

# ELECTRON-IMPACT DOUBLE IONIZATION OF TUNGSTEN ATOMS AND IONS AT LOW IONIZATION STAGES

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Received 20 October 2009; revised 11 December 2009; accepted 18 December 2009

The double ionization of neutral tungsten atoms and  $W^{2+}$ ,  $W^{4+}$ , and  $W^{6+}$  ions by electron impact is investigated. The calculations are performed taking into account the Auger decay of single ionization states with inner vacancies in the 4d, 4f, 5s, 5p, and 5d shells as well as the direct double ionization using sudden perturbation model. The direct single ionization cross-section is obtained in relativistic distorted wave or binary-encounter-dipole approximations; the contribution from the indirect ionization by excitation-autoionization process is included too. It is shown that both processes – Auger decay of inner vacancy states and direct double ionization – play an important role in the double ionization process. The calculated cross-sections are compared with the experimental data.

**Keywords:** tungsten, electron-impact double ionization, Auger transitions, sudden perturbation

**PACS:** 34.50.Fa, 52.20.Fs, 31.15.A-

## 1. Introduction

Due to a very low sputtering rate of tungsten it is used as the first wall material in tokamaks, especially in the divertor region. However, some its ions migrate to the central region of discharge and may be a reason of large radiation losses. Thus the modelling of the production of tungsten ions is an urgent task. Much theoretical and experimental work has been done in investigating the electron impact single-ionization of tungsten ions [1–6]. In the case of neutral atoms the experimental data are absent, only the calculation results are available [2, 6]. The double ionization of tungsten ions at low ionization stages was measured and described by the semi-empirical formula [7]; no *ab initio* calculations till now have been made.

There are two possible processes giving contribution to the multiple ionization: Auger transitions following the deexcitation of the initial state with a vacancy in an inner shell as well as the direct multiple ionization. The importance of the Auger and radiative cascades in plasma modelling was stressed in [8, 9]. It was pointed out [8] that the essential part of the level population in some highly charged ions originated from inner shell ionization; this process was not yet considered in the database ADAS. Thus it is up-and-coming to investi-

gate the role of the Auger decay of the states with a vacancy and the direct double ionization in the production of tungsten ions during their interactions with electrons.

The aim of this work is the investigation of double ionization of tungsten atoms and ions at the low ionization stages, namely, for W,  $W^{2+}$ , and  $W^{4+}$  in the region of electron energies from the process threshold up to 1000 eV and for  $W^{6+}$  up to 5000 eV.

## 2. Results of calculation and their discussion

We will consider the double ionization of ions as a two-step process: the initial single ionization by electron impact and the following autoionization or additional ionization due to a sudden perturbation of electronic shells.

Calculations of energy level spectra, cross-sections of single ionization and excitation by electron impact as well as of Auger transitions rates have been performed using the Flexible Atomic Code [10]. In this complex of programs the relativistic Dirac–Fock–Slater wave functions are used. The mixing of all relativistic configurations corresponding to the same nonrelativistic configuration is taken into account. The results of calculations with this code of single and double ionization thresholds for the considered ions correspond with the

average accuracy of 0.13 eV to their values obtained by multiconfiguration Dirac–Fock code [7], but differ by 1–2 eV from experimental and empirical values compiled in [11].

