

## ELECTRON IMPACT IONISATION OF MULTIPLY CHARGED IONS

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### RESUME

La formation d'ions dans les plasmas est presque totalement dominée par les processus d'ionisation par impact d'électrons. Les observations ont montré la présence d'ions fortement chargés dans les plasmas naturels (observations du soleil) ou dans les études liées aux applications de la fusion thermonucléaire.

Au cours des dernières années, l'amélioration des techniques expérimentales et théoriques a permis d'obtenir des résultats fiables utiles à la compréhension des mécanismes d'ionisation simple ou multiple pour un grand nombre d'espèces ioniques.

Dans ce rapport, nous présentons une discussion de l'importance des différents processus d'ionisation dans le cas de deux séries isoélectroniques, celle de l'hélium et celle du lithium. Dans la première, la contribution des ions formés dans des états métastables est aussi prise en considération.

Quelques développements récents de l'étude des processus d'ionisation double sont aussi présentés.

### ABSTRACT

The ion formation in plasmas is totally dominated by the electron impact ionisation of atoms and ions. This holds for natural plasmas and for artificial ones like thermonuclear fusion plasmas. Since a long time, multiply charged ions have been observed in the various hot plasmas, so that the investigation of the mechanisms of production of these ions have been given a decisive effort in the last few years : reliable theoretical and experimental methods have produced precise results for a wide spectrum of ions.

In this report the importance of various single ionisation processes is discussed for the heliumlike sequence and for the lithiumlike sequence. The contribution of ions formed in metastable states is also taken into account.

Some recent developments of double ionisation studies are also presented.

The electron impact ionisation of multiply charged ions has received a great attention in the last few years because of its large importance in plasma physics. Both astrophysical studies and thermonuclear fusion research have shown a large influence of impurity ions on plasma equilibrium. Collisional processes involving these impurities are responsible for large energy losses which prevent the plasma to reach the expected ion temperature.

The main mechanism of ion production is the electron impact ionisation process that has been extensively studied.

Various experimental approaches have been used for these studies. One is based on a quantitative analysis of the time evolution of plasma emitted radiation. This method gives information on ionization and excitation rates and requires additional measurements of electron temperature and density and the knowledge of the other processes taking place in the plasma. This makes the detailed information very difficult to obtain from those observations.

A second approach makes use of ion traps in which a well defined ion species can be stored in various configurations, electron beams, electric and magnetic fields. Some of those devices are also used as ion sources and the time evolution of the charge spectra can produce quantitative information on the ionization process.

The third approach involves two beams interacting in a restricted volume. In those experiments the particles are in a well defined state when they interact. The actual relative velocity is also well-known so that detailed information can be obtained.

This crossed electron ion beam method is able to produce cross-sections values with an uncertainty of a few percents only [1]. For these experiments the primary ion beam is extracted from an ECR ion source giving the opportunity to study ion charge state up to 20 for gaseous and solid atomic species including metals that have been seen to play a fundamental role in fusion plasmas.

Experimental studies often follow isoelectronic sequences in order to estimate the relative importance of indirect ionisation processes with increasing nuclear charge.

In contrast to the complicated many-electron systems, the single ionisation of simple ions has not received a great attention from the different groups involved in this subject in the last few years. Hydrogen-like ions have been completely neglected. The lowest charge states of the helium isoelectronic sequence only have been experimentally analysed, that is up to the case of nitrogen 5+. The first ion of the sequence, the singly charged lithium ion was already studied in details twenty years ago by Lineberger *et al.*, [2], Peart and Dolder, [3] and Peart *et al.*, [4]. Data for the heliumlike boron, carbon and nitrogen ions were presented by the Oak Ridge group in 1979 [5] and by our group for nitrogen [6].

Ionisation cross-section have also been derived by Donets and Ovsyannikov [7] from the observation of ion charge state spectra evolution in an Electron Beam Ion Source (EBIS). This method has produced partial data - that is, for electron impact energies larger than 2 keV - for the heliumlike carbon, nitrogen, oxygen, neon and argon ions.

The first crossed beam data have been recently obtained by our group [8] for  $O^{6+}$ . They are presented in Figure 1 together with the EBIS data [7] and also with the theoretical predictions in the DWBA [9].

