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The (e,2e) spectroscopy, achievements and perspectives

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Abstract Electron-electron coincidence experiments have experienced over the past twenty years an increasingly rapid expansion and application to many different fields. Among them, the (e,2e) spectroscopy has been successfully used to investigate either the target electronic structure or the ionisation dynamics. The capability of this spectroscopy to investigate single particle properties in atoms, molecules and solids is discussed by the help of some recent results. Correlation effects in bound states and in the continuum have been the subject for several recent (e,2e) investigations. Single and double ionisation coincidence experiments are reviewed focusing the attention on correlation effects. The interference between direct and resonant ionisation channels has been also the objective of recent electron coincidence experiments. Perspective and developments of the (e,2e) spectroscopy are briefly discussed.

1. INTRODUCTION

The ionisation of atoms, molecules and solids by electron impact is one of the most significant processes in the field of interaction of charged particles with matter. These collisions play a significant role in many situations of current interest, especially in the chemistry and physics of plasmas, gas laser media, thin film deposition and radiation chemistry. The investigation of ionisation processes started in the early '30 and since then it has continuously attracted the interest of both experimentalists and theorists (1). In spite of all these efforts, ionisation by electron impact is still one of the most demanding processes to be properly described as far as the collision dynamics is concerned. Ionisation experiments are usually performed detecting and analysing only one of the final unbound particles of the reaction $e^- + A \rightarrow A^+ + e^- + e^-$. This way the finer details in the ionisation cross section are averaged out by integration over the dynamical variables of the undetected particles.

In recent years coincidence experiments, usually abbreviated (e,2e), have allowed to completely determine the kinematics of the ionisation process, giving the largest information possible from the quantum mechanical point of view. This implies a serious challenge to theory. The experiments were firstly started in Rome and Kaiserslautern more than twenty years ago (2,3). Since then they have been applied to perform spectroscopic and dynamical studies in a variety of systems and energy domains, from the threshold ionisation of He to the core ionisation of heavy atoms at relativistic energies. Briefly, the experiment consists of measuring simultaneously the energy E_0 of the incident electron, the energies E_1 and E_2 of the two final electrons and the probability of them being emitted into solid angles $\Omega_1(\theta_1, \phi_1)$ and $\Omega_2(\theta_2, \phi_2)$, i.e. the triple differential cross section (TDCS). An electron colliding with a target exchanges a continuum of energy and momentum, ranging from vanishing momentum transfer (dipolar limit), to the case in which the momentum is totally transferred to an individual target electron (binary limit) (4). By choosing the energy loss E and the momentum transfer \mathbf{k} , a large variety of kinematics can be selected. This allows the spectroscopic or the dynamical information to be enhanced or diminished in the cross section. The coincidence technique offers both the opportunity to study in detail the collision dynamics of the ionisation process and the possibility of a powerful analytical tool to probe the electronic structure of the target. Recent achievements and perspectives in this field will be discussed by the help of few selected examples that identify common denominators to the large variety of experiments performed in the past few years. Among the others, the following topics will be addressed in the discussion: single particle and many body properties, electron correlation in the target and short/long range correlation in the continuum of the collisional process.

One might argue whether it is worth spending a substantial experimental effort in studying a break-up reaction, like the ionisation, of a quantum system whose Hamiltonian is known. The answer is positive, because the study is relevant to the Coulomb n -body problem, that is not exactly solvable not even for n as small as 3. Hence comes the necessity for developing and testing approximated solutions to the problem (5).

The simplest approximation to the triple differential cross section for the fast electron impact ionisation is the first order interaction for the reaction mechanism and the single particle description for the target structure (6)). Within this approximation and for a plane wave description of the fast electrons, the cross section factorizes in a kinematical term times a form factor that is fully related to the structure of the target:

$$\frac{d^3\sigma}{d\Omega_1 d\Omega_2 dE} = \frac{k_2 k_1}{k_0} |F|^2 \quad (1)$$

The interpretation of the form factor becomes particularly simple in two limiting conditions: the dipolar limit, at vanishing momentum transfer \mathbf{k} , and the impulsive limit, at high momentum transfer and kinetic energy of the ejected electron of the order $k_2^2 \sim E_2$. The last condition

corresponds to the Bethe ridge condition, where the electron-atom interaction reduces to a two-body electron-electron interaction.

In the dipolar limit

$$|F|^2 = \frac{4}{k^4} \left\langle \Psi_{f(\mathbf{k}_2, z-1)} \left| \sum_i e^{i\mathbf{k} \cdot \mathbf{r}_i} \right| \Psi_{i(z)} \right\rangle^2 \quad (2)$$

and the process reduces to a pseudo-photoionisation of the target. The fast electron is scattered at small angle, with small energy loss, and one of the target electrons is ejected, with much lower energy, in two opposite lobes (binary and recoil) along the direction of the momentum transfer. The ratio of intensities of these lobes approaches the unity as $k \rightarrow 0$, so that the angular distribution of the ejected electron around \mathbf{k} equals the distribution of the photoelectron around the electric field in photoionisation. Like in that case, it is possible to define an asymmetry parameter β .

