IONIZATION OF LASER ORIENTED SODIUM ATOMS BY POLARIZED ELECTRONS

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1. INTRODUCTION

The most stringent tests for theories describing low and intermediate energy ionization processes by electron impact would be provided by measurements in which the quantum mechanical states of both the reaction participants and products were resolved. As a step in this direction, several experiments have been performed over the last decade in which the initial state of the e-atom system has been partially or completely resolved [1, 2, 3, 4]. Recently, we reported on the first $(e, 2e)$ measurement of laser excited sodium atoms by polarized electrons under symmetric kinematics where the two final state electrons had equal energies [5]. In the present work, we extend this study by presenting experimental and theoretical results under asymmetric kinematics at an intermediate incident electron beam energy E_0 of $151eV$.

Sodium was chosen as the target atom for these experiments for a number of reasons. Firstly, it is a light atom, which suggests that at an intermediate energy regime, such as the one considered in the present study, relativistic interactions for both continuum and bound electrons can be neglected. Secondly, the details of the laser pumping scheme for the 3S-3P sodium transition are well understood and the transition frequency is easily accessible to high power tunable lasers. Lastly, previous work on the ionization of laser excited sodium atoms by an unpolarized electron beam showed that the dependence of the (e,2e) ionization cross section on the initial target orientation can be properly described at this energy regime within a Distorted Wave Born Approximation (DWBA) framework [6, 7]. This latter point motivated the present study which emphasizes the ionization of laser oriented sodium atoms by polarized electrons in a kinematical region where comparison with the DWBA theoretical predictions is meaningful.

In the (e,2e) ionization of laser oriented sodium atoms by spin polarized electrons, the presence of non-zero target orientation results in a more complicated structure for the ionization cross section than that for ionization with spin polarized electrons of spin polarized atoms possessing zero orientation [1, 8]. In the latter case the triply differential cross section (TDCS) depends solely on the relative spin orientations of the projectile electron and bound valence electron it removes in the collision. This dependence is usually related to a spin asymmetry, whose behavior reflects the contribution of the singlet and triplet cross section to the spin averaged cross section, the only quantity accessible to conventional (e,2e) experiments (without initial state preparation). In the present experimental situation, we show that it is possible to extract from measurements the singlet and triplet cross sections corresponding to a given initial atomic angular momentum state, thus probing deeper into the mechanism driving the ionization process. To emphasize the dependence of the cross sections on a particular initial state preparation and highlight new aspects of the collision dynamics revealed by the present measurements, we introduce a set of tensorial parameters for the description of the excited state ionization. This set of parameters reveals a new dependence of the TDCS on the initial state of the electron-atom system, observed for the first time experimentally and termed magnetic dichroism.

2. EXPERIMENTAL ARRANGEMENT

Fig. 1 shows schematically the kinematical arrangement employed for the present measurements and the main features of the apparatus. As a detailed description of the apparatus has been given elsewhere [9], only a brief outline will be given here.

The primary polarized electron beam used to induce the ionization process is generated by photo-emission from a cesium and oxygen coated GaAs crystal under illumination by a 810nm circularly polarized laser radiation. The degree of polarization, perpendicular to the scattering plane, achieved for the present measurements was P_e=0.24±0.03 and remained stable over the course of the measurements. The inversion of the polarization of the incident electron beam is achieved by reversing the helicity of the laser radiation field. This is achieved through rotation of the quarter wave plate interposed between the diode laser producing the radiation field and the GaAs photocathode.

A frequency modulated beam of 589nm circularly polarized radiation is used to orient and excite the sodium atoms. After a few excitation/decay cycles the atoms gather exclusively in the two state system

$$\begin{array}{c} 3s^1\ {}^2S_{1/2}(F=2, m_F=+2) \\ \leftrightarrow 3p^1\ {}^2P_{3/2}(F=3, m_F=+3) \\ \text{for pumping by } \sigma^+ \text{ radiation,} \end{array} \tag{1}$$

$$\begin{array}{c} 3s^1\ {}^2S_{1/2}(F=2, m_F=-2) \\ \leftrightarrow 3p^1\ {}^2P_{3/2}(F=3, m_F=-3) \\ \text{for pumping by } \sigma^- \text{ radiation.} \end{array} \tag{2}$$

For the two excited states $m_F = +3$ and $m_F = -3$ both the polarization and orientation vectors associated with the target beam are directed normal to the scattering plane and are parallel to one another. The excitation fraction obtained is around 40%.

