Experimental investigations of quenching of ionization states in a freely expanding, recombining laser-produced plasma

Andrea Thum-Jaeger,[1] B. K. Sinha,[2] and K. P. Rohr[1]
[1]*Fachbereich Physik, Universitat Kaiserslautern, D-67663 Kaiserslautern, Germany*
[2]*Laser and Plasma Technology Division, Bhabha Atomic Research Centre, Mumbai 400 085, India*
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The experimental measurements of the average ionization states of the plasma produced from slab targets of carbon, aluminum, titanium, nickel, molybdenum, and tantalum, using a 5 ns–Nd:YAG laser, at a laser intensity in the vicinity of 5×10^{10} W/cm^2, are reported. The experimental results have been compared with those of the theoretically calculated ones using the steady state collisional radiative (CR) ionization model. It is observed that a significant difference between the two sets exists which leads to the conclusion of quenching of the ionization states through recombination. From physical considerations one is inclined to consider the possibility of recombination processes quenching the ionization states faster before detection as one goes towards higher and higher values of the atomic number of the target element. The limitations of the ionization model with reference to the plasmas of hydrogenlike ions, the plasma opacity and the consideration of the transient states of ionization and recombination coefficients are discussed. The difference between the experimentally estimated values of ionization states by means of time of flight spectra and theoretical values of these states at the center of the plasma, may provide an interesting experimental tool to investigate the three-body, radiative, and dielectronic recombination processes inside the plasma core.

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I. INTRODUCTION

For more than a decade laser produced plasma has drawn the attention of researchers mainly in three intensity-regimes of laser-matter interaction. First, between the laser intensities of 10^{16} to 10^{19} W/cm^2, the interests were directed towards investigations of laser-fusion, x-ray lasers, shock-related phenomena and generation and studies of super-hot electrons and their distribution functions. In the second regime between 10^{12} to 10^{16} W/cm^2, sometimes, leading even to the intensity of 10^{19} W/cm^2, the investigations were directed towards the studies of laser-plasma and wave-wave interaction along with the studies of energy-transfer processes and energy transport. The third regime is a regime of comparatively low laser intensities ranging from 10^9 to 10^{12} W/cm^2. This regime has not drawn as much attention as the first two but it is very useful for the material scientists in the fields of material preparation such as fabrication of thin films of high-T_c superconductors, oxides, semiconductors, and diamond-like carbon [1–17].

In the hurried quest for laser-fusion and related high-profile physics problems we have not given sufficient and due attention to the experimental investigations of basic plasma properties such as thermodynamic equilibrium, equipartition of energy between electrons and ions and the ionization state of the ions, which depend very significantly on the electron temperature and the electron density. In many investigations on laser-produced plasmas and, especially, on the studies of x-ray lasers one has to know exactly what the ionization states of the plasma under various conditions of plasma temperature and plasma density are. In the investigations related to x-ray lasers many workers [18–22] have theoretically obtained the average ionization states of the plasma taking into account the balance between the ionization rate on the one hand and the radiative, three-body and dielectronic recombination rates on the other as a function of the plasma temperature and density. As experimental data on the average ionization states of various materials with varying atomic number under conditions of various plasma temperatures and densities are scarcely to be found in the literature, one is not sure how near or how far the theoretically determined average ionization states are from the experimentally estimated ones. One is also curious to know how the three recombination processes, the radiative, the three-body and the dielectronic, influence the ionization state under different situations of the plasma density and the plasma temperature.

In the present work we have experimentally measured the average ionization states of the plasma produced from slab targets of carbon, aluminum, titanium, nickel, molybdenum and tantalum using a 5 ns Nd:YAG laser at the laser intensity in the vicinity of 5×10^{10} W/cm^2. We have compared these results with those of theoretically calculated ones using the steady state collisional radiative (CR) ionization model and observed a significant difference between the two sets. From physical considerations we consider the possibility of recombination-processes quenching the ionization states faster before detection as one goes towards higher and higher values of the atomic number of the target element. We have discussed the limitations of the ionization model with reference to the plasmas of hydrogenlike ions, the plasma opacity and the consideration of the transient states of ionization and recombination coefficients.

