Optical emission diagnostics of C_{60}-containing laser-ablated plumes for carbon film deposition

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Optical-emission studies of laser-ablated carbon plasma used for deposition of carbon films is reported at irradiance in the range 2.4×10^8–9.0×10^{10} W/cm^2 using 1.064-, 0.532-, and 0.355-μm laser wavelengths in the presence of ambient gas. The presence of argon gas has a pronounced effect on the emission. The vibrational temperature calculated from molecular C_2 Swan bands is found to peak at an intermediate laser irradiance. C_{60}-containing carbon films are deposited on a silicon substrate at various ambient gas pressures and the correlation between their formation and laser-produced plasma parameters is discussed.

I. INTRODUCTION

Pulsed-laser-induced ablation or vaporization from material surfaces has been a subject of immense interest for quite some time in view of its importance in the context of our understanding the nature of laser-solid interactions.[1–4] Laser-induced vapor plasmas are becoming increasingly important in the processing of advanced materials and as a medium for fundamental high-temperature experiments. Laser-ablated plasmas have made significant progress in their application to many fields of basic research and materials technology such as production of microclusters, growth of thin films, annealing, etching, and chemical modification of surface layers, and in the fabrication of microelectronics devices.[5] Pulsed-laser deposition has become a leading technique for thin-film deposition due to its various advantages such as high deposition rates, low-temperature processing, retention of stoichiometry, and great versatility of processing variables. Despite the variety of applications our physical understanding of the pulsed-laser ablation is limited to a narrow range of irradiation regimes. There are various reports in the literature on the production and characterization of high-irradiance plasma, however, the comparative studies of low- and high-irradiance plasmas are rare.[6]

Laser-solid interaction depends strongly on laser and interaction parameters (wavelength, irradiance, pulse wave form, irradiated spot size, angle of incidence, etc.), material characteristics (composition, optical, and thermal properties, etc.), and environmental conditions (pressure, flow field, acceleration, etc.).[7] It is possible to get atoms in highly excited states or molecular clusters from the laser-ablated plume depending on laser power and pulse duration. Low-irradiance ($\sim10^8$– 10^9 W/cm^2) laser-created plasma is mostly used for pulsed-laser deposition of diamondlike carbon films,[8–10] high-T_c superconductors,[11] polymeric thin films,[12] and production of clusters,[13,14] etc. Its use at high irradiance includes mainly the generation of high-density atomic beam source, source of x rays and VUV continuum, sample composition analysis, and understanding the hydrodynamics of plasma.[15–18]

The emission characteristics of laser-produced plasma are influenced to a large extent by the surrounding atmosphere.[19–22] There are reports in the literature on the theoretical and experimental aspects of the interaction of the laser-produced plasma with ambient gases.[23,24] Laser-ablated plasmas in an ambient atmosphere have been used for depositing high-quality thin films. High-quality crystalline and stoichiometric TiN films have been obtained by the interaction of Ti laser-produced plasma in presence of nitrogen gas.[25] Presence of C_{60}, C_{70}, and C_{84} fullerenes in laser-ablated carbon have also been reported.[26] The laser-produced plasmas in an ambient gas are also being studied as a gain medium for x-ray lasers.[27]

Laser-induced plasmas from a solid target have been extensively used to study and characterize the clusters of neutral and ionic species. Since the report of Kroto *et al.*[28] on the presence of C_{60} in the mass spectrum of laser-ablated graphite, there has been much interest in laser-ablated graphite.[29] C_{60} clusters are observed to be dominant in laser-evaporation sources when formed in the presence of ambient gases; however, the correlation of carbon clusters with plasma dynamics is not well understood. Creasy and Brenna[30] have observed fullerene ions from laser ablation of diamondlike carbon (DLC) films demonstrating that C_{60} ions can be formed in a variety of systems. The deposited films are usually characterized using mass spectroscopy, x-ray diffraction, uv-visible, Raman spectroscopy, scanning electron microscopy, TEM, etc.

In pulsed-laser deposition, the plasma source plays a key role in controlling the growth, structure, and properties of the deposited films. Optimum DLC films are obtained only above a critical threshold irradiance $\sim10^8$ W/cm^2 where molecular C_2 emission is most dominant. To understand the formation of DLC films, it is necessary to understand the formation and dominance of C_2 species at such low irradiance and its dependence on various parameters such as laser wavelength and pressure of background gas, etc. The presence of ambient gas during deposition effectively enhances the possibility for cluster-

ing and growth reaction. In diamondlike carbon films, the variation in diamondlike character is due to different temperature of various species in the laser plasmas. An estimate of temperature of dominating species of the film helps to optimize the film properties. It has been found that pulsed-laser deposition of high-T_c superconductors using a short-wavelength laser yield superior quality films.[31] We have reported the wavelength dependence of photoablation of carbon at low irradiance in the presence of helium gas.[32] In general, the properties of the carbon films prepared by pulsed-laser ablation vary from soft and graphitic to hard and diamondlike, depending on the energetic particles present in the plasma plume and on the deposition parameters such as laser-power density, laser wavelength, background conditions, and substrate temperature. In the present paper, we report a comprehensive study of the emission characteristics of the laser-produced plasma used for carbon-film deposition in presence of ambient atmosphere at both low and high irradiances. The characterization of emission plume along with the deposited films can yield important information on the pulsed-laser-deposition process and to a better understanding of the deposited films.

