Pathways to charge equilibration following multiple electron exchange between highly charged ions and atoms

de Nijs, Gerardus

Chapter 1

Introduction

The interest for the dynamics of collisions between highly charged ions and gaseous targets lies in the fields of plasma, astro- and fundamental physics. To further improve the understanding and subsequent control of magnetically confined fusion plasmas it is necessary to obtain knowledge about the dynamics of collisions including multicharged plasma impurities. The light we can observe from both astrophysical and laboratory plasmas results from various physical and chemical processes. Part of the light is emitted after slow collisions between highly charged ions and neutral atomic or molecular gas targets. With the advent of sources for highly charged ions around 1982 (Geller and Jacquot 1982), it became possible to investigate in detail the reactions between neutral particles and highly charged ions of species relevant for plasma research.

The possibility to address multicharged ion physics problems experimentally has led to significant improvements in the understanding of processes occurring in highly charged ion-atom collisions. Because of the higher charge state of multicharged ions electron transfer processes are especially important in collisions between multicharged ions and neutral atoms or molecules. After a good ten years of dedicated research, one-electron transfer from neutrals to multicharged ions is well understood. In contrast to this basic knowledge of processes in which more electrons are involved is still limited, although it is realized that multiple electron processes are important channels in highly charged ion collisions (Guillemot *et al* 1990).

This thesis is focussed on experimental studies of the dynamics of *multiple* electron capture by slow highly charged ions after collisions with rare gas targets. The collisions are studied by measuring and analyzing the spectra of the electrons emitted during the collision. The spectra are measured in coincidence with the charge state of the remaining target particles after the collision.

In general the studied collisions are of the form

$$\mathrm{A}^{q+} + \mathrm{B} \rightarrow (\mathrm{AB})^{q+} \rightarrow \mathrm{A}^{(q-t)+} + \mathrm{B}^{r+} + \Delta\mathrm{E} + (r-t)\ \mathrm{e}^- + \mathrm{h}\nu \qquad (1.1)$$

where A and B denote the projectile and target, respectively. During the collision

the q-fold charged projectile captures electrons. Both the projectile and the target can be left in an excited state, which decays by emitting photons or electrons. If long after the collision the projectile is $q-t$ fold charged and the target r fold, then $r-t$ electrons are ejected from the system. If the binding energy of the electrons in the target is different from the binding energy of these electrons, whenever they are captured into the projectile, then the energy difference ΔE between target and projectile binding energy can be transferred to the motion of the collision partners. A 'perfect' atomic physics experiment measures all these entities in coincidence, enabling a full identification of the collision $(AB)^{q+}$. It is, however, by no means trivial to do so. It is difficult to design an experimental setup in which, under all scattering angles, all the various reaction products can be measured in coincidence with each other.

The number of electrons which participate in a collision is a measure for the complexity of the collision system. In general a subset of the reaction products are measured. It is possible to measure the energy of the photons emitted by the projectile or target (see e.g. Hoekstra 1995), to measure the energy of the emitted electrons (Bordenave-Montesquieu *et al* 1984, Mack *et al* 1989) or to determine the energy gain of the projectile after the collision (Schmeissner *et al* 1984). Their experiments gave already much insight in the dynamics of electron capture by highly charged ions. A lot of these experiments were done using one- or two-electron targets (H, H_2 or He). One-electron capture processes are extensively studied, experimentally (see e.g. Folkerts *et al* 1994) as well as theoretically (Harel and Jouin 1987, Fritsch 1994, Fritsch and Lin 1991) and are fairly well understood.

The problem which is met with two-electron (and also with multi-electron) targets is that the measured energy gain, photons or electrons, can result from one- as well as from more-electron transfer processes. Double-electron capture processes are much more difficult to describe than single electron capture, but much work has already been done on the theoretical side (Kimura and Olson 1984, Harel and Jouin 1992) as well as for the experiments (Nakamura *et al* 1995, Posthumus *et al* 1992, Stolterfoht *et al* 1990, Cederquist 1991, Bordenave-Montesquieu *et al* 1994). Especially for the case of measurements on the projectile energy gain it is needed to distinguish between one- and two-electron processes. This can be done by determining the charge of the target in coincidence with the projectile energy gain (Barat *et al* 1987, Cederquist *et al* 1992). The energy gain and scattering angle, selected on target charge state, can provide better insight into charge exchange processes. Measurements of energy selected photons in coincidence with the target charge state (Martin *et al* 1993) as well as electron target ion coincidences (Posthumus and Morgenstern 1992, De Nijs *et al* 1994) provide complementary information on the collision dynamics.

