PERGAMON

Journal of Quantitative Spectroscopy & Radiative Transfer 65 (2000) 645–653

Journal of Quantitative Spectroscopy & Radiative Transfer

www.elsevier.com/locate/jqsrt

Temperature and electron density gradient in Xe laser produced plasmas, by spectral analysis

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Received 5 July 1999

Abstract

We have performed a spectroscopic analysis on laser produced plasmas in Xe at intermediates pressures (less than 1 atm), resolved both spectrally and temporally, also discriminating different regions of the plasma. This is produced by tightly focusing the radiation of a Q-switched Nd : YAG laser on a cell equipped with an observation window at right angles with respect to the laser. Our results confirm that there is a gradient in electron density in the plasma ball formed by the laser. This gradient almost defines at least two regions, an outer layer of higher electron number density, and an inner core of lower electron number density. The electron temperature gradient has the same sign. These conclusions are supported by evidence taken using the spectral results mentioned above.

1. Introduction

The laser produced plasmas (LPP) in gases provided by some time a bright source for electromagnetic radiation in the UV spectral region [1–3]; since the interest on soft X-Rays sources was increasingly growing, the concern on these plasmas shifted towards solid target — LPP because of the higher plasma densities achievable in these conditions [2–4].

Nevertheless, a number of facts about the LPP reveal that some interesting information regarding the mechanisms of the plasma energy dissipation, can be gathered from medium-density gaseous targets, since the energy of the laser has been already absorbed by the gas at the end of the breakdown process [5]. The mechanism of gas heating and of energy dissipation, which are responsible of most of the emission process in the optical region has been little explored for the ranges of fluences as the ones used in the present work (on the order of 10^{13} W cm^{-2}).

We have presented in Ref. [6], experimental evidence on the role played by the radiative recombination (RR) channel in this post-heating pulse temporal stage, evidenced by the continuum

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0022-4073/00/$ - see front matter
PII: S0022-4073(99)00130-2

spectra emitted by such plasmas long after the laser pulse has ended. The importance of the RR channel evidenced itself as peaks in the continuum spectrum which were assigned to resonance in the RR from free-bounded level transitions and from auto-ionizing levels of the ions.

In Ref. [7], we have used a similar equipment and the main goal there was to interpret the emission of a continuum and the main channels of radiation in LPP's at nearly atmospheric pressures, focusing our attention to the line spectrum, both time and frequency resolved of this plasma ball which is bright enough to give rich information on the physical processes involved. The same type of spectroscopic analysis on the plasma emission were performed in the present paper.

By a careful analysis of the spectra taken in such conditions, in this work we are able in Ref. [7], to identify the RR [8] as the main energy dissipation channel for the LPP, compared with de-excitation by collisions or by hydrodynamic or diffusion expansion, at least in the first hundreds of nanoseconds following the free evolution of the plasma, i.e. once the laser has ended. The conclusions mentioned allow us to identify the processes taking place in the plasma for different ion species and for different atomic structure in different ions.

The present paper is devoted to the analysis of a spatial structure of the plasma, possibly due to the complex process of the laser radiation absorption. In fact, we shall present below an analysis which is based on new spectroscopic data and using the data of Refs. [6,7], which we think is sufficient evidence of temperature gradient and thus of an electron number density gradient, manifested in several ways which we will enumerate. This structure would consist in at least two regions, a core of lesser electron density and a periphery of higher electron density. According to our description of the phenomena, these regions would have been generated during the breakdown but due to the mechanisms involved in the energy dissipation of the plasma, the core would last much longer than the laser pulse. Thence, the lines involving the transitions of the ions Xe II and Xe III display long lasting tails, the Xe V ions appears after the laser pulse attains its maximum; accordingly, the spatial discrimination of the plasma ball, display different temporal evolution for different ion species. We also show the evolution of the continuum, featuring a shift towards lower frequencies with time, a peculiar maxima distribution and the peaks due to auto-ionizing levels.

2. Experiments

Fig. 1 displays the main features of the experiment. As in Refs. [6,7], we have employed the Nd:YAG laser (650 mJ at 10 Hz) emitting in the 1.06 μm. Attempts to form the plasma using its second harmonic gave results which are consistent with the ones using the infrared radiation, and so will not be presented in the present work.

