Appl. Phys. B 61, 63–67 (1995)

Applied Physics B Lasers and Optics

Line-emission enhancement in laser-ablated carbon plasmas

Abhilasha, R. K. Thareja

Department of Physics and Centre for Laser Technology, Indian Institute of Technology, Kanpur 208 016, India
(Fax: + 91-512/25-0260, E-mail: THAREJA@iitk.ernet.in)

Received: 9 May 1994/Accepted: 13 September 1994

Abstract. The role of recombination and charge-exchange processes in enhancing the line emission of various ionic species of laser-ablated carbon in the presence of helium and argon are presented.

PACS: 52.50.Jm; 52.40.Nk; 52.25. – b; 52.20. – j

A high-density, high-temperature plasma is created when a Q-switched laser is focussed onto a solid target which subsequently expands with a transfer of the initial thermal energy of the constituent ionic species and electrons to the kinetic energy of radial expansion [1]. In the presence of a low-pressure background gas, the radiation from the laser-ablated plasma photoionizes the gas, and the plasma streams radially outward through the resulting ambient plasma. There has been great interest in studying the interaction between charged ions emanating from expanding laser-ablated plasmas with a background-gas medium [2, 3]. Various interaction processes such as recombination, charge exchange, momentum transfer, micro-instabilities, etc. play an important role in line emission [4–6] and are of great interest in the understanding the hydrodynamics of the expanding plasma [7–9], in exploring the possibility of laser-generated controlled thermonuclear reaction [10] and in sample-composition analysis [11]. Hot, dense laser-produced plasmas have been used as a source of high intensity X-rays [12], VUV continuum in rare gases and highly charged ions [13, 14]. Recently much interest is being shown in laser-produced plasmas as a gain medium for X-ray lasers [15, 16] and for deposition of thin films [17]. Seely et al. [18] have used a charge-exchange scheme between C VI and Ar III in expanding carbon plasma for soft X-ray laser in carbon. Qi et al. [19] have demonstrated laser action in C III ions at 217.7 nm and 216.3 nm by resonant photoexcitation where Mn line radiation from a laser-produced Mn plasma was used to resonantly pump the C III ions.

We have earlier reported [3] laser-ablated carbon plasma studies in vacuum and in air. In the present paper we report on spectroscopic studies on both spatial and temporal behaviour of the emission spectrum of laser-ablated carbon plasma in the presence of helium and argon gas. The role of recombination and a charge-exchange process in enhancing the line emission of various ionic species of carbon in presence of ambient gas are discussed. It is noticed that the presence of argon gas significantly enhances the intensity of the C III transition $3d^1D - 4f^1F^0$ at 216.3 nm. These studies are helpful in the diagnostics of the plasma gas-interaction regime, in laser microspectral analysis and in the development of X-ray or UV lasers where the background gas is used for populating the excited states of the ions.

1 Experimental setup

Experimental setup used in the present study is similar to the one described elsewhere [20]. We employed a Nd:YAG laser (DCR-4G) with a Gaussian-limited mode structure (1.06 μm) delivering upto 1 J of energy in 8 ns (FWHM) with a repetition rate of 10 pps to produce the carbon plasma. A 50 cm focal length lens focussed the laser beam onto the graphite target to a spot of 240 μm diameter. The target was mounted in a vacuum chamber which could be evacuated to a pressure better than 10^{-3} Torr and then filled in with helium and argon gases at desired pressures in the range of 10^{-3} to 100 Torr. The target rod was continuously rotated and translated with an external motor so that each laser pulse falls on fresh graphite surface every time. Plasma radiation was imaged onto the entrance slit of the monochromator (HRS-2, Jobin Yvon) with a lens of focal length of 15 cm so as to have one-to-one correspondence with the plasma and its image onto the slit of monochromator. The output from the monochromator was detected with a photomultiplier tube (PMT) (1P28, Hamamatsu, Japan) and recorded on a strip chart recorder or displayed on the counter timer oscilloscope (11302A, Tektronix). A micro-processor

