1s-2p Excitation of Atomic Hydrogen by Electron Impact Studied Using the Angular Correlation Technique

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(Received 11 July 1997)

Excitation of the $2p$ state of atomic hydrogen by electron impact was studied using the electron-photon angular correlation technique with the aim of resolving a long-standing and serious discrepancy between theories and previous experiments at large scattering angles. At a scattering angle of 100°, where the discrepancy was greatest, the present result shows excellent agreement with the theoretically predicted correlations. [S0031-9007(97)04312-3]

PACS numbers: 34.80.Dp

"There has been a very long-standing discrepancy at large scattering angles between theory and measurements of the 2^2P angular correlation parameters for 54.4 eV electron-impact excitation of atomic hydrogen. In our view, this is the most outstanding problem in fundamental electron-atom scattering. The development of most electron-atom scattering theories use the e-H system as a testing ground for dealing with the more sophisticated problems. The discrepancy with experiment here undermines the basic building blocks of such theories. For this reason it is imperative that this problem be resolved as soon as possible" [1]. This quote is typical of a number made by various authors in recent years and it would be difficult for the present authors to put the present experimental study more vividly in context.

Study of electron impact excitation processes using either the electron-photon angular correlation method, in which the angular distribution of the decay photon is measured in coincidence with the electron scattered in a particular direction, or the electron-photon polarization correlation method, where the polarization state of the emitted radiation is determined for a specific scattered electron momentum, is now well established [2]. In principle correlation methods can provide a complete quantum mechanical description of excitation processes. In practice this is true for only a limited number of cases, for example, S-P excitation in helium. Spin averaged correlation studies, such as those reported here, for S-P excitation in hydrogen are incomplete because the different spin channels can be resolved using spin polarized beams. Nevertheless these measurements provide the most stringent test of theoretical models for excitation of hydrogen presently available.

Experimentally, correlation measurements for hydrogen have been hampered by the difficulties of producing suitable beams of hydrogen atoms, stable over a long period of time and by the depolarization of the radiation due to fine structure. Only three independent experimental research groups have made correlation measurements for the $(1S$-$1P)$ excitation process [3–10]. Of these only the data of Weigold *et al.* [5] and Williams [6,7] have been made over a wide range of electron scattering angles at an incident electron energy of 54.4 eV. There is accord between these two sets of data at all scattering angles but because of the large statistical uncertainties in the data of Weigold *et al.* [5] further discussion of that work is not given here. Conversely this excitation process has been the subject of many theoretical studies using a variety of different approaches in this intermediate energy range. More recent examples which give comparison with correlation experiments include the multipseudostate close coupling method of van Wyngaarden and Walters [11], a second-order distorted-wave calculation by Madison *et al.* [12], an intermediate energy R-matrix theory of Scholz *et al.* [13], the convergent close coupling (CCC) method of Bray and Stelbovics [14], and a close coupling calculation of Wang *et al.* [15].

It has become standard practice to express the results of correlation experiments using parameters defined directly in terms of the excitation amplitudes f_M and their relative phases (see [16] for the case of hydrogen) or in terms of the shape and dynamics of the excited state [2]. The latter is particularly useful in developing physical insight into the excitation process. However, comparisons between experiment and theoretical predictions are best made directly with the measured correlations. Since it is serious discrepancies between theory and previous experiments which we investigate here, we restrict the discussion in this Letter to a comparison between measured and theoretically predicted angular correlations. Although the angular correlation method, unlike the polarization correlation method, gives no direct information on the dynamics (angular momentum transfer) of the excitation process, it is ideally suited to investigate the large scattering angle discrepancies between theory and experiment. Its greatest strength is that the measured correlations give a direct measure of the position of the correlation minimum, usually with small uncertainties unless the correlation is nearly isotropic. The long-standing difference at scattering angles greater than 70° manifests itself most clearly in the position of the correlation minimum, which corresponds in magnitude to the angle

γ of the excited state charge cloud relative to the electron beam direction [2].

The situation can be illustrated most dramatically by comparing the experimental and theoretical results at a scattering angle of 100° (Fig. 1). It can be clearly seen that the experimental and theoretical results are almost exactly out of phase with each other. There is excellent agreement between the theoretical data of Scholz *et al.* [13], Bray and Stelbovics [14], and Wang *et al.* [15]. However the earlier close coupling calculation of van Wyngaarden and Walters [11] and the second-order distorted-wave calculation of Madison *et al.* [12] are also in substantial agreement with the other theories.

