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

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Chapter 7

Conclusions and outlook

In this thesis charge exchange processes occurring in collisions between highly charged ions and gas targets were investigated. Conclusive experimental data were obtained by means of coincidence measurements of electrons and target-ion distributions. The reactions studied may be characterized by the following scheme:

$$A^{q+} + B \rightarrow (AB)^{q+} \rightarrow A^{(q-t)+} + B^{r+} + (r-t)\ e^{-} \qquad (7.1)$$

where A denotes the q-fold charged ion and B the neutral target, which becomes r times ionized after the collision .

This type of collisions is subject of extensive research in many laboratories since the early eighties, when sources to produce beams of intense highly charged ions became available (Geller and Jacquot 1982). By measuring the decay of excited ions formed in the collision combined with the measurements on the distribution of projectile and target ions lead to the discovery of an unexpected phenomenon (Roncin *et al* 1991), which is known as radiative stabilization. As described in the introduction, the configuration interaction of symmetric doubly excited (n_1,n_2)=(4,4) states with the asymmetric (3,n) series may allow a flux from (4,4) to (3,n) and vice versa, i.e. electrons captured in the doubly excited autoionizing manifold (4,4) will be transferred to the radiative stabilizing (3,n) series. Since then a lot of experimental (see e.g. Bachau *et al* 1992, Martin *et al* 1994, Ali *et al* 1993) as well as theoretical investigations (see e.g. Van der Hart *et al* 1995) have been performed on various collision systems exhibiting this effect.

The goal of the research presented in this thesis is to investigate the autoionizing properties of systems exhibiting radiative stabilization. One of such collision systems is N^{7+}-Ar. Capture of the two most loosely bound electrons, the most probable double electron capture process, is predicted to be into the (4,4) manifold of N^{5+}. Roncin *et al* (1991) investigated this system by means of energy gain measurements. The radiatively stabilizing fraction was determined to be about 30%. This collision system, as all systems including bare projectile

ions, turns out to be very suited for electron spectroscopy due to the degenerate final state of the ion.

The electron spectra for double electron capture show autoionization electrons over a wide range of energies. This range is much broader than expected from capture of electrons into the (4,4) manifold only. The measurements show electrons resulting from the (3,n) series with n=4 and 5. It has been shown that it is not possible to populate these states from the flux (4,4)→(3,n). The population of these doubly excited states can however be explained by assuming electron capture accompanied by the excitation of the target. This leads to a direct population of the (3,4) and the (3,5) states.

To test this description further we collided C^{6+}-ions on Ar. In this case double electron capture is predicted to be into the (3,5) manifold of C^{4+}. The (3,5) configurations do not overlap with any other doubly excited series, and thus the possibility for radiative stabilization of doubly excited states is low. Therefore this system is suited for testing target excitation. For double electron capture the measurements show indeed excitation of the target. For multi-electron capture it is shown that a reconstruction can be made up five-electron transfer to C^{6+}, also directly showing the uniqueness of the coincidence technique. The partial electron spectra measured using this method give deep insight in multi-electron capture processes. For example, the different pathways leading to the decay of excited carbon show that the final charge state of carbon ions is five or four, despite capture of up five electrons in C^{6+}. In the identification of the pathways, target excitation has been included and turns out to be of major importance in multi-electron capture processes.

The effect of target excitation can also be observed in charge state distributions of target particles. The measurements show a small but clear yield of Ar^{8+} in C^{6+}-Ar collisions. The fact that the target charge state is *higher* than the projectile charge state may be explained by taking inner-shell electron capture, i.e. target excitation, into account. Inner-shell electron capture is possible due to the 1s vacancies in the bare C^{6+} projectile ions. Six fold ionized projectiles with an occupied K-shell (like O^{6+} or Ne^{6+}), show a maximum target charge state depending on the projectile core structure. The core structure of the projectile was found to be crucial for the final charge state distribution.

