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Oxygen ion spectra in a two-elemental laser plasma

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Abstract. **The mass–charge and energy spectra of oxygen ions are studied in a plasma produced by laser irradiation of targets containing heavy elements with different masses (Sc, Ce, Lu) and close thermal parameters. The dependence of the maximum multiplicity of O^{2+} oxygen ions on the laser radiation intensity is determined. The maximum multiplicity is found to be independent of the elemental composition of a two-element target. The variation of the O^{+} and O^{2+} ion energy spectra and the broadening of the spectra of Sc, Ce, and Lu ions with multiplicities $Z = 1 - 3$ towards higher energies with increase in the mass of a heavy component (Sc, Ce, Lu) is found to occur owing to the energy exchange between light (O) and heavy (Sc, Ce, Lu) plasma ions.**

Keywords: *oxygen ions, laser plasma, mass – charge spectra.*

1. Introduction

The investigation of oxygen ion spectra in plasmas produced by laser irradiation of targets containing heavy elements with different masses and close thermal properties is of interest for the following reasons. The oxygen-containing media show promise for efficient sources of multiply charged ions and nuclei, high-temperature superconductors, plasma lasers, methods of laser elemental analysis, and nonlinear-optical media.

A wealth of theoretical and experimental studies on laser radiation–matter interaction have been performed to date (see, e.g., Refs [1 – 11]). The radiation intensity, wavelength, and focusing conditions, as well as the nature of the material, the density and the defectiveness of the target structure were found to affect the production of ions of different multiplicity in a laser plasma [1 – 8]. Many papers have been devoted to laser plasmas obtained with monoelemental metal samples and little attention has been given to the study of multielemental laser plasmas.

Note that the mass – charge, energy, and angular distributions of multiply charged ions were comprehensively studied for monoelemental plasmas in a broad laser-radiation intensity range for targets of different nature [2]. These experiments underlie the design and efficient operation of laser sources of multiply charged ions at accelerator complexes and facilities for elemental material analysis [2]. It was found that monoelemental plasmas are a source of multiply charged ions with multiplicities Z up to 25 and energies up to 40.0 keV and that their expansion occurs in a narrow solid angle.

In Refs [4, 9], the ion spectra of a multielemental plasma containing gold and silver ions at low concentrations were studied. The elemental target composition was found to affect the production of multiply charged ions upon ionisation and recombination. This effect consists in a selective reduction of recombination losses of highly charged ions of gold and silver in the presence of light hard elements of the base. The method of analysis of multiply charged ions by mass spectra [9] proposed on the basis of these data provides a 100-fold increase in the concentration sensitivity with retention of all the advantages of the laser mass-spectrometric analysis by singly charged ion spectra [2].

Note that the ion spectra of gases and the role of mutual influence of the ions of different elements that are simultaneously involved in ionisation, recombination, and 'freezing' are poorly studied in the literature [6, 7]. The authors of Refs [6, 7] analysed in detail the production of gas ions (hydrogen, nitrogen) and their energy and angular distributions depending on to the laser radiation intensity and the nature of a multielemental target.

In this paper, we consider the spectrum of oxygen ions and their charge and energy distributions in the laser irradiation intensity range $q = 0.1 - 100$ GW cm^{-2}. We also study the mutual influence of light oxygen ions and heavy ions of Sc, Ce, and Lu with close thermal parameters in a two-elemental laser plasma.

Experiments were performed using a time-of-flight laser mass spectrometer combined with an electrostatic analyser. The experimental setup is shown schematically in Fig. 1. We used in our experiments a neodymium laser with a pulse duration of 15 ns, wavelength of 1.06 μm, and a pulse energy of $\sim$ 0.5 J. The angle of a laser beam incidence on the target surface relative to the normal was $\sim 18^{\circ}$, and the radiation intensity on the target surface was $q = 0.1-$ 100 GW cm^{-2}. The plasma ions were detected at a distance of 100 cm along the target normal; the slit of the electrostatic analyser was 0.4 mm in width. The two-elemental targets under study were placed in the ion source chamber. The chamber design permitted a change of the irradiation spot on the target after every laser shot. The targets were Sc_2O_3, Ce_2O_3, and Lu_2O_3 oxygen-containing materials prepared as 5.0-mm thick tab-