II. EXPERIMENTAL DETAILS

The experimental setup used in the present study is similar to the one described elsewhere.[33] A Nd:YAG (yttrium aluminum garnet) laser (DCR-4G) and its harmonics delivering up to 1 J of energy in 8 ns (full width at half maximum) at its fundamental with a repetition rate of 10 pps was used to produce carbon plasma. The laser beam was focused onto the graphite target using a spherical lens of 50 cm focal length and the target rod was continuously rotated and translated with an external motor so that each laser pulse encountered a fresh graphite surface every time. The laser energy was monitored by using a laser power meter (Ophir Model 30A) by placing the power meter in the path of the main beam. Laser energy was varied by changing the voltage on the laser oscillator and amplifier. The target was mounted in a vacuum chamber which could be evacuated well below 10^{-3} Torr and then filled the argon gas at desired pressure in the range 10^{-3}–100 Torr. Plasma radiation was imaged onto the entrance slit of the monochromator (HRS-2, Jobin Yvon) with a lens of 15 cm focal length so as to have one-to-one correspondence with the plasma and its image onto the slit of the monochromator. The output from the monochromator was detected with a photomultiplier tube (1P28, Hamamatsu) and recorded on a strip chart recorder. A microprocessor controlled scan system was used for controlling the scan speed of the monochromator. For thin-film deposition, the laser beam was line focused onto the graphite target enclosed in vacuum chamber using cylindrical lens of focal length 25 cm. The ablated carbon was deposited on silicon substrate placed at about 1 cm away and parallel to the target surface in vacuum and at various ambient gas pressures.

III. RESULTS AND DISCUSSION

The emission spectra of laser-ablated carbon was recorded at both low and high irradiances in the presence of argon gas pressures from 10^{-3} to 100 Torr at various distances from the target. The emission spectra was taken by moving the monochromator in the horizontal plane in a direction perpendicular to the plume direction. The emission lines were identified from the available literature.[34] The plasma emission from the target at higher laser irradiances was found to be dominated by various atomic/ionic species from C IV to C I. The electron temperature of carbon plasma, in vacuum, estimated from the relative intensities of the spectral lines assuming plasma to be in thermal equilibrium lies between 2–10 eV. The electron density estimated by measuring the Stark width was found to be in the range 10^{16}–10^{17} cm^{-3}.[17,26] To see the effect of argon gas on the high-irradiance laser-generated plume, we recorded the emission spectra at various distances away from and parallel to the target surface with 532 nm laser wavelength at 9×10^{10} W/cm^2 laser irradiance. The relative intensity of the plume was found to be sensitive to a range of experimental parameters, particularly laser fluence, the distance from the target surface, and argon gas pressure. The addition of argon during ablation has a strong influence on the plume properties such as change of color and reduction of plume length. The emission at 247.8 nm (C I) and 229.6 nm (C III), corresponding to the transitions ($2p^2\,{}^1S$-$3s\,{}^1p^0$) and ($2p\,{}^1p^0$-$2p^2\,{}^1D$), respectively, are chosen to study the effect of background gas. Figure 1 shows the normalized emission intensity for the above transitions as a function of argon gas pressures at distances 2 and 8 mm from the target surface. The apparent size of the plume as seen visually was about 15 mm in vacuum and reduced to about 3 mm at 100 Torr of argon gas pressure using 532 nm laser wavelength at 9×10^{10} W/cm^2 laser irradiance. It is observed that the intensity of the lines attains a maximum value at some pressure and then decreases with further increase in argon gas pressure. The line emission enhancement can be attributed to various in-

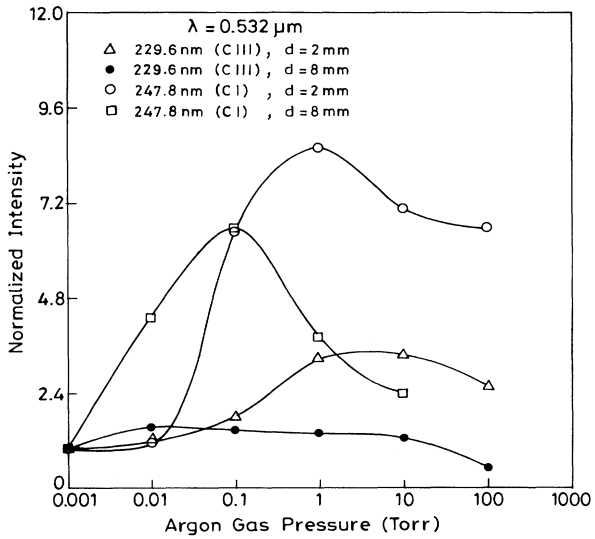

FIG. 1. Dependence of peak intensity of C I (247.8 nm) and C III (229.6 nm) transitions on argon gas pressure for 1.06-μm wavelength at 9×10^{10} W/cm^2 laser irradiance.