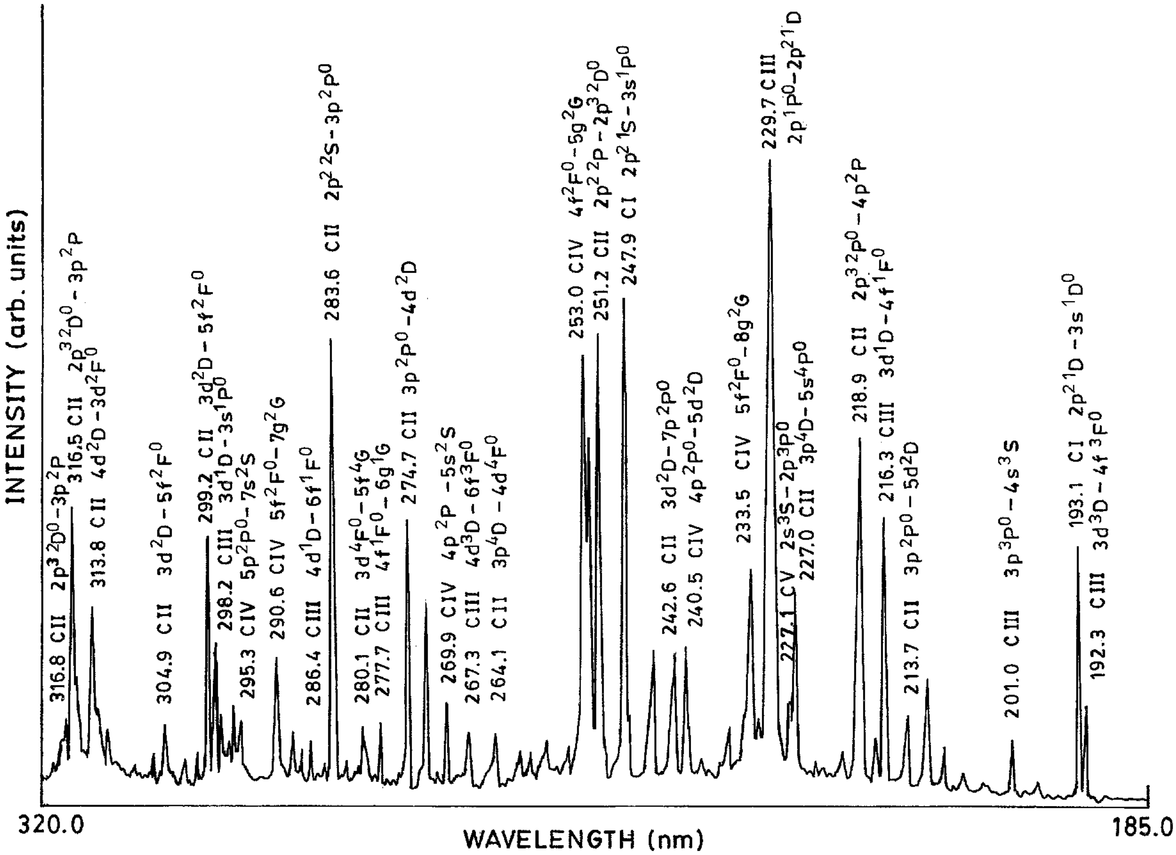

Fig. 1. Typical emission spectrum in the wavelength range 190–300 nm in vacuum

controlled scan system was used for controlling the scan speed of the monochromator. The signals were digitized using a digitizing video camera (Tektronix, DCS 01) attached to the oscilloscope, digitized signals were fed to personal computer for further data processing.

