Magnetic and Orbital Dichroism in $(e, 2e)$ Ionization of Sodium

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We present the first measurement of $(e, 2e)$ ionization cross sections for a laser oriented atomic target by spin polarized electrons. Cross sections are presented as a function of target orientation and polarization direction of the incident electron beam. This study provides insight into mechanisms by which angular momentum is transferred from the valence electron to the *two* final-state continuum electrons in both singlet and triplet spin channels, by comparing measurement with distorted wave Born approximation and the dynamically screened three Coulomb wave calculations.

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Since the early days of quantum theory electron scattering experiments have been employed as a prototype example to understand and demonstrate various quantum mechanical phenomena. Nowadays, the underlying physics of electron atom elastic and excitation processes are fairly well understood [1]. In contrast, various aspects of electron impact ionization are still unclear due to conceptual and technical difficulties. On the theoretical side, ionization upon electron impact leads to a nonseparable spin-dependent many-body problem (two interacting electrons moving in the field of the residual ion). It is only in recent years that elaborate analytical and numerical approaches have been developed that can describe a considerable range of the continuum spectrum of the electron pair in the field of the residual ion [1–4]. Nevertheless, and as shown in this work, certain gaps in the theoretical treatment and understanding of the ionization processes by electron impact still remain. On the experimental side, a kinematically complete study of ionization requires the energies and directions of the two emerging electrons to be determined in a coincidence experiment. In addition, to completely specify the spectrum of the emitted electron pair, the individual spin projections of the initial- and final-state atoms and continuum electrons need to be determined. For light isotropic atoms a significant step towards this ultimate goal was reported by Baum *et al.* [5,6]. In that experiment, in addition to the vector momenta of the two escaping electrons, the spin projections of the incoming electron and the valence electron of the atom were determined. The experiment consisted of measuring the respective coincidence rates for collisions in which the spin polarization vectors of the incident electron and atomic beams were parallel or antiparallel. From these measurements they determined values for the *spin asymmetry* A_s, a parameter whose origin is purely quantum mechanical and which is closely connected with the Pauli exclusion principle. For light atoms A_s vanishes if either the impinging electron beam or the atomic beam is unpolarized [7].

We now demonstrate, experimentally and theoretically, that this situation changes if the valence electron possesses a defined sense of orbital rotation, i.e., if the direction of the orbital angular momentum is preferentially orientated in space. Previously it was revealed that for an unpolarized electron beam the ionization cross section depends in a characteristic way on the orientation of a target ensemble [8]. This study unravels two novel independent features when a polarized electron beam ionizes an oriented target, namely, a quantity that depends on the polarization of the incoming electron beam but not on the helicity of the exciting photon used to orient the target and an observable that resembles the spin asymmetry A_s but differs markedly in behavior due to the presence of orbital orientation.

Figure 1 shows schematically the experimental and kinematical arrangement described in detail in a previous publication [9]. A collimated target beam of sodium atoms is produced by effusion through a 1 mm diameter exit nozzle of an Ohmically heated recirculating sodium oven. Spin polarized electrons are produced by the photoemission from a cesium and oxygen coated GaAs crystal under illumination by 810 nm circularly polarized laser radiation. The degree of polarization P_e was 0.24 ± 0.03. Inversion of the electron beam polarization from into to out of the scattering plane is achieved by reversing the helicity of the radiation field through rotation of a quarter wave plate.

Pairs of scattered electrons emitted in the scattering plane, defined by the sodium and electron beams, are detected in two independently rotatable electrostatic analyzers on opposite sides of the incident electron beam. Each

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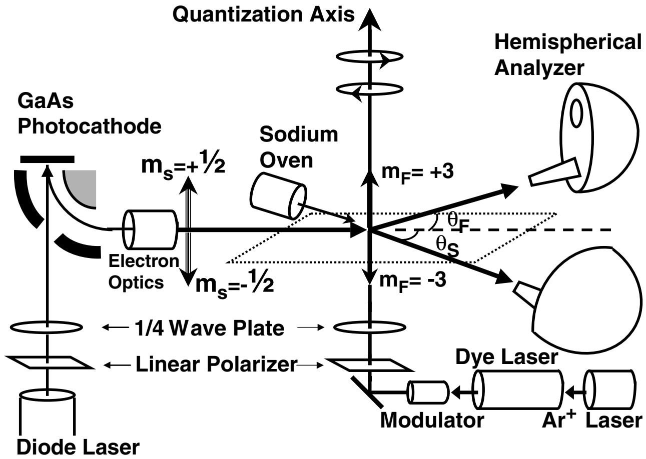

FIG. 1. Schematic representation of the $(e, 2e)$ experiment.

of the two analyzers measures electrons over a 6 eV energy band with an energy resolution of around 300 meV.