teraction processes like collisional excitation, charge transfer, and recombination processes, etc. Our results can be qualitatively explained on the basis of recombination which occur either due to radiative or the three-body recombination (TBR) process. The functional dependence of recombination rate for radiative and TBR can, respectively, be expressed as $n_e n_i \bar{Z}^2 T_e^{-3/4}$ and $n_e^2 n_i \bar{Z}^3 T_e^{-9/2} \ln\sqrt{\bar{Z}^2+1}$ where $\bar{Z}$, n_e, and T_e represent the ion charge, electron density, and electron temperature, respectively.[35] It can be seen that the radiative process is important only close to the target whereas TBR is a dominant process beyond a few mm from the target surface. The background gas basically provides a heat sink so that the recombination can continue for a longer period. The observed decrease in intensity of emitted lines at higher pressures is due to a very high cooling rate. Also, the excitation of molecules of background gas results in reducing the electron energy, increasing collisional cooling, which in turn increases the TBR rate. This results in populating the excited neutral and ionic carbon species in the presence of background gas.

Figure 2 shows the variation of the line intensity of the C I transition at 247.8 nm and the C III transition at 229.6 nm with distance from the target surface at various argon pressures. The intensity of both the transitions is found to increase as the pressure of argon increases from 10^{-3} to 1 Torr but decrease on increase in pressure beyond 1 Torr. In vacuum, the plasma expands freely; however, it is confined to a small region in the presence of background gas which results in reduced expansion rate and hence enhanced cooling rate. Similar results have been reported for excimer-laser[21] and nitrogen-laser[36] ablated plasmas.

At low irradiance ($\sim 10^8$ W/cm^2), the spectrum is dominated by C_2 emission in the $\Delta v = -2, -1, 0, +1, +2$ sequence of the Swan ($d^3\pi_g$-$a^3\pi_u$) bands and $\Delta v = 0$ and $+1$ sequence of the Deslandres-d'Azambuja ($C^1\pi_g$-$A^1\pi_g$) bands.[37] No cometary bands at 405 nm were seen with or without argon gas in our experiment, though, few C I and C II lines were present in addition to molecular C_2 bands. All the C_2 Swan band heads have been recorded which include $\Delta v = -2, -1, 0, +1, +2$ where $\Delta v = v' - v''$ is the difference of the vibrational quantum number between the upper (v') and lower (v'') states of transition. The C_2 d-a $\Delta v = 0$ Swan band sequence in vacuum consisted of Swan band heads (0-0) at 516.5 nm, (1-1) at 512.9 nm, and (2-2) at 509.7 nm only, while for $\Delta v = 1$, the bands (1-0) at 473.7 nm, (2-1) at 471.5 nm, (3-2) at 469.7 nm, (4-3) at 468.4 nm, and (5-4) at 467.8 nm are found to be prominent. We also observed the $\Delta v = -2$ Swan band sequence with the Swan band heads of (0-2) at 619.1 nm, (1-3) at 612.2 nm, (2-4) at 605.9 nm, (3-5) at 600.4 nm, and (4-6) at 595.9 nm. The emission spectra obtained are in close agreement with those observed during laser ablation of graphite using KrF (248 nm) excimer laser by Chen, Mazumdar, and Purohit.[38] The only difference is that we observed a few ionic lines at an irradiance level well below their threshold value. The appearance of the C_2 bands at such low irradiance suggest that they are probably formed from atomic carbon recombination or due to electron impact excitation of ground-state C_2 molecules emitted directly from the target or formed from the fragmentation of higher clusters in the plasma.

Figures 3 and 4 show the emission spectra of C_2 $\Delta v = -1$ Swan band sequence for 1.06-μm laser wavelength at 10^{-3} and 10^{-1} Torr of argon gas at 22, 33, 44, and 66 mJ of laser energies, respectively. The laser irradiance was obtained from the measured spot area 1.13×10^{-2} cm^2 and pulse duration 8 ns. The spectra were recorded at a distance of 3 mm away and parallel to the target surface. It is observed that the intensity of band heads first increases with energy, attains a maximum value, and then decreases. Film deposition at this optimum energy may optimize the diamondlike film properties. Similar variation in C_2 emission intensity with laser energy was observed using 0.532- and 0.355-μm laser wavelengths. Figure 5 shows the intensity of C_2

