**TRANSFER EXCITATION AND CAPTURE OF INNER TARGET ELECTRONS IN COLLISIONS
WITH SLOW MULTIPLY CHARGED IONS**

H.R. KOSLOWSKI, H. LEBIUS and B.A. HUBER

*Institut für Experimentalphysik AG II, Ruhr-Universität, D-4630
Bochum, F.R.G.*

Abstract

Single electron capture by triply charged Ne projectiles in He and Ne has been studied at low collision energies (600 eV) showing the dominant mechanisms to be due to transfer excitation and capture of inner target electrons. The results are compared with predictions of various classical models and improved multi-channel LZ calculations. Information is obtained on the coupling strength relevant for both types of processes.

1) Introduction

Single electron capture reactions have been studied very extensively in experiments and by theory during recent years. The basic type of reaction is described by the following equation:

Here q describes the charge state of the projectile and the entity ΔE represents the energy defect of the reaction. The mechanism of this process is well established, nevertheless, looking for finer details, the selection of individual reaction channels and their relative intensity distribution remains still unexplained in some collision systems.

Within the so-called classical model /1-3/ the target electron is allowed to be transferred to the projectile only, if the potential barrier between both particles is reduced below the binding energy of the active electron. Therefore a minimal approach or distance of both colliding particles is required, determining the size of the cross section as well as the population of the final projectile states. On the one hand this model has been extended by Barany /4/ in order to describe the capture of several electrons. On the other hand Niehaus /5/ has refined the description by considering molecular electrons and by taking into account the uncertainty principle with respect to the transition time and the height of the potential barrier. By this means a wider range of nuclear distances - the so-called reaction window - is defined where electron transfer occurs.

Kimura et al /6/ derived another type of reaction window using the Landau Zener formalism (LZ). Within this frame LZ-cross sections for individual reaction channels can be represented as a product of a transmission function T and a geometrical cross section, whereby T depends on the collision velocity as well as on the nuclear distance R_c where the crossing of the involved potential curves takes place, and the strength of the coupling matrix element H_{12} . As H_{12} may be

expressed in terms of R_c according to the relation given by Olson and Salop /7/, the transmission function T depends on R_c only. The maximum of the transmission curve shifts towards smaller crossing distances with higher velocities, thus favouring electron capture reactions with larger exoergicities. Taulbjerg /8/ introduced the dependence of H_2 on the quantum numbers of the atomic state populated in the electron capture reaction and therefore obtained a 'reaction window' where the position depends on the principal and azimuthal quantum numbers n and l .

Collision systems which are characterized by many exit channels have been described with a multi-channel Landau Zener formalism (MCLZ) which has been formulated by Olson and Salop /9/. Very recently this type of calculation has been performed including the above mentioned Taulbjerg factors /10,11/.

These models which describe many experimental findings remarkably well suggest a large reaction probability if the potential curve crossing of the involved molecular states lies within the reaction window. However, further selection and propensity rules regarding the total electron spin, the symmetry of the molecular states and the ion core of the projectile have to be considered.

In the following the predictions of these models will be discussed by means of two collision systems where 2-electron processes or the capture of inner target electrons are dominating.

II) Experimental method

The collision processes have been analysed with the aid of the translational energy spectroscopy technique, which is described in more detail elsewhere /12/. Briefly, a mass-selected ion beam being analysed with respect to charge state q and kinetic energy E collides with a static gas target. The change of the kinetic energy as well as the charge state of secondary ions is measured with a double hemispherical analyser. The translational energy spectra which are discussed below are taken in forward direction. These spectra contain information on the final atomic states as well as on the initial internal state of the projectile.

The composition of the primary ion beam with respect to ground-state ions and ions in various excited metastable states has been determined by applying the translational attenuation method /13/ and by analysing collision induced excitation and deexcitation processes /14/. Furthermore, charge transfer reactions are studied for various electron energies E_e in the ion source.

III) Multichannel Landau Zener calculations (MCLZ)

In the following we will briefly describe an extended multichannel LZ-model, which we have modified in order to reproduce the experimental results. This method takes into account, that in the experiment projectile ions in different internal states are present. Therefore more than one incoming channel is considered and the coupling between different entrance channels via the strong repulsive exit channels is included in these calculations.

