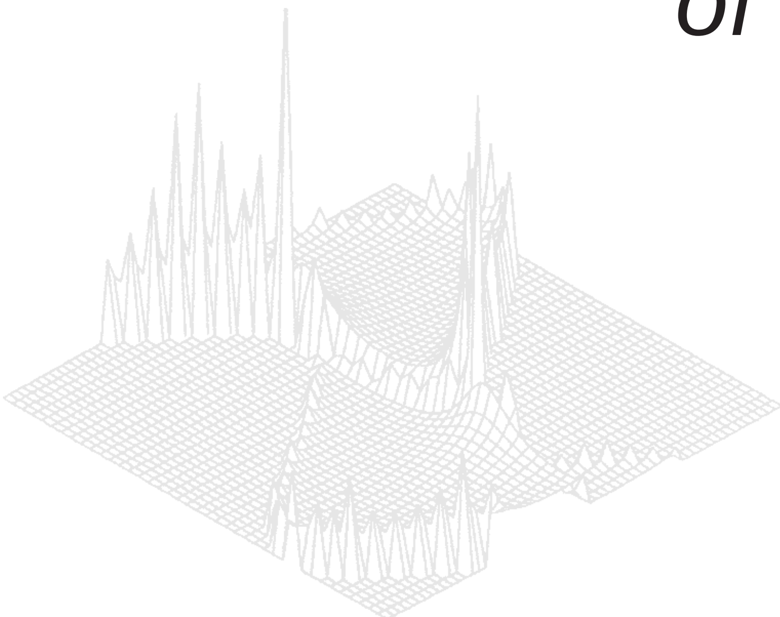
C S I R O P U B L I S H I N G

Australian Journal of Physics

Volume 51, 1998



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Aust. J. Phys., 1998, **51**, 679–89

Investigating Inner Shell Ionisation via the (e, 2e) Technique*

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Abstract

The (e, 2e) technique has been applied successfully to valence shell ionisation of many targets, but studies of inner shell ionisation by this technique have been limited. The triple differential cross section for the latter process exhibits behaviour which is very different to that observed for valence shell ionisation, particularly when the energy of the slow ejected electron is decreased below the binding energy of the inner shell orbital. Our recent results for inner shell ionisation of argon and krypton will be discussed, and comparisons made with distorted wave calculations.

1. Introduction

The importance and utility of the (e, 2e) technique in investigating electron impact ionisation may be gauged from the range of target species and kinematical conditions over which measurements have been performed. The primary theme of this workshop is the application of the (e, 2e) technique in electron momentum spectroscopy (EMS), where structural information about the electron motion in the target may be obtained. However, another emphasis in the application of the (e, 2e) technique is in studies of the dynamics of the ionisation process itself. Aspects of the collision such as distortions in the incident and outgoing channels, the influence of multiple scattering, decay processes following the ionisation and electron–electron correlation effects such as post collision interaction have been studied. Valence shell ionisation of numerous atomic targets has been studied in considerable detail, with most attention being directed to simple targets such as helium and hydrogen, where the target wavefunctions are well known. For other atoms, uncertainties in the target description can complicate the theoretical description of the collision; nevertheless good progress has been made in the theoretical approaches to this problem.

For larger atoms, core ionisation becomes a possibility. Associated with the primary core-hole formation is the possibility of Auger emission. Indeed, for targets other than very heavy atoms, the Auger decay process dominates over photon decay. The process may be represented as follows:

* Dedicated to Professor Erich Weigold on the occasion of his sixtieth birthday.

 10.1071/PH97093 0004-9506/98/040679$05.00

$$e_{\text{incident}}(E_0,\, \boldsymbol{k}_0) + A(\text{target atom}) \rightarrow A^{+*} + e_{\text{scattered}}(E_{\text{a}},\, \boldsymbol{k}_{\text{a}}) + e_{\text{ejected}}(E_{\text{b}},\, \boldsymbol{k}_{\text{b}})$$

$$\hookrightarrow A^{++} + e_{\text{Auger}}\,,$$

where the incident electron has energy E_0 and momentum $\boldsymbol{k}_0$, and the scattered and ejected electrons have energies and momenta of $E_{\text{a}}, \boldsymbol{k}_{\text{a}}$ and E_{b}, $\boldsymbol{k}_{\text{b}}$ respectively. The momentum transfer to the target in the initial ionisation process is $\boldsymbol{K} = \boldsymbol{k}_0 - \boldsymbol{k}_{\text{a}}$. Energy conservation gives

$$E_0 = E_{\text{a}} + E_{\text{b}} + \epsilon_{\text{i}}\,,$$

where ϵ_{i} is the binding energy of the orbital under investigation. The Auger electron is emitted with a fixed energy determined by the particular decay transition.

