Supplementary absolute differential cross sections for the excitation of atomic hydrogen's $n=3$ and 4 levels by electron impact

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We have conducted measurements of absolute differential cross sections for the excitation of hydrogen atoms to their $n=3(3S+3P+3D)$ and $4(4S+4P+4D+4F)$ levels. A modulated, crossed-beam method was employed, and the impact energies were 40 and 60 eV. Comparison of our results with those of others is quite favorable.

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I. INTRODUCTION

For more than a century now studies of the hydrogen atom and simple related systems have been essential in advancing our understanding of the small-scale nature of matter. The earliest of these studies culminated with Balmer's discovery of a simple empirical formula to characterize H's visible line spectrum [1]. Accurate description of this spectrum's wavelengths in a way naturally incorporating Rutherford's nuclear model of the atom was the main motivation for the success of Bohr's "old quantum theory" [2,3]. Proper prediction of the same spectrum and its associated transition intensities from first principles led to the widespread acceptance of Schrödinger's formulation of quantum mechanics as a replacement for the old quantum theory [4].

Helium—the next simplest atom—soon thereafter became the focus of quite a lot of attention with Hylleraas's research on its bound states [5]. But it was not until the late 1950s that Pekeris finally developed approximation methods sophisticated enough to treat this system accurately [6]. Extension of these basic approaches to increasingly more complicated bound-state problems gradually resulted in the emergence of quantum chemistry as a mature, quantitative scientific discipline.

Letting one of helium's electrons become unbound gives the next simplest atomic problem—the electron-hydrogen collision system. Since this system is the archetype of all real atomic and molecular collision problems, a proper understanding of it underpins the correct characterization of all more complicated collision processes. Such processes are crucial in engineering and applied science since they underlie all gaseous-discharge and plasma phenomena. But successful treatments of these collision problems have taken a lot longer to develop than their bound-state counterparts. This is largely due to the difficulty in mathematically formulating and solving scattering problems. Such difficulty is particularly acute for the case of ionization during the scattering process, where the asymptotic forms of the wave functions can be especially cumbersome to handle [7,8]. The plethora of approaches invoked over the years has been discussed extensively at many levels of sophistication in all parts of the physics literature (see, e.g., Refs. [9–13]). The older approximation methods produced results of limited accuracy. But in the past decade substantial improvements to these methods have been made, as a result many atomic physicists believe that calculations on the unbound quantum three-body problem can now be done to quite high levels of accuracy. Two particularly promising techniques developed during this time were Bray and Stelbovics' convergent close-coupling (CCC) method[1] and Rescigno and colleagues' exterior complex scaling technique [14,15].

Just like the theoretical treatments, electron-hydrogen collision experiments have been slow to progress. This was due primarily to the difficulty in producing high-intensity, high-purity atomic hydrogen beams. But with the recent introduction of unusually inert materials into the construction of atomic-beam sources, this difficulty has been largely overcome [18]. During the past few decades there have been several elastic collision measurements, including those of Williams and of Shyn and colleagues [19–22]. Differences among the backscattering rates of these two different measurements have led to some amount of controversy.

Excitation measurements have also been performed in the last few decades. For the case of $n=2$ excitation these include Williams' treatment, Williams and Willis's experiments, the measurements of Khakoo and colleagues, the treatment of Doering and Vaughn, and our experiments [23–28]. Agreement between the results of Khakoo and colleagues and ours is excellent, even high into the backscattering region. This is an especially promising development as the two groups used entirely different means of normalization. The agreement of these two sets of experimental results with the CCC method predictions of Bray was also quite good. Unfortunately, the results of Williams and Willis did not agree well with the others in the backscattering region.

The next step is to treat the excitation of higher levels. We recently reported measurements of $n=3$ and 4 level excitation at low energies [29]. Agreement of our results with the few available theoretical predictions was good. Since then we have made additional measurements of the excitation of these levels at higher energies. Due to the small signal strengths and long integration times involved, these are the

[1]Clarification of the correctness of the approximations employed in the CCC approach has recently led to some controversy in the physics literature [16,17].

last measurements of these processes possible with our apparatus. In this article we report the results. Agreement with the only available theoretical results is good. Unfortunately, no other experimental results are available for comparison.