2 Results and discussion

The emission spectra of carbon plasma in the wavelength range 190–800 nm in vacuum, and in the presence of helium and argon gases were recorded at different distances (z) away from the target surface by moving the monochromator in the horizontal plane in the direction perpendicular to the direction of the expanding plume. It was made sure that the background gas does not breakdown and absorb the laser radiation [21]. Figure 1 shows a typical emission spectrum in the wavelength range 190–300 nm in vacuum. The emission lines were identified using the information available in the literature [22]. Transitions up to C V have been observed. The intensity of the emitted lines increased in the presence of a background gas [23]. A detailed investigation on C I, C II and C III transitions which have applications for laser [4, 18, 19, 26] was undertaken. Figure 2 exhibits normalized intensity as a function of distance from the target for the transitions $2p^2\,{}^1D - 3s^1D^0$ at 193.1 nm of C I, $3p^2P^0 - 5d^2D$ at 213.7 nm of C II and $3d^2D - 4f^3F^0$ at 192.3 nm of C III species at a pressure of $\leq 10^{-3}$ Torr. Every point in the figures to follow is an average of five observations. It is observed that for the C III specy, the line intensity peaks at 3 mm from the target surface and decreases rapidly as z increases whereas for the C II and C I species the intensity is maximum at z = 4 mm from the target and decreases slowly as z decreases. That is to say that the intensity of higher species decreases rapidly as they recombine to give lower species. Figure 3 displays the variation of line intensity of the C I transition $2p^2\,{}^1D - 3s^1D^0$ with distance from the target at various helium gas pressures ranging from 10^{-3} to 10 Torr. It is observed that line intensity is higher in the presence of helium gas than that in vacuum. The intensity of the C I specy increases when the helium pressure is increased from 10^{-3} to 1 Torr but decreases for 10 Torr helium pressure. In vacuum, the plasma expands freely whereas in the presence of background gas the plasma is confined to a small region which results in a reduced expansion rate and hence an enhanced cooling rate. The rate of change of electron temperature is sum of three terms [24], viz. the elastic collision, electron heating due to collisional de-excitation of metastable ions and the recombination of ions. The rate of loss of electron energy is dominated by the elastic collision term $Q_{\Delta T}$ given by

$$Q_{\Delta T} = \frac{2m_e \sigma_{ea} n_B}{M_B} \left(\frac{8kT_e}{\pi m_e}\right)^{1/2}, \tag{1}$$

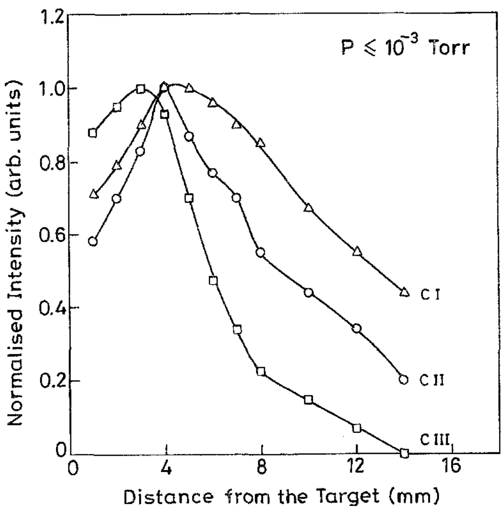

Fig. 2. Normalized intensity as a function of distance from the target for transitions $2p^{2\,1}D - 3s^1D^0$ at 193.1 nm of CI, $3p^2P^0 - 5d^2D$ at 213.7 nm of C II and $3d^3D - 4f^3F^0$ at 192.3 nm of C III species at a pressure of less than 10^{-3} Torr