Figure 1 shows a simplified schematic potential curve diagram representing M entrance channels ($j = 1 \dots M$) according to the groundstate and further metastable states of the projectile and N exit channels ($i = 1 \dots N$) characterizing all possible final states. Hence the collision system is described by a grid of $N \times M$ potential curve

Figure 1: Schematic potential curve diagram for M entrance and N exit channels

crossings where transitions between different molecular states occur and where the incoming particle flux ϕ is distributed on different paths. At each curve crossing the partition of ϕ is determined by the Landau Zener probability $p^{i,j}$, which gives the probability to stay on the diabatic potential curve while passing the crossing point. The coupling matrix elements H_{ij} which are included in $p^{i,j}$ have been calculated using the following expression / 7,8/:

$$H_{ij} = 9,13 \text{ b} f_{b,i}^{1,j} q^{-1/2} \exp \{-1,324 R c^{1,j} (2I/q)^{1/2}\}, \quad (2)$$

where I represents the ionisation potential of the target and $f_{b,i}$ the so-called Taulbjerg factor. The factor b is included in order to take into account various selection rules, setting b equal to 1 or 0. Furthermore, by varying the value of b between 1 and 0 the coupling strength can be reduced, which turns out to be useful in the case of multi-electron processes as will be discussed below.

For each curve crossing (i,j) four different fluxes can be defined in a recursive manner: Two for the case of approaching particles ($\phi_{in}^{i,j}$, $\phi_{up}^{i,j}$) and two for the separating quasimolecule ($\phi_{out}^{i,j}$, $\phi_{down}^{i,j}$). Considering the appropriate boundary conditions the evolution of the incoming flux can be calculated for a given impact parameter. The normalized flux in a specific final state is integrated over the impact parameter range yielding absolute state-selective cross sections.

The main advantage of this procedure is the automatic inclusion of the coupling between incoming channels due to the crossings with the Coulomb curves of the exit channels. Therefore, for a given entrance channel other parallel curves represent competitive exit channels leading to the excitation or deexcitation of the projectile. Due to the additional loss of flux the electron capture cross sections are expected to become somewhat smaller.

IV) Discussion of results

The following reactions will be considered below:

The atomic states which are present in the ion beam are: ($4S_{3/2}$), ($2D^0$) and ($2P^0$), whereby the excitation energy of the metastable states amounts to 5.1 eV and 7.7 eV, respectively. Due to the high electron energy ($E_e = 500$ eV) the relative fractions are close to a statistical distribution, i.e. $f(4S^0)/f(2D^0)/f(2P^0)$ corresponds roughly to 2:5:3.

b) $Ne^{3+} + He$

Single electron capture by Ne^{3+} in He has been studied recently by Tunnell et al /17/ and Schmeissner et al /18/ showing two dominant reaction channels caused by Ne^{3+} projectiles in the groundstate ($4S^0_{3/2}$):

and

Both reactions exhibit unfavourable energy defects with respect to the reaction window, i.e. the potential curve crossings lie at rather large or rather small internuclear distances (9.2 a. and 2.0 a.). Furthermore, both channels represent two electron processes as an electron configuration $2s2p^5$ is created in the final state, i.e. one electron is captured into the 2p level and a further 2s electron is excited into 2p. Therefore the weak intensity measured in this experiment seems to be plausible.

Figure 2: Energy gain spectrum of Ne^{2+} ions formed in Ne^{3+}/He collisions. $E=600$ eV, $E_e=500$ eV. The positions of spin allowed and spin forbidden channels are marked as full and broken vertical lines, respectively.

However, the dominant population of the $^1P^0$ state compared to the $^3P^0$ state seems to be inexplicable for two reasons. On the one hand the classical model requires a minimal approach of 5 atomic units in order to transfer the electron on a classically allowed path, i.e. the energy gain should be larger than 10 eV. On the other hand reaction (5) violates Wigner's spin rule. Whereas the total spin in the incoming channel amounts to $3/2$, it is $1/2$ in the exit channel only.

In order to test these objections we have studied the same system with higher resolution. The result is shown in Figure 2b. Like in the previous measurements the dominant final state turns out to be the $2s2p^5 \ ^1P^0$ state of Ne^{2+} , however, the electron capture is due to Ne^{3+}

projectiles in the metastable ($^2P^o$) or ($^2D^o$) states. The dominant reaction channel is described by the equation:

Additional support for this identification has been obtained by the near threshold translational energy spectroscopy /15/ performed in a He/Ne target gas mixture (compare section c). These measurements clearly show reaction (7) being due to the second metastable state $^2P^o$ of Ne^{3+} .