Pairs of scattered electrons emitted within the scattering plane, defined by the sodium and electron beams, are detected in two independently rotatable electrostatic analyzers on opposite sides of the incident beam. Each analyzer incorporates channel plate position-sensitive electron detectors enabling simultaneous measurement of electrons over a 6 eV energy band with an energy resolution of around 300 meV. A total (e,2e) coincidence energy resolution of around 0.9 eV was achieved. This resolution is sufficient to accurately distinguish between ionizing events involving respectively atoms in their ground and excited states, which are separated in binding energy by 2.14eV.

The experiment consisted of measuring (e,2e) counts as a function of the scattering angle of one of the two final state electrons and for

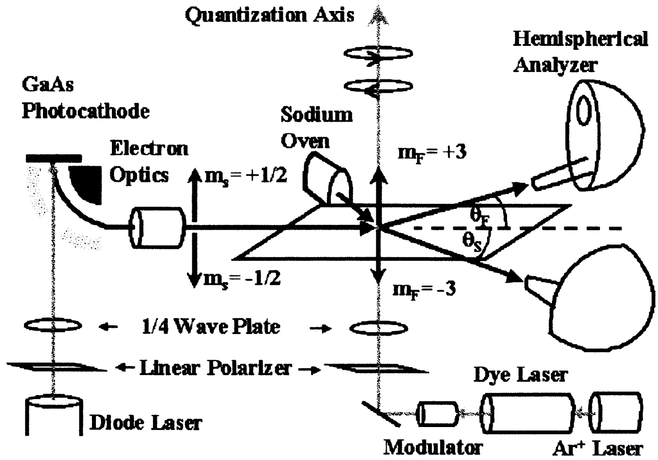

Figure 1 Schematic representation of the (e,2e) experiment on laser excited sodium atoms

each of the four combinations of atomic and electron beam polarization directions, namely

$$e(\uparrow) + A(m_F = +3), e(\uparrow) + A(m_F = -3),$$
$$e(\downarrow) + A(m_F = +3), e(\downarrow) + A(m_F = -3).$$

In the present paper, we focus on the ionization of the excited sodium atoms. Results for the ionization of polarized ground state atoms by polarized electrons which are collected simultaneously will be described in a later publication.

3. SINGLET AND TRIPLET CROSS SECTIONS

The formalism developed to describe spin polarized inelastic scattering experiment on oriented sodium atoms [10] can be readily applied to the present situation. This framework naturally relates the count rates obtained for the four experimentally accessible initial states of the e-atom system to singlet and triplet initial atomic-state-resolved cross sections as:

$$N^{(\uparrow\downarrow)\Uparrow} \equiv \frac{K}{4}\left[(3+P_e)t_{11} + (1-P_e)s_{11}\right], \tag{3}$$

$$N^{(\uparrow\downarrow)\Downarrow} \equiv \frac{K}{4}\left[(3-P_e)t_{-1-1} + (1+P_e)s_{-1-1}\right]. \tag{4}$$