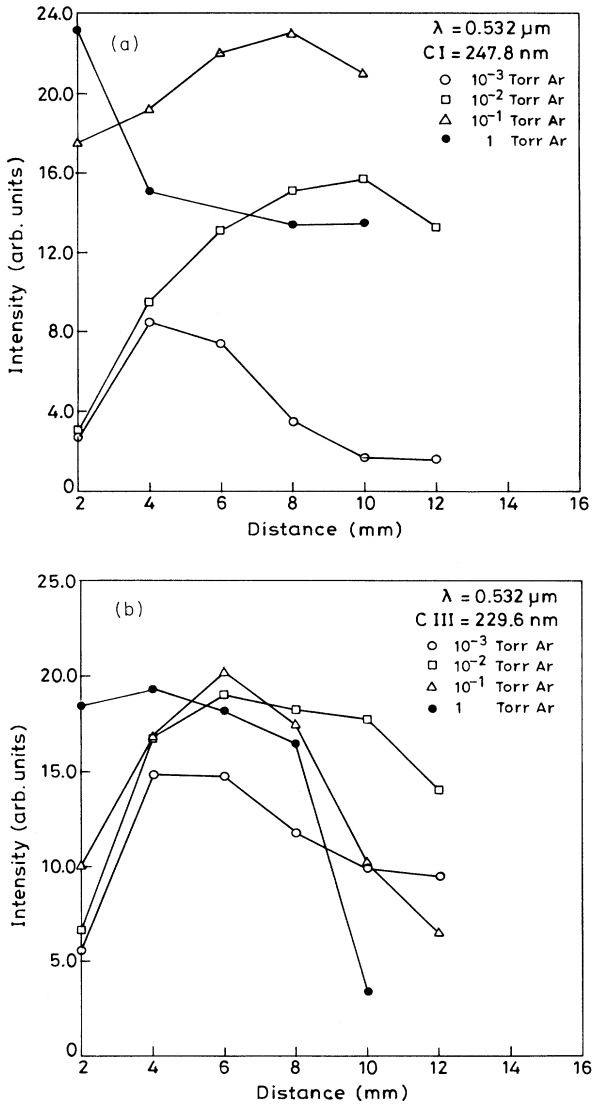

FIG. 2. Variation of intensity with distance from the target surface at various argon gas pressures for the transitions (a) C I (247.8 nm), (b) C III (229.6 nm) for 1.06-μm wavelength at 9×10^{10} W/cm^2 laser irradiance.

Swan band heads of $(v'-v'')$, i.e., (0-1) at 563.5 nm and (1-2) at 558.5 nm for 0.532- and 0.355-μm laser wavelengths at 10^{-1} Torr of argon gas, recorded at a distance of 3 mm away and parallel to the target surface. For all the wavelengths, the intensity peaks at some intermediate energy. Also the peak intensity maximum shifts towards low energies with decrease in wavelength which may be due to the photofragmentation of higher clusters in plasma. The mass-ablation rate has also been reported to increase at shorter wavelengths.[39]

To see the effect of ambient gas on molecular C_2 emission, emission spectra of C_2 Swan bands was recorded at various argon gas pressures at different laser energies. Figure 6 shows the C_2 Swan band spectra at 10^{-1}, 1, and 10 Torr of argon gas pressures at 33 mJ of 1.06-μm laser wavelength, recorded at a distance of 3 mm from the target surface. The intensity of bands increased as the pressure of the argon gas increased from 10^{-3} to 100 Torr. Thus the incorporation of the ambient gas helps to cool the molecular species and increase the recombination rate. Figure 7 shows the variation in intensity of Swan band head, i.e., (0-1) at 563.5 nm for 1.06-, 0.532-, and 0.355-μm laser wavelengths in pressure range of 10^{-3}–100 Torr of argon gas, recorded at 3 mm from the target surface. The intensity increases with pressure for

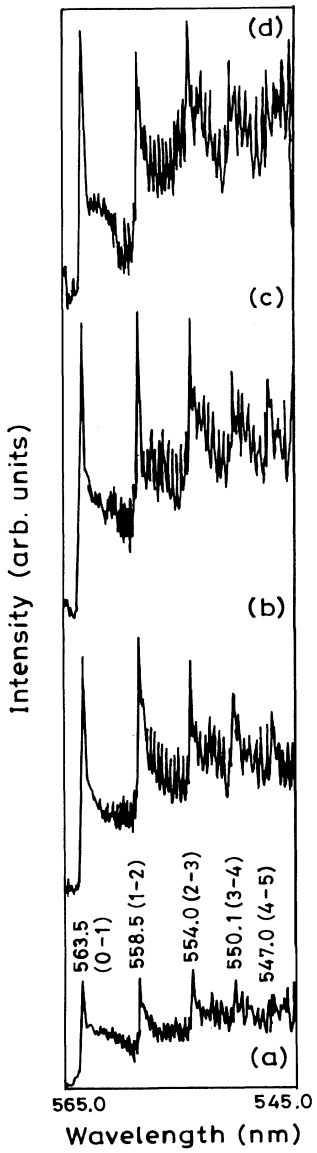

FIG. 3. C_2 *d-a* Swan band sequence $\Delta v = -1$ at 10^{-3} Torr of argon gas pressure at (a) 22, (b) 33, (c) 44, and (d) 66 mJ of laser energy for 532-nm laser wavelength, recorded at a distance of 3 mm from the target surface.

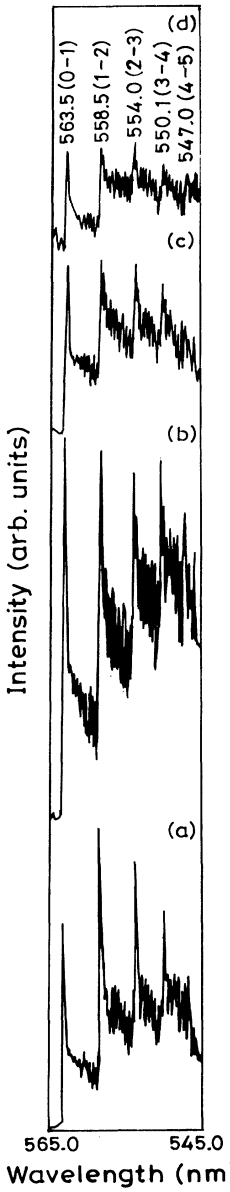

FIG. 4. C_2 *d-a* Swan band sequence $\Delta v = -1$ at pressure of 10^{-1} Torr of argon gas at (a) 22, (b) 33, (c) 44, and (d) 66 mJ of laser energy. The intensity in (c) and (d) is twice that of (a) and (b). The spectra is recorded at a distance of 3 mm away and parallel to the target surface.

