Electron correlations and spin effects activated by excitation of an inner 3d electron in zinc

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Abstract. Excitation of the 4s4p^3P_0,1,2 states of zinc was studied in the energy region of the lowest 3d^{10} autioinizing state using spin polarized incident electrons. The energy dependence of angular differential excitation intensities was measured for incident electrons with spin up and down. Negative ion resonances with a 3d^94s^24p^2 electron configuration show prominent structure in the angular differential cross section and also show significant spin dependence which is prominent especially at 90°.

1. Introduction

Experiments with incident spin polarized electrons have revealed subtle details of momentum coupling in a target atom and how this coupling may influence the scattering dynamics [1]. Our previous studies of inelastic scattering of polarized electrons from free zinc atoms explored the spin effects in the scattering process of ionization-with-excitation of open and closed 3d-shell states [2]. Also measurements of the three Stokes polarization parameters [3] of the 3d^{10}4s4d^1D_2 → 3d^{10}4s4p^1P_1 decay photons observed and disentangled the spin orbit and exchange effects in excitation via 3d core-excited negative ion resonances. Here we report on the difference in the angular differential cross sections for excitation of the 3d^{10}4s4p^3P states for spin-up and spin-down incident electrons in the energy region where excitation is affected by 3d core excited negative ion resonances. A combination of different experimental techniques provides complementing information.

Asymmetries for electrons scattered left and right in elastic and inelastic collisions have been measured previously for a series of atoms [4,5,6]. In elastic scattering from atoms with a closed shell ground state electron configuration the polarization effects are caused only by the spin-orbit interaction of the scattered electron in the field of the target atom, that is, Mott scattering. In inelastic scattering, polarization effects may be caused also by the exchange interaction between the scattered and atomic electrons, the inter-atomic spin-orbit interaction and their combined effect [1]. Left/right asymmetries for zinc have been measured previously only for elastic scattering [4] and when experimental conditions were not adjusted to observe the effects of negative ion resonances. Here we study excitation via a negative ion resonance with the open 3d shell structure of a zinc atom for which there is a significant distortion of the atomic charge cloud such that the associated angular momentum effects can be observed [4,7]. In this way a clear distinction of the spin-orbit interaction via fine structure effect and via Mott scattering is possible.
2. Experimental

The apparatus used in the present study consists of a scattering chamber incorporating a cylindrical 127° scattered electron analyzer, an evaporative source of free zinc atoms and a source of spin polarized electrons. The scattering chamber and components used for detection and characterization of the collision products, scattered electrons and decay photons, are described elsewhere [8]. Briefly, electrons scattered by the beam of free zinc atoms are collected, energy analyzed and detected at a variable angle using a single channel electron multiplier. Two different modes of operation of the electron spectrometer were used, namely an energy loss mode to identify and isolate a particular scattering process and secondly an excitation function mode to observe the energy dependence of the angular differential cross section. In the latter mode, the incident electron energy and residual scattered electron energy were scanned simultaneously over a fine energy mesh to observe negative ion resonances.

![Figure 1](image.png)

Figure 1. Representation of the spin polarized electron source and beam. The left-hand section shows the source and scattering chambers with dotted lines within which are (h) the electrostatic 180° energy selector; (i) the isolation valve between UHV source chamber and the scattering chamber; (j) the electron optical lenses for transfer and (k) focusing of electrons into the scattering region. The right-hand section shows the components used to obtain the transversely spin polarized electrons: (a) laser; (b) linear polarizer; (c) liquid crystal variable retarder; (d) circularly polarized laser light passing through the vacuum chamber window; (e) GaAs crystal; (f) electrostatic 90° deflector from which electrons initially longitudinally spin polarized emerge with spins $\mathbf{P}$ perpendicular to their momentum vector $\mathbf{k}$ as indicated by the vector diagram.

