Electron-photon angular correlation measurements for the 2P state of hydrogen at 35 eV

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Electron-photon angular correlations have been measured for excitation of the 2P states of hydrogen at an incident energy of 35 eV. The data presented relate to electron-scattering angles from $\theta_e = 8^\circ$ to $\theta_e = 120^\circ$ and yield values for the parameters λ and R. The experimental results are compared with several theories. In general, none of the theories is found to be adequate at this energy.

INTRODUCTION

The study of electron-impact excitation processes using the electron-photon delayed coincidence technique is now well established in experimental atomic collision physics. Its primary importance lies in its capability of measuring the interference between coherently excited magnetic sublevels. The experimental data from coincidence experiments relate directly to the way in which angular momentum is transferred to the atom during the collision process, and is thus intimately associated with the collision dynamics. These experiments provide very sensitive tests of theoretical approximations developed to predict the excitation process.

Over the last two decades, the study of excitation processes in the "intermediate"-energy region has received considerable attention from both experimentalists and theoreticians alike. This energy domain is loosely defined as extending from some low energy (typically a few eV above threshold) below which a truncated eigenfunction expansion of the scattering wave function can be expected to provide as adequate theoretical description, to a sufficiently high energy for the first-Born approximation (FBA) to be valid. The latter limit has always been the subject of some controversy, though it is now generally accepted that the FBA is not accurate below 1-keV incident energy. A proliferation of scattering approximations has resulted, each attempting to provide an accurate theoretical description of collisions at these energies, with a concomitant demand for experimental measurements of as many collision parameters as possible and of a quality sufficient to check the range validity of any particular approximation. Within this context, the

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electron-photon coincidence technique as a means for measuring target or source parameters of the excitation process has become increasingly important in delineating the strengths and weaknesses of any particular theoretical model.

Because of its simplicity, atomic hydrogen has always played a central role in experimental and theoretical studies of atomic collisions. Since hydrogenic wave functions are exact, a theoretical description of the collision process depends only on an adequate treatment of the scattering dynamics, and an imprecise description of the collision target does not contribute any uncertainty to the theoretical results. Recently, excitation of the 2P state of atomic hydrogen has received considerable attention from both experimentalists and theoreticians. Dixon et al.,¹ Hood et al.,² and Weigold et al.³ have reported coincidence data for electron-impact energies in the range 40-200 eV. Hollywood et al.⁴ and Williams⁵ have made similar measurements at 54.4 and 100 eV, while Slevin et al.⁶ have reported data for small electron scattering angles at 55 and 100 eV. In this paper, we present an extensive series of measurements at an incident electron energy of 35 eV, and compare our data with available theoretical results. This energy is in the most difficult part of the intermediate-energy range for theory. It is below the generally accepted lower limit for distorted wave, eikonal, and second-Born approximations, which are essentially high-energy models. Further, the existence of resonances in the pseudostates commonly used in close-coupling calculations to take account of the atomic states excluded from the expansion severely complicates and limits the usefulness of these essentially low-energy approximations. These coincidence data can thus be expected to provide a target for the development of theoretical techniques to bridge the gap between the low- and high-energy regions.

The theory and method of electron-photon coincidence have been treated extensively in earlier publications (see, for example, Macek and Jaecks,⁷ Eminyan *et al.*,⁸ Slevin *et al.*,⁹ and Williams⁵), and only a brief summary is given here of the relevant features of the experimental technique and interpretation of the data.