In recent years, attention has been directed to exploring the inner shell ionisation process, and the subsequent Auger decay, via the (e, 2e) technique. Electron–electron coincidence studies of the Auger decay process are usually given the acronym $(\text{e}, \text{e}'\text{e}_{\text{Auger}})$, indicating that one of the outgoing electrons from the primary ionisation is detected in coincidence with the subsequently emitted Auger electron. A number of studies of this type have been performed, detecting either the scattered electron or the ejected electron in coincidence with the Auger electron (see Lohmann 1996 and references therein; also Avaldi *et al.* 1995; Waterhouse and Williams 1997*a*, 1997*b*). The energy of the unobserved electron is known, but the results are an average over the angular distribution of this electron. Interesting post-collision interaction effects between the ejected electron and the Auger electron have been observed, resulting in energy shifts and possibly affecting the angular distributions of one or both electrons. Ideally, a complete investigation of this core ionisation/decay process would involve measuring all three outgoing electrons in coincidence. Such experiments have been performed (Ford *et al.* 1995) but they are very difficult and the studies are limited.

The initial core-hole formation can be studied by using the (e, 2e) technique. In this case, the scattered and ejected electrons produced in the inner shell ionisation are detected in coincidence. The measured cross section is known as the triple differential cross section (TDCS). The energies of the two electrons can be adjusted so as to minimise or enhance the influence on the cross section of any subsequently emitted Auger electrons. Relatively few results for this process have been published as the cross section for inner shell ionisation is much smaller than for outer shell ionisation, and there is a large background of electrons produced by ionisation of outer-lying shells. The first systematic study of inner shell ionisation was published by Lahmam-Bennani *et al.* (1984), in which they investigated Ar(2p) ionisation at an incident energy of 8 keV, ejected electron energy of 150 eV and momentum transfers ranging from 0·802 a.u. to 3·066 a.u. The results, performed in asymmetric coplanar kinematics, showed some surprising behaviour, in particular the presence of a large recoil peak (corresponding to electrons being ejected into a direction roughly anti-parallel to the momentum transfer direction). A simple, binary electron–electron collision cannot result in electrons being ejected into the backward direction. The presence of such electrons is taken to be an indication of multiple scattering processes, with an additional elastic scattering occurring at some stage of the collision process. In particular, under these kinematical conditions, the recoil peak is attributed to a backward elastic scattering of the ejected electron off the core. Lahmam-Bennani *et al.* found that first order calculations were unable to describe correctly the results they observed, despite the high incident energy at which the experiments were performed. Bickert *et al.* (1991) extended the inner shell (e, 2e) experiments on atoms to lower incident energies. They measured the TDCS for Ar(2p) ionisation at incident energies in the range 2–3 keV and much higher momentum transfers than employed by Lahmam-Bennani *et al.* Again, they found that first order theories could not explain the observed results, although a first Born approximation with Coulomb waves was found to give reasonable agreement at the higher incident energies, and in fact was able to describe quite well the 8 keV data of Lahmam-Bennani *et al.* A theoretical breakthrough came with the application of the distorted wave Born approximation (DWBA) to inner shell ionisation (Zhang *et al.* 1992). The DWBA naturally incorporates distortion in the incoming and outgoing electron waves, as well as multiple scattering effects. Good agreement was observed between the DWBA calculation and the data of Bickert *et al.* (see Zhang *et al.* 1992). However, further inner shell ionisation measurements for 4d ionisation in xenon were performed by Avaldi *et al.* (1993), who found that for some kinematical conditions the best that could be said about the comparison between the DWBA and the experiment was that the dips and bumps were more or less in the right place (Avaldi *et al.* 1993).

Very recently, new measurements have been produced for inner shell ionisation in argon and krypton (Cavanagh and Lohmann 1997, 1998; Taouil *et al.* 1998). These results, and comparisons with a number of theoretical approximations, will be discussed in Section 3.

2. Experiment

The results presented in Section 3 have been measured with two different electron coincidence spectrometers. The measurements on argon and krypton at an incident energy of around 1 keV have been performed at Griffith University, Brisbane, in a 'conventional' (e, 2e) apparatus, while the argon measurements at an incident energy of around 5 keV have been performed on a multichannel toroidal (e, 2e) spectrometer at the Université de Paris XI, Orsay. A brief description of each experimental apparatus is in order. Further details may be found in Lohmann *et al.* (1992), Cavanagh and Lohmann (1997) and El Marji *et al.* (1997).