II. EXPERIMENT

Over the past three decades our apparatus and experimental procedures have been discussed extensively in the physics literature (see, e.g., Refs. [30–34]). We therefore provide only a brief treatment here. The entire collision spectrometer is housed in a differentially pumped, dual-chamber vacuum system that is evacuated by turbomolecular pumps backed by two-stage rotary-vane pumps. Three mutually perpendicular sets of Helmholtz coils surround the entire system and attenuate unwanted magnetic fields to less than 20 mG in any direction in the electron-atom interaction region.

Molecular hydrogen is piped into the upper chamber via Teflon and stainless steel tubing. It is dissociated into a mixed H and H_2 beam by a microwave discharge source, and then enters the interaction region located in the lower chamber after passing through a beam chopper and a Teflon double skimmer. The dissociation fraction in the interaction region was consistently $55\pm3\%$ as measured by a quadrupole mass spectrometer.

A gun based on a tungsten filament produces the electrons used for the measurements. They are filtered by a 127° cylindrical energy selector which has electron lenses at both its entrance and exit pupils. They are next accelerated to the required energy, which is calibrated using the 19.34 eV resonance of helium. The beam thus produced has an energy spread of 180 meV full width at half maximum, an angular spread of ±3°, and can be rotated from −90° to 160° with respect to the detector. This detector is fixed to the lower chamber's wall. It contains a 127° energy selector with electron lenses at both its entrance and exit pupils, and ends with a Channeltron electron multiplier.

When measurements are made the scattering angle and impact energy are fixed, while the energy-loss window of the detector is swept repeatedly over the region of interest. All this is controlled by a dedicated microcomputer, which also accumulates the data and performs the signal subtraction required by the beam modulation.

III. DATA ANALYSIS

The modulated nature of the hydrogen beam makes the data analysis a little complicated, as the beam contains both atomic and molecular components. The method of handling this is treated in detail elsewhere [35,36]. The upshot of this treatment is that the absolute excitation differential cross section (DCS) $d\sigma_{\mathrm{H},n=3,4}/d\Omega$ is given as

$$\frac{d\sigma_{\mathrm{H},n=3,4}}{d\Omega} = \frac{S_{\mathrm{H},n=3,4}}{S_{\mathrm{H+H_2,elas}}}\left[\frac{d\sigma_{\mathrm{H,elas}}}{d\Omega} + \left(\frac{1-D}{\sqrt{2}D}\right)\frac{d\sigma_{\mathrm{H_2,elas}}}{d\Omega}\right]. \tag{1}$$

Here $S_{\mathrm{H},n=3,4}$ and $S_{\mathrm{H+H_2,elas}}$ are the $n=3$ or 4 excitation and the elastic signal strengths, respectively. The dissociation fraction D is related to the signal strengths for the microwave-discharge source "on" and "off" as

$$D = 1 - \frac{J^{\mathrm{on}}_{\mathrm{H_2}}}{J^{\mathrm{off}}_{\mathrm{H_2}}}, \tag{2}$$

where $J^{\mathrm{on}}_{\mathrm{H_2}}$ and $J^{\mathrm{off}}_{\mathrm{H_2}}$ are the neutral beam intensities with the microwave-discharge source on and off, respectively. $d\sigma_{\mathrm{H,elas}}/d\Omega$ and $d\sigma_{\mathrm{H_2,elas}}/d\Omega$ are the absolute elastic differential cross sections for atomic and molecular hydrogen, respectively. Before analysis the measured spectra were corrected for the nonconstant transmission efficiency of the detector. Absolute values for the elastic cross sections were those previously previously measured by our group [37,21,22].

Values for the uncertainties involved are provided in Table I. Since the uncertainty sources were independent, they were added in quadrature to determine the net uncertainty. This net uncertainty is also provided in the table.

TABLE I. Uncertainties in the $n=3$ and 4 excitation cross sections.

Source	Contribution (%)
Raw data (statistics)	20
Dissociation fraction	3
Transmission efficiency	4
Elastic DCS uncertainty	15
Total	25

TABLE II. Absolute differential cross sections for excitation of atomic hydrogen's $n=3$ level by electron impact. Units are 10^{-18} cm^2/sr.