The dominant role of reaction channel (7) is explainable as the total electron spin is conserved and the energy defect is more adequate. The surprising fact that reaction channel (6) does not contribute at all is due to the symmetry of the involved molecular states. Whereas the approaching particles will form a $^4\Sigma^-$ molecular state only, the secondary ions can interact via $^2,4\Sigma^+$ and $^2,4\Pi$ potential curves. If we assume that radial coupling holds at these low collision energies, reaction (6) is forbidden because of symmetry violation. The same is true for reaction channel (5). However, if metastable projectile states are considered a molecular coupling between different Π or Σ^+ states is possible, fulfilling all selection rules.

Figure 3: State-selective single electron capture cross sections for Ne^{3+}/He collisions. Full bars: experiment, striped bars: MCLZ calculation, broken curves: 2-state LZ result with different f_{n1} and b factors, full and dotted curves: Niehaus model.

From the energy gain spectrum shown in Figure 2b and the primary beam fractions absolute cross sections are estimated for the 4 dominant reaction channels which are shown in Figure 3 in comparison with various theoretical results.

Reaction channel 1 representing a 'pure' single capture process is described well by the 2-state LZ curve with the corresponding f_{n1} value of 0,58. However, the intensities of channels 2 to 4 which involve multi-electron processes are not well described by the reaction window with $f_{n1}=0,7$ according to capture into the 2p level. This window suggests channels 2 and 3 to be of the same importance, whereas peak 4 should not occur at all. An excellent agreement is obtained, however, if within a two state LZ-calculation the factor b in equation (2) is set to 0,3. Obviously, electron capture with simultaneous excitation of a further projectile electron has to be described by a much lower coupling strength; in the present case it is smaller by a factor of ≈ 3 .

The striped bars show the result of a MCLZ calculation, whereby channel 1 is described by $f_{n1} = 0,58$ and $b=1$ and channels 2 to 4 by $f_{n1} = 0,7$ and $b=0,3$. As the experimental and theoretical values are determined on an absolute scale the obtained agreement is rather encouraging. In the present case even the simple LZ calculation yields good results as the number of final states is very limited and the size of the cross sections is relatively small.

c) $\text{Ne}^{3+} + \text{Ne}$

The translational energy spectrum of Ne^{2+} ions obtained in Ne^{3+}/Ne collisions is shown in figure 4. A surprising feature of this spectrum is the large exoergicity of the dominant reaction channels, which are mainly due to the capture of an inner 2s target electron in agreement with earlier results of optical measurements /19/. The dominant channel is caused by Ne^{3+} ions in the ground state ($4S_{3/2}$):

The same target state is populated by three additional reactions:

Further contributions due to the formation of the $(2s2p^5\ ^1,^3P^{\circ})$ states of Ne^{2+} are found.

A comparison with theoretical considerations is made in Figure 4b. Again we find reasonable good agreement with our multi-channel calculation, if we take into account that the 2s target electron has an enhanced ionization energy which shifts the region of strong coupling between the incident and outgoing channels to larger exoergicities. These details are not included in the simple LZ calculation which would favour reaction channels with much lower energy gains (see figure 4b). The classical model according to Niehaus /5/ in principle also describes the capture of 'inner' target electrons (see lower dotted curves in figure 4b), however, this model does not include the capture of a 2s target electron. When during the approach of both particles three 2p electrons have become molecular the system is totally symmetric and capture of more tightly bound electrons is not possible (during a further approach two corresponding electrons from projectile and target become molecular simultaneously and have to occupy their initial states while the quasimolecule separates).

Whereas the formation of the $3S^{\circ}$ states seems to be hindered by the core conservation propensity rule, the MCLZ calculation yields too large cross sections for the reactions characterized by $Q = 11 \text{ eV}$ and 13 eV . This discrepancy is not understood at present.

Conclusion

We have measured translational energy spectra obtained in charge changing collisions between triply charged Ne ions and various rare gas atoms. A careful analysis, which explains ambiguities in the interpretation of some spectra reported recently, also shows that certain features of single electron capture processes are not explained by existing simple models.

