Alignment of the (3d^{10}4s5s)^{3}S_{1} State of Zn Excited by Polarized Electron Impact

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Abstract

We have measured the integrated Stokes parameters of light from Zn $(4s4p)4^3P_{0,1}$ - $(4s5s)5^3S_1$ transitions excited by transversely-polarized electron impact at energies between 7.0 and 8.5 eV. Our results for the electron-polarization-normalized linear polarization Stokes parameter P_2, between incident electron energies 7.0 and 7.3 eV, are consistent with zero, as required by basic angular-momentum coupling considerations and by recent theoretical calculations. They are in qualitative disagreement with previous experimental results for the P_2 parameter.

Since the Franck-Hertz experiment in 1914 [1], experiments studying electron-atom collisions have served as test-beds for quantum mechanics [2], and have provided basic data for topics ranging from technologically-oriented plasma physics [3] to planetary atmospheres [4]. Electron-atom scattering physics is, at its core, an exemplification of the ubiquitous many-body long-range force problem in its most basic form, and attempts to bring a diverse array of experimental results in line with state-of-the-art theoretical calculations are the most important endeavor in the field. As experimental sophistication has increased, our knowledge base of such collisions has become more and more detailed. Researchers have done numerous "complete" experiments [5], in which all the quantum numbers of a collisional system are measured, they are using "reaction microscopes" to determine many or all of the kinematic variables in multi-component collisions [6], and have used polarized collision partners to provide unprecedented detail about spin-dependent magnetic and Pauli-exclusion forces in these collisions [7]. With the caveat that there remain significant uncertainties in our understanding of the collisional dynamics of complex systems such as, e.g., low-temperature hydrogen plasmas [8], it is safe to say that the problem of single collisions of electrons with one- and two-valance-electron atoms is largely solved. Our understanding of scattering from complex targets in the "great outback" of the periodic table of the elements, however, is still in its infancy.

This state of affairs was called into question recently by the experiments of the Perth group [9] in which they bombarded a light, quasi-two-electron atomic target, Zn, with transversely spin-polarized electrons. The Zn was excited from its

$(3d^{10}4s^2)4^1S_0$ ground state to the $(3d^{10}4s5s)5^3S_1$ state by electron impact and exchange, and the relative "integrated" Stokes polarization parameters [10] of the subsequent fluorescence from the decay of the 5^3S_1 state to the fine-structure-resolved $(3d^{10}4s4p)4^3P_{0,1,2}$ multiplet were measured. ("Integrated" in this context refers to the fact that the scattered electrons were not detected in coincidence with the fluorescence photons.) Integrated experiments of this type, while having the disadvantage that they average over scattered electron trajectories and thus lose information about the Coulombic dynamics of the excitation, have distinct benefits as well: they have much higher counting rates than electron-photon coincidence experiments and can thus yield more precise data, they are not subject to many of the systematic errors endemic to low-count rate, variable-detection-angle scattering experiments, and, perhaps most importantly here, they provide a clean signature of spin-dependent interactions in the collision, unmasked by the much larger Coulombic effects. These advantages were first pointed out in a seminal paper by Bartschat and Blum (BB) [11] and a subsequent series of papers by our group [12 - 15].

The key insight of the BB paper is this: in integrated Stokes parameter measurements of the type described above, and in the absence of either target or continuum electron spin-orbit coupling during the collision, the integrated Stokes parameter P_2 must be identically zero. The P_2 parameter corresponds to the difference in the intensity of linearly-polarized light at 45^o and 135^o to the incident electron beam (see Fig. 1). It results from a tilting of the excited state quadrupole moment in the x-z plane away from the beam axis. Any deviation from zero of P_2 must be due to spin-orbit coupling during the collision that manifests itself because

(a) the excited state is not well LS-coupled,

(b) the LS-coupled excited stated is produced by the decay of either a higher-lying neutral atomic state or a negative-ion resonance that is not well-LS coupled, or

(c) strong spin-orbit forces act on the continuum electron during the collision, causing its spin to rotate in the motional B-field it experiences. This latter effect is essentially Mott scattering.

If case (c) is not relevant, e.g., in the case of scattering from low-Z atoms like Zn, the fluorescing state in question must be excited in an exchange reaction with the polarized electron beam for P_2 to be non-zero [16].

