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# Velocity-resolved recombination dynamics in a laser-produced Ta plasma†

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**Abstract.** Ion energy spectra of a laser-produced Ta plasma have been investigated as a function of the flight distance from the focus. The laser (Nd:YAG, 20 ns, 210 mJ) is incident obliquely (45°) and focused to an intensity of about $10^{11}$ W cm$^{-2}$. The changes in the ion distributions have been analysed for the $Ta^{+}$ to $Ta^{6+}$ ions in an expansion range 64–220 cm. With increasing distance from the target, a weak but monotonic decrease is observed for the total number ions, which is essentially due to the decrease in number of the more highly charged species. For the $Ta^{+}$ and $Ta^{2+}$ ions the net changes approximately cancel. A more sophisticated picture of the recombination dynamics is obtained, however, if the changes within individual groups of ions expanding with different velocities are compared. Here, in the same spectrum, both increasing and decreasing ion numbers can be observed. This can be interpreted as direct evidence of recombination and its dependence on temperature, density and charge.

## 1. Introduction

Laser-initiated ablation of neutral species, ions and clusters has gained considerable importance in the past in a series of applications like laser-induced mass analysis (Adrain and Watson 1984), generation of particle beams for secondary experiments (Vertes *et al* 1990) even of atomic species that are difficult to produce by other means (Carroll and Costello 1986, Campbell *et al* 1990) and above all material synthesis techniques for fabricating thin films of semiconductors, high-$T_c$ superconductors, or diamond-like carbon (Paine and Bravman 1990) for example. The quality of these films depends critically on the charge and energy distribution of the ions in the plasma beam (Pompe *et al* 1992). Since, however, the degree of ionization is changing during the expansion, for an optimization of these applications reliable data on the recombination dynamics and its dependence on experimental parameters are essential. Despite the many existing investigations on the ion emission of laser-produced plasmas, so far only a few experiments have been published, that could demonstrate directly the effect of recombination in the ion beam during the expansion in regions far away from the focal range. Tallents (1980) has shown the strong decrease of the ions of charge 3–6 of a carbon plasma. In this experiment the charge state distribution appears to be frozen out at a flight distance of about 1.5 m. On the other hand, Golubev *et al* (1984) have observed a continuous decrease in the total charge over the whole expansion range investigated (more than 3 m), although they started with about the same initial temperature.

So far no experimental data have been published in which the development of the charge state distributions in the asymptotic range has been investigated in detail. In the present experiment we have analysed for the first time the dynamics of recombination of all individual ion species, and, which we think is more interesting, within separate velocity groups. Thereby a distinct picture of the expansion process is obtained, which could stimulate quantitative theoretical models of the expansion mechanism and allow an extrapolation to the short decimetre range that is typical for material synthesis techniques, but not accessible in general to direct measurements of the ion dynamics.

## 2. The experiment

A schematic representation of the experimental arrangement is given in figure 1. The plasma is created by a Nd:YAG $Q$-switch pulse ($\tau = 20$ ns) in the $TEM_{00}$ mode incident at a fixed angle of $\Theta = 45°$ onto a flat Ta target inside a vacuum chamber. The laser energy is 210 mJ, which is focused to a corresponding intensity of about $10^{11}$ W cm$^{-2}$. The particles of the freely expanding plasma are detected at an angle of $\phi = -10°$

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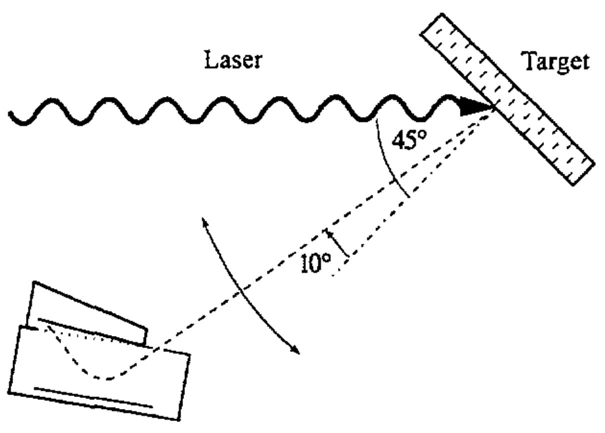


**Figure 1.** A schematic diagram of the experimental set-up. The laser pulse is incident at a fixed angle of 45°. Ion analysis is in the plane of incidence by a dynamical multichannel analyser at flight distances in the range 64–220 cm.

relative to the target normal at four distances in the range 64–220 cm from the target. In this direction the highest degree of ionization is expected (Pitsch *et al* 1981, Thum *et al* 1994) and hence recombination between many ionic states should be observable. Detection of the ions is by means of a multichannel dynamical analyser (Eicher *et al* 1983, Rupp 1994), which allows for complete analysis of charge and energy distributions of each plasma pulse, thereby leading to an accuracy, unachievable by the usual time-of-flight/electrostatic methods. This enhanced accuracy and sensitivity, allowing for direct detection of absolute particle numbers over more than five decades, has turned out to be essential for the success of the present experiment.