Intersecting the scattering plane at right angles and completely encompassing the interaction region is a frequency modulated beam of 589 nm circularly polarized laser light whose purpose is to excite, spin polarize, and orient the target sodium atoms. After a few excitation/decay cycles, the atoms gather exclusively in the two state system

$$\begin{aligned} &3s^1\,{}^2S_{1/2}(F = 2, m_F = +2(-2)) \\ &\leftrightarrow 3p^1\,{}^2P_{3/2}(F = 3, m_F = +3(-3)) \end{aligned} \tag{1}$$

for pumping by left-hand σ^+ (right-hand σ^-) circularly polarized radiation. 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. In this study we focus on the ionization of the excited sodium atoms.

The experiment consisted of measuring $(e, 2e)$ counts as a function of the emission angle θ_S of one of the two final-state electrons, the emission angle θ_F of the other being fixed at 37°, for each of the four combinations of atomic and electron beam polarization directions, namely,

$$\begin{aligned} &e(\uparrow) + \mathrm{Na}(m_F = +3), \qquad e(\uparrow) + \mathrm{Na}(m_F = -3), \\ &e(\downarrow) + \mathrm{Na}(m_F = +3), \qquad e(\downarrow) + \mathrm{Na}(m_F = -3). \end{aligned} \tag{2}$$

The formalism developed to describe spin polarized superelastic scattering experiments on oriented sodium atoms [10] can be generalized to the present situation. Since we are dealing with a light atom and low energies of the escaping electrons, spin-orbit coupling can be neglected [7,11]. In this case the spin degrees of freedom are decoupled and we can then relate the count rates obtained for the four experimentally accessible initial states (2) of the electron-atom system to singlet and triplet initial atomic-state-resolved cross sections as

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

$$\sigma_{t,\Uparrow(\Downarrow)} \equiv K\left[\left(\frac{1}{P_e} + 1\right)N^{\uparrow\Uparrow(\downarrow\Downarrow)} - \left(\frac{1}{P_e} - 1\right)N^{\downarrow\Uparrow(\uparrow\Downarrow)}\right], \tag{4}$$

where $\sigma_{s,\Uparrow(\Downarrow)}$ and $\sigma_{t,\Uparrow(\Downarrow)}$ stand, respectively, for the initial atomic-state resolved singlet and triplet cross sections for positive $\Uparrow$ ($m_F = +3$) and negative $\Downarrow$ ($m_F = -3$) 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. Thus relations (3) and (4) show that the experimental count rates provide all the information required to obtain the different singlet and triplet cross sections, to an overall normalization factor.

Figures 2(a) and 2(b) show the state-resolved singlet and triplet cross sections obtained from measurements performed under symmetric kinematics where the incident electron beam energy E_0 is set at 83 eV and the two outgoing electrons are measured over the energy bands $E_S = E_F = 40 \pm 3$ eV. The cross sections are presented as a function of the ejection angle of one of the two outgoing electrons while the other is fixed at 37° on the opposite side of the incident beam. These figures show the singlet cross sections depend strongly on the initial target orientation ($\Uparrow, \Downarrow$), while the statistical uncertainty resulting from the small magnitude of the triplet cross sections precludes any conclusion being drawn on whether they too exhibit any orientation dependence.

The experimental results are compared to two theoretical models, namely, the distorted wave Born approximation (DWBA) [12] and the dynamically screened three-body Coulomb wave (DS3C) method [4]. The DWBA method accounts for short range interactions in both the entrance and exit channels, i.e., static and exchange with the atom in the entrance channel and with the ion in the exit channels, while treating the two outgoing electrons as independent particles, hence neglecting the electron-electron final-state correlation. In contrast, the DS3C method approximates the three-body final state as three two-body subsystems and accounts for multiple scattering within these subsystems to infinite order. The coupling between the individual two-body subsystems is then described by a dynamical shielding of the interaction strength within the individual two-body subsystems.