The laser beam was focused onto a cell filled with spectroscopic Xe at nearly 300 Torr (and up to 2 atm in some control experiments) by means of an antireflection coated quartz lens of 20 cm focal length. The cell consisted of a Pyrex glass tube with two windows at the ends, and a third window at right angle with the tube axis which is used to gather the light coming from the plasma. The laser produced a very stable plasma ball which was arranged to be behind the third window. The emitted light from the plasma was gathered by another quartz lens of 20 cm focal length, and then analyzed by a monochromator of Resolution = 300,000 equipped with a photomultiplier (PM) with known responsivity curve. All the results were thus corrected with it.

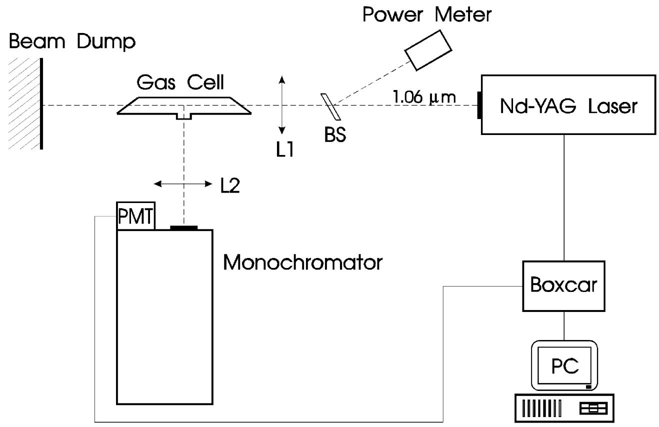

Fig. 1. Schematics of the experimental setup.

The signal produced by the PM is then analyzed by a Box-Car integrator with a 10 ns gate width, connected with a PC which stores the information. The signal processing was made by normal statistical-fitting software.

Our results can thus be subdivided into three main subjects, the temporal analysis made at different wavelength position of the monochromator, the spectral analysis at different times for the position of the Box-Car integrator window, and the spectra taken at different regions of the plasma ball. Altogether, there were two filling pressures at which all these spectra were taken, one higher pressure for which only continuum was observed, and a lower one for which mixed, continuum and line spectra were recorded.

3. Experimental results

As was anticipated, a detailed analysis of the time- and frequency-resolved spectra was done on the Xe plasma; both analysis were also done in two symmetrical location of the ball by placing a convenient diaphragm after the output window of the gas cell. This allows us to study the glowing periphery alone and the central portion of the plasma (which unavoidably includes some of the periphery due to the symmetry of the plasma).

At filling pressures above 760 Torr, the plasma is very bright and the study of the emitted light consent us to conclude that there is no line emission from any ion during the observation time reported here. All the light is emitted in the form of a continuum, which in turn was studied previously [6]. Reducing the filling pressure, the continuum produced signal becomes lower and the line transitions between ion levels are detected superimposed to the continuum. The information of the time evolution of these spectral features is interesting and will be described later.

In the case of the continuum, taken from 2700 to 6000 Å, the records were elaborated using the temporal analysis, so the spectra could be constructed for different times after the breakdown has occurred, we found this procedure less time consuming and more reliable than the spectrally resolved at different time for a wide range of data. However, it is interesting to study the spectral evolution in time directly to minimize errors in considering line width or line shapes. Different types of consistency test were done to assure that the results were not affected by the techniques used to record data.

4. Analysis of data

The first observations that we made on these experiments show that there is a continuum emission with peaks due to resonant recombination from auto-ionizing levels of Xe higher ions, and in some cases we observed a line spectrum coming from different ion species and with characteristic time evolution, which depend on the transition being involved. In the present experiments, we resolved in time both spectra emerging from the plasma (i.e., the continuum and the line ones), and were able to follow the resonant recombination peaks, which enable to follow the main deactivation channels [6,7]. The continuum evolution is constructed from the time-resolved spectra taken at different wavelengths, revealing that the time evolution of the entire continuum depends on the frequency. This result is evidenced in Fig. 2, where it is also evident a "global" structure of the spectrum, displaying local maxima at wavelength roughly coincident with the emission of each ion, separated by approximately 1000 Å, and also that the plasma evolves in such a way that the emission is shifted toward lower frequencies drifting with time. It has to be stressed that these peaks are not lines, since the latter were eliminated in the data gathering as in Refs. [6,7].

