Measurement of absolute differential cross sections for the excitation of atomic hydrogen to its $n=2$ level by electron impact

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Using a crossed-beam method, we have measured absolute differential cross sections for the excitation of atomic hydrogen to its $n=2(2^2S+2^2P)$ level by electron impact. The angular range covered was from 12° to 156° in 12° increments, while the impact energies treated were 15, 20, 30, and 40 eV. Absolute integrated excitation cross sections were calculated from the differential ones. Agreement of our data with other recent data and calculations is quite good, but there are still some discrepancies among our results and the older data.

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I. INTRODUCTION

Perhaps the most important unresolved problem in atomic collision processes is the angular behavior of electrons inelastically scattered from atomic hydrogen $[1,2]$. There has been a voluminous amount of effort directed toward solving this problem theoretically for decades $[3-5]$. But it has only been in the past few years with the convergent closecoupling (CCC) method of Bray and co-workers that the computations have approached a satisfactory state $[6-8]$. Experimentally the situation is worse. Only three other research groups beside ours have conducted absolute cross-section measurements, and in all cases these were limited to $n=2$ excitation. Williams and Willis conducted measurements over an impact energy range from 13.87 to 680 eV $[9-11]$, while measurements at 100-eV impact were made by Doering and Vaughn $[12]$. Most recently Khakoo and co-workers reported experimental results in the 30–100-eV range [13,14]. (A fourth group also made measurements, but they were relative, not absolute $[15]$.) Considering this dearth of data, it would be beneficial to have more data to provide better insight into what the correct cross sections values are.

In this paper we shall present the results of our absolute differential cross section (DCS) measurements for the excitation of atomic hydrogen's $n=2$ level by electron impact. Discrimination between the 2 ${}^{2}S$ and 2 ${}^{2}P$ states' excitations was not made. We employed a modulated crossed-beam method, and covered the impact energy range of 15–40 eV, and the angular range of 12° through 156° in 12° increments. Absolute integrated cross sections (ICSs) were calculated from the measured differential ones. A comparison of our results and those of others—both experimental and theoretical—is provided.

II. EXPERIMENT

Our apparatus and experimental procedures have been described extensively elsewhere, so here we give only a brief accounting of them $[16–18]$. Our system is housed in a differentially pumped, dual-vacuum-chamber enclosure. Three mutually perpendicular sets of Helmholtz coils surround this enclosure. They attenuate unwanted magnetic fields to less than 20 mG in any direction within the enclosure's interaction region. Research-grade molecular hydrogen is introduced into this enclosure from a commercial storage cylinder. It is then dissociated in an extended Evenson cavity by microwave discharge. The mixed beam thus produced was measured by a quadrupole mass spectrometer to be consistently $55\pm3\%$ atomic hydrogen in the interaction region, where electron collisions occur. (Improved methods which boost the dissociation fraction to more than 80% and maintain high beam intensity have become available since we conducted our measurements $[19]$.) Just before entering the interaction region, the beam is chopped at audio frequencies by a toothed wheel.

Electrons are produced by a gun based on a tungsten filament and pass through a 127° cylindrical energy selector before being accelerated to the required impact energy. The beam thus produced can be rotated continuously from -90° to 160°, has an angular spread of ± 3 ° full width at half maximum (FWHM), and has an energy spread of 180-meV FWHM. The mean energy of the electrons in the beam was established with help from the 19.34-eV resonance of helium. Scattered electrons are received by a detector fixed to the lower vacuum chamber's wall. This detector subtends a solid angle of about 5×10^{-4} sr, and is based on a 127° cylindrical energy analyzer and a Channeltron electron multiplier.

During measurements the scattering angle and impact energy are fixed, while the energy-loss acceptance window of the detector is swept over the energy-loss region of interest under the control of a dedicated microcomputer running locally developed software. This computer also performs the signal subtraction required by the beam modulation and accumulates and stores the data. Data are collected over the prescribed energy and angular ranges. The results are energy-loss spectra like the one displayed in Fig. 1.