where $s_{11,(-1-1)}$ and $t_{11,(-1-1)}$ stand respectively for the initial atomic-state resolved singlet and triplet cross sections for positive ($m_l = 1$) and negative ($m_l = -1$) target orientations. $N^{\uparrow\Uparrow}$ ($N^{\downarrow\Uparrow}$) stand respectively for the measured coincidence count rates for ionization of positive orientation $\Uparrow$ target atoms by an electron beam whose polarization vector is parallel $\uparrow$ (antiparallel $\downarrow$) to the target orientation. In the same manner $N^{\downarrow\Downarrow}$ and $N^{\uparrow\Downarrow}$ stand respectively for the count rates corresponding to ionization of negative orientation $\Downarrow$ target atoms by an electron beam whose polarization vector is parallel $\downarrow$ (antiparallel $\uparrow$) to the target orientation. K is a normalization constant arising from the fact that the present (e,2e) measurements are relative and not absolute. The relations (3) and (4) are derived invoking the Percival Seaton hypothesis [11], which assumes that the hyperfine structure of the atoms have no dynamical contribution to the reaction. The spin-orbit interactions are further assumed to have negligible effects on the parameters accessible experimentally. $s_{m_l,ml}$ and $t_{ml,ml}$ stand respectively for the initial atomic-state resolved singlet and triplet cross sections which in turn reflects the parity of the unsymmetrised initial atomic-state-resolved scattering amplitude $f_{ml,ml}$ under exchange of the two outgoing electrons

$$s/t_{mlml} = |f_{mlml} + (-1)^S g_{mlml}|^2. \tag{5}$$

$g_{ml,ml}$ is the exchange amplitude obtained by reversing the role of the two outgoing electrons in the exit channel while S is the total spin of the e-atom system. The relations (3,4) illustrate the nature of the information that is accessible in the present experimental arrangement. As can be readily seen from the relations (3,4), the experiment distinguishes between parallel and antiparallel spin projections for the incident and atomic electrons for each accessible atomic state. Consequently, the experiment directly accesses the triplet cross section and the average of the triplet and singlet cross sections for each of the two angular momentum projection states of the atomic electron. Furthermore, the relations (3,4) also highlight the dependence of the measured count rates on the degree of polarization of the incident electron beam.

Inverting relations (3,4) leads to the following expressions for the singlet and triplet cross sections expressed in terms of count rates

$$s_{11(-1-1)} \equiv K^* \left[(\frac{3}{P_e} + 1) N^{\downarrow\Uparrow(\uparrow\Downarrow)} - (\frac{3}{P_e} - 1) N^{\uparrow\Uparrow(\downarrow\Downarrow)} \right], \tag{6}$$

$$t_{11(-1-1)} \equiv K^* \left[(\frac{1}{P_e} + 1) N^{\uparrow\Uparrow(\downarrow\Downarrow)} - (\frac{1}{P_e} - 1) N^{\downarrow\Uparrow(\uparrow\Downarrow)} \right]. \tag{7}$$

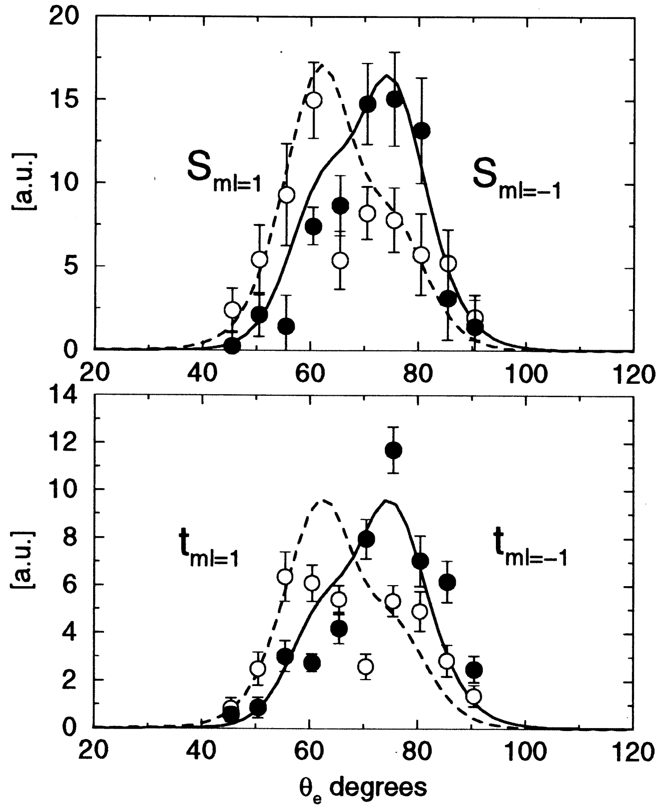

Figure 2 Variation of the singlet (a) and triplet (b) atomic-state-resolved TDCS as a function of the ejected electron angle. E_0 = 151eV, E_s = 121eV E_e = 27eV and $\theta_s = 20°$. (solid line) $m_l = -1$, (dashed) $m_l = 1$.