In the Perth experiment, P_2 was measured for all three well-resolved multiplet lines in the $4^3P_{0,1,2}$ - 5^3S_1 transitions. Between the threshold for excitation of the 5^3S_1 state (6.7 eV), and the threshold for excitation of the lowest state that can cascade into it, the $(3d^{10}4s5p)5^3P$ states at 7.6 eV, they measured P_2 values of about -0.12(1), 0.06(1) and -0.02(1) for decay to the J = 0, 1, and 2 multiplet components of the lower 4^3P state, respectively. (All uncertainties quoted in this Letter correspond to the standard error with a 68% confidence limit.) Note that when combined with their statistical weights of 1,3, and 5, these values have a sum consistent with zero, within their experimental uncertainty, as expected; radiation from an unresolved ^3S multiplet cannot be polarized. Over the same energy range, P_1 was measured to be zero for all multiplet transitions. Above the first cascading threshold, the P_2 values are reduced slightly in magnitude, while the P_1 data increase to significant non-zero values.

The non-zero results for P_2 below the first cascading threshold are forbidden by the BB dynamical symmetry argument, because the 5^3S_1 Zn state is extremely-well LS coupled, and Mott scattering is certainly negligible for this system [9,17,18]. The Perth result is particularly surprising in light of the fact that a host of experimental evidence from the last three decades has shown that transitions involving excited states that are well-LS coupled and are unaffected by the decay of negative ion resonances or cascading exhibit no significantly non-zero P_2 values, while those that are intermediately coupled always do [7,12 – 14, 19 - 21], in agreement with BB. In the present case, three state-of-the-art theoretical calculations predict P_2 values of order 10^{-4}, three orders of magnitude below those observed [9,17,18]. Since the constraints imposed by BB are based on analytical Clebsch-Gordan algebra (as opposed to a dynamical calculation), it is difficult to see where much more can be done on the theoretical side. There is some ($3d^94s5snd$) configuration mixing in the 5^3S_1 state [22], but it remains better than 99.9% LS-coupled.

The Perth experiment has been carefully checked and redone with different components and reassembled apparatuses thrice [9,23]. One possible explanation for the discrepancy is that there might be a strong negative ion resonance in the energy region between 6.7 and 7.6 eV that decays into the 5^3S_1 state and that is not well-LS coupled. The Perth P_2 data show no obvious resonant behavior, but they have not published optical excitation functions corresponding to their Stokes parameter measurements that might better shed light on this possibility. One previously published excitation function [24], for the $4^3P_2 - 5^3S_1$ transition at 481.1 nm, which is in good agreement with our data, does exhibit a prominent unclassified resonance-related feature above 7.18 eV, but this structure is also well-reproduced by theory [25], and doesn't yield a corresponding prediction of P_2 significantly greater than 10^{-4}.

The Perth group has sketched several explanations for their anomalous results that invoke new phenomena related to Berry's phase [21]. These suggestions have been rejected in the literature [17,18]. Thus the major disagreement between theory and this experimental result has not been resolved. This Letter reports results from our experiment that sought to reproduce the Perth result.

We used a standard GaAs polarized electron source [26] (Fig. 2) to produce beams of electrons with a polarization, P_e, of 0.25(1) and an energy width of 0.4 eV. After initial extraction, electrons passed through a 90° electrostatic bend which converted the initially longitudinally-polarized beam into a transversely-polarized one. A series of electrostatic lenses then guided the electrons from the source chamber through a differentially-pumped transport section to a target chamber that housed the Zn target oven. A 100-turn solenoidal coil, whose longitudinal axis was along the direction the electron beam's momentum, was used to rotate the electron spin in a plane perpendicular to the electron momenta such that the light observed in the downstream collision region was along the direction of the electron spin. This process caused some weak beam defocussing, resulting in a loss of, at most, 40% of the beam.

The Zn target beam was produced by an oven and a separately heated effusive channel that directed it at right angles to both the fluorescence observation direction and the electron beam axis. The zinc oven, which was based on one designed for Cd [27], consisted of a titanium crucible which held a ~40 g zinc charge, and a 0.34 mm ID nozzle for beam formation. Zinc pellets were cut from 99.9% pure metal purchased from the Goodfellow Corporation. Both the crucible and nozzle were wound with independent bi-axial heating wire (ARI Industries, Inc.), which produced relatively low residual magnetic fields. A zinc catcher opposite the oven was cooled with 5^{o}C chilled water. Additionally, various critical components of the apparatus were covered with Kapton sheet to prevent the deposition of Zn on them.

