Absolute Differential Cross Sections for the Electron Impact Excitation of the $1^2S \to 2^2S + 2^2P$ Levels of Atomic Hydrogen at 50 and 100 eV

M. A. Khakoo,[1] M. Larsen,[1] B. Paolini,[1] X. Guo,[1] I. Bray,[2] A. Stelbovics,[3] I. Kanik,[4] and S. Trajmar[4]

[1]*Department of Physics, California State University, Fullerton, California 92834*
[2]*Flinders University of South Australia, GPO Box 2100, Adelaide, South Australia 5001, Australia*
[3]*Center for Atomic, Molecular and Surface Physics, Murdoch University, Perth 6150, Australia*
[4]*Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California 91109*
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Absolute experimental differential cross sections for the electron impact excitation of the $1^2S \to 2^2S + 2^2P$ levels of H at 50 and 100 eV incident energy are obtained using an application of the method of mixtures and available accurate He ($n = 2$) experimental electron impact excitation differential cross sections. The determination of the number density composition of the mixed beam is made from energy loss measurements of the mixed beam at 200 eV and 25° scattering angle using accurate H and He theoretical differential cross sections obtained from the distorted-wave Born approximation [D. H. Madison (private communication)] and convergent close coupling [I. Bray and A. Stelbovics, Phys. Rev. A **46**, 6995 (1992); D. V. Fursa and I. Bray, Phys. Rev. A **52**, 1279 (1995)]. [S0031-9007(99)09129-2]

Electron impact excitation of H is considered to be one of the most fundamental problems that needs to be resolved at the present time. Recent electron-photon coincidence measurements show that the convergent close-coupling theory (CCC) of Bray and Stelbovics [1] is excellent for modeling electron-He [2,3] scattering and *possibly* as good for electron-H collisions [4]. However, some doubt exists with regard to the reliability of the CCC method for providing highly accurate scattering amplitudes for electron-H scattering. One reason which supports this doubt is that H has a considerably higher dipole polarizability (0.67×10^{-30} m^3) than He (0.2×10^{-30} m^3) and the long-range polarization potential could represent difficulty for the CCC in the case of H. There are significant discrepancies between the absolute measurements of Williams and Willis [5] (regarded to be the best measurements to date) and the CCC for the electron impact excitation of the $n = 2$ manifold of H at the electron impact energy (E_0) of 54.4 and 100 eV. There are also disagreements between the results of Williams and Willis [5] and other experiments [6] for both elastic and inelastic differential cross sections (DCSs) for H. This is understandable since there are significant experimental difficulties in working with atomic hydrogen beams and with the process of normalizing inelastic scattering intensities to elastic scattering DCSs. Normalization of the inelastic scattering intensities to absolute cross sections could be achieved by utilizing known elastic scattering cross sections. However, beams of H are generated from dissociation of H$_2$, and they always contain an H$_2$ component. This H$_2$ fraction has to be precisely known in order to determine elastic scattering from H alone. Fortunately, this problem is not present when one monitors only inelastically scattered electrons, since one can clearly separate the H and H$_2$ signals

(or for that matter H/H$_2$ and other target gas signals) using electron energy loss spectroscopy. Based on this fact, a better solution to the problem of normalization of DCSs is, therefore, to use an inelastic, rather than elastic, electron scattering calibration standard. Here, we present new and accurate measurements of the absolute DCSs for excitation of the $n = 2$ manifold of H based on measurements of inelastic scattering by a mixed H and He beam [7] and normalization to known He DCSs.

Our apparatus will be briefly discussed here (for more details, see Ref. [8], and references therein). The atomic beam is generated by a single glass capillary needle and made to cross a monoenergetic beam of electrons of incident energy E_0 from an electron gun in a conventional electrostatic electron spectrometer. Scattered electrons can be detected by the spectrometer's electrostatic analyzer as a function of energy loss ΔE and scattering angle θ. The spectrometer is operated with typical currents of ≈ 0.15–0.3 μA and with an energy resolution of about 170–200 meV (FWHM). This apparatus has proven to be extremely stable over long periods (~ 1 yr). The unit is baked at ≈ 110–120 °C to maintain stability which is important for the measurements concerned. It is enclosed in a double Mumetal shield which reduces the Earth's magnetic field below 5 mG. Its data acquisition and control system is computerized (angle settings, multichannel sweep, pressure monitoring, etc.), thus allowing for the continuous (overnight) collection of data. Precision in the location of our experimental θ is within $\pm 1.0°$.

