J. Phys. D: Appl. Phys. 27 (1994) 1791–1794. Printed in the UK

RAPID COMMUNICATION

Two-component structure in the angular emission of a laser-produced Ta plasma

A Thum, A Rupp and K Rohr

Universität Kaiserslautern, FB Physik, D 67663 Kaiserslautern, Germany

Abstract. The particle flux produced by an obliquely incident Nd *Q*-switched pulse (20 ns) on a Ta target has been analysed with regard to its angular distribution resolved for both its neutral and ion components. The laser intensity has been varied in the range between about 10^{10}–10^{11} W cm^{-2}, which is appropriate for many low-irradiance applications. It is observed that, at all emission angles and for the whole range of laser intensities, the number of neutral species clearly dominates the composition of the particles. At 1.3×10^{10} W cm^{-2} the total number of emitted particles is 4×10^{14}, scaling as $E_L^{3/4}$ with the laser energy. While for relatively low laser energies the angular distribution shows the usual smooth cos-behaviour, an additional strong directive emission cone, superimposed upon the cos-distribution, develops if the laser energy is enhanced. Both the strength and the width strongly depend on the laser intensity. While at lower intensities a fit by a $\cos^n$ function with $n \approx 10$ seems appropriate, n increases to 26 at an intensity of 10^{11} W cm^{-2}. It can be assumed that secondary energy transfer processes that are not yet fully understood are responsible for this anomalous emission.

Interest in detailed information on the particle composition of laser-produced plasmas is being stimulated by a series of applications. Among these laser ablation and deposition techniques have gained considerable importance in recent years as unique methods for fabricating thin films of high-T_c superconductors, oxides, semiconductors and diamond-like carbon. For optimization of these applications reliable data about the angular emission characteristics of the plasma pulse and its dependence on the experimental situation are essential (Pompe *et al* 1992).

Considerable confusion originates, however, from published measurements of angular distributions of the particle flux, ranging from $\cos^1$ to $\cos^{170}$ fits (Neifeld *et al* 1988, Gorbunov and Konov 1990, Sajjadi *et al* 1990, Champeaux *et al* 1993). To a large extent, this unsatisfactory situation arises from the fact that most of the data have been deduced from densitometric investigations of the deposited material which can represent the originally ablated particle beam only indirectly. Densitometric methods, in addition, give only averaged information concerning the ionic charge and kinetic energy distributions and their interaction during expansion. Thus they can scarcely give the information that is needed for theoretical modelling of the particle dynamics and flow characteristics, for example.

In the present paper we have therefore detected directly the particle emission of a laser-produced Ta plasma into the half-space. The plasma itself was created under experimental conditions and laser intensities typical for the above-mentioned applications. For the first time, angular distributions of the particles are presented in absolute quantities, resolved according to the numbers of neutrals and each ion charge. The results, in addition, show that the recently discussed two-component structure in the emission characteristic

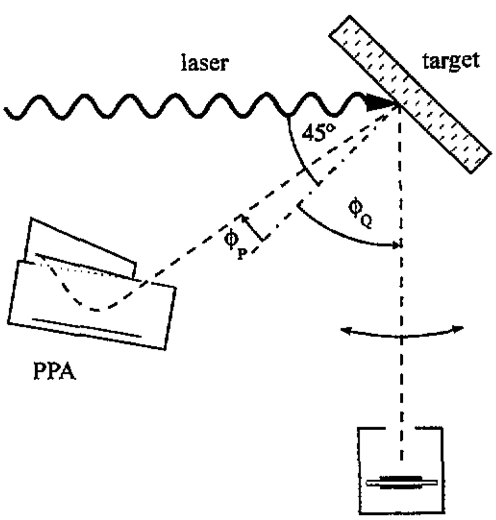

Figure 1. Schematic diagram of the experimental set-up. The laser pulse is incident at a fixed angle of 45°. Particle analysis is in the plane of incidence by a combination of a multichannel dynamical ion analyser (PPA) and a Quartz crystal (QC).