The optical polarimeter used in this experiment comprised a very thin BK7 glass window, a collection lens, a rotating birefringent polymer 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, dark-count selected photomultiplier tube (Hamamatsu R943-02). The upstream window of the optical train had no measurable birefringence. The transition of interest was selected by one of two narrow-band interference filters with center wavelengths (and bandwidth values) of 468.0 nm (0.3 nm) and 472.2 nm (0.3 nm) for the Zn $4s4p^3P_{0,1}$ - $4s5s^3S_1$ transitions, respectively.

The relevant specifications of the polarizer and retarder (fast axis, transmission axis, extinction ratio, retardance) were measured on a separate optical bench and *in situ*. The Stokes parameters for light emitted from the Ne $(2p^5 3s)^3P_2$ - $(2p^5 3p)^3D_3$ optical decay were measured using a filter with a center wavelength of 640.2 nm and a 0.9 nm bandwidth. They were found to be qualitatively consistent (within the statistical precision of the measurements to two standard errors (95% confidence limit)), with our earlier results [13] and those of Hayes *et al*. [19].

Possible sources of systematic error were investigated thoroughly, including the effects of radiation trapping, collisional depolarization, beam tuning, non-linearity of the photomultiplier tube, exotic excimers [28], and Hanle rotation [14]. These were found to have no measurable influence on the data. We were particularly concerned about systematic error due to the Hanle effect, and eliminated extraneous magnetic fields (to a level below 10^{-6} T) which could cause Hanle rotation and subsequent mixing of the linear polarization fractions P_1 and P_2. The Zn density of this experiment was about 10^{12} cm^{-3}, as determined by comparing the observed intensity of Zn fluorescence to the theoretical Zn cross-sections of Ref. [29]. At these pressures, radiation trapping and collisional depolarization are negligible.

The results of our integrated Stokes parameter measurements for the $(4s4p)^3P_0$ - $(4s5s)^3S_1$ optical decay (468.1 nm) are shown in Fig. 3. These data are corrected for the effects of an imperfect-polarizer and a non-quarter-wave retarder, as well as hyperfine depolarization and the effect of collecting light into a finite solid angle. The error bars (68% confidence limit) on these data account for statistical counting uncertainty including the Fourier fit error of each measurement, as well as the uncertainty in the extinction ratio of the polarizer and the retardance of the waveplate. The energy scale indicated in Fig. 3 was set by measuring the optical excitation function for the 468.1 nm transition, and determining the voltage applied to the electron-Zn interaction volume that was necessary to see a signal distinct from the background with 95% statistical confidence.

Our results are in substantial agreement with those of the Perth Group, except for the values of P_2/P_e for energies below the first cascading threshold. Here, our results are in quantitative agreement with theory and are in qualitative disagreement with those of ref [9]. Small variations in the energy dependence we observe in contrast with that of the Perth group appear to be due largely to our broader electron beam energy width (0.40 eV *vs*. 0.25 eV). We note also that our single-energy measurement of the integrated Stokes parameters for the $(4s4p)^3P_1$ - $(4s5s)^3S_1$ optical decay (472.2 nm) are again in agreement with Perth except for the P_2/P_e datum below the first cascading threshold at 7.6 eV, where we measure P_2/P_e = -0.006(6), as compared with 0.049(7) that was reported in Ref. [9]. We also investigated values of P_2 and P_3 (which should both be proportional to P_e) for incident unpolarized electrons. All of these measurements gave results consistent with zero.

Two other experiments have made integrated Stokes measurements of the Zn transitions considered here. Eminyan and Lampel [30] measured the integrated Stokes parameter P_3/P_e for the $(4^3P_J - 5^3S_1)$ transitions using polarized electrons at energies between threshold (6.7 eV) and 10 eV, and Suzuki et al. [31] measured

P_1 for the same transitions using unpolarized electrons for a single energy. The reported P_3/P_e values of Eminyan and Lampel agree with the values of this work and those of the Perth group. The measurements of Suzuki *et al.*, if we assume an energy calibration offset of >0.6 eV and a systematic error resulting in a sign flip of their reported P_1 values, agree with both results as well.