During the experiment continuous tests were performed concerning the transmission function of the detection system, the temporal stability of the laser and reproducible conditions in the interaction regime at the target. The $nl$ product (residual gas density $\times$ plasma flight distance) has been chosen to be more than two orders of magnitude below its critical value at which collisions begin to influence the ion spectra (Clement *et al* 1980). For the present case of a Ta ion beam, recently measured charge exchange cross sections for the low energy eV amu$^{-1}$ range of Schwarz *et al* (1994) have been used as guideline data (typically $10^{-14}$ cm$^2$). Further details of the experimental arrangement and the measurement procedure have been described previously (Mann and Rohr 1992).

## 3. Results

Energy spectra of the $Ta^+$ to $Ta^{6+}$ ions were measured in absolute particle numbers at four flight distances. For a distance of 118.4 cm these are converted in figure 2 to relative spectra according to the kinetic energies in order to emphasize the well-known group structure for the different ion species. Apparently, for each ion species, an energy range can be found within which the respective charge survives and even dominates. This principal species behaviour holds for all distances.

In figure 3 the numbers of the $Ta^+$, $Ta^{3+}$ and $Ta^{5+}$ ions are plotted as a function of the flight distance for a set of kinetic energies. Thereby the changes in the ion distributions are resolved according to the ions' velocities. Such an approach suggests that, during the expansion, recombination should take place preferentially between particle groups expanding with about the same velocity. Straight lines are drawn through the data points as visual guides. Although at first glance the general behaviour seems to be confusing,

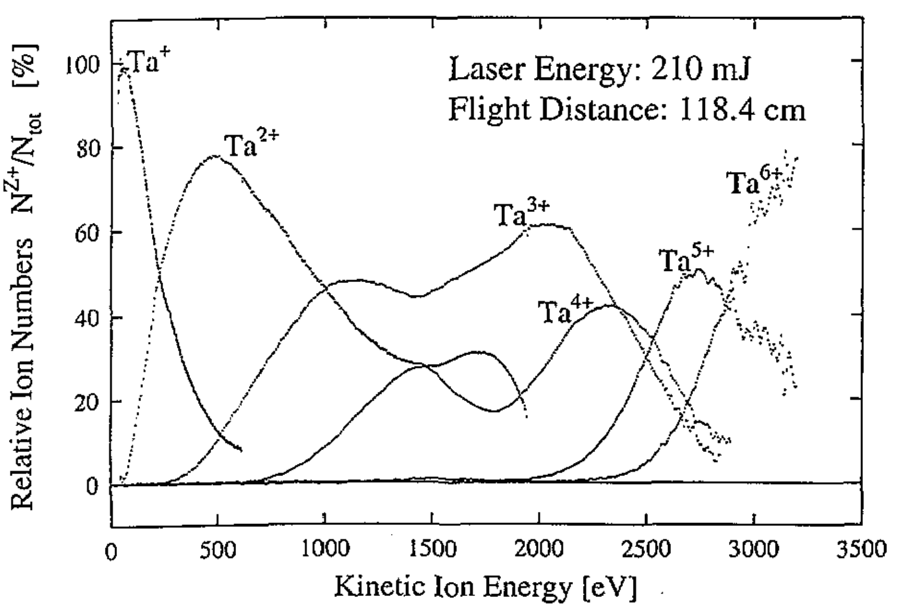


**Figure 2.** Spectra of the relative ion numbers $N^{Z+}/N_{tot}$ for $Ta^+$ to $Ta^{6+}$ at a flight distance of 118.4 cm. The laser energy is 210 mJ (about $10^{11}$ W cm$^{-2}$). Note that there is a characteristic energy region in which each charge state dominates.