In the second limiting case, because of the rapid oscillation of the exponential operator $\exp(i\mathbf{k} \cdot \mathbf{r})$, the matrix element is non vanishing only if in the final state the wave number of the free ejected electron is comparable to k (7). It means that the first order interaction furtherly reduces to a two body interaction and

$$|F|^2 = \frac{4}{k^4} \left\langle \Psi_{f(z-1)} \left| e^{i\mathbf{q} \cdot \mathbf{r}} \right| \Psi_{i(z)} \right\rangle^2 \quad (3)$$

where $\mathbf{q} = \mathbf{k}_0 - \mathbf{k}_1 - \mathbf{k}_2$ and $\Psi_{f(z-1)}$ is the final ionic state. Therefore, if the single particle description holds, the form factor becomes the squared modulus of the Fourier transform of the wave function of the initial bound state, i.e. the electron momentum density. Upon validity of these conditions (impulse approximation) the (e,2e) angular distribution, which is now confined into a single lobe (binary), realises a unique electron momentum spectroscopy (EMS). Let's start discussing that class of experiments

2. ELECTRON MOMENTUM SPECTROSCOPY

The possibility of extracting electron momentum densities from the (e,2e) TDCS, upon validity of the impulse approximation, was firstly verified in symmetric, coplanar and non coplanar, geometry (8,9) and then extended to asymmetric kinematics that satisfies the Bethe ridge condition (10). By now EMS has been applied to the study of a large variety of atoms and molecules, for outer and inner orbitals (6). The capability to discriminate among different quality wave functions has also been proved, particularly in the case of bonding molecular orbitals. For what concerns the application to solid targets, even though the first EMS experiment was performed on an amorphous carbon thin film (8), it is only recently (11,12,13) that it has been applied to the study of bands in solids. In particular, new multichannel spectrometers and improved energy resolution, recently achieved, have allowed for a complete

reconstruction of the band structure in amorphous grafitic carbon (14). The presence of sp^2 hybridisation accompanying the more usual sp^3 , was evidenced by these measurements. For completeness, two recent applications of (e,2e) on solids are still to be mentioned. The first one is an experiment performed in highly asymmetric kinematics (15), again on amorphous carbon thin film, where the experimental results have been interpreted in terms of pseudo-photoionisation and contributions from plasmons decay into electron pairs have been evidenced. The second experiment (16) was performed in reflection kinematics, on a target of W(100). It was found that events of emission of two coincident electrons exist. They require a certain threshold energy of the primary electron which is not equal to twice the work function of the sample surface.

There is an aspect of the (e,2e) technique, which is particularly relevant to the EMS. It is related to the finite experimental acceptances, that influence sensibly the measurement of the electron momentum distribution close to zero and might mask structural effects. Although in the first EMS experiment this effect had been pointed out and taken into account in comparing the measured and calculated momentum distributions (8), over the years this problem had attracted marginal attention. Recently it has been reconsidered in the attempt to explain an unexpected result on the outer orbital of the H_2O molecule. Accurate numerical simulation of the effect of the finite target size and of the experimental acceptances has removed the apparent discrepancy between calculated and measured momentum distribution (17).

It has to be finally pointed out that electron momentum distributions can be obtained, with good confidence level, up to a momentum value of the order of 2 a.u. Above, the factorisation approximation is questionable and further interactions with the target are no more negligible (18). The largest momentum allowable for validity of the EMS reconstruction is still a debated issue.

One of the most attractive perspectives for EMS is the possibility of measuring the momentum density for oriented molecules. The potentiality of such an experiment was pointed out already in 1979 (19), but up to now not a single experiment has been performed where the momentum distribution of a spatially oriented molecule was measured. A possible experimental approach to get oriented molecules is represented by the interaction with a surface and, indeed, electron impact experiments on adsorbed molecules have recently started (20). On the contrary, the alignment of atoms with a laser beam is quite an established technique in spectroscopy (21). EMS on laser excited states allows to explore in the momentum space normally unoccupied orbitals. By this method the momentum distributions of the 3p excited states of Na and Cd have been measured (22). At present, however, the very low target densities, and high dilutions, achievable seem to be a severe limitation for coincidence experiments.

The subject of momentum spectroscopy can not be left without mentioning the present efforts toward measuring inner shell momentum distributions. Bharathi et al. (23) have recently measured molecular inner shell momentum distributions in a number of targets. The peculiarity of core momentum distributions is that they extend to large momenta, in a region where the interference between neighboring atoms shows up becoming a relevant feature of the

measured cross section. A further open field of investigation concerns the possibility of measuring momentum distributions of inner orbitals of heavy atoms at relativistic energies. The subject has been recently reviewed by Nakel (24). His work has shown that, at least for the experiments performed up to date, there are still dynamical effects not properly taken into account by the theories, that mask the structural information.

