Polarization Correlation Measurements of Electron Impact Excitation of H$(2p)$ at 54.4 eV

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(Received 14 October 1997)

First direct measurements are reported of the linear reduced Stokes parameters $\overline{P}_1, \overline{P}_2$ for H(2p) excited by electron impact at the benchmark energy of 54.4 eV. The results differ significantly from previous values deduced from angular correlation measurements which are in serious conflict with all sophisticated theoretical approaches. Our results support the trend of theoretical predictions for $\overline{P}_2$ and confirm that its value is negative at electron scattering angles above 100°, as predicted by theory. [S0031-9007(98)05388-5]

PACS numbers: 34.80.Dp

Polarization correlation measurements employ the electron-photon delayed coincidence technique to permit measurements of the polarization of atomic radiative decay corresponding to electron scattering into a well defined direction. Coincident detection of photon and scattered electron permits a subensemble of the total excited atomic decay radiation to be selected for examination. The polarization correlation method is intimately related to the angular correlation technique in which the intensity variation of the decay radiation corresponding to a particular electron scattering direction is mapped as a function of photon emission direction. Such detailed measurements provide a highly stringent test of scattering theories. However, despite notable recent theoretical successes in describing the scattering of electrons by helium [1] and alkali atoms (Li, Na) [2–5], there has remained a very long standing discrepancy at large electron scattering angles between all sophisticated theoretical calculations [6–10] and the experimental measurements of Williams [11] and Weigold *et al.* [12] of the angular correlation parameters for electron impact excitation of H(2p) at 54.4 eV. Data at this energy have acquired benchmark status for the comparison of experiment with theory at intermediate energies. Given that the hydrogenic system is the only one for which the wave functions are analytically known, the electron-hydrogen system represents a prototype for calculation of more complex interactions. Thus any discrepancy here must be viewed as serious. It is clearly of critical importance to determine whether this discrepancy is due to experimental shortcomings or deficiencies in the theoretical treatment, or to a combination of both.

We report in this Letter the first direct measurements of the linear Stokes parameters of Lyman-α decay radiation (121.6 nm) resulting from electron impact excitation at an incident energy of 54.4 eV. The primary motivation for these measurements is to provide complementary measurements to those previously obtained using the angular correlation technique, and thus to shed new light on the outstanding discrepancy between experiment and theory at large scattering angles.

The electron impact excitation of the 2^2P_j states of hydrogen can be characterized by three independent parameters in addition to the angular differential scattering cross section σ, which have traditionally been taken as

$$\lambda = \frac{\langle |a_0|^2 \rangle}{\sigma}, \qquad R = \frac{\mathrm{Re}\langle a_1 a_0^* \rangle}{\sigma}, \qquad I = \frac{\mathrm{Im}\langle a_1 a_0^* \rangle}{\sigma},$$

where the brackets denote an average over unobserved electron spins of the transition amplitudes a_M for different magnetic substates $|LM\rangle$ of the excited ${}^2P_j$ state. An equivalent parametrization is the set of so-called reduced Stokes parameters $\overline{P}_i$, which describes the nascent excited charge cloud, i.e., immediately following instantaneous excitation at $t = 0$. These two sets are related by [13]

$$\overline{P}_1 = 2\lambda - 1, \qquad \overline{P}_2 = -2\sqrt{2}R, \qquad \overline{P}_3 = 2\sqrt{2}I.$$

