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Ionization of Atoms with Spin Polarized Electrons


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Abstract. The most detailed insight into the process of electron impact-induced ionization of atomic species is provided by measurements in which both kinematical and quantum mechanical variables are determined. Here we describe recent (e,2e) experimental and theoretical studies involving the ionization of xenon and argon by spin-polarized electrons in which the fine-structure levels of the ion are energetically resolved. Such investigations shed light on the mechanisms driving the ionization reaction and the role of exchange and relativistic processes.

Keywords: Spin Polarized Electrons, Electron Impact-Induced Ionization
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INTRODUCTION

Continuing progress in areas of technological interest, such as plasma formation, gas discharge and laser physics, as well as our understanding of the physics and chemistry of the upper atmosphere, depend crucially on our understanding of the process of electron impact-induced ionization. The most detailed insight into electron impact-induced ionization of atomic species is provided by measurements performed within a kinematically complete framework. In the case of single ionization, this can be achieved through (e,2e) measurements in which pairs of electrons derived from a common ionization event are identified by their correlated arrival times at two detectors [1] or through detection of one scattered electron and the residual ion in an electron-ion coincidence measurement [2]. In recent years a new class of (e,2e) measurements have emerged in which both kinematical and quantum mechanical variables are determined. This has been achieved through preparation of the quantum projection state of the electron-atom system prior to collision. These measurements employ beams of spin-polarized electrons [3–4] and/or spin polarized targets achieved through laser preparation or magnetic selection techniques [5–6]. The addition of quantum state specificity to ionization experiments means that the constituents of the ionization process can be isolated and highlighted.
Numerous refinements in theoretical techniques have accompanied these experimental developments. Electron exchange, relativistic effects and the long-range Coulomb-interaction of the charged particles can now be treated accurately with modern theoretical tools. This paper will focus on calculations performed within the framework of Distorted Wave Born Approximation (DWBA) to interpret the experimental data. Here we describe recent progress in investigations into the electron-atom system and explain how spin-resolved studies can provide insight into the role of electron exchange in electron-impact-induced ionizing collisions.

FINE STRUCTURE EFFECT

Spin dependent effects in the scattering of electrons from atomic targets can arise through three mechanisms, namely electron exchange, spin-orbit interaction of scattered electrons in the field of the atom/ion and through angular-momentum coupling within the target atom and/or residual ion in the case of ionizing collisions [7,8]. For ionization experiments performed with spin polarized electrons and in which the fine-structure levels of the residual ion are resolved, Jones et al [9] showed that even in the non-relativistic limit where spin-orbit interaction of scattered electrons in the field of the atom/ion is negligible, a strong dependence of the ionization cross section on the spin projection of the projectile electron could be expected. In their model, the spin dependence results from an interplay of exchange between the projectile and ejected target electron, angular-momentum coupling and the impact-induced orientation of the residual ion. The model predicts zero spin asymmetries in the limit of negligible exchange, and in analogy to the “Fine-structure Effect” in excitation [10,11], zero spin asymmetry for the case where the individual fine-structure transitions are energetically unresolved. Subsequent measurements on xenon [12,13] and later, more sophisticated calculations [14,15], while confirming a strong spin-dependence in the ionization cross sections as originally predicted, have revealed additional and significant contributions from many-body exchange effects which tend to mask the signatures for a “pure” fine-structure effect. Notwithstanding this fact, that [9] predicts zero spin asymmetry in the absence of exchange means that the spin asymmetry parameter is a very sensitive test of the role and nature of electron exchange in electron-impact-induced ionizing collisions, and a sensitive test to the magnitude of contributions from relativistic effects which can also contribute to a non-zero result, even in the absence of exchange.

EXPERIMENT

The experimental apparatus is shown schematically in Figure 1 with details to appear separately [16]. Longitudinally polarized electrons are created by the photo-excitation of valence electrons from a strained gallium arsenide photo-cathode under illumination by circularly polarized laser light [17]. These electrons are extracted, deflected through 90° and focused to produce a beam of transversely polarized electrons. Inversion of the electron beam polarization from into (spin down) to out of (spin up) the scattering plane is achieved by reversing the helicity of the laser light by...
means of a liquid crystal retarder. Here the scattering plane is defined by the momentum vectors for the incident and measured scattered electrons. After deflection through 90°, the electron beam is accelerated to around 1 keV and transported at high energy through a differential pumping stage before entering the main collision chamber in which the electron spectrometer is housed. Inside this scattering chamber, the electron beam is decelerated to the experimental collision energy \( E_i \) by means of a five element electrostatic lens and focused at the grounded interaction volume defined by the overlap of the electron and target beams. The atomic target beam is formed by effusion through a 1mm internal diameter needle orientated orthogonally to the scattering plane. The required beam energy \( E_i = eV_c \) is set by adjusting the photocathode potential \( V_c \).