Thus four orientation dependent singlet and triplet cross sections are extracted from experiment to an overall normalization factor K^*. This analysis of the experimental data enables comparison to be made between theoretical predictions and experimental measurements for each of the individual processes contributing to the averaged cross section accessed by a conventional (e,2e) experiment (i.e. without initial state preparation).

In Fig. 2, we compare the experimental singlet and triplet cross sections obtained in asymmetric kinematics, where the initial electron beam energy is set at $E_0 = 151$ eV, while the scattered and ejected electrons are detected with respective average energies $E_s = 121$ eV and $E_e = 27$ eV. The cross sections are presented as a function of the ejected electron angle, θ_e, while the scattering angle is fixed at $\theta_s = 20°$. As the experimental measurements are not absolute, a single normalization of the experimental data to the theoretical average cross section is performed. Consequently, the relative contribution of each of the state resolved cross sections to the average cross section is preserved. The experimental results are compared to DWBA calculations [6, 7].

Within the DWBA framework, the scattering from the Na atom is reduced to a three-body problem by considering only the active (valence) electron of the Na atom. A Slater representation of the sodium Hartree-Fock orbital, generated using the Hartree-Fock program of Fischer [12], is used for the target radial orbitals. This representation is used to generate the bound-electron radial orbital as well as the static potential in the calculation of the distorted waves. In the present calculations, the equivalent spin-average local exchange potential of Furness and McCarthy [13]is used to generate the exchange part of the distorting potential in both the entrance and exit channels. The Coulomb potential is added when the distorted waves are considered as electron-ion states. As such, no final state electron-electron correlation is included in the present model.

Inspection of Fig. 2 reveals that the relative strength of the singlet to triplet cross sections is well accounted for within the DWBA model. Further, both the experimental measurements and the theoretical model predict a strong dependence of both singlet and triplet cross sections on the initial atomic state preparation. Such a dependence of the experimental cross section is well depicted by the theoretical model, also some overestimation of the t_{11} cross section is however clearly noticeable. This dependence which was termed orientational dichroism [14] was previously observed for the spin averaged cross sections in experiment involving the (e,2e) ionization of laser oriented sodium atoms by an unpolarized electron beam [3]. The present analysis clearly shows that this effect is present in each of the spin channels. It should also be noted that the theoretical model predicts atomic-state-resolved cross sections, for both singlet and triplet cross sections symmetric around the momentum transfer. The origin of this feature can be traced back to the influence of the Coulomb interaction on the scattered electron [9]. Within a Born description of the reaction, it is well known that when the scattered electron is described by a plane wave, the resulting cross section is symmetric around the momentum transfer. In the DWBA model used in the present study, while the ion potential is used to calculate the scattered electron distorted wave, the typical behavior of the state resolved cross sections is an indication of the neglegible influence of the Coulomb interaction on this electron. The experimental singlet cross sections support such an analysis. Consequently, the origin of the disagreement between the theoretical and experimental t_{11} cross section remains unclear as, under these kinematic conditions, the inclusion of the electron-electron interaction in the final state is expected to have little influence.