At first we will consider the single ionization of ions, because in the following its cross-section will be used for the calculation of double ionization of ions. For  $W^{4+}$  and  $W^{6+}$  the direct single electron impact ionization is calculated in the distorted wave approximation (DWA). For near neutrals the DWA tends to overestimate the experiment [5] and more accurate results are given by the binary-encounter approximation [6]. Thus in the case of W and  $W^{2+}$  the binary-encounter-dipole (BED) model [12] is used. Also the indirect ionization process of excitation-autoionization is included; it gives for such tungsten ions a significant contribution to the single ionization cross-section [4, 5, 7]. It is supposed that W and  $W^{2+}$ ,  $W^{4+}$  ions are in the initial ground state. In the case of  $W^{6+}$  ion we accept the assumption made in [4, 7] that most of the ions are ionized from the metastable states of the excited  $5p^6 4f^{13} 5d$  configuration. The lowest level of this configuration is  $^3D_2$ , however, it can be deexcited to  $5p^6 4f^{14}$  configuration by the electric quadrupole transitions. Thus we consider the ionization from the second long-living level  $^3D_5$ . At the energies of electron beam up to about thousand electronvolts used in the experiment [7] for  $W^{2+}$ ,  $W^{4+}$  ions, the single ionization of 4d, 4f, 5s, 5p, and 5d shells is necessary to take into account [5]. In the case of  $W^{6+}$  the cross-sections of single and double ionization were measured up to 5000 eV; then the ionization of deeper shells becomes possible, however, their contribution to both cross-sections is small.

In W and  $W^{2+}$  the largest contribution to the direct single electron impact ionization cross-section is given by the ionization of 5d shell. In W the ionization of 6s shell also plays an important role (Figs. 1, 2). For  $W^{4+}$  and  $W^{6+}$  the role of 4f ionization increases due to a contraction of 4f orbital and decrease of 5d electron number in this shell (Figs. 3, 4).

In calculations of the indirect single ionization process the largest contributions are given by the following excitations: 6s–6p, 5d–6p, and 5p–5d for W; 5d–6p, 5p–5d, 6s for  $W^{2+}$ ; 5p–5d and 5d–6p, 5f for  $W^{4+}$ , and 5d–5f, 5p–5d, 4f–5d for  $W^{6+}$ . The excitation-ionization part of single ionization cross-section augments its value given by the direct ionization. The contribution of two-step process to the cross-section peak value increases with the charge of ion: 14.7% for W, 32.1% for  $W^{2+}$ , 58.8% for  $W^{4+}$ , and 59.5% for  $W^{6+}$ .

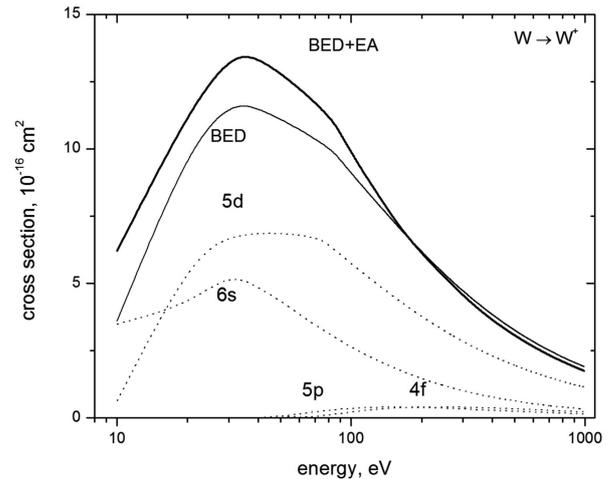


Fig. 1. Electron-impact single ionization cross-section of neutral W atoms. Results of calculation: dotted curves, contributions from separate shells to the direct ionization; full curve, total direct ionization; solid line, single ionization including direct ionization and excitation-autoionization. Direct ionization is calculated in BED approximation.

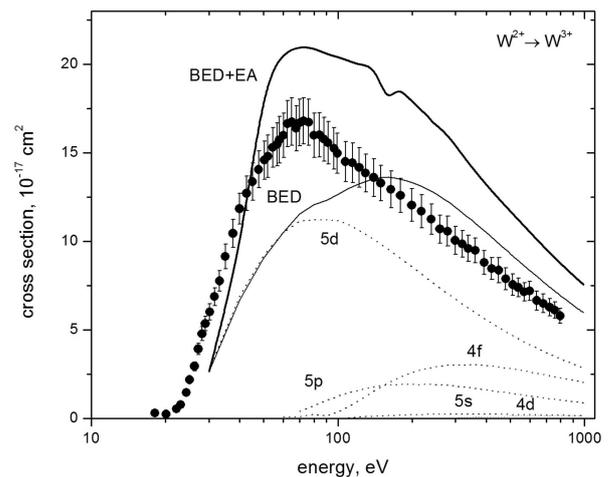


Fig. 2. Electron-impact single ionization cross-section of  $W^{2+}$  ions. Experiment [3], points with error bars. Results of calculation: dotted curves, contributions from separate shells to the direct ionization; full curve, total direct ionization; solid line, single ionization including direct ionization and excitation-autoionization. Direct ionization is calculated in BED approximation.

