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Effect of irradiation angle on the efficiency of formation of multiply charged ions in a laser-produced plasma

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Abstract. Mass spectrometry is used to investigate the emission behaviour and the characteristics of multiply charged ions in a plasma produced at small angles of incidence of laser radiation ($\alpha \sim 20^\circ$) and also at grazing incidence ($\alpha \sim 85^\circ$). It is found that upon grazing incidence of the laser radiation onto a target, the efficiency of production of multiply charged ions is reduced compared to that for $\alpha \sim 20^\circ$. However, this geometry of laser irradiation of solids can be used for the elemental analysis of surface layers of a sample.

1. Introduction

A strong electric field is known to arise from the resonance oscillation of plasma electrons for certain parameters of laser radiation and radiation-target interaction geometry [1, 2]. Multiphoton effects, such as, ionisation and emission of charged particles [3], which depend on the angle of radiation incidence relative to the surface, also make a contribution to the formation of near-surface electric fields. All these phenomena affect the velocity, the expansion dynamics, and the number of ions accelerated in these fields.

We note that investigations of the characteristics of charged-particle emission from a plasma were restricted so far primarily to the two cases: studies upon sharp focusing of laser radiation on a target and at a relatively small angle of incidence of laser radiation ($\alpha < 45^\circ$). Only in a few (e.g., see Ref. [4]) papers on mass spectrometric analysis of materials, upon multiphoton ionisation of vaporisation products, the laser beam was directed along the target surface, i.e., perpendicular to the direction of ion expansion. To improve the reliability and sensitivity of the layer-to-layer elemental analysis of materials and determination of the composition of films adsorbed on the target surface, it is necessary to augment the ion-optical emissive capacity of the ion source of the mass spectrometer. This requires a change in either the irradiation parameters or the experimental geometry.

This work is concerned with experimental investigation of the emission behaviour and the characteristics of ions in a plasma produced at oblique incidence (nearly grazing incidence relative to the target surface) of laser radiation.

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2. Experimental

The studies were carried out using a setup with a laser ion source, which comprised a time-of-flight mass spectrometer combined with an electrostatic analyser. The ion drift path was ~ 150 cm, and the mass resolving power was ~ 100. The plasma was produced by a Q-switched Nd laser. The pulse length ($\tau = 80$ ns) and the pulse energy ($E = 2$ J) were maintained constant throughout the experiment. The angle of radiation incidence was limited by tangency of the generatrix of the light channel cone with the surface of the samples, as shown in Fig. 1. Provision was made to change the orientation of the polarisation plane relative to the target normal.

The laser radiation was focused with a lens of focal length $f = 300$ mm. The radiation with a Gaussian distribution over the cross section, a beam diameter of 15 mm, and a divergence $\theta = 10^{-3}$ rad was incident on a sample at an angle $\alpha \sim 85^\circ$. The beam was focused to produce on the surface an oval spot of length $l \sim 5$ mm and width $d \sim 0.5$ mm.

The samples were rolled tungsten plates and high-temperature superconducting $YBa_2Cu_3O_{7-x}$ ceramics formed into disk tablets 10 or 25 mm in diameter. The target was attached to a coaxial electrode, with provision made to change the location of the irradiation area on the sample surface. A collector in the form of a 50-mm-long cylinder 30 mm in diameter and located on the front side of the target, carried a noninductive resistor load with a resistance $R = 4.3\ \Omega$.

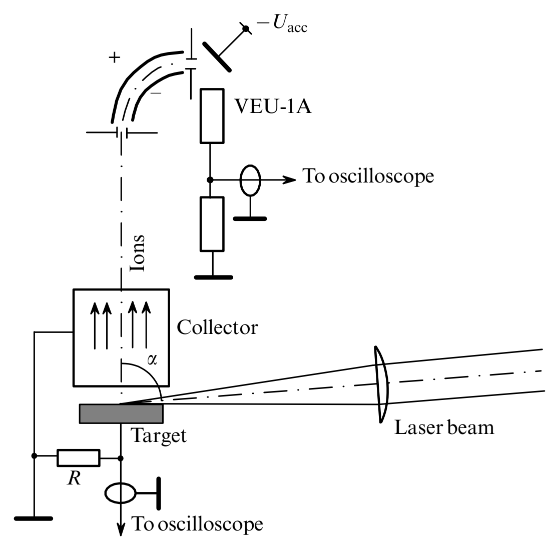

Figure 1. Schematic of the mass spectrometer with a laser ion source for grazing incidence of the laser radiation on the target.