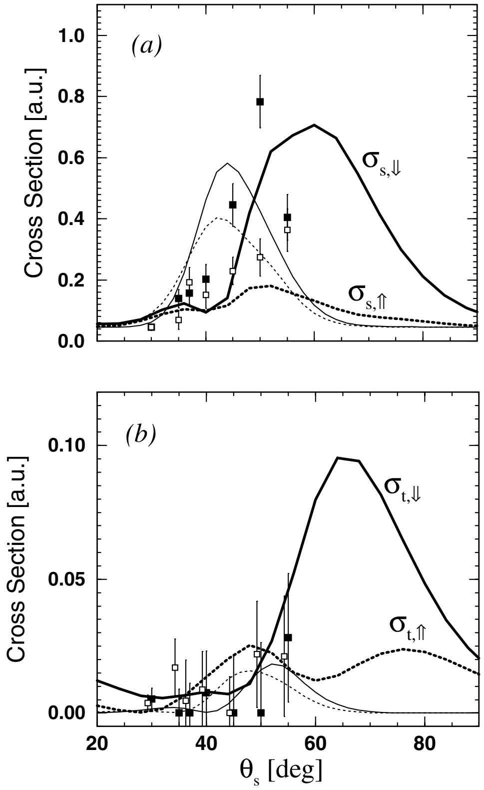

FIG. 2. Singlet $\sigma_{s,\Uparrow(\Downarrow)}$ (a) and triplet $\sigma_{t,\Uparrow(\Downarrow)}$ (b) state resolved $(e, 2e)$ cross sections as a function of electron emission angle θ_S for positive (open squares and dashed lines) ($\Uparrow$) $m_l = +1$ and negative (full squares and solid lines) ($\Downarrow$) $m_l = -1$ target orientations. Primary electron energy E_0 is 83 eV and the fixed emission angle θ_F is 37°. DWBA (thin solid and dashed lines) (multiplied by 0.05) and the DS3C (thick lines) cross sections are calculated at the mean emission energies $\bar{E}_S = \bar{E}_F = 40$ eV.

The absolute scale on the figures is given by the DS3C calculation with the DWBA calculation multiplied by 0.05. The spin averaged cross sections [Fig. 3(a)] are used to normalize the data to the DS3C theory by requiring the best fit for the two peak points. This normalizes all the data.

The DS3C results, and despite the relatively low energies of the measurements also the DWBA results, appear to reasonably describe the relative magnitudes of the various experimental cross sections and confirm the dependence of both the singlet and triplet cross sections on the initial atomic state orientation. For the singlet cross sections the DWBA results peak at significantly smaller angles compared to experiment, whereas the DS3C calculations peak at larger angles. Inclusion of the electron-electron correlation in the description of the final state does not bring the predictions of theory closer to measurement. While the maxima of the theoretical cross sections shift to larger angles, the DS3C method predicts a larger dependence of the singlet cross sections on the initial-state preparation than is observed experimentally. Nevertheless, both theory and experiment show conclusively that under the present kinematics involving symmetric energy sharing between

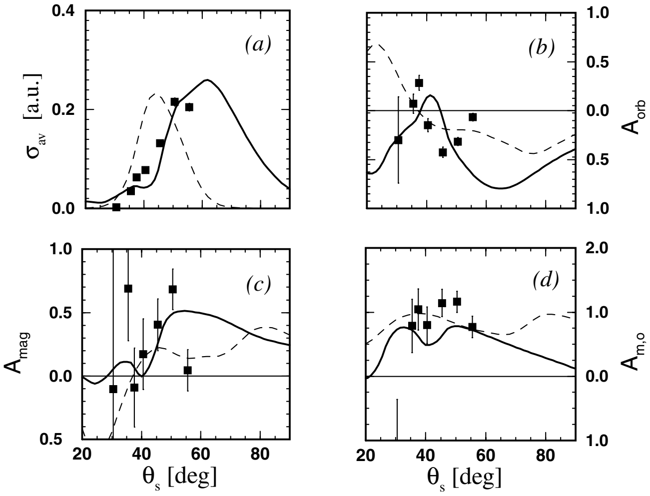

FIG. 3. Variation of the parameters σ_{av} (a), A_{orb} (b), A_{mag} (c), and $A_{m,o}$ (d) as a function of scattering angle. The DWBA (dashed lines) and DS3C (solid lines) are shown. The same normalization as in Fig. 2 is applied in (a).

the two final-state electrons, the collision dynamics is characterized by a dominance of singlet over triplet scattering.