The reduced Stokes parameters $\overline{P}_i$ can be derived from experimentally measured Stokes parameters S_{i}, provided account is taken of the depolarization inherent in the evolution of the excited state under the influence of internal forces over its lifetime ($\tau = 1.6$ ns). The experimentally measured Stokes parameters are operationally defined as

$$S_1 = \frac{1}{\varepsilon}\frac{I(0) - I(90)}{I(0) + I(90)}, \qquad S_2 = \frac{1}{\varepsilon}\frac{I(45) - I(135)}{I(45) + I(135)},$$
$$S_3 = \frac{1}{\varepsilon}\frac{I(\sigma^-) - I(\sigma^+)}{I(\sigma^-) + I(\sigma^+)},$$

where $I(\alpha)$ represents the intensity of radiation transmitted by a linear polarization analyzer whose transmission axis is oriented at an angle α degrees with respect to the quantization axis provided by the direction of the electron beam. $I(\sigma^+)$ and $I(\sigma^-)$ represent the transmitted intensities of radiation characterized by helicity $+1$ and -1, respectively, and ε is the polarization efficiency of the analyzer. As far as the two equivalent experimental techniques are concerned, measurements of angular correlations in the scattering plane without regard to polarization analysis of the radiation yield values for only two independent parameters λ and R. A measurement of the circular polarization of the radiation is required to specify I. Such measurements of the circular polarization have been reported by Williams [14] and Nic Chormaic *et al.* [15]. We are concerned here with the linear parameters $\overline{P}_1$ and $\overline{P}_2$ only.

Our experimental geometry is shown in Fig. 1, the coordinate frame shown being the so-called *collision frame*. The momenta $\vec{k}_{\text{in}}, \vec{k}_{\text{out}}$ of the incident primary electrons and detected scattered electrons, respectively, define the scattering plane. Emitted photons are detected in a direction at 90° to $\vec{k}_{\text{in}}$ and at an elevation angle of 45° to the scattering plane, (i.e., $\theta = 90°$, $\phi = 135°$ in the collision frame). Lyman-α radiation from the discharge tube prevents positioning of the polarization analyzer at the obvious position perpendicular to the scattering plane ($\theta = 90°, \phi = 90°$). The general expressions relating the reduced Stokes parameters $\overline{P}_i$ to experimentally measured Stokes parameters S_i for radiation propagating in the direction (θ, ϕ) can be obtained using Eqs. (4.3.11) of Blum and Kleinpoppen [16] and conversion factors for state multipoles and independent parameters given by Anderson *et al.* [13]. A series of purely algebraic substitutions gives two equations which, for the particular analysis direction specified, reduce to

$$\overline{P}_1 = \frac{25S_1 - 3}{3(3 - S_1)}, \qquad \overline{P}_2 = \frac{6\sqrt{2}}{3 - S_1} S_2 . \tag{1}$$

The basic apparatus used in the present experiment is similar to that used previously in this laboratory [15]. A thermal beam of deuterium atoms produced by the dissociation of molecular deuterium in an rf discharge is intersected by an electron beam of energy 54.4 eV (energy spread approximately 0.5 eV) and diameter ~1 mm. (Deuterium is preferred as a target in order to minimize the small effect of the hyperfine structure, which is assumed to be negligible in the development of the reduced Stokes parameter relations.) The discharge source provides 60% dissociation and an atomic density at the interaction point of $\sim 5 \times 10^{11}$ cm^{-3}. Electrons scattered at a given angle are selected for an energy loss of 10.2 eV, corresponding to $n = 2$ excitation, using two consecutive 127° electrostatic analyzers. The circular entrance aperture of the electron energy analyzer subtends a solid angle at the interaction region of 0.002 sr. Lyman-α photons emitted in the direction $\theta = 90°$, $\phi = 135°$ are collected by a VUV polarization analyzer and detected by a channel electron multiplier coated with CsI, which enhances the Lyman-α detection efficiency. The linear polarization analyzer [17] consists of a quartz (fused silica) reflector whose degree of polarization was measured as 83.5 ± 0.5%, using a polariscope arrangement. The polarization analyzer was preceded by a LiF lens of focal length 10 mm at 121.6 nm which increases the solid angle subtended by the analyzer at the point of intersection of the beams (focal point of lens) to 0.6 sr. The large collection solid angle of the polarization analyzer requires that Eqs. (1) be modified to account for this. This was done using the procedure of Goeke *et al.* [18], by integrating the analytical expressions for the relevant Stokes parameters over the acceptance angle of the detector ($\theta \pm \delta, \phi \pm \delta$, with $\delta = 22.5°$) to obtain

$$\overline{P}_1 = \frac{25S_1 - 2.87}{3(2.87 - 0.86S_1)}, \qquad \overline{P}_2 = \frac{8.55}{2.87 - 0.85S_1} S_2 . \tag{2}$$