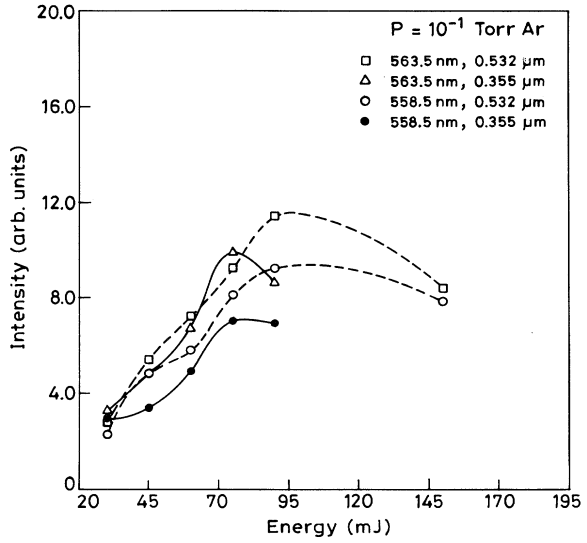

FIG. 5. Intensity of C_2 Swan band heads (0-1) at 563.5 nm and (1-2) at 558.5 nm at various laser energies of 0.532- and 0.355-μm laser wavelengths at 10^{-1} Torr Ar pressure, recorded at a distance of 3 mm from the target surface.

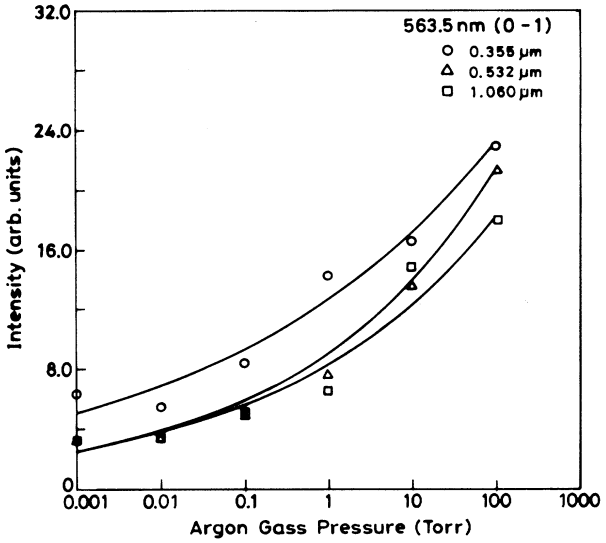

FIG. 7. Variation in intensity of Swan band heat (0-1) at 563.5 nm as a function of argon gas pressure for 1.06-, 0.532-, and 0.355-μm laser wavelengths, recorded at a distance of 3 mm from the target surface.

all the laser wavelengths. The observed intensity of the band is largest at 0.355 μm at all pressures. This probably is due to a change in optical penetration depth which decreases with wavelength. The decreased volume of the material with which the laser can interact results in a more effective coupling to the target. We have also estimated the vibrational temperature of C_2 bands at various argon gas pressures using different laser energies.

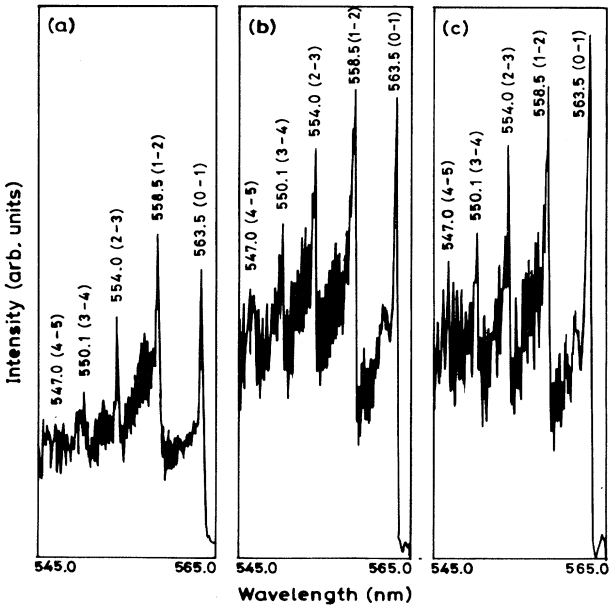

FIG. 6. C_2 d-a, $\Delta v = -1$ Swan band sequence at argon gas pressures of (a) 10^{-1}, (b) 1, and (c) 10 Torr for 1.06-μm laser wavelength, recorded at a distance of 3 mm from the target surface.

