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# ELECTRON AND PHOTON IMPACT IONIZATION AND RELATED TOPICS 2004

Proceedings of the International Conference
Louvain-la-Neuve, 1–3 July 2004

183

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# Electron and Photon Impact Ionization and Related Topics 2004

**Proceedings of the International Conference on Electron and Photon Impact Ionization and Related Topics Louvain-la-Neuve, Belgium, 1-3 July 2004**

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## Preface

This book is a collection of the contributions of most of the invited speakers to the International Conference on "Electron and Photon Impact Ionization and Related Topics" which was held in Louvain-la-Neuve, Belgium, from 1st to 3rd July 2004.

This meeting which takes place every year brought together about 80 scientists, from 19 different countries, working in the field of simple and multiple ionisation by electron and photon impact as well as related topics. The program included 22 invited talks and 54 posters on recent experimental, theoretical and computational achievements in the realization, interpretation and modelling of correlated processes involving a wide range of targets : atoms, molecules, clusters or surfaces. Beside the regular talks, three review talks have been organized on three hot topics: anisotropy and polarization in Auger-electron emission, multiple ionisation of atoms in strong fields and theoretical and practical aspects of photoionization with excitation.

Financial support from the "Faculté des Sciences de l'Université catholique de Louvain" as well as from the "Fonds National de la Recherche Scientifique de la Communauté Française de Belgique" is gratefully acknowledged. This financial support permitted the participation of young scientists from eastern and Latin American countries.

I thank the members of the International Scientific Committee and my colleagues, in particular Xavier Urbain, as well as the students from the "Laboratoire de Physique atomique, moléculaire et optique de l'Université catholique de Louvain" for their help in the organization of this meeting.

I am also indebted to Ugo Ancarani from the Université de Metz who shared with me his experience in organizing such meeting.

Finally, I want to thank all speakers who contributed to these proceedings and all participants who gave their valuable time in carefully refereeing all contributions.

**B. Piraux**
November 2004

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*Paper presented at Int. Conf. Electron and Photon Impact Ionization and Related Topics, Louvain-la-Neuve, 2004*



# Identification of double ionization mechanisms, results bearing on electron correlation measurements

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**Abstract**

Electron impact double ionization of atoms and molecules offers a way to directly measure two-electron correlation, provided a number of conditions are satisfied. The ionizing collisions should take place in the binary, impulsive regime so that all of the energy and momentum lost by the projectile electron in each of the collisions is fully transferred to the target electrons. It is also necessary that the target electrons acquire sufficient kinetic energy from the ionizing collisions to leave the target atom or molecule in times short compared to an electron orbital period. Finally, the double ionization mechanisms must be well understood and separated from interfering processes. Measurements of the double ionization cross-sections for the two 3s electrons of magnesium have led to the identification and separation of two double ionization mechanisms. From the measurements the means by which two-electron momentum distributions can be obtained are clear.

## 1. Introduction

The most direct experimental insight into electron correlation is through measurement of two-electron probability densities, the square modulus of the two-electron wave function. Ezra and Berry (1983) and Krause and Berry (1985) first clearly demonstrated the sensitivity of two-electron position probability densities to electron correlation in atoms and were able to assign, in a qualitative way, correlated modes of motion to different states of two-electron atoms. Krause et al. (1987) demonstrated the equivalent sensitivity of two-electron momentum probability densities. These results established the foundation for measurements of electron correlation and

a rigorous test of theories and calculations. As an example we show in Fig. 1 the results of calculations of two-electron momentum densities for the pair of 3s electrons in magnesium. The figure shows plots of the probability of finding one of the electrons with a momentum $\mathbf{q}_1$ of a fixed magnitude 0.0, 0.5, and 1.0 a.u. and the other electron with a momentum $(q_x, q_y)$ where $q_x$ is the component of momentum parallel to $\mathbf{q}_1$and $q_y$ is the component of momentum perpendicular to $\mathbf{q}_1$. The calculations are based on a simple 5-term CI wave function.