The apparatus used in the present study is a conventional electron-photon coincidence spectrometer. Apart from the atom source, it is similar to that used for a wide range of correlation studies in helium [17]. Briefly, a beam of electrons with energy spread ≤ 0.5 eV and current $\leq 1 \times 10^{-6}$ A crosses at right angles a beam of hydrogen atoms. A microwave discharge source is used as the H-atom source. It is of the type discussed in detail by McCullough *et al.* [18] with the microwave power (at 2.45 Ghz) coupled to a Pyrex glass discharge tube through two slotted line radiators. This device is capable of producing a dissociation fraction of 95% and atom beam densities of 4×10^{13} cm^{-3}. In our case the H atoms were transported to the interaction region through a Pyrex capillary of length 35 mm and internal diameter 0.5 mm. Scattered electrons which have excited $\mathrm{H}(n = 2)$ states are selected by a hemispherical electrostatic analyzer and detected by a channel electron multiplier. Lyman-alpha photons from decay of $\mathrm{H}(2p)$ are detected by a channel electron multiplier preceded by a LiF window to minimize the random coincidence signal due to shorter wavelength photons and by a system of grids to prevent charged particles reaching this detector. Both the scattered electron and photon detectors are rotable over a wide range of angles about the interaction center. Standard coincidence methods are used to measure the reported angular correlations with the coincidence signal determined for a range of photon detector angles at fixed electron scattering angles.

Numerous procedures, tests, and checks have been performed similar to those previously carried out for a wide range of correlation data reported for helium and krypton from this laboratory ([17,19], and references therein). For example, the scattered electron signal was used to start the ramp of a time-to-amplitude converter with the true coincidence signal being normalized to the scattered electron signal. This eliminates the effects of any variations in the incident electron and atom beam fluxes and the electron detector efficiency. Small variations in the photon detector efficiency were minimized by averaging the coincidence signal at each photon angle from a number of scans of the photon angular range in opposite directions. The apparatus was initially checked out by measuring 2^1P angular correlations in helium at 80 eV over a range of scattering angles using a conventional capillary to produce the helium beam. These measurements were then repeated by flowing helium through the hydrogen discharge assembly. Identical results were obtained

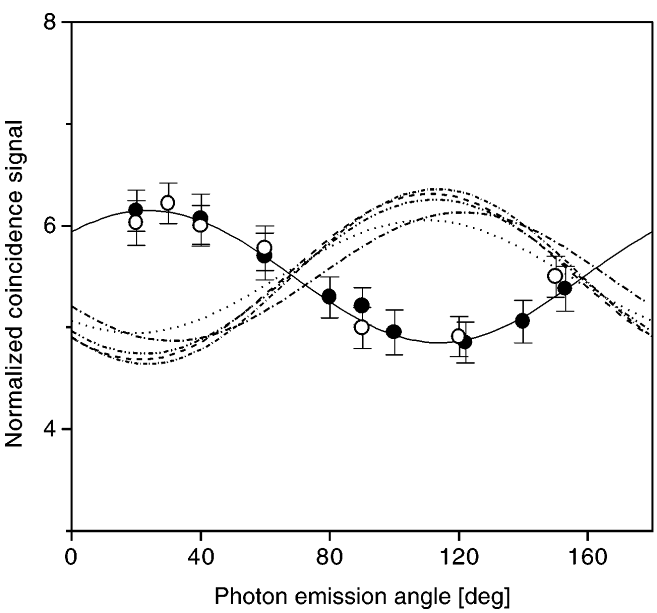

FIG. 1. A comparison between previous experimental and theoretical predictions of the $\mathrm{H}(2p)$ angular correlation at 54.4 eV and an electron scattering angle of 100°. Experimental data of Williams [6,7], circles and solid line fit to data. Theories: dotted line [11]; dash-dotted line [12]; short dash-dotted line [13]; dashed line [14]; dash-double-dotted line [15].

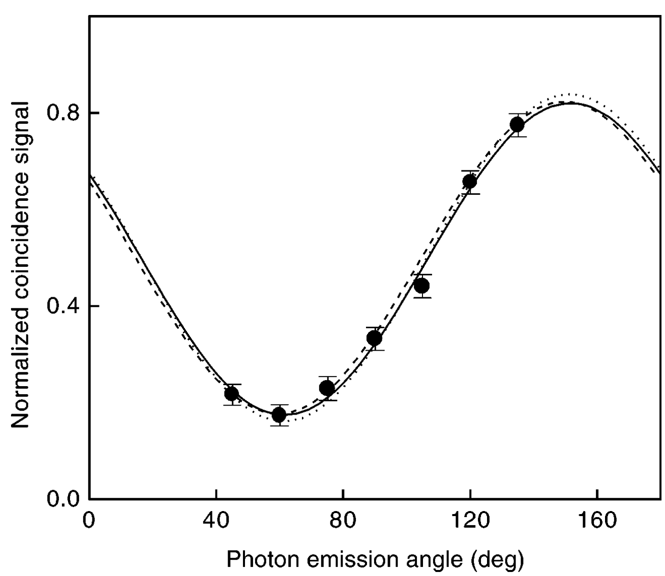

FIG. 2. Angular correlation for excitation of the 2^1P state of helium at 80 eV and a scattering angle of 25°. Present data, circles and solid line fit; dotted line, fit to data of Hollywood *et al.* [20]; dashed line, CCC calculation of Fursa and Bray [21].

which agree well with previous data [20] and recent calculations [21,22].

A typical angular correlation is shown in Fig. 2, obtained with the helium beam from the H source. This agreement gives general confidence in the performance of the coincidence spectrometer.