2.1. EMS and electron correlation

The independent particle model is the approximation most widely used in describing the bound states of quantum systems, the ionisation process and is also the basis for the interpretation of most of the spectroscopies used to investigate electronic structure of atoms, molecules and solids. Nevertheless, it was soon realised that also valuable information on the many body properties of the target can be deduced by these experiments. The main effect shows up as the presence of "satellite" peaks in the $(e,2e)$ energy separation spectrum. These peaks correspond to leaving the residual ion with at least one electron excited, which means that more than one electron has been involved in the ionisation process. Bound state electron correlations are responsible for these effects, in addition to the non orthogonality of the initial and final state. Electron correlations, often referred to as internal state correlations (ISC), can be shortly defined as whatever is not accounted for by the independent particle approximation. Radial electron-electron correlations can still be described by self consistent field treatments, whereas angular correlations require the use of wave functions fully dependent on the relative electron positions or can be taken into account by Configuration Interaction (CI) methods. He and Ar are classical examples to be referred. In He correlations are present only in the initial neutral state and are responsible for the population of ionic states with the residual electron excited to the levels $n>1$. The states $n=2$, $n=3$ have been clearly observed in the $(e,2e)$ energy separation spectra, in addition to the main peak. The electron momentum distributions for these satellites have been measured in impulsive conditions and have been found to differ from each other and from the main peak distribution (6,25). Comparison with the calculated cross sections allows for discriminating among different correlated wave functions. Only good quality wave functions reproduce the measured momentum profiles. In the spectrum of Ar, on the contrary, the satellite peaks can be ascribed either to initial or to final (ion) state correlations. The measure of the momentum distribution, in that case, has shown to be capable of discriminating between the different origins(26).

It has been pointed out that direct double ionisation experiments, using the coincidence techniques $(e,3e)$ or $(h\nu,2e)$, give a more direct insight on bound state correlations. Indeed that process can take place only in correlated systems and the cross section vanishes when uncorrelated wave functions are used for the target. Neudatchin et al. (27) proposed these experiments and shown that in the high energy limit the cross section factorises, similarly to the binary $(e,2e)$ case, allowing the direct determination of the spectral density of the Green's two particle function and of the Fourier amplitude of the e-e pair correlator. Such high energy

double ionisation experiments have not been realised yet, even though their feasibility at low energy transfer has been already shown for both $(e,3e)$ (28) and $(h\nu,2e)$ experiments (29).

It is worth mentioning an interesting problem that has been raised up, i.e. whether the satellite spectrum measured by $(e,2e)$ is identical or not to that measured by photoemission(30). On the basis of the reducibility of the electron scattering operator to the dipole operator, in the limit of vanishing momentum transfer, this equivalence should be expected. The experimental evidence for such an equivalence has not been established yet (30) and more experiments would be desirable. However, in the binary regime, the $(e,2e)$ technique can select q -values of the bound electron momentum close to zero, which are never accessed by the photoemission spectroscopy. Amusia (31) has discussed this subject and has shown that the interaction terms contributing to the impulsive $(e,2e)$ are different from those contributing to the photoemission cross section. In particular in his picture, only the $(e,2e)$ technique can measure the spectroscopic factors and the full equivalence of the energy spectra is excluded.

3. INTERACTION MECHANISM

The $(e,2e)$ experiments have shown up, since the early stage, as a powerful tool to investigate the reaction mechanism in electron impact ionisation. The study was initially confined to high incident energies, in order to apply first Born theories, but it appeared quite soon that the scheme of first order interactions, plane waves and one electron wave functions for the target was too simplified and could rarely interpret in a satisfactory way the outcomings of the experiments. The measured cross sections were found to exhibit systematic deviations from the calculated ones, consisting in angular shifts, higher ratios of the recoil to binary lobe intensity and the presence of minima and structures in the angular distributions. Also the absolute values of the cross section, when measured, were often found to be in disagreement with the predicted values. These findings have quite soon attracted the attention of theorists, because it was evident that fast electron collisions with atoms were not at all a well understood subject and that the $(e,2e)$ experiments were relevant to the unsolved Coulomb three body problem, with the further complication that one of the three particles has internal structure. That requires to take into account: i) distortion of the free particles wave functions in the region close to nucleus, due to the atom/ion potential; ii) effects of the electron-electron correlations that can be important either in the bound state of the target and in the continuum state of the final electrons. Because of the mutual repulsion experienced up to infinity, the electrons can not be considered as independent particles. This aspect of the collision has been treated in turn with a semiclassical approach, basically leading to a modification of the electron trajectories, or quantum-mechanically allowing the three body Coulomb wave function in the continuum state to have the correct asymptotic behaviour.

Many different calculations have been developed, emphasising one or more of the collisional features of the ionisation process. He and H have been very often considered as targets, for the easy handling of calculations and simplicity in the interpretation of results, although the

effects on the cross section are often observed to be larger for many-electron atoms. As already mentioned in the introduction, in the early stage of development of the (e,2e) experiments the attention was mostly focused on the asymmetric kinematics close to dipolar conditions, whereas symmetric kinematics were mostly applied to perform EMS. During the years the (e,2e) technique has continuously extended to the study of the dynamics of ionisation down to energies close to the Wannier threshold region. An overview of these studies is given in the following, keeping in mind that detailed discussions of the recent achievements are provided by the individual authors' contributions to this conference.

3.1 Intermediate-high energy (e,2e)

The knowledge of the single ionisation mechanism of outer shell electrons from light atoms at intermediate-high energies (i.e. above 4 times the threshold) is based upon a large body of measurements, on various targets and in various kinematics (32,33,6) and is essentially complete. From this point of view it is interesting the survey of the interaction mechanism given in the paper by Rosel et al. (34). The energy range covered is large and the evolution of the TDCS in coplanar symmetric conditions is followed continuously from the impulsive limit to the near threshold region.

