Near-threshold measurement of integrated Stokes parameters for Kr excited by polarized electrons

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We have made high-accuracy measurements of the integrated Stokes parameters for resonance fluorescence from polarized electron-impact excitation of the 4p55p(5/2) 3D3 and 4p55p(5/2)2D5 states of Kr. We report measurements in the region within 0.6 eV of threshold, which is below the first cascade threshold. We also present theoretical calculations of these Stokes parameters using a recently developed relativistic Breit-Pauli R-matrix code. In well LS-coupled systems, nonzero values of the integrated Stokes parameter P2 signal relativistic effects (like continuum spin-orbit coupling, i.e., Mott scattering). A single value of P2 = 4(4) × 10−3 at 12.0 eV was previously reported in this energy range [Furst et al., Phys. Rev. A 47, 3775 (1993)]. We have now measured P2 at six different energies in this region to comparable precision. These results are consistent with P2 = 0 and with the theory. We discuss the effect of the electron-beam energy width on the accuracy of the measurements. Even when such effects are accounted for, serious discrepancies remain between theoretical and experimental results for excitation of the intermediately coupled 3D2 state. [S1050-2947(99)07907-X]

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I. INTRODUCTION

With the advent of convergent-close-coupling (CCC) calculations [1,2], electron scattering amplitudes can be calculated very accurately over a broad energy range for H and He targets, as well as the light alkali-metal atoms. However, calculations involving heavy targets are hampered by the necessary inclusion of a larger number of target electrons as well as relativistic effects such as internal spin-orbit coupling and spin-orbit coupling to the continuum electron. Some progress has been made by using the Breit-Pauli R-matrix technique [3–5], particularly for impact excitation of heavy noble gases (HNGs). In this method, relativistic effects are accounted for perturbatively through the one-electron terms of the Breit-Pauli Hamiltonian [6].

We have been particularly interested in the relative Stokes parameter P2 in angle-integrated transversely polarized electron-impact experiments. Under these conditions, observing a nonzero value of P2 provides a clean test of relativistic effects, involving either atomic fine-structure or spin-orbit coupling to the continuum electron. If the spin and orbital angular momentum of the excited state are decoupled, only spin-orbit coupling of the free electron to the atom will produce nonzero values of P2 [7–9]. Several years ago, we made a number of attempts to measure nonzero P2 values in the HNGs Ne, Ar, Kr, and especially Xe, where the angle-integrated Mott scattering asymmetry should be largest. The statistical precision of those measurements was hampered by low-density targets which produce low count rates. Moreover, the theoretical calculations at that time predicted immeasurably small values of P2 [8–10]. Since our initial work, other groups have also made measurements of integrated Stokes parameters in the HNGs, using incident polarized electrons [5,11–16].

More recently, several theoretical groups have investigated polarized electron-impact excitation of HNGs [5,17,18]. Of these theories, the R-matrix calculations clearly have the best chance of correctly predicting results within 1 eV of the excitation threshold. Interestingly, R-matrix calculations of Zeman et al. [5] predict values of P2 as large as 6% near the excitation threshold of the well LS-coupled states 4p55p(5/2)3D3 in Kr and 5p56p(5/2)3D3 in Xe. This provided us with the motivation to make additional high-precision measurements of P2 in Kr, where the gap between the excitation threshold of the 3D3 state and the excitation of the next higher state which can decay into the 3D3 state is 0.7 eV. While the predicted values of P2 are higher for Xe, the corresponding gap is only 0.3 eV, indicating that the count rates would be very low in the region where our measurements are free of contamination from cascades [19].

The Stokes parameters P1, P2, and P3 are closely related to the detailed structure of the excited-state charge cloud. Generally, atomic excitation processes create anisotropic charge clouds whose detailed shape and angular momentum coupling are completely characterized by state multipoles. The angular distribution and polarization of the dipole emis-
sion from these excited states carries information about the monopole, dipole, and quadrupole moments of the charge cloud [20]. However, the number of nonzero multipole moments depends on the collision symmetries in the experimental apparatus [21]. In the present experiment we excited the Kr target with a transversely polarized electron beam, but did not detect the scattered electrons in coincidence with the emitted photons (we made an "angle-integrated" measurement). Because of this planar symmetry, the number of possible independent multipole moments is reduced from eight to four integrated multipole moments: \(<T^0_{00}\), \(<T^4_{20}\), \(<T^1_{11}\), and \(<T^2_{21}\).