The agreement with the EBIS data at 2250 eV and 2950 eV is surprisingly good: the two sets of data nearly coincide. That situation is slightly different from what has been frequently observed in other cases where the EBIS data are often larger than the crossed beam data [10]. A non-zero apparent cross-section

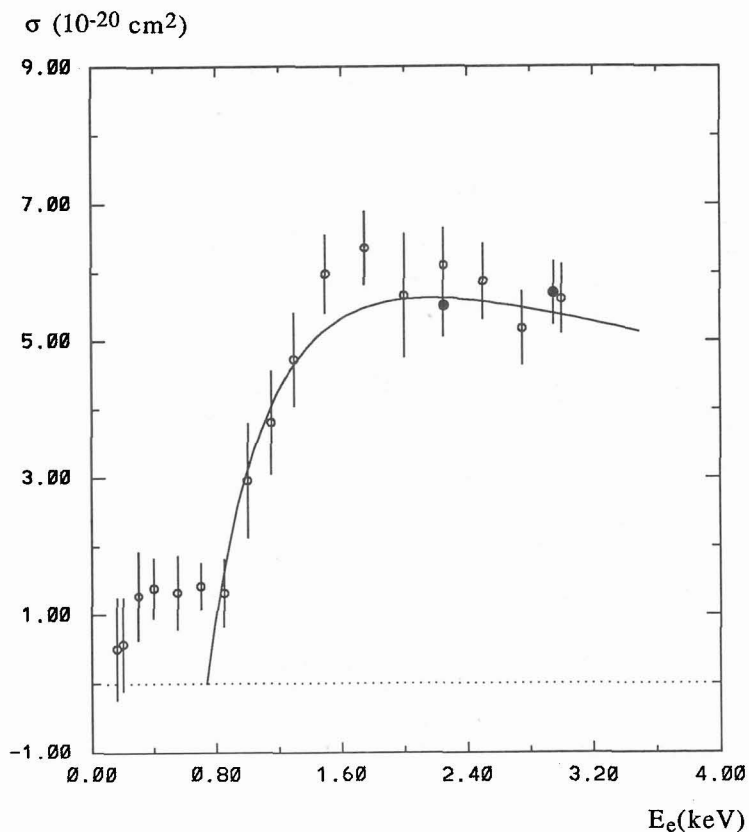


FIGURE 1

Absolute cross-section for electron impact ionisation of  $O^{6+}$ : (O) our measurements, (•) Donets and Ovsyiannikov [7], (---) DWBA calculation, Younger, [9].

is also observed below the threshold for the ground state ionisation (739.3 eV) and the zero signal is obtained below the ionisation threshold for the two existing metastable states, 178.2 eV and 170.5 eV for the  $2^3S$  and for the  $2^1S$  states, respectively.

The estimation of the metastable state contribution requires knowledge of their population and of their electron impact ionisation cross-section.

Although the life-time of these states is very long (according to the fact that it is not reduced by a dipole transition), it may be affected by transitions that have strong Z-dependence. As a consequence, the population in the collision region may differ slightly from the initial one.

Three particular decay modes play a role for these states : magnetic dipole emission, electric dipole and spin forbidden, two photon emission. The rates for

these processes are known from calculations that include relativistic effects. The data obtained by Lin *et al.* [11] in the relativistic random phase approximation have been used in order to characterise the actual metastable population in the beam.

From this reference the life-time is  $4.33 \times 10^{-7}$  sec for the  $2^1S$  state and  $8.43 \times 10^{-4}$  sec for the  $2^3S$  state. The ion time of flight from the source to the collision region is  $6.34 \mu$  sec, so that the singlet state population is totally eliminated while the attenuation of the triplet state is negligible.

The knowledge of collisional processes involving multiply charged ions in excited states is very fragmentary, but the particular case of heliumlike ions has already been subject to theoretical investigations allowing a detailed description.

The structure of these ions is similar to the lithium-like ion structure, so that the same ionisation processes take place :

- (i) the direct ejection of an electron from the K-shell and from the L-shell;
- (ii) excitation to doubly excited states followed by autoionisation.

The direct ejection of the electron belonging to the 2s shell is the principal ionisation process for heliumlike ions in a metastable state. The ejection of the K-shell electron should be of lower importance according to the large difference between the corresponding ionisation thresholds. The direct electron impact ionisation cross-section  $s_d$  has been calculated very recently in the Coulomb-Born approximation including exchange effects (CBX) by Attaourti *et al.* [12] for various members of the helium isoelectronic sequence. In addition, the contribution of the direct electron ejection from the K-shell may be estimated by the Lotz formula [13]

Indirect processes may also play a non-negligible role through the excitation of the ion from its initial singly excited metastable state to a doubly excited state belonging to the configurations  $(2l, 2l')$ . The collisions strengths for these processes have been calculated by Goett and Sampson [14] in the Coulomb-Born-Exchange Approximation.