The (e, 2e) spectrometer at Griffith University is comprised of two electrostatic hemispherical electron energy analysers, mounted coplanar with the electron gun (defining the scattering plane), and at right angles to the target gas beam. The electron gun is fixed in position while the two hemispherical analysers are independently rotatable. The experiments are performed in a crossed-beam configuration, where a well-defined electron beam crosses a (reasonably) well-defined gas beam at right angles. Electrons emitted from the interaction region are retarded in energy by five-element electron optical lenses before entering the hemispherical analysers where they are energy analysed and detected by channel electron multipliers. The experiments are 'single-channel' in energy, and the angular distribution is measured by rotating the ejected electron energy analyser in the scattering plane.

The (e, 2e) apparatus at Orsay is a new generation instrument which allows simultaneous detection of emitted electrons over almost the full in-plane angular range. Again, it is an asymmetric, coplanar crossed beam experiment in which the electron beam crosses a target gas beam at right angles. The scattered electrons are detected at small scattering angles by a 127° cylindrical electron energy analyser. The ejected electrons emitted into the scattering plane are detected by a toroidal electrostatic energy analyser which has been 'split', so that one-half plane of emitted electrons is imaged into one half-toroid, placed above the scattering plane, while the other half-plane of emitted electrons is imaged into the other half-toroid, which is placed below the scattering plane. At the exit of the half-toroids are position sensitive detectors, which detect the arrival (time and position) of an electron. As the toroidal geometry preserves angular information, this apparatus can simultaneously detect electrons over a wide range of ejected electron angles, thereby substantially improving the efficiency of an (e, 2e) experiment.

The kinematic conditions employed in the measurements reported here are summarised in Table 1.

Table 1. Kinematic conditions employed in the Griffith University measurements of the TDCS for inner shell ionisation

Target	E_0 (eV)	E_a (eV)	E_b (eV)	θ_a (deg.)	K (a.u.)	θ_K (deg.)
Argon 2p	1249	880	120	15	2·76	48·9
	1179	880	50	15	2·59	53·5
	5719*	5460	10	0·5	0·5	20·4
Krypton 3d	1046·4	880	72	15	2·31	64·3
	1024·4	880	50	10	1·59	61·6

* Measurement made at the Université de Paris XI.

3. Results and Discussion

The measured TDCS for Kr(4p) ionisation for the case $E_a = 880$ eV, $E_b = 30$ eV and $\theta_a = 5°$ ($\epsilon_i = 14\cdot4$ eV) is shown in Fig. 1. This cross section illustrates some typical features of the TDCS for valence shell ionisation measured under similar kinematics to those used in the inner shell ionisation measurements. In particular, the cross section exhibits a large peak in the 'binary' region which is almost symmetric about the momentum transfer direction $\boldsymbol{K}$. The cross section in the binary region is dominated by direct electron–electron collisions, where there is little interaction with the ion. It is notable that there is essentially no emission into the recoil region (no 'recoil peak'), indicating that multiple scattering processes involving the ion do not play a significant role in valence shell ionisation under these conditions. One may contrast this with the situation for inner shell ionisation of krypton (see Fig. 3*b* below). One thing which is immediately apparent is the presence of a very large recoil peak in the cross section, an observation which will be explored in more detail below.

A number of theoretical approximations will be compared with the experimental results, and it is appropriate to briefly summarise them. All the measured cross sections, except the measurement at high incident energy in Table 1 made at the Universite de Paris XI, are compared with DWBA calculations. The DWBA calculations were performed using a program code provided by McCarthy (1995), and details of the approximation and the calculation may be found there and in references therein. Additionally, one of the TDCS measurements for Kr(3d) ionisation has been performed under bound Bethe ridge conditions, where the momentum transferred to the target is completely taken up by the ejected electron. This corresponds to an impulsive collision regime, where the distorted wave impulse approximation (DWIA) has been shown to work successfully for valence shell ionisation. Hence we have also included a DWIA calculation in one case. The calculation again was performed using the program code of McCarthy, and the formulation of the DWIA may be found in McCarthy (1995).

