PHYSICAL REVIEW A **83**, 040701(R) (2011)

Unexpected effects in spin-polarized electron-impact excitation of the $(3d^{10}4s5s)^3S_1$ state in zinc

L. Pravica,[1,2] J. F. Williams,[1,2,*] D. Cvejanović,[1,2] S. Samarin,[1,2] K. Bartschat,[3] O. Zatsarinny,[3] A. D. Stauffer,[4] and R. Srivastava[5]

[1]*Centre for Atomic, Molecular and Surface Physics (CAMSP), School of Physics M013, University of Western Australia, Crawley, Perth 6009, Australia*

[2]*ARC Centre for Antimatter-Matter Studies (CAMS), School of Physics M013, University of Western Australia, Crawley, Perth 6009, Australia*

[3]*Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA*

[4]*Department of Physics and Astronomy, York University, Toronto, Ontario, Canada M3J 1P3*

[5]*Department of Physics, Indian Institute of Technology, Roorkee 247667, India*

(Received 2 August 2010; published 8 April 2011)

Observations of the $(3d^{10}4s^2)^1S_0 \rightarrow (3d^{10}4s5s)^3S_1$ excitation process in zinc, with incident spin-polarized electrons, show significant deviations from expectations for a pure electron exchange transition. This result, derived from values of up to 10% of the Stokes parameter P_2 (aligned linear polarization) of the light emitted in the $(3d^{10}4s5s)^3S_1 \rightarrow (3d^{10}4s4p)^3P_{0,1,2}$ optical decays, is very different from predictions of near-zero values from general theory as well as sophisticated, relativistic close-coupling and distorted-wave numerical models. Although the linear light polarization P_2 may be nonzero by parity conservation, it was expected to vanish due to dynamical symmetries.

DOI: 10.1103/PhysRevA.83.040701 PACS number(s): 34.80.Dp

Spin-polarized electron beams are well known as an excellent tool to unravel effects in physics that would go undetected if a potentially preferred spin projection is either not prepared in the initial state or not observed in the final state of a scattering experiment. In these cases, one averages over the initial spin orientations and sums incoherently over the final ones. The result is a loss of potentially valuable information that could, in principle, be obtained according to the laws of quantum mechanics. A comprehensive treatment of spin-polarized electron physics was given by Kessler [1] and updated with examples of "complete experiments" by Andersen and Bartschat [2]. The key uses of spin-polarized electrons take advantage of the facts that (i) the spin can sometimes be used to "mark" an electron and thus obtain additional information about exchange processes, and (ii) the spin is associated with a magnetic moment and hence can be used to study magnetic interactions, including relativistic effects such as the spin-orbit interaction. The area of possible applications is enormous, ranging from very fundamental studies such as parity violation in elementary particle, nuclear, and atomic physics, as well as the investigation of exchange, spin-orbit, and other spin-dependent interactions in atomic and molecular structures and collisions, all the way to the study of magnetic materials, thin films, surfaces, and material research in general.

Due to the multiple effects that can, in principle, be studied with spin-polarized electrons, an important aspect regarding their use in obtaining the most detailed information concerns the possibility, or lack thereof, of studying a particular effect that is sensitive to the electron spin. In electron-scattering experiments, two very significant spin-related effects are electron exchange and the spin-orbit interaction. For instance, it is well known that electron-impact excitation of a light target, such as helium in its $(1s^2)^1S_0$ ground state, from a singlet to a triplet state can only occur through electron exchange. On the other hand, an unpolarized electron beam scattered elastically from a heavy target, such as Hg in its $(6s^2)^1S_0$ ground state, becomes spin polarized due to the spin-orbit interaction, an effect known as Mott scattering [1]. In general, however, the two effects are entangled and it is usually impossible to determine unambiguously which of the two aspects are the most responsible for a certain outcome that requires at least one of them. An example of such an effect is the electron-impact excitation of the $(6s6p)^3P_1$ state in mercury. In this case, the target is sufficiently heavy so that the spin-orbit interaction for the projectile should not be ignored. In addition, this state should be described in "intermediate coupling." Whereas the total orbital angular momentum ($L = 1$) is a reasonably good quantum number, the total spin ($S = 1$) is definitely not. As a result, there is a singlet ($S = 0$) admixture in the description of the state.