This change amounts to a correction of at most 5% for the parameters measured. An important experimental check is that measurements at small electron scattering angles (up to 20°) were made both with and without the use of the LiF lens. Without the lens the acceptance solid angle of the polarization analyzer was restricted to 0.008 sr. Good agreement in both sets of measured Stokes parameters was obtained, thus giving confidence that the lens does not spuriously affect the measured polarizations.

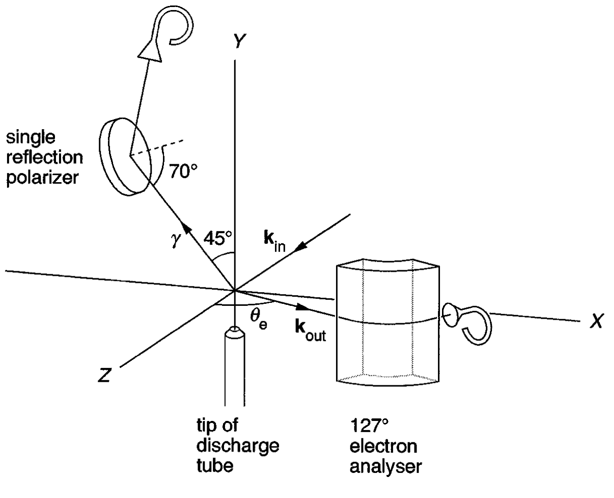

FIG. 1. Schematic diagram of the experimental geometry.

An experimental measurement consists of a series of consecutive measurements at each of the four polarizer orientations. Measurements at low electron scattering angles can be completed to reasonable statistical accuracy in a matter of hours, while the results at high scattering angles are the result of approximately eight weeks of data accumulation at each angle. This is a direct consequence of the sharp decrease in the angular differential cross section for H($2p$) excitation. As is standard practice, all measured photon intensities are normalized to the scattered electron count rate to account for possible variations in electron beam intensity and target density over the course of a run.

As a matter of course in these measurements it is also possible to measure approximately the corresponding noncoincident Stokes parameters (i.e., Stokes parameters for the decay radiation averaged over all electron scattering angles) simultaneously with the coincident parameters. This provides an important experimental check of systematics, in particular, of any system misalignment. The noncoincident S_1 parameter is required to be zero by virtue of the axial symmetry of the measurement. All measurements reported here were supported by a noncoincident S_2 measurement within the range ±0.03. The noncoincident

S_1 parameter is also routinely measured and compared with the established experimental value of 0.12 ± 0.02 [19], but less reliance is placed on this check as our measurement is subject to molecular contamination (~20% of the total photon flux originates from molecular excitation) and atomic cascade contributions. Nonetheless, we typically recorded values in the range 0.11–0.15. These values are also corrected to account for the large acceptance solid angle of the polarization analyzer.

Standard consistency checks have also been satisfied. The electron and photon count rates and the coincidence rate were linear with electron beam intensity and target density. The possibility of Lyman-α resonance trapping was tested at low electron scattering angles by reducing the atomic target density by a factor of 3 and looking for variation in the measured Stokes parameters. No significant variation was observed. The possibility of any polarization sensitivity of the channeltron photon detector was also excluded after a systematic search for any such effect.

Directly measured Stokes parameters and the reduced Stokes parameters derived from these using Eqs. (2) are presented in Table I. Each value represents a least squares combination of several independent experimental measurements. Errors here represent statistical uncertainties combined with the uncertainty in the measured polarization efficiency ε, and are quoted at one standard deviation. Figure 2 compares our measured values for the reduced Stokes parameter $\overline{P}_1$ with the values derived from the angular correlation measurements of Williams [11] and Weigold *et al.* [12]. Also shown are the results of various recent theoretical calculations. Figure 3 displays the same comparisons for $\overline{P}_2$.