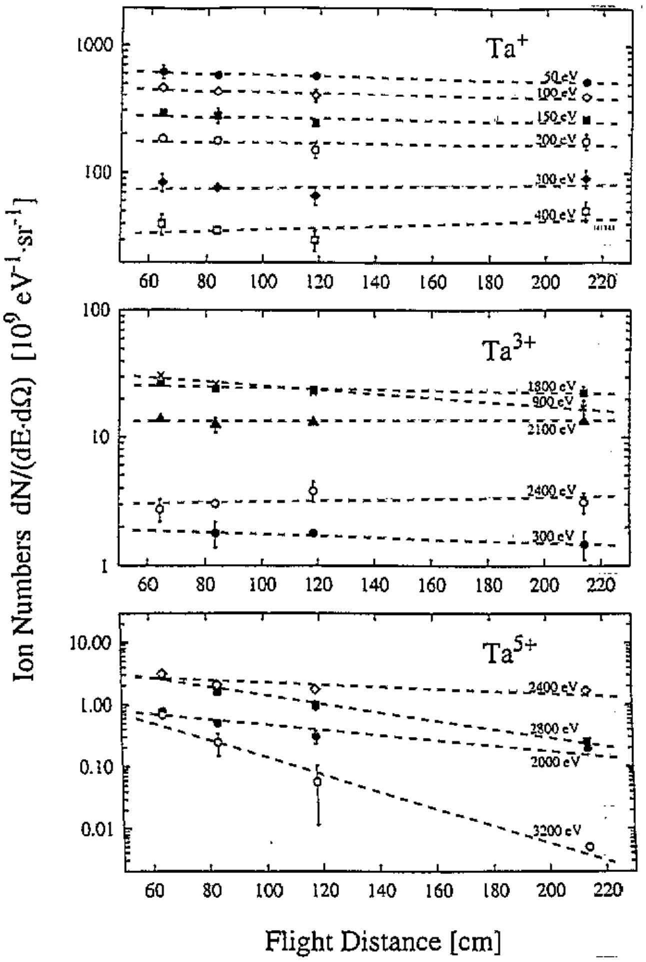


**Figure 3.** Numbers of the ions $dN/(dE d\Omega)$ of $Ta^{+}$, $Ta^{3+}$, and $Ta^{5+}$ at several kinetic energies as a function of the flight distance. Straight lines are drawn through the data points as visual guides. No unique trend is observed, instead of that the behaviour is dependent on the charge, density and velocity of the local groups.

some simplified conclusions can be drawn in terms of the three essential parameters that determine the recombination rate, namely the charge, density and temperature (Hinnov and Hirschberg 1962). As could be expected from the temperature-dependence, a decrease is observed for all ion species in the low-energy tail of the spectra. Similarly, a relatively strong decrease is observed for the $Ta^{5+}$ ions for all kinetic energies, which should primarily be correlated with the $Z$-dependence of the recombination rate. Apparently, both trends can effectively be modified by the density of the corresponding more highly charged ions at this energy, as can be concluded with the help of figure 2. Striking examples are for the $Ta^{+}$ at about 400 eV and the $Ta^{3+}$ in the range about 2000–2300 eV. Surprisingly, at these energies the ion numbers increase with the flight distance.

By using the interpretation in terms of atomic processes, there is no need to assume, as for very high-intensity laser–plasma interactions, that two different ion components, fast and thermal, correlated to different temperatures or lateral heat flow effects, are responsible for the structures in the energy spectra in this experiment.

In order to get data comparable with the two published measurements (Tallents 1980, Golubev *et al* 1984), the energy spectra of the different ion species have been integrated to give total particle numbers. In figure 4 results are presented for the $Ta^{+}$ to $Ta^{6+}$ components and for all ions (upper curve) as a function of the flight distance. For the $Ta^{+}$ and $Ta^{2+}$ ions the net change in the particle numbers is essentially balanced at all target distances. This might suggest a frozen state, at first sight. However, for the ion species with charge $Z > 2$ a continuous decrease is observed over the whole expansion range. The effect becomes stronger, the greater the ions' charge and the closer the distance to the target. Figure 4 shows in addition the usual behaviour that the ion numbers strongly decrease for higher charge states (Demtröder and Jantz 1970, Mann and Rohr 1992). For the present experimental situation and a laser energy of 210 mJ, the numbers of the singly and doubly charged ions are nearly $10^{14}$ $sr^{-1}$, representing more than 80% of the total ion flux and therefore the effect on the decrease of the total numbers of ions, which corresponds to an increase in number of the neutral species (Thum *et al* 1994) remains relatively weak (about 5%) within the measurement range. No frozen state is observed, however, in agreement with the results of Golubev *et al* (1984). In this experiment, in which the change in the total charge was investigated, a continuous decrease was obtained up to flow distances of more than 3 m. The findings are in contrast, on the other hand, to the experiment of Tallents, who observed an abrupt freezing at about 1.5 m although there the focal intensity was higher by about an order of magnitude. Using scaling laws of Donaldson *et al* (1979) and Sinha and Gopi (1979), one would similarly expect a higher initial plasma temperature, which should finally result (above all due to the strong $T^{-9/2}$-dependence of the three-body recombination rate) in an inhibited recombination.