Band head intensities[40] of C_2 Swan bands have been used for estimating the vibrational temperature. The relative population in each vibrational level can be found using theoretical Franck-Condon factors which in our case are from Spindler.[41] The relative population of the upper level as derived from the measured intensities was plotted against the vibrational quantum number, the slope of the curve gives the vibrational temperature. Figure 8 shows the relative population of the upper vibrational level against vibrational quantum number at 10^{-1} Torr of argon gas pressure at various laser energies of 0.532-μm

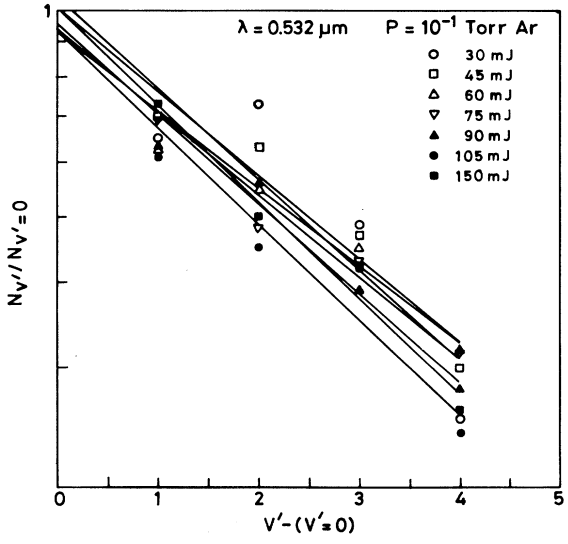

FIG. 8. Relative population of the upper vibrational level of C_2 Swan band sequence $\Delta v = -1$ against vibrational quantum number for 10^{-1} Torr argon gas pressure at various laser energies of 0.532 μm.

laser wavelength, the slope gives the vibrational temperature.

Figure 9 shows the variation of vibrational temperature at 10^{-1} Torr of argon gas pressure with laser energy for 0.532- and 0.355-μm laser wavelengths. The vibrational temperature is maximum at an optimum laser energy. The results are consistent with an earlier report on vibrational temperature by Chen, Mazumdar, and Purohit.[38] It has been observed by various investigators[9] that the degree of diamondlike character in diamondlike carbon films varies due to different temperatures in the laser plasma, the deposition of thin films at this optimum energy may help in optimizing the film quality. It is also evident from Fig. 9 that a higher vibrational temperature is obtained by short-wavelength laser irradiation which may be due to small absorption depth of the uv photon and to higher absorption by the ablated fragments. UV laser-deposited carbon films are reported to be superior to those prepared by long-wavelength lasers with regard to their mechanical hardness and optical properties.[42]

To see the effect of ambient environments the measurements were also carried out in the presence of helium gas. Figure 10 shows the variation of the relative intensities of C_2 d-a $\Delta v = -1$ Swan band heads at 563.5 nm (0-1) and 558.5 nm (1-2) using 0.355-μm laser at 40 mJ of laser energy at various helium and argon gas pressures. The spectra was recorded at a distance of 3 mm away and parallel to the target surface. The intensities of the band heads are found to increase with pressure of the ambient gases; however, the enhancement is more pronounced in presence of argon gas. The vibrational temperatures calculated from C_2 Swan band heads are also found to be larger in the case of argon than that in helium. This difference could be due to the confinement effect. The atomic mass of argon is larger than that of helium. Therefore, a greater restriction force will be exerted by argon on the plume expansion than in helium, and hence more intense emission in the presence of argon than helium. Figure 11 shows the variation of C_2 Swan band heads intensity as a function of 0.355-μm laser energy at 10^{-1} Torr of helium and argon gas pressure, recorded at a distance of 3 mm from the target surface. It can further be seen that the intensity, in presence of argon gas, is more than that in helium at all laser energies and increases with increase of laser energy.

Carbon films deposited on silicon and glass substrates in the presence of helium and argon gas using 0.532- and 0.355-μm wavelengths were characterized by x-ray diffraction and scanning electron microscopy.[43] The

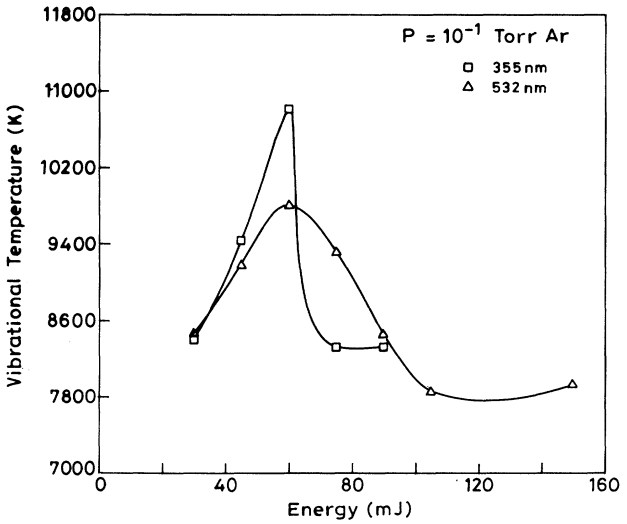

FIG. 9. Vibrational temperature calculated from C_2 Swan bands as a function of laser energy for 0.532- and 0.355-μm laser wavelengths at 10^{-1} Torr of argon.