The phenomena described in this thesis can be studied further by modifying and further improving the experimental setups. One of the possible extensions to the electron coincidence setup, described in chapter 3, is to measure also the projectile charge in coincidence with the electron emission and target ion distribution. It is recommended that the target ion distribution is measured using high resolution time-of-flight measurements as described in chapter 3. Using a similar setup Dörner *et al* (1994) could distinguish between electron-electron and nucleon-electron interactions in fast ($\sim$ 1 MeV) He^{+}-He and 130 eV e^{-}-He collisions. They were also able to measure the precise spatial distribution of the momentum of the target ions. The method may also be used to determine more information on, for example, double electron capture. The observed autoioniza-

tion electrons resulting from the (3,4) manifold in N^{7+}-Ar collisions (chapter 4) which are accompanied by target-excitation processes should be measured in coincidence with a larger recoil energy for the target ions than for those ions when electrons initially are captured into the (4,4) manifold. The extra information obtained in this way leads to a more profound understanding of the mechanisms leading to target excitation. Coincidences between electron, projectile ions and target ion distributions will therefore give conclusive evidence on target excitation. It is from this information possible to determine precisely the radiative stabilization ratio for double electron capture of the outer two electrons.

The recoil energy of the target ions is dependent on the distance of closest approach. It is expected that the widths of the peaks in the target-ion distributions depend on the observed target charge state. Measurements of the charge state distributions of target ions, presented in chapter 5, show more or less a charge state dependence of the width of the measured peaks. The width of the peaks is in this case mainly determined by the length of the ion bunches. It is therefore recommended to improve the bunch width of the chopped ion beam. Especially for collisions between C^{6+}-Ar, where Ar^{8+} was observed, the recoil energy dependence on the charge state can be determined with high accuracy.

Throughout this thesis it was shown that the overbarrier model (Niehaus 1986) seems to be capable of predicting fairly well the initially populated manifolds. The model is (semi) classical, so for an improved description of the collision dynamics complex quantum mechanical calculations should be carried out. A full quantum mechanical description of a collision system should take into account all correlations between electrons and ions in the collision system. Also the movement of the projectile needs to be evaluated. For one-electron capture processes quite some work has already been done and these processes are fairly well understood (Fano and Lichten 1965, Lichten 1963, Niehaus 1980, Harel 1995, Zouros *et al* 1987, Jain *et al* 1989, Hoekstra 1995, McCullough *et al* 1995, Fritsch and Lin 1991, Folkerts *et al* 1993). The theoretical approaches are based on calculating the energies and lifetimes of various multiply excited states as function of the internuclear distance.

For multi-electron systems it is rather difficult to calculate the binding energies accurately. For two-electron processes some work has been done. For collisions between He^{2+} and He it is expected that electrons are mainly captured resonantly into the ground state and with low probability in excited states of He^{2+}. The measurements on the ejected electrons show however, apart from some autoionizing lines, a continuous 'background'. In figure 7.1 the results are plotted for 20 keV He^{2+} projectiles. Part of the electron spectrum can be described by the semi-classical description of Miller (1970) which makes use of calculated energies (Fritsch 1994) and lifetimes (Sato and Hara 1980,1984) of the excited states as input parameters. The solid curve in the spectrum is the result of this modelling. It is clear that this approach leads to a rather well description of the measured data.

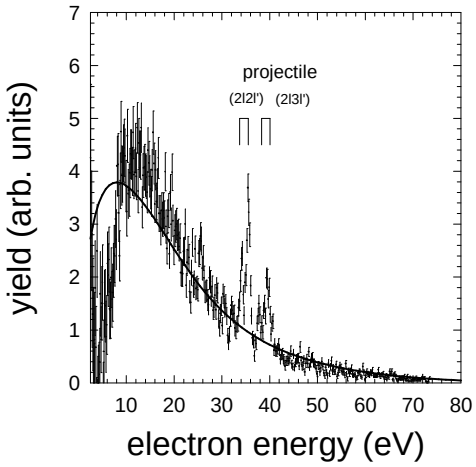

Figure 7.1: Electron spectrum resulting from collisions between He^{2+} and He. On top of the continuous spectrum peaks can be identified which correspond to autoionizing electrons from initial electron capture in the (2,2) and (2,3) manifold emitted by the projectile. The continuous spectrum turns out to be reproduced rather well by calculations which include the binding energy of the electrons as well as the lifetime of the molecular state as function of the internuclear distance.

To summarize, it is shown that the measurements presented in this thesis show the importance of target excitation. This implies that one has to include *all* electrons present in a collision system to do proper predictions on several effects which are measured in highly charged ion collisions. There is a need for proper quantum-mechanical calculations on binding energies and lifetimes of electrons in *multi*-electron systems.

In conclusion, some light is shed on the collision dynamics of highly charged ions and neutral atoms so the pathways to charge equilibration are more visible now.

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