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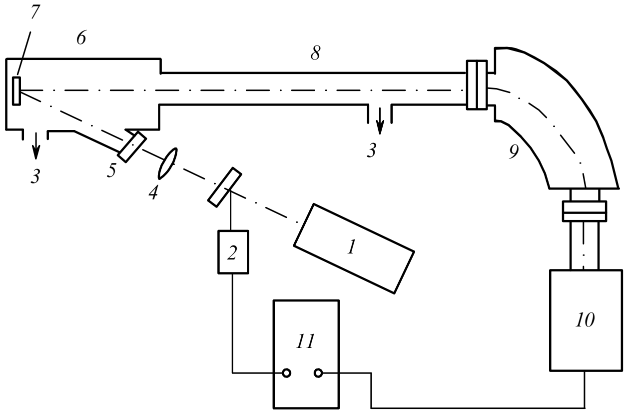

Figure 1. Schematic of the experimental setup: (*1*) solid-state laser; (*2*) coaxial photocell; (*3*) evacuation; (*4*) focusing lens; (*5*) entrance window; (*6*) vacuum chamber; (*7*) target; (*8*) time-of-flight tube; (*9*) electrostatic mass separator; (*10*) VEU-1B vacuum photomultiplier; (*11*) double-beam oscilloscope.

lets 10 mm in diameter. The operating pressure in the chamber was 10^{-6} Torr. The relative measurement error for a number of measurements $n = 4$ and the relative error in determining the ion signal amplitude were equal to 15 %; the relative error in determining the total number of ions $\Delta N/N$ was $\sim$ 30 %.

Our experiments yielded the mass–charge and energy ion distributions of a two-elemental laser plasma in which oxygen ions fly apart together with heavy Sc (Ce, Lu) ions. We studied the dependences of the characteristics of the resultant two-elemental plasma of interest to us on the laser radiation intensity and the elemental target composition. Relying on the experimental data obtained, we determined several physical features of the production and expansion of oxygen ions in a two-elemental plasma. These features consist in the characteristic changes of the mass – charge and energy ion distributions of both oxygen and the heavy component, in this case, Sc, Ce, and Lu. In our view, these changes are caused by the energy transfer between oxygen ions and heavy Sc (Ce, Lu) ions [10] during their joint expansion in a two-elemental plasma.

An analysis of the experimental results obtained by investigating the mass spectra of two-elemental plasmas containing oxygen ions showed that the form of the mass spectrum depends not only on the intensity of laser irradiation, but to a large degree on the elemental target compostion and the ion energy. Irrespective of the nature of a two-elemental target under study, the resultant mass spectra exhibit well-defined ion peaks of oxygen in different ionisation stages. The maximum intensities and charge multiplicities ($Z_{\max} = 2$) of oxygen ions in a two-elemental plasma are produced for $q = 100$ GW cm^{-2} and in the range of relatively low energies ($E = 40 - 120$ eV). In this case, the maximum multiplicity of oxygen ions $Z_{\max} = 2$ is independent of the nature of a two-elemental target under study in the $q = 0.1 - 100$ GW cm^{-2} range.

Fig. 2 shows typical mass – charge ion spectra of a two-elemental plasma produced by a laser beam with $q = 100$ GW cm^{-2} from CeO_2 and Lu_2O_3 for $E/Z = 25$ and 150 eV. One can see that the $Z = 1, 2$ oxygen ion peaks and the $Z =$ 1 peaks of Ce and Lu for $E/Z = 25$ eV and the $Z = 1$ oxygen ion peaks and the $Z = 1 - 3$ peaks of Ce and Lu for $E/Z = 150$ eV emitted by the two-elemental plasma are well

