure to minimize energy. It is speculated that SCG has a high potential to produce carbon en-
riched with complex heavier molecules using PLD. The pulsed laser ablation of SCG target should be interesting from the point of view of the characteristics of deposited films, the dynamics of the laser ablated plume, and the correlation between film properties and plasma characteristics.

Because the dynamics of laser ablated plasma plume affects the characteristics of deposited thin film, there are many investigations using optical emission spectroscopy (OES), time of flight mass spectrometry, laser-induced fluorescence spectroscopy, interferometry, and the Langmuir probe method to understand the PLD process and to find out the correlation between deposited film properties and plasma parameters. Among these various diagnostic techniques, the OES has definite advantages such as species identification, and high spatial and temporal resolution without perturbation of the plasma. From the intensities and/or wave forms of the observed emission spectra in the OES, various plasma parameters, such as electron temperature and electron density, kinetic energy of ablated species, and vibrational temperature of molecules, can be estimated. Therefore, the OES is very useful for in situ monitoring during preparation of films using PLD. In spite of several investigations, however, the mechanism of the PLD process has not yet been understood in detail, specially in the case of DLC deposition. In order to prepare high-quality thin films and nanoparticles and to improve the potential of PLD, it is desirable to diagnose and establish correlation between plasma composition (atoms, ions, and molecules in the expanding plasma plume produced by laser irradiation) and properties of thin films. The comparative study of laser ablated plasma plumes during the PLD using the SCG and the a-C targets is especially important.

We have systematically investigated the characteristics of hydrogen-free DLC films preparation using KrF excimer laser ablation of the SCG and the a-C targets. The deposited DLC films have been characterized using nanohardness measurements and Raman spectroscopy. The properties of laser-ablated plasma plume have been diagnosed using optical emission measurements and Langmuir probe measurements. Our goal is to correlate the properties of the deposited DLC films with the characteristics of laser-ablated plasma plumes and to more completely understand the process of these materials through these systematic investigations. In this article, we describe mainly the comparative study of the optical emission measurements for laser ablated plasma plumes originated from the SCG and the a-C targets during KrF excimer PLD in high vacuum and nitrogen background. Spatially and spectral resolved optical emission is investigated by means of a monochromator equipped with an intensified optical multichannel analyzer with gating function. The Langmuir probe measurements and the characterization of deposited films are also employed to understand the PLD process. Quantitative measurements of optical emissions from atomic carbon, ionic carbon, diatomic carbon molecules, and carbon nitride molecules in plasma plumes from SCG and a-C targets are reported. We discuss possible correlations between plasma characteristics and film properties.

II. EXPERIMENT

The schematic diagram of the experimental apparatus used in this study is shown in Fig. 1 of Ref. 1. Only the experimental procedure is briefly described here. A stainless steel chamber is evacuated to about $10^{-7}$ Torr by a turbomolecular pump equipped with a mechanical booster pump and a rotary pump. The SCG target (10 mm square, 5 mm thick) or the amorphous carbon (a-C) target ($\phi 1 \text{ in.} \times 0.25 \text{ in.}$, 99.995%) is placed in the center of the chamber and is constantly rotated at 6 rpm during experiments to avoid pitting. High purity nitrogen gas was fed into the chamber and the pressure up to 600 mTorr was sustained during the experiment by a mass flow controller. The Lambda-Physik KrF excimer laser, EMG201 ($\lambda = 248 \text{ nm}$, max. output of 600 mJ, pulse duration of 25 ns at full width at half maximum) is used to irradiate the target with an incident angle of 45°. The laser energy density ($E_L$) on the target surface is set 1.0–5.0 J/cm² corresponding to an average power density in the 10$^8$ W/cm² range. Diagnostics of plasma plume include optical emission spectroscopy and a triple Langmuir probe measurement described elsewhere. Optical emission from the ablation plasma plume is collected at a right angle to plasma expansion and imaged onto an end of a bundled optical fiber ($\phi 1 \text{ mm}$) by a biconvex lens ($f = 100 \text{ mm}$). The 2:1 correspondence with the viewing area of the plasma plume and the image gives the spatial resolution of $\phi 2 \text{ mm}$. The other end of the fiber is coupled with an entrance slit of a 0.32 m monochromator (Instrument SA Inc., HR-320) with a 1200 g/mm holographic grating. An intensified diode array (Princeton Instruments, Inc., IRY-700S/B) is attached on the exit port of the monochromator. An optical multichannel analyzer system (Princeton Instruments, Inc., OMA) is used to detect the spatial and spectral resolved optical emission from the plasma plume. The OMA system consists of an intensified diode array (IRY-700S/B), a detector controller (ST-120), a high voltage gate pulser (PG-10) for the intensifier, and operating software. The emission spectra are integrated over 20 laser shots to improve the signal to noise ratio and then recorded on a personal computer. The excimer laser and the OMA system are synchronously controlled at a repetition rate of 10 Hz by a delay/pulse generator (Stanford Research Systems, Inc., DG535). The observed line emissions and band emissions are identified using the information available in the Refs. 9 and 29–31. During the DLC film deposition, the substrate is placed parallel to the target surface at 5 cm separation. The DLC films are deposited on the Si (100) substrate at room temperature under various nitrogen pressures up to 500 mTorr at 10 Hz repetition rate for 15–30 min. The DLC characteristics of the deposited films are studied by Raman scattering spectroscopy and correlated with nanohardness measurements.