A pulsed current generated by the laser-produced plasma was measured simultaneously with the ion signal recorded at the output of the mass spectrometer.

A pressure $p \leqslant 1.3 \times 10^{-5}$ Pa was maintained in the system. To elucidate the role of the angle of radiation incidence on the target, the investigation was carried out for $\alpha \sim 85$ and $\sim 20°$, other factors being invariable.

3. Results

The data obtained at oblique incidence of the radiation focused onto the target surface, with the plane of polarisation normal to the plane of the target, differ significantly from those obtained at traditional (near-normal) angles of radiation incidence on a sample. In accordance with the focusing conditions, a nearly round crater is produced at the target surface for $\alpha \sim 20°$ and the crater diameter does not exceed the cross section of the focal spot. For $\alpha \sim 85°$, the imprint is a 5-mm-long oval with a maximum width of 0.5 mm and a significantly (by an order of magnitude and over) smaller depth than for a small angle relative to the normal.

The ion mass spectra of the plasma produced by laser irradiation at $\alpha \sim 85°$ are more fully representative of the elemental composition of the layer adsorbed at the surface of a tungsten sample (Fig. 2). In this case, the maximum multiplicity Z_{max} of the multiply charged ions that correspond to the sample matrix is always lower than Z_{max} for $\alpha \sim 20°$.

Upon a sequential irradiation of a sample by laser pulses at an angle $\alpha \sim 85°$, both the number and the intensities of the ion signals of impurity elements decrease. Simultaneously, the amplitude of the signal of tungsten ions and the ion multiplicity increase. For instance, in the case under discussion, the $O^+, O^{2+}, C^+, N^+, Na^+, K^{2+}, S^+, S^{2+}, Co^+$, and Co^{2+} ion lines of impurity elements are recorded in the mass spectra after the first pulse, while the multiplicity of W ions does not exceed three. After the third pulse, Z_{max} reaches four for W ions and only C and O ions are recorded of the impurity elements. For $\alpha \sim 20°$, the mass spectra are virtually identical over the sequence of ten pulses; for tungsten ions $Z_{max} = 5$.

The energy distributions of multiply charged ions strongly depend on the angle of target irradiation. For a nearly grazing incidence, the energy distribution range is significantly narrower than for acute angles of incidence. For instance, one can see from Fig. 3 that energy E_{max} of the W^+ and W^{4+} ions produced for $\alpha \sim 85°$ does not exceed 500 eV and $\sim$ 1 keV,{w respectively, whereas E_{max} amounts to $\sim$ 4 keV for these ions for $\alpha \sim 20°$.

Similar changes are observed in the ion energy spectra of the elements on which the high-temperature superconducting ceramics is based. For instance, E_{max} for oxygen ions produced for $\alpha \sim 85°$ does not exceed $\sim$ 100 eV but achieves $\sim$ 500 eV for $\alpha \sim 20°$. Also evident from Fig. 3 are significant changes in the energy spectra themselves: the spectrum has only one maximum for $\alpha \sim 85°$, while additional recombination maxima are observed for small α. This is indicative of a higher initial plasma charge.

The plasma-generated currents were also recorded. Unlike Ref. [1], our measurements were performed employing a cylindrical collector with a low-inductance ohmic load located near the target [5]. For different angles, the currents differ greatly. For instance, the pulsed current I_{max} amounts to $\sim$ 100 A at grazing incidence ($\alpha \sim 85°$) but $I_{max} \leqslant 45$ A for $\alpha \sim 20°$.