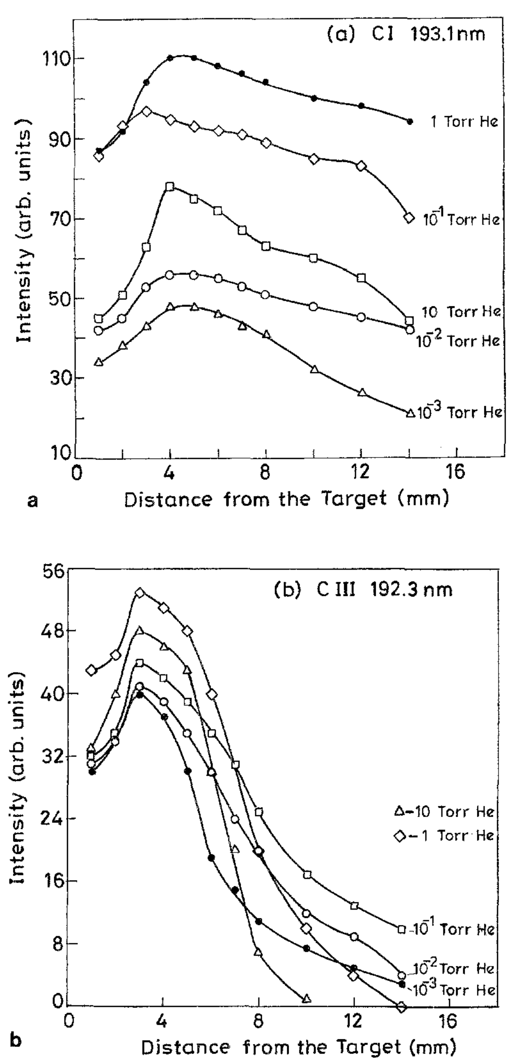

Fig. 3a, b. Variation of line intensity of (**a**) C I transition $2p^2\,{}^1D$–$3s^1D^0$ and (**b**) C III transition $3d^3D$–$4f^3F^0$ with distance from the target at various helium pressures

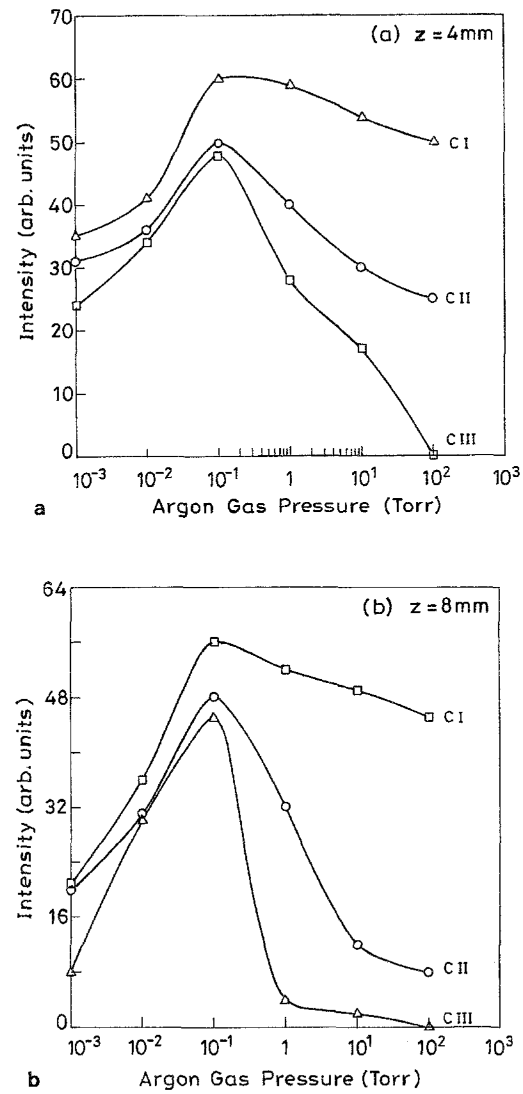

Fig. 4a, b. Intensities of C I transition $2p^2\,{}^1D$–$3s^1D^0$ at 193.1 nm. C II transition $3p^2P^0 - 5d^2D$ at 213.7 nm and C III transition $3d^3D$–$4f^3F^0$ at 192.3 nm at a distance of (**a**) 4 mm and (**b**) 8 mm from the target surface

where σ_{ea} is the elastic scattering cross-section between the electrons and the atoms, n_B is the density of background gas, and M_B is the mass of the background gas atom. It follows from (1) that cooling is inversely proportional to M_B, and hence lighter gases are efficient for rapid cooling. It is also observed that as the gas pressure increases beyond a certain point, 1 Torr in our case, the enhancement slows down. Timmer et al. [25] have also observed a similar behaviour for an A1 I transition in N_2 gas. Figure 3b shows the variation of line intensity of the C III transition $3d^3D - 4f^3F^0$ with distance from the target. It is observed that as the gas pressure is increased from 10^{-3} to 1 Torr, the intensity increases and peaks at about 3 mm from the target, but increasing the pressure from 1 Torr to 10 Torr decreases the line intensity.