III. RESULTS

A. Ablation in high vacuum

Optical emission spectra of the carbon plasma plume originating from the SCG and the a-C targets are shown in Figs. 1(a) and 1(b), respectively. The plasma plumes were generated at $E_L = 3.0 \text{ J/cm}^2$ in high vacuum ($\sim 10^{-7}$ Torr).
These optical spectra were observed at a distance \( d \sim 10 \) mm from the target surface, and integrated for 2 \( \mu \)s after the laser pulse. As shown in Figs. 1(a) and 1(b), atomic carbon lines C I, such as 437.14, 477.13, 493.20, 505.39, 538.03 nm, and carbon ion line emission at C II \((\lambda = 401.54 \text{ nm})\), C III \((\lambda = 407.04, 426.84 \text{ nm})\), C IV \((\lambda = 422.03 \text{ nm})\) can be observed in both cases of the SCG and the a-C target. However, it should be noted that the Swan band emission of the \( \text{C}_2 \) molecule \((d^3 \Pi_{g} - a^3 \Pi_u, \Delta \nu = 0:516.5 \text{ nm}, \Delta \nu = +1:473.4 \text{ nm})\) is observed only in the plasma plume originating from the SCG target [as shown in Fig. 1(a)], while no \( \text{C}_2 \) emission from the a-C target is detected [as shown in Fig. 1(b)]. In a previous work, we reported that the plasma plume in vacuum from the a-C target consists mostly of monatomic carbon ions and neutral atoms. The presence of \( \text{C}_2 \) emission for the SCG case under similar laser irradiation conditions, however, suggests a greater population of heavy particles in the plasma plume compared with the a-C case.

Closer to the target surface \((d = 5 \text{ mm})\), weak \( \text{C}_2 \) emission can be observed even in the plume originating from the a-C target. The \( \text{C}_2 \) emission intensity for both target cases varies drastically with laser energy density. Figure 2 shows the optical emission spectra at \( d = 5 \) mm of the plasma plume from the SCG target as a function of \( E_L \). At \( E_L = 1.1 \text{ J/cm}^2 \), weak \( \text{C}_2 \) emission is observed, while no emission lines from monatomic carbon neutrals are detected. At \( E_L = 2.1 \text{ J/cm}^2 \), the \( \text{C}_2 \) emission intensity becomes larger than that at \( E_L = 1.1 \text{ J/cm}^2 \), some C I lines appear and the intensity of C I \((505.39 \text{ nm})\) is as comparable as that of the \( \text{C}_2 \) Swan band head \((516.5 \text{ nm})\). At still higher laser energy density \((2.8 \text{ J/cm}^2)\), the \( \text{C}_2 \) emission intensity decreases while the C I emission intensity increases. In this range of laser energy densities, the optical emission spectra, as well as the ablated species and their fractions, behave much differently.