For our gas source, we have used a recently developed, intense, and very stable H source which is detailed in a recent publication [9]. The H source is an extended cavity microwave discharge of 99.999% purity H$_2$, operating at 2450 MHz, with a Teflon tubing to conduct the atoms to the collision region, where the tubing is terminated by

the aforementioned outside-silvered glass capillary needle (0.5–0.7 mm internal diameter). This source delivers H with a dissociation fraction of approximately 82%–85%. This fraction is stable over periods exceeding a month. Approximately a 0.3:0.2 to a 0.5:0.5 mixture (by pressure) of H_2 to He was used and was introduced into the discharge tube through separate precision leak valves. The discharge was usually allowed to stabilize over a day and was checked for stability by monitoring scattering intensities using the electron spectrometer [9]. The pressure fluctuation of the discharge during the entire experiment (for H and He together) did not exceed 2% as measured upstream of the discharge using a temperature stabilized capacitance manometer. At the working pressure (typically 0.5 Torr of H_2 + He), the experimental background pressure increased from a base of 8×10^{-8} Torr to $\approx 1.8 \times 10^{-6}$ Torr.

Our measurements comprise of electron energy loss spectra covering the energy loss range of 9.7 to 12.2 eV (H, $n = 2$ features) and 19.5 to 22.0 eV (He, $n = 2$ features) at $E_0 = 50$ and 100 eV and in the range of 10° to 127°. Given the stability of our system, we took electron energy loss spectra for complete sets of θ from 10° to 127° in 5° intervals or less (in single runs lasting approximately two days). Spectra were remeasured several times to check reproducibility. The ratio $R_{H/He}$ of the scattering intensities for the energy loss feature for the $1^2S \rightarrow 2^2S + 2^2P$ levels of H and the $1^1S \rightarrow 2^1S + 2^3P + 2^1P$ levels of He was determined from each energy loss spectrum. This ratio is related to the respective DCSs:

$$R_{H/He}(E_0, \theta) = \frac{I_H^s(E_0, \theta)}{I_{He}^s(E_0, \theta)}$$
$$= \frac{T_H(E_R)I_0 n_H(V_{eff})DCS_H(E_0, \theta)}{T_{He}(E_R)I_0 n_{He}(V_{eff})DCS_{He}(E_0, \theta)}. \quad (1)$$

Here I_0 is the incident electron current, n_H and n_{He} are the respective number densities for H and He in the collision region, V_{eff} is the "effective" overlap of the electron beam, the gas beam, and the spectrometer analyzer acceptance view cone. $I_H^s(E_0, \theta)$ and $I_{He}^s(E_0, \theta)$ are the scattered electron intensities and $DCS_H(E_0, \theta)$ and $DCS_{He}(E_0, \theta)$ are the DCSs for excitation of the H $1^2S \rightarrow 2^2S + 2^2P$ and the He $1^1S \rightarrow 2^1S + 2^3P + 2^1P$ transitions, respectively. $T_H(E_R)$, $T_{He}(E_R)$ are the electron analyzer detection efficiency for electrons of residual electron energy $E_R(= E_0 - \Delta E)$.

The success of using this experimental setup depends on several factors: (i) The electron energy loss spectra associated with the constituents of the mixture do not interfere with each other. This is truly the case for the features in question here, where the H ($n = 2$) feature is isolated on a smooth H_2 $b\,^3\Sigma_u^+$ continuum and the He ($n = 2$) features lie on a flat H/H_2 ionization continuum. Thus, both $n = 2$ features can be easily separated from background features. (ii) The flow of constituent atoms in the mixture

is stable, which implies a stable H source; i.e., the dissociation fraction of H in the source is constant. This is also the case for the present experiment, where runs of several days could be made with essentially identical flow conditions with about 2% variation. (iii) The transmission of the electron analyzer remains constant during the course of the experiment. This was ensured in this experiment with baking the system and was demonstrated by the fact that the analyzer did not need retuning during the experiment to successfully reproduce ratios.