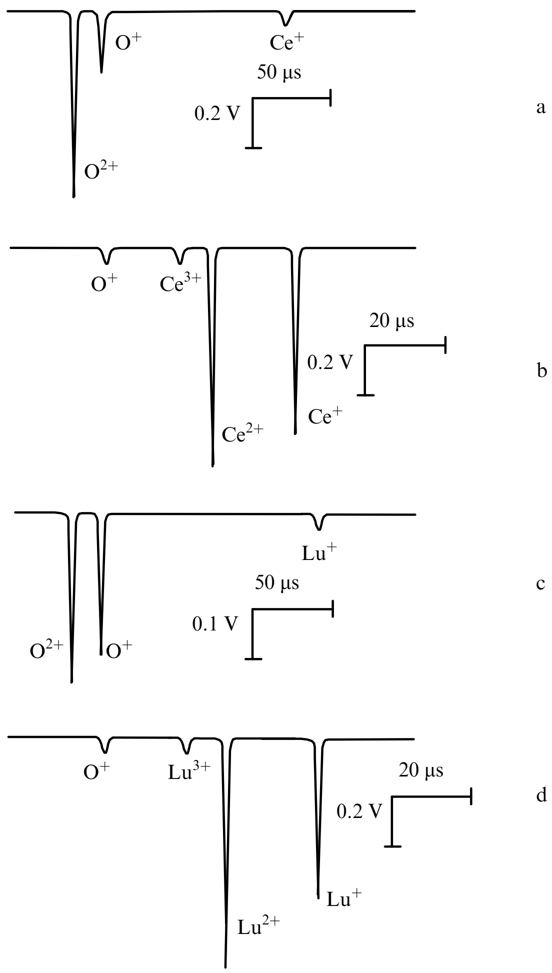

Figure 2. Mass–charge ion spectra of two-elemental plasmas of Ce_2O_3 (a, b) and Lu_2O_3 (c, d) produced by laser radiation with $q =$ 100 GW cm^{-2}; E/Z = 25 (a, c) and 150 eV (b, d).

pronounced and well resolved. Note that the ion beam intensity, the charge multiplicity, and the energies of the ions generated in the two-elemental plasmas increase as q is increased from 0.1 to 100 GW cm^{-2}.

We determined the most optimal energy interval (25 – 50 eV) in which oxygen ions of maximum multiplicity ($Z_{\max}$ = 2 and the ions of heavy elements (Sc, Ce, Lu) with a multiplicity $Z = 1$ can be simultaneously detected. For relatively high ion energies (over 150 eV), the peaks of oxygen ions with multiplicities $Z \geqslant 2$ vanish, while the $Z = 1 - 3$ peaks of the heavy elements (Sc, Ce, Lu) are clearly observed.

Fig. 3 shows typical energy spectra of O^+ and O^{2+} ions accelerated in the two-elemental plasmas produced from Sc_2O_3, Ce_2O_3, and Lu_2O_3. One can see that the type and width of the energy spectra of the O^+ and O^{2+} ions and their $E_{\max}$ depend on the nature of a two-elemental target. For all oxygen-containing targets under study, the largest width of the energy spectrum of the oxygen ions is observed for $q = 100$ GW cm^{-2}. One can see from Fig. 3, that the width of the energy spectrum of the $Z = 1$ oxygen ions is largest for a plasma produced of Ce_2O_3 and is smallest for a plasma of Sc_2O_3.

The maximum energy $E_{\max}$ of singly charged ions O^+ in a two-elemental plasma depends on the mass of ions of the second plasma component (Sc, Ce, Lu). For instance, in the case of a Ce_2O_3 target, $E_{\max}$ of the O^+ oxygen ions is two