The radiative decay of doubly excited states can also be taken into account by a method that was applied to the study of lithiumlike ions [15], that is, by introducing in the calculation theoretical values of Auger and radiative decay rates (Vainshtein and Safronova, [16]). For the particular oxygen case the cross-section reduction is of low importance. The branching ratio is 0.88.

The total ionisation cross section of  $O^{6+}$  in the  $2^3S$  metastable state is presented in Figure 2

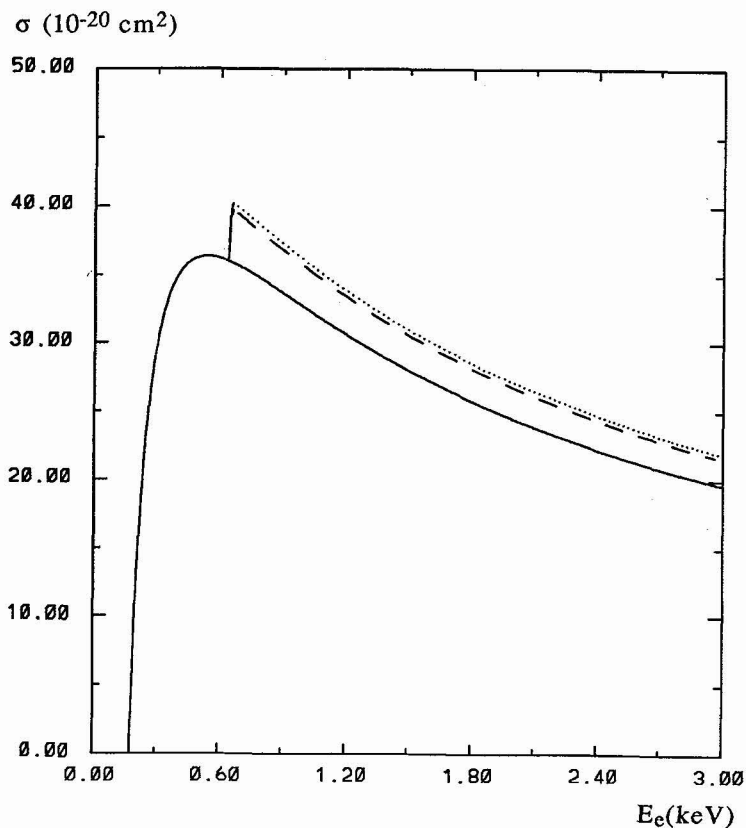


FIGURE 2

Theoretical estimation of electron impact ionisation cross-section of  $O^{6+}$  in the  $2^3S$  metastable state. (—) direct only, (.....) direct plus excitation-autoionisation, (- - -) direct plus excitation-autoionisation including the branching ratio for radiative decay.

The direct comparison of experimental and theoretical data is not possible, because the metastable population is not known. That population is estimated from the comparison of theoretical and experimental data taken in the electron energy range where only the metastable state contributes to the signal, that is below the ground state ionisation threshold. The population estimate is 3.9 %.

The lithiumlike sequence has been extensively studied experimentally and theoretically. In addition to the direct ejection process, this sequence offers the possibility of electron excitation from the ground state to the doubly excited states of the  $(1s, 2\ell, 2\ell')$  configurations. These indirect processes have been seen to contribute significantly to the ionisation signal. From astrophysical and fusion plasma observation, it is well known that radiative decay of autoionising doubly excited states competes with autoionisation for highly charged ions. This effect reduces slightly the excitation autoionisation cross section. For  $Ne^{7+}$ , that reduction has been estimated to some 30% and the branching ratio at the excitation threshold has been seen to have a  $Z^{-1}$  dependence [15].

Resonant processes have also been predicted to play a role in the ionisation mechanism. The first step of these processes is the resonant excitation, that is, the capture of the incident electron simultaneously with the electron excitation of the target ion. This process can only take place for an incident electron energy corresponding to the formation of a doubly excited state.

The second step is the spontaneous ejection of two electrons. This ejection can take place in two successive autoionization. It was first predicted by Lagatuta and Hahn<sup>[17]</sup> and is called resonant Excitation Double Autoionisation (REDA). The two electron ejection can also take place in a single step. This process was first proposed by Henry and Msezane<sup>[18]</sup> and is called Resonant Excitation Auto Double Ionisation (READI).