The obtained total single ionization cross-section  $\sigma^+(\varepsilon)$  for  $W^{4+}$  and  $W^{6+}$  corresponds well to the experimental data. For  $W^{2+}$  the cross-section  $\sigma^+(\varepsilon)$  is obtained about 1.2 times higher at its peak than the experimental value [7] (Fig. 2).

According to the diagram of energy level spectra (Fig. 5) the additional single and even double ionization of tungsten ions is possible by Auger transitions from the states with the inner vacancy in the  $4l^{-1}$  and  $5l^{-1}$  shells. Our results of the double ionization limits for  $W^{2+}$  (61.3 eV) and  $W^{4+}$  (114.0 eV) practically

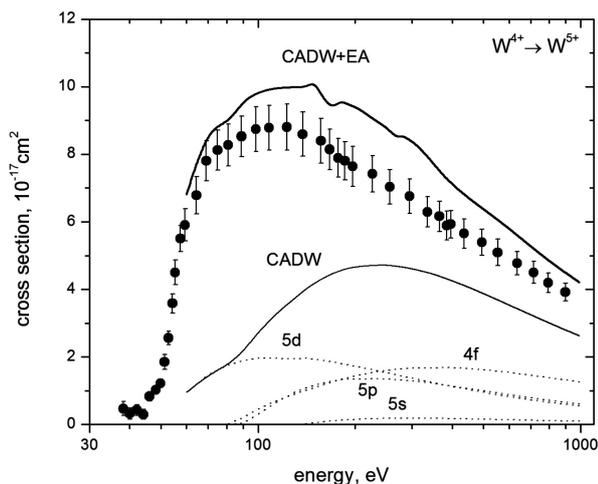


Fig. 3. Electron-impact single ionization cross-section of  $W^{4+}$  ions. Experiment [3], points with error bars. Results of calculation in DWA approximation: dotted curves, contributions from separate shells to the direct ionization; full curve, total direct ionization; solid line, single ionization including direct ionization and excitation-autoionization.

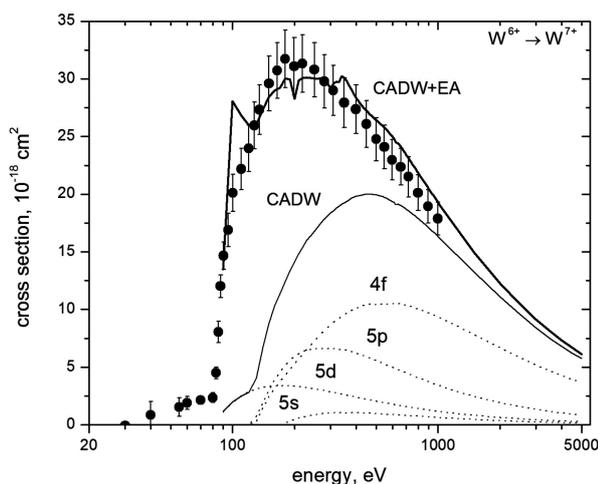


Fig. 4. Electron-impact single ionization cross-section of  $W^{6+}$  ions. Experiment [3], points with error bars. Results of calculation in DWA approximation: dotted curves, contributions from separate shells to the direct ionization; full curve, total direct ionization; solid line, single ionization including direct ionization and excitation-autoionization. It is supposed that most of the ions are ionized from the metastable states of the excited  $5p^6 4f^{13} 5d$  configuration.

coincide with the values 61.4 eV and 114 eV calculated by multiconfiguration Dirac–Fock method [7].