THEORY

When an electron of given energy E is scattered in a particular direction θ_e relative to the incident electron beam, the electron-photon angularcorrelation function for an *S-P* transition in hydrogen can be written (see Morgan and McDowell¹⁰):

$$N(E,\theta_e,\theta_\gamma) = 4 + 3\lambda + 3(1-2\lambda)\cos^2\theta\gamma - 3\sqrt{2}R\sin 2\theta_\gamma, \qquad (1)$$

where θ_{γ} specifies the direction of photons emitted by the excited atoms in the plane of scattering: $R = \operatorname{Re}\langle a_0 a_1 \rangle / \sigma$, the quantity $\langle a_m a_{m'} \rangle$ denoting the spin-averaged product of the scattering amplitudes for exciting the magnetic sublevels of the 2P state (l=1); with $\langle a_m a_m \rangle = \sigma_m$, the cross section for excitation of the *m* sublevel and $\sigma = \sigma_0 + 2\sigma_1$, the full differential cross section for *S*-*P* excitation; $\lambda = \sigma_0 / \sigma$. From the definitions of λ and *R*, it is easily shown that the function $N(E, \theta_e, \theta_\gamma)$ is restricted to the interval

$$4 \leq N(E, \theta_e, \theta_v) \leq 7$$
.

Thus a measurement of the angular-correlation function, fitted to the functional form of Eq. (1), allows a determination of the two parameters λ and Rfor a particular electron-incident energy E and scattering angle θ_e . Equation (1) is true only for infinitesimally small electron and photon detector solid angles. The averaging effect of the finite aperture of the photon detector can be allowed for exactly. Since the dependence of λ and R on θ_e depends on a knowledge of the collision dynamics, the values of λ and R derived from the raw experimental data must be assumed to be averaged over the small range of scattering angles intercepted by the electron detector.

The angular-correlation data can also be analyzed to yield values of the alignment and orientation parameters defined by Fano and Macek,¹¹ as well as the multipole moments of the excited state (Blum and Kleinpoppen¹²).

EXPERIMENTAL METHOD

The experimental method consists of crossing an atomic hydrogen beam with an energy-selected beam of electrons. Delayed coincidences are observed between electrons scattered inelastically with the 10.2-eV energy loss corresponding to 2P excitations and the Lyman- α photons which result from the decay of the excited atoms. The experimental geometry is coplanar, with electron and photon detectors located in the same plane.

The hydrogen atoms are produced in an rf discharge with approximately 95% dissociation and the atom beam is formed by effusion from a 1-mm capillary joined to the discharge tube.¹³ A conventional electron gun is used to produce an electron beam, and two 127° analyzers, joined in tandem, are used to analyze the scattered electrons. The interaction volume, approximately 1.5 mm³, is located about 0.25 mm above the atom-beam capillary. The Lyman- α radiation is detected by a channel electron multiplier, preceded by a LiF window to prevent light of shorter wavelength than 121 mm from reaching the detector. The density of hydrogen atoms in the interaction region is $\sim 2 \times 10^{12}$ atoms cm^{-3} , a sufficiently low pressure to avoid the problem of trapping of the resonance radiation. The angular resolution of the electron analyzer is limited by an entrance cone, and the acceptance solid angle varied from about 3×10^{-3} sr to 1.0×10^{-2} sr for the data reported here.

Pulses from the electron and photon detectors are amplified by 300-MHz amplifiers and conventional electronics are used to measure the coincidence rate of electrons scattered at a particular angle with 10.2-eV energy loss and Lyman- α photons resulting from the decay of the 2*P* excited states. The angular correlation is determined by measuring the coincidence rate as a function of photon detector angle for a fixed incident energy and electron scattering angle. A complete description of the experimental apparatus and method is outlined in an earlier publication.⁹

The energy-loss spectrum (Fig. 1) obtained by scanning the voltage slit of the analyzer, and displayed on a multichannel analyzer, shows the operation of the rf discharge source.¹³ With the discharge on, a large peak occurs at 10.2 eV corresponding to 2P excitations, followed by a broad maximum at 12.3 eV corresponding to $n \ge 3$ excita-



FIG. 1. Energy-loss spectrum at 35 eV at an electron scattering angle $\theta_e = 5^\circ$. •, rf discharge on; \bigcirc , rf discharge off.