Figures 3(a) and 3(b) show the emission intensities of C I \((505.39 \text{ nm})\) and the \( \text{C}_2 \) Swan band head \((516.5 \text{ nm})\) as a function of laser energy density, respectively, detected at \( d = 5 \) mm for 2 \( \mu \)s after the laser pulse. In Figs. 3(a) and 3(b), the open and black circles indicate the SCG target and the a-C target cases, respectively. The threshold for the observation of C I emission is about 1.1–1.4 J/cm\(^2\), and the emission intensity increases with \( E_L \) increase for both cases. On the other hand, the \( \text{C}_2 \) emission intensity changes for the SCG and the a-C targets are quite different. For the SCG case, the \( \text{C}_2 \) emission intensity changes dramatically with \( E_L \) up to about 3 J/cm\(^2\) and achieves a maximum value near 2 J/cm\(^2\). With further increase in \( E_L \) up to 5.2 J/cm\(^2\), the \( \text{C}_2 \) intensity increases gradually from a minimum near 3 J/cm\(^2\). For the a-C case, the \( \text{C}_2 \) emission intensity increases with
increase of $E_L$ up to 2 J/cm$^2$, then is nearly constant in the $E_L$ range of 2–5 J/cm$^2$. The emission intensity ratio of C$_2$/C obtained from Figs. 3(a) and 3(b) is plotted in Fig. 4 as a function of laser energy density $E_L$. The open and black circles indicate the C$_2$/C intensity ratio for the SCG and the a-C target cases, respectively. For the a-C target case, the C$_2$/C intensity ratio decreases with $E_L$ increase. At low energy density below $E_L=2.1$ J/cm$^2$, the C$_2$/C intensity ratio changes greatly. Above this value the C$_2$/C intensity ratio decreases gradually with laser energy density increase. Many articles$^{10,14–16}$ have shown that higher laser energy density leads to the higher $sp^3$ bond fraction in the DLC film deposited by PLD using an a-C target. This result suggests that the C$_2$ molecule in the ablated plasma plume is not conducive for high quality DLC films deposited by PLD using an a-C target, and is in good agreement with our previous result.$^1$ As for the SCG case, the C$_2$/C intensity ratio decreases greatly with increase of $E_L$ up to 3.0 J/cm$^2$, then increases slightly with $E_L$ increase. The critical value (where the curve changes slope) of 3.0 J/cm$^2$ is larger than that for the a-C case. Below 3.0 J/cm$^2$ for the SCG case, the C$_2$ emission intensity is relatively large as shown in Fig. 3(b) compared with the a-C case. In our Langmuir probe measurements,$^{32}$ the velocities of ions in the plasma plume generated in vacuum at $E_L=3.0$ J/cm$^2$ are found to be 5 $\times$ 10$^4$ m/s for the a-C case and 2.5 $\times$ 10$^4$ m/s for the SCG case. Also, it is suggested that slightly less ionization for the SCG case occurs than for the a-C case at the same $E_L$. It should also be noted that the substrate temperature rises to 85 $^\circ$C during the film deposition using the SCG target, while only about 50 $^\circ$C for the a-C case. From these results, it is conjectured that there may be many heavy particles in the SCG plasma plume, especially at relatively lower laser energy density below 3.0 J/cm$^2$.

Furthermore, the optical emission data suggests that the formation mechanism of excited C atoms and C$_2$ molecules resulting in these optical spectra change drastically at $E_L=2.1–3.0$ J/cm$^2$ corresponding to 0.84–1.2 $\times$ 10$^8$ W/cm$^2$. Muller et al. have proposed that the threshold of laser fluence for the formation of diamond-like character film is 3.0 $\times$ 10$^9$ W/cm$^2$ using a KrF excimer laser.$^{18}$ More recently, Tabbal et al. have presented the effect of laser intensity on characteristics of DLC film deposited by KrF excimer PLD.$^{33,34}$ In the literature, DLC films having the $sp^3$ content exceeding 33% (maximum 60%) have been obtained at the laser intensity over 0.9 $\times$ 10$^8$ W/cm$^2$. The nanohardness of our deposited films, described later, shows a relatively high value of 35 GPa when the deposition is performed above 3 J/cm$^2$, although the $sp^3$ content in these films is not as of yet confirmed. Therefore, we suggest that the threshold of laser fluence for the formation of diamond-like character film using KrF excimer PLD may be as low as 2.1 J/cm$^2$ (0.84 $\times$ 10$^8$ W/cm$^2$) using an a-C target and 3.0 J/cm$^2$ (1.2 $\times$ 10$^8$ W/cm$^2$) for a SCG target, respectively.

We have deposited DLC films on a Si (100) substrate at 5 cm from the target surface in vacuum. Figure 5 shows the nanohardness of the DLC films deposited using the SCG (solid line) and the a-C (broken line) targets at various laser energy densities. The average nanohardness of the DLC film deposited by a-C target ablation is found to be 32 GPa for $E_L=3.0$ J/cm$^2$ and 48 GPa for $E_L=4.2$ J/cm$^2$, respectively. For the a-C case, the nanohardness increases with laser energy density increase, which is consistent with the laser energy density dependency of the diamond-like character mentioned earlier. For the SCG case, however, the nanohardness of the DLC film deposited at 3.0 J/cm$^2$ is found to be 55 GPa and higher than that of the DLC film deposited at 4.3 J/cm$^2$. This suggests the degradation of the diamond-like character of the DLC film deposited using SCG target at relatively high laser energy density. The nanohardness of the deposited films as a function of the C$_2$/C intensity ratio is shown in Fig. 6. The open and the black circles indicate DLC films deposited using SCG and a-C targets, respectively. The nanohardness, as one measure of diamond-like character, sug-