Figure 1 shows the geometry of the present experiment, where the spin polarization $\boldsymbol{P}_e$ defines a plane that contains also the linear momentum $\boldsymbol{k}_0$ of the incident electrons. The polarization patterns of the emitted photons can be described by the Stokes parameters P_1, P_2, and P_3 defined as

$$P_1 = [I(0^\circ) - I(90^\circ)]/[I(0^\circ) + I(90^\circ)], \quad (1)$$

$$P_2 = [I(45^\circ) - I(135^\circ)]/[I(45^\circ) + I(135^\circ)], \quad (2)$$

$$P_3 = [I(\sigma^-) - I(\sigma^+)]/[I(\sigma^-) + I(\sigma^+)]. \quad (3)$$

Here $I(\theta)$ corresponds to the photon intensities measured for the linear polarizations, with the polarizer transmission axis oriented at an angle θ with respect to the incident electron-beam direction, and for circular polarization P_3 with the positive (σ^+) or negative (σ^-) helicity. If a transversally spin-polarized electron beam is used, the *geometrical* planar symmetry allows for the linear and circular light polarizations

*jfw@cyllene.uwa.edu.au

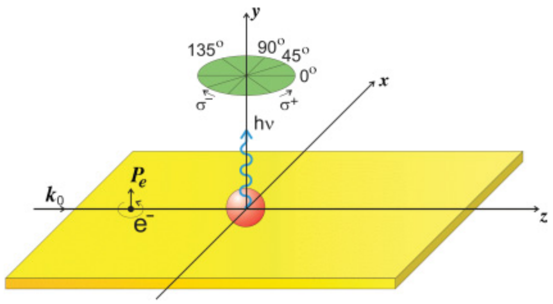

FIG. 1. (Color online) Geometry of the experiment. The symbols are explained in the text. The scattered electrons are not observed.

to be nonzero. Indeed, this was measured many years ago for the particular transition in Hg mentioned above [3], and for many other transitions subsequently [4].

The original purpose of our experiment was to test the dynamical assumptions made by Bartschat and Blum [6], in particular to find out to what extent P_2 deviates from exactly zero in reality. There are a few special cases for which very general predictions about the light polarizations can be made, even without explicit numerical calculations. These arguments are based on assumptions regarding additional *dynamical* symmetries about the excitation process. One such case concerns the excitation of the $(4s^2)^1S_0 \to (4s5s)^3S_1$ transition in Zn. The total electronic angular momentum $J = 1$ is simply a mathematical coupling of what are generally believed to be good quantum numbers, namely $S = 1$ and $L = 0$. Populating this state in a straightforward excitation process, below the threshold for possible cascade populations from higher-lying states, should result in the outcome

$$P_1 = P_2 = 0, \quad P_3 = f P_e. \tag{4}$$

Here f is a factor that depends on the final state of the optical decay, effectively a Wigner $6j$ symbol, and P_e is the component of the electron polarization vector along the direction of light observation.

Due to its simplicity, this very system was suggested, and has been used, for an optical determination of the electron-spin polarization [5]. Bartschat and Blum [6] further analyzed such excitations and showed that the above result, especially the vanishing of P_2, is based on the assumptions that the spin-orbit interaction for the projectile is negligible and the spin-orbit interaction in the target is sufficiently small that L, S, and J are all good quantum numbers. We reemphasize that by purely *geometric* arguments, based on the plane defined by the initial electron momentum and the transverse-electron polarization, P_2 could be nonzero without violating parity conservation. By that argument alone, P_2 would need to be proportional to P_e, while P_1 is independent of P_e. However, the additional *dynamical* symmetries of this particular transition would require P_2 to vanish nevertheless.