Theoretically the group structure in the ion energy spectra of figure 2, sometimes according to a misleading expression called ion separation, has been consistently interpreted as an effect of the atomic interaction dynamics during the expansion, namely of the ionization and three-body recombination processes (Goforth and Hammerling 1976, Matzen and Pearlman 1979, Kunz and Mulser 1982, Caruso *et al* 1983, Latyshev and Rudskoi 1985, Kunz 1986, Stevefelt and Collins 1991). Even under crude assumptions about the initial state of the plasma, it emerges from these calculations that the initially faster ions experience less recombination, which finally produces energetically separated charge groups. The relative size and the dynamical behaviour of the individual ion spectra and consequently the appearance of a frozen state-like situation depend critically, however, upon assumptions about the degree of recombination heating (Kunz and Mulser 1982, Kunz 1986), that is the amount of energy that is conserved in the three-body recombination process, or some limitations in the heat conduction (Payne *et al* 1978), both shifting the plasma expansion more or less from adiabatic to isothermal (Zel'dovich and Raizer 1967,

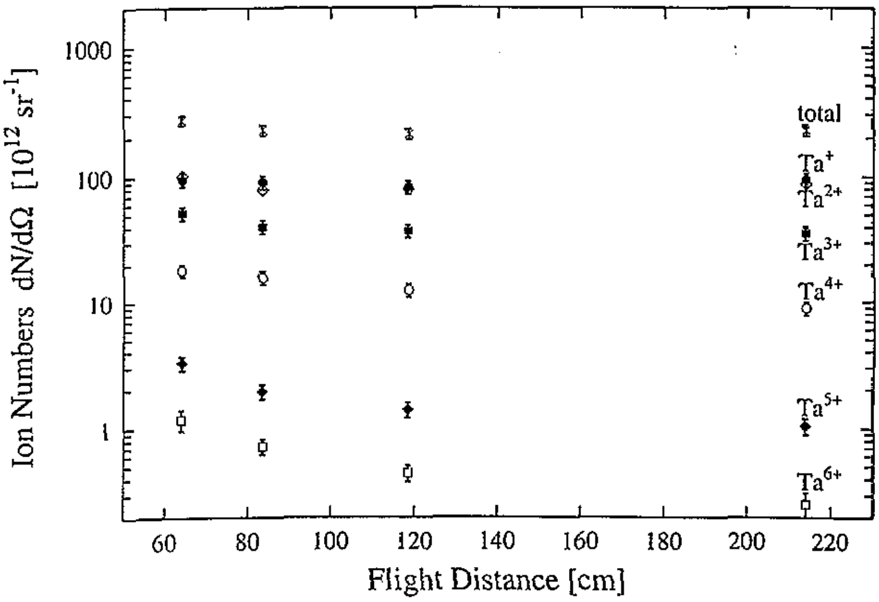


**Figure 4.** Energy-integrated ion numbers $dN/d\Omega$ for $Ta^{+}$ to $Ta^{6+}$ (lower curves) and for the total particle current (upper curve) as a function of the flight distance. The data points are connected by visual fits. Although no frozen state is observed, the overall decrease in the number of ions with flight distance is relatively small due to the fact that the net changes in number of the singly and doubly charged species, whose contribution is about 80% of the total, nearly cancel.

Kunz and Mulser 1982). Until now there have been neither quantitative experimental foundations for these assumptions nor possibilities for a feedback between calculated and experimental ion data. Detailed results like those of the present experiment should allow for the first time quantitative tests of model calculations in the asymptotic range.

## 4. Conclusions

We have measured with high precision the development of the ion distribution in a laser-produced Ta plasma during the expansion in a range up to about 2 m. The changes in the ion composition have been measured in quantitative units and resolved according to the ion velocities. While for the energy-averaged ion numbers a frozen state-like situation might be assumed, a much more sophisticated picture of the complex recombination dynamics is obtained from the resolved data, from which it becomes directly apparent, that the recombination rate is indeed dependent upon the charge and density structure in individual local ion groups, each expanding with its own velocity. It can be expected that these data will stimulate realistic theoretical model calculations and be helpful for an optimization of practical applications of laser-produced ion beams.

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