When viewing the excited-state fluorescence along the direction of the transverse electron polarization, the relative integrated Stokes parameters of the dipole radiation can be expressed as follows:

\[
P_1 = \frac{I(0) - I(90)}{I(0) + I(90)} = \frac{2(-1)^{J_f+J} \sqrt{2} G^2_{J_f}}{3\sqrt{2J+1}} \langle t^2_{20}\rangle,
\]

(1)

\[
P_2 = \frac{I(45) - I(135)}{I(45) + I(135)} = \frac{2(-1)^{J_f+J} \sqrt{2} \text{Re}\langle t^2_{21}\rangle}{3\sqrt{2J+1}} \langle t^2_{20}\rangle,
\]

(2)

and

\[
P_3 = \frac{I(\sigma^-) - I(\sigma^+)}{I(\sigma^-) + I(\sigma^+)} = \frac{2(-1)^{J_f+J} \sqrt{2} \text{Im}\langle t^1_{11}\rangle}{3\sqrt{2J+1}} \langle t^2_{20}\rangle,
\]

(3)

where \(J\) is the excited-state angular momentum, \(J_f\) is the optical transition’s final-state angular momentum, and the \(G^J_{J_f}\) are factors that determine depolarization due to hyperfine nuclear interactions. The photon intensity \(I(\Theta)\) is for light transmitted through a perfect linear polarizer with its pass axis aligned at an angle \(\Theta\) with respect to the incident beam, while \(I(\sigma^-)\) and \(I(\sigma^+)\) are the intensities of light with positive and negative helicity along the optical axis of the detector [22]. The terms \(\langle t^J_{K\ell}\rangle = \langle T^J_{K\ell}\rangle/\langle T^0_{00}\rangle\) are the "relative integrated-state multipoles." Because of the spin dependence of these multipoles both \(P_2\) and \(P_3\) are proportional to the transverse spin, while \(P_1\) is independent of spin [21].

II. NUMERICAL METHOD

The calculations reported here were performed along the lines described earlier by Zeman and co-workers [5,23,24]. Briefly, the \(N\)-electron target states \(\Phi_i\) were represented as multi-configuration expansions

\[
\Phi_i(\mathbf{r}_1, \ldots, \mathbf{r}_N) = \sum_k c_{ik} \phi_k(\mathbf{r}_1 \ldots \mathbf{r}_N).
\]

(4)

The expansion coefficients and the approximate target energies \(E_i\) were obtained by diagonalizing the target Hamiltonian \(H^N\) according to

\[
\langle \Phi_i | H^N | \Phi_j \rangle = E_i \delta_{ij}.
\]

(5)

The configurations \(\phi_k\) were constructed from a bound orbital basis consisting of self-consistent-field (SCF) orbitals whose radial components, \(P_{nl}(r)\), were obtained using the CIV3 atomic structure package of Hibbert [25] with the nonrelativistic Hamiltonian \(H^N\) used in the optimization procedure. After the orbitals were obtained, the approximate Breit-Pauli Hamiltonian

\[
H^N_{BP} = H^N + H^N_{mass} + H^N_{D} + H^N_{SO},
\]

(6)

consisting of the nonrelativistic Hamiltonian and the one-electron relativistic mass correction, Darwin, and spin-orbit terms, was used in the description of the target states.

The input for starting the optimization procedure consisted of the Hartree-Fock orbitals for the respective ground states, as given in the tables of Clementi and Roetti [26]. Further valence orbitals were then constructed for the 4\(p^55s\) and 4\(p^25p\) states, with simultaneous reoptimization of the 4\(p\) orbital in the ground-state configuration. The re-optimization ensured that this orbital was also a reasonable approximation for use in the excited states of interest. In addition, a 4\(d\) and a 6\(s\) valence orbital were constructed to account for the most important channel coupling effects for all the states of interest.

The results presented below were obtained in a 31-state close-coupling approximation, including all states with the configurations 4\(p^5\), 4\(p^55s\), 4\(p^55p\), 4\(p^54d\), and 4\(p^56s\). The calculated energy level splittings of the states compared very well with experimentally determined values [27], but in order to correct as much as possible for the missing details in the structure calculation, we adjusted the diagonal terms of the Hamiltonian matrix by the very small amounts necessary to obtain the experimental thresholds for the channels coupled to the target states of interest.