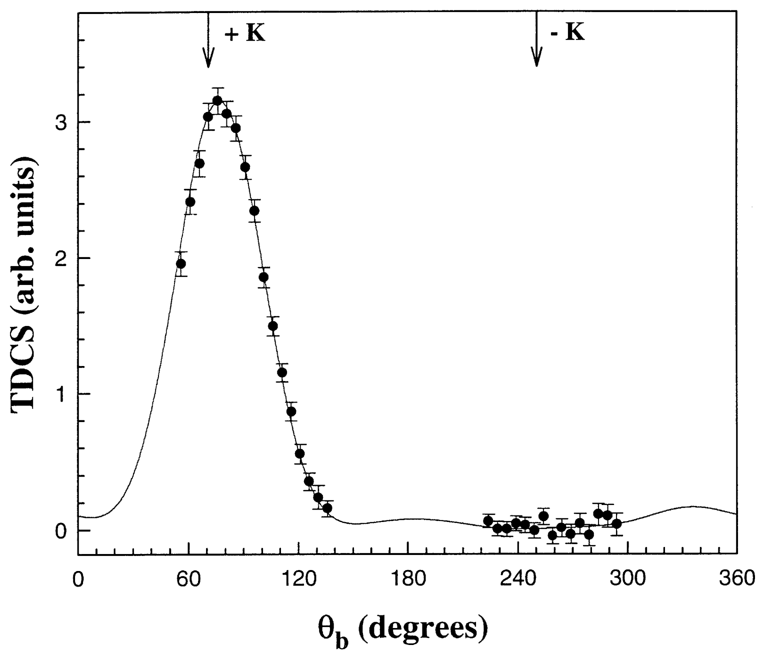
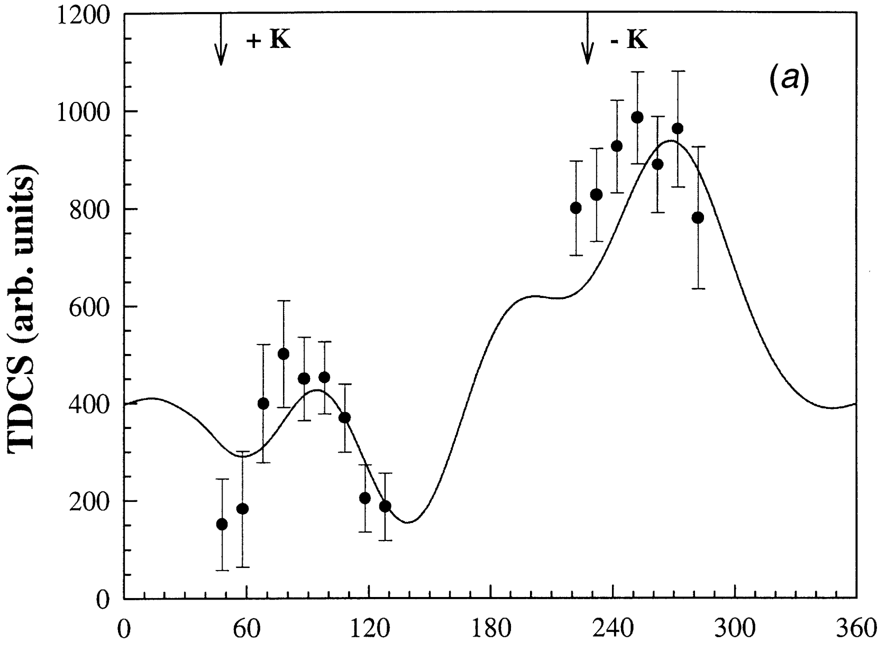
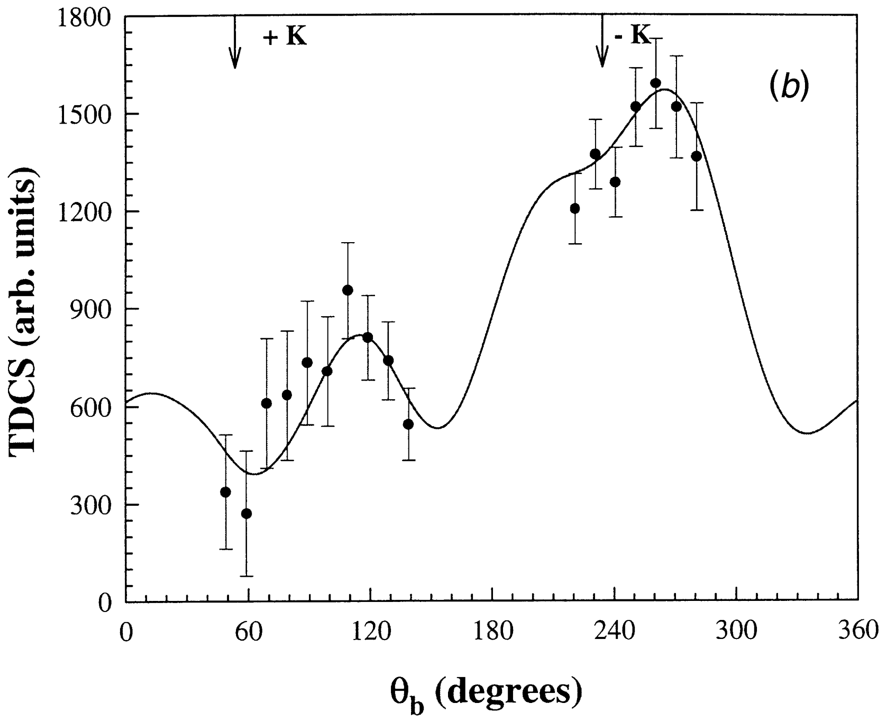