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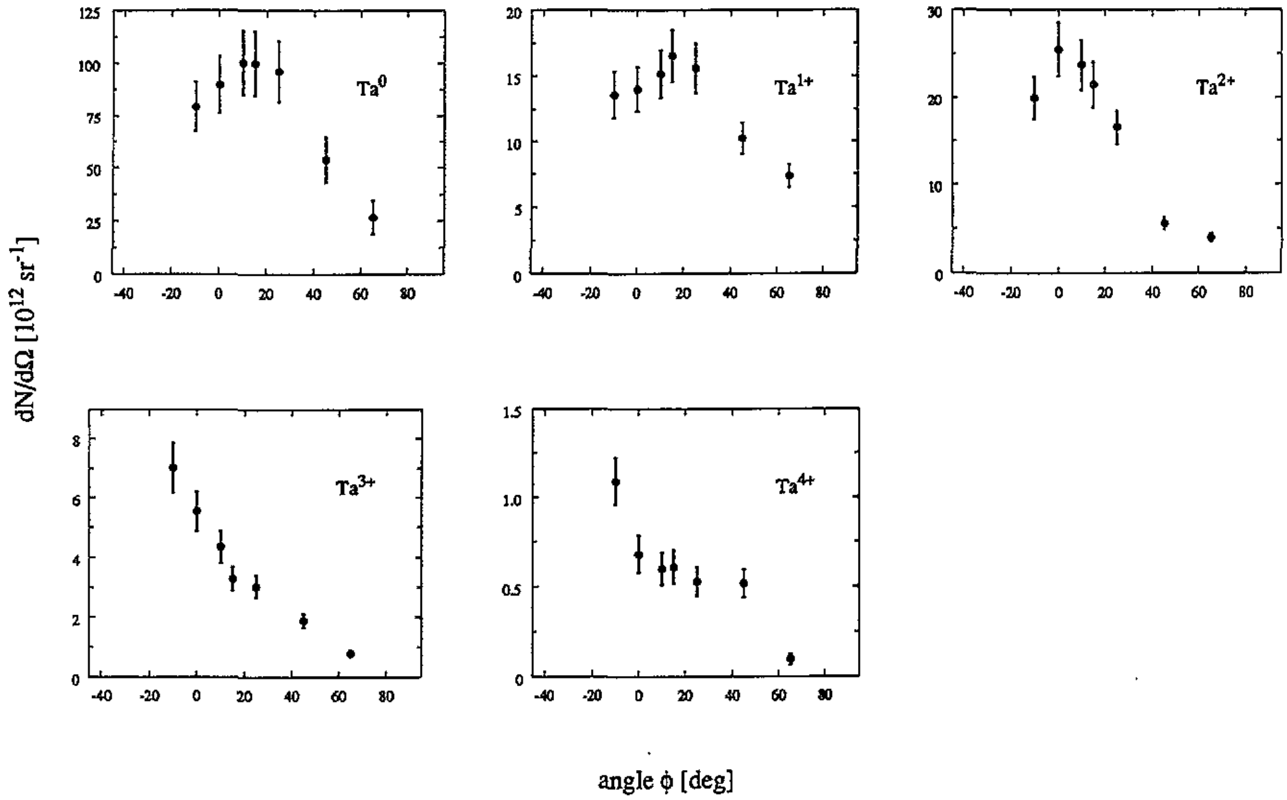

Figure 2. Angular dependences $dN/d\Omega$ of the emitted particles Ta^0 to Ta^{4+} at laser energy 21 mJ corresponding to an intensity of 1.3×10^{10} W cm^{-2}. The number of neutral species is obtained by the difference between the total particle signal of the crystal and the summed ion signal of the dynamical analyser. The vast majority of the particles are neutral species and ions of low charge.

(Venkatesan *et al* 1988, Urbassek and Sibold 1993) is a general feature in no way limited to multi-atomic species, which, however, is strongly dependent on the laser intensity. This latter observation might in part be able to explain the discrepancies in existing data.

A schematic representation of the experimental arrangement is given in figure 1. The plasma is created by an Nd:YAG Q-switched pulse ($\tau = 20$ ns) in the TEM_{00} mode of variable energy incident at a fixed angle of $-45°$ onto a flat Ta target inside a vacuum chamber. The laser energy is in the range 20–200 mJ and is focused to corresponding intensities about 10^{10}–10^{11} W cm^{-2}, which is large enough to avoid formation of clusters and droplets. The particles of the freely expanding plasma are detected in the angular range between $\phi = 65°$ and $-10°$ relative to the target normal by moving the analysers around within the plane of incidence at a distance of 64 cm from the target. Detection of ions is by means of a multichannel dynamical analyser (Eicher *et al* 1983, Rupp 1994), which allows 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. The neutral component is deduced from the difference of the total particle signal obtained from the frequency change of a quartz crystal after the plasma has been deposited and the number of ions measured by the dynamical analyser.

In order to increase the reliability of the results some effort has been essential concerning above all the transmission function of the detection systems, temporal stability of the laser and reproducibility of conditions in the interaction regime at the target. The linearity of the frequency response, the sticking efficiency and the effects of temperature on the crystal have been carefully observed. We assume that the remaining overall uncertainty of the data thus obtained lies between 10% and 40%, depending on the signal strength. Further details of the experimental arrangement and measurement procedure have been described previously (Mann and Rohr 1992).