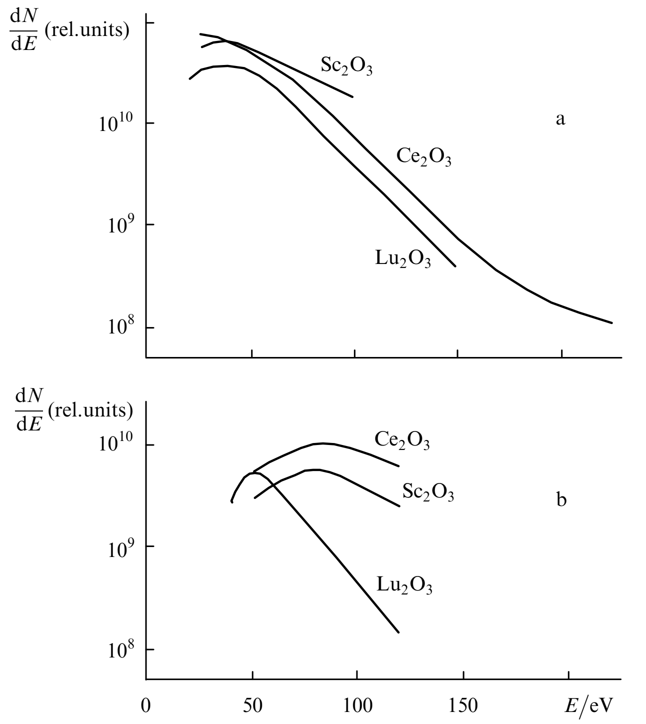

Figure 3. Energy spectra of the O^+ (a) and O^{2+} (b) ions accelerated in two-elemental plasmas of Sc_2O_3, Ce_2O_3, and Lu_2O_3 recorded for $q = 100$ GW cm^{-2}.

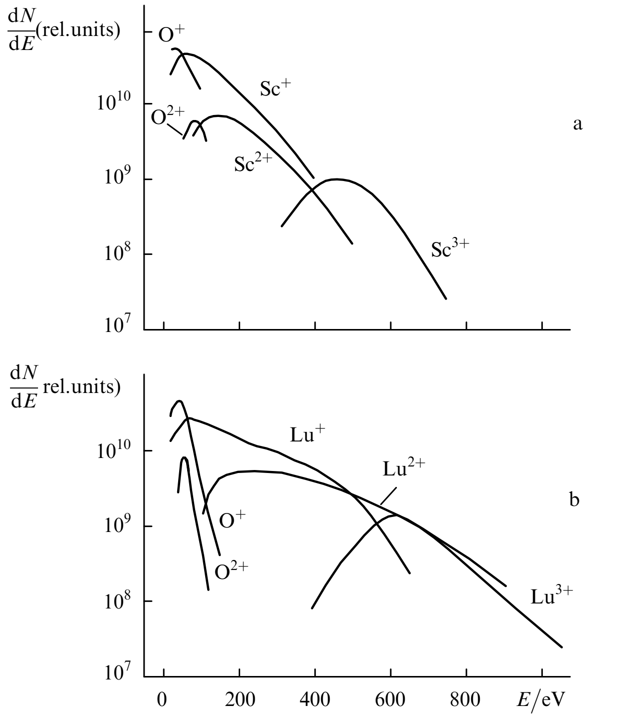

Figure 4. Energy spectra of O and Sc (Lu) ions accelerated in two-elemental plasmas of Sc_2O_3 (a) and Lu_2O_3 (b) obtained for $q = 100$ GW cm^{-2}.

times E_{max} of these ions produced from Sc_2O_3 for the same laser intensity. A comprehensive analysis of the energy spectra of the O^{2+} ions obtained with targets of different nature revealed that the distribution has one maximum and the energy range of E_{max} is independent of dN/dE, but depends on the nature of the second component (Sc, Ce, Lu) of a two-elemental target (see Fig. 3b). For the O^{2+} ions, the dependence of dN/dE on the target nature manifests itself most clearly in the 60 – 120 eV energy range, where recombination losses are enhanced. These losses are insignificant in the 40 – 60 eV range dominated by ionisation processes.

Fig. 4 shows typical energy spectra of the O^+ and O^{2+} oxygen ions generated in two-elemental plasmas of Sc_2O_3 and Lu_2O_3 for $q = 100$ GW cm^{-2}. One can see that the spectra are grouped in two ion 'packets' located in different energy ranges. Oxygen ions with multiplicities $Z = 1, 2$ are located in the low-energy range and the Sc (Lu) ions in the high-energy range. We note that the passage from Sc_2O_3 to Lu_2O_3 is attended with clearly defined changes of the O^+ and O^{2+} oxygen ion spectra and a shift of the Lu^+ and Lu^{2+} ion energy spectra (relative to the Sc^+ and Sc^{2+} ion spectra) towards higher energies. The width of the energy spectra of both O and Sc (Ce, Lu) ions is largest for $q = 100$ GW cm^{-2} for all oxygen-bearing two-elemental targets under investigation. Reducing the intensity of laser irradiation of the surface of an oxygen-bearing target results in a narrowing of the energy distribution of the O and Sc (Ce, Lu) ions accelerated in the two-elemental plasmas.