Usually the method of global characteristics of spectra [13] is used for the calculation of ion yields produced by Auger cascade between complex configurations. However, the energy spectra of initial and final configurations of considered tungsten ions with vacancies in outermost shells are not distant energetically or they even overlap. Thus it is necessary to perform more

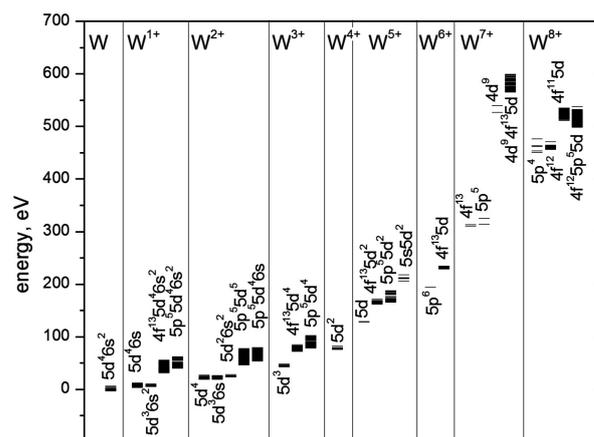


Fig. 5. Energy level spectra of ions involved in Auger transitions after the ionization of initial ion. The positions of levels are given with respect to the ground level of neutral atom W.

complex level-by-level calculations. The wave functions of Auger electrons as well as of bound states have been calculated in the central potential of autoionizing ion (before autoionization) using its lowest lying relativistic configuration. According to the obtained results the most probable Auger transitions end in the configurations of the next ionization stage.

The probability of double ionization by Auger transitions in the tungsten atom and first ions mainly depends on the energetic position of  $4f^{-1}$  and  $5p^{-1}$  vacancies' states with respect to the autoionization limit. In  $W^+$  the transitions from all such states to the states of  $W^{2+}$  are energetically permitted. In  $W^{3+}$  only the Auger decay from some levels of  $5p^5 5d^4$  configuration is possible in the single configuration approximation, thus the  $W^{4+}$  ions are mainly obtained by the decay of  $5s^{-1}$  and  $4d^{-1}$  vacancy states. However, after taking into account a rather strong interaction between  $4f^{-1}$  and  $5p^{-1}$  configurations one highly populated level of  $5p^5 5d^4$  configuration is shifted through autoionization threshold, leading to the essential increase of Auger transition contribution to the double ionization cross-section of  $W^{2+}$  ion. In  $W^{5+}$  already the Auger transitions from all levels of  $4f^{-1} 5d^2$  as well as  $5p^{-1} 5d^2$  configurations are forbidden in single configuration approximation. By taking into account the interaction of these configurations as well as of  $W^{6+} 5p^6$  with 5–7 neighbouring configurations, the indicated level is shifted to the autoionization threshold, but remains 3 eV below it. However, it is probable that including more correlation effects this level can give a contribution to the double ionization cross-section. In  $W^{7+}$  the levels of  $4f^{-1}$  and  $5p^{-1}$  configurations are lying essentially below the autoionization threshold and the main contribution to the production of

$W^{8+}$  ions is given by the Auger decay of  $4d^{-1}$  vacancy states.

The additional electron can also be removed from an ion or excited as a result of its strong perturbation at the creation of initial vacancy. We have approximately estimated the contribution of these shake-off and shake-up processes to the double ionization in the average configuration approximation. The probability for all electrons to remain in  $nl^N$  shell at the production of  $n_0l_0^{-1}$  vacancy is expressed as follows:

$$A_0(n_0l_0^{-1} nl^N) = \langle nl_K | nl_{K_1} \rangle^{2N}, \quad (1)$$

where the subscript indicates the configuration, the wave functions of which are used for the calculations of overlap integral:  $K$  is the initial configuration without vacancies and  $K_1 \equiv K n_0l_0^{-1}$ . Thus the probability that one electron will be removed from  $nl^N$  shell and all its other electrons will remain unperturbed at the single ionization of  $n_0l_0^{N_0}$  shell is expressed as

$$A(n_0l_0^{-1} nl^{-1}) = N [1 - \langle nl_K | nl_{K_1} \rangle^2] \times \langle nl_K | nl_{K_1} \rangle^{2(N-1)}. \quad (2)$$