Fig. 1. Relative TDCS for Kr(4p) electron impact ionisation at $E_0 = 924{\cdot}4$ eV, $E_{\rm a} = 880$ eV, $E_{\rm b} = 30$ eV and $\theta_{\rm a} = 5°$. The points are the experimental data; the curve is a DWBA calculation. Arrows indicate the direction of $\boldsymbol{K}$, the momentum transfer, and of $-\boldsymbol{K}$.

For an incident energy of 5719 eV we were not able to perform a DWBA calculation, as the high incident and scattered electron energies mean that too many partial waves are required.

The measurements of the TDCS for ionisation of the argon 2p shell are shown in Fig. 2. In each case, the recoil peak is the dominant feature in the cross section. The ejected electron energy varies from (*a*) 120 eV to (*b*) 50 eV to (*c*) 10 eV. The binding energy for the Ar(2p) shell (fine-structure averaged) is 249 eV, hence the ejected electron energies are much lower than the binding energy, and significant distortion of the ejected electron wave may be expected due to increased interaction with the core and correlation between the outgoing electron and the remaining target electrons in the outer orbitals. The curve in





Figs 2*a* and 2*b*

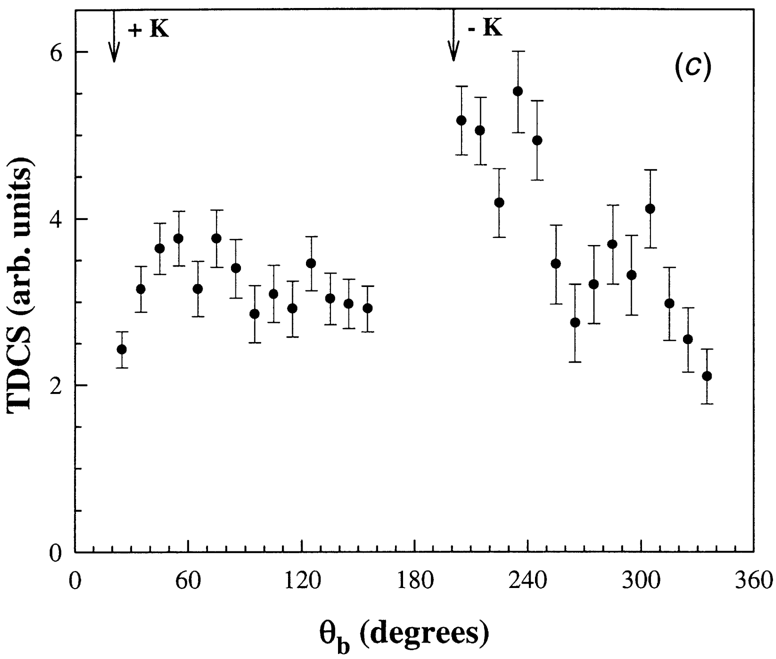


Fig. 2. (a) Relative TDCS for Ar(2p) electron impact ionisation with $E_0 = 1249$ eV, $E_{\rm a} = 880$ eV, $E_{\rm b} = 120$ eV and $\theta_{\rm a} = 15°$. The points are the experimental data; the curve is a DWBA calculation. (b) As for (a) except $E_0 = 1179$ and $E_{\rm b} = 50$ eV. (c) Measured relative TDCS for Ar(2p) electron impact ionisation with $E_0 = 5719$ eV, $E_{\rm a} = 5460$ eV, $E_{\rm b} = 10$ eV and $\theta_{\rm a} = 0{\cdot}5°$.

each graph is the theoretical calculation. In Figs 2a and 2b the DWBA is in good agreement with the experimental cross section, although one must keep in mind that the measurements are relative, and hence have been normalised to the theory to give the best visual fit. Nevertheless, the DWBA does a good job of describing the shape of the cross section, and in particular gives the correct recoil to binary ratio.