II. THEORETICAL CONSIDERATION

Using the steady-state collisional radiative ionization model, the ion densities in the two consecutive charge states n_{z+1} and n_z are related as [21,23]

$$\frac{n_{z+1}}{n_z}=\frac{S_z}{\alpha_{z+1}+D_{ez+1}+n_e\beta_{z+1}}, \tag{1}$$

where S_z = the collisional ionization coefficient corresponding to the ionic charge state z, α_{z+1} = the radiative recombination coefficient corresponding to the ionic charge state $z+1$, D_{ez+1} = the dielectronic recombination coefficient corresponding to the ionic charge state $z+1$, β_{z+1} = the three-body recombination coefficient corresponding to the same charge states as above, n_e = density of the plasma electrons.

Though there are many expressions for ionization coefficient S_z, Gupta and Sinha [21], in their recent work on x-ray laser gain calculations, found that the expression of S_z due to McWhirter [24] is the most suitable one. Therefore, we have used the expression of S_z due to McWhirter as

$$S_z=2.43\times10^{-6}\xi_z T_{ev}^{-3/2} \times\lfloor \exp(-\chi_z/T_{ev})/(\chi_z T_{ev})^{7/4}\rfloor \ \mathrm{cm}^3\,\mathrm{s}^{-1}, \tag{2}$$

$$\alpha_{z+1}=5.2\times10^{-14}(\chi_z/T_{ev})^{1/2}(z+1)\times\left[0.429+\frac{1}{2}\ln(\chi_z/T_{ev})+0.469(T_{ev}/\chi_z)^{1/2}\right]\mathrm{cm}^3\,\mathrm{s}^{-1}, \tag{3}$$

$$\beta_{z+1}=2.97\times10^{-27}\xi_z/\{T_{ev}(\chi_z)^2[4.88+(T_e/\chi_z)]\}\ \mathrm{cm}^6\,\mathrm{s}^{-1}, \tag{4}$$

where χ_z and T_{ev} are the ionization potential for ions of charge z and electron temperature, respectively, expressed in electron volts. The term ξ_z represents the number of electrons in the outer most layer corresponding to the ionic charge state z. The values of the ionization potential for different ionization states are taken from the works of Moore [25] and Fraga *et al.* [26]. Though there is a slight difference in the values for χ_z according to the two works, this difference has been ignored as they do not cause any noticeable difference in the end results of the calculations. The expressions for α_{z+1} and β_{z+1} used by the earlier workers [20,21] are due to Kolb and McWhirter [27].

The reliable values of the dielectronic recombination coefficient for a given ionization state of an element of a given atomic number under conditions of various plasma temperatures and densities are difficult to obtain. Kunz [28] and Kunz and Mulser [29], based on the works of Burgess [30], have given an expression for the estimation of the said coefficient, which involves either a knowledge or estimation of oscillator strengths for all the transitions from the ground to the upper levels of an ion of a given ionization state, which are difficult to obtain. Based on the works of Apruzese *et al.* [31], Whitten *et al.* [19], and Hagelstein *et al.* [18] we have concluded that the dielectronic recombination coefficient $D_{e(z+1)}$ can be safely assumed to vary between 1×10^{-12} to $1\times10^{-11}\,\mathrm{cm}^3\,\mathrm{s}^{-1}$. Therefore, in our calculations, we have considered three values for the dielectronic recombination coefficient as $D_{e(z+1)}=1\times10^{-11}$, 5×10^{-12}, and $1\times10^{-12}\,\mathrm{cm}^3\,\mathrm{s}^{-1}$.

Recently Graham *et al.* [32] and Hahn [33,34] have reported extensively on recombination of atomic ions. Hahn [33], in his review work, has reported improved version of Kramers formula [35] on radiative recombination rate. The original formula of Kramers was found to be reliable for principal quantum numbers $n>10$. He reported a slightly improved version for the radiative recombination rate extending its applicability to lower n. Using this formula [Eq. (3.2) of Ref. [33]] we found that the radiative recombination rates calculated from this varies within 10 to 20% of the rates calculated from Eq. (3) of this paper.