Figure 2 shows the emission behaviour of differently charged particles of the tantalum plasma Ta^0 to Ta^{4+} in an angular range from $-10°$ to $65°$. The data points represent the energy-integrated numbers of each species at this emission angle. The focal intensity (FWHM) obtained by the pin-hole method (Dinger *et al* 1986) is 1.3×10^{10} W cm^{-2}, corresponding to a laser energy of 21 mJ. Obviously, the majority of the detected particles are neutral species. This general behaviour holds for the whole laser energy range investigated. At laser intensity 1.3×10^{10} W cm^{-2} the contribution of the neutral species is about 70% for all angles. If the laser intensity is increased to 2×10^{11} W cm^{-2} (280 mJ) then the relative contribution of the ions gradually increases to about 40%, which, however, is essentially due only

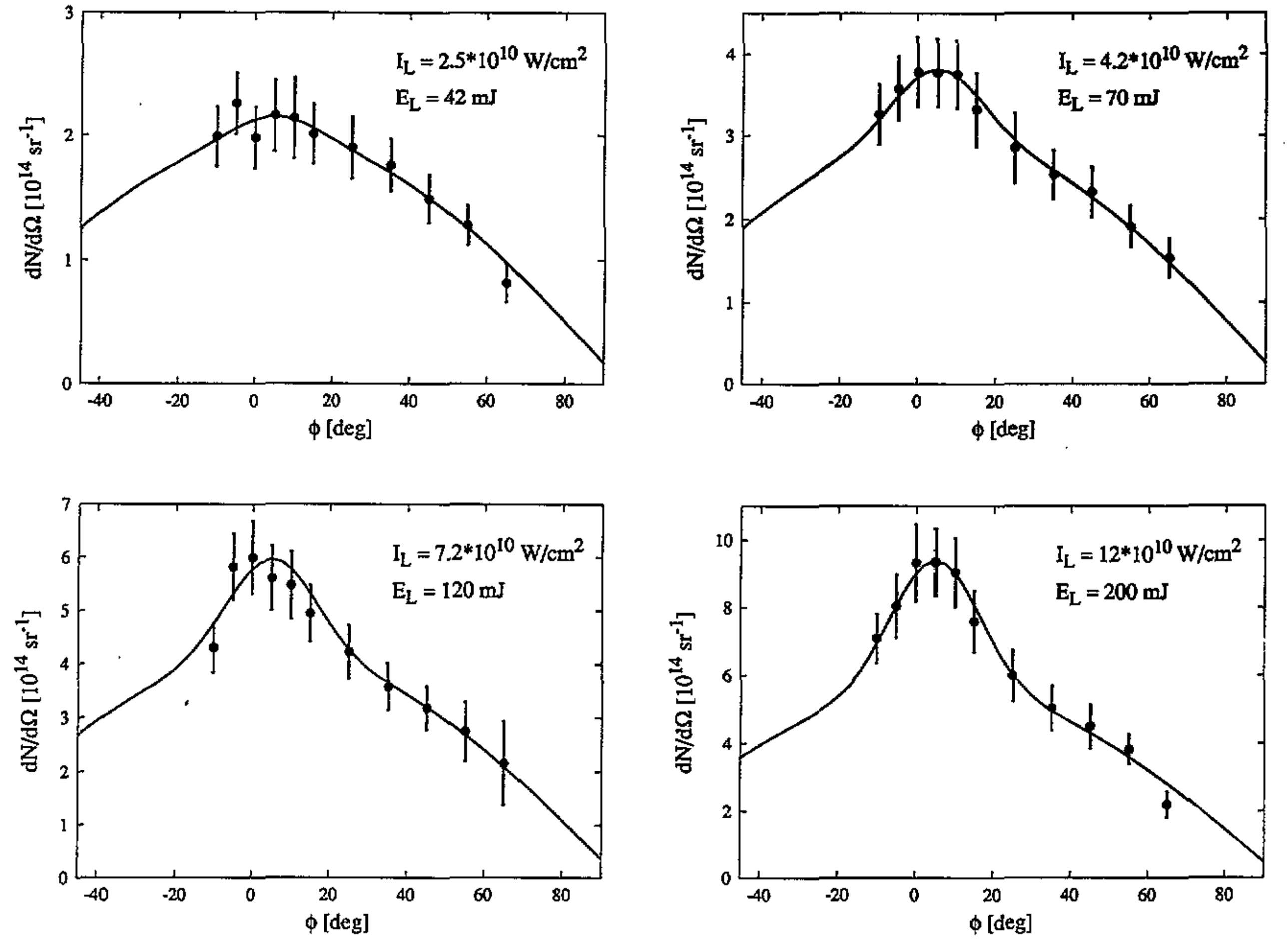

Figure 3. Angular dependences $dN/d\Omega$ of the total number of emitted particles at four laser energies (intensities). It is observed that, with increasing laser energy the flow characteristic in the range of the target normal is steepened by an anomalous emission. The full curves represent $a\cos^1 + b\cos^{26}$ fit functions.

to a relative enhancement of the higher charged species Ta3+ and still higher charges.