The theoretical approaches considered fall into one of two categories: nonperturbative close-coupling calculations [6–9] or perturbative distorted-wave approximations [10]. The various close-coupling calculations differ mainly in their treatment of the continuum via the use of pseudostates, the most ambitious of this type being the 36 state convergent close-coupling calculation of Bray and Stelbovics [6]. The single distorted-wave calculation of Madison *et al.* [10] is exact to second order and includes a treatment of second-order exchange. It can be seen that all of these sophisticated calculations are in broad agreement as to the values of $\overline{P}_1$ and $\overline{P}_2$ over the entire electron scattering range.

Measurements at low electron scattering angles (up to 30°) were undertaken mainly in order to establish that our experimental technique is free of any significant systematic errors, since previous experiment and theory are in reasonable agreement over this range. Indeed, during the long accumulation times required at higher electron scattering angles (~2 months integration), we regularly remeasured small angle values to ensure against any systematic changes in the apparatus. Our measurements of $\overline{P}_1$ are in fair agreement with the previous angular correlation results over the entire angular range measured and confirm that there exists a minimum in the electron scattering range of 90°–120° which is deeper than that predicted by any recent calculation.

It is in values for $\overline{P}_2$ above 90° that the discrepancy between previous experiment and theory is most pronounced, and this has been the primary motivation for this experiment. As can be seen in Fig. 3, the various theoretical approaches all predict negative values for $\overline{P}_2$ in the scattering range above 90°, falling to a minimum of ~ -0.6 at ~130°, while both sets of angular correlation data record only positive mean values in this angular range. In the case of Williams' results [11], the possibility of negative values is strongly rejected on the basis of his reported statistical uncertainties. The current polarization correlation results at 110° and 120° strongly contradict the trend of the angular correlation data and qualitatively support the theoretically predicted variation of $\overline{P}_2$. Error bars on the current data are unfortunately too large to be entirely conclusive; however, on the assumption that the uncertainties are normally distributed, the datum at 110° expresses a 93% probability, and the datum at 120° an 85% probability, that $\overline{P}_2$ is negative in value at these scattering angles. At 120° there is only a 2% probability that the

TABLE I. Directly measured Stokes parameters S_1, S_2 at electron scattering angles θ_e, and reduced Stokes parameters $\overline{P}_1, \overline{P}_2$ derived from these using Eqs. (2). Numbers in brackets represent statistical uncertainty in the least significant digits expressed as 1σ.

θ_e	S_1	S_2	$\overline{P}_1$	$\overline{P}_2$
2°	0.259(015)	−0.053(014)	0.459(048)	−0.169(045)
4°	0.192(014)	−0.174(015)	0.244(047)	−0.553(048)
6°	0.131(009)	−0.242(009)	0.052(029)	−0.748(030)
8°	0.078(016)	−0.249(018)	−0.115(052)	−0.762(056)
10°	−0.013(019)	−0.229(021)	−0.369(061)	−0.681(068)
15°	−0.054(009)	−0.266(010)	−0.480(030)	−0.780(030)
20°	−0.039(019)	−0.221(021)	−0.445(062)	−0.681(068)
30°	0.075(029)	−0.223(028)	−0.120(090)	−0.680(090)
90°	0.153(032)	0.055(033)	0.115(100)	0.170(102)
110°	0.118(038)	−0.055(036)	0.010(114)	−0.169(113)
120°	0.162(057)	−0.062(060)	0.143(175)	−0.194(189)