To highlight the interesting new aspects of the ionization collision dynamics revealed by the present measurements we introduce the four tensorial parameters

$$\sigma_{av} = K[N^{\uparrow\Uparrow} + N^{\uparrow\Downarrow} + N^{\downarrow\Uparrow} + N^{\downarrow\Downarrow}] = KN_\Sigma , \quad (5)$$

$$A_{orb} = \frac{1}{N_\Sigma}[N^{\uparrow\Uparrow} + N^{\downarrow\Uparrow} - N^{\uparrow\Downarrow} - N^{\downarrow\Downarrow}], \quad (6)$$

$$A_{mag} = \frac{1}{N_\Sigma P_e}[N^{\uparrow\Uparrow} + N^{\uparrow\Downarrow} - N^{\downarrow\Uparrow} - N^{\downarrow\Downarrow}], \quad (7)$$

$$A_{m,o} = \frac{1}{N_\Sigma P_e}[N^{\uparrow\Downarrow} + N^{\downarrow\Uparrow} - N^{\uparrow\Uparrow} - N^{\downarrow\Downarrow}]. \quad (8)$$

The parameter σ_{av} is a scalar which describes the ionization cross section averaged over the spins of the electrons and the sense of orbital rotation and is independent of the helicity of the laser light. The quantity A_{orb}, defined for a beam of unpolarized electrons, is proportional to the spin averaged *orbital dichroism*. It is a polar vector with respect to inversion of the laser's helicity but a scalar in the spin space of projectile electron and results from the dependence of the ionization cross section on the *orientation* of the atomic target ensemble. In contrast the parameter A_{mag}, 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. (7)]. It describes a spin up-down asymmetry for a polarized beam of electrons from an *aligned* ensemble of target atoms. Its origin, however, is more complicated than that for A_{orb}, resulting from the m_l dependency of the ionization cross section in both singlet and triplet spin channels, and as demonstrated in this work its magnitude can be large. If the individual singlet and triplet cross sections show no m_l dependence, i.e., if $\sigma_{s/t,\Uparrow} = \sigma_{s/t,\Downarrow}$, the magnetic dichroism vanishes, as can be seen using (7) and (3),(4). 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" [13].

Finally the independent parameter $A_{m,o}$ is needed to fully characterize the present measurements. It is an exchange induced antiparallel/parallel spin asymmetry 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. (8). In contrast to the spin asymmetry A_s, which results from the electron impact ionization of spin polarized electrons from spin polarized targets with no orbital orientation [5], our present parameter $A_{m,o}$ depends in a subtle way on A_{orb}. To illustrate this we recall that for the ionization from isotropic states and in the doubly symmetric kinematics (i.e., $\theta_S = \theta_F$ and $E_S = E_F$) the triplet cross section vanishes due to its odd symmetry behavior with respect to exchange of the two escaping electrons [5]. For this case $A_{m,o}$ tends to unity. In contrast, however, for oriented targets the triplet cross sections are generally finite in the case of doubly symmetric kinematics, because the symmetry of space is broken by the presence of the laser light, i.e., the exchange of the two electrons does *not* correspond to a symmetry operation under which our experiment is invariant (π rotation around the incoming beam direction in absence of the laser). Therefore $A_{m,o}$ does not need to tend to unity in the doubly symmetric kinematics. On the other hand if A_{orb} is indeed zero, for the doubly symmetric kinematics the experiment becomes invariant under a π rotation around the incoming beam leading to a vanishing triplet cross section and therefore $A_{m,o} \rightarrow 1$. Within the first Born approximation this is the case when the wave vector of the photon, the momentum transfer vector, and the vector momentum of the secondary electron are linearly dependent, i.e., are confined to the same plane.