The collision calculation was performed using the \(R\)-matrix (close-coupling) method that is based upon the partitioning of configuration space into two regions whose boundaries intersect at a specified radial distance \(r=a\). In the internal region \((r \leq a)\) exchange electron exchange and correlation between the scattered electron and the \(N\)-electron target are considered important, and the \((N+1)\)-electron system is treated as a closed system similar to an atomic structure problem. In the external region \((r > a)\) exchange between the scattered electron and the target can be neglected, and hence the calculation is simplified dramatically.
The Breit-Pauli \( R \)-matrix code of Berrington, Eissner, and Norrington [28] was used to perform the inner-region calculation. Accounting for partial waves up to a total (target plus projectile) electronic angular momentum of \( J_{\text{tot}} = 9/2 \), with 25 continuum orbitals for each orbital angular momentum of the projectile, ensured converged results for energies up to about 5 eV above the thresholds of interest. The calculation for the external region was performed using the flexible asymptotic \( R \)-matrix (FARM) package by Burke and Noble [29]. For each collision energy, this yields the reactivity (\( K \)), scattering (\( S \)), and transition (\( T \)) matrices from which the Stokes parameters of interest were calculated following the procedure outlined by Bartschat et al. [30].

III. EXPERIMENTAL METHOD

For the measurements reported here, we investigated excitation of the well \( Ls \)-coupled \( 4p^5S5p[5/2] \) \( ^3D_3 \) and the intermediately coupled \( 4p^5S5p[5/2] \) \( ^3D_2 \) states of Kr with polarized electrons having energies up to 1.2 eV above the respective excitation thresholds of these states. The threshold of the \( ^1S_0 \) state which can cascade into the \( ^3D_3 \) level is 12.11 eV. Likewise, the threshold of the \( ^1D_2 \) level which can cascade into the \( ^3D_2 \) level is 12.03 eV. Thus, there are 0.7 eV and 0.6 eV “windows” of cascade-free fluorescence above each respective excitation threshold.

As a source of polarized electrons (Fig. 1), we used the (100) face of bulk \( p \)-type Zn-doped GaAs [31,32]. We produced a longitudinally polarized electron beam by photoemission from the GaAs using a circularly polarized GaAlAs diode laser at 780 nm (Lasiris model DLS-500-780-50). To obtain efficient photoemission, we activated the crystal to negative electron affinity using the yo-yo technique described by Tang et al. [33]. The longitudinal polarization of the beam was transformed into transverse polarization by bending the beam 90° with a unique electrostatic deflector, which rotated its momentum without affecting its spin. We have observed emission currents of 40 \( \mu \)A with 40 mW of laser power and electron polarizations of 28(3). A more detailed description of this polarized electron source can be found in Ref. [32].

The electron beam was transported down the beamline using electrostatic lenses and magnetic steering coils (see Fig. 1). The beam also passed through a solenoidal spin rotator which could correct for any spurious rotation of the electron spin. After being transported 50 cm the electron beam entered a stainless steel target cell through a 1 mm diameter beam-defining aperture, excited the target gas, emitted through a 2.0 mm diameter aperture, and was detected on a series of downstream lens elements.

Our target cell was constructed of a stainless steel can. The top of the cell was isolated from the chamber by a gas-tight connection to a ring of Macor and was capped by a Viton seal to the photon collection lens. Target gas was delivered to the cell through a stainless steel tube which passed through the outer wall of the cell. The pressure was controlled by a Granville Phillips 203 leak valve. A pressure buffer composed of a wad of crumpled and sooted copper mesh was placed over the gas entrance at the bottom of the cell.

The resonance fluorescence produced in the vertical direction (along the electron spin axis) was focused into a parallel beam by the photon collection lens (51 mm diameter, 101 mm focal length) at the top of the gas cell. These photons then passed through an optical polarimeter composed of a linear polarizer / quarter-wave plate combination similar to that described by Berry et al. [34]. The photons were then counted by a GaAs photomultiplier tube (Hamamatsu model R943-02).

Since we knew that the linear polarization fraction \( P_2 \) is quite small [Furst et al. [8,9]] measured it to be \( 4(4) \times 10^{-3} \) at 0.7 eV above threshold, we had to design our apparatus and experimental procedure to reduce statistical uncertainty and eliminate spurious systematic effects. The target cell was carefully designed to measure the relative Stokes parameters near threshold precisely. We chose to use a static gas cell in the present experiments rather than the effusive beam target of Furst et al. to increase the target density-length product from \( 5 \times 10^{10} \) cm\(^{-2} \) to \( 5 \times 10^{12} \) cm\(^{-2} \). Because of the Kr pressures in the target cell, it was necessary to add a differential pumping stage to the beam line so that the pressure in the source region would not diminish the GaAs quantum efficiency. We were able to increase the pressure ratio between the target cell and the source chamber to \( 10^4 \) by using a 55 l/s turbo pump and two 5 mm apertures at the entrance and exit of the differential pumping chamber. The higher target density resulted in a significantly higher count rate and an accompanying improvement in the statistical precision of our data.