Given the large amount of evidence that supports the BB symmetry argument, our results are not terribly surprising. There is a simpler way to understand why P_2 must be zero below the cascading threshold, however. A non-zero P_2 can occur only of there is a quadrupole distribution of excited-state atomic oscillators that is tilted in the x-z plane away from the z-axis (see Fig. 1). A 5^3S_1 state can have such a quadrupole in principle, because J > ½. Because the atom is in an S state, though, any alignment of the system must come from a spin quadrupole, which in turn must be the result of spin-exchange excitation. But exchange excitation of a triplet state from a singlet state by a transversely-polarized electron beam can only result in an orientation along $\hat{y}$, not an alignment in the x-z plane [10,16]. Thus, both P_1 (which corresponds to an alignment along $\hat{z}$) and P_2 must be identically zero for our experimental geometry. Cascade population by decay of the 5^3P state can result in a tilted 5^3S alignment if (a), the 5^3P state is not well-LS coupled, (b), it is excited by exchange, and (c), its orbital angular momentum has a quadrupolar distribution along $\hat{z}$ as a result of the collision. The nascent spin orientation of the ^3P system can then rotate the P-state alignment away from the z-axis in the x-z plane. Upon decay this tilted quadrupole is converted to a spin alignment of the ^3S state which yields a non-zero P_2. This mechanism is apparently operative, given the non-zero values of P_2 we and the Perth group observe above the cascade threshold energy. As such, it represents a rare and interesting example of spin-alignment leading to linear polarization in an atomic collision process [32].

The fact that P_2 is non-zero below the first cascade threshold for the analogous 6^3P_2 - 7^3S_1 transition in Hg should not be taken as support for the Perth results [17,18,23,33]; Hg is much heavier, the 7^3S_1 state is intermediately coupled, and some level of Mott scattering is very likely.

The reasons for the discrepancy between our results and those of the Perth group remain unclear. Residual, uncharacterized magnetic fields are always a potential concern in these measurements, but the Perth reports would appear to preclude this possibility. Poorly-characterized effects due to secondary electrons or unfocussed primaries can be a potential problem with triplet excitation [34], but both data sets have been taken at sufficiently low pressures that this seems unlikely as well. One way forward now would be for theorists to calculate the effect of cascading on the integrated Stokes parameters, although here, both experiments agree. A detailed understanding of how intermediate coupling coefficients can be used to predict P_2 values for specific atoms and transitions might also prove useful. The dynamic interaction that leads from intermediate coupling to a rotated excited state quadrupole is still not well understood. For the time being, however, we argue that, based on our experimental results, the Bartschat-Blum symmetry argument is valid, and that no new physics is needed to explain below-cascade threshold values of P_2 in the Zn system.

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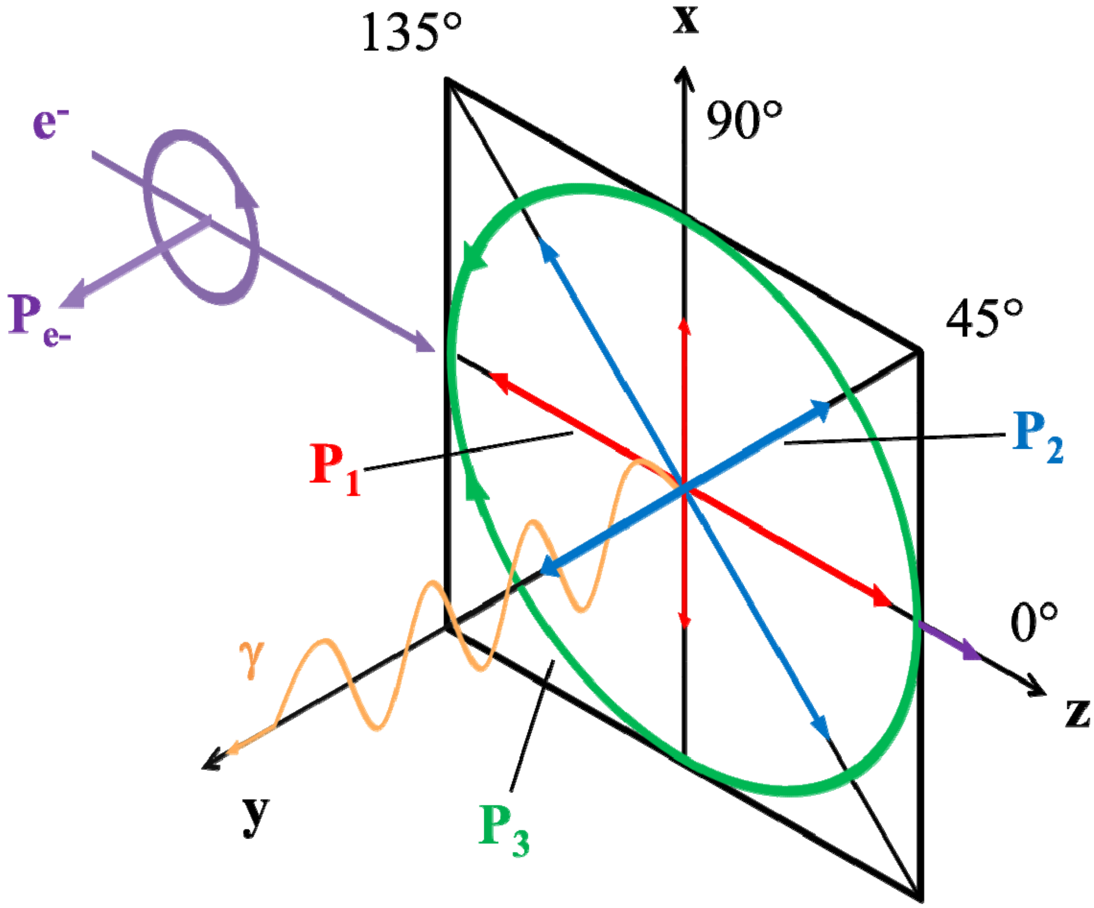