It can be said that the ionisation process can be quite well described, if the kinematics is confined to asymmetric coplanar conditions and if terms higher than the first in the interaction potentials are included. This has been done with different approaches. Second Born approximations, (35,36) have been introduced, where the interaction of the fast electron with the bound electron and with the core is taken at the second order and the slow electron wavefunction is solution of the atomic hamiltonian in the bound and in the continuum state. Distorted wave Born approximations, DWBA, (18,37), use the atomic/ionic potential to calculate the free electron wave functions, thus including the effects of the electron-core interactions, and limit the electron-electron terms to the first order. Distorted wave impulse approximation (6), DWIA, consider the electron-electron interaction to all orders but implies the factorisation approximation, which is no more valid for low momentum transfer. Therefore its application has to be limited to kinematics close to the Bethe ridge conditions. On the simpler targets, H and He, all the mentioned models have shown to be able to satisfactorily describe the experiments. while for more complex targets only distorted wave calculations are feasible. The common aim of the models is to take into account the short range multiple scattering terms in the inter-particle potentials of the three body system. This is done to the expense of the exact asymptotic behaviour of the wave function, which is determined by the long range part of the Coulomb potential. The importance of dynamic Coulomb correlation in the continuum, often addressed as continuum final state correlation (CFSC), was firstly suggested (38) to explain the reduction in the TDCS systematically measured at the lower angles in coplanar symmetric (e,2e) experiments. More rigorous treatments of these effects were made for asymmetric kinematics and/or lower incident energies (39,40). Wave functions, that satisfy exact boundary

conditions for the three body Coulomb continuum state of electron impact ionisation, were used. A very good agreement was obtained with cross section data on hydrogen at medium energies. The model has, however, inherent disadvantages. The applicability is limited in practice to very simple targets. Moreover, the wave functions are now not as good at r_1, r_2 small, so that, when the short range effects of the multiple scattering terms of the potentials are preminent, the theory is expected to fail. A model that could merge the short range and long range Coulomb interactions is desirable. In fig. 1 absolute TDCS on He are reported, measured in asymmetric kinematics at incident energy of 300 eV (41). The experiment is compared with a DWBA calculation and shows a remarkable overall agreement everywhere but at the smaller angles of ejection of the slow electron. The agreement at small angle improves when raising the energy of the free electrons. This finding is persistent throughout the various kinematics investigated and for different targets (42,43), and indicates an insufficient description of the long range e-e interaction. How to incorporate the exact boundary conditions for the wave functions in the distorted wave Born approximation is one of the current subjects of investigation.

An interesting study has been carried out on the satellite peak $n=2$ of He, measuring the TDCS in asymmetric conditions. In identical kinematics, the TDCS of the main $n=1$ peak has also been measured, in order to evidetiate target and continuum state effects (44,45). In He ISC are present only in the initial state, which is the same for both peaks, while the final states can be affected by CFSC only. The TDCS measured for the two peaks have shown striking differences in the angular dependence. The dynamical effects, that are typically observed in these measurements, as modification of the ratio between the intensities of the binary and recoil lobes or as angular shifts in the low energy electron distributions, are larger in the ionisation excitation process than in the pure ionisation. A similar study was performed by Dupre' et al. at higher energy (46). In this latter experiment the absolute value of the cross section was also determined. Four different theoretical approaches have been used to predict these cross sections: i) a first Born orthogonalised Coulomb wave model (OCW), where the final state is characterised by an effective charge, which is fixed to the asymptotic value ($Z=1$) for the main peak and used as an adjustable parameter ($1 < Z < 2$) for the $n=2$ peak; ii) a first Born multichannel model, where the R-matrix techniques are used to evaluate the initial and final target eigenstates; iii) a calculation (47) where the final state wave function describing the free electrons properly accounts for the asymptotic behaviour); iv) a parametric potential method (48), that adopts a description of the ejected electrons by distorted waves obtained from channel dependent parametric potentials. All these calculations are carried out in first Born approximation. The overall result is that, even when the $n=1$ TDCS is well described, the cross section for the process of ionisation accompanied by excitation ($n=2$ satellite) is only qualitatively predicted. Its shape changes with the relative amount of p-type and s-type correlation present in the He wave function and is extremely sensitive to the wave function used to describe the continuum. Once more the $(e,2e)$ cross section appear to be a fine tool to

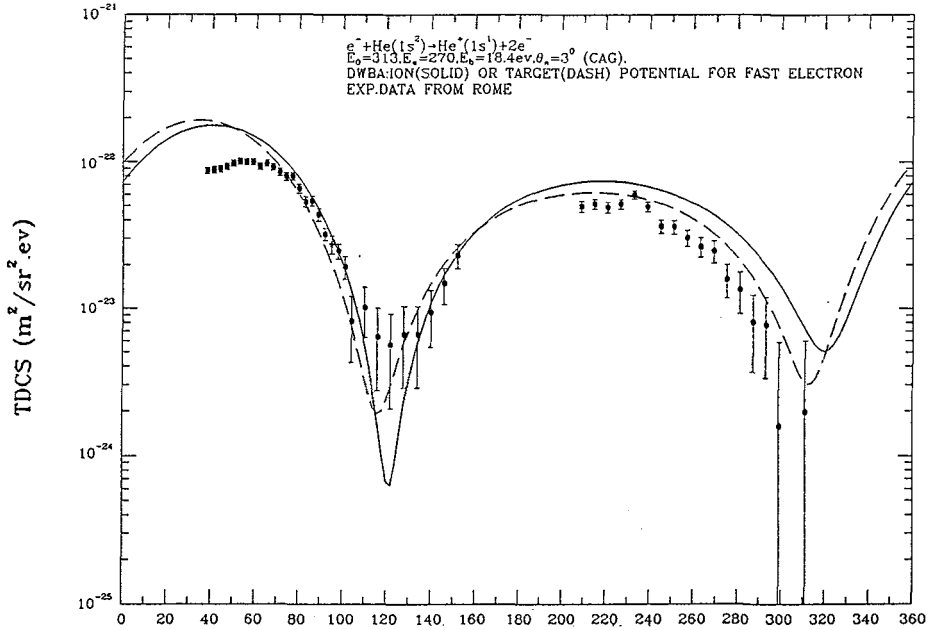


Figure 1A: Absolute triple differential cross section of He at $E_1=270$ eV, $E_2=18.4$ eV and $\theta_1=3^\circ$. The solid and dashed lines are DWBA with the distorted wave of the scattered electron generated in the ion and target potential, respectively.