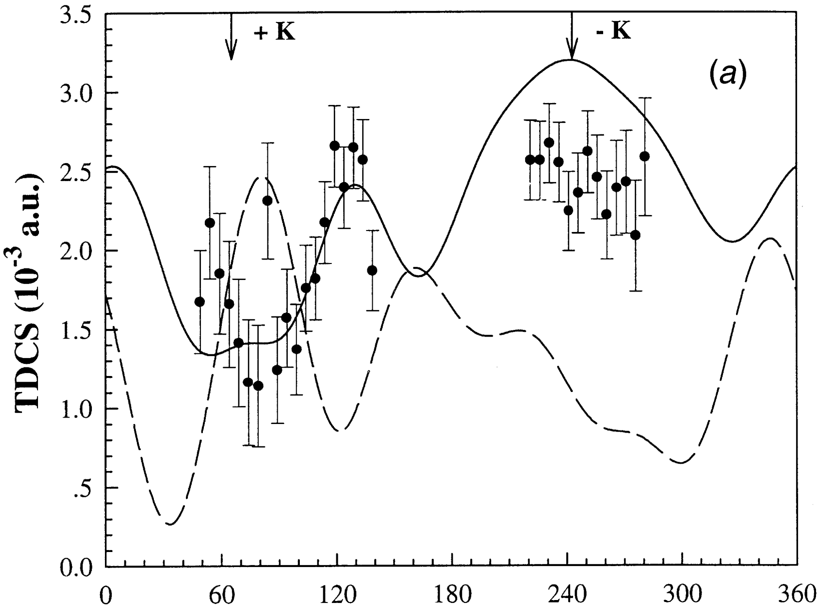
Fig. 2c shows the measured TDCS for an ejected electron energy of 10 eV. The experimental cross section again exhibits a large recoil peak, and there appears to be some structure in both the binary and recoil peaks. The measured distribution is very similar to one obtained by Stefani *et al.* (1986) at an ejected electron energy of 7 eV and an incident energy of ~8 keV, that is, under similar kinematics. This gives added confidence in the reliability of the measurements. The present measurement corresponds to a ratio of 1 : 25 of ejected energy to binding energy, hence one would expect a very strong interaction with the core. In terms of this ratio, the present experiment is equivalent to a helium experiment where the ejected electron would carry only 1 eV kinetic energy. This corresponds to a regime where the DWBA, for example, is quite unsatisfactory in describing the ionisation process (Whelan *et al.* 1993), and hence these highly asymmetric measurements for Ar(2p) ionisation should provide a stringent test of theory.

The good agreement between the DWBA and the Ar(2p) data at lower incident energies, but rather poor agreement of the DWBA with the Xe(4d) data of

Avaldi *et al.* (1993) prompted us to investigate inner shell ionisation in a target of intermediate size, i.e. krypton. In Figs 3*a* and 3*b* we present measurements of the TDCS for Kr(3d) ionisation (binding energy of $94\cdot4$ eV, averaged over the $3d_{3/2}$ and $3d_{5/2}$ levels). In these plots, the data have been normalised to the theory, rather than the theory being normalised to the data. For the krypton measurements a normalisation between the two data sets has been experimentally determined (Cavanagh and Lohmann 1998). Thus the relative magnitude of the two cross sections is fixed, and they cannot be independently normalised to the theoretical calculation; instead the normalisation to the theory is such that the best visual fit across *both* data sets is obtained.

It is immediately apparent that the recoil peak is not as large compared with the binary peak as was the case in argon. The recoil to binary ratio in argon ranges from $1\cdot5$–$2\cdot2$, while for krypton it is closer to $1:1$. The fact that we are ionising from a *d* orbital rather than a *p* orbital may be responsible for this difference. The TDCS in Fig. 3*a* was measured under conditions corresponding to bound Bethe ridge kinematics, and thus we have included a DWIA calculation in the plot (dashed curve). There is essentially no agreement between the theory and the experiment. This is perhaps not too surprising as the incident energy in these experiments is relatively low compared with the binding energy, and we may be seeing a breakdown of the factorisation approximation. The solid curve in Figs 3*a* and 3*b* is the DWBA calculation. For the lower ejected electron energy of 50 eV (Fig. 3*b*) the calculation is in good agreement with the experimental results. At an ejected energy of 72 eV (Fig. 3*a*), the agreement is somewhat poorer, with the theory overestimating the recoil peak slightly, and predicting a minimum in the cross section near 50°, while the experimental cross section appears to be heading for a maximum.