Line spectra shown in Fig. 3, reveal that the lines of the first and second ion of Xe (Xe II and Xe III) have a long tail that last for times much longer than the laser pulse. Although this feature is

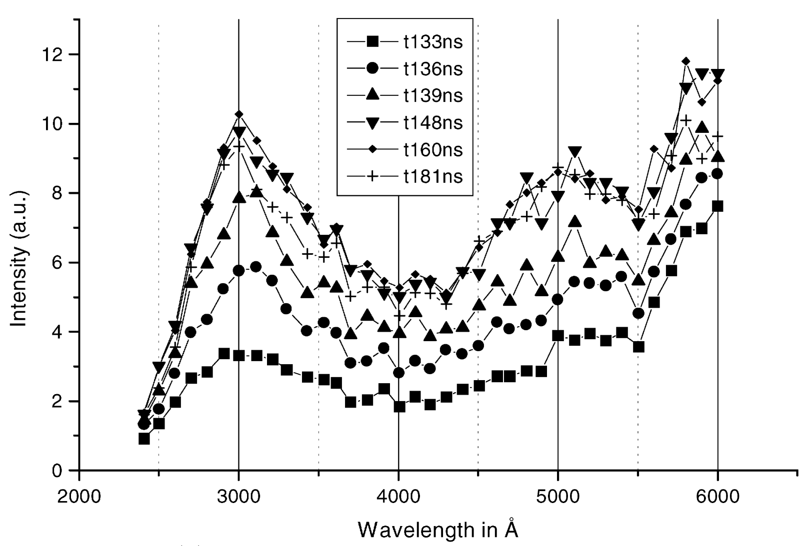

Fig. 2. Time evolution of continuum emission in the region between 2700 and 6000 Å. there are broad local maxima 1000 Å apart, and sharp peaks due to recombination from auto-ionizing levels. During the evolution, these peaks are maintained throughout. the emission maxima shift in time towards the lower frequencies.

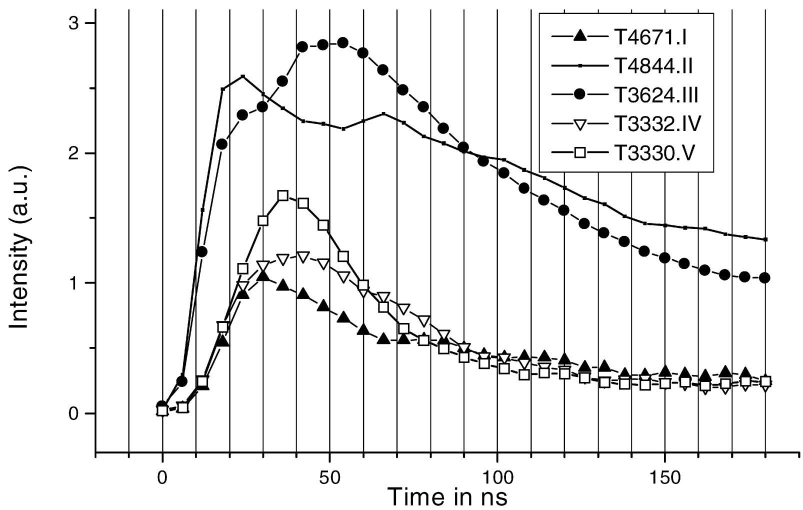

Fig. 3. Time-resolved line spectra for different ion species at the central region of the plasma ball. It is clearly seen that: (a) there are early peaks and long tails for some ions, (b) lines coming from Xe V peak during the turning off of the laser pulse, (c) Xe I lines go behind Xe II and Xe III lines, and (d) Xe IV lines display a small "bump" revealing a recombination mechanism involved.

observed directly in the center of the plasma for several lines, there are transitions of the same ion for which apparently this does not happen. However, the lines that seemingly have no tail, are those that shift the center of emission with time. Following its evolution both in time and in frequency, however, reveals that they last for very long times compared to the laser pulse duration. In the periphery, the time-resolved spectra shows no long tail. These facts are important as evidences of the electron number density gradient, as will be discussed later.