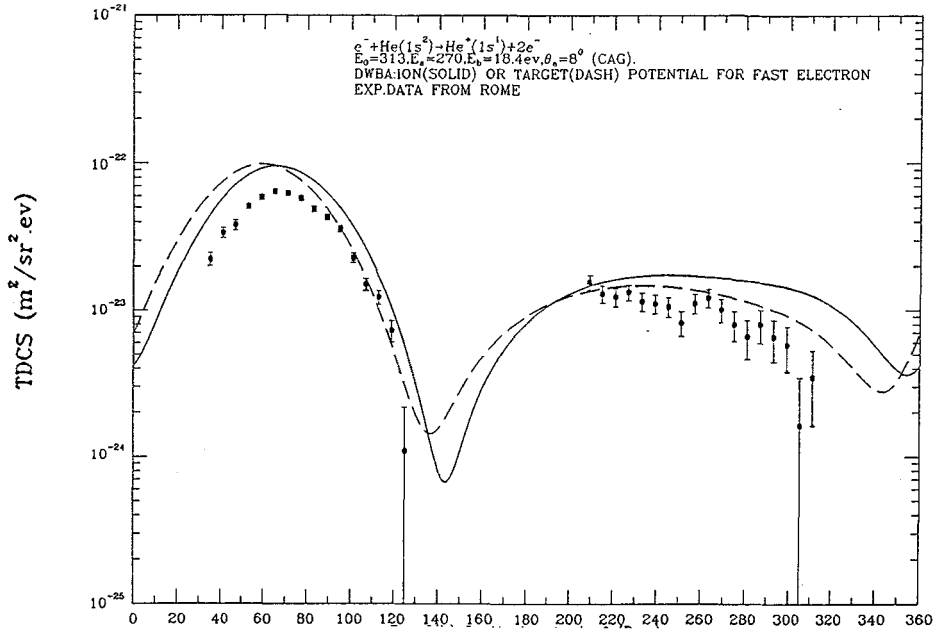


Figure 1B: Absolute triple differential cross section of He at $E_1=270$ eV, $E_2=18.4$ eV and $\theta_1=8^\circ$. The solid and dashed lines are DWBA with the distorted wave of the scattered electron generated in the ion and target potential, respectively.

probe the static electron correlations in the target and the dynamical correlations in the continuum.

A good fraction of the capability of the TDCS to discriminate between different models, even when they reproduce the shape of the measured distribution, comes from the knowledge of the absolute cross section. Various different experimental methods have been developed over the years ⁽³²⁾. Most of the TDCS are brought to the absolute scale by normalisation procedures, based on the assumption that the double or triple generalised oscillator strength (GOS) converges to the optical limit at vanishing momentum transfer (Lassetre theorem). Some caution has to be adopted when applying this method. First of all these procedures are reliable provided that enough experimental points are obtainable by the experiment in the region where the convergence is achieved. Furthermore, it has been recently suggested ⁽⁴⁹⁾ that the assumption of the convergence of the GOS to the optical limit may not be valid. The analysis is based on a parametrisation of the multiply differential cross section ⁽⁵⁰⁾, and has shown that for the ionisation of He at 8 KeV, even for vanishing scattering angles, the contributions to the GOS of terms beyond the dipolar one can not be neglected

3.2 Backward scattering

According to simple first Born models the coplanar symmetric TDCS should be negligible at very large scattering angles (backward kinematics). Early experiments by Pochat et al. ⁽⁵¹⁾ had shown evidence of a relative maximum at large scattering angles and a sharp minimum around 80°, already predicted by a second Born calculation ⁽⁵²⁾. This feature is interpreted as due to a collision of the fast electron with the nucleus characterised by large momentum transfer, which is followed or preceded by the collision with the ejected electron. More recent absolute measurements, performed at Brest ⁽⁵³⁾ and Kaiserslautern ⁽⁵⁴⁾ with better angular resolution, have pointed out that, at medium-high impact energies, the relative maximum at large scattering angle and the sharp minimum are quite well reproduced by a DWBA calculation, both in symmetric and energy sharing conditions. It is interesting to note that, while in the second Born calculation the angular minimum comes from an interference effect of the electron-electron term and of the electron-nucleus term, in the DWBA it is due to the combined low efficiency of the two contributions.