CHANNEL

NUMBER

tions. With the discharge off, a broad peak occurs at 12.9 eV corresponding to excitation of the $B^{1}\Sigma_{u}^{+}$ and $C^{1}\Pi_{u}$ molecular states. The electron analyzer was operated in a low-resolution mode in order to minimize transit-time variations in the scattered electron trajectories, and the overall energy resolution of the system was about 1.0 eV.

The quality of the hydrogen source is demonstrated in Fig. 2, where the coincidence count rate between scattered electrons and Lyman- α photons is plotted as a function of electron energy loss. With the discharge off, a broad peak occurs at 12.9 eV as with the energy-loss spectrum corresponding to excitation of molecular states. The Franck-Condon factors for the decay of these states allow the wavelength of the corresponding photons to fall within the bandpass of the photon detector. With the discharge on, a large peak appears at an energy loss of 10.2 eV corresponding to 2P excitations, while the peak at high-energy loss almost disappears. The absence of a coincidence signal at the higher-energy loss in this case is due to the wavelength cutoff of



FIG. 2. Electron-photon coincidence count rate as a function of energy loss at 35 eV at an electron scattering angle $\theta_e = 5^\circ$. •, rf discharge on; \bigcirc , rf discharge off.

the LiF window, which prevents photons for the $n \ge 3$ atomic decays from reaching the photon detector. The data in Fig. 2 permit a lower limit to be determined for the dissociation fraction of the



FIG. 3. Electron-photon angular correlations for (a) $\theta_e = 12^\circ$ and (b) $\theta_e = 100^\circ$ at 35 eV. Error bars represent two standard deviations. Full curve represents a least-squares fit of Eq. (1) to the data.

hydrogen source, which in the case of the data shown is 92%.

Two typical angular-correlation curves are shown in Fig. 3 for electron-scattering angles $\theta_e = 12^\circ$ and 100° at 25 eV. The full curve in each represents the normalized computer fit of Eq. (1), corrected for the infinite photon aperture, to the data points. These curves are shown to illustrate the quality of the data. The total time taken to accumulate the data shown for these curves was ~ 2 h for $\theta_e = 12^\circ$ and ~ 4 d for $\theta_e = 100^\circ$. In comparison with similar data reported for 2^1P excitations in helium from this laboratory,⁹ these integration times are significantly better and reflect the efforts made to optimize all apparatus functions in order to maximize the coincidence signal.

EXPERIMENTAL RESULTS AND DISCUSSION

The experimental results are summarized in Tables I and II. Table I contains a selection of the raw data from which values of the parameters λ and R were deduced. These data can be compared with theoretical results, and agreement between

theory and experiment interpreted in terms of the χ^2 probability of the fit. Table II summarizes all the results at 35 eV for the two parameters λ and R.

The data are displayed graphically in Figs. 4 and 5 along with the results of several theoretical calculations. With the exception of the distorted-wave results of Madison,¹⁴ all the calculations used the close-coupling approximation. Morgan¹⁵ has used both a 3-state and a 12-state basis to expand the scattering wave function, but does not include exchange. Kingston et al.¹⁶ have included exchange in a 3-state 1s-2s-2p close-coupling expansion, and Edmunds and McDowell¹⁷ have used a 10-state basis without exchange. In the case of the λ parameter all the theories are in excellent agreement with the experimental data for small electron scattering angles $\theta_e < 70^\circ$, but only Morgan's¹⁵ 12-state calculation predicts the experimentally observed largeangle behavior. For the parameter R, theory and experiment are in disagreement over the entire range of electron scattering angles. While the distorted-wave model is not expected to provide an adequate description of the excitation at such a low energy, suitably formulated close-coupling models might be expected to offer the best theoretical ap-