	θ (deg)								
E (eV)	12	24	36	48	60	72	84	96	108
40	18.1	4.1	0.69	0.30	0.22	0.15	0.12	0.10	0.10
60	20.6	2.7	0.47	0.15	0.13	0.077	0.055		

IV. DISCUSSION OF RESULTS

The results of our experiments are provided in Tables II and III. Figure 1 shows the $n=3$ excitation cross sections, along with those calculated by Bray with the CCC method [38]. Agreement is very good for both 40 and 60 eV impact.

TABLE III. Absolute differential cross sections for excitation of atomic hydrogen's $n=4$ level by electron impact. Units are 10^{-18} cm^2/sr.

	θ (deg)				
E (eV)	12	24	36	48	60
40	7.5	2.6	0.39	0.16	0.13
60	5.9	1.0	0.23	0.053	0.029

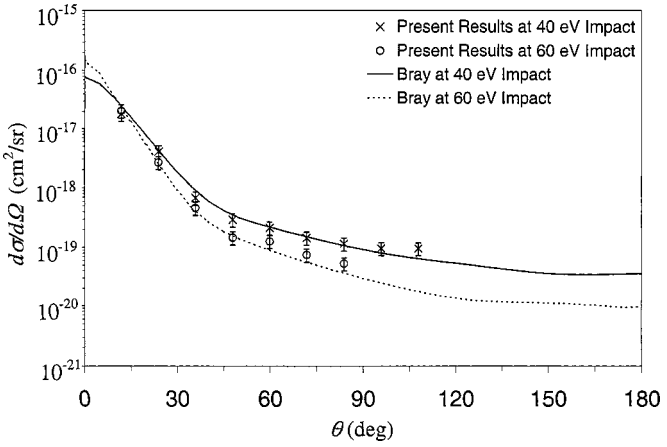

FIG. 1. Absolute differential cross sections for the excitation of atomic hydrogen's $n=3$ level by electron impact.

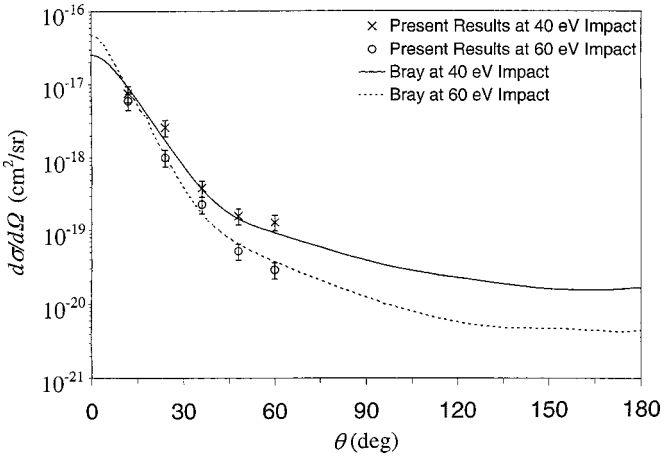

FIG. 2. Absolute differential cross sections for the excitation of atomic hydrogen's $n=4$ level by electron impact.

In the majority of cases, the theoretical predictions fall within the experimental uncertainty. As the scattering angle increases, there is a tendency for the theory to run below experimental error bars, but this tendency is only slight. The 60 eV data show diminished backscattering compared to the 40 eV results for both experiment and theory. This diminished backscattering with increased energy is exactly what one would expect.

Figure 2 provides our $n=4$ excitation cross sections, along with Bray's CCC cross sections [38]. Again, agreement is quite good between the experimental and theoretical results. In most cases, theory and experiment agree to within the experimental uncertainty. In the few where they do not, they are still quite close. At both 40 and 60 eV, our cross sections possess similar character, dropping by two decades in magnitude as the angle 60° is reached. The 60 eV cross sections drop more rapidly than the 40 eV cross sections, which again is to be expected at higher impact energies.

V. CONCLUSION

We have extended our previous $n=3$ and 4 level excitation measurements for atomic hydrogen's electron-impact excitation to higher impact energies. Comparison of our results with the only other available results is favorable. There is a tendency for our results to exceed the CCC results as the scattering angle increases in all cases, but at the angles we were able to probe this tendency is only slight. Unfortunately, the low signal strengths and long integration times for these measurements make higher-angle measurements impossible with our apparatus. We thus welcome additional research into these processes by others.