Three independant attempts have been made recently in order to analyse in details the relative importance of excitation autoionisation to states of the configurations ( $1s, 2\ell, 2\ell'$ ). In addition, attention was given to the possible presence of peaks corresponding to resonant processes.

In the first of those, the Lithium-like oxygen ion ( $O^{5+}$ ) was studied by Rinn *et al* <sup>[19]</sup>. The electron energy is scanned by fine energy steps ( $\approx 1$  eV) over two separate energy ranges. The first one (540-570 eV) includes the excitation-autoionisation thresholds and the second one (430-455 eV) includes two predicted READI processes ( $1s 2s^2 2p(^3P)$  and  $1s 2s 2p^2(^3D)$ ). The structure of cross-section at the excitation autoionisation threshold was well observed but contributions of READI processes are only suggested.

In the second attempt, indirect ionisation mechanisms for the  $N^{4+}$  ion were studied by our group <sup>[20]</sup>. For this purpose the classical crossed beam method was slightly adapted in order to eliminate systematic uncertainties during the electron energy scan.

The usual procedure has been modified in the following manner :

- (i) The electron energy is swept continuously from 380 eV to 450 eV by applying a symmetric linear ramp voltage to the cathode. The scanning period is 20 seconds.
- (ii) The total count rate, that is signal plus background, is recorded in a multichannel analyser as a function of electron energy. The spectrum is divided in two parts energy corresponding respectively to the increasing and the decreasing energy range. Each of the 255 channels corresponds to an energy band with of 0.7 eV.
- (iii) The recording was held continuously during a period of 55 hours. The electron beam sweeping frequency is 38 Hz, so that about 60.000 crossings between both beams occurred for each energy channel.
- (iv) During the energy scan, the electron beam focusing and sweeping voltages were not adjusted. Before the start of the experiment, it was checked that the beam profile was not drastically altered.

Finally, relative cross-sections are put onto an absolute scale by comparison with previous classical measurements <sup>[6]</sup>.

Results are presented in figure 3. Although error bars are still large the spectrum clearly shows structures at energies which correspond to expected excitation-autoionisation thresholds. In addition a large peak is superimposed to the smooth direct ionisation cross-section. This peak can only be attributed to some of the resonant excitation processes.

The third attempt has been made very recently by A. Müller and his collaborators at Giessen University<sup>[21]</sup>. In this case lithium-like ions are studied by scanning the electron energy over a range that includes the resonant excitation levels and the most fundamental excitation autoionisation thresholds. Relative uncertainties are reduced down to 0,15% so that contributions of individual doubly excited states were clearly observed.

The results show also the presence of REDA structure associated with these doubly excited levels. In addition, peaks corresponding to individual READI processes are undoubtedly observed. Details of this experiment are presented at this conference.

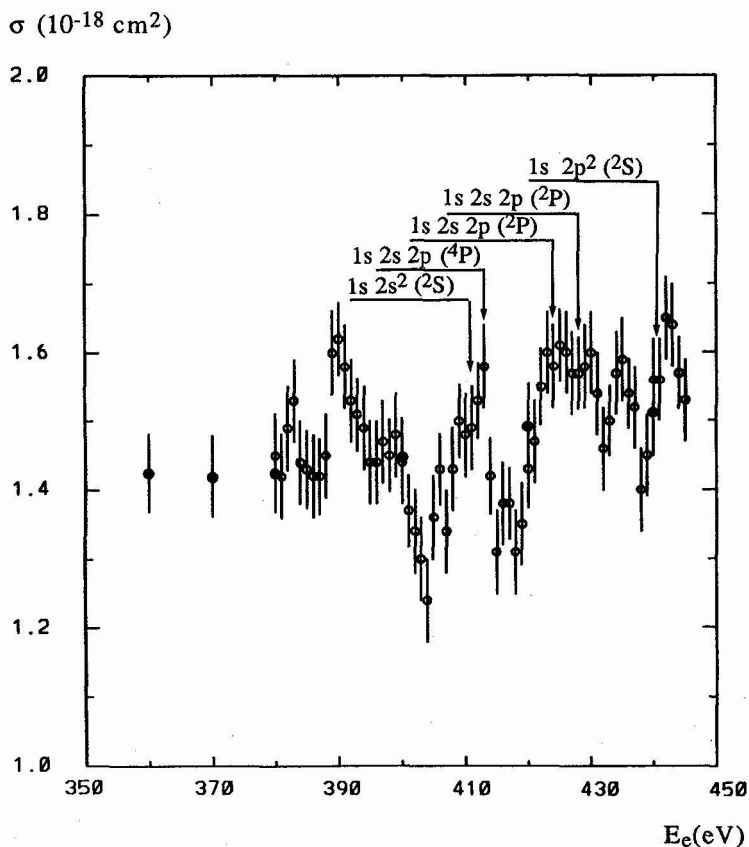


FIGURE 3

Absolute cross section for electron impact ionisation of  $N^{4+}$ . (o) this measurement, (o) values used for normalisation. The position excitation-autoionisation thresholds are also indicated.