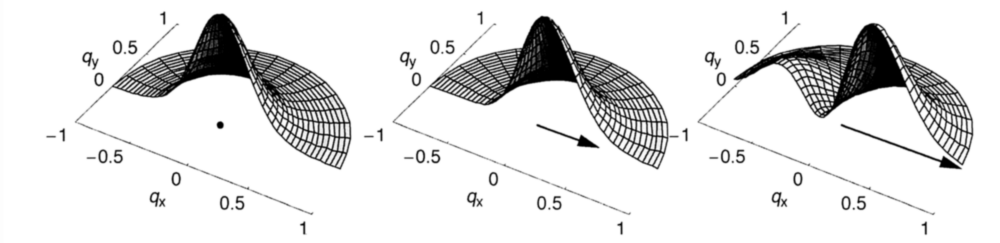


**Figure 1**. Momentum densities for the 3s electrons of magnesium. The momentum vectors for the electron with fixed momentum of 0.0, 0.5, and 1.0 a.u. are shown pointing in the positive $q_x$ direction.

These model calculations show that two-electron momentum densities are sensitive to the correlated motion of electrons. The determination of two-electron momentum densities requires that the momentum of one of the electrons be determined for a fixed magnitude and direction of the momentum of the second electron. These measurements must be done in a way that does not perturb the motion of the electrons in the atom; they must be the equivalent of "snap shots" of the atomic electrons.

**2. Double ionization**

Electron-impact ionization as a nonperturbative probe of electron motion was first demonstrated in the 1970's (McCarthy and Weigold 1976). Extensive experimentation demonstrated that the cross section for electron-impact ionization with relatively high-energy (1 keV) projectile electrons, large momentum-transfers (2-5 a.u.) and impulsve collisions can be accurately separated into the product of a scattering term and a target structure term (Pinkás et al. 1998). Single electron momentum densities have been extracted from measurements of the (e,2e) cross sections for a wide variety of atoms and molecules (Coplan et al. 1994, 1996).

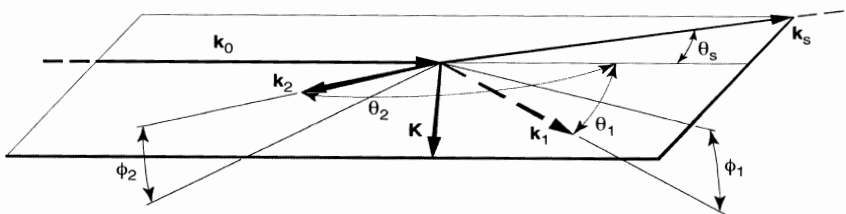


**Figure 2**. Vector diagram for double ionization collisions. The momenta of the incident scattered, and ejected electrons are $\mathbf{k}_0$, $\mathbf{k}_s$, $\mathbf{k}_1$, and $\mathbf{k}_2$. $\mathbf{K}$ is the momentum transferred from the incident electron to the target. The directions of $\mathbf{k}_s$, $\mathbf{k}_1$, and $\mathbf{k}_2$ are specified by $\theta_s$, $\theta_1$, $\phi_1$, $\theta_2$, and $\phi_2$. $\mathbf{k}_0$, $\mathbf{k}_s$, and $\mathbf{K}$ all lie in a single plane, the scattering plane.

Anticipating the success of (e,3e) experiments, Glassgold and Ialongo (1968), Smirnov et al. (1978), Amusia (1993), Popov et al. (1994), and others speculated that if two electrons were simultaneously knocked out of a target atom, the two-electron momentum density could be measured and used to learn about the correlated motions of the electrons in the atoms. The double ionization (e,3e) process is described in the vector diagram shown in Fig. 2.

If two atomic electrons are ejected in an impulsive fashion, it is, in principle, possible to determine information about their initial momenta, $\mathbf{q}_1$ and $\mathbf{q}_2$, from the measurement of their final momenta, $\mathbf{k}_1$ and $\mathbf{k}_2$, and a knowledge of the scattering kinematics ($\mathbf{k}_0$, $\mathbf{k}_s$, and $\mathbf{K} = \mathbf{k}_0 - \mathbf{k}_s$, the incident-electron momentum, the scattered-electron momentum and the transferred momentum). (e,3e) experiments for the investigation of correlated electron motion pose difficulties and complications in addition to those encountered in (e,2e) experiments. To start with, it is necessary to know the mechanism by which the two electrons are ejected from the target in order to calculate their initial motion from measurements of their motion following ejection. Beyond this fundamental requirement of many-body mechanics, there are practical experimental difficulties. To achieve the necessary conditions it is necessary that the energies of the ejected electrons exceed their binding energies by a factor of 2 to 3 at the minimum, however under impulsive conditions the (e,3e) cross section decreases as the inverse third power of the ejected-electron energies, so the signal from the experiment will decrease rapidly as the requisite ejected electron energies and momentum transfer are approached.