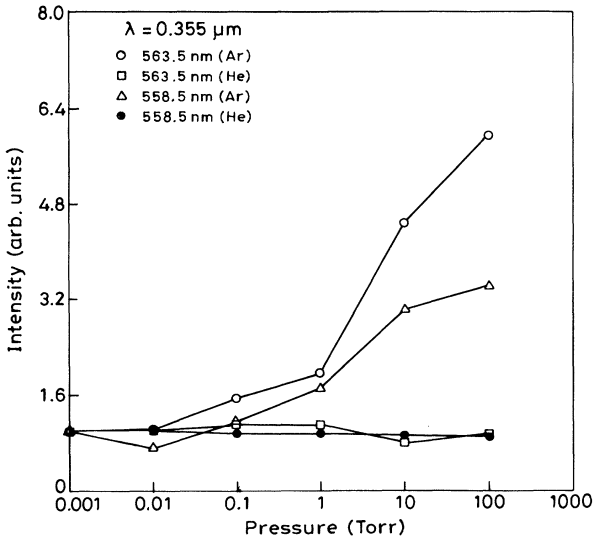

FIG. 10. Intensity of C_2 Swan band heads, (0-1) at 563.5 nm and (1-2) at 558.5 nm for 0.355-μm laser at various helium and argon pressures, recorded at a distance of 3 mm from the target surface.

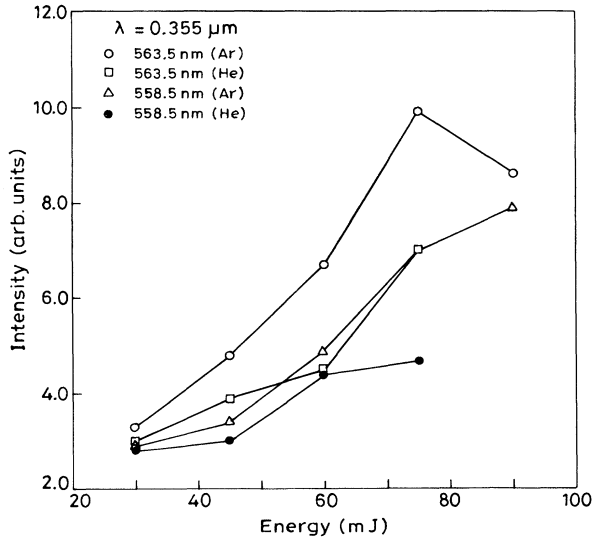

FIG. 11. Dependence of C_2 Swan band heads intensity at 563.5 nm (0-1) and 558.5 nm (1-2) with laser energy at 10^{-1} Torr of helium and argon gas pressures, recorded at a distance of 3 mm from the target surface.

structural properties of the deposited films were investigated in order to determine the crystalline orientation. The x-ray diffraction pattern of the carbon films deposited on silicon substrate gave peaks at 10.3°, 11°, 17.9°, and 21° showing the presence of (100), (002), (110), and (112) crystalline planes of C_{60}. The observed peaks showed a strong dependence on pressure. The intensity of the peaks was found to be maximum at 100 Torr of helium gas pressure. The variation in the surface structure of the films with ambient gas pressures was studied by scanning electron microscopy. We observed a marked difference in the composition and morphology of the films with the pressure of the ambient gas. It is found that the density of the microcrystalline clusters increases with ambient gas pressures and attains a maximum at an intermediate pressure. It is attributed to cooling of plume particles in the presence of background gas. The variation in morphology may be due to the variation of temperature in laser-ablated carbon plasma at various ambient gas pressures.

IV. CONCLUSIONS

The investigations demonstrate that the formation of atomic, ionic, and molecular species is sensitive to laser irradiance, laser wavelength, and pressure of background gas. The plasma emission is dominated by molecular C_2 at low irradiance while various atomic/ionic species dominate the emission at higher irradiances. It is found that the plume emission can be enhanced significantly in the presence of argon gas. The molecular C_2 emission observed could be due to either plasma excitation at low irradiances or recombination processes at high irradiances. The intensity of C_2 Swan band heads increases with decrease in laser wavelength. The vibrational temperature calculated from C_2 Swan bands is maximum at intermediate laser energy. The deposited films showed a strong dependence on ambient gas pressures. The density of microcrystalline clusters increases with ambient gas pressure and shows a maximum at an intermediate pressure. These studies are helpful for correlation of plume carbon cluster formation with plasma dynamics and film quality.