FIG. 5. Nanohardness of the deposited films using SCG (solid line) and a-C (broken line) targets at 3.0–4.3 J/cm$^2$.
suggests a decreasing trend with C$_2$/C intensity ratio increase regardless target type. This observation is consistent with our earlier Raman scattering results.1,2

In order to further identify properties of the ablation plasma plumes originated from SCG and a-C targets, we have estimated the vibrational temperatures of the C$_2$ molecule using a relative emission intensity method assuming a Boltzmann distribution in the density at various excited levels of the molecule.11,31 In our previous article, we have described in detail the procedure for estimating the vibrational temperature and its utility as applied to the ablation plasma plume. Figure 7 shows the vibrational temperature of the C$_2$ molecules at d=5 mm as a function of $E_L$. The plasma plumes were generated from the SCG target (open circle) and the a-C target (black circle) in high vacuum as low as 2x10$^{-7}$ Torr. For the SCG case, the C$_2$ vibrational temperature is almost constant about 0.5 eV at low laser energy density below 3.0 J/cm$^2$, and increases slightly to about 0.6 eV in the range of 3.0–5.2 J/cm$^2$. The critical value of 3.0 J/cm$^2$ is the same as described earlier. The C$_2$ vibrational temperature for the a-C case increase with $E_L$ increase. At $E_L$ = 5.0 J/cm$^2$, the C$_2$ vibrational temperature for the a-C case is estimated to be 1.1 eV and higher by a factor of 2 than that for the SCG case. In the range of 3.0–4.8 J/cm$^2$, we have estimated the electron temperature of the plasma plume using the Langmuir probe measurement (part II of this article). In this range, the electron temperature for the SCG case is found to be 10%–20% larger than that for the a-C case.3,32 This is consistent with the features of the C$_2$ vibrational temperatures estimated here, although the C$_2$ vibrational temperatures show different characteristics.

B. Ablation in nitrogen background

The formation of CN in nitrogen background pressure and its incorporation into the DLC films is also interesting from the viewpoint of nitrogen doping as well as formation of carbon nitride phases. The α and β forms of crystalline carbon nitride have been speculated to posses hardness exceeding that of diamond. Also, a recent paper describes carbon nanotube formation during PLD in nitrogen.17 Thereby, PLD in a nitrogen atmosphere is also important for synthesis of novel materials and structures. Figure 8 shows the optical emission spectrum at d=10 mm in the ablation plasma plume originating from the SCG target at $E_L$ = 3.0 J/cm$^2$ in a nitrogen background of 100 mTorr. This spectrum was obtained for the first 2 µs after the laser pulse. In this figure, CN violet band ($B^2Σ^+ - X^2Σ^+$, $Δν$ = –1) emission is observed in addition to C I lines and C$_2$ Swan band emission (d$^3Π_u$ – a$^3Π_g$, $Δν$ = 0:516.5 nm, $Δν$ = 1:473.4 nm). This spectrum is very similar to that from the a-C target [Fig. 1(b) in Ref. A], which was obtained under the same conditions.

It is well known through spectroscopic and electrostatic measurements that the plasma plume in the presence of background gas has two distinct velocity distributions of ablated species.12–14 In order to clarify the different phenomena corresponding to two distributions, the emission spectra were taken as a function of nitrogen pressure at various gate widths and delay times from the laser pulse. In previous1 as well as present studies, we have measured the time-integrated optical emission for 2 µs after the laser pulse, and for 50 µs after the laser pulse with 2 µs delay. Figures 9(a) and 9(b) show the emission spectra in a nitrogen background for the SCG and the a-C cases, respectively. The plasma plumes were generated at $E_L$ = 3.0 J/cm$^2$ in 500 mTorr and the emission spectra were observed at d=10 mm. In the a-C plasma plume [Fig. 9(b)], C I emission lines (493.20, 504.3, and 505.49 nm) and C$_2$ Swan band emission are observed in both cases for the first 2 µs and the subsequent 50 µs. On the other hand, no C I emission from the SCG plasma plume [Fig. 9(a)] is detected for the subsequent 50 µs, while C I emission lines and C$_2$ emissions are observed for the first 2 µs.