In Fig. 4, instead, the line spectra was taken at the periphery. In this case, it is evident the lack of the long tails for Xe II and Xe III lines. It is also apparent that Xe V has the same shape as in Fig. 3. It is also clear that the decay of this line is not affected by recombination, since it displays a normal exponential behavior. Lines coming from Xe IV in this spectral region display, instead a small but evident longer tail due to recombination. This fact is an important evidence of the conclusion discussed later, that the Xe V is the higher ionic species during the stages following the breakdown. It could serve as a measurement of the maximum energy of the electrons, although more and better knowledge of the cross sections for the formation mechanisms are needed, as well as an evaluation of those mechanisms.

Using the data taken from experiments done for each transitions, the Fig. 5 can be constructed showing an important result, which is that the lines of Xe II (and Xe III, not shown in the figure) widens as the time increases, while Xe V lines has decreasing widths instead. A behavior like Xe V line is consistent with a time-decreasing electron density. Although these results are displayed for three lines, it can be shown valid for all of the lines inspected. We consider this fact an important evidence of the electron number density gradient mentioned above. In fact, Fig. 6 is an evaluation of the electron number density from an approximate model and shows that the plasma has to have, at least for the first hundred nanoseconds, regions with different temperature and electron density,

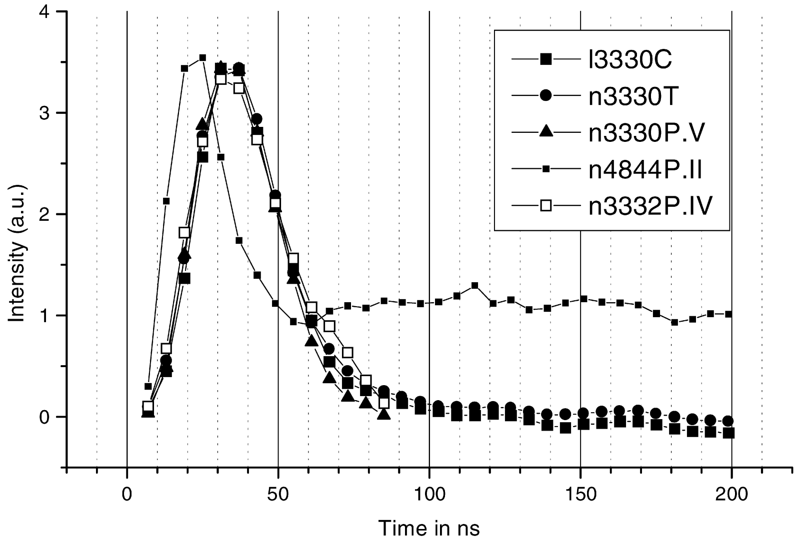

Fig. 4. Time-resolved line spectra for different ion species at the periphery region of the plasma ball. It is clearly seen that: (a) there are early peaks but the long tails are missing, (b) lines coming from Xe V peak during the turning off of the laser pulse, and have the same type of behavior as in the central regions, (c) Xe I lines go behind Xe II and Xe III lines as in the central region, and (d) Xe IV lines display a small "bump" revealing a recombination mechanism involved, as in the central region. Line intensities are normalized to the maximum in order to compare the shape of the Xe V line in all the regions inspected.