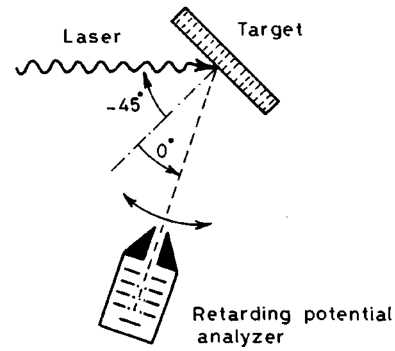

FIG. 1. The scheme of the experimental setup.

Hahn [34] has further reported improved rate formulas for dielectronic recombination (DR) by fitting all of the existing DR data for ions with core charge Z_c less than 50 and the number of electrons N in the target ions less than 13. Because of the gaps in the available data the formulas are reported to be reliable only for $N<13$ and $Z_c<50$. He further stressed the need for additional bench mark data for ions with $5<N<9$ and $N>12$. Moreover, the applicability of the rates given is somewhat limited in view of the potentially large effects of plasma densities and fields for C, O, Mg, Ar, Fe, Se, and Mo ions and the relevant values fall approximately within the range of 10^{-11} to $10^{-12}\,\mathrm{cm}^3$/sec. Even some variations in the values of these coefficients will not have much effect on the estimation of the average ionization as discussed in the following Sec. IV.

III. EXPERIMENT

A schematic representation of the experimental arrangement is given in Fig. 1. The plasma is created by a Nd:YAG Q-switch pulse ($\tau=5$ ns, $\lambda=1.06\,\mu$m) in the TEM_{00} mode of variable energy incident at a fixed angle of $-45°$ onto flat, rotating targets inside a vacuum chamber. The investigated materials are C ($M=12, A=6$), Al ($M=27, A=13$), Ti ($M=48, A=22$), Ni ($M=59, A=28$), Mo ($M=96, A=42$), and Ta ($M=181, A=73$). The laser energy ranges from about 20 to 180 mJ and is focussed to intensities from about 10^{10} to $10^{11}\,\mathrm{W/cm}^2$. The freely expanding ions of the plasma are measured in an angular range between about $\theta=-17.5\%$ to 60° relative to the target normal by moving the detector upon the plane of incidence at a distance of 37.5 cm from the target. Analysis of the ion velocity distribution and charge is performed by means of a time of flight retarding potential detector whose transmission function has been controlled carefully. Some additional effort has been necessary to increase the reliability of the results and to be able to obtain obsolute values of the ion flux. This includes the $n\times1$ product (residual gas density x plasma flight distance) which is at least an order of magnitude below a typical criti-

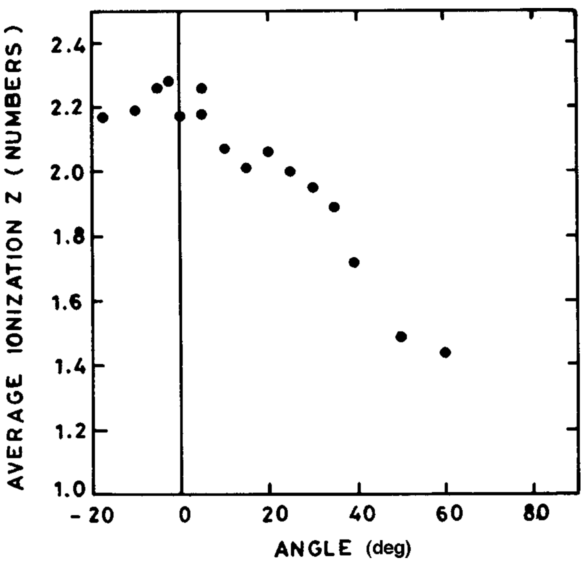

FIG. 2. Average ionization Z as a function of emission angle for carbon at incident laser energy of 180 mJ.