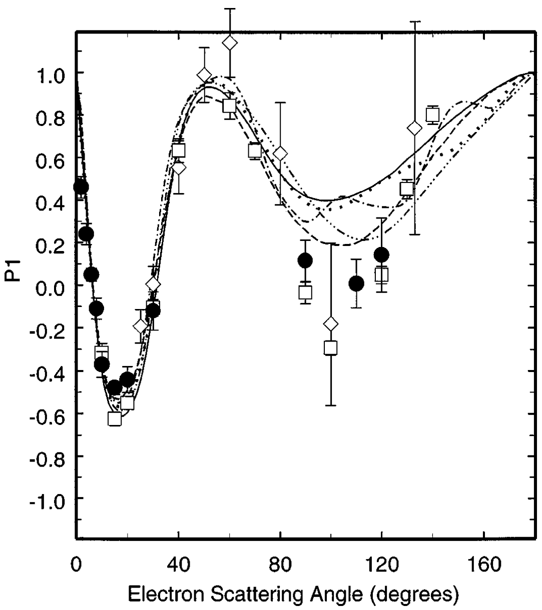

FIG. 2. Reduced Stokes parameter $\overline{P}_1$ as a function of electron scattering angle. The reported polarization correlation measurements (●) are compared with those deduced from the angular correlation measurements of Williams (□) [11] and Weigold *et al.* (◇) [12]. Error bars represent statistical uncertainties quoted at 1 standard deviation. Also shown are the results of recent calculations *viz.*, 36 state convergent close-coupling results of Bray and Stelbovics (—) [6], 17 state close-coupling calculation of Wang *et al.* (· · ··) [7], multipseudostate close-coupling calculation of van Wyngaarden and Walters (–···–) [8], intermediate energy *R* matrix calculation of Scholz *et al.* (– · –) [9], and the second-order distorted wave calculation of Madison *et al.* (-----) [10].

value of $\overline{P}_2$ is as high as the mean value reported by Williams. It is equally clear, however, that the current data are not in accord with theory. Indeed, the probability of a $\overline{P}_2$ value of -0.5 at 120° is estimated to be only 5%. There seems little point, therefore, in discussing the extent of disagreement with the individual theoretical approaches.

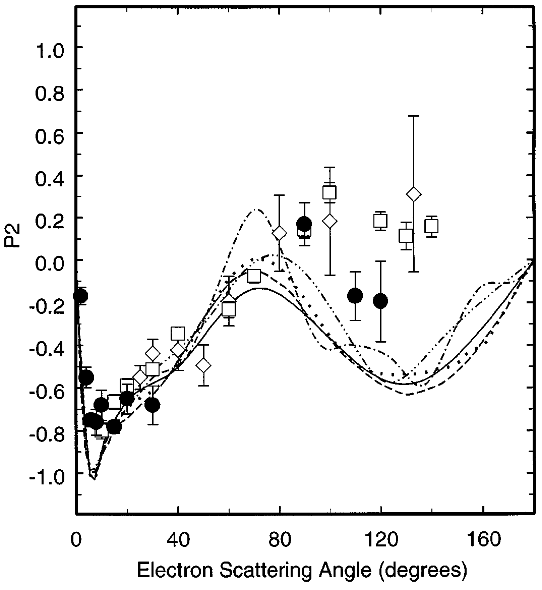

FIG. 3. Reduced Stokes parameter $\overline{P}_2$ as a function of electron scattering angle. Symbols are the same as in Fig. 2.

It is our conclusion that the angular correlation experiments of both Weigold *et al.* and Williams were in error in their measurements of the correlation parameter *R*, and that the resulting discrepancy between experiment and theory, while still not satisfactorily resolved, has been significantly diminished by the polarization measurements reported here. Further experimental effort will be required to confirm this conclusion.

This work was carried out with the support of Forbairt (Ireland) and the EU Human Capital and Mobility program.

Note added—Since the submission of this Letter, Yalim *et al.* [20] have published important and directly relevant angular correlation data for electron impact excitation of H(2*p*) at 54 eV and at scattering angles of 10°, 30°, and 100°. Their measurement at 100° is in good agreement with theory, and, like the present work, strongly suggests that the earlier angular correlation data of Weigold *et al.* [12] and Williams [11] is in error. The new data from both Yalim *et al.* and our own group provide strong evidence of the essential validity of current theories.

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