Here the term excluding the excitations into the occupied states is omitted, because it is practically negligible at the production of vacancies in the outer shells. Probability  $A(n_0l_0^{-1} nl^{-1})$  has been calculated using quasirelativistic wave functions [14]. While most of the states with a vacancy and an excited electron autoionize into a state of the next ion, Eq. (2) gives the estimation of shake process probability to the double ionization of ion. Then the cross-section of double ionization due to shake process after electron impact ionization of  $n_0l_0^{N_0}$  shell gets:

$$\sigma^{++}(\varepsilon) = \sum_{n_0l_0} \sigma_{n_0l_0}^+(\varepsilon) \sum_{nl} A(n_0l_0^{-1} nl^{-1}), \quad (3)$$

where  $\varepsilon$  is the energy of incident electron. Only energetically possible shake transitions must be included. The probability  $\sum_{nl} A(n_0l_0^{-1} nl^{-1})$  obtains its largest value, exceeding 15%, for  $4f^{-1}$  vacancy in neutral atoms (Fig. 6). The sudden perturbation of outer shells is weaker at the production of  $5l^{-1}$  vacancies. The probability to remove an additional electron diminishes on increasing the ionization degree.

In Figs. 7–10 the results of calculation of electron-impact double ionization cross-sections for the considered ions are presented. The total cross-section of double ionization is obtained smaller than the experimental data [7]: the peak value is underestimated by 25% for

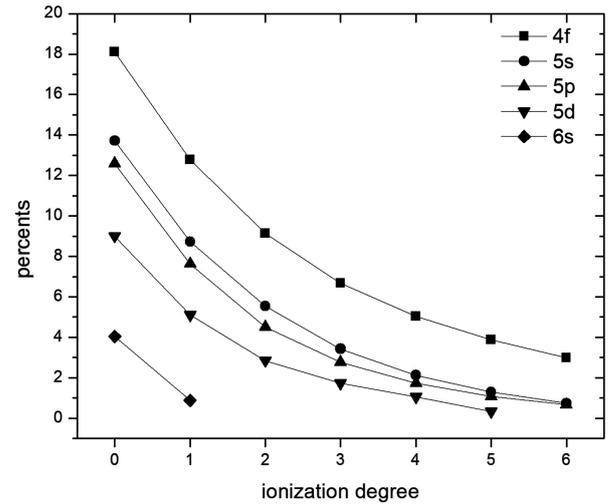


Fig. 6. The values of the probability  $\sum_{nl} A(n_0l_0^{-1} nl^{-1})$  to remove one electron from any other shell of ion at the production of  $n_0l_0^{-1}$  vacancy. Results of calculation according to Eq. (2) in quasirelativistic approximation.

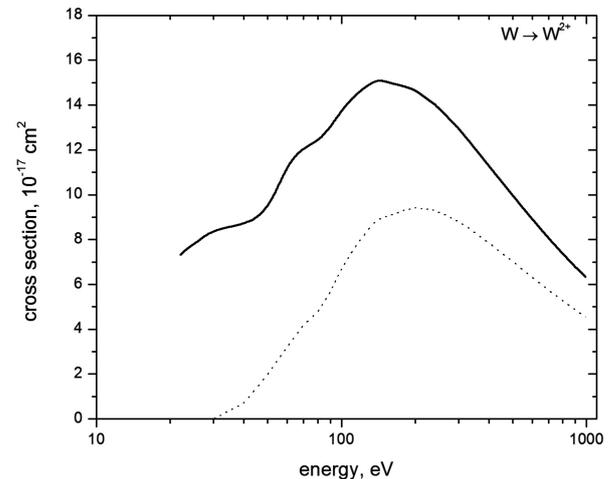


Fig. 7. Electron-impact double ionization cross-section of neutral W atoms. Results of calculation: dotted curve, single ionization following the Auger transitions; full curve, total double ionization.