The apparatus described in detail in [7] are ideal for this investigation. A transversely polarized electron beam, produced by photoemission from a GaAs crystal by a circularly polarized 830-nm laser light, excites Zn atoms from a resistively heated oven in a crossed-beams geometry. The spin polarization is typically 66% or 29% from strained or unstrained GaAs crystals, respectively. The light emitted in the decay of the $(4s5s)^3S_1$ state to the $(4s4p)^3P_{J_f}$ final states, with total electronic angular momenta $J_f = 0,1,2$ with wavelengths of 468.1, 472.3, and 481.1 nm, respectively, is detected by an EMI 9863QB photomultiplier. Wavelength selection is achieved by interference filters or an acousto-optic tunable filter, and the light polarization is analyzed by a combination of a liquid-crystal variable retarder with a linear polarizer or by a rotating linear polarizer.

The Stokes parameters P_1, P_2, and P_3 measured for the decay of the $(4s5s)^3S_1$ state into each member of the $(4s4p)^3P_{0,1,2}$ fine-structure multiplet are shown in Fig. 2. As expected, an unpolarized electron beam yields the required (by parity conservation) zero result for P_2. However, we draw attention to the significantly nonzero values of P_2/P_e in the cascade-free region within about 1 eV above threshold. At first sight, these data for this excitation process simply suggest a serious violation of the assumptions made by Bartschat and Blum [6] regarding the dynamics of the process. We will see, however, that both P_1 and P_3 conform very well to these expectations. Furthermore, even the most sophisticated, current numerical collision models (see below) are unable to reproduce the experimental findings.

We now discuss the experimental results in some detail. To begin with, the large step seen in all the Stokes parameters at about 7.6 eV is caused by cascade from the $(4s5p)^3P_{0,1,2}$ states to the $(4s5s)^3S_1$ state, as observed independently by Eminyan and Lampel [5] for P_3. The values of P_1 at 8.0 eV, i.e., just above the step, in the three observed transitions are 0.058 ± 0.002, -0.031 ± 0.002, and 0.01 ± 0.004. These agree well, within experimental uncertainties, with the calculated relative factors of 1, $-1/2$, and $1/10$ from the respective $6j$ symbols that determine the optical decay [2]. Before the onset of cascade effects, P_1 vanishes within the experimental uncertainties, as one would expect from the spherical electron charge cloud of a well-LS-coupled $^3S_1$ state. These results provide the first strong support for the validity of the linear polarization measurements, since both P_1 and P_2 were measured under the same experimental conditions.

After correcting for the depolarization due to hyperfine interaction, the circular polarization P_3 for the three transitions in the cascade-free region is expected as $-P_e$, $-P_e/2$, and $P_e/2$, respectively [5]. Once again, the factors -1, $-1/2$, and $1/2$ are due to a $6j$ symbol in the formula for the optical decay. The observed P_3/P_e values of -0.986, -0.487, and 0.482 agree with these expectations within our experimental uncertainties. This provides further confidence in the measurements.

It thus appears that the excitation from the ground state $(3d^{10}4s^2)^1S_0$ to the excited state $(3d^{10}4s5s)^3S_1$, in which no net orbital angular momentum is transferred, proceeds primarily via electron exchange. Nevertheless, the small difference from the expected values allows for the possibility of some angular momentum being transferred via a different mechanism.