**FIGURE 1.** Schematic representation of the apparatus used to measure spin-resolved cross sections.

Scattered electrons emitted within the scattering plane are momentum analyzed in one of two toroidal-sector electrostatic-energy-analyzers located on opposite sides of the primary electron beam. Each analyzer is terminated by a micro-channel-plate electron-multiplier pair followed by a crossed delay line detector from which the spatial and temporal arrival coordinates \((x_i, y_i, t_i)\) for each measured electron are determined [18]. From these coordinates, pairs of electrons derived from common \((e,2e)\) ionization events are identified by their correlated arrival times at the two separate detectors and their initial momenta \((p_1, p_2)\) are deduced. One analyzer accepts electrons over the azimuthal angular range \( \theta_1 = -20^\circ \rightarrow -60^\circ \), the second over the range \( \theta_2 = +20^\circ \rightarrow +120^\circ \) (-240° → -340°), where \( \theta_1 \) and \( \theta_2 \) are measured within the scattering plane and with respect to the direction of the primary electron beam. Due to the limited size of the electron-multipliers (80 mm diameter), parallel measurements can only be performed over a 40° angular capture range in the larger analyzer. However, its multiplier/detector assembly is rotatable which enables access to its full 100° acceptance range. Background resulting from electrons derived from separate ionization events are subtracted using standard statistical techniques [1]. Recently, the spectrometer performance was greatly improved due to suppression of stray-electron background. The small statistical fluctuations of recent valence-shell ionization-data reflect this fact.
The present series of measurements concern the investigation of exchange effects in the electron-atom ionization of the two atomic species xenon and argon. In the case of xenon, we consider the reaction leading to the removal of a target electron in the 5p\(^6\) valence shell leading to the spin-orbit split Xe\(^+\) 5p\(^5\) 2P\(_{1/2}\) or 5p\(^5\) 2P\(_{3/2}\) final ion states. In the case of argon, a hole is created in the inner-shell 2p\(^6\) orbital and cross sections have been measured for transitions leading to the Ar\(^+\) 2p\(^5\) 3p\(^6\) 2P\(_{1/2}\) and 2P\(_{3/2}\) ion states. By varying the reaction kinematics and target structure, we aim to obtain a deeper insight into the exchange process.

Xenon was the first closed-shell target atom for which the measurement of non-zero spin asymmetries was established [12,13]. Since that time disparities between experiment and theory have narrowed, leading to an enhanced understanding of scattering dynamics. Calculations performed in the early 90’s and subsequent works have shown that while exchange between the incident and ejected target electron, electron impact-induced orientation of the ion, and angular momentum coupling within the residual ion all contribute to the observed asymmetries, as predicted by [9], the form of the asymmetry function is strongly influenced by many-body effects involving exchange of the final-state continuum electrons with the residual electrons in the ion. The fact that significant discrepancies between measurement and theory still remain raises questions about our ability to accurately describe many-body exchange processes in electron-atom scattering. However, the origin for the disparity may lie elsewhere in theory or experiment.

Figure 2 shows the results of recent measurement and calculation. The reaction kinematics are presented in the figure caption. The experimental results are compared to non-relativistic Distorted Wave Born Approximation (DWBA) calculations in two different forms. In both cases, exchange between the incident and ejected target electron are taken into account as well as exchange between the two final-state
continuum electrons and the remaining electrons in the residual ion. This latter many-body exchange effect is accounted for in the calculation of the distorted waves by making a local approximation for the non-local exchange potential. In the calculation labeled DWBA, the Coulomb interaction in the final state is calculated through angle dependent effective charges [19]. In the more sophisticated 3DW calculation [20] the final-state Coulomb interaction is included exactly and constitutes a distorted wave version of the BBK theory [21]. For both cases the agreement with measurement is reasonable, however the remaining discrepancies suggest that a better theoretical treatment of the exchange effect between the continuum electron and passive electrons in the ion may be required.

Given the difficulties in theoretically describing the complex many-body nature of the exchange process for the xenon measurements, kinematics and a target were proposed where the exchange process, to a very good approximation, should reduce to a two-body exchange between projectile and ejected target electron [8]. Thus by performing investigations under those conditions, a clearer understanding might be achieved of the relationship between spin asymmetries and mechanisms of exchange. The proposed experiment involved measuring fine-structure-resolved spin-asymmetries for ionization of the 2p⁶ inner shell of argon with respective energies of 1949 eV and 500 eV for the ejected and scattered electrons. These energies were deemed large enough to ensure a two-body exchange mechanism, but small enough to ensure relativistic spin flip processes, which play a significant role in the K-shell ionization of heavy targets [22], remain small. As the cross section for the proposed kinematics was extremely small, we decided to adopt modified energies of 600 eV and 60 eV for the final-state electron-pair where the cross section is higher [23], hoping that a simplified exchange mechanism would still remain. Preliminary experimental results are shown in Figure 3.

\[ \text{FIGURE 3. Spin asymmetry for the ionization of argon leading to the } ^2\text{P}_{1/2} \text{ (solid circles) and } ^2\text{P}_{3/2} \text{ (open circles) final ion states. The incident beam energy is } 910 \text{ eV, the two outgoing electrons of energies 600 eV and 60 eV leave on opposite sides of the incident beam. For this figure, the fast 600 eV electron scatter at an angle } \theta_1=36^\circ \text{ and the spin asymmetry is measured as a function of the scattering angle } \theta_2 \text{ of the slow 60 eV electron.} \]

Of particular interest is the relationship between asymmetries for the $^2\text{P}_{1/2}$ and $^2\text{P}_{3/2}$ ion states, which for a “pure” fine-structure effect should follow the relationship [9]
\[ A_{1/2} = -2 A_{3/2}. \] \hspace{1cm} (1)

Whilst the error bars in Figure 2 are too large to substantiate this relationship, the spin asymmetry derived from averaging over our full data set (not presented here), corresponding to a range of scattering angles $\theta_1$ and $\theta_2$, suggests that relationship (1) is closely approximated. This suggests that the experiment may indeed describe a simpler scattering process. Comparison of the data with Relativistic Distorted Wave Born Approximation (rDWBA) calculations will appear in a future publication [24].

**CONCLUSION**

The study of ionization of closed-shell atoms with spin polarized electrons provides insight into target coupling, exchange and relativistic effects in electron-atom collisions. Through a judicious choice of the reaction kinematics and target, contributions from relativistic or exchange processes can be isolated in measurement. Continued improvements in experimental and theoretical tools promise greater insight into the dynamics of electron-atom collisions in the coming years.

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**REFERENCES**
