Measurement of the Integrated Stokes Parameters of Zn 468 nm Fluorescence Excited by Polarized-Electron Impact

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Abstract. The integrated Stokes parameters P_1, P_2, and P_3 of Zn ($4^3P_{0,1} - 5^3S_1$) fluorescence resulting from transversely-spin-polarized electron impact excitation of the Zn $(4s5s)5^3S_1$ state have been measured. This work was motivated by similar studies reported several years ago, in which non-zero values of the integrated Stokes parameter P_2 between the threshold for the $(4s5s)5^3S_1$ excitation and the first cascading $(4s5p)5^3P_J$ threshold were measured. We observe optical excitation functions in agreement with previous experimental and theoretical results, but find integrated P_2 Stokes parameter values which are consistent with zero and inconsistent with those measured previously.

1. Introduction

In 2011, a group at the University of Western Australia (UWA) published a remarkable paper [1]. It described a measurement of non-zero values of the P_2 "integrated Stokes parameter" for light emitted after near-threshold excitation by transversely-polarized electrons in the reaction: $e^- + Zn(3d^{10}4s^2)4^1S_0 \rightarrow (3d^{10}4s5s)5^3S_1 \rightarrow (3d^{10}4s4p)4^3P_{0,1,2} + \gamma$. This result is fundamentally at odds with established theory [1-4]. We describe here experiments conducted to confirm or refute that result.

The term "integrated" when used to refer to Stokes parameters means that the scattered electrons are not detected. In our experiment, a polarized electron beam is crossed with an atomic Zn target beam. We define the electron direction of travel to be along the z-axis with a polarization vector $\vec{P}$ parallel to the y-axis. Emitted photons are then observed along the y-axis. In general, the light emitted by an atom after excitation by electron impact in such an experiment is elliptically polarized. The size, shape, orientation, and the direction of rotation of the ellipse can be described by the three relative Stokes parameters P_1, P_2, and P_3 [5].

2. Theory

Collisions of this kind were the subject of a paper by Bartschat and Blum in 1982 [2]. From symmetry arguments and the properties of Wigner 9-j coefficients, they concluded that a non-vanishing value of P_2 can only be observed if spin-orbit effects during the collision are significant. This can be the case if either (i) the excited target is not well L-S coupled or (ii) the continuum electron experiences sufficiently strong spin-orbit forces that its spin undergoes precession during the collision (i.e. Mott scattering). A non-zero measurement of P_2 thus reveals very specific dynamical information about the spin-dependent coupling operative during the atomic collision.

The Zn $(4s5s)5^3S_1$ state is known to be well-LS coupled [1,6] and Zn has a low enough Z that spin-orbit forces are too small to cause significant continuum-electron spin precession. All state-of-the-art theoretical calculations that have been brought to bear on this problem [1,3,4] have predicted $P_2/P_e < 10^{-4}$, whereas Pravica *et al.* [1] measured $P_2/P_e \approx 10^{-1}$. (Here P_e is the incident electron spin

polarization.) With regard to intermediate coupling, there could be admixtures from other configurations that could also result in J=1 excited states, e.g. $(4s6s)^3S_1$, $(4p^2)^3P_1$, $(4p5p)^1P$ and $(4s4d)^3D_1$. However, the largest of these has a mixing coefficient with a magnitude of 0.00002 [1], so vanishing linear light polarizations P_1 and P_2 would still be expected [2].

It should be pointed out that there is another mechanism by which a non-zero P_2 is allowed. If a well-coupled L-S state becomes populated by decay of an intermediate non-well L-S coupled state such as a negative-ion resonance, a non-zero P_2 can be measured. If the lifetime of this intermediate state is longer than the fine-structure relaxation time, then L and S may not be good quantum numbers in the protracted collision. Another mechanism which can cause $|P_2| > 0$ is the excitation of a non-well-LS-coupled excited state above the level of interest that can subsequently decay into that level. Thus, the energy and energy-width of the incident electron beam must be well understood in measurements of this kind.

3. Apparatus

Our experimental apparatus consists of four major components: the polarized electron source chamber, the electron transport chamber, the zinc target chamber, and the optical polarimeter. The GaAs polarized electron source used in this experiment is a heavily modified version of that described in Ref. [7]. It produced beams of electrons with a polarization of 0.25(1) and an energy FWHM, ΔW, of ca. 300 meV. After initial extraction, electrons pass through an electrostatic 90° bender which converts the initially longitudinally-polarized beam into a transversely-polarized one. Electrostatic lenses then guide the electron beam from the source chamber to an intermediate differentially-pumped transport chamber, and ultimately to a target chamber which houses the Zn oven. A spin rotator in the transport section is used to rotate the electron spin in a plane perpendicular to the beam such that the light observed in the subsequent collisions is along the direction of the electron spin. The atomic Zn target was produced by a Zn oven and a heated effusive channel that directed the atomic beam at right angles to both the fluorescence observation direction and the electron beam axis.

