Segregation of neutrals and ions in laser ablated plasmas from binary targets

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Abstract

In the present experiment we have investigated in detail emission characteristics of laser produced plasma pulses ablated from monoatomic targets Al, Ti, Cu, W and their compounds TiAl and WCu of different stoichiometric mass ratios. The plasma was produced by obliquely incident Q-switched pulses ($E_L = 130$ mJ, $\tau = 5$ ns and $\lambda = 1.06$ μm) focused to a focal area of 0.3 mm². Angular emission distributions of the compounds have been compared with those of the pure components, resolved according to the total number of particles and the neutrals and ions of different charge states. The following major results are observed: (1) All emission distributions can be well fitted by Gauß-functions. (2) The emission cones of both, ions and total number of particles, steepen as a function of the average atomic mass A and can be well approximated by $\Gamma = k/A + c$, whereby k systematically depends on the size of the focal area. This should allow quite simply predictions of emission distributions of arbitrary compounds for many cases of practical applications

Introduction

Laser initiated ablation of neutral particles, ions and clusters has gained considerable importance in materials synthesis techniques. Above all angular emission distribution of multiatomic materials are of particular interest for a straightforward optimization. In the present contribution we present, for the first time, systematic comparative measurements of angular emission distributions from two series of binary targets TiAl and WCu of variable stoichiometric ratios. The measurements give results which are differential according to absolute particle numbers (masses), charges and kinetic energies and reveal several basic scaling laws for the emission of laser produced plasmas from compounds.

Experiment

The plasma is created by a Nd-YAG Q-switch pulse ($E_L = 130$ mJ, $\tau = 5$ ns and $\lambda = 1.06$ μm) incident at a fixed angle of -45° onto flat targets inside a vacuum chamber. The laser is

focused to an area of about 0.3 mm^2 (1/e^2 width). Target materials have been TiAl and WCu compound systems and pure ^{27}Al, ^{48}Ti, ^{64}Cu and ^{184}W metals. Stoichiometric mass ratios of the compounds have been 25/75, 34/66, 50/50 and 75/25 for TiAl and 60/40, 80/20 and 90/10 for WCu, respectively. Thereby the (average) atomic mass ranges from 27 to 184. The emission of ions and neutrals are detected angular resolved within the plane of incidence by moving around the analyzers at a distance of 35 cm from the target. The ion spectra are fully resolved by the time-of-flight/retarding potential method which makes it possible to obtain the absolute number of each species. The total number of particles are determined from the frequency change of the crystal after the plasma has been deposited. The number of the neutral particles can be deduced from the difference between the total particle signal and the summed spectra of the different ion species. Considerable effort was invested in the components and the measurement procedure to increase the reliability of the results.

Results

Figures 1 compares measured angular emission distributions of the ablated mass pulses: on the left, pure Al and Ti and on the right, four compounds TiAl with stoichiometric ratios 25/75, 34/66, 50/50 and 75/25. The mass values are deduced from the frequency changes of the quartz detector after deposition of the particle pulses. In the case of the monoatomics the results directly correspond to the total number of particles. In all cases the observed distributions are essentially those of the neutral part of the plasma pulse, which represents more than 90% of the total number of particles under the present conditions. All spectra are fitted by Gauß functions and obviously, this approximation is particularly suitable to describe the shape of the emissions in each case quantitatively. Moreover, the wide angle measurements , in principle up to 90°, show that all distributions, total particle numbers, neutrals and ions, can be well approximated by simple Gauß-functions $a \cdot e^{-(\Theta-\varepsilon)^2/2\cdot\Gamma^2}$.

This finding that i.e. the shape of the emission can be fully described by one single parameter, namely the width Γ, considerably simplifies comparison of emission distributions and allows to formulate scaling laws of increased significance. In the past, emission cones have often been discussed, both in experimental and in theoretical contributions, as a two-component structure, approximated by the superposition of cosine functions, e.g. $a \cdot \cos(\Theta-\varepsilon) + b \cdot \cos^n(\Theta-\varepsilon)$. As far as this approximation has been considered as an approach to discuss emission shapes from different experiments it has proven to be of minor usefulness. Certainly part of the confusing data for e.g. the exponent n of the peaked