The studies were also done with argon as a background gas. Argon has been used for pumping of soft X-ray lasers in carbon [18]. Figure 4a exhibits the intensities of the C I transition $2p^2\,{}^1D - 3s^1D^0$ at 193.1 nm, the

C II transition $3p^2P^0 - 5d^2D$ at 213.7 nm and the C III transition $3d^3D - 4f^3F^0$ at 192.3 nm at a distance of 4 mm from the target at various argon pressures. For C I and C II species, the intensity increases rapidly upto 0.1 Torr and decreases slowly beyond 0.1 Torr. Whereas the C III line decreases rapidly beyond 0.1 Torr of argon gas pressure, decaying completely at 100 torr. Figure 4b shows the intensities of C I, C II and C III lines at a distance of 8 mm from the target at various argon pressures. It is observed that the intensity of the C III transition is almost insignificant beyond 1 Torr of argon pressure. This reveals that the presence of background gas affects higher species most. Qi et al. [26] observed enhanced fluorescence at 217.7 nm, 216.3 nm and 192.3 nm in C III ions by photoexcitation with the Mn VI line radiation at 31 nm. We observed a significant enhancement of the intensity of the C III transition $3d^1D - 4f^1F^0$ at 216.3 nm in the presence of argon gas. Figure 5 exhibits the intensity variation of the C III transition at 216.3 nm with the distance from the target at various argon pressures. It is observed that the line intensity reaches a maximum value at a pressure of about 1 Torr at 4 mm from the target surface, and then decreases with further increase in pressure upto 100 Torr. The curve shows that with the increase in argon pressure cooling of the gas increases rapidly. The decrease of intensity at high pressure is due to very high cooling rate. We observed more line enhancement for this transition in the presence of argon than in the presence of helium gas. It agrees well with the earlier results [18] where it is shown that an interaction process like charge exchange between C VI and Ar III in expanding carbon plasma can result in soft X-ray lasing. The temporal profiles of the C III transition $3d^1D - 4f^1F^0$ at 216.3 nm where recorded at different distances and at various argon pressures. Figure 6 depicts the variation of time delay in the peak-emission intensity at various distances and for various argon pressures. The velocity of plasma emission front decreases with the increase in collisional frequency as argon pressure is increased. Moreover, the confinement of the plasma in the presence of argon gas increases the duration of emission thereby decreases the velocity.

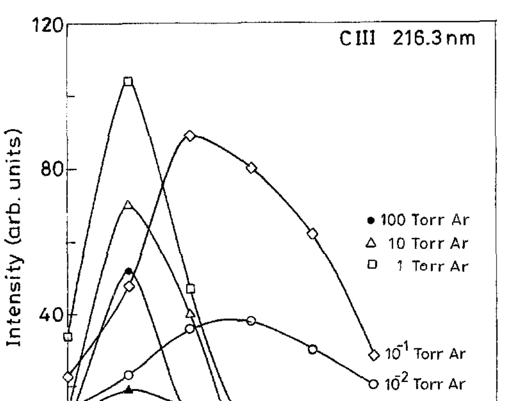

Fig. 5. Intensity variation of C III transition $3d^1D$–$4f^1F^0$ at 216.3 nm with distance from the target at various argon pressures

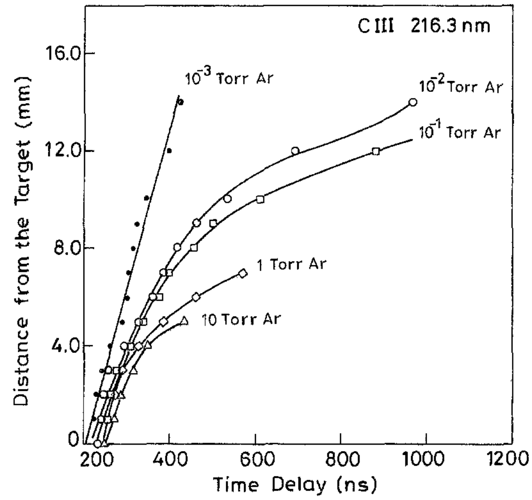

Fig. 6. Variation of time delay in the peak emission intensity with distance from the target for various argon pressures