There have been a number of (e,3e) measurements (Dorn et al. 2002, 2003, Taouil et al. 1998, Lahmam-Bennani et al. 1989, 1991), but until recently, no fully impulsive (e,3e) measurement has been reported, nor has it been possible to unambiguously identify and isolate an impulsive mechanism that would be suitable for the measurement of two-electron momentum densities. The situation has changed recently, as results have become available from an (e,3e) apparatus that has been operating in our laboratory (van Boeyen et al. 2004). A particular feature of this device is that (e,3e) cross sections can be measured both in and out of the scattering plane. Moreover, this apparatus is multiplexed in such a way that up to 28 triple-coincidence measurements can be carried out and recorded simultaneously. Within the past two years the sensitivity of the apparatus has been increased by over two orders of magnitude while the background has been reduced by a factor of ten. We have reported the cross sections measurements for the impulsive ejection of two 62 eV electrons from magnesium by 1059 eV electron impact (van Boeyen et al. 2004); here we report additional measurements for the ejection of two 120 eV electrons by 2175 eV incident electrons. These results establish, we believe, the practical feasibility of the (e,3e) technique for the study of the motion of pairs of correlated electrons in atoms.

### 3. Impulsive (e,3e) with identification of mechanisms

The application of (e,3e) electron-impact double-ionization to the determination of the two-electron momentum densities requires experimental conditions that restrict the mechanism to two successive single-ionizing collisions of the incident electron with each target electron. Each of the single-ionizations must satisfy impulsive collision conditions with high incident–electron energy and large momentum-transfer. When these conditions are achieved, and the incident-, scattered-, and ejected-electron momenta are determined for each collision, the perpendicular component of the momentum of each ejected electron at the instant of its ejection from the target can then be obtained by subtracting the momentum transferred to the ejected electron from the

measured ejected-electron momentum. Our experiment has been designed to isolate mechanisms of double ionization suitable for use in the experimental measurement of two-electron momentum densities.

The two pathways for impulsive, doubly-ionizing collisions are TS1, where the incident electron interacts with only one of the target electrons that in turn interacts with the second (Tweed, 1992), and TS2, where the incident electron interacts with each electron in sequence. While effects observed in fully differential cross sections have been attributed to TS1 and TS2 (Dorn et al. 2002, 2003, Jia et al. 2002, 2003, El-Marji et al. 1997, Lahmam-Bennani et al. 2003), we have sought to measure TS1 and TS2 ionization involving impulsive, binary collisions. In the analysis below we develop the basis for our experimental measurements of TS1 and TS2. For simplicity we neglect the binding energies of the target electrons, assume that they are stationary at the instant of collision, and that they have equal ejected energies.

Following the first collision in the TS1 mechanism, the incident electron is scattered with a momentum $\mathbf{k}_s$, transferring momentum $\mathbf{K}$ to the first target electron. The $\mathbf{K}$ vector is perpendicular to $\mathbf{k}_s$. The first ejected electron, now with momentum $\mathbf{K}$, strikes the second target electron and, sharing its momentum, is scattered with momentum $\mathbf{k}_1$. The second target electron is ejected with momentum $\mathbf{k}_2 = \mathbf{K} - \mathbf{k}_1$. For equal ejected-electron energies, both $\mathbf{k}_1$ and $\mathbf{k}_2$ will lie in a plane that contains $\mathbf{K}$ with the angles between $\mathbf{k}_1$ and $\mathbf{K}$ and $\mathbf{k}_2$ and $\mathbf{K}$ both equal to 45°. This plane can assume any orientation about $\mathbf{K}$.