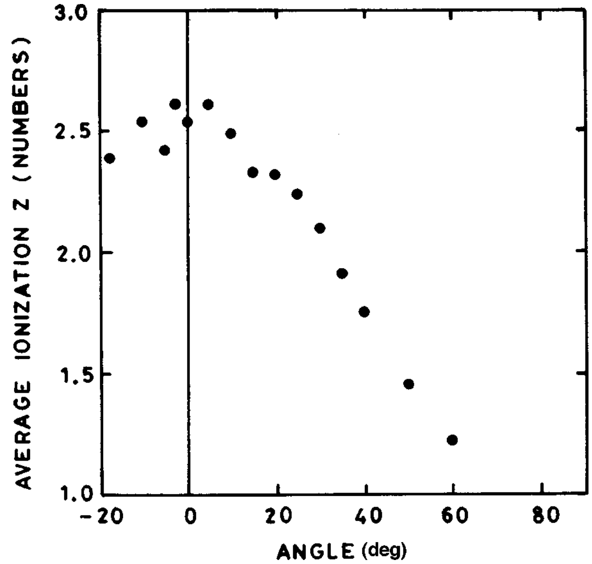

FIG. 3. Average ionization Z as a function of emission angle for aluminum at incident laser energy of 180 mJ.

cal value of $5\times10^{13}\,\mathrm{cm}^{-2}$, deduced from cross section data of Schwarz *et al.* [36] surface contaminations of the target which are removed by defocussed prepulses, and changes of the ion emission distribution due to cratering which are minimized by rotating the target after a certain number of shots empirically determined to be below 10.

The signal of the detector is stored at a digitizing rate of 100 MHz, and, by multiscaling, an effective resolution of about 10 bits is obtained. The total uncertainty of the absolute flux of the evaluated differential ion spectra is estimated to range from 5% for the low energetic part to about 30% for the highest charges and kinetic energies of the ions. The laser energy is determined by a calorimeter in absolute units (type RJP 700, Laser Precision Corporation). It is monitored continuously and has a shot-to-shot variation within 3%. The laser pulse is essentially TEM_{00} (spatially controlled by a CCD camera). The beam has a divergence of about 0.3 mrad with a FWHM of 5 nsec. Complete details of the experimental arrangement and the measurement procedure have been described in the works of Mann and Rohr [7].

IV. RESULTS AND DISCUSSION

The experimental results of the average ionization Z, which is given by the expression $\Sigma\xi_z N_i^{z+}/\Sigma N_i^{z+}$, where N_i^{z+} represents ions with charge $z+$, are displayed for carbon, aluminum, titanium, nickel, molybdenum, and tantalum in Figs. 2–7, respectively. The results were obtained at angles θ varying from $-17.5°$ to 60° with reference to the target normal and the ions were collected at a distance of 37.5 cm by the ion-collector system referred to in the preceding section. An incident laser energy of 180 mJ corresponded to a laser intensity of about $5\times10^{10}\,\mathrm{W/cm}^2$. We observe that the average ionization Z is not isotropic with reference to the angle of observation but it is anisotropic. It has about the maximum value at the detection angle of 0° and in a narrow cone about it and, then, decreases as θ goes far from 0° and away towards 60°. This trend is observed starting from carbon $(A=6)$ to $\mathrm{Ta}(A=73)$.

In Fig. 8 we have displayed the values of maximum average ionization as a function of laser energy for the elements carbon, aluminum, titanium, nickel, molybdenum, and tantalum. The maximum ionization has been plotted for all the elements as a standard reference. Here we find a trend which has an element of consistency. For low A values such as carbon $(A=6)$ and aluminum $(A=13)$ the average ionization increases with energy. For elements such as $\mathrm{Ti}(A=22)$ and $\mathrm{Ni}(A=28)$ the average ionization increases up to an incident laser energy of 40 mJ and then decreases as the energy increases from 40 to 180 mJ. For elements such as molybdenum $(A=42)$ and tantalum $(A=73)$ the average ionization is maximum at an incident laser energy of 20 mJ and slowly decreases as the energy increases up to 180 mJ.