Although the gross features of the electron spectra resulting from the collisions of N^{7+} and Ar can be explained by evaluating the total non-coincident electron spectra (Benoit-Cattin *et al* 1988), the experiments, described in chapter 4 (De Nijs *et al* 1994), show with more detail unexpected features in the

partial electron spectra, measured in coincidence with the target charge state.

For multi-electron processes the theory is not so far developed that it is possible to predict in detail the electron capture dynamics. The overbarrier model, described in chapter 2 (Niehaus 1986), is capable of explaining the gross features of the electron spectra (Barat and Roncin 1992, Posthumus and Morgenstern 1992, De Nijs *et al* 1994). This model gives good predictions of the states in which the electrons are captured. No information on populated substates can be extracted from the model and therefore it is only suitable for rather qualitatively considerations of the electron capture dynamics. The key to the model is resonant electron capture, i.e. electrons are independently captured into states of the projectile ion with binding energies almost equal to their binding energies in the target.

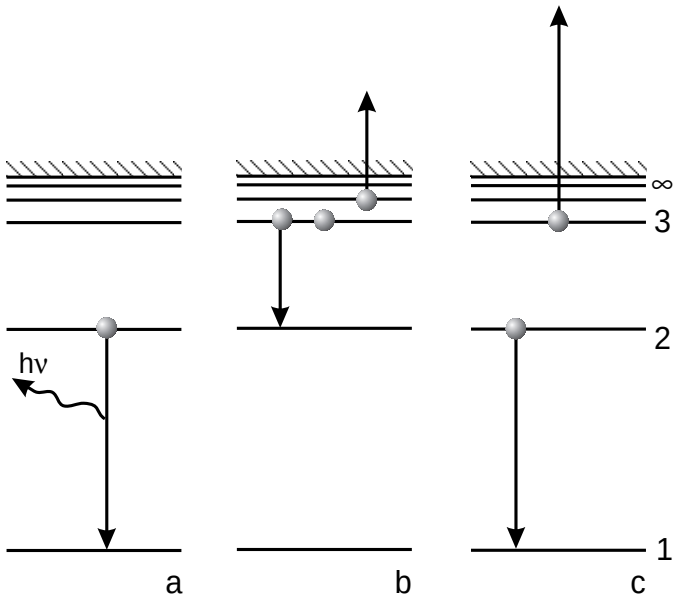

Figure 1.1: (a) After capture of one electron the excited projectile will stabilize radiatively. Multiply excited states preferably decay via autoionization. For example three captured electrons decay in two steps. In (b) the first step is drawn. The two electrons exchange energy and one of them escapes, whereas the other makes a transition to a lower state. The resulting ion decays further (c) emitting another electron. The energy of the emitted photons and electrons is a fingerprint of the initially populated states.

In figure 1.1 a schematic energy diagram of electrons captured into excited states of a projectile ion is plotted. If only one electron is captured in an excited state, figure 1.1a, it will decay toward the ground state accompanied by photon emission corresponding to the energy difference between the initial and the final state. Two- or more-electron capture into excited states will result not only in the emission of photons, but preferably in the emission of electrons. This situa-

tion is drawn in figure 1.1b as an example for the case of three-electron capture. Two electrons are captured into the $n = 3$ and one into the $n = 4$ orbital of the projectile. In the first step of the stabilization process one electron decays from $n = 3$ to $n = 2$. The energy difference $E_3 - E_2$ will be exchanged to the electron in the $n = 4$ orbital. This one will be emitted provided its binding energy E_4 is less than the energy difference.