4. ORBITAL AND MAGNETIC DICHROISM

To highlight the interesting new aspects of the collision dynamics revealed by the present measurements we introduce the following four tensorial parameters to describe the excited state ionization:

$$\sigma_{av} = \frac{K^*}{2}\left[(3t_{-1-1} + s_{-1-1}) + (3t_{11} + s_{11})\right], \quad (8)$$

$$A_{orb} = \left[(3t_{11} + s_{11}) - (3t_{-1-1} + s_{-1-1})\right]/(4\sigma_{av}), \quad (9)$$

$$A_{mag} = \left[(s_{-1-1} - s_{11}) - (t_{-1-1} - t_{11})\right]/(4\sigma_{av}), \quad (10)$$

$$A_{m,o} = \left[(s_{-1-1} + s_{11}) - (t_{-1-1} + t_{11})\right]/(4\sigma_{av}). \quad (11)$$

In terms of count rates Eqs.(9-12) can be expressed as:

$$\sigma_{av} = K^*\left[N^{\uparrow\Uparrow} + N^{\uparrow\Downarrow} + N^{\downarrow\Uparrow} + N^{\downarrow\Downarrow}\right] = KN_{\Sigma}, \quad (12)$$

$$A_{orb} = \frac{1}{N_{\Sigma}}\left[N^{\uparrow\Uparrow} + N^{\downarrow\Uparrow} - N^{\uparrow\Downarrow} - N^{\downarrow\Downarrow}\right], \quad (13)$$

$$A_{mag} = \frac{1}{N_{\Sigma}P_e}\left[N^{\uparrow\Uparrow} + N^{\uparrow\Downarrow} - N^{\downarrow\Uparrow} - N^{\downarrow\Downarrow}\right], \quad (14)$$

$$A_{m,o} = \frac{1}{N_{\Sigma}P_e}\left[N^{\uparrow\Downarrow} + N^{\downarrow\Uparrow} - N^{\uparrow\Uparrow} - N^{\downarrow\Downarrow}\right]. \quad (15)$$

In Fig. 3 we compare the behavior of these parameters obtained from experimental count rates with the theoretical prediction of the DWBA model. Fig. 3a shows the variation of the σ_{av} parameter. The parameter σ_{av} is a scalar which describes the ionization cross section averaged over the spin of the electrons and the sense of orbital rotation for the ionized valence electron and is independent of the helicity of the laser light. This quantity is directly proportional to the cross section measured in a conventional (e,2e) experiment. The DWBA calculations describe well the position of the maximum of the experimental TDCS. However, the details of the variation of the experimental cross section as a function of ejected electron angle between 60 and 80 degrees are not accounted for by the present model. The analysis performed in the previous section

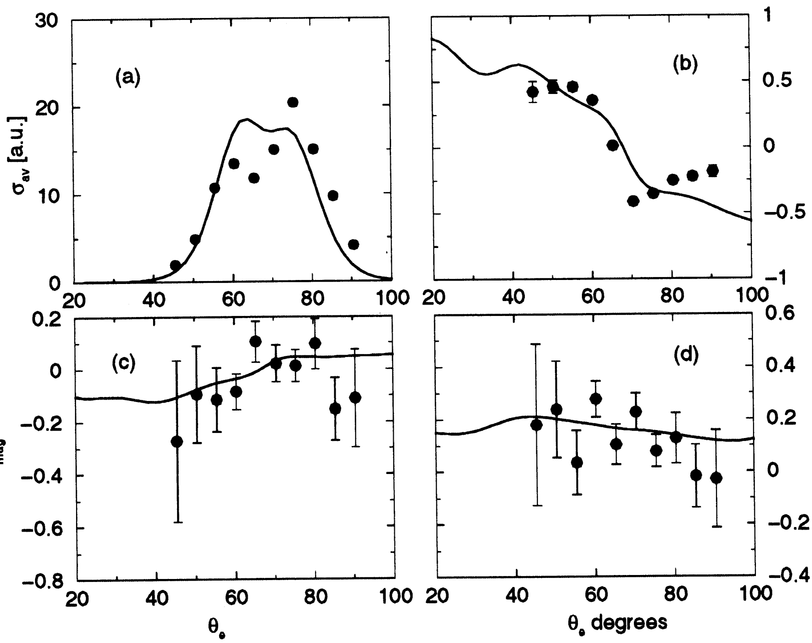

Figure 3 Variation of the scattering parameters A_{avg}, (a), A_{orb}, (b), A_{mag}, (c), and $A_{m,o}$, (d) as a function of scattering angle. The same normalization as in fig1 is applied to the DWBA result in (a).

indicates that this discrepancy can be traced back to the difference in behavior of the experimental and theoretical t_{11} cross section.