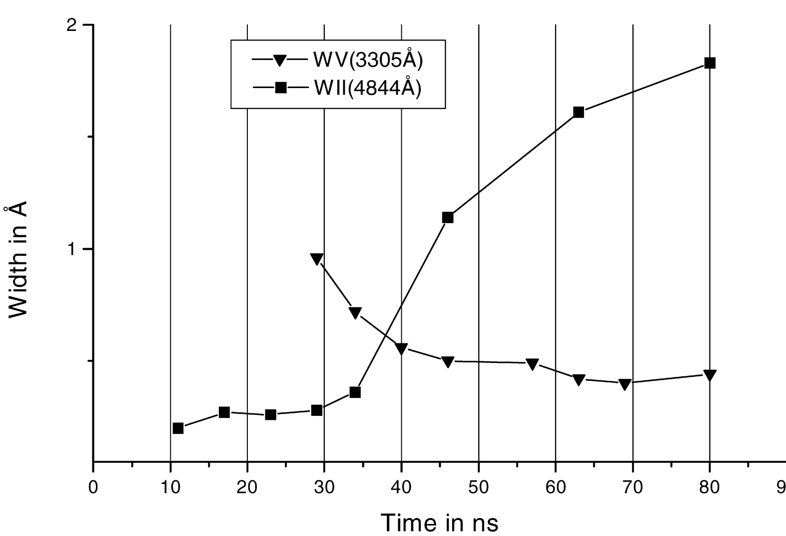

Fig. 5. Spectral widths of two lines as time is increasing from the breakdown. It is clearly evident that The Xe II line (with long tail) have a line width that at a certain time jumps almost by a factor of three, while for Xe V this width is steadily decreasing.

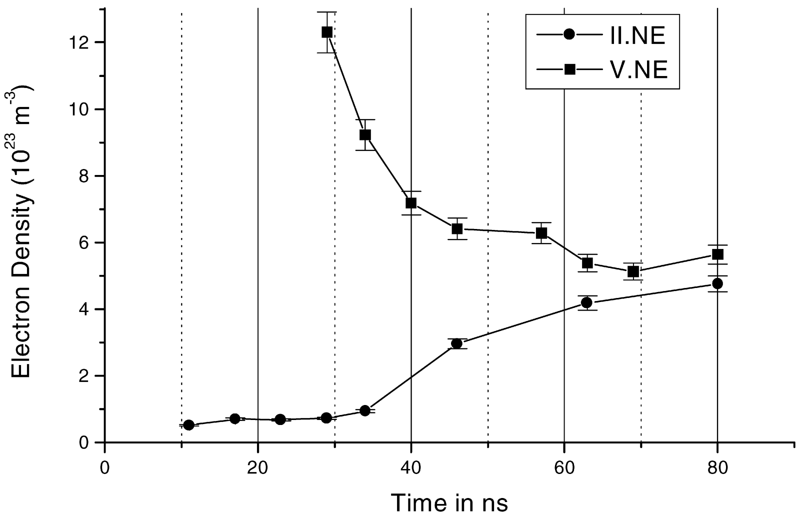

Fig. 6. Electron number density vs. time evaluated from a LTE model. At approximately 100 ns, the regions tend to a common electron number density.

as we show in the appendix. The model used assumes LTE and hence the validity of Saha Equations; this is questionable, since the radiative–collisional model should be preferred [9]. However, the numerical solutions of the radiative–collisional model [10] indicates that these equations proved to work fine as a scaling law, for plasmas defined by electron temperatures T_e ranging from 0.5 to 30 eV and electron number densities, N_e, in the range 10^{15}–10^{18} cm^{-3} which is our case.

We have also made some experiments with the laser doubled in frequency. The results obtained in that way are qualitatively the same except for those related specifically to the fluence, for example, the type of plasma generated by the laser. All other results support our description.

5. Conclusions and discussions

We have shown what we believe constitutes evidences that there are at least two regions of very different electron density in the plasma that coexist for times much longer than the heating laser pulse. This assertion is supported by the evidence taken from experiments done resolving different regions of the plasma. The spectral widths of several lines of different Xe ions (particularly Xe II and Xe III) have a characteristic feature that we shall use as evidence of the same fact, being the long tail of some lines spectrally wider than other ions, increasing their width with time instead of decreasing as it should be expected for a uniform plasma; in fact, this is the behavior of lines coming from Xe V transitions. The analysis performed to the Xe V lines, which altogether show no evidence of dependence on the region of the plasma under study, and whose intensities peaked at times after the laser has reached its maximum, is another evidence of the existence of a gradient in the electron density, also this feature allows us to conclude that the Xe V is the highest ion in the plasma at times between the onset of breakdown and the end of the plasma ball. We therefore conclude that all the evidence fits to a model of at least two regions in the plasma, one with high electron density number at the external layer and an internal core of lower density number. All these evolve to a uniform plasma at times much longer than the laser duration.