The significant enhancement of emission lines in the presence of background gas indicates an important role of the gas in the interaction processes. Various interactions which can result in populating the upper state involved in the observed transitions could be collisional excitation, charge-transfer and the recombination process. A dense carbon plasma expands adiabatically as the electron thermal energy is converted into streaming motion of the ions. In our studies, we observed the emission up to an axial distance of 15 mm, whereas the collisional excitation process is found to dominate in the region $z < 1$ mm from the target surface [27, 28]. Furthermore, the collisional excitation from the ground state requires very large excitation energy to populate the upper states. Hence, the possibility of collisional excitation process playing the main role in enhancing the line emission can safely be ruled out. The charge transfer between the plasma ions and the atoms of the background gas can also populate the excited high − n states. It was experimentally found that for ion streaming velocity in the range of 10^7 cm/s, the charge-transfer rate [18] is of the order of $R \cong (10^{-11}\ \mathrm{cm^3/s})N_1$ where N_1 is the density of C III ions. It follows that due to plasma expansion the charge transfer rate decreases as r^{-3}, where r is the radial distance away from the target. The calculations have shown that the charge-transfer process dominates beyond $z > 1$ cm [18, 29]. This is because the dense hot plasma near the target surface is a strong source of continuum radiation which can easily be absorbed in the background gas and can ionize the background gas. The small difference in the charge state of the plasma ions and the gas atoms reduces the probability of charge transfer. Moreover, in our experiment, it was difficult to ascertain the exact charge-transfer process as the onset of shock and radiation heating of the background gas greatly limit the density at which the charge-transfer interaction can take place.

It is found that recombination processes are much more likely to occur due to the presence of low-energy electrons from the ionization of the background gas. The recombination can occur either due to the radiative or the three-body recombination process [30, 31]. The recombination rates for the radiative and three-body recombination are $2.7 \times 10^{-13} n_e n_1 \bar{Z}^2 T_e^{-3/4}$ cm^3/s and $9.2 \times 10^{-27} n_e n_i^2 \bar{Z}^3 T_e^{-9/2} \ln (\bar{Z}^2 + 1)^{1/2}$ cm^6/s, respectively, where n_e and n_1 are the electron and ion densities, respectively T_e is the electron temperature in eV and $\bar{Z}$ is the ionic charge. It is seen that the radiative process is important only close to the target, whereas the three-body recombination remains the dominant recombination process beyond a few millimeters from the target surface [32] during adiabatic expansion. The density of plasma decreases on expansion, but still a significant rate of the three-body recombination can exist at distances of a few mm due to simultaneous reduction in temperature of the adiabatically expanding plasma. The rate of the three-body recombination, however, starts decreasing at much larger distances when the recombination heating [33] becomes important, resulting in a slower decrease of temperature during expansion and leading to a freezing of the ionization [30]. The background gas essentially provides a heat sink so that the recombination process can continue for a longer period. Theoretical estimates [33] suggest that a background gas at a pressure of 0.1–10 Torr can significantly increase the cooling rate of electrons in the plasma region of 1–10 mm away from the target. Excitation of the molecules of the background gas results in reducing the electron energies thereby increasing collisional cooling. Thus, it follows from (1) that the role of the background gas lies in increasing the cooling rate of the plasma in the expansion region. Enhanced cooling of the expanding plasma increases the three-body recombination rate, resulting in populating the excited neutral and ionic carbon species in the presence of a background gas. Hence, the three-body recombination process is mainly responsible for the enhancement of the emission lines.

3 Conclusions

Laser-ablated carbon-plasma studies in presence of helium and argon indicated the line emission enhancement of various ionic species of carbon. The role of various interaction processes in enhancing the line emission was discussed. The C III transition at 216.3 nm showed significant enhancement in the presence of argon gas and therefore, the spatial and temporal behaviour of this transition was studied.

Acknowledgements. The work is partly supported by the Department of Science and Technology, New Delhi.