Fig. 3b shows the variation of the A_{orb} parameter. The quantity A_{orb} is proportional to the spin averaged *orbital dichroism*. Inspection of relations (10,14) reveals an average over the spin projection of the initial electrons. This quantity can be accessed experimentally by ionization of a laser-oriented sodium atom by an unpolarized electron beam. Consequently, A_{orb} is a polar vector with respect to inversion of the laser's helicity but a scalar in the spin space of the projectile electron, and exhibits the dependence of the ionization cross section on the *orientation* of the atomic target ensemble. The behavior of the A_{orb} parameters is well accounted for in the DWBA model. At small ejected electron angle, θ_e, A_{orb} is large and positive, changing sign around 70 degrees to become large and negative. As can be seen from Fig. 2, this reflects the variation of both the singlet and triplet state resolved cross sections and their respective symmetry around the momentum transfer. It has been noted earlier that A_{orb} is zero when the wave vector of the photon, the momentum transfer vector and the vector momentum of the scattered electron are linearly dependent i.e. are confined to the same plane [3].

In contrast, the parameter A_{mag} shown on Fig. 3c, hereafter referred to as the *magnetic dichroism*, changes sign when the polarization of the incoming electron beam is inverted but remains invariant under a change of the helicity of the photon (cf.Eq. 10 and 14). It describes a spin up-down asymmetry due to a polarized electron beam ionizing an ensemble of *aligned* target atoms. Its origin, however, is more involved than that for A_{orb}, resulting from the m_l dependency of the ionization cross section in both singlet and triplet spin channels. The results, both theoretical and experimental show that this quantity is non-zero over the angular range presented. If the individual singlet and triplet cross sections show no m_l dependence, i.e. if $s/t_{11} = s/t_{-1-1}$, the magnetic dichroism vanishes, as can be seen using (14) and (10). It is also important to stress that the origin of this asymmetry is a pure spin effect, the atomic ensemble defined in equation Eq. 10 possessing finite alignment but zero orientation. A similar effect appears in the electron impact excitation process and the ionization of closed shell systems by polarized electrons and was categorized as “fine structure effect” [15]. This new effect, observed for the first time in the present experimental arrangement is consistent with the DWBA prediction.

Finally the independent parameter $A_{m,o}$ is needed to fully characterize the present measurements. It is an exchange induced antiparallel/parallel spin asymmetry. This quantity is the equivalent of the spin asymmetry parameter introduced to describe (e,2e) ionization of spin polarized targets without orbital orientation by polarized electrons [1], [8]. However, in contrast to the latter spin asymmetry A_s, our present parameter $A_{m,o}$ depends in a subtle way on A_{orb} and as such changes sign if the helicity of the photon is flipped or if the polarization of the incoming beam is inverted, as is clear from Eq.(15). The parameter $A_{m,o}$ is equivalent to the spin asymmetry A_s, only when A_{orb}=0. As previously described, only under this condition does the value of $A_{m,o}$ yield the relative magnitudes of singlet and triplet scattering amplitudes [1]. As can be expected from the results presented on Fig. 2, such a relative contribution appears also well accounted for.

5. CONCLUSION

We have presented (e,2e) ionization cross sections of laser-oriented sodium atoms by polarized electrons under asymmetric reaction kinematics. These results are obtained at an intermediate impact energy of E_0=151 eV where a comparison with the prediction of the DWBA approximation is justified. This comparison, made at the level of each individual cross sections contributing to the average cross section accessible in a conventional (e,2e) experiment, reveals some anomalies for the triplet cross section t_{11} that can not be explained with the present model. The results also show, for the first time, that the initial state resolved ionization cross section depends on both the relative spin projections of the incident and bound state electrons and on the orientation of the target electron. A novel effect termed magnetic dichroism is also demonstrated.

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