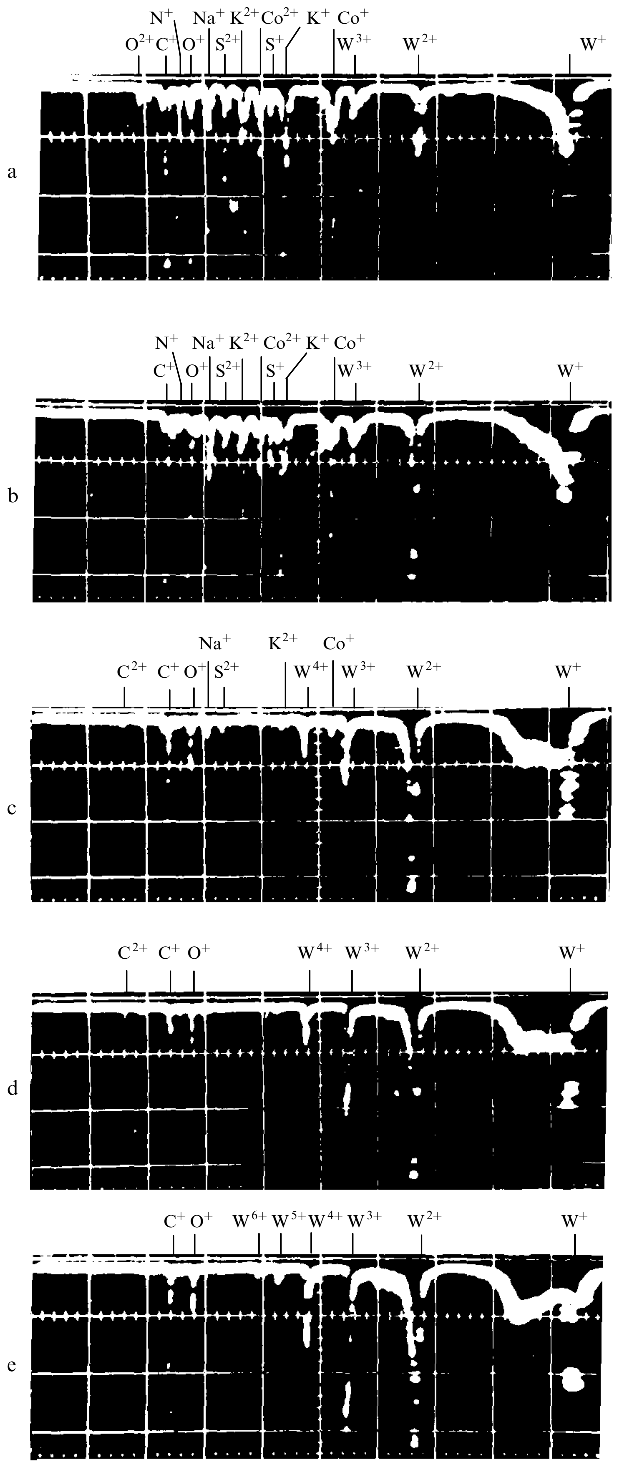

Figure 2. Mass spectra, recorded employing an oscilloscope, of the surface layer adsorbed on the surface of a tungsten sample for the first (a), second (b), third (c), and fourth (d) laser radiation pulse for an angle of incidence $\alpha \sim 85°$ and the mass spectrum for ten exposures to the laser radiation at $\alpha \sim 20°$ (e).

4. Discussion

Therefore, the qualitative form and the quantitative characteristics of the mass spectra, as well as the energy distributions of multiply charged ions and the plasma-generated currents depend significantly on the angle of incidence of laser radiation on a target. The effects revealed in this way

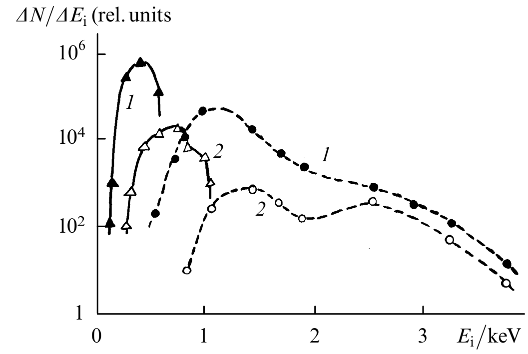

Figure 3. Energy spectra of tungsten ions for angles of laser irradiation $\alpha \sim 20^\circ$ (dashed lines) and 85° (solid lines), $E = 2$ J, $\tau = 80$ ns, $Z = 1$ (1) and 4 (2).