After the first collision in the TS2 mechanism, the incident electron is scattered with a momentum $\mathbf{k}_s'$, transferring momentum $\mathbf{k}_1$ to the first target electron. In the second collision, the incident electron, now with momentum $\mathbf{k}_s'$, is scattered with momentum $\mathbf{k}_s$ transferring momentum $\mathbf{k}_2$ to the second target electron. The three scattering planes defined by $\mathbf{k}_0/\mathbf{k}_s'$, $\mathbf{k}_s'/\mathbf{k}_s$, and $\mathbf{k}_0/\mathbf{k}_s$ are constrained to form an irregular tetrahedron with the fourth side bounded by $\mathbf{K}$, $\mathbf{k}_1$ and $\mathbf{k}_2$. Under these circumstances, the $\mathbf{K}/\mathbf{k}_1/\mathbf{k}_2$ plane can have only a fixed orientation with respect to the $\mathbf{k}_0/\mathbf{k}_s$ scattering plane. This means that TS2 events can only be observed by detectors that lie in the $\mathbf{K}/\mathbf{k}_1/\mathbf{k}_2$ plane.

The geometry and kinematics of the experiment were chosen to accommodate electrons ejected from the target atom by both the TS1 and TS2 mechanisms. The target atom was magnesium, a pseudo-two-electron atom with two 3s electrons outside of a closed shell. Identical ejected electron energies of 62 and 120 eV were chosen to meet the requirement of being significantly greater than their binding energies. These energies also fall within the energy regions where Auger electrons are absent. Table I lists the kinematics that were used with a scattering angle of 20° and a momentum transfer direction of –70° with respect to the incident electron direction.

Table I

| $E_0$ (eV) | $E_s$ (eV) | $E_1=E_2$ (eV) | $k_0$ (a.u.) | $k_s$ (a.u.) | $k_1=k_2$ (a.u.) | K (a.u.) |
|---|---|---|---|---|---|---|
| 1059 | 912 | 62 | 8.8 | 8.2 | 2.1 | 3.0 |
| 2175 | 1912 | 120 | 12.6 | 11.8 | 3.0 | 4.3 |

An incident electron current of 1 to 2 μA was used. Scattered electrons were collected by a tandem hemispherical analyzer with an entrance aperture ±2.25° by ±2°, and an energy resolution of 12 eV FWHM. Ejected electrons with momentum directed 45° from $\mathbf{K}$ were collected by a single truncated spherical analyzer with symmetry axis coincident with $\mathbf{K}$ and

detected by an array of eight detectors as illustrated in Fig. 3. Detector locations are labeled by the angular position about **K** relative to the ($\mathbf{k}_0$, $\mathbf{k}_s$) scattering plane. The energy resolution was 8 eV FWHM and the analyzer entrance apertures accepted electrons over ±7° by ±4.5° about the nominal scattering angle. There are 28 possible pair combinations of ejected-electron detectors, all of which are monitored simultaneously in triple coincidence with the scattered electron with nanosecond timing resolution. The maximum observed triple-coincidence rate for $E_0$ = 1059 eV was 4 × $10^{-4}$ events/s. In this geometry for the energies and momenta listed in Table I, the signature of TS1 ionization is triple coincidence events between ejected electron detectors separated by 180°. TS2 ionization gives triple coincidence events only for the detectors at 90° and 270°.

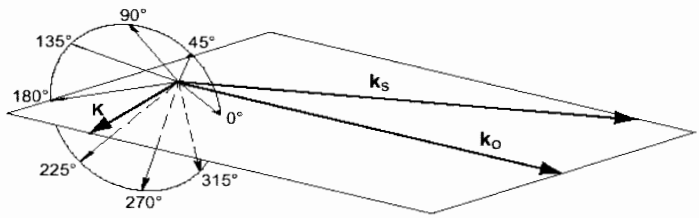


**Figure 3**. Experimental geometry for the observation of impulsive high momentum transfer double ionization collisions.

The measured fully differential differential cross sections for the $3s^2$ double ionization of magnesium at $E_0$ = 1059 eV and $E_0$ = 2175 eV for the geometry of Fig. 3 are shown in Fig. 4. The heights of the vertical panels are proportional to the magnitudes of the cross sections and the panel orientations indicate the two ejected-electron detector angles for which the cross sections were measured. The cross sections were placed on an absolute scale with an accuracy of ±50% using known single ionization cross sections.