$W^{2+}$ , 58% for  $W^{4+}$ , and 33% for  $W^{6+}$ . The contributions from the Auger transitions and from the sudden perturbation are of the same order, though the last process plays a smaller role, especially for the higher ions. For W,  $W^{2+}$ , and  $W^{4+}$  the double ionization due to sudden perturbation is dominated by the initial ionization of 4f shell, however, at the ionization of metastable states of  $W^{6+}$  the influence of the production of  $5d^{-1}$  vacancy becomes also essential.

### 3. Conclusions

The large scale calculations have been performed in order to obtain the theoretical double ionization cross-

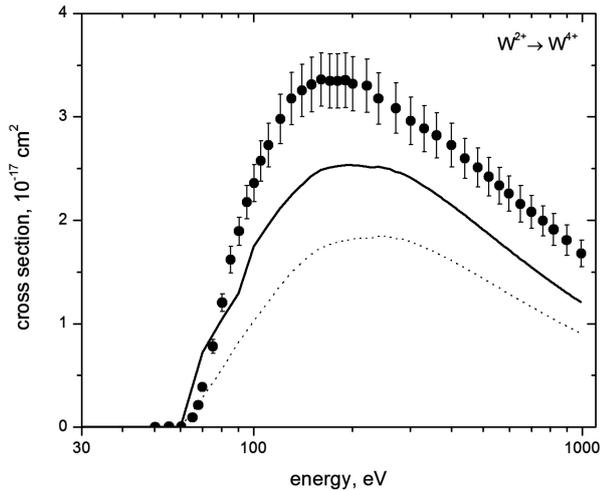


Fig. 8. Electron-impact double ionization cross-section of  $W^{2+}$  ions. Experiment [7], points with error bars. The meaning of other points and curves is the same as in Fig. 7.

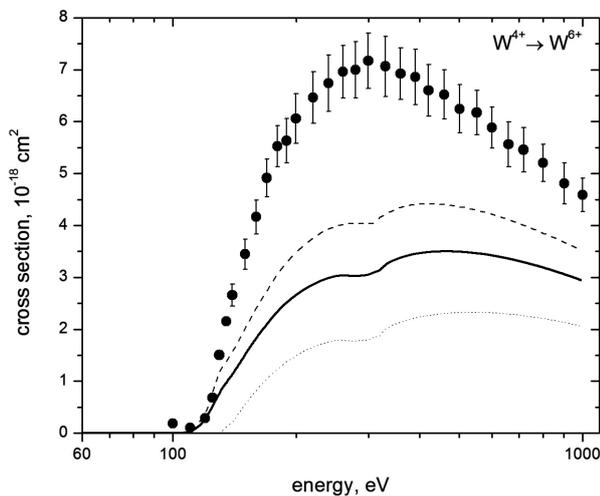


Fig. 9. Electron-impact double ionization cross-section of  $W^{4+}$  ions. Experiment [7], points with error bars. The meaning of other points and curves is the same as in Fig. 7. Dashed curve, the total double ionization after the energy of  $5p^6$  level for  $W^{6+}$  ion is lowered by 3 eV.

sections for the tungsten atoms and for  $W^{2+}$ ,  $W^{4+}$ , and  $W^{6+}$  ions; till now only the empirical results were known for the considered ions and data absent at all for the neutral atoms. Two possible mechanisms of double ionization by electron impact have been investigated. After the single ionization of 4d, 4f, 5s, and 5p shells some their states can decay by Auger transitions into the states of ions with one and even two higher ionization stages. Because such transitions take place between neighbouring or overlapping configurations the detailed level-by-level calculations have been performed. The direct double ionization of ions has been considered as a two-step process: the initial single ionization by