Moreover, one more trend is clearly discernable. As the value of A, the atomic number increases, the experimentally

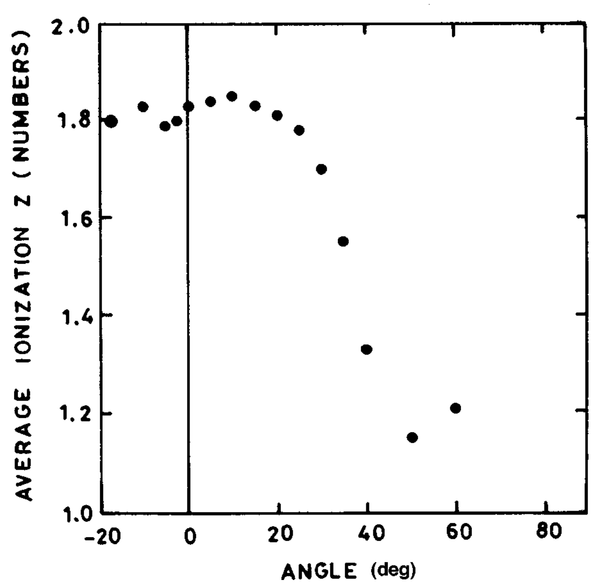

FIG. 4. Average ionization Z as a function of emission angle for titanium at incident laser energy of 180 mJ.

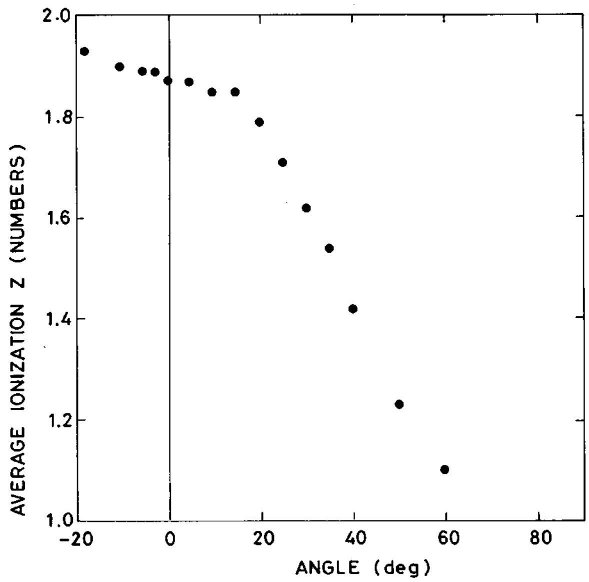

FIG. 5. Average ionization Z as a function of emission angle for nickel at incident laser energy of 180 mJ.

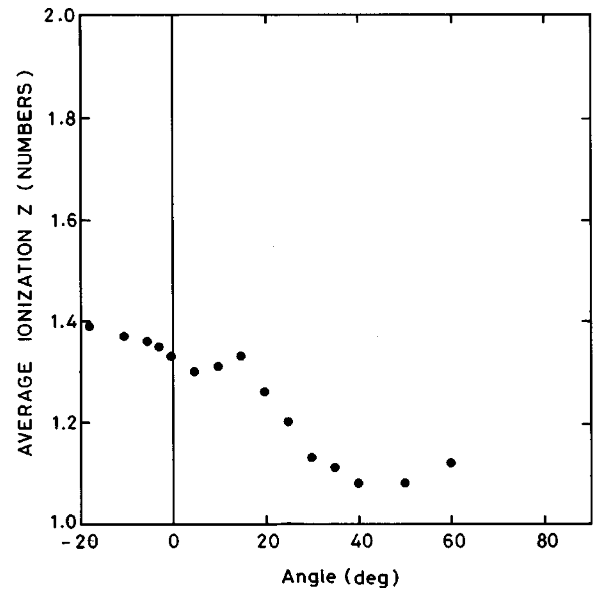

FIG. 7. Average ionization Z as a function of emission angle for tantalum at incident laser energy of 180 mJ.

determined values of maximum average ionization, or, in a sense, the values of average ionization decrease for a given incident laser energy. We will discuss these observations in the next paragraphs after examining the theoretical values of average ionization obtained on the basis of steady state collisional radiative ionization model as used by the previous workers [18–22].