A new feature of the present experiment, different from the one previously used in our laboratory for the study of negative ion resonances in excitation of zinc [8], is the source of spin polarized electrons. While the general design principle of this source is the same as described by [9,10], here we employ a 180° electrostatic electron energy selector to reduce the energy spread in the incident electron beam. The need for a high efficiency in data collection and the relatively large natural width of the observed features justified the use of an energy resolution of 120 meV. The electron optics controlling the electron beam and the 180° energy selector are shown in the left-hand section of figure 1. The source of the spin polarized electrons is indicated on the right hand section of the figure.
The spin polarized electrons are obtained via photoemission from the surface of a GaAs crystal irradiated by circularly polarized 830 nm wavelength laser light. A spin polarization $P_c=28\%$ was used. The photo electrons are emitted with the spins parallel or anti-parallel to the incident laser light direction. This initially longitudinal spin polarization is changed to transverse by deflecting the electron beam through 90° so that in the interaction region the electron spin is perpendicular to the scattering plane. Spin orientation, from spin-up to spin-down, is changed using a liquid crystal variable retarder controlled by data acquisition software. For each incident electron energy the intensity of electrons scattered at a given angle for spin up, $I^\uparrow$, and spin down, $I^\downarrow$, was recorded over a period of a few seconds in repeated energy scans. The spin up-down asymmetry is determined from the relation

$$S_A=I/P_c(1-I^\uparrow)/(1-I^\downarrow)$$  \hspace{1cm} (1)

The energy scale was calibrated by detecting the photons from the decay of the 4s4d$^1D_2$ state while scanning the incident electron energy through the excitation threshold which for this state has a step-like shape [11]. The cross section is then fitted to theoretical BSRM data [12] as described previously [13] to obtain a calibration of the incident electron energy scale.

$\begin{align*}
\text{Figure 2. Electron energy loss} \\
\text{spectrum in zinc using spin polarized} \\
\text{incident electrons with spin up and spin} \\
\text{down with respect to the scattering} \\
\text{plane defined by the momentum vectors} \\
\text{of the incident and detected scattered} \\
\text{electrons.}
\end{align*}$

3. Results and discussion

The electron energy loss spectrum, measured at an incident electron energy of 11.3 eV and scattering angle of 54° for spin up and spin down electrons, is shown in figure 2. The energy resolution and relative intensities of different excitation processes are well shown. The energy loss peaks, corresponding to the excitation of the 4p$^3P_0,1,2$ and 4p$^1P_1$ states studied here, are well isolated from each other and from other excitations and also from the elastic peak, (not shown in the spectrum). From this spectrum one can see the following features. At an electron impact energy of 11.3 eV and a scattering angle of 54°, a non-zero spin up-down asymmetry in excitation of both the 4s4p$^3P_0,1,2$ and 4p$^1P_1$ states is indicated by different intensities for spin-up and spin-down incident electrons. For these specific scattering conditions of the incident energy and scattering angle, the asymmetry is negative for the triplet state and positive for the singlet state. Also, the variation of the asymmetry is observed along the 4p$^3P_0,1,2$ peak where there are larger values in the high energy wing. The fine structure components, the $J=0$ (at 4.006 eV), $J=1$ (at 4.030 eV) and $J=2$ (at 4.078 eV), of the lowest excited 4s4p$^3P_0,1,2$ states are not resolved and the observed variation of asymmetry might be associated predominantly with the $J=2$ excitation. In contrast, the asymmetry associated with the 4s4p$^1P_1$ excitation does not vary along the corresponding energy loss peak, as expected.
Figure 3. Scattered electron intensities following excitation of $4s4p^3P_{0,1,2}$ states using unpolarized electrons (top row) and spin polarized electrons with spin up (triangles) and spin down (closed circles) (middle row). The spin up-down asymmetry [calculated using expression (1)] is shown in the bottom row. The vertical bars and labels indicate the negative ion resonance values recommended by Napier et al 2009 [14].

The energy dependence of the angular differential scattered electron intensities and spin up-down asymmetries for excitation of the $4s4p^3P_{0,1,2}$ states are shown in figure 3 for scattering angles of 30°, 54° and 90°. The top spectrum at each angle was obtained using a source of unpolarized electrons [8] which was optimized for low energy electrons. The middle spectrum was measured with the new source (see above) of spin polarized electrons. The two sets of data correspond to the two orientations of spin up and down. As shown on the figure, the smoothly varying continuous part of the cross section in the experiments with unpolarized and spin polarized electron beams is very similar. That indicates a negligible change of transmission of electron optical parts in the energy region studied here. Also the appearance of structures caused by the scattering via negative ion resonances is in good general agreement for unpolarized and spin polarized electron beams is very similar. That indicates a negligible change of transmission of electron optical parts in the energy region studied here. Also the appearance of structures caused by the scattering via negative ion resonances is in good general agreement for unpolarized and spin polarized electron beams is very similar.