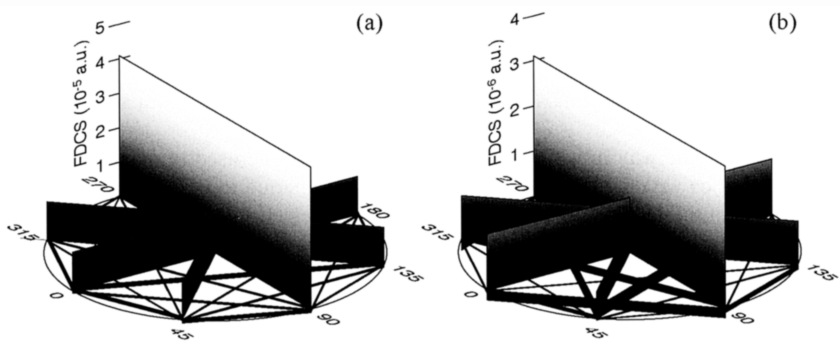


**Figure 4**. Double ionization five-fold differential cross sections (FDCS) as a function of ejected electron angle pairs. a.1059 eV incident electron energy and 62 eV ejected electron energy. b. 2175 eV incident electron energy and 120 eV ejected electron energy.

For both incident energies the (e,3e) cross sections are very small for detector pairs not separated by 180°. This means that for the geometry and kinematics of this experiment, there are no ionization mechanisms that do not involve discrete impulsive collisions. In particular, Auger and shake-off processes that would give signals between all pairs of ejected electron detectors do not contributing to double-ionization in this regime. The cross sections for the (0°, 180°), (45°, 225°), and (135°, 315°) pairs are all of the same magnitude within the experimental uncertainties and correspond to the ejection of a pair of electrons with momentum vectors coplanar with the momentum-transfer vector. The mechanism for the ejection of these electrons is identified as TS1.

The cross sections are largest for the pairs of electrons ejected in the planes that are perpendicular to the scattering planes, that is, one electron detected at 90° and the other at 270°. The (90°, 270°) cross section appears to result from a combination of TS1 and TS2 ionization. Assuming a quarter of the 90°/270° pairs of electrons are ejected by the TS1 mechanism, the remaining three quarters can be identified with TS2. For $E_0$ = 1059 eV, the TS1 cross section has a value of $1.1 \times 10^{-5}$ a.u.; for $E_0$ = 2175 eV the average value of this cross section is an order of magnitude smaller. Subtracting the TS1 values from the (90°, 270°) values gives $3 \times 10^{-5}$ a.u. for the TS2 cross-section at $E_0$ = 1059 eV and $2 \times 10^{-6}$ a.u. at $E_0$ = 2175 eV. For this experiment, assuming impulsive, binary collisions, the cross sections for both the TS1 and TS2 ionizations should scale approximately as the energy of the ejected electrons to the inverse third power. This gives a ratio of the TS1 and TS2 cross sections at $E_0$ = 1059 eV to those at $E_0$ = 2175 eV of 0.12 compared with the experiment values of 0.076. The calculated ratio of the TS1 to TS2 cross sections for the kinematics of the experiment is 0.25. From the experimental measurements we infer a value of 0.24 We believe that the measured angular dependence of the (e,3e) cross sections along with the measured energy dependence and relative cross sections provide strong evidence for the TS1 and TS2 mechanisms and the binary, impulsive collision regime.

**4. TS2 and two-electron momentum densities**

The TS2 mechanism is the appropriate process for the investigation of the relative motion of pairs of atomic electrons because information about the momenta of the ejected electrons in the atom is preserved in the course of the two sequential collisions. In this regard it is important to realize that the angle between the ejected-electron momentum vectors is approximately a right angle for the TS1 mechanism, while for TS2, the angle between the ejected-electron momentum vectors can be varied from about 0° to 180° simply by changing the choice of the incident-electron scattering angle. It is thus possible to isolate ionization by TS2 from all other ionization processes.