For the studies with atomic hydrogen we have maintained similar operating conditions as for helium. Under these conditions the source displays a number of features which are important for correlation measurements. It operates continuously over several months and is highly stable over that period as monitored by frequent measurements of the energy loss spectrum. Stray radiation from the source, detectable by the photon detector, is also small. The source is not operated to maximize dissociation fraction or beam flux. Further work is required to establish the extent to which the present operating conditions are due to the lower conductance of the Pyrex capillary compared with those reported in [18].

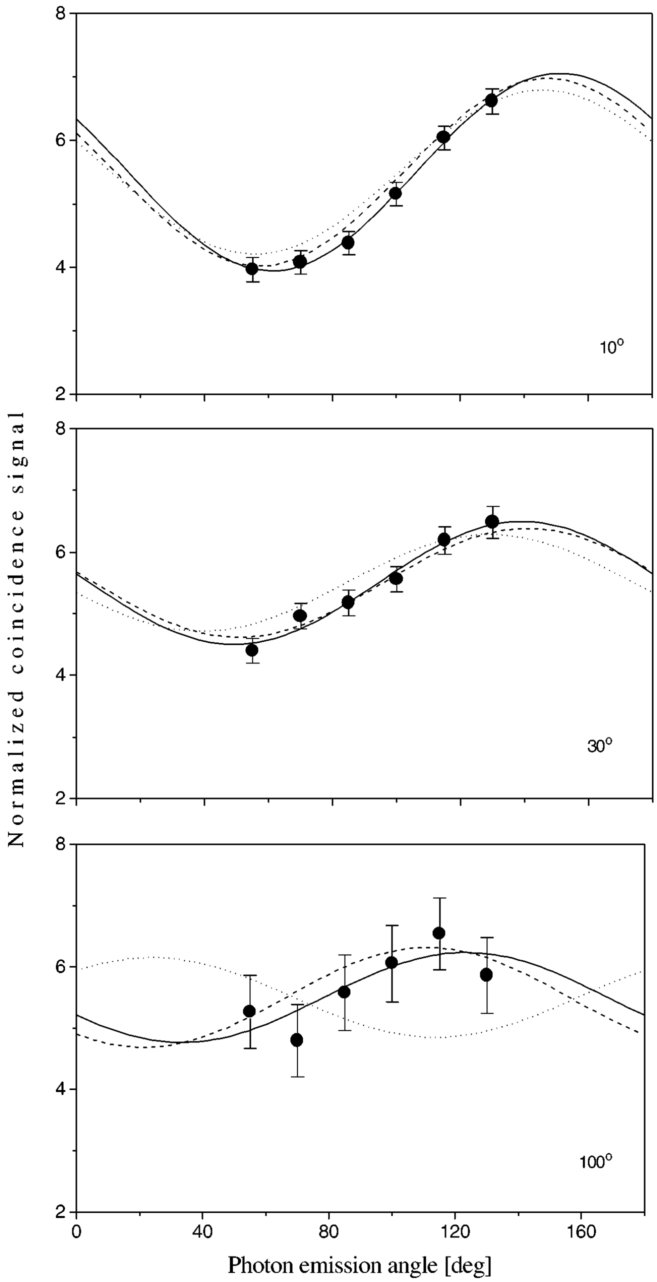

FIG. 3. Angular correlations for excitation of the 2^2P state of hydrogen at 54.4 eV and scattering angles of 10°, 30°, and 100°. Present data, circles and solid line, fit; dotted line, best fit to data of Williams [6,7]; dashed line, CCC calculation of Bray and Stelbovics [14].

Figure 3 shows the present $H(2p)$ angular correlations measured at scattering angles of 10°, 30°, and 100° at an incident electron energy of 54.4 eV, compared with a recent typical calculation. It can be seen that excellent agreement is obtained with the CCC correlations at all three scattering angles. At small scattering angles, the new data simply confirm the previous agreement between theory and experiment. At 100° the previous experimental data could not be reproduced. This result cannot be considered as a major surprise, since confirmation of the previous experimental result would have raised fundamental questions about our understanding of simple atomic collision processes. What is unclear is the reason for the discrepancy between the previous experimental data and theory and now the present data at scattering angles greater than 70°. Only the difficulties of these measurements due to the low differential cross sections at large angles and the small amplitudes of the correlations due to fine structure depolarization can be emphasized.

In conclusion we have presented new experimental data which resolve the long-standing and fundamental disagreement between a range of respected theoretical models and previous experiments at a scattering angle of 100°. This program of work is continuing with the aim of covering the scattering angular range 10°–170°, complemented by polarization correlation studies.

This work is supported by the U.K. Engineering and Physical Science Research Council. We were first persuaded to develop this experiment by Professor D. H. Madison. We acknowledge the guidance of Dr. R. W. McCullough and Professor J. Geddes in setting up the H-atom source. The experiment would not have been possible without the technical support of A. Bott, C. Burrow, R. Ridley, J. Corner, and M. Black.

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