Based on the equations (1)–(4) we have displayed the theoretically estimated average ionization for carbon, aluminum and nickel in Figs. 9, 10, and 11, respectively, as a function of electron density and at an electron temperature of 30 eV for three values of dielectronic recombination coefficient as discussed in the preceding section.

For the discussion of the present experimental results, based on the works of Sinha *et al.* [37] and those of Sakabe *et al.* [38] at a laser intensity in the vicinity of 10^{11} W/cm^2, the time- and space-integrated temperature of about 30 eV has been estimated and density of 10^{18}–10^{20} particles/cm^3 has been assumed. We could not get the required data on ionization energy for tantalum from the works of Moore [25]

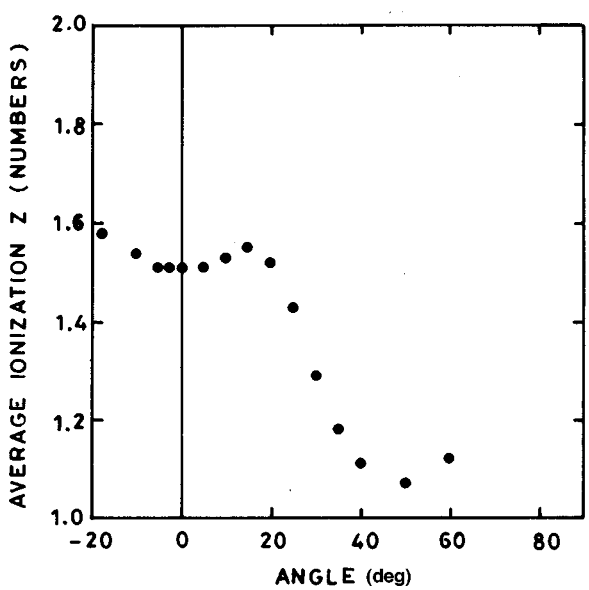

FIG. 6. Average ionization Z as a function of emission angle for molybdenum at incident laser energy of 180 mJ.

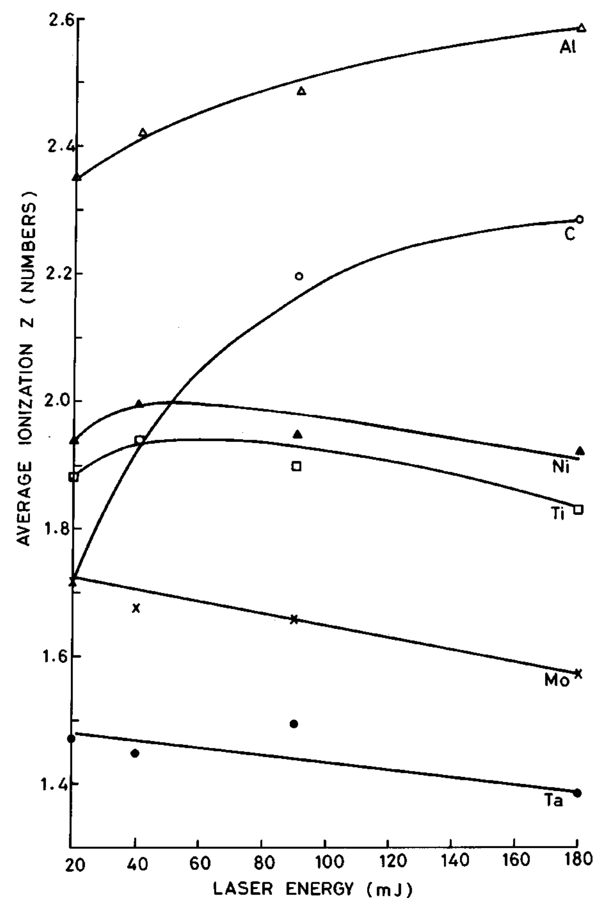

FIG. 8. Maximum value of average ionization in the vicinity of emission angle $\theta=0°$ as a function of incident laser energy for carbon, aluminum, titanium, nickel, molybdenum, and tantalum.