To illustrate the way in which the TS2 mechanism can be used to measure two-electron momentum densities we let $\mathbf{K}_1$ and $\mathbf{K}_2$ be the momenta imparted to the two target electrons in each collision with the incident electron. If the two target electrons are stationary at the instant of ejection from the atoms, the electrons will be ejected with momenta $\mathbf{k}_1 = \mathbf{K}_1$ and $\mathbf{k}_2 = \mathbf{K}_2$. If, on the other hand, electron 1 initially has momentum $\mathbf{q}_1$ and electron 2 initially has momentum $\mathbf{q}_2$, the electrons will be ejected with momenta $\mathbf{k}_1 = \mathbf{K}_1 + \mathbf{q}_1$ and $\mathbf{k}_2 = \mathbf{K}_2 + \mathbf{q}_2$. For ejected-electron energies selected such that $|\mathbf{k}_1| = |\mathbf{K}_1| = |\mathbf{k}_2| = |\mathbf{K}_2|$, as shown in Fig. 5, the initial momenta of the electrons can be determined from the relations $|\mathbf{q}_1| = 2|\mathbf{K}_1| \sin(\beta_1/2)$ and $|\mathbf{q}_2| = 2|\mathbf{K}_2| \sin(\beta_2/2)$, where $\beta_1$ and $\beta_2$ are the detection angles of the ejected electrons relative to $\mathbf{K}_1$ and $\mathbf{K}_2$. From the geometry of Fig. 5, it can be seen that the measured momenta, $\mathbf{q}_1$ and $\mathbf{q}_2$ can be either parallel or anti-parallel. This is the most useful case to study as it corresponds to conditions where

correlation effects are expected to be greatest and most clearly reflect the nature of the correlated electron motion (see Fig. 1). Two-electron momentum densities can be obtained by recording triple-coincidence rates for detection of ejected electrons at a range of angular positions about $\mathbf{K}_1$ with ejected electrons at a range of angular positions about $\mathbf{K}_2$. The geometry is similar to that of Fig. 3, however, to exclude non-TS2 ionization events, the included angle between $\mathbf{k}_1$ and $\mathbf{k}_2$ is

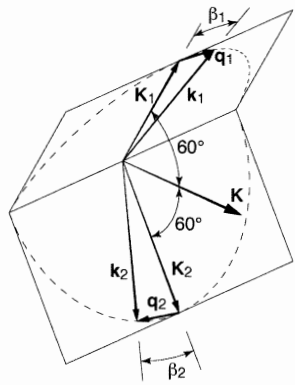


**Figure 5**. Double ionization kinematics for two-electron momentum density measurements.

increased from 90° to 120°. The signal from the desired sequential double-ionization mechanism will not be affected by this change in included angle. There is a direct relationship between the measured (e,3e) cross section and the two-electron momentum density. This is shown in the model calculations of Fig. 6 in which the (e,3e) cross section is shown as a function of detection angle $\beta_2$ for a fixed value of detection angle $\beta_1$ for the geometry of Fig. 5. The conditions are $E_0$ = 1000 eV, $E_s$ = 912 eV, $E_e$ = 62 eV, $\theta_s$ = 14°, and $\beta_1$ = 77° ($q_{1\parallel}$ = 0.4 a.u.). Positive values of $q_{2\parallel}$ indicate $\mathbf{q}_2$ parallel to $\mathbf{q}_1$, negative values indicate $\mathbf{q}_2$ antiparallel to $\mathbf{q}_1$. The asymmetry of the cross section about $\beta_2$ = 270° indicates a preference for the two electrons to travel in the same direction under the specified conditions. Cross sections and probability densities were calculated with a correlated CI wave function.

To measure two-electron momentum densities the present tandem spherical analyzer will be retained for scattered electron analysis, however, two toroidal electrostatic analyzers will replace the single truncated spherical analyzer of the present experiment. The analyzers will be positioned to collect electrons ejected into planes that are rotated about the scattered direction by +60° and –60° relative to the plane containing $\mathbf{k}_0$, $\mathbf{k}_s$, and $\mathbf{K}$. An array of up to 8 electron multipliers positioned at the image plane of each analyzer will accept a range of ejected-electron angles about the directions $\mathbf{K}_1$ and $\mathbf{K}_2$ to be studied as well as increase the data rate by a factor of 64. Preliminary measurements will be made at coarse momentum resolution using existing electron detectors. Final measurements will be performed with an array of electron multipliers custom designed to achieve a momentum resolution of ±0.1 a.u. These detectors will cover a range from 0 to 1 a.u. with a resolution of ±0.1 a.u. The pass energies and energy resolutions of the analyzers are fixed by the need to maintain impulsive collision conditions while at the same time defining the final state of the residual doubly charged ion and excluding Auger electrons.

For magnesium, the large separation in the energies of the ion states means that it will be possible to measure two-electron momentum densities for the ejection of two 3s electrons while unambiguously establishing the final ion state.