To summarize our results, we arrive to the conclusion that the plasma ball generated by the laser at the fluences noted earlier, present at least two regions of different electron number density, this assertion is supported by several facts, which we enumerate: (1) the longer tail on the temporal evolution of lines of Xe II and Xe III, which indicates that there exist recombination for times much longer than the laser pulse duration, (2) the fact that such long tail is missing in the periphery, (3) the fact that the Xe V line has the same temporal evolution regardless to the region of plasma being observed, (4) the fact that the continuum evolution last for times comparable to the duration of the tails, (5) the continuum duration is shorter than the duration of the long tails of some lines, and (6) the increasing spectral width of Xe II and Xe III lines with time. All these evidence, together with some approximate evaluation of the density are obviously pointing to the conclusion stated above.

Now, we speculate about the reasons of these regions being present in the plasma. In the first tenths of nanoseconds after the laser pulse hit the target, tightly focused, the neutral Xe is unstable with respect to the Xe II, Xe III (and, perhaps, Xe IV and Xe V) as stated by Lambropoulos [11]. Thus, the gas breaks down, which in turn produces an optically inhomogeneous region; since the laser is still increasing the power, it will deflect in the transverse directions and (probably) shift the focus position in the axial direction. During the spreading it still continuously generates ions, up to some time almost coincident with the total pulse duration. The expansion of the plasma ball is, thence no hydrodynamic, but an optical process, incidentally, this fact is supported indirectly by the fact that the emission neither at the periphery, nor at the core is Doppler shifted whatsoever [6,7]. Once the plasma ball is established, the radiation of the outer layer follows the laser pulse. However, since the core remained, the emission coming from it, is not influenced by the laser, and proceeds through the radiative recombination. This, in turn, is completed at a lower electron number density and so, its linewidth increases with time until it dies out.

Finally, we want to emphasize that the analysis above has been carried solely by spectral evidence taken during experiments conducted in laser generated plasmas.

Acknowledgements

Work partially supported by Grants of: CONICET (PMT-PICT 0104 (BID)), and UNCPBA, through an Instituto de Física “Arroyo Seco” Grant.

Appendix

According to Saha equations, at $T_e \approx 1.5$ eV, the ion population would predominantly be of Xe II, while for $T_e \approx 2.4$ eV there would be similar proportion of Xe II and Xe III. Instead, for $T_e \approx 3.5$ eV there is practically only Xe III. Besides, the predominance of Xe IV implies a $T_e \approx 6$ eV and if Xe V is the predominant ion, $T_e \approx 8$ eV.

With these estimations of T_e, the N_e electron number density can be inferred from the line widths using Griem's semi-empirical formula [8], applied for ionic transitions and considering the electron impact width. For the FWHM(w), we use

$$w = 2.22 \times 10^{-22} N_e T^{-1/2} [\langle i|r^2|i\rangle + \langle f|r^2|f\rangle] g(x),$$

where $x = 1.5kT/E$ and $g(x)$ is the Gaunt factor; for $E \leq 2$ eV we use $g(x) = 0.2$ (which means that Xe II is the predominant ion); and $g(x) = 0.3$ for Xe V as predominant ion [9]. In the equation, w is in cm^{-1}, N_e is in m^{-3}, and T is in K.

From the Xe II experimental widths (using $T_e = 2.5$ eV, since it is clear from the experiment that coexist temporally and spatially with the Xe III) we can draw the curves of Fig. 6 corresponding to the Xe II ion. If instead we use the $T_e = 7$ eV (which is valid for an equal population of Xe IV and Xe V), we can draw the other line. The difference in both is pointing to a separation of the regions where each ion is predominant. Similar arguments can be used to the spatial gradient of this magnitude.

The error estimated in the above calculations is not greater than 15%, which means that the analysis is precise enough to sustain our conclusions.

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