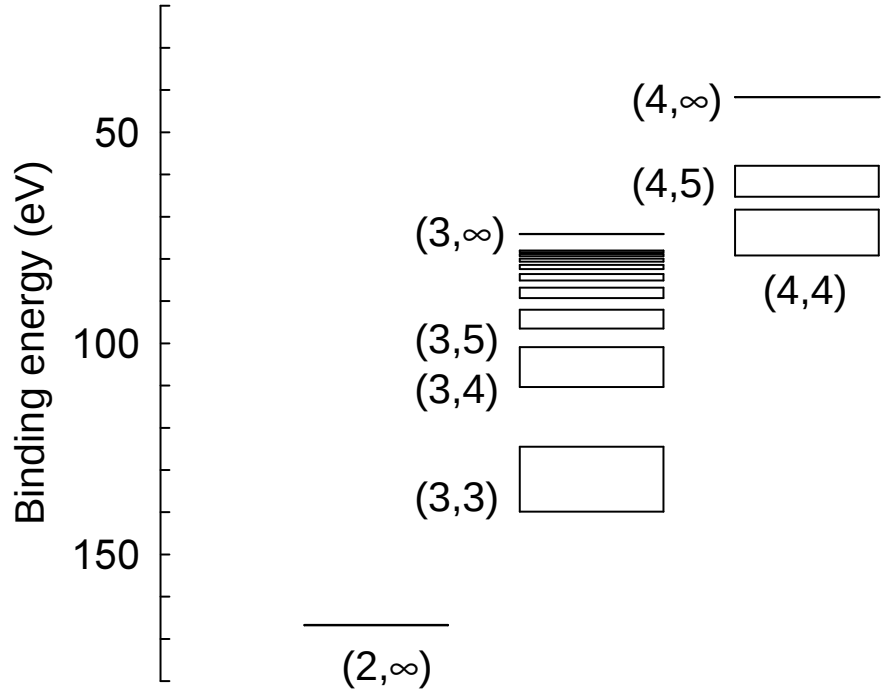

Figure 1.2: Part of the energy level diagram of doubly-excited states in N^{5+} calculated using the Cowan code (Cowan 1984). Note that the (3,n) configurations overlap the (4,4) structure giving the possibility of radiative stabilization after populating the (4,4) manifold.

Collision systems with bare projectile ions are the most suitable for precise identification of initially populated states after double electron capture by means of electron spectroscopy. This is because the final states of an ion after an autoionizing decay is degenerate, thus facilitating the analyzes the electron spectra. A collision system including such a projectile ion is N^{7+} - Ar. In figure 1.2 the binding energies are plotted of several doubly excited states in N^{5+}. The (n_1, n_2)=(4,4) manifold overlaps with the (3,n) series for $n > 9$. This energy degeneracy allows a strong configuration interaction between the (4,4) manifold and the (3,n) series, implying that the asymmetric (3,n) configurations can be populated from the symmetric (4,4) manifold and vice versa. The overbarrier model predicts initial population of the (4,4) manifold, due to the almost equal

binding energies of the electrons in the target. This symmetric manifold will decay mainly via autoionization. However, due to the overlap with the (4,4) the (3,n) configuration can be populated which preferably stabilizes radiatively. Thus despite of the initial population of the (4,4) manifold, radiative stabilization is possible due to the flux of intensity from (4,4) to the (3,n), $n > 9$, series.

All systems exhibiting an overlap between symmetric configurations and asymmetric configurations seem to have an increased radiative stabilization. This feature has motivated many theoretical (Vaeck *et al* 1992, Roncin *et al* 1993, Van der Hart *et al* 1995) and experimental (Bachau *et al* 1992, Martin *et al* 1994, Ali *et al* 1993) investigations.

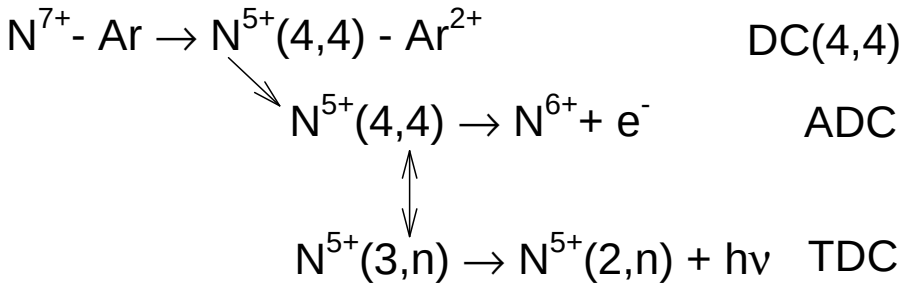

Figure 1.3: The reaction N^{7+}-Ar. Double electron capture into the (4,4) manifold is denoted by DC(4,4). ADC indicates autoionizing double capture, i.e. capture into (4,4) and decay emitting an electron and thus changing the charge state. TDC means true double capture, i.e. the final charge state is not changing after capture of two electrons. For $n > 9$ the (3,n) series overlap energetically with (4,4) (fig 1.2), giving rise to radiative stabilization.

A possible way to study this can be exemplified by the reaction scheme shown in figure 1.3. Two electrons initially captured into the (4,4) manifold, indicated as DC(4,4), can decay via autoionization (Autoionizing Double Capture: ADC) or decay radiatively (True Double Capture: TDC). The first yields a six-fold the latter a five-fold charged projectile. Both double electron capture processes leaves the target doubly ionized. Coincidence measurements between the charge of the target and projectile give the information on the competition between the two sketched processes.