One effect which is not included in the DWBA, but which may affect the shape of the TDCS (Avaldi *et al.* 1993), is the influence of the Auger electrons emitted after the inner shell ionisation. In order to investigate this effect, the data in Fig. 3*b* were measured at an ejected electron energy (50 eV), which is just below the $M_{4,5}$–$N_{2,3}N_{2,3}$ manifold of Kr Auger lines which extend from 51 to 57 eV (Fig. 4). Thus one might expect to see an influence of these Auger electrons on the angular distribution of the ejected electrons. The good agreement seen between the DWBA and the data in Fig. 3*b* suggests that such effects are negligible. To emphasise any impact on the cross section from the Auger electrons, we have also performed a measurement of the TDCS for an ejected electron energy of $52\cdot65$ eV, which is within the energy range spanned by the $M_{4,5}$–$N_{2,3}N_{2,3}$ manifold of Auger lines. The large number of Auger electrons entering the ejected electron analyser (which has an energy resolution of about $1\cdot5$ eV FWHM) makes the experiment more difficult as it increases the background, however we have obtained a preliminary measurement of the TDCS. The results are shown in Fig. 5, where they are again compared with the DWBA calculation. There appears to be essentially no impact on the *shape* of the cross section due to the Auger electrons, as shown by the good agreement between theory and experiment. Note that the measured TDCS in this case includes true coincidences between the scattered electron and the Auger electrons emitted after the decay of the ion, hence our only means of determining the contribution of the Auger electrons is to compare the measured distribution with the DWBA calculation, which does *not* take into account the presence of the Auger electron.



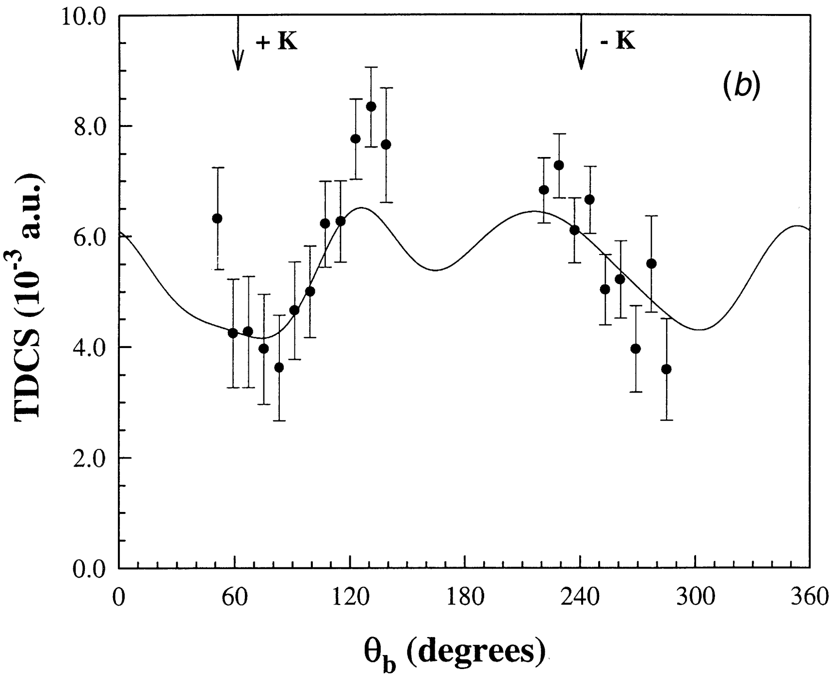


Fig. 3. (a) TDCS for Kr(3d) electron impact ionisation with $E_0 = 1046 \cdot 4$ eV, $E_a = 880$ eV, $E_b = 72$ eV and $\theta_a = 15°$. The points are the experimental results; the solid curve is a DWBA calculation and the dashed curve is a DWIA calculation. See text for details of the normalisation. (b) As for (a) except $E_0 = 1024 \cdot 4$, $E_b = 50$ eV and $\theta_a = 10°$.