The main result of the present work is seen in the center column of Fig. 2, which reveals significant nonzero values of P_2/P_e within the first eV above threshold. In this energy regime, the measured values of $P_2/P_e = -0.12 \pm 0.005$, 0.06 ± 0.007, and -0.015 ± 0.012 for $J_f = 0,1,2$ are in agreement with the depolarization factors expected for linear

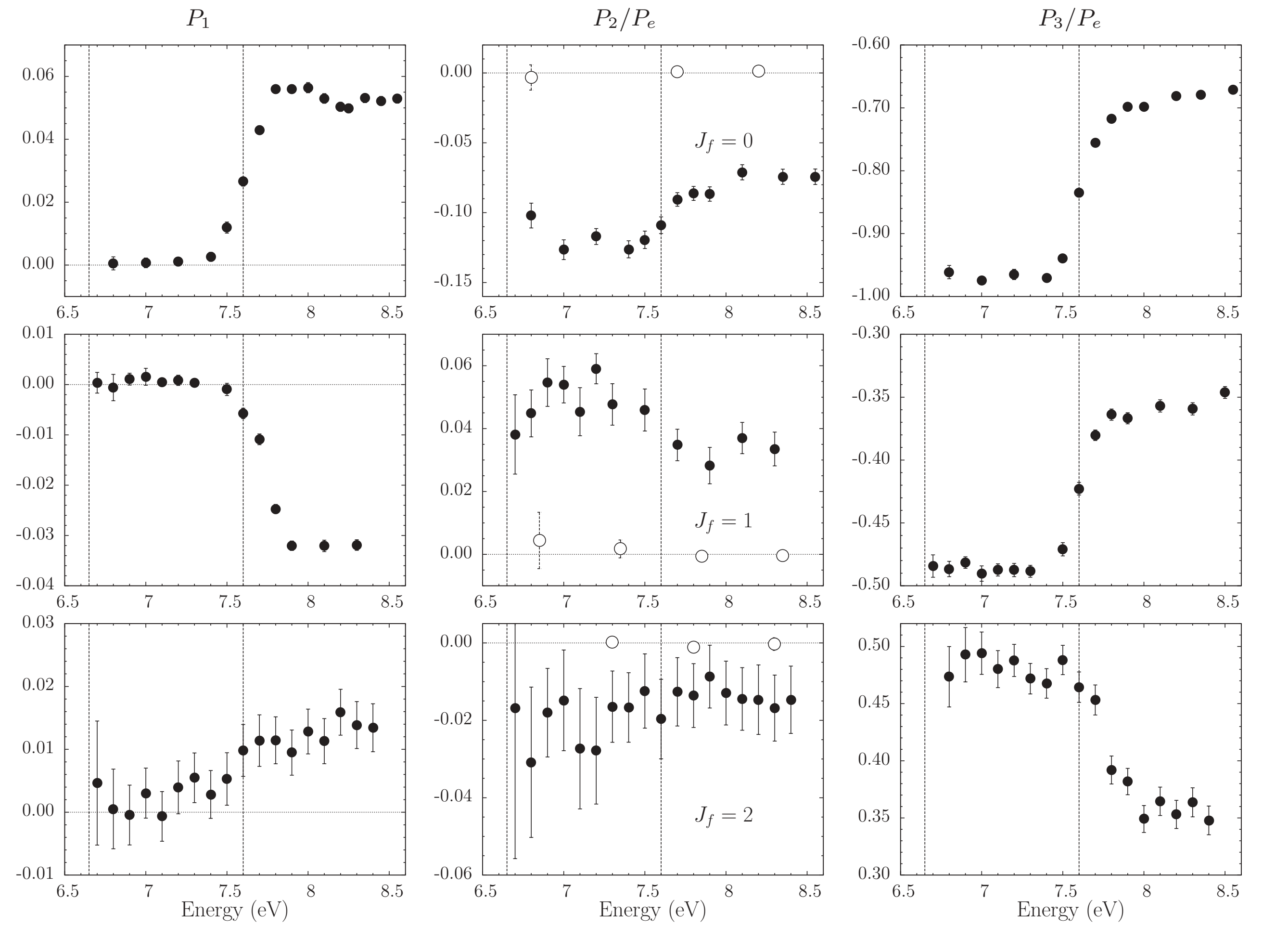

FIG. 2. Stokes parameters P_1 (left column), P_2/P_e (center column), and P_3/P_e (right column) for the $(4s5s)^3S_1 \rightarrow (4s4p)^3P_{0,1,2}$ decay transitions in zinc. The vertical lines at 6.65 and 7.6 eV indicate the excitation thresholds of the $(4s5s)^3S_1$ state and the first cascading $(4s5p)^3P_J$ state, respectively. The filled circles represent our data with incident spin-polarized electrons, while the open circles are those obtained with an unpolarized electron beam. By dividing the measured values for P_2 and P_3 by P_e (when $P_e \neq 0$), these results are normalized to $P_e = 100\%$. Note the offset zero and change of scale between panels.