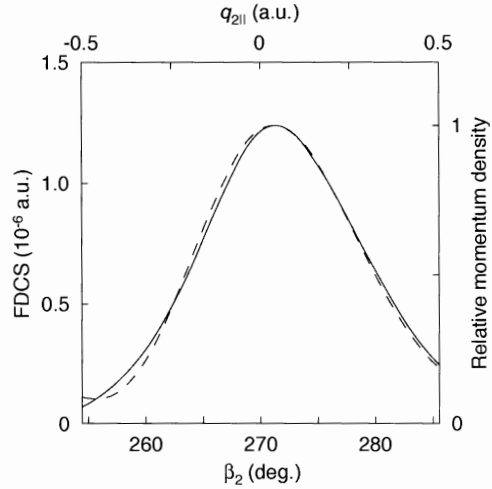


**Figure 6**. Mg $3s^2$ two-electron momentum probability density (solid line). (e,3e) five-fold differential cross section (FDCS) (dashed line).

## 5. Signal rates and momentum resolution

The feasibility of a triple coincidence measurement is determined by data accumulation time required to achieve a desired statistical precision, rather than just the coincidence rate. Statistical precision depends on both the true triple coincidence electrons and the coincidence rate due to randomly correlated electrons (Lahmam-Bennani 1991). Once the random coincidence rate equals the true coincidence rate, data accumulation times required to achieve a desired precision increase with increasing incident current. This limit can be raised by improving coincidence timing resolution, analyzer efficiency, and the uncorrelated background signal.

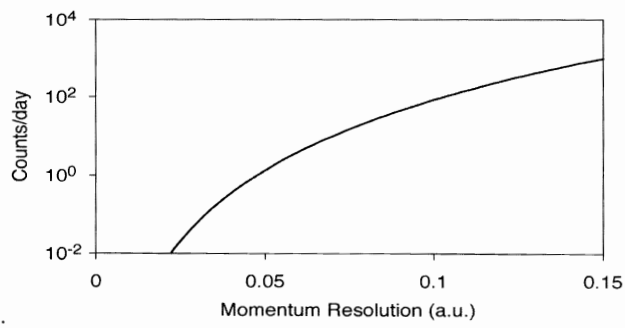


**Figure 7**. Calculated double ionization signal rate as a function of the experimental momentum resolution.

With readily attainable improvements in these areas a 36-fold increase in the true to random ratio can be achieved over the present spectrometer and incident currents up to 20 μA can be used. A plot of the expected true coincidence rate as a function of momentum resolution is shown in Fig. 7. The coincidence rate was calculated for a true to random ration of 1:1, which is obtained at an incident current of 20 μA. At a momentum resolution of ±0.1 a.u. the maximum true coincidence rate is expected to be 86 counts/day. Table II summarizes the operating parameters of the proposed experiment.

Table II

| Parameter | $\Delta q = \pm .0.15$ a.u. | $\Delta q = \pm .0.10$ a.u |
|---|---|---|
| Electron Energy Resolution | 8.0 eV | 8.0 eV |
| Angular Resolution, Scattered Electrons | ±1.1° | ±0.7° |
| Angular Resolution, Ejected Electrons | ±4.1° | ±2.7° |
| Signal Rate | 978 counts/day | 86 counts/day |
| Signal-to-Random Ratio | 1:1 | 1:1 |
| Incident Current | 20 μA | 20 μA |

## 6. Conclusions

In order to measure two-electron momentum densities using electron impact double ionization it is necessary to work in a kinematic regime where the ionizing collisions occur impulsively and the double ionization mechanism preserves the information about the momenta of the target electrons at the instant of their ejection. Two impulsive double ionization mechanisms, TS1 and TS2, have been identified from the angular distributions of the ejected electrons about the momentum transfer direction. Of the two mechanisms, only TS2 is suitable for momentum density measurements. TS2 can be isolated from TS1 by changing the angle between the momenta of the ejected electrons and the momentum transfer vector at which the ejected electrons are detected. An instrument based on the results and realistic calculations will be able to directly measure two-electron momentum densities with a resolution of from ±0.15 to ±0.10 a.u. over data accumulation times from 1 to 10 days. It is anticipated that these measurements will offer the first experimental measurement of the correlated motion of electrons in atoms.

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## 8. References

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