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[1] J. J. Balmer, Astrophys. J. **5**, 199 (1897).
[2] E. Rutherford, Philos. Mag. **21**, 669 (1911).
[3] N. Bohr, Philos. Mag. **26**, 1 (1913).
[4] E. Schrödinger, Ann. Phys. (Leipzig) **79**, 361 (1926).
[5] E. A. Hylleraas, Z. Phys. **54**, 347 (1929).
[6] C. L. Pekeris, Phys. Rev. **112**, 1649 (1958).
[7] C. W. McCurdy, T. N. Rescigno, and D. Byrum, Phys. Rev. A **56**, 1958 (1997).
[8] C. W. McCurdy and T. N. Rescigno, Phys. Rev. A **56**, R4369 (1997).
[9] T.-Y. Wu and T. Ohmura, *Quantum Theory of Scattering* (Prentice-Hall, Englewood Cliffs, NJ, 1962).
[10] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions*, 3rd ed. (Oxford University Press, London, 1965).
[11] L. I. Schiff, *Quantum Mechanics*, 3rd ed. (McGraw-Hill, New York, 1968).
[12] B. L. Moiseiwitsch and S. J. Smith, Rev. Mod. Phys. **40**, 238 (1968).
[13] J. J. Sakurai, *Modern Quantum Mechanics*, rev. ed. (Benjamin/Cummings, Menlo Park, CA, 1994).
[14] I. Bray and A. T. Stelbovics, Phys. Rev. Lett. **70**, 746 (1993).
[15] T. N. Rescigno, M. Baertschy, W. A. Isaacs, and C. W. McCurdy, Science **286**, 2474 (1999).
[16] V. L. Shablov, V. A. Bilyk, and Yu. V. Popov, Phys. Rev. A **65**, 042719 (2002).
[17] I. Bray and A. T. Stelbovics, Phys. Rev. A **66**, 036701 (2002).
[18] B. P. Paolini and M. A. Khakoo, Rev. Sci. Instrum. **69**, 3132 (1998).
[19] J. F. Williams, J. Phys. B **8**, 1683 (1975).
[20] J. F. Williams, J. Phys. B **8**, 2191 (1975).

[21] T. W. Shyn and S. Y. Cho, Phys. Rev. A **40**, 1315 (1989).
[22] T. W. Shyn and A. Grafe, Phys. Rev. A **46**, 2949 (1992).
[23] K. G. Williams, in *Proceedings of the Sixth Annual Conference of the Physics of Electron-Atom Collisions*, edited by I. Amdur (MIT Press, Cambridge, MA, 1969), p. 731.
[24] J. F. Williams and B. A. Willis, J. Phys. B **8**, 1641 (1975).
[25] J. P Doering and S. O. Vaughn, J. Geophys. Res., [Atmos.] **91**, 3297 (1986).
[26] M. A. Khakoo, M. Larsen, B. Paolini, X. Guo, I. Bray, A. Stelbovics, I. Kanik, and S. Trajmar, Phys. Rev. Lett. **82**, 3980 (1999).
[27] M. A. Khakoo, M. Larsen, B. Paolini, X. Guo, I. Bray, A. Stelbovics, I. Kanik, S. Trajmar, and G. K. James, Phys. Rev. A **61**, 012701 (1999).
[28] A. Grafe, C. J. Sweeney, and T. W. Shyn, Phys. Rev. A **63**, 052715 (2001).
[29] C. J. Sweeney, A. Grafe, and T. W. Shyn, Phys. Rev. A **64**, 032704 (2001).
[30] T. W. Shyn, R. S. Stolarski, and G. R. Carignan, Phys. Rev. A **6**, 1002 (1972).
[31] T. W. Shyn, C. J. Sweeney, A. Grafe, and W. E. Sharp, Phys. Rev. A **50**, 4796 (1994).
[32] C. J. Sweeney and T. W. Shyn, Phys. Rev. A **53**, 1576 (1996).
[33] C. J. Sweeney and T. W. Shyn, Phys. Rev. A **56**, 1384 (1997).
[34] T. W. Shyn and C. J. Sweeney, Phys. Rev. A **62**, 022711 (2001).
[35] J. L. Tate and P. T. Smith, Phys. Rev. **39**, 270 (1932).
[36] W. L. Fite and P. T. Brackman, Phys. Rev. **112**, 1141 (1958).
[37] T. W. Shyn and W. E. Sharp Phys. Rev. A **24**, 1734 (1981).
[38] I. Bray (private communication).