3.3 . Inner shell ionisation

For complex targets only the distorted wave approach has shown to be feasible. Therefore, the DWBA has been used to calculate the TDCS for inner shells. Apart from the experiments mentioned in the EMS chapter, devoted to measure momentum densities, experiments on inner orbitals have been performed by the Orsay ⁽⁵⁵⁾, Rome ⁽⁵⁶⁾ and Wurzburg ⁽⁵⁷⁾ groups, mostly on the rare gases, but also on few molecules. The calculations in DWBA, DWIA and distorted Coulomb ⁽⁵⁸⁾ approximation are quite good, particularly for the L-shell of Ar and the K-shell of Ne⁽⁵⁹⁾. All of these inner shell cross sections are characterised by a transfer of intensity from

the binary to the recoil lobe and by a bending of the recoil symmetry axis which is opposite to the one observed in outer shell ionisation. A double collision picture has been introduced to explain the peculiarities of these features, based on the fact that for inner orbitals the collision takes place closer to the nucleus and then the scattering of the fast electron with the atomic core becomes more important. Another example of experiment from which fine information on the ionisation dynamics is extracted, is that carried out on Xe at energy losses capable to excite the giant resonance in the 4d ionisation. The resonance is originated from the trapping of the f partial wave of the ejected electron in the centrifugal barrier of the experienced potential. It is well known that in the same region the ionisation cross sections of the 5p and 5s electrons show a similar, although less pronounced, enhancement, explained via an interchannel coupling mechanism, where the 4d electron is virtually excited and then decays leaving a hole in the shell $n=5$. The electron ejected has the same kinetic energy of the direct process so that the two contributions are energetically indistinguishable. In this region of energy the electron-electron angular distributions have been measured in coplanar asymmetric conditions both for the ionisation of the 4d orbital and the 5p (⁶⁰), fig. 2. The calculated DWBA cross section is in reasonably good agreement with the results obtained for the 4d electron, in the various kinematics. It is also good for the 5p distribution measured in impulsive condition. A clear discrepancy is observed however in the dipolar case, although the two experiments, on the 4d and 5p electrons, were conducted in similar conditions, differing only for the kinetic energy of the ejected electron which is higher in the latter case. The calculations, performed in DWBA, are accurate. Distortion of the free electrons is taken into account both in the initial and the final channel, polarisation and exchange are included and the resonant contribution of the f-wave is insured by the partial wave expansion. Only the long range interaction between the final electrons is not explicitly treated. What is however completely out of the model is the interchannel coupling mechanism. It is reasonable to expect some effect of it in the 5p angular distribution measured at small momentum transfer, that in fact exhibits features (like intense structured recoil) present also in the correspondent 4d distribution, not reproduced by the calculation. These many body effects disappear when the momentum transfer increases, as the TDCS is now mostly determined by the single particle properties of the 5p orbital. Once more there is evidence that the (e,2e) experiments can be tuned to enhance single particle and many body properties, by selecting the kinematics.

3.4 Relativistic (e,2e)

(e,2e) experiments on relativistic orbitals at relativistic energies have been performed by the Tübingen experimental group and have been recently revised in a paper by Nakel (²⁴), to whom we refer for a detailed discussion of the subject. The results achieved up to now can be summarised in the following way. Although good relativistic bound state wave functions are used, it is not yet possible to describe correctly the TDCS and the dynamics of the process. Several types of distorted wave and Coulomb Wave Born approximations have been tried,

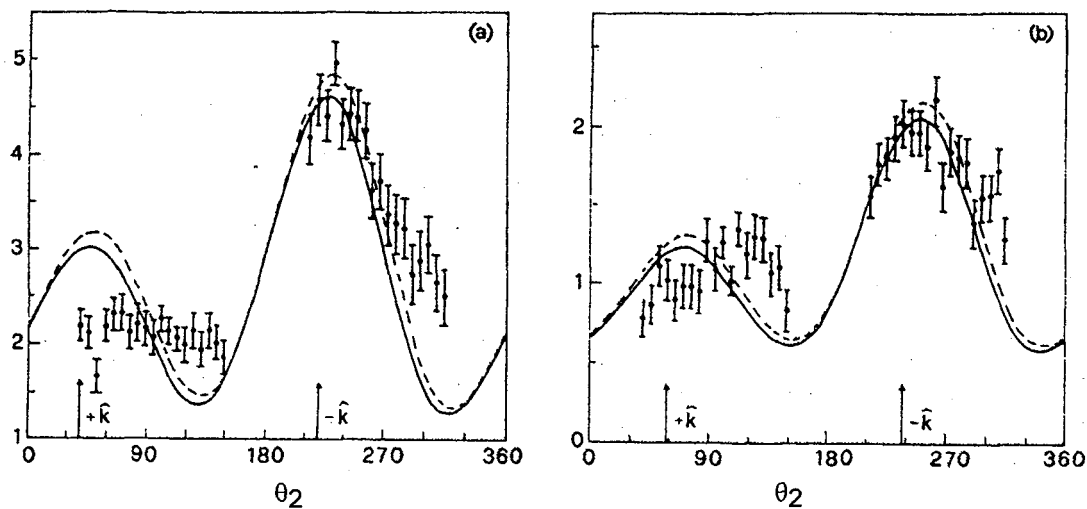


Figure 2A: Relative triple-differential cross sections of Xe 4d at $E_1=1000$ eV, $E_2=20$ eV and $\theta_1=2^\circ$ (a), 4° (b). The solid and dashed lines are DWBA calculations with the scattered-electron wave function calculated in the target or ion potential, respectively.