In order to improve the theoretical description we have made some extensions of recent multichannel LZ calculations. Firstly, different

Figure 4: a) Energy gain spectrum of Ne^{2+} produced in Ne^{2+}/Ne collisions. Full vertical lines mark those energy gains which are correlated with the formation of specific Ne^{2+} states. An asterisk indicates the capture of a 2s target electron. b) State-selective cross sections. Full bars: experiment, open bars: MCLZ calculation, other notations see figure 3.

incoming channels may influence each other due to an adiabatic coupling at curve crossings with the repulsive potential curves. Our modified MCLZ calculation takes into account all possible transitions between several incoming and several outgoing channels.

The second extension concerns the semi-empirical expression for the coupling matrix element H_{ij} . A new factor b has been introduced which reduces the coupling strength in the case of multi-electron processes. E.g. the population of the $(2s2p^5)$ and $(3s3p^5)$ configurations by single electron capture - starting from $(2s^2 2p^3)$ and $(3s^2 3p^3)$ projectile configurations - is predicted well if b is chosen to be 0.3, i.e. if the coupling strength according to Taulbjerg is reduced by a factor of ≈ 3 .

An adequate description of the capture of an inner target electron has to consider the correct ionization energy of the active electron. Due to the larger binding energy the coupling strength becomes smaller and hence the reaction window is shifted towards larger exoergicities. Therefore, in some collision systems the selection of final states as well as the size of the total cross section deviates remarkably from the predictions of simple models.

Acknowledgments

The authors would like to thank Prof. K. Wieseemann for valuable critical discussions concerning different aspects of this work. These experiments have been supported by a grant from the Deutsche Forschungsgemeinschaft which is gratefully acknowledged.

References

1. N.Bohr and J.Lindhard; K.Dan.Vidensk.Selsk.Mat.-Fys.Medd.28,1 (1954)
2. H.Ryufuku and T.Watanabe; Phys.Rev.A 18 , 2005 (1978)
3. R.Mann, F.Folkmann and H.F.Beyer; J.Phys.B 14 , 1161 (1981)
4. A.Barany, G.Astner, H.Cederquist, H.Danared, S.Huldt, P.Hvelplund, A.Johnson, H.Knudsen, L.Liljeby and K.G. Rensfeldt; Nucl.Instr.Meth. B9, 397 (1985)
5. A.Niehaus; J.Phys.B 19 , 2925 (1986)
6. M.Kimura, T.Iwai, Y.Kaneko, N.Kobayashi, A.Matsumoto, S.Othani, K.Okuno, S.Takagi, H.Tawara and S.Tsurubuchi; J.Phys.Soc.Japan 53, 2224 (1984)
7. R.Olson and A.Salop; Phys.Rev.A 14, 579 (1976)
8. K.Taulbjerg; J.Phys.B 19, L367 (1986)
9. A.Salop and R.Olson; Phys.Rev.A 13, 1312 (1976)
10. S.M.Wilson, R.W.McCullough and H.B.Gilbody; J.Phys.B 21,1027 (1988)
11. M.Boudjema, P.Benoit-Cattin, A.Bordenave-Montesquieu and A. Gleizes; J.Phys.B 21, 1603 (1988)
12. B.A.Huber; Comments At.Mol.Phys. 21, 15 (1987)
13. B.A.Huber and H.J.Kahlert; J.Phys.B 16, 4655 (1983)
14. Y.Itoh, N.Kobayashi and Y.Kaneko; J.Phys.Soc.Japan 50, 3541 (1981)
15. J.Binder, H.R.Koslowski, B.A.Huber and K.Wieseemann; J.Phys.B 20 2713, (1987)
16. J.Hansen and W.Persson; J.Phys.B 19, L367 (1986)
17. L.N.Tunnel, C.L.Cocke, J.P.Giese, E.Y.Kamber, S.L.Varghese and W.Wagoner; Phys.Rev.A 35, 3299 (1987)
18. C.Schmeissner, C.L.Cocke, R.Mann and W.Meyerhof; Phys.Rev.A 30, 1661 (1984)
19. E.Bloemen, H.Winter, F.J.deHeer, R.Fortner and A.Salop; J.Phys.B:At.Mol.Phys. 11, 4207 (1978)
20. J.Puerta, H.-J.Kahlert, H.R.Koslowski and B.A.Huber; Nucl. Instr. and Meth. B9, 415 (1985)