This interplay between A_{orb} and $A_{m,o}$ is confirmed in Fig. 3(b) and 3(d). At $\theta_S = 37°$ we reach the doubly symmetric kinematics, and $A_{m,o}$ should tend to unity if $A_{\mathrm{orb}} = 0$. This is the case within the DWBA model, however within the DS3C model $A_{\mathrm{orb}} \neq 0$ at $\theta_S = 37°$ and hence $A_{m,o} \neq 1$. In fact the DS3C theory anticipates (in contrast to the DWBA model) $A_{\mathrm{orb}} = 0$ at $\theta_S \approx 38°$ and again at $\theta_S \approx 44°$ with a maximum in between at $\theta_s \approx 41°$ that leads to the minimum in $A_{m,o}$ at $\theta_S \approx 41°$. The behavior of A_{orb}, shown in Fig. 3(b), results in the double structure of $A_{m,o}$. The theoretical results of the DS3C model give reasonable predictions of the shapes of the measured A_{orb} and $A_{m,o}$, but some deviations in the magnitudes and positions of the peaks are observable.

Figure 3(a) shows the measured parameter σ_{av}. The reasonable agreement between both theories and experiment is not surprising as σ_{av} is an averaged quantity.

Figure 3(c) shows the measured magnetic dichroism along with the theoretical results. The DS3C theory shows a double hump structure due to the small minimum in A_{mag} around $\theta_S \approx 40°$. The reason for this minimum can be understood from Eqs. (7), (3), (4), and Fig. 2(a), in which the singlet cross sections are depicted. As is clear from Fig. 2(a) at $\theta_S \approx 40°$ the singlet cross sections become insensitive to the inversion of the helicity of the laser, i.e., $\sigma_{s,\Downarrow} = \sigma_{s,\Uparrow}$ at $\theta_S \approx 40°$. From Eqs. (3), (4), and (7) it is then readily deduced that A_{mag} diminishes in this case [the triplet cross sections are quite small in the present case as is clear from Fig. 2(b)].

We have presented experimental results for $(e, 2e)$ cross sections where the initial state of the projectile-target system is completely resolved by employing a polarized electron beam and an atomic beam prepared in a well defined quantum state by laser pumping. The measurements involve the ionization of the $3p\,^1\,^2P_{3/2}(F = 3, m_F = +3$ or $m_F = -3)$ hyperfine states of sodium by spin polarized electrons. The results show for the first time that the initial state resolved ionization cross section depends both on the relative spin projections of the incident and bound state electrons and on the orientation of the target electron. While aspects of the experimental cross sections are reproduced by the two theoretical models, a lack of detailed agreement is observed in both cases. For the DS3C calculations, where agreement with the shape of the nonstate resolved cross section is good, the large deviations observed in the spin and orbital angular momentum resolved partial cross sections suggest that the state specific measurements provide the most stringent tests to the theory of the ionization process.

[1] I. E. McCarthy and E. Weigold, *Electron-Atom Collisions* (Cambridge University Press, Cambridge, England, 1995).
[2] I. Bray and A. Stelbovics, Adv. At. Mol. Phys. **35**, 209 (1995).
[3] T. N. Rescigno, M. Baertschy, W. A. Isaacs, and C. W. McCurdy, Science **24**, 2474 (1999).
[4] J. Berakdar, Phys. Rev. A **53**, 2314 (1996).
[5] G. Baum *et al.,* Phys. Rev. Lett. **69**, 3037 (1992).
[6] M. Streun *et al.,* J. Phys. B **31**, 4401 (1998).
[7] J. Kessler, *Polarized Electrons* (Springer-Verlag, Berlin, 1985), 2nd ed.
[8] A. Dorn *et al.,* Phys. Rev. Lett. **80**, 257 (1998).
[9] J. Lower *et al.,* Phys. Rev. A **62**, 012706 (2000).
[10] I. V. Hertel, M. H. Kelley, and J. J. McClelland, Z. Phys. D **6**, 163 (1986).
[11] I. Percival and M. J. Seaton, Philos. Trans. R. Soc. London A **251**, 113 (1958).
[12] I. E. McCarthy, Aust. J. Phys. **48**, 1 (1995).
[13] S. Jones, D. H. Madison, and G. F. Hanne, Phys. Rev. Lett. **72**, 2554 (1994); X. Guo *et al., ibid.* **76**, 1228 (1996).