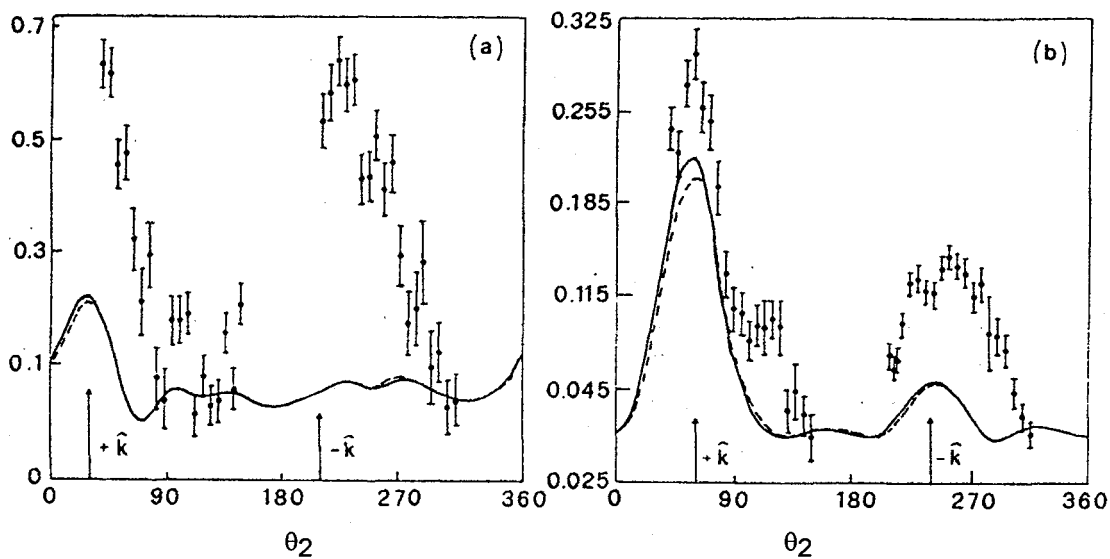


Figure 2B: Relative triple-differential cross sections of Xe 5p at $E_1=1000$ eV, $E_2=80$ eV and $\theta_1=1.5^\circ$ (a), 5° (b). The solid and dashed lines are DWBA calculations with the scattered-electron wave function calculated in the target or ion potential, respectively.

which are able to describe the shape of the TDCS, but badly fail in predicting the order of magnitude of the cross section. Walters et al. (61) have compared absolute TDCS for K-shell ionisation of gold, silver and copper with a number of simple first order approximations. The theoretical calculation shows that spin flip effects, neglected in previous work, are important in symmetric geometry. A simple first order interaction model has also been applied (62). It uses relativistic eikonal impulse approximation to describe the short range interaction and a semiclassical non relativistic correction for the long range electron-electron interaction. This work enlightens that final state interactions, that represent the relevant effect in coplanar symmetric non relativistic experiments, are less important in deep core ionisation. The distortion of the continuum wavefunction due to the interaction with the atomic potential, seems to be the most relevant effect.

3.5 Auger decay

The Auger decay has also been matter of investigation with the electron coincidence technique. All the final unbound electrons should be detected in the complete experiment by a triple coincidence. Such experiments, that belong to the general class of the (e,3e) experiments, are not yet feasible, due to the very low cross section associated with inner shell ionisation. However, due to the fast progresses that are being made in developing high luminosity multi-coincidence apparatuses, the minimum detectable cross section value is continuously decreasing and they could become feasible in a near future. Up to date experiments have been performed where only the scattered electron-Auger electron pair is detected (e,e'Auger). An example for application of a multichannel coincidence spectrometer to (e,e'Auger) experiments (144 independent coincidence channels) is the investigation of the angular dependence of the post collisional energy shift of the Ar L₃MM(¹D₂) Auger transition (63). The high luminosity of the spectrometer has allowed to measure the variation of the Auger transition energy as a function of the relative angle between the slow ejected electron and the Auger electron. The same effect has been recently observed also by Lohmann et al. (64)

4. DOUBLE IONISATION AND (e,3e) EXPERIMENTS

(e,3e) studies of the valence shell double ionisation in Kr, Ar and at moderate ejected energies have recently become available (65). Similarly to the (e,2e) case these experiments are expected to provide structural information on correlated properties of the target for high impact energy and energy transfer (66). At low ejected electron energies (e,3e) and (e,(3-1)e) experiments, where an arbitrary pair of electrons is detected irrespectively of the direction of the third unobserved one, provide a very sensitive test of the double ionisation mechanism. Pioneering experiments of this kind (29) were conducted with modest energy and angular resolution and the kinematics achievable by the experimental set up was limited. Nevertheless

it was not only possible to assess their feasibility but also to collect interesting physical information. In the fig. 3 the four fold differential cross section for double ionisation of Ar is shown (67), as measured by detecting in coincidence the two slow ejected electrons at an incident energy of about 5 KeV. The angle at which one of the two electrons is detected is fixed (θ_b), while the other one is varied (θ_c).