As a consequence the spin up/down asymmetry increases progressively with increase of scattering angles from a variation inside the -0.10 to +0.07 range at 30°, to a -0.25 to +0.35 range at 90° respectively.
The resonance structure indicated by vertical bars in figure 3 and labeled a a’, b b’, c c’ c’’ and d, was investigated previously, and here is observed clearly in the intensities but less clearly in the asymmetry functions. The asymmetries at all scattering angles in figure 3 are dominated by a broad smooth variation with energy with a hint of a possible doublet structure for the first broad peak at 30°, and a clearly resolved doublet at 54° where the maxima align best with the resonances a’ and b. No clear indication of individual effects of any of the resonances a to d is observed at 90°.

While the observed variation of the asymmetry function is related clearly to negative ion resonance phenomena and indicates the spin dependence in the creation and decay of these short-lived negative ions, an association with individual a to d resonances is less clear. The behavior of the spin up-down asymmetry at 90°, and to some extent at other angles, can be viewed as an unresolved contribution of several broad overlapping and possibly interfering resonances, but another explanation in terms of the existence of shape resonances as inferred from both the elastic scattering [14] and excitation [8,15] needs to be considered. Most of the resonances in the lowest autoionizing region, shown in figure 3 with labels a to d, have been assigned a 3d^94s^24p^2 electronic configuration. The angular differential elastic scattering indicates that the labeled a’,b b’, c c’ resonances can be characterized by total angular momentum J=5/2 and 3/2. In the absence of any theoretical modeling the symmetries of corresponding states are discussed by comparison to the corresponding autoionizing states of a Gallium atom. In view of these results some of the properties of the asymmetry function can be discussed further using a simple picture [1,16]. Based on conservation of angular momentum, the collisional alignment of the excited atomic particle and the assumption that spin and orbital momentum have well defined values, the resonance scattering of electrons with spin-up and spin-down will be different and will excite preferentially negative ions with J=5/2 or J=3/2 if the scattered electrons are detected at a positive scattering angle. This observation will correspond to the orientation of orbital momentum and spin in the direction of the positive z-axis. If the effect of a resonance was to enhance the cross section, then this would lead to an absence or reduction of one of the J=5/2 or J=3/2 components and would be observable in the spin up/down asymmetry. For example, in the case of the 54° scattering in figure 3, the structures in the asymmetry values seem to be associated best with the a’ and b resonance energies which were determined separately from electron transmission measurements [14]. Consequently their positive asymmetries indicate J=5/2 state assignments for both of them. This deduction is in agreement with the assignments for b but not for a’ by Napier et al [14] who proposed J=5/2 and 3/2 respectively. The proposed a’ assignment is also contrary to the situation in Ga where the J=3/2 state is lower in energy for the equivalent isoelectronic auto-ionizing states. However, if angular dependent interference exists, either with non-resonant scattering or between the resonances themselves, the picture would be less straightforward to interpret and theoretical modeling would be essential. For a meaningful comparison with experimental data there is a need for theoretical models to investigate the effects of negative ion resonances and their associated spin effects in both the integral and angular differential cross sections.

The previous experimental studies using spin polarized electrons reported the angular behavior of asymmetries in elastic scattering but the widely spaced energies did not permit the detection of resonances. Measurement of spin dependent Stokes polarization parameters by detecting the 4d^1D_2→4p^1P_1 decay photons by Pravica et al [3] was the first to observe a spin dependence associated with the unresolved resonances b b’ and also c’ specific to this scattering channel. That study used predictions from Bartschat and Blum [15] to identify and disentangle the roles of the spin orbit and exchange interactions. Similar theoretical analyses for the spin asymmetries are not yet clear. The present data with overlapping unresolved resonances and possible interference effects requires a theoretical study specifically for zinc atoms.

4. Acknowledgements
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