The fraction TDC/DC between the production of N^{5+} and the total double capture (DC) production is a measure for the radiative stabilization fraction of the autoionizing manifold. For N^{7+} ions impinging on Ar this ratio has been found to be about 30 % (Roncin *et al* 1991). It is presumed that the total double capture probability is mainly determined by capture of electrons into the (4,4) manifold. The goal of the research presented in this thesis is to explore the autoionizing features of systems exhibiting an enhanced radiative stabilization ratio.

In chapter 4 measurements are presented which identify the initially populated manifold by determining the energy of autoionizing electrons in coincidence with Ar^{2+}. The measurements show clearly that beside double electron capture

in the (4,4) manifold a large contribution of autoionizing electrons stem from configurations which do *not* stabilize radiatively. This implies that the radiative stabilization ratio TDC/DC(4,4) is even larger than the average ratio TDC/DC.

The effect of target excitation should not only be present in the N^{7+}-Ar case but also in other collision systems. Chapter 5 shows the results for collisions between C^{6+} on Ar. This collision system has a low radiative stabilization fraction for double electron capture. The kinetic energy of the electrons measured in coincidence with Ar^{2+} show also contributions of electron capture accompanied by target excitation. This chapter presents the analyzes of partial electron spectra in coincidence with capture of two, three, four and five electrons. The measurements show that the electrons are primarily captured into manifolds predicted by the overbarrier model. Also shown is a deconvolution of the possible pathways leading to charge equilibration by emission of electrons after multiple electron capture. A remarkable property is that despite capture of five electrons in C^{6+}, thus yielding C^{1+}, the final charge state of the projectile, due to a number of autoionizing decay steps, is C^{4+}.

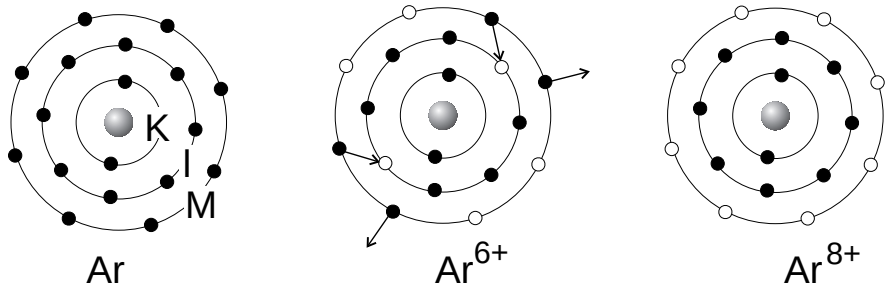

Figure 1.4: A simplified picture of the autoionizing decay of the Ar target after capture of inner shell electrons by C^{6+}. The left picture is Ar before the collision, the middle figure is directly after electron capture by the ion. In this figure it is assumed that six electrons are captured including two L-shell electrons. The right figure shows the result of the autoionizing decay of target electrons.

The importance of inner-shell electron capture is shown in chapter 6. When electrons in the more outer lying shell remain on the target these remaining electrons can be viewed as being 'excited'. In this chapter the results of measurements on the final charge state distributions of the target are shown, measured with a high resolution mass/charge spectrometer. The collisions of C^{6+} on Ar result in a charge state distribution of the target up to 8+. With a certain probability the final charge state of the target is therefore *higher* than the initial one of the projectile, i.e. that more electrons are removed from the target than could be captured by the projectile. In the collision energy range of our experiments the probability for direct ionization is small (Cocke *et al* 1981, Justiniano *et al* 1981), so the observed phenomena must be a result of charge redistribution

within the collision system. The interpretation of this effect is the following. During the collision the internuclear distance between the bare C^{6+} ion and the Ar target is so small that two electrons in the L-shell of Ar can resonantly be captured into the K-shell of C^{6+}. In figure 1.4 a schematic picture is sketched of this inner-shell electron capture and the decay afterwards. Due to the absence of any electrons in C^{6+} the L-shell electrons of Ar can be captured.

The measurements presented in this thesis provide information on the dynamics of multiple electron capture by highly charged ions. It is shown that the capture of electrons often is accompanied by target excitation. The measurements clearly show the importance of inner-shell electron capture. Different pathways to charge equilibration are shown in the presented collision systems which give information on the validity of the overbarrier model.

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