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FIG. 1. A typical electron-energy-loss spectrum for excitation of the $n=2$ level of atomic hydrogen by electron impact. The impact energy was 20 eV, while the scattering angle was 24°. The leftmost peak represents elastic scattering. The peak to its right, at about 10.2-eV energy loss, represents $n=2$ excitation. The peak at about 12.1-eV energy loss represents $n=3$ excitation. Peaks representing $n=4$ and 5 excitations are barely discernible near 12.7- and 13.0-eV energy loss, respectively. There is an axis break in the figure to emphasize the inelastic excitation features.

III. DATA ANALYSIS

The mixed character of our hydrogen beam makes analysis of our data slightly complicated, since the signal contains both atomic and molecular hydrogen contributions. The method of handling this problem by modulating the beam is treated in detail elsewhere $[20,21]$. The result is that

$$
\frac{d\sigma_{\text{H},n=2}}{d\Omega} = \eta \frac{S_{\text{H},n=2}}{S_{\text{H}+\text{H}_2,\text{elas.}}} \left[\frac{d\sigma_{\text{H,elas.}}}{d\Omega} + \left(\frac{1-D}{\sqrt{2}D} \right) \frac{d\sigma_{\text{H}_2,\text{elas.}}}{d\Omega} \right],
$$
\n(1)

where $d\sigma_{H,n=2}$ / $d\Omega$ is the cross section for $n=2$ excitation, η is a factor accounting for the nonconstancy of the detector's efficiency with respect to energy loss, and $S_{H,n=2}$ and $S_{\text{H+H}_2 \text{elas}}$ are the signal strengths for the $n=2$ and elastic peaks, respectively. $d\sigma_{\text{H,elas.}}/d\Omega$ is the elastic cross section for atomic hydrogen, *D* is the dissociation fraction, and

TABLE I. Sources of uncertainty and net uncertainty in our measurements.

Uncertainty source	Value $(\%)$			
Raw data (Statistics)	5			
Dissociation fraction	3			
Transmission correction	4			
Elastic DCS's	15			
Net for $n=2$ excitation DCS's	17			
Angular extrapolation	10			
Net for $n=2$ excitation ICS's	20			

 $d\sigma_{\text{H}_2\text{elas}}/d\Omega$ is the elastic cross section for molecular hydrogen. For these last two cross sections we chose values we determined from previous measurements $[22-24]$.

Once we obtained the absolute differential cross sections, we employed the trapezoid rule to calculate absolute integrated cross sections with the formula

$$
\sigma_i = \int d\Omega \frac{d\sigma}{d\Omega},\tag{2}
$$

where σ_i is the integrated cross section. This required that we extrapolate our results to both 0° and 180°, which we did in a semiexponential manner. Uncertainty introduced by this, as well as uncertainties in other quantities, are provided in Table I. This table also gives net uncertainties. Since the uncertainties were independent of each other, the net uncertainty was determined by the addition of their values in quadrature.

IV. DISCUSSION OF RESULTS

Values for all the cross sections we determined are presented in Table II. Figure 2 shows DCSs at 15-eV impact. Significant backscattering is apparent in these. Williams and co-workers $[9-11]$ measured the DCSs at nearby impact energies, but we chose not to show them. This is because the proximity to threshold energy and presence of resonances in this energy region makes a comparison between our data and those of Williams and co-workers meaningless here. For the same reason we chose not to display the calculations of Bray and co-workers $[6-8]$ at nearby energies.

Figure 3 shows DCSs at 20-eV impact, along with those

TABLE II. Absolute cross sections for the excitation of atomic hydrogen's $n=2$ level by electron impact. Units for the differential cross sections are 10^{-18} cm²/sr, while those for the integrated cross sections are 10^{-18} cm². Parentheses enclose extrapolated values.

	θ (deg)														
	12	24	36	48	60	72	84	96	108	120	132	144	156	168	σ_i
E (eV)															
15	44	19	9.0	3.6	2.2	1.6	1.4	1.2	1.4	1.5	1.8	2.4	2.8	(3.2)	48
20	68	22	7.7	3.4	2.2	1.5	1.5	1.3	1.4	1.3	1.4	1.4	1.7	(1.9)	57
30	110	22	4.3	2.2	1.4	1.0	0.79	0.59	0.51	0.47	0.45	0.39	0.32	(0.30)	64
40	140	17	3.1	1.5	0.70	0.44	0.31	0.29	0.26	0.19	0.18	0.16	0.13	(0.10)	67