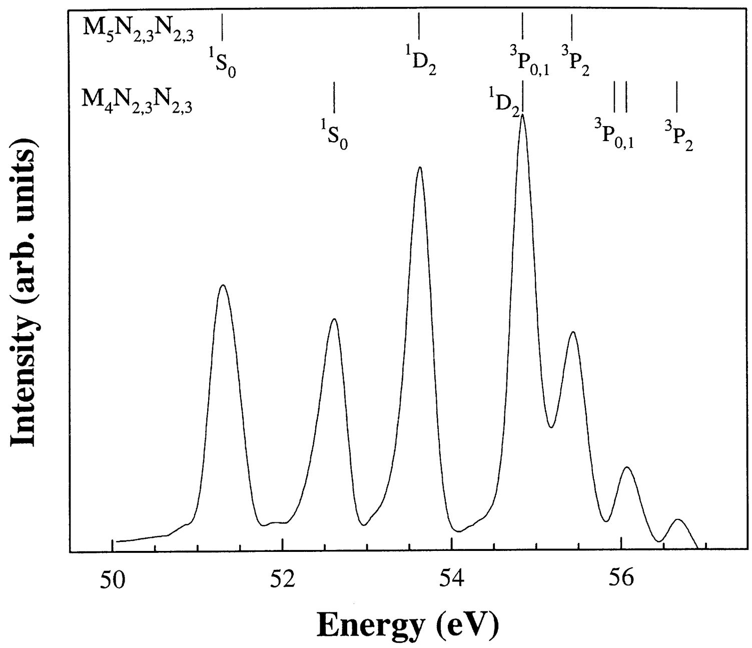


Fig. 4. Measured (non-coincidence) intensity of the $M_{4,5}$–$N_{2,3}N_{2,3}$ Auger lines in krypton.

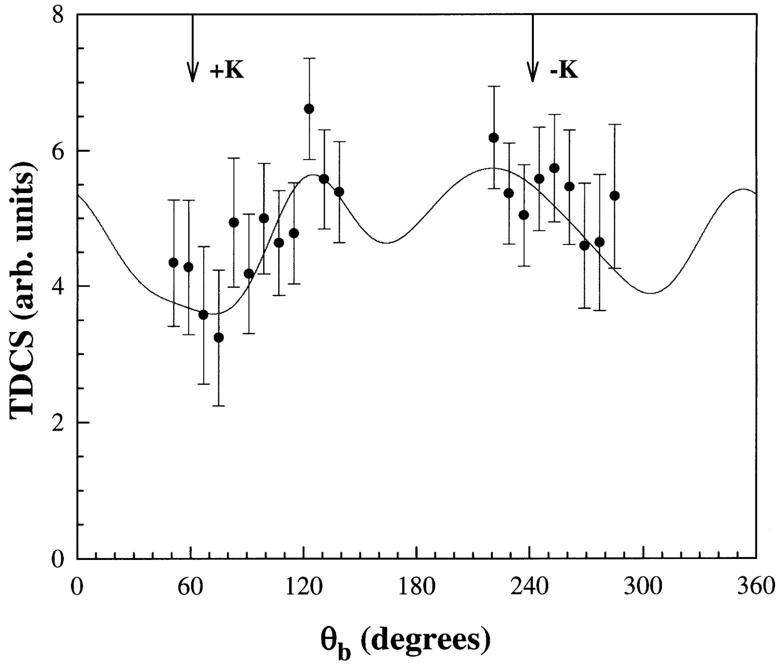


Fig. 5. Preliminary measurement of the TDCS for Kr(3d) electron impact ionisation with $E_0 = 1027 \cdot 05$ eV, $E_a = 880$ eV, $E_b = 52 \cdot 65$ eV and $\theta_a = 10°$. The points are the experimental data; the curve is a DWBA calculation.

4. Conclusions

Recent (e, 2e) measurements for inner shell ionisation of argon and krypton have revealed a rich structure in the triple differential cross section, reflecting interferences between the quantum mechanical amplitudes for various scattering processes. The distorted wave Born approximation has been shown to provide a good representation of the data for incident electron energies of approximately 1 keV, and ejected electron energies down to 50 eV. Results obtained in very asymmetric conditions (incident energy 5 keV and ejected energy 10 eV) show similar features to the triple differential cross section measured at lower incident energies; however, we await a suitable calculation for comparison with the data. The measurements should provide an interesting challenge for current theories.

Acknowledgments

The authors would like to thank Professor Ian McCarthy for providing the DWBA code. BL would like to thank the Université de Paris XI for the opportunity to visit as a Professeur Invité, and the Australian Academy of Sciences for the award of a Bede Morris Fellowship.

The work at Griffith University was supported by the Australian Research Council.

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Manuscript received 17 December 1997, accepted 11 March 1998