structure obtained from experimental results, which is ranging between one and more than two hundred originates from this fit procedure.
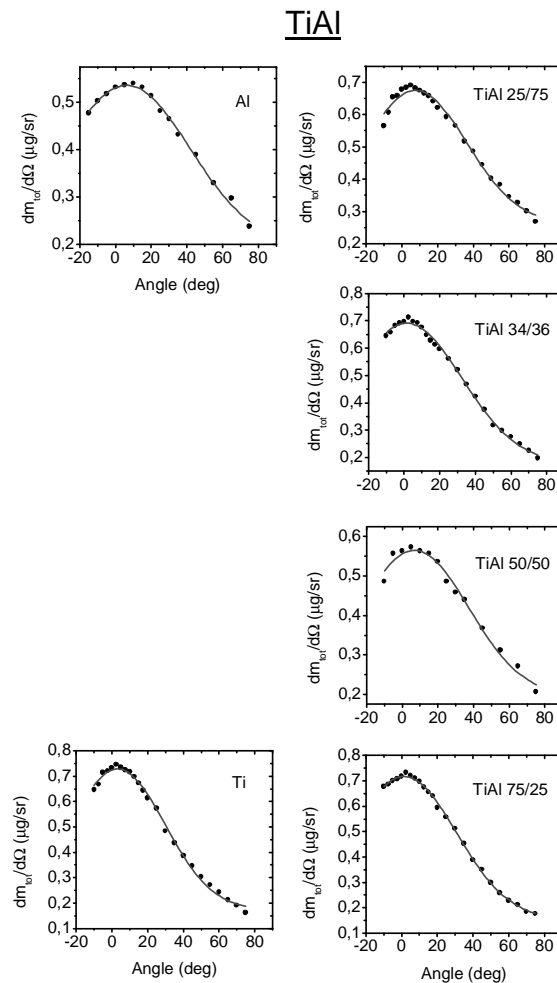
TiAl

Figure 1: Angular emission distributions of laser ablated particle masses from pure Al and Ti targets (left column) and from TiAl compounds of variable stoichiometry (right column). All data can be well fitted by Gauß functions (solid lines).

Obviously the emission cones in figure 1 get smaller continuously with increasing (average) mass. This finding holds for the whole range of target masses from 24 to 184. In figure 2 measured widths (open triangles) are depicted as a function of the average atomic mass of the target materials. Analytically, the dependence of Γ on the atomic mass A can be well approximated by a hyperbola function $\Gamma = k/A + c$ (full line). Comparison with independent foregoing experiments [1, 2] show that this dependence seems to be valid universally, whereby it was found that k systematically depends on the size of the focal area. This suggest, that it should be able to predict emission widths of any materials, in particular of complex or difficult to handle species. As a test we have compared calculated widths of the

compound systems by using a simple superposition of single components results for Al, Ti, Cu and W and considering the corresponding stoichiometric ratios (open circles and dashed line in fig.2)
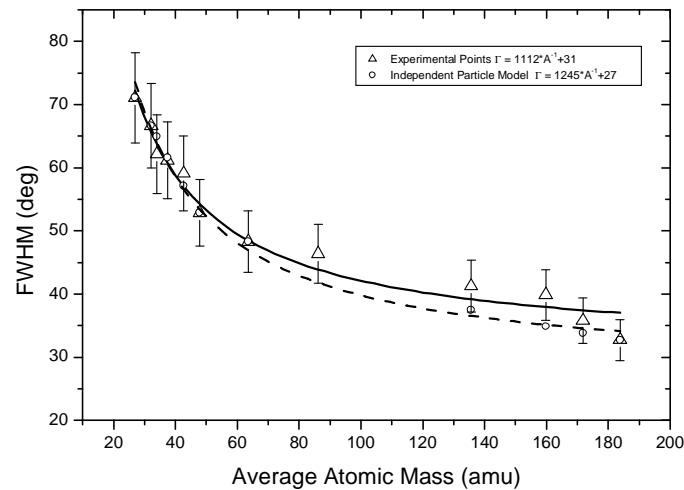

Fig. 2: Comparison of measured Gaussian widths Γ of the emission cones (open triangles), approximated by a hyperbola function $\Gamma = 1112/A + 31$ (solid line) with widths calculated by the superposition of the widths of the corresponding monoatomics with the appropriate stoichiometry ratios (open circles), approximated by $\Gamma = 1245/A + 27$ (dashed line).

Conclusion

The following major findings are observed:

- All emission distributions can be well approximated by a Gauß-fit which simplifies comparison with different results. .

- Widths of the emission cones of all kinds of particles, ions and neutrals behave as the inverse of the average atomic mass. Data can be well fitted by a approximated by a hyperbola function $\Gamma = k/A + c$ where k systematically depends on the focal spot size.

- The measured widths of the compounds can be well approximated by a simple superposition of the widths of the corresponding monoatomics considering the right stoichiometry which allows generally predictions of emission distributions of arbitrary compounds in many cases of practical applications.

References

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