The procedure utilized here ensures that the term V_{eff} in Eq. (1) is identical for both gases, since the collisional dynamics for both gases are identical. The detection efficiency can be determined as described below. Given that condition (ii) (above) is valid, the ratio in Eq. (1) becomes directly proportional to the DCS ratio of the two gases. Thus, relative DCSs for the unknown gas (H in our case) may be obtained from absolute DCSs of the calibration gas (He in this experiment). Absolute experimental DCSs for He at $E_0 = 50$ eV were obtained from combining the recent relative DCSs of Roder et al. [10] and the absolute measurements of Hall et al. [11]. These two sets of data are in agreement on a relative scale to better than 5%. The Hall et al. DCSs are specified to an absolute accuracy of 12%. DCSs at $E_0 = 100$ eV were taken from the recent measurements of Cartwright et al. [12] and Trajmar et al. [12]. These data are accurate on a relative scale of 5% and an absolute scale of 9%.

In Fig. 1 we show our measured $R_{H/He}$ taken from our electron energy loss spectra. The experimental errors on our $R_{H/He}$ values include statistical and fitting errors due to the determination of the pertinent line intensities (typically 1%–3%, combined) as well as discharge stability (2%–3%) and the contribution due to background gas to be less than 2% for $\theta > 20°$ and maximum of 8% at our smallest θ of 11°. Our measured $R_{H/He}$ overall errors are typically ±5% at most θ except for a markedly increased ±11% value at the smallest θ of 5°. From the He($2^1S + 2^3P + 2^1P$) DCSs of Refs. [10,11], in combination with our measured $R_{H/He}$, we can determine the relative H($2^2S + 2^2P$) DCSs. Combining the ratio and the He DCS errors we obtain typical errors in our relative H DCSs which range from ±7% at most except for the increased ±12% value at the smallest θ of 5° at 100 eV.

In the next stage our relative H ($n = 2$) DCSs were made absolute by measuring repetitive and consecutive spectra for the H and He mixtures at 50, 100, and 200 eV at the scattering angle of 25°. Spectra were repeated with at least five cycles for reproducibility checks. From the 200 eV spectra we determined the number density ratio n_H/n_{He} needed in Eq. (1), using the average of the H DCSs from the distorted wave Born approximation (DWBA) [13] and the CCC [1] at 200 eV and 25°, where agreement between these two very different models for H is better than 2% for $\theta \leq 40°$, thus providing an excellent calibration point. For He, the 200 eV and 25° DCSs were taken from the CCC [2] and have already been found to
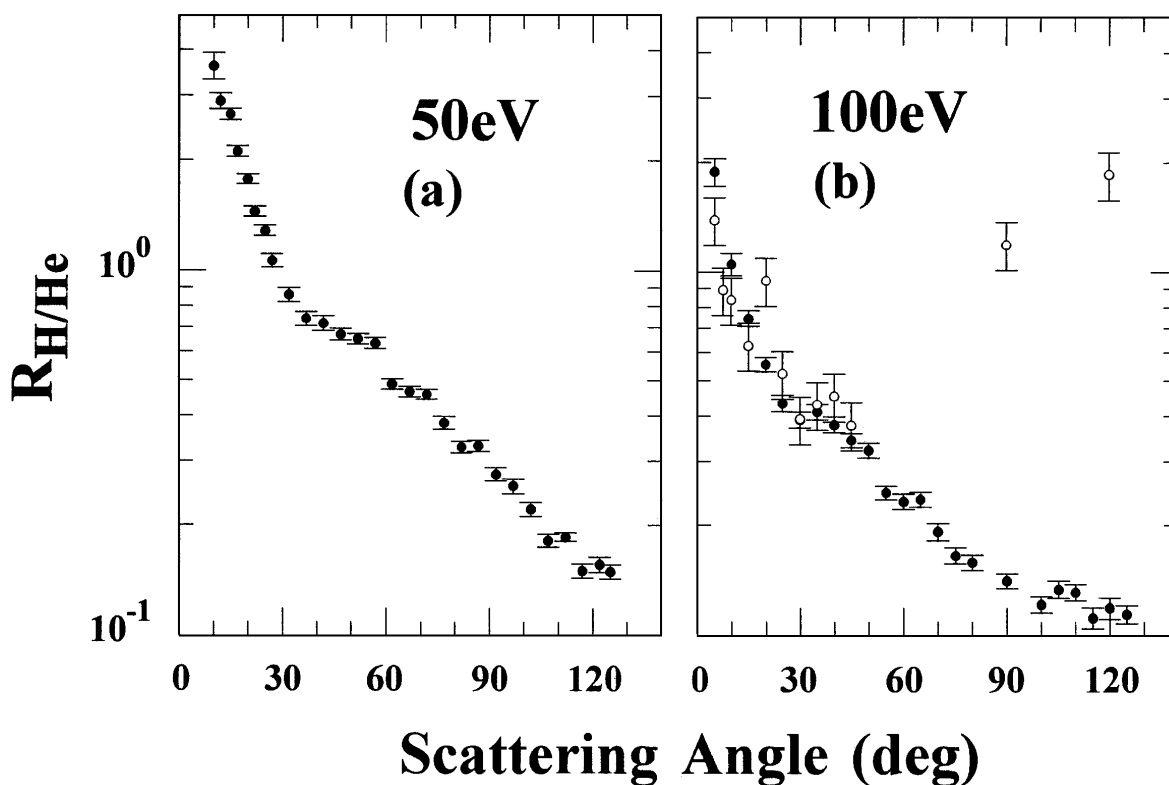
FIG. 1. (●) $R_{H/He}$ values determined from the present experiment. (a) $E_0 = 50$ eV and (b) $E_0 = 100$ eV. (○) are the experimental $R_{H/He}$ values of Doering and Vaughan [7]. See text for discussion.