polarization measurements involving those final states, i.e., the same factors as for P_1. We note that the degree of polarization, $P = \sqrt{P_1^2 + P_2^2 + P_3^2}$, must be less than unity within experimental uncertainties. For $J_f = 0$, we obtain $P = 0.99 \pm 0.09$. This value indicates that the vector polarization is near a pole on the Poincaré sphere. Hence a small deviation of P_3 from unity may be just as important as a much larger variation in P_2.

Expecting to explain these results as the simple need of going beyond the analytical assumptions made by Bartschat and Blum [6], we set up a number of numerical calculations in which relativistic effects were accounted for, either at the level of first-order perturbation theory by calculating matrix elements of the Breit-Pauli Hamiltonian between states generated from nonrelativistic one-electron orbitals, or by calculating both the target structure and the wave function of the projectile using the Dirac-Coulomb approach. Two semirelativistic *R*-matrix (close-coupling) models were employed. The first one was based on the work described by Sullivan *et al* [8]. Here the well-known Belfast *R*-matrix code [9] was used to couple the lowest seven target states of Zn, i.e., $(4s^2)^1S_0$, $(4s4p)^3P_{0,1,2}$, $(4s5s)^3S_1$, and $(4s5s)^1S_0$. The second calculation was performed with the much more sophisticated *B*-spline *R*-matrix (BSR) code [10]. For *e*-Zn collisions [11], 23 physical states and pseudostates were coupled, and the model was later extended to 49 states [12]. These models seemed very appropriate for the near-threshold excitation and generally provided excellent agreement with the many observed resonance features [11].

The spin-orbit interaction of the projectile and the possibility of intermediate coupling in the description of the target states are contained within the semirelativistic calculations. There could be admixtures from other configurations with the same ^3S LS term, plus contributions from ^3P, ^1P, or ^3D that could also result in $J = 1$. The latter were found to be extremely small (the largest coefficient had a magnitude of 0.00002), and hence one would still expect vanishing linear light polarizations P_1 and P_2 according to the Bartschat and Blum [6] theory. Even the other ^3S admixtures were small, resulting in a nearly pure (>99.8%) $(3d^{10}4s5s)^3S_1$ state.

Fully relativistic distorted-wave (RDW) [13] calculations for the process were also performed. All one-particle relativistic effects, including spin-orbit coupling, were included in the bound states and the scattered wave. The GRASP2K program [14] was used to generate the ground and the excited $^3S_1$ state, including the configurations $3d^{10}4s^2$, $3d^{10}4s4d$, $3d^{10}4s5s$, $3d^94s^24d$, and $3d^94s^25s$. Again the $3d^{10}4s5s$ state was found to be very well described in pure single-configuration LS coupling.

Based on the dominance of the LS-coupled $4s5s$ configuration in the description of the $^3S_1$ state, it is not surprising that the largest values of $|P_2/P_e|$ obtained in any of our calculations were in the range of 10^{-4}. This is three orders of magnitude smaller than the observations. The remaining numerical values, $|P_1| \leqslant 10^{-5}$ and $P_3/P_e = -1, -0.5$, and $+0.5$ depending on J_f of the final state, were also in excellent agreement with the analytical predictions of the Bartschat and Blum model [6]. Hence, we do not show results from these calculations.