FIG. 2. Absolute differential cross sections for the excitation of atomic hydrogen's $n=2$ level at 15-eV impact.

measured by Williams and co-workers at 19.58-eV impact, and those calculated by Bray and co-workers at the latter impact energy. Agreement among all three sets of results is satisfactory at low angles, while there is a slight tendency for the experimental values to run below the theoretical ones at middle angles. But at higher angles our results and those of Bray and co-workers correspond to each other well, with Williams's being somewhat greater. This is curious, as we used our own elastic cross section values to normalize our $n=2$ results. Had we used Williams and co-workers' values, we would have arrived at lower $n=2$ cross section values at high angles, disagreeing with Bray and co-workers' predictions and increasing our discrepancy with Williams and coworkers' values even more. There are other calculations—for example those of Madison $[25]$ at 20-eV impact and those of Scholz and co-workers $[26]$ at 19.59-eV impact. We chose not to display these to keep the figure uncluttered. We have very good agreement with the latter even at high angles, but come in with lower cross sections than former, especially at high angles. The values of the former are greater than ours by about a factor of 2 over the entire angular range. This is

FIG. 3. Absolute differential cross sections for the excitation of atomic hydrogen's $n=2$ level at 20-eV impact, along with those of Williams and co-workers $[9-11]$ at 19.58-eV impact and Bray and co-workers $[6-8]$ at 19.58-eV impact.

FIG. 4. Absolute differential cross sections for the excitation of atomic hydrogen's $n=2$ level at 30-eV impact, along with those of Khakoo and co-workers $[13,14]$ and Bray and co-workers $[6-8]$ at this impact energy.

not surprising, as the distorted-wave second-order Born approximation used to generate them is not expected to be accurate at such low energies [25].

Figure 4 shows our data at 30 eV, along with the measurements of Khakoo and co-workers $[13,14]$ and the computations of Bray and co-workers. Agreement among all three sets of results is quite good, except in the vicinity of 100°, where the experimental DCSs are consistently slightly smaller than the theoretical ones.

Figure 5 gives our DCSs at 40-eV impact. Those measured by Khakoo and co-workers and those calculated by Bray and co-workers are also provided. Especially encouraging is the agreement among our results and those of Khakoo and co-workers. This agreement occurs not only at the forward and middle angles, but also for the case of backscattering. Again there are other calculated results that are not shown to keep the figure uncluttered. These include the results of Scholz and co-workers, which match our values quite well over the entire angular range. Madison's calcu-

FIG. 5. Absolute differential cross sections for the excitation of atomic hydrogen's $n=2$ level at 40-eV impact, along with those of Khakoo and co-workers $[13,14]$ and Bray and co-workers $[6-8]$ at this impact energy.

FIG. 6. Absolute integrated cross sections for the excitation of atomic hydrogen's $n=2$ level by electron impact.

lated DCS's again exceed ours by about a factor of 2 over the entire angular range.

Figure 6 gives our ICSs. They increase gradually over the impact energy range we treated. Comparison of our 15-eV impact data with Bray and co-workers' 13.58-eV predictions is not made, as the known presence of resonances in this region and the difference in residual electron energy by nearly a factor of 2 so close to threshold would make the comparison meaningless.

Bray and co-workers' CCC method has been shown to provide excellent predictions for electron-He scattering

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 $[27,28]$, but there has been some question as to its accuracy for electron-atomic H scattering. The doubt partially came from the substantially larger electric-dipole polarizability that H has $(0.67 \times 10^{-30} \text{ m}^3)$ when compared to He (0.2) $\times 10^{-30}$ m³). Presumably this would lead to difficulty in handling the effects of the long-range polarization potential of H in calculations. The agreement of our data with Bray and co-workers' predictions is thus especially encouraging. The agreement of our data with those of Khakoo and coworkers is also encouraging, as they used an entirely different normalization scheme than we did.

V. CONCLUSION

Using a modulated crossed-beam technique, we have measured absolute differential cross sections for the excitation of atomic hydrogen's $n=2$ (2²S+2²P) level by electron impact. Our results agree quite well with the recent measurements of Khakoo and co-workers, but have discrepancies with the earlier low-energy data of Williams and co-workers in the backscattering region. Comparison of our results with the CCC calculations of Bray and co-workers shows good agreement, even well into the backscattering region.

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