be in excellent agreement to within 5% at small θ with available experiments. Another factor to make our relative H DCSs absolute is the analyzer detection efficiency for various residual energies $T(E_R)$ in Eq. (1). This was made with He alone, immediately following the mixed beam experiments by measuring elastic and inelastic energy loss spectra at $E_0 = 200, 100, 75, 60, 50,$ and 40 eV at 25° as well as $E_0 = 30$ eV at 90°. The DCS values of experimental Ref. [2] (at 200 eV) and [10,11] (at other E_0) were used, from which we determined the relative detection efficiency $T(E_R)$ as a function of E_R. $T(E_R)$ was found to be a smooth function which changed by 20% for E_R ranging from 9 to 191 eV and was reproducible within 2%. The resulting errors from this normalization procedure are 3% for the transmission function [for the two features in quadrature in Eq. (1)] and additional <2% for the statistical errors which were additionally compounded with errors in the cross sections used for He($2^1S + 2^3P + 2^1P$) and H($n = 2$). The reproducibility (1 standard deviation) in the measurement of the H DCSs at the required E_0 values at a 25° scattering angle was in the region of <±5%, which demonstrates the excellent stability of the apparatus.

Our average DCS uncertainties are typically 15.5%–17.0% at $E_0 = 50$ eV and 13.5%–15.0% at $E_0 = 100$ eV. Comparison of our measurements and the CCC and DWBA results is shown in Fig. 2. At $E_0 = 50$ eV [Fig. 2(a)] reduced chi-squared values (using our experi-

mental errors) between our measurements and the CCC and DWBA are 0.51 and 7.0, respectively. From this we conclude that our DCSs are in excellent agreement with the CCC, but not in good agreement with the DWBA. Agreement in shape between the experiment and both theories is excellent. At $E_0 = 100$ eV [Fig. 2(b)] we compare our results to the DWBA, the CCC, and the absolute experimental DCSs of Williams and Willis [5] and Doering and Vaughan [7]. Agreement with the CCC and the DWBA is excellent with reduced chi-squared values of 0.61 with the CCC and 1.14 for the DWBA, i.e., showing a somewhat better agreement with the CCC. Good agreement is also observed with the other experimental DCSs [5,7], but for $\theta \gtrsim 90°$ we find these experiments deviating sharply from our results and the theoretical values. We draw attention to the important fact that our DCSs are significantly more closely spaced in θ than any previous experimental data.