In light of these serious discrepancies between experiment and theory, extensive tests were made to ensure the validity, accuracy, and precision of the measurement techniques. The complete set of results, taken as a whole, provides further, almost uncontestable proof, since it is indeed fully self-consistent. If any one result were incorrect, it would have affected many others.

Measurements showed that the minimum P_2/P_e values obtained for transitions from the adjacent $(4s4d)^1D_2$ and $(4s6s)^1S_0$ states were about 0.0007 ± 0.0007, respectively. This is two orders of magnitude smaller than the reported observed P_2/P_e value for the $(4s5s)^3S_1$. Studies of several transitions in other atoms [15] also showed high precision and accuracy of the measurements and correct techniques.

The measurements were repeated several times over a six-year period, during which the entire apparatus was taken apart and reassembled in a different location. The scattering apparatus was enclosed in mu-metal, with residual magnetic fields of the order of 10^{-7} T, causing no observable effects. The experiments were repeated with normal-crystal 29% polarization, strained-crystal 66% polarization, and unpolarized electrons.

All elements of the optical detection system were replaced, and every optical element was tested for zero birefringence, for all conditions of tension in the components.

Further confirming the validity of the measurements are the values of P_1 and P_3, which show the expected and predicted results. Also, before the onset of cascade effects, P_1 vanishes within the experimental uncertainties, as expected from the spherical electron charge cloud of a well-LS-coupled $^3S_1$ state. If nonzero values of P_2 were caused by instrumental effects, it is very likely that P_1 or P_3 would have been affected as well.

In summary, our measurements reveal nonzero values of P_2/P_e. The size of the deviations from zero suggests some effect that is not accounted for in the most sophisticated, currently available theories for electron-atom collisions. It is unlikely that relativistic effects are the origin, and even the interaction with an open $3d$ subshell does not provide an obvious explanation. Hence, further studies of this and similar systems seem highly desirable in order to shed more light on the origin of these very unexpected results.

This research was supported by the Australian Research Council and the University of Western Australia. K.B. and O.Z. acknowledge support from the US National Science Foundation under Grants No. PHY-0757755 and No. PHY-0903818. A.D.S. received financial support from NSERC Canada. R.S. acknowledges grants from the Council of Scientific and Industrial Research (CSIR), New Delhi, and IAEA Vienna.

[1] J. Kessler, *Polarized Electrons*, 2nd ed. (Springer, Heidelberg, 1985).
[2] N. Andersen and K. Bartschat, *Polarization, Alignment, and Orientation in Atomic Collisions* (Springer, New York, 2001).
[3] K. Bartschat, G. F. Hanne, and A. Wolcke, Z. Phys. A **304**, 89 (1982).
[4] J. F. Williams *et al.*, J. Phys. Conf. Ser. **235**, 012005 (2010), and references therein.
[5] M. Eminyan and G. Lampel, Phys. Rev. Lett. **45**, 1171 (1980).
[6] K. Bartschat and K. Blum, Z. Phys. A **304**, 85 (1982).
[7] L. Pravica, J. F. Williams, D. Cvejanovic, and S. A. Napier, Phys. Rev. A **75**, 012721 (2007).
[8] J. P. Sullivan *et al.*, New J. Phys. **5**, 159 (2003).
[9] K. A. Berrington, W. B. Eissner, and P. H. Norrington, Comp. Phys. Commun. **92**, 290 (1995).
[10] O. Zatsarinny, Comp. Phys. Commun. **174**, 273 (2006).
[11] O. Zatsarinny and K. Bartschat, Phys. Rev. A **71**, 022716 (2005).
[12] S. A. Napier *et al.*, Phys. Rev. A **79**, 042702 (2009).
[13] S. Kaur *et al.*, J. Phys. B **31**, 157 (1998).
[14] P. Jönsson *et al.*, Comput. Phys. Commun. **177**, 597 (2007).
[15] L. Pravica, D. Cvejanovic, J. F. Williams, and S. A. Napier, Phys. Rev. A **75**, 030701(R) (2007).