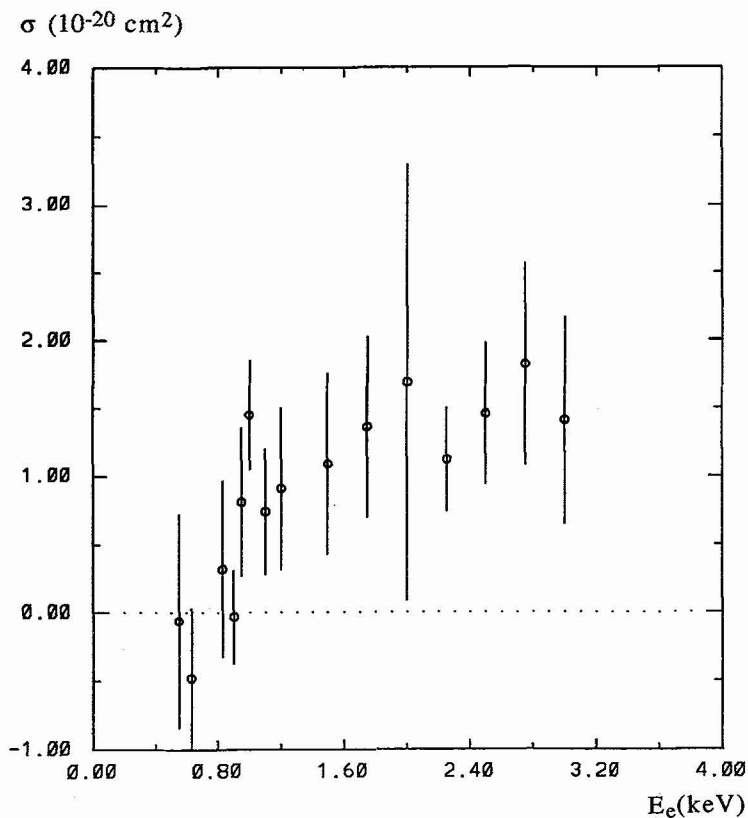
The multiple ionisation process is generally much less important than the single one, except for many electron systems.

The simplest process for multiple ionisation is the direct ejection of two electrons. This process is also very weak. The classical description of multiple ionisation takes into account the possibility of two successive "binary encounter" collisions inside the ion electronic cloud. In a recent experiment<sup>[22]</sup> the double

ionisation of  $\text{Ar}^{7+}$  - a sodiumlike ion - has been investigated over an energy range that excludes contributions of indirect processes.

From Clementi E. and Roetti C. tables<sup>[23]</sup>, the five existing thresholds for double ionisation are estimated. Their value depends on the shells from which electrons are ejected : 566 eV (3s and 2p), 641 eV (3s and 2s), 846 eV (2p and 2p), 921 eV (2s and 2p) and 996 eV (2s and 2s).

Results are presented in figure 4. The apparent threshold is located well above the lowest expected ionisation threshold. This indicates that the direct double ionisation takes place with a greater probability for electrons belonging to the same shell.



**FIGURE 4**  
Absolute cross section for double ionisation of  $\text{Ar}^{7+}$  by electron impact.

In addition, various indirect processes may play a role in the multiple ionisation.

The first one is the innershell ionisation where the ion is left in an excited state that lies above the next ionisation threshold. In this case, autoionisation may follow.

In the second indirect process, the primary ion is excited to a state that can eject two electrons in two separate single autoionisations or in auto double ionisation.

Finally, resonant capture may also be followed by three electron ejection, leading to resonance structure.

For the first time the importance of resonant in direct processes in double and triple ionisation indirect processes has been clearly demonstrated for heavy metal ions ( $\text{Cs}^+$ ,  $\text{Ba}^{2+}$  and  $\text{La}^3$ ) by A. Müller<sup>[24]</sup>.

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