Precautions were taken to accurately determine signal rate very near threshold where the background rate is a large portion of the total count rate. First, we cooled the photomul-
tiplier tube to $-26^\circ$C, which reduced the dark count rate to $\approx 10$ Hz. Next, we made several efforts to reduce the background due to stray photons. The entire target cell, including the copper mesh pressure buffer, was heavily sooted using an acetylene torch. The mesh absorbed photons from the interaction region that were emitted directly away from the photon collection lens. Furthermore, a series of light apertures were used to define the visible interaction volume and absorb reflected photons. Along with the electron beam entrance and exit apertures, these light apertures limited the visible interaction volume to a cylinder 8 mm long and 1.5 mm in diameter. Additionally, we improved our estimate of the background count rate by measuring the background at several energies below threshold for each excitation function. Finally, we minimized the problems associated with drifting experimental parameters by taking the data quickly using a computer to control the electron energy accurately and repeatedly. Each data set was composed of several excitation functions, one for each combination of linear polarizer position and electron polarization. The data were analyzed in several steps. First, the dark counts were removed. Then, the data were normalized to pressure and current to remove the effects of drifting experimental parameters. Finally, the residual background was removed and the modified excitation functions were combined and analyzed to yield polarization measurements as a function of energy.

Carefully accounting for the background in this way allowed us to obtain data consistent with the kinematically required threshold value of $P_1 (0.41)$ for the $^3D_3$ state. In this regard, we note that the threshold is defined as being one-half step (0.05 eV) below the energy at which the signal rate becomes statistically distinct from the background. This convention has the advantage that the lowest-energy polarizations should be consistent with the kinematic threshold values, regardless of the energy distribution of the electron beam. Furthermore, graphs of the measured polarizations begin at the actual energy threshold and can be compared directly to any theoretical calculation, after appropriate electron beam energy profile convolutions have been made (see below).

Additionally, we carefully examined our apparatus for possible systematic errors and eliminated them. We corrected for the small relative reduction ($\leq 1.3\%$) in the Stokes parameters due to the finite collision volume, slightly divergent electron beam ($3.5^\circ$ half angle), and finite photon aperture ($11^\circ$ half angle) [35]. Additionally, we used magnetic coils outside the vacuum chamber to cancel stray magnetic fields in the target region, which could have altered the fluorescence radiation through the Hanle effect [20]. Figure 2 demonstrates the effect of a magnetic field on the linear polarization fractions $P_1$ and $P_2$ when the field vectors, electron spin, and photon emission are all along the same axis. For the data presented here, the $^3D_3$ state was excited with unpolarized electrons which would produce pure $P_1$ polarization in the absence of a magnetic field. The nonzero values of $P_2$ in Fig. 2 were produced when the plane of polarization was rotated due to the precession of the excited states in the magnetic field. It is clear that large magnetic fields destroy both $P_1$ and $P_2$, and that even small magnetic fields can produce spurious results. These results emphasize the necessity of carefully eliminating magnetic fields for accurate Stokes parameter measurements.

We averaged four measurements of $P_1$ and $P_2$ at the four equivalent pairs of linear polarizer angles to eliminate rotational optical asymmetries. We also made each measurement of the spin-dependent parameters $P_2$ and $P_3$ with the spin polarization of the incident electron beam both parallel and antiparallel to the optical axis of the polarimeter. This optical spin reversal technique is particularly effective in eliminating spin-independent instrumental asymmetries and effects due to possible spurious magnetic fields in the interaction volume.

We investigated the dependence of the measured polarization on target pressure (Fig. 3), and found that depolarization became significant at pressures above $4 \times 10^{-4}$ Torr. This is consistent with the results of Chilton et al. [36]. All of the results reported here were obtained with target pressures below $1.5 \times 10^{-4}$ Torr. It is probable that there is no significant pressure dependence of the Stokes parameters below $3 \times 10^{-4}$ Torr. But even in a worst case, for polarization rising linearly to zero pressure, measurements made at $1.5 \times 10^{-4}$ Torr would be systematically low by only about 0.3% for the situation illustrated in Fig. 3, or about 1.3% of the quoted value. Such errors are not significant given the scatter and statistical uncertainty of our data.