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[1]R. W. Dreyfus, R. Kelly, and R. E. Walkup, Appl. Phys. Lett. **49**, 1478 (1986).
[2]R. Viswanathan and I. Hussla, J. Opt. Soc. Am. B **3**, 796 (1986).
[3]M. Berti, L. F. Dona dalle Rose, A. V. Drigo, C. Cohen, J. Siejka, G. G. Bentini, and E. Jannitti, Phys. Rev. B **34**, 2346 (1986).
[4]J. T. C. Yeh, J. Vac. Sci. Technol. A **4**, 653 (1986).
[5]*Laser-Induced Plasmas and Applications,* edited by L. J. Radziemski and D. A. Cremers (Dekker, New York, 1989).
[6]K. Dittrich and R. Wennrich, Prog. Anal. Atom. Spectrosc. **7**, 139 (1984).
[7]T. P. Hughes, *Plasmas and Laser Light* (Wiley, New York, 1975).
[8]E. B. D. Bourdon, W. W. Duley, A. P. Jones, and R. H. Prince, in *Diamond and Diamondlike Films and Coatings,* edited by R. E. Clausing *et al.* (Plenum, New York, 1991).
[9]D. L. Pappas, K. L. Saenger, J. Bruley, W. Krakow, and J. J. Cuomo, J. Appl. Phys. **71**, 5675 (1992).
[10]T. Sato, S. Furuno, S. Iguchi, and M. Hanabusa, Jpn. J. Appl. Phys. **26**, L1487 (1987).
[11]C. Champeaux, P. Marchet, J. Aubreton, J.-P. Mercurio, and A. Catherinot, Appl. Surf. Sci. **69**, 335 (1993).
[12]E. Dyer and R.-J. Farlay, J. Appl. Phys. **74**, 1442 (1993).
[13]E. A. Rohlfing, D. M. Cox, and A. Caldor, J. Chem. Phys. **81**, 3322 (1984).
[14]U. Naher, H. Gohlich, T. Lange, and T. P. Martin, Phys. Rev. Lett. **68**, 3416 (1992).
[15]H. Katsuragawa and T. Minowa, Nucl. Instrum. Methods **43B**, 289 (1989).
[16]B. Soom, H. Chen, Y. Fisher, and D. D. Meyerhofer, J. Appl. Phys. **74**, 5372 (1993).
[17]Abhilasha, P. S. R. Prasad, and R. K. Thareja, Phys. Rev. E **48**, 2929 (1993).
[18]G. Mehlman, D. B. Chrisey, P. G. Burkhalter, J. S. Horwitz, and D. A. Newman, J. Appl. Phys. **74**, 53 (1993).
[19]V. Kumar and R. K. Thareja, J. Appl. Phys. **67**, 3260 (1990).
[20]Y. Iida, Appl. Spectrosc. **43**, 229 (1989).
[21]H. P. Gu, Q. H. Lou, N. H. Cheung, S. C. Chen, Z. Y. Wang, and P. K. Lin, Appl. Phys. **58B**, 143 (1994).
[22]C. Timmer, S. K. Srivastava, T. E. Hall, and A. F. Fucaloro, J. Appl. Phys. **70**, 1888 (1991).
[23]O. B. Anan'in, Y. A. Bykovskii, E. L. Stupitskii, and A. M. Khudaverdyan, Sov. J. Quantum. Electron. **17**, 1474 (1987).
[24]T. P. Wright, Phys. Rev. Lett. **28**, 268 (1972).
[25]J. C. S. Kools, C. J. C. M. Nillesen, S. H. Brongersma, E. Van de Riet, and J. Dieleman, J. Vac. Sci. Technol. A **10**, 1809 (1992).
[26]P. S. R. Prasad, Abhilasha, and R. K. Thareja, Phys. Status Solidi A **139**, K1 (1993).
[27]C. Steden and H. J. Kunze, Phys. Lett. A **151**, 534 (1990).
[28]H. W. Kroto, J. R. Heath, S. C. O'Brian, R. F. Curl, and R. E. Smalley, Nature **162**, 318 (1985).
[29]G. Meijer and D. S. Bethune, J. Chem. Phys. **93**, 7800 (1990).
[30]W. R. Creasy and J. T. Brenna, J. Chem. Phys. **92**, 2269 (1990).
[31]G. Koren, A. Gupta, R. J. Baseman, M. I. Lutwyche, and R. B. Laibowitz, Appl. Phys. Lett. **55**, 2450 (1989).
[32]Abhilasha, R. K. Dwivedi, and R. K. Thareja, J. Appl. Phys. **75**, 8237 (1994).
[33]R. Tambay, R. Singh, and R. K Thareja, J. Appl. Phys. **72**, 1197 (1992).
[34]A. R. Striganov and N. S. Sventitskii, *Tables of Spectral Lines of Neutral and Ionized Atoms* (Plenum, New York, 1968).
[35]P. T. Rumsby and J. W. M. Paul, Plasma Phys. **16**, 247 (1974).
[36]K. Kagawa, O. Ohtani, S. Yokoi, and S. Nakajima, Spectrochim. Acta **39B**, 525 (1984).
[37]E. A. Rohlfing, J. Chem. Phys. **89**, 6103 (1988).
[38]X. Chen, J. Mazumdar, and A. Purohit, Appl. Phys. A **52**, 328

(1991).
[39]R. Fabbro, E. Fabre, F. Amiranoff, C. Garban-Labaune, J. Virmont, M. Weinfeld, and C. E. Max, Phys. Rev. A **26**, 2289 (1982).
[40]L. L. Danylewych and R. W. Nicholls, Proc. R. Soc. London Ser. A **339**, 197 (1974).
[41]R. J. Spindler, J. Quant. Spectrosc. Radiat. Transfer **5**, 165 (1965).
[42]F. Xiang and R. P. H. Chang, in *Novel Forms of Carbon,* edited by C. L. Renschler, J. J. Pouch, and D. M. Cox, MRS Symposia Proceedings No. 270 (Materials Research Society, Pittsburgh, 1992), p. 451.
[43]S. Sen, M. Tech. thesis, Indian Institute of Technology, Kanpur, 1994.