tion $egin{array}{c} heta_{\gamma} \ N \end{array}$ $\theta_e = 12^\circ$ 70 50 90 105 120 135 4.36 4.83 5.56 6.02 6.38 6.53 σ 0.02 0.02 0.02 0.02 0.04 0.04 θ_{γ} $\theta_{e} = 20^{\circ}$ 40 55 75 95 115 135 N 4.57 4.42 4.64 5.16 5.77 6.39 0.07 0.10 0.08 0.08 0.03 0.06 σ θ_{γ} $\theta_e = 40^\circ$ 30 40 60 80 100 120 N 4.80 4.86 5.37 5.72 5.99 6.13 0.10 0.05 σ 0.13 0.12 0.06 0.12 $\theta_e = 60^\circ$ θ_{γ} 40 60 70 80 100 120 Ň 5.06 6.23 6.39 6.93 7.05 6.14 σ 0.19 0.14 0.12 0.12 0.16 0.15 $\theta = 80^{\circ}$ θ_{γ} 40 60 80 100 120 140 N 6.09 6.21 6.40 5.84 5.04 4.66 0.14 0.09 σ 0.14 0.09 0.14 0.08 θ_{γ} $\theta_e = 100^\circ$ 40 60 80 100 120 140 Ń 5.71 5.96 5.50 5.18 5.15 5.11 0.16 σ 0.21 0.17 0.24 0.17 0.24

TABLE I.	Values of	the normaliz	ed coinci	dence rate	N as a	function	of the ph	oton d	letec-
tion angle θ_{γ} (a	degrees) at	t different θ_e f	for 35-eV	electrons.	Errors	represent	one stan	dard d	levia-
tion									

TABLE II. Values of the parameters λ and R at different electron scattering angles θ_e for an incident energy of 35 eV. Error bars represent one standard deviation.

θ_e	$\lambda = \sigma_0 / \sigma$	$R = \operatorname{Re}\langle a_0 a_1 \rangle / \sigma$
8	0.71±0.03	0.13±0.01
12	0.53 ± 0.02	0.26 <u>+</u> 0.01
16	0.36 ± 0.02	0.28 ± 0.01
20	0.35 ± 0.02	0.22 ± 0.01
25	0.36 ± 0.02	0.21 ± 0.01
30	0.37 ± 0.04	0.14 ± 0.02
40	0.66 ± 0.03	0.11±0.01
60	1.03 ± 0.06	0.02 ± 0.02
70	0.83 ± 0.06	-0.07 ± 0.02
80	0.72 ± 0.04	-0.017 ± 0.02
90	0.61 ± 0.05	-0.06 ± 0.02
100	0.51 ± 0.05	-0.09 ± 0.02
110	0.36 ± 0.04	-0.02 ± 0.03
120	0.33 ± 0.08	0.09±0.04

proach in this energy range. The sharp contrast between theory and experiment for the two parameters λ and R in the case of Morgan's calculations is an indication of the inadequacy of current theory at this energy. Thus, whereas the 12-state expansion provides the best agreement for λ , a reduced 3-state basis is much closer to experiment for R. The generally poor agreement for R may reflect the fact



FIG. 4. Variation of λ with scattering angle at 35 eV. Experimental data, error bars representing two standard deviations. —, Morgan (Ref. 15) (12 state); ---, Morgan (Ref. 15) (3 state); $-\cdot - \cdot - \cdot$, Kingston *et al.* (Ref. 16). — —, Madison (Ref. 14); $\cdot \cdot \cdot \cdot$, Edmunds and McDowell (Ref. 17).



FIG. 5. Variation of R with scattering angle at 35 eV. Symbols as in Fig. 4.

that this particular parameter depends strongly on the relative phases of the excitation amplitudes. Morgan¹⁵ points out that while all close-coupling calculations contain the dominant long-range interactions, the phases may be sensitive to shortrange effects which are only poorly accounted for in these models.

Although no other experimental data is available at this energy, Weigold *et al.*³ and Williams⁵ have carried out a similar series of measurements at 54.4 eV. Their data is qualitatively similar to that presented here. Both experiments show that λ exhibits two deep minima at small and large scattering angles, while *R* has a sharp maximum at small values of θ_e and is negative over an extended range of large angles. Theoretical predictions for *R* are again particularly poor, and this parameter thus appears to provide the most demanding test of theory.

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