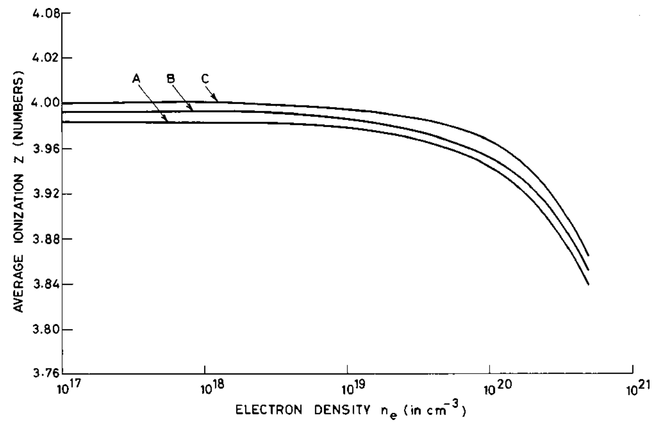

FIG. 9. Theoretically estimated average ionization for carbon as a function of electron density and at an electron temperature of 30 eV. The symbols *A*, *B*, and *C* represent three values of dielectronic recombination coefficients which are given by $D_e = 1\times10^{-11}$, 5×10^{-12}, and $1\times10^{-12}\,\mathrm{cm^3\,s^{-1}}$.

and Fraga *et al.* [26], hence, similar results, as in Figs. 9–11, could not be obtained for tantalum as it required too many extrapolations. However, based on linear extrapolation of a few initial values of the ionization energy, the average ionization for tantalum was calculated to be in the vicinity of 12 at the values of temperature and density under consideration, which is much more than the experimental values obtained from measurements and estimates based on time-of-flight spectra of the ions.

From Figs. 9–11 it is observed that as the density increases, the average ionization slowly falls and the fall is sharper between a density of 10^{20} to $5\times10^{20}\,\mathrm{cm^{-3}}$. Between densities of 10^{17} to $10^{19}\,\mathrm{cm^{-3}}$ the fall is very slow. That is to say, the effect of the three recombination precesses is profound at a density higher than $10^{20}\ \mathrm{cm^{-3}}$. The effect of dielectronic recombination coefficient does not seem to vary much with the density. In the density range of 10^{17} to $5\times10^{20}\ \mathrm{cm^{-3}}$, as we vary the dielectronic recombination coefficient from 10^{-11} to $10^{-12}\ \mathrm{cm^3\,s^{-1}}$, the variation in average ionization state is of the order of 10% or less for all the three elements. This point should be noted because, as mentioned earlier, the exact values for dielectronic recombination coefficient are difficult to obtain.

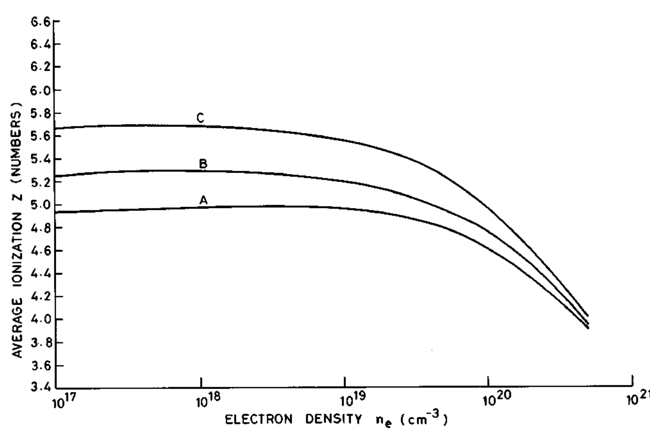

FIG. 10. Theoretically estimated average ionization for aluminum as a function of electron density. The other conditions and symbols are the same as in Fig. 10.