The preferential direction for emission of the particles is rotated relative to the target normal, depending on the charge of particles. This finding is well known for experiments with obliquely incident laser pulses. It can be explained by the unsymmetrical interaction of the laser with the ablating plasma current. Therefore the effect becomes stronger the smaller the laser energy and is absent if the laser is incident normally (Mulser *et al* 1973, Dinger *et al* 1980) or if the pulse length is short relative to the plasma expansion time (Bykovskii *et al* 1988).

In order to get data comparable with published measurements of angular distributions, the spectra of the different species are summed to give the emission characteristics of the totally ablated particle numbers. In figure 3 results are presented for four laser intensities between 2.5×10^{10} and 1.2×10^{11} W cm^{-2}. For the lowest intensity one finds a relatively smooth angular behaviour, which can be approximated by a simple cos function. If the laser intensity is increased, however, an additional, steeper emission cone develops. At higher laser energies this anomalous distribution can be well fitted by a $\cos^{26}$ function superimposed on the cos fit. At smaller laser energies the exponent of the fit function could likewise be of the order of 10 and below. The direction of the maximum is relatively stable and, for the range of laser intensities of the present experiment, positioned between 10° and 0°.

Assuming rotational symmetry, which has been confirmed by single wide-angle and out-of-plane spot measurements, the angular spectra could be integrated over the half-space and summed for the different species. The total numbers of particles thus obtained range from about 4×10^{14} to 2.8×10^{15} for laser energies 21–280 mJ. The relative contribution of the particles in the highly directed structure remains negligible, however, reaching only about 5% at maximum laser intensity. The functional behaviour of the particle numbers can be well approximated by a simple power law of the laser energy, scaling as $E_L^{3/4}$ for the cos distribution, and approximately scaling as E_L for the narrow distribution. In both cases this essentially represents the distribution of neutral species and singly and doubly charged ions.

The presently observed two-component structure can be compared with the angular distribution deduced by Venkatesan *et al* (1988) from a deposited multi-atomic film of superconducting material. In that experiment similarly, a broad cos distribution is observed, which is superimposed upon a steep component for which an angle-dependence 'sharper than $\cos^{11}$' is estimated. However, in contrast to the present experiment, there the steep angular structure appears already at a laser intensity about three orders of magnitude smaller. This might be due to the strong dependence of laser light absorption on the surface structure, which is quite different in the two cases (Rupp and Rohr 1991).

Referring to the ions only, it is well known that a smooth behaviour, in both angular behaviour and energy spectra is characteristic of ion beams produced at low and medium laser intensities. In particular, differential measurements of the ion yield from laser-produced C plasmas show that the emission characteristic can be fitted approximately by $\cos^{(2z+1)}$ dependences, where z is the ions' charge (Mann and Rohr 1992). Generally, in this range the ion spectra can be consistently interpreted if atomic interactions, namely ionization and three-body recombination, are assumed to be the essential processes dominating the expansion dynamics (Goforth and Hammerling 1976, Caruso *et al* 1983, Kunz and Mulser 1982, Kunz 1986, Stevefelt and Collins 1991). When the laser intensity is enhanced to above about 10^{11} W cm^{-2} additional structures have been observed in the ion energy distributions (Ehler 1975, Rohr *et al* 1989, Mann and Rohr 1992). These structures, sometimes called thermal and fast ion groups, have been correlated with the different electron temperatures that have been shown to exist in the focal region (Eidmann 1975, Stenz *et al* 1977). In addition it was observed that the angular distribution of the fast ions was more peaked than the thermal distribution (Church *et al* 1982). However, these effects are closely correlated with the charge of the ions, while in the present experiment the neutral species massively dominate the particle distribution.

On the other hand, there have been several attempts to explain the structures of the deposited films. Kelly and Dreyfus (1988) argue that the highly directed component is a result of the formation of a Knudsen layer, which should shift the particle spectrum to higher velocities for normal emission and narrow the angular distribution. Urbassek and Sibold (1993) and Sibold and Urbassek (1993) have recently studied gas-phase collisions of a gas desorbed from finite-area targets using a three-dimensional Monte Carlo simulation. They show that, for both mono- and polyatomic targets a desorption jet is formed, in which fast particles are focused towards the jet axis, and influence the angle and energy distribution in the beam. In this calculation the steepening of the emission increases with deposited laser energy, in agreement with the present observation.

It seems that the simulation can explain the steep angular structure of the present experiment, in which the large neutral species component dominates the plasma composition. The broad cos distribution, on the other hand, might be, as with the ionic results, essentially affected by atomic interactions.

Acknowledgment

This work was supported by the Deutsche Forschungsgemeinschaft.

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