can be interpreted as follows. Let the radiation be incident on the target surface in the direction close to the target normal or at an acute angle relative to the normal. Then, for its intensity $q = 10^9 - 10^{10}$ W cm^{-2}and pulse length $\sim 10^{-8}$ s, a material layer gains an energy which far exceeds the heat of vaporisation of this material [6]. The resultant overheated layer acts as an explosive on the underlying target. A shock wave propagates inside the sample with a velocity $v \leqslant 10^5$ cm s^{-1}, resulting in vaporisation of the material (the dumping wave). The increase in vapour temperature results in its ionisation and a rapid increase of the absorption coefficient. Eventually the target surface is screened, and the internal energy of the resultant plasma builds up in this case. The formation of a plasma layer takes a very short time, and therefore the entire course of the process is governed primarily by the laser radiation-plasma interaction. During hydrodynamic expansion, the hot plasma additionally heats and vaporises the target ma-terial. A crater of certain size forms at the focal spot in the end.

Upon oblique (at an angle $\alpha \geqslant 80^\circ$) radiation incidence, the irradiation intensity decreases owing to the increase in the area of interaction with the target. A simplified calculation shows that the intensity q for $\alpha \geqslant 80^\circ$ is lower by approximately two orders of magnitude, i.e., $10^7 - 10^8$ W cm^{-2}. However, the effect does not amount exclusively to a reduction in the energy input. If the beam incident at $\alpha \sim 20^\circ$ is defocused to bring the spot area close to the area of the crater produced at oblique ($\alpha \sim 80^\circ$) radiation incidence on the sample surface, the radiation intensity will be no greater than 10^8 W cm^{-2}. In this case, for tungsten ions $Z_{\max} \leqslant 2$, i.e., lower than in the case of grazing incidence. Recorded in the mass spectrum are the ion signals of impurity elements (Na, S, Co), which were not recorded for higher $q > 10^9$ W cm^{-2}. However, the number of peaks recorded and their intensities for $\alpha \sim 20^\circ$ are much lower than for $\alpha \sim 85^\circ$.

The higher charge state of W ions and the increase in the number of detectable impurity elements recorded for $\alpha \sim 85^\circ$ is supposedly explained by the following. When the radiation is incident on the target at $\alpha \geqslant 85^\circ$, there occur a desorption of adsorbate gases contained in the surface layer of impurity elements (alkali metal ions, in particular) and a vaporisation of some quantity of the basic target material. Ions, neutral particles, and clusters ejected from the target surface find themselves in the laser radiation field of higher intensity ($q \geqslant 10^9$ W cm^{-2}), which exists in the volume of the lens caustic, and are ionised to the state of a high-temperature plasma.

Therefore, the laser radiation at grazing incidence brings about primarily a removal of surface impurities without appreciable erosion of the target material itself.

Significantly higher intensities and number of the ion peaks of impurity elements recorded at grazing incidence of the radiation can be explained by the following two factors. First, by a more efficient photoeffect (when the field intensity vector of the polarised laser radiation coincides with the preferential direction of particle escape from the target surface). Second, the impurity elements vaporised from the surface find themselves in a high-intensity radiation field. When the laser radiation is defocused (for $\alpha \sim 20^\circ$), the region of maximum radiation intensity ($q \geqslant 10^9$ W cm^{-2}) is offset from the target by a distance approximately three times longer than the caustic length of the lens employed. In our case, this distance is ~ 15 mm. Since the ejection velocity of the material vaporised by the laser beam does not exceed 10^5 cm s^{-1} [1], the vaporised particles do not enter the laser radiation field of maximum intensity during the pulse ($\tau \sim 80$ ns). The effect is that a higher fraction of the vaporised material persists as neutral particles or weakly ionised atoms than for $\alpha \sim 85^\circ$. That is why the mass spectrum of the plasma produced in this case contains a smaller number of peaks of impurity elements.

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