A comparison of the mass and energy spectra of O and Sc (Ce, Lu) ions generated in a two-elemental laser plasma shows that the production and acceleration of both O^+, O^{2+} and $Sc^+ - Sc^{3+}$ ($Ce^+ - Ce^{3+}, Lu^+ - Lu^{3+}$) ions is determined by the elemental target composition. We note that the targets of Sc_2O_3, Ce_2O_3, and Lu_2O_3 employed in experiments and their elements Sc, Ce, and Lu are rather close to each other in several thermal parameters (such as work function, electrical conduction, melting heat, heat capacity, ionisation potentials of Sc, Ce, and Lu up to the fourth multiplicity). However, these two-elemental targets differ strongly in the mass of the second component (the masses of Sc, Ce, and Lu atoms exceed the mass of oxygen atoms by factors of 3, 9, and 11, respectively. This factor plays a significant part in the formation of a two-elemental plasma and, hence, in the energy exchange between O and Sc (Ce, Lu) atoms. Referring to Fig. 4, the O^+ and O^{2+} ion spectra overlap with those of the $Sc^+ -$ $Sc^{3+}, Ce^+ - Ce^{3+}$, and $Lu^+ -$ Lu^{3+} ions in a narrow energy range. It is characteristic that the increase in mass of the second component of a two-elemental plasma results in a rise of the energy exchange between the ions of O and Sc (Ce, Lu). Eventually the ion energy spectra expand towards higher energies as the mass of the second target component (Sc, Ce, Lu) increases. We note that E_{max} for the $Z = 1 - 4$ ions increases with increase in mass of the second component atoms (Sc, Ce, Lu) of a two-elemental plasma (Fig. 5).

Since the ions of a two-elemental plasma exhibit a broad energy spectrum with a single maximum, the processes occurring in a two-elemental plasma may be thought of as occurring in two stages. At the first stage (prior to the distribution peak), an intense ionisation is observed, which is accompanied with a rise in the production probability of multiply charged ions of both O and Sc (Ce, Lu) contained in the target. The second stage (after the distribution peak) is characterised by a rise in recombination rate, i.e., in energy exchange between light (O) and heavy (Sc, Ce, Lu) ions of a two-elemental plasma. This is in qualitative agreement with theoretical calculations of the expansion of a two-elemental plasma in vacuum [10].

The experiments showed that the formation of charge and energy spectra of multiply charged O and Sc (Ce, Lu) in a two-elemental plasma is determined not only by ionisation, recombination, ionisation state freezing, and ion acceleration, but also by the mutual influence of the mass- and charge-different ions present in the plasma. In this case, it is difficult tell which of the ions plays the part of greatest significance in this process. According to Ref. [11], the energy E_k of the ions of sort k in a collisionless multi-elemental plasma is determined the similarity law $E_k \sim Z_k^2/M_k$, where Z_k and M_k are the ion charge and mass.

As shown by the authors of Ref. [10], in the 'collisional' limit the velocities of all components become equal, the charge dependence of the ion energy vanishes, and there arises a proportionality between mass and energy, i.e., $E_k \sim M_k$. Therefore, we may say that our experimental data are in qualitative agreement with the theory [10, 11]: the peak multiplicity of oxygen ions (O^{2+}) is independent of the elemental composition of a two-elemental target, the energy spectrum of the O^+ and O^{2+} ions changes, and the energy spectrum of Sc, Ce, and Lu ions with multiplicities $Z = 1-3$ expands towards higher energies with increase in mass of the second component (Sc, Ce, Lu) of the two-elemental plasma. In addition, the ion E_{max} increases with a rise in Z and the mass of ions of the heavy plasma component.

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