We also investigated the energy width ($\Delta E$) of the photon

![FIG. 2. Integrated Stokes parameters $P_1$ and induced $P_2$ as a function of magnetic field in the target region for the 811 nm transition excited by unpolarized electrons at 12.4 eV (see text). The data are represented by triangles, and theoretical Hanle effect values by lines. The magnetic-field measurement is accurate to 20%. ($P_1$, solid triangles and solid line; $P_2$, open triangles and dashed line.)](image)

![FIG. 3. Depolarization of 811 nm resonance fluorescence vs target pressure at 12.4 eV. The solid line represents an error-weighted least-squares fit to the data. The dotted lines represent the uncertainty in the fit. Pressure is accurate to 25%.](image)
to emitted electron beam. Knowledge of this width is crucial in this experiment, where we are comparing measurements with theoretical results having features as narrow as 0.1 eV. Furthermore, reports in the literature have indicated that the FWHM can grow with both larger emission current [37] and higher GaAs temperature [38]. We determined our beam’s ΔE by scanning its energy across the narrow (≤25 meV) 2s2p 2 2D negative-ion resonance in He [39] and observing the subsequent 3 3D→3 3P fluorescence. A typical data set is plotted in Fig. 4. Because of the narrow resonance width, the beam energy width is essentially given by the width of the fluorescence peak. Using this technique, we determined ΔE [full width at half maximum (FWHM)] to be 0.3(0.05) eV for the operating conditions of our source. Unlike Ref. [37] we found no statistically significant variation of ΔE with extracted beam current over the range in which we operated (≤10 μA).

IV. RESULTS

In this section, we compare the measured integrated Stokes parameters for the 811 nm transition from the well LS-coupled 3D3 state and the 878 nm transition from the intermediatively coupled 3D5 state to those predicted by the 31-state R-matrix calculation. To compare these results directly, we needed to account for the effect of the finite electron beam energy width ΔE on the calculated polarizations. Since we observed the fluorescence through polarization optics, the convolution of the calculation with the electron beam profile must be performed on the individual polarization components before they are combined to yield the Stokes parameters. We assumed an electron beam profile of a Gaussian with an asymmetric low-energy tail, similar to the profiles observed by Kolac et al. [37].

We discuss the integrated Stokes parameters of the 811 nm transition first. Figure 5 is comprised of our present data, the data of Furst et al. [9], and our calculation. The three sets of data for the linear polarization fraction P1 agree qualitatively (notice that the figure has a suppressed zero). The gross features of the present experimental data also compare well with the R-matrix calculation, i.e., the present data have the same configuration of bumps and dips. However, the bump at 12.25 eV in the present data is more pronounced than that of Furst et al. There also seems to be an energy shift among the results, which might explain why the polarization of the present data begins to fall at a lower energy than that of Furst et al., and why there seems to be a misalignment with the convoluted theoretical curve. More importantly, though, there are places where the two experimental data sets disagree by several standard deviations. We can offer no explanation for this, but must attribute it to the significant differences between the two apparatuses we used.

The present P2 data are consistent with the datum of Furst et al. For energies below 12.11 eV, the first cascading threshold for the 3D3 state, our data are consistent both with zero and the convoluted theory. Only above 12.3 eV do the data deviate significantly from zero. However, this may be the result of cascading (only one of the lowest-lying cascading levels is a Russell-Saunders state). Assuming the theory is correct, it should be possible to measure a nonzero value of P2 at about 11.6 eV. However, this is a very difficult measurement because the polarization is small and the background and signal rates are roughly equal. The P3 data of Furst et al. (Fig. 5), like the present data for P1 and P2, are qualitatively similar to the convoluted theory curve (note again the suppressed zero).

The integrated Stokes parameters of the 878 nm transition in Kr (4p3/2p [3/2] 3D5→4p3/2p [3/2] 3P1) are presented in Fig. 6. Here, the agreement between the measured results and
For the case of electron-impact excitation of the well $LS$-coupled $4p^5p^3D_3$ state in Kr, the Breit-Pauli $R$-matrix approach does a good, qualitative job of describing the integrated Stokes parameters $P_1$, $P_2$, and $P_3$, with the caveat that cascading, which is not accounted for in the theory, is responsible for the discrepancies above 12.1 eV incident energy. For the intermediately coupled $4p^5p^3D_2$ state, none of the integrated Stokes parameters are predicted satisfactorily. It is thus evident that the present theoretical description, although being the state-of-the-art method for treating these collisions, needs to be improved. This could be achieved by a relatively straightforward (though computationally very challenging) extension of the $R$ matrix with pseudostate (RMPS) method, described by Bartschat et al. [40], to include relativistic effects. In such calculations, the description of both the collision process and the target structure would likely be improved. On the other hand, it is also clear that experimentalists must possess good knowledge of the characteristics of their apparatuses in order to allow for meaningful comparisons between experiment and theory at a detailed level.

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