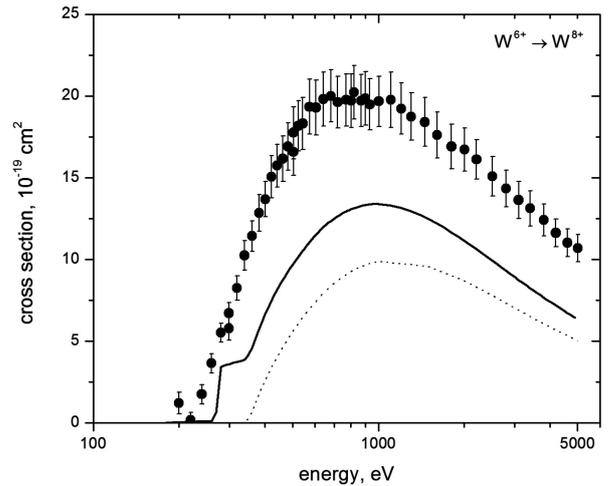


Fig. 10. Electron-impact double ionization cross-section of  $W^{6+}$  ions. Experiment [7], points with error bars. The meaning of other points and curves is the same as in Fig. 7.

electron impact and the additional ionization due to a sudden perturbation of electronic shells. The contributions from the Auger transitions and from the sudden perturbation to the double ionization cross-section are obtained to be of the same order, though the last process plays a smaller role, especially for the higher ions.

### Acknowledgements

We gratefully acknowledge Prof. E. Salzborn for the permission to reproduce experimental data of single and double ionization cross-section for the tungsten ions. Our work was partly supported by the Joint Taiwan–Baltic Research project and the Ministry of Education and Science of Lithuania, agreement No. SUT-683. The calculations were partly funded by the European Commission project RI026715 BalticGrid, and also by the Lithuanian State Science and Studies Foundation in the frame of LitGrid and GridTechno.

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## DVIKARTINĖ VOLFRAMO ATOMŲ IR PIRMŪJŲ JONŲ JONIZACIJA ELEKTRONŲ SMŪGIU

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### Santrauka

Volframas, kaip labai atspari danga, naudojamas naujos kartos tokamako įrenginiuose, numatomas naudoti ir projektuojamame ITER tokamake. Tačiau net nedideli jo kiekiai, patekę nuo įrenginio sienelių į plazmą, sukelia nemažus radiacinius nuostolius, todėl yra svarbu ištirti įvairaus kartotinumų volframo jonų susidarymo galimybes. Iki šiol, sprendžiant balanso lygtis, nebūdavo atsižvelgiama į dvikartinę volframo atomų jonizaciją, jiems sąveikaujant su plazmos elektronais. Pateikiami pirmieji tokio proceso šiam elementui skerspjūvių teorinių skaičiavimų rezultatai.

Tiesioginės vienkartinės jonizacijos skerspjūvis buvo skaičiuotas reliatyvistiniu sutrikdytųjų bangų ir dvinario susidūrimo metodais, taip pat buvo atsižvelgta į dvipakopės jonizacijos, sukuriant sužadintus lygmenis, o po to vykstant jų autojonizacijai, indėlių. Po

4d, 4f, 5s ir 5p sluoksnių vienkartinės jonizacijos galimi Auger šuoliai, kurių metu atomo jonizacijos laipsnis padidėja vienetu ar dviem. Kadangi tokie šuoliai vyksta tarp energetiškai gretimų ar persiklojančių konfigūracijų, reikia atlikti detalius šuolių tarp lygmenų skaičiavimus; jie buvo vykdomi reliatyvistiniu konfigūracijų maišymosi artutiniu. Tiesioginė dvikartinė jonizacija buvo nagrinėjama kaip dvipakopis procesas: vienkartinė jonizacija elektronų smūgiu ir papildoma jonizacija dėl elektronų staigaus trikdžio. Auger šuolių ir staigios perturbacijos indėliai į dvigubos jonizacijos skerspjūvį gaunami panašios eilės, tačiau antrasis procesas vaidina mažesnę vaidmenį, ypač didesnio jonizacijos laipsnio jonuose. Ap-skaičiuoti skerspjūviai kokybiškai atitinka eksperimentinius duomenis.