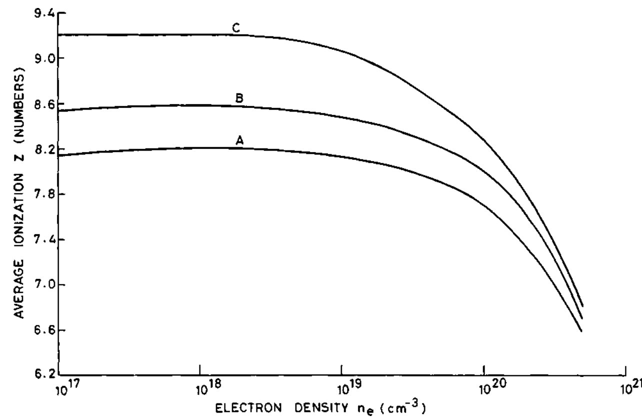

FIG. 11. Theoretically estimated average ionization for nickel as a function of electron density. The other conditions and symbols are the same as in Figs. 10 and 11.

The most significant results of the present investigation, hitherto unknown and hitherto unreported, are contained in Figs. 8 and 9–11. Figure 8 itself is derived from Figs. 2–7. We keenly observe that as the laser energy increases from 20 to 80 mJ, the average ionization of carbon ($A=6$) and aluminum ($A=13$) increases from 1.72 and 2.35 to 2.3 and 2.6, respectively. This seems to be in order as with the increase in laser energy, the plasma temperature increases and, hence, the ionization state. But for the second set of elements titanium ($A=22$) and nickel ($A=28$) average ionization increases slightly from values of 1.88 for Ta and 1.94 for Ni up to an incident laser energy of 40 mJ and then slowly decreases as the laser energy increases up to 180 mJ. In the third set of the elements molybdenum ($A=41$) and tantalum ($A=73$) the picture is completely different. Here as the laser energy is increased from 20 to 180 mJ, the average ionization (1.48 for Ta and 1.72 for Mo at 20 mJ) slowly decreases instead of increasing.

Moreover, from Figs. 9–11 one notes that theoretically calculated average ionizations are in the vicinity of 4.0, 5.2, and 8.6 for carbon, aluminum, and nickel. For tantalum it has been separately calculated to be in the vicinity of 12.

From the physical considerations it seems that the recombination effects are fast and profound as one goes to elements with higher atomic number. At the initial stage of plasma production it is possible that the plasma is produced with a higher value of ionization state, which gets quenched to lower values due to possibly faster recombination processes. One also notes that the values of ionization energy of different ionization stages gets smaller and smaller as one goes towards higher values of *A*. As a result, for a given laser energy, for ions of higher *A* values, ionization states are higher and, hence, the electron densities which enhance the recombination rate before the detection system is in a position to detect them. Additionally, one has to examine the limitations of the plasma model used to derive and obtain Eqs. (1)–(4). These relations are based on the steady-state collisional-radiative ionization model which has the following chief limitations. This model has been mainly used for hydrogenlike ions and no data are available for other groups of ions [23]. Secondly, the formulations are strictly valid for optically thin plasmas. An optically thick plasma, partly or wholly, absorbs the radiation produced in the plasma and modifies the population densities of the energy levels and, hence, the values of various ionization and recombination coefficients. The third limitation is that the time dependence of the ionization and recombination processes has been ignored. Thus, our experimental results provide a sufficient impetus to look for a suitable plasma model which takes into account the nonhydrogenlike character of the plasma ions, its opacity with reference to the radiation emitted in the interior of the plasma and the completely transient state of the ionization and recombination processes and, hence, the transient state of the ionization and recombination coefficients.

In this connection it is relevant and useful to draw the attention to the works of Matzen and Pearlman [39] in which they pointed out that the time dependent rate effects can be important for predicting the signals from both x-ray and ion diagnostics. They compared the steady state and rate equation ion density distributions and concluded that whereas the time delays in the ionization of the plasma can affect x-ray calculations, the calculations of the freezing out of the recombination processes in the plasma is necessary to predict signals from ion diagnostics.

As the theoretical estimates of the ionization state at the plasma core give the relevant figures before the outset of the recombination processes, and the experimental ones give the expression after the recombination processes cease to play a role, the experimental results provide an interesting tool for the investigations of these processes and their differential importance on the quenching of the ionization states. One may solve the ionization and recombination rate equations and arrive at approximate values for the quenched ionization states at the end of the plasma expansion and compare them with the experimental ones.

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