The light polarization analysis system used in this experiment is comprised of a very thin BK7 glass window, a collection lens, a birefringent polymer retarder (ideally a quarter-wave retarder), a dichroic linear polarizer, an interference filter to select the fluorescent transition under study, and lenses to refocus the collimated light onto the photocathode of the photon-counting PMT (Hamamatsu R943-02). The lens and window elements of the optical train were tested for birefringence and exhibited none. The transition of interest was selected by one of two narrow-band interference filters with center wavelengths (and bandwidth values) of 468.1 nm (0.3 nm) and 472.3 nm (0.3 nm) for the Zn $(4s4p)4^3P_{0,1} - (4s5s)5^3S_1$ transitions, respectively. The polarization of the transition to the $(4s4p)4^3P_2$ state was not studied because it was expected to be a factor of six smaller in magnitude than that of the transition to the J = 0 state [1].

Several possible sources of systematic error were investigated, including the effect of radiation trapping, beam-tuning-related issues, non-linearity of the photomultiplier tube, effects of exotic excimers, and the Hanle effect. These effects were found not to influence the measurement on the order of the statistical uncertainties. We carefully eliminated extraneous magnetic fields in the target volume which could cause significant Hanle rotation; the residual magnetic field in the target volume was less than 10^{-6} T. The Zn density of this experiment was 5×10^{-5} Torr, as determined by comparing the observed intensity of Zn fluorescence to the theoretical Zn cross-sections of Ref. [8]. Radiation trapping was thus not a likely source of systematic error.

4. Results

Excitation function measurements of the transition to the 4^3P_0 state (468.1 nm) are shown in Fig. 1, as well as theoretical predictions of Napier *et al.* [8] and the previous experimental results of Kontrosh *et al.* [9] for the transition to the 4^3P_2 state (481.1 nm). These results have been normalized to the peak value of our data. The FWHM energy width, ΔW, of Kontrosh *et al.*'s electron beam was 0.08-0.20 eV. The theoretical work of Napier *et al.* was convoluted with an assumed ΔW = 0.25 eV. Our

excitation function measurements are in qualitative agreement with the theoretical predictions of Napier *et al*., and have the same threshold energy value as that of Kontrosh *et al.* due to a lack of noticeable contact potential effects. However, we observe the peak of the excitation function at a somewhat higher energy. We attribute our failure to observe a secondary maximum at ~0.5 eV above threshold to our poorer experimental energy resolution.

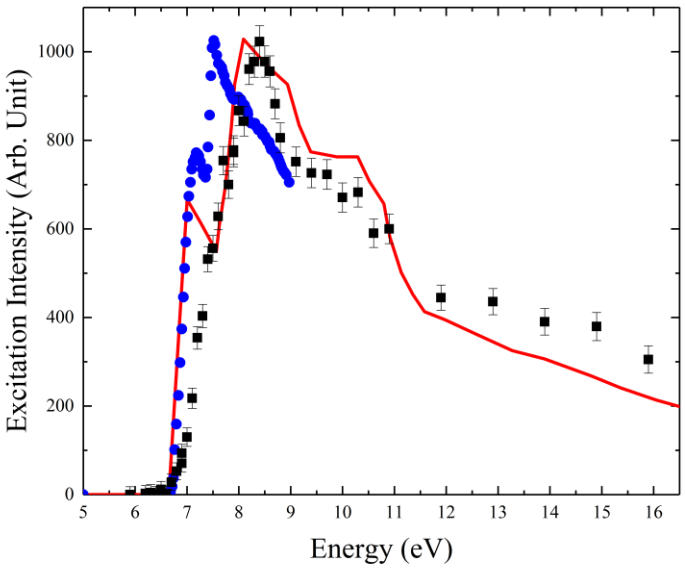

Figure 1. Excitation function measurements at 468.1 nm for the transition to the Zn 4^3P_0 state (squares) and theoretical (line) and experimental (circles) excitation function curves at 481.1 nm for the transition to the Zn 4^3P_2 state of Refs. [8] and [9], respectively. The energy dependence of the optical excitation functions for the 468.1 nm and 481.1 nm transitions should be largely the same. The excitation function is used to determine the energy scale of subsequent Stokes parameter measurements.