FIG. 1. Experimental collision geometry for the measurement of the integrated Stokes parameters when transversely-polarized electrons excite a target and the subsequent fluorescence is observed along the direction of the initial spin polarization. The three relative Stokes parameters P_1, P_2, and P_3 correspond to linear polarization relative to the beam axis, linear polarization rotated from the beam by 45°, and circular polarization [9].

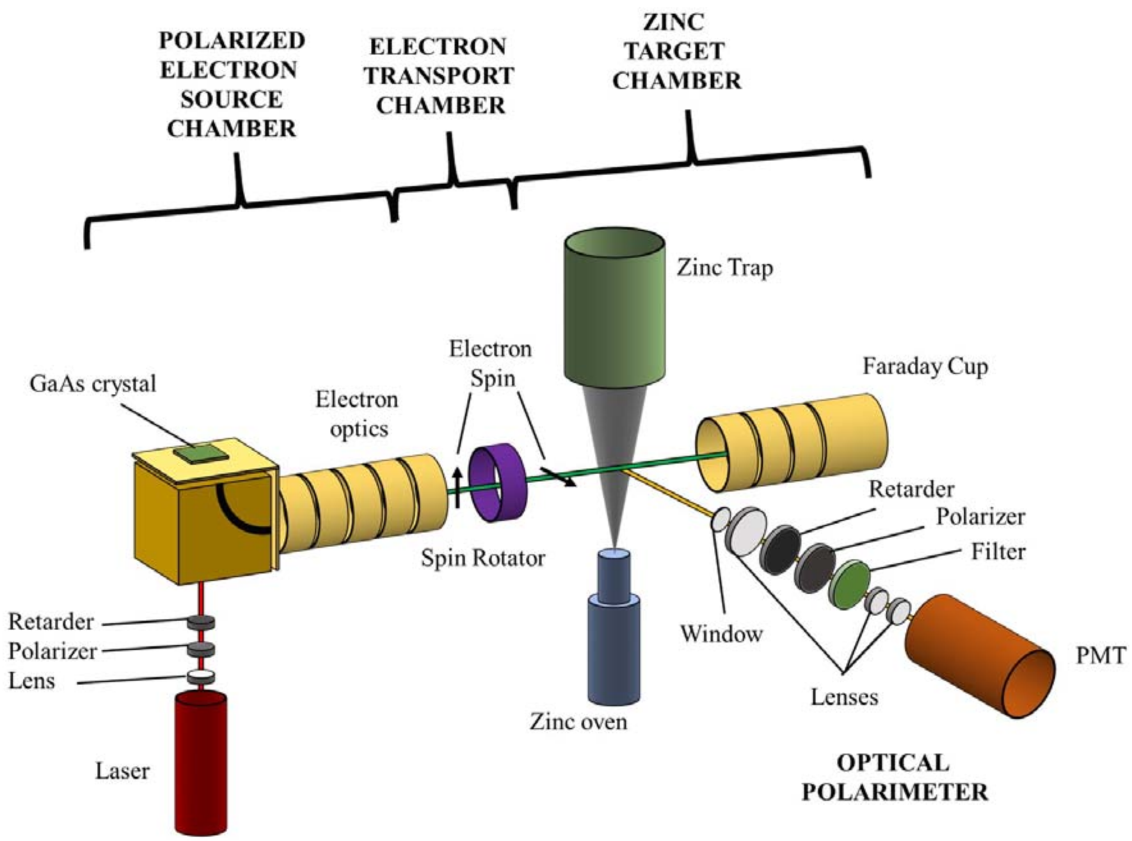

FIG. 2. Schematic diagram of the apparatus.

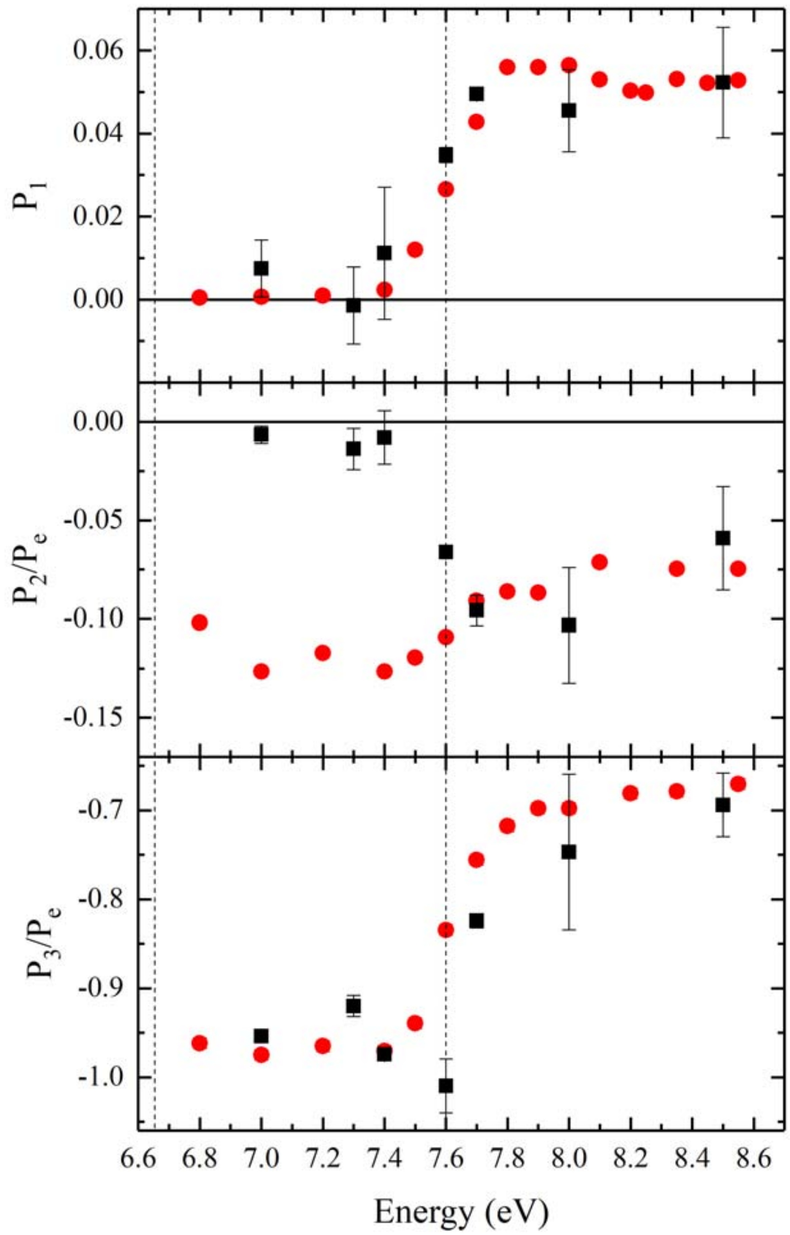

FIG. 3. Integrated Stokes parameters for the Zn (4s4p)4^3P_0 – (4s5s)5^3S_1 (468.1 nm) transition. Vertical lines at 6.65 eV and 7.60 eV denote the excitation thresholds of the 5^3S_1 state and the first cascading 5^3P_J state, respectively. Circles are data of Ref. 8; squares are data of this work.