References

1. J.M. Dawson: Phys. Fluids **7**, 981 (1964)
2. D.W. Koopmen: Phys. Fluids **14**, 1707 (1971)
3. Abhilasha, P.S.R. Prasad, R.K. Thareja: Phys. Rev. E **48**, 2929 (1993)
4. R.H. Dixon, R.C. Elton: Phys. Rev. Lett. **38**, 1072 (1977)
5. T.N. Lee, W.A. Molander, R.C. Elton: Phys. Rev. A **33**, 1202 (1986)
6. P.A. Naik, P.D. Gupta, S.P. Khumbare: P Phys. Rev. A **43**, 4540 (1991)
7. B.C. Boland, F.E. Irons, R.W.P. McWhirter: J. Phys. B **1**, 1180 (1968)
8. F.E. Irons, R.W.P. McWhirter, N.J. Peacock: J. Phys. B **5**, 1975 (1972)
9. F.E.Irons: J. Phys. B **8**, 3044 (1975)
10. L.Z. Barabash, Yu.A. Bykovskii, A.A. Golubev, D.G. Kosyrev, K.I. Krechet, Yu.I. Lapitskii, S.V. Latyshev, R.T. Haydarov, B.U. Sharkov, A.V. Shumshirov: Laser Particle Beams **2**, 49 (1984)
11. Y.M. Kim: In *Laser Induced Plasmas and Applications*. ed. by L.J. Radziemski, D.A. Cremers (Dekker, New York 1989) Chap. 8
12. C.M. Brown, J.O. Ekberg, U. Feldmenn, J.F. Seely: J. Opt. Soc. Am. B **4**, 533 (1987)
13. P. Laporte, N. Damany, H. Dameny: Opt. Lett. **12**, 987 (1987)
14. C.L.S. Lewis, P.F. Cunningham, L. Pina, A.K. Roy, J.M. Ward: J. Phys. D **15**, 69 (1982)
15. M.D.J. Burgen, R. Dragila, B. Luther-Daview, K.A. Nugent, A.J. Perry, G.J. Tallents, M.C. Richardson, R.S. Craxton: Phys. Rev. A **32**, 2899 (1985)
16. M.C. Richardson, R. Epstein, O. Barnouin, P.A. Jaanimagi, R. Keek, H.G. Kim, R.S. Mayoribanks, S. Noyer, J.M. Souees, B. Yaakobi: Phys. Rev. A **33**, 1246 (1986)
17. D.L. Pappas, K.L. Saenger, J. Bruley, W. Krakow, J.J. Cuomo, T. Gu, R.W. Collins: J. Appl. Phys. **71**, 5675 (1993)
18. J.F. Seely, W.B. McKnight: J. Appl. Phys. **48**, 3691 (1977)
19. N. Qi, M. Krishnan: Phys. Rev. A **39**, 4651 (1989)
20. R. Tambay, R. Singh, R.K. Thareja: J. Appl. Phys. **72**, 1197 (1992)
21. V. Kumar, R.K. Thareja: J. Appl. Phys. **67**, 3260 (1990)
22. A.R. Striganov, N.S. Sventitskii: *Tables of Spectral Lines of Neutral and Ionized Atoms* (Plenum, New York 1968)
W.L. Wiese, M.W. Smith, B.M. Glennon: *Atomic Transition Probabilities*, Vol. I, Natl. Bur. Stand. Ref. Data. Ser., Natl. Bur. Stand. (USA), Circ. No. 4 (US GPO, Washington, DC 1966)
23. Abhilasha: Unpublished.
24. A. Khare, R.K. Thareja: IEEE J. QE-**24**, 2525 (1988)
25. C. Timmer, S.K. Srivastava, T.E. Hall, A.F. Fucaloro: J. Appl. Phys. **70**, 1888 (1991)
26. N. Qi, H. Kilic, M Krishnan: Appl. Phys. Lett. **46**, 471 (1985)
27. R.A. Andrews: In *Progress in Lasers and Laser Fusion*, ed. by A. Perlmutter, S.W. Widmeyer (Plenum, New York 1975)
28. J. Davis, K.G. Whitney: Appl. Phys. Lett. **29**, 419 (1976)
29. J.L. Giuliani, M. Mulbrandon, E. Hyman: Phys. Fluids B **1**, 1463 (1989)
30. P.T. Rumsby, J.W.M. Paul: Plasma Phys. **16**, 247 (1974)
31. M. Mattioli: Plasma Phys. **13**, 19 (1971)
32. F.E. Irons, N.J. Peacock: J. Phys. B**7**, 2084 (1974)
33. A.W. Ali, W.W. Jones: Phys. Lett. A **55**, 462 (1976)
34. W. Furukane, K. Sato, T. Oda: J. Phys. D **22**, 390 (1989)