In conclusion, we have presented absolute DCSs for electron impact excitation of the H($2^2S + 2^2P$) levels from the ground state, using the method of mixtures and calibrating our H ($n = 2$) scattered electron intensities by using an inelastic He DCS standard. These absolute (relative) results have uncertainties which average 15% (7%) and show clearly that the CCC theory is more accurate compared to the DWBA for the excitation of the $n = 2$ levels of H. Our method could not be applied at the most popular E_0 of 54.4 eV since there are no experimental He DCSs here.
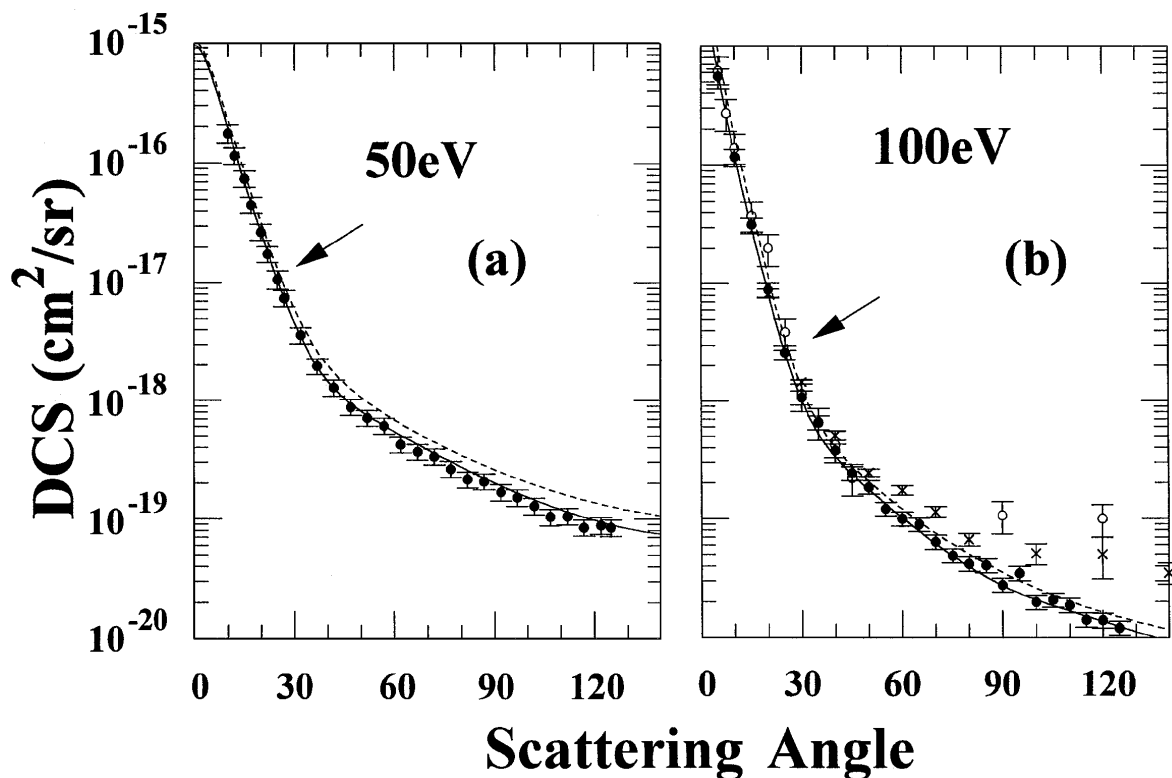
FIG. 2.   Absolute electron impact DCSs for excitation of the $H(1^2S) \rightarrow H(2^2S + 2^2P)$ transition determined from this work ($\bullet$) compared to the present CCC (——) and the DWBA (– – – –) at (a) $E_0 = 50$ eV and (b) $E_0 = 100$ eV. ($\times$) The experimental DCSs of Willams and Willis [5] and ($\bigcirc$) Doering and Vaughan [7]. The normalization points are arrowed.

The above result is also useful in that it opens up new possibilities for measuring accurate inelastic (and possibly elastic) DCSs using the CCC H ($n = 2$) as a calibration standard especially at $E_0 < 21$ eV, i.e., below those of the He excitation threshold. We are thus considering using the above method to measure absolute DCSs for other atomic (e.g., Ne, Ar) and dissociatable molecular targets (e.g., N_2, H_2O, CO with the molecules are entered in the postdischarge region).

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