The excitation function curves served to set the energy scale of subsequent Stokes measurements by determining the energy at which photons were first detected with statistical significance above background and setting that energy equal to the threshold energy of the excitation process. The error of the experimental data in Fig. 1 is given by the statistical counting error of each measurement. These data have been normalized to the incident electron beam current and corrected for the polarization dependence of the emitted radiation [10].

The Stokes parameter measurements (Fig. 2) have been corrected for the effects of both an imperfect-polarizer and a non-quarter-wave retarder, as well as for hyperfine depolarization. They have not been corrected for either the effects of the finite photon collection angle or finite electron beam divergence because such effects are, again, quite small compared with the reported uncertainties. The value of the incident electron polarization, $P_e = 0.25(1)$, was determined by neon optical polarimetry. This value, but not its uncertainty, is folded into the P_2/P_e and P_3/P_e measurement uncertainties presented here.

For the transition to the 4^3P_0 state (468.1 nm), Pravica *et al*. and this work report the same P_1 and P_3/P_e values between the excitation threshold of the $(4s5s)5^3S_1$ state (6.7 eV) and the excitation threshold energy of the first cascading state (7.6 eV). The theoretically-predicted values for P_1 and P_3/P_e are in good agreement with these experimental results. However, our measured P_2/P_e values differ from the UWA result. Whereas Pravica *et al.* report a non-zero P_2/P_e for this energy range, we report P_2/P_e values which are consistent with zero and qualitatively inconsistent with their values. Our result is consistent with the theoretical predications of Refs. [1-4].

Integrated Stokes measurements of the transition to the Zn 4^3P_1 state (472.2 nm) at 7.2 eV follow a similar trend. The measured P_1 and P_3/P_e values of both Pravica *et al.* and this work agree with the theoretically predicted values. However, our measured P_2/P_e value is consistent with zero and is inconsistent with that of Ref. 1, which is 4.9(7)%. Our integrated Stokes measurements with an unpolarized electron beam are also consistent with zero.

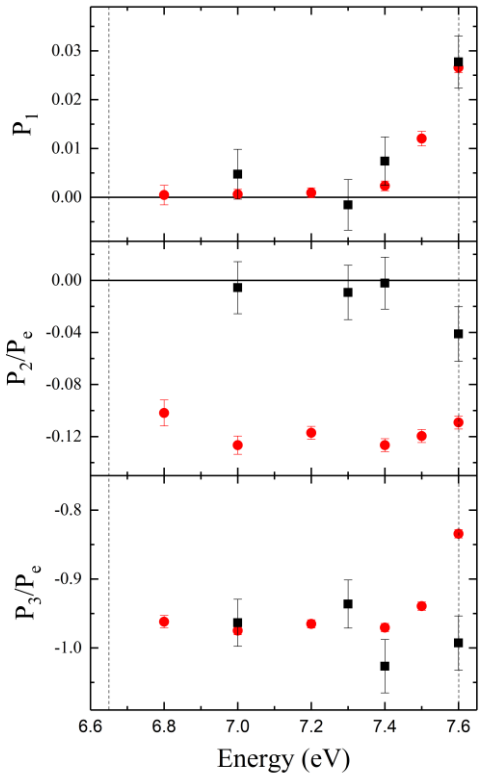

Figure 2. Integrated Stokes parameters for the Zn $(4s4p)4^3P_0$ – $(4s5s)5^3S_1$ (468.1 nm) transition. Vertical lines at 6.65 eV and 7.60 eV denote the excitation thresholds of the 5^3S_1 state and the first cascading 5^3P_J states, respectively. Circles are data of Ref. [1]; squares are data of this work. The FWHM of the energy widths of the incident electron beams, ΔW, are ~ 300 meV for the Nebraska data and 250 meV for the UWA data. Below the first cascade threshold, symmetry arguments and the properties of Wigner 9-j coefficients require that P_1 and P_2/P_e equal zero [2], and that $P_3/P_e = 1$ (not accounting for hyperfine depolarization) [5,6].

5. Conclusion

Pravica *et al*.'s work and this work measure different P_2/P_e values below the energy of the first cascade threshold. Although we cannot explain this difference, the agreement between our results, the angular-momentum coupling constraints pointed out by Bartschat and Blum [2], and the highly successful [1,3,4,8] theoretical calculations that have been applied to this problem are perhaps the best evidence of our results accuracy. The agreement with theory and the care we took in considering the various possible sources of systematic error outlined here give us confidence that the seeming disconnect between theory and experiment suggested by the results of Pravica *et al*. has been eliminated.

Acknowledgments

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