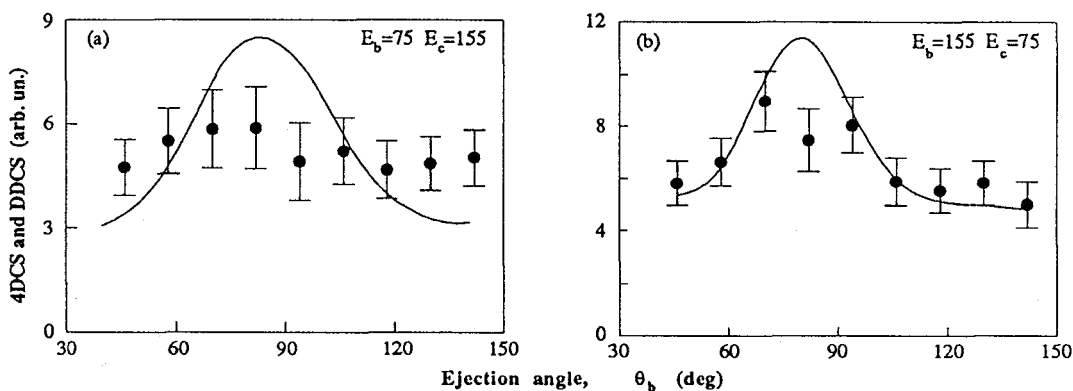


Figure 3: (e,(3-1)e) fourfold differential cross section (full circles) for double ionisation of Ar in coplanar geometry at $E_0 = 5.5$ KeV. (a): $E_b = 5$ eV and $E_c = 75$ eV. (b): $E_b = 75$ eV and $E_c = 5$ eV (65). See text for details.

Two processes can be at the origin of the observed cross section: a direct double ionisation or a shake off process. Though both of them are energetically allowed, within the experimental resolutions, they are expected to yield different angular distributions. On the basis of the results, the authors were able to discriminate the shake-off process as mainly responsible, upon the kinematics selected by the experiment, for the double ionisation process. These experiments, although in their infancy, have already stimulated new theoretical efforts aimed at elucidating the electron impact double ionisation mechanism.

5. (e,2e) AT LOW IMPACT ENERGY

Recently symmetric (e,2e) experiments have been conducted at low impact energies both in the scattering plane (equatorial plane) and in the plane perpendicular to it (azimuthal plane). Once more, H and He, being the simplest targets, have attracted much attention. The spectrometers developed at Manchester (68) and Kaiserslautern (69) permit accurate measurements of the full spatial distribution of the triple differential cross section and accurate investigation of the ionisation process. Absolute measurements in He performed at energies as low as 2 eV above threshold (34) have shown that the TDCS is no longer nearly rotationally symmetric with respect to the momentum transfer axis. An accurate knowledge of the non

coplanar TDCS is necessary to analyse the few relevant partial wave amplitudes. Contributions due to distortions in the incident channel, to electron-electron repulsion in the final state, or to the singlet or triplet components of the cross section can be tested by these experiments. Similar experiments, that have been carried out at incident energies up to 80eV (70), have shown that also in this case the usual versions of the Born approximations are not yet valid. All the complexities of the incoming and outgoing channel distortions and short and long range correlation must be embedded in the model describing the ionisation process. The out of plane geometry is the most sensitive one to all of these details. First order interaction models predict vanishing cross sections in the perpendicular plane. Hence TDCS in these kinematics are dominated by higher order interaction effects. Future developments are to be expected in this field. Namely, measurements on atomic hydrogen and argon could clarify the role of correlations, distortion and polarisation. Theoretical efforts mostly aimed at describing the interaction to all orders and to incorporate the exact asymptotic boundary conditions are desirable.

6. CONCLUSIONS

The rapid growth of the (e,2e) field has demanded for several review papers in order to keep track of the developments over the past few years. The most recent one (32) is only two years old and yet there was so much new material to be accounted for, that this paper, meant to highlight the recent achievements and possible future trends in the field, had to select, mostly to the taste of the authors, the examples to be discussed. Fortunately these subjects are fully discussed by other papers presented at this conference. Autoionising states studied by (e,2e) (71) and the ionisation mechanism at threshold (72) are subjects not discussed by this paper but extensively treated in this conference. The use of a polarised electron beam together with polarised targets is another field that, in spite of the experimental difficulties, has recently produced the first measurements of spin asymmetry for scattering of electrons on lithium (73). These experiments give new information about the ionisation process and new developments are to be expected in the near future.

In summarising the subjects discussed in this paper it is to be pointed out that the recent studies on the ionisation mechanism at high/intermediate energies both for valence and core shells have greatly contributed to elucidate the dynamics of few body Coulomb systems in multiple continua. For the future more progress are expected in the description of electron electron correlation both in the continuum and in the bound state. As the research will move from simple atoms to more complex systems, metal vapors, molecules and solids, the bound state correlation is expected to become more and more relevant. At that stage (e,3e) and (hv,2e) experiments are expected to become dependable spectroscopies of target electron correlation.

The body of (e,2e) data already collected is very large and for the needs of the investigation on the dynamics of few Coulomb body interactions more data will come in the near future. To

describe the cross sections in a concise manner suitable parametrisations of the cross section are of key relevance. The work on this subject has already been started. A natural evolution of the investigation on the ionisation mechanism is expected towards collisions in external fields and positron scattering.

Electron momentum spectroscopy is likely to evolve toward excited states, solids, oriented molecules and possibly molecules on solids.

All of the above mentioned evolutions imply measurements of very small cross sections with poor signal to noise ratio. With the present generation of coincidence spectrometers these experiments will be, if at all possible, extremely time consuming. The development of novel multicoincidence and/or multichannel coincidence spectrometers, fully computer controlled and optimised, is a compulsory step towards the wealth of new results to harvest in the (e,2e) field.

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