Figure 10 shows the emission intensity of C$_2$ (516.5 nm) from the SCG and the a-C plasma plumes as a function of nitrogen pressure up to 500 mTorr. The plasma plumes were generated at $E_L$ = 3.0 J/cm$^2$, and observed at d=10 mm. Open (and black) circles and squares indicate the emission intensities for the first 2 µs and for the subsequent 50 µs from the SCG (and a-C) plasma plume, respectively. The C$_2$ emission intensity for the SCG case increases with nitrogen pressure increase in both the first 2 µs and the subsequent 50 µs. For the a-C case, the C$_2$ emission intensity increases linearly with nitrogen pressure increases up to 200 mTorr for the first 2 µs and 300 mTorr for the subsequent 50 µs. As the pressure is increased further, these intensities are nearly constant. At relatively high pressure, especially at 500 mTorr,
the intensity for the SCG case is much larger than that for the a-C case for both the first 2 ms and subsequent 50 ms cases. Figure 11 shows the vibrational temperature of the C₂ molecules at \( d = 10 \) mm as a function of nitrogen pressure. The plasma plumes were generated from the SCG target or the a-C target at \( E_L = 3.0 \) J/cm². Open and black circles and squares indicate the emission intensities for the first 2 ms and for the subsequent 50 ms from the SCG (a-C) plasma plume, respectively. The C₂ vibrational temperatures decrease slightly with the pressure increase for all cases. As for the a-C case, the C₂ vibrational temperatures are about 0.5 eV for the first 2 ms and 0.4 eV for the subsequent 50 ms, indicating no significant difference. On the other hand, the C₂ vibrational temperatures are about 0.8 eV for the first 2 ms and 0.5 eV for the subsequent 50 ms of the SCG case. As for the a-C case, we suggest that the formation of excited C₂ molecules is the result of molecular recombination of C atoms and ions. However, the big differences of the C₂ emission intensity and vibrational temperature between the SCG and the a-C targets suggest that additional effects play a role in the formation of excited C₂ molecules, which include fragmentation of heavy particles in the SCG plasma plume. The ion saturation current to the Langmuir probe in the carbon ablation plasma plume in nitrogen also shows double peaks in the time-resolved profile. In high vacuum, only a single peak for both the SCG and a-C cases are observed. The velocity of carbon ions in high vacuum was estimated to be as high as \( 5 \times 10^4 \) and \( 2.5 \times 10^4 \) m/s for the a-C case and the SCG case, respectively. In nitrogen background, the time separation between the two observed peaks for the SCG case is slightly larger than that for the a-C case. Also, as nitrogen pressure continues to increase up to 500 mTorr, the ion velocities determined by the first peak for the SCG plasma plume decay much more slowly than those for the a-C plasma plume. These Langmuir probe data suggest that the SCG plasma plume includes a larger number of heavy particles than the a-C plume.

IV. CONCLUSION

A comparative study of ablation plasma plumes originating from SCG and a-C targets has been performed using optical emission spectroscopy during the DLC film preparation by KrF excimer PLD in vacuum and nitrogen background. In high vacuum, the emission lines of carbon neutral C and ions of C⁺, C²⁺, and C³⁺ can be observed from both SCG and a-C plasma plumes. Otherwise, the emission intensities of C and C₂ from the SCG plasma plume change drastically with the laser energy density compared with the a-C plasma plume, especially at lower laser energy density around 2 J/cm². The nanohardness of deposited films decreases with the increase of the C₂/C emission intensity ratio. This suggests that for both SCG and a-C cases the C₂ molecule in the ablated plasma plume may not play an important role in producing high quality DLC films. It is proposed that the threshold of laser fluence for the formation of diamond-like character film using KrF excimer PLD is 2.1 J/cm² (0.84 \( \times 10^8 \) W/cm²) for the a-C target and 3.0 J/cm² (1.2 \( \times 10^8 \) W/cm²) for the SCG target. In high
vacuum, the $C_2$ vibrational temperature of the SCG plasma plume is almost constant, while that of the $a$-C plasma plume increases with laser energy density increase. In nitrogen background, the $C_2$ vibrational temperature for the first 2 $\mu$s of the SCG plasma plume is estimated to be about 0.8 eV, higher than that of about 0.4 eV for the subsequent 50 $\mu$s, whereas the $C_2$ vibrational temperatures are nearly the same for these times in $a$-C case. From the optical emission spectroscopy and Langmuir probe measurements in vacuum and nitrogen background, it is conjectured that there is a large concentration of heavy particles in the SCG plasma plume, especially at relatively lower laser energy density below 3.0 J/cm$^2$. Optical emission spectroscopy is a powerful tool to diagnose the characteristics of transient plasma plumes and to monitor the film deposition process and resulting properties.

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