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Total cross sections for positron scattering from H₂ at low energies

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This paper revisits positron scattering from molecular hydrogen, in an attempt to provide accurate total cross-section data against which theoretical calculations might be benchmarked. The present data were measured over the energy range 0.1–50 eV and, where possible, are compared to results from previous experiments and calculations. Agreement with the earlier data was typically very good at energies above 10 eV but becomes progressively more marginal as we go to lower energies. None of the current theories quantitatively reproduce our measurements over the entire energy range, although at a qualitative level the main features driving the scattering dynamics are apparent.

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I. INTRODUCTION

Molecular hydrogen (H₂) represents, in many respects, the prototypical species for studying positron or electron-scattering phenomena in molecules. It contains nearly all the elements that one must address when dealing with the complexities of multicentered targets in scattering computations and yet, because H₂ has only two bound electrons, its structure can be calculated to relatively high accuracy with present computational resources. This utility therefore enables theoreticians to test the efficacy of, for example, the model polarization potential that they employed in the scattering part of their calculation. Note that H₂ has another advantage in that as it is a homonuclear diatomic it does not possess a permanent dipole moment, which can rather complicate the description of the scattering process. Furthermore, understanding positron and electron low-energy scattering from H₂ is the first step toward a deeper understanding of matter and matter-antimatter chemistry, as these are the simplest chemical reactions involving molecules. New high-precision experimental data thus provide new standards for both future experimental and theoretical investigations.

Experimental total cross-section (TCS) measurements of the positron-H₂ system are now quite dated. With the exception of the results of Sullivan *et al.* [1], where measurements were made over a very narrow energy range (~2 eV), the most recent total cross-section data are those by Zhou *et al.* [2] over a decade ago. Other total cross-section data that we are aware of are those measured by Charlton *et al.* in 1983 [3], over the energy range of 2–20 eV, and those covering the much larger range of 1–500 eV by Hoffman *et al.* [4] in 1982. Note that the measurements of Zhou *et al.* and Hoffman *et al.* both originate from the same group in Detroit, USA. As we shall see later, the total cross sections from these groups [2–4], at positron energies between ~1–10 eV, all exhibit a rather high degree of “scatter” in their respective values. This should not be interpreted as any

implied criticism of their results [2–4] by us. Rather, we simply note that in the last two or so decades our understanding of the techniques needed to produce stable low-energy positron beams has grown significantly and, in addition, technology developments, such as the availability of sources of higher activity, have aided us in realizing these techniques in a practical sense. Nonetheless, investigating the scatter in previous low-energy data [2–4] was one of the rationales behind the present study. A second rationale for this work was to extend the available TCS data to as low an energy as possible with the present spectrometer to assist theoretical colleagues in the development of their models.

The three data sets mentioned above [2–4] have been used as tests for various calculation methods over the years [5–10]. The interactions between positrons and matter involve a positive static potential and a negative polarization potential which, to some extent, can be thought of as counterbalancing one another. Therefore, comparison of theoretical total cross sections with reliable experimental results is a good evaluation for the quality of the polarization potential used. The majority of theoretical data published ranges in energy from ~0.1 eV to the positronium formation threshold at ~9 eV. These computations include a Kohn variational method (KVM) calculation from Armour *et al.* [5], Danby and Tennyson [7] gave results from an application of the *R*-matrix method and Gibson [8] put forward a distributed positron model (DPM) in order to treat short-range correlation effects. The calculation of Reid *et al.* [10] using a complex model potential was the first, and still only, theory to be used over an extended range of energies (1–1000 eV). This work of Reid *et al.* also demonstrated that GAUSSIAN could be used to generate accurate molecular charge densities for molecular hydrogen, implying that the independent atom model was no longer necessary to generate diatomic molecular charge densities from two individual atomic charge densities. Furthermore Sanchez and Lima [11] demonstrated an undeniable effect, by adding more diffuse *f*-type

functions to the molecular wave function, on the integral elastic cross section and the annihilation parameter. The most recent calculations give partial cross sections, either integral elastic [12] or integral inelastic [6,9,11], and are still largely concerned with developing an accurate model of the target, in terms of the description of the wave function [13], or accounting for the nuclear motion (rotation and vibrations) [9], in order to improve their results.

In the following section, a brief description of the experimental apparatus is given (Sec. II A) and an account of our data manipulation techniques (Sec. II B) is outlined. Section III contains the present data for the total cross sections of positron scattering from H₂, over the energy range of 0.1–50 eV, and these results are discussed here. A comparison to the results from theory and other experiments is also given in this section. Finally, conclusions from the present study are drawn in Sec. IV.

II. EXPERIMENTAL DETAILS AND DATA ANALYSIS PROCEDURES

A. Experimental details

The transmission positron spectrometer [14] at the University of Trento was used to measure the total cross-section data presented in this paper. The basis of all linear transmission experiments is the Beer-Lambert law, which is defined as

$$I_1 = I_0 \exp\left(\frac{-(P_1 - P_0)L\sigma}{kT}\right), \quad (1)$$

where σ is the total cross section, I_1 is the positron beam count rate at P_1 , the pressure measured in the scattering cell when the H₂ target gas is admitted, and I_0 is the positron beam count rate at P_0 , the pressure in the scattering cell when the H₂ is diverted to the vacuum chamber. We recall that in this latter configuration, the attenuation in the scattering chamber is negligible (10^{-3} of the attenuation with P_1). Finally, k is Boltzmann's constant, T is the temperature of the target gas (in K), and L is the length of the scattering cell (22.1 ± 0.1 mm).

The details of this spectrometer have been described elsewhere [14], as have the data analysis techniques (see [15,16], for example). Therefore, only those points required for completeness are given here. This apparatus utilizes a ²²Na radioactive source (~ 2.7 mCi) in conjunction with a nickel moderator. This moderator only recently replaced the previously employed tungsten moderator, in order to give an improved energy resolution. The current 2- μ m-thick nickel (Ni) moderator typically gives an energy resolution of 0.1–0.15 eV full width half maximum (FWHM), which is a significant improvement from that obtained with the tungsten moderator that was typically 0.3 eV (FWHM). However, it was found that the performance of the Ni moderator deteriorated much more rapidly over time so that reconditioning was needed every 7–14 days, in order to maintain sufficient (and stable) positron intensity for accurate cross-section measurements. Full details of the moderator stability and conditioning technique will be given elsewhere [17]. The

positron beam produced from this source is guided into the interaction region by charged particle optics, with an axial magnetic field usually of ~ 11 G being used to confine the positrons and to achieve an additional tunable focusing onto the scattering cell region.

As a standard practice in the Trento laboratory, the performance of the spectrometer is verified periodically with a preliminary TCS measurement for positron scattering from N₂. This practice has been established as there are accurate sets of data already available in the literature, e.g., [4]. N₂ cross sections have also been measured in this laboratory since 2006 with great consistency [18] and these routine measurements provide a benchmark against which the performance of the apparatus, in terms of the measured absolute cross section, can be tested.

A further experimental consideration that needs to be taken into account for the physical application of Eq. (1) is to ensure that the TCS measured is independent of the pressures used. To achieve this, i.e., to keep double-scattering events below a 1% limit, the linearity of the logarithm of the positron count rate against pressure is established. In practice, this means that we operate at pressures so that the ratio I_1/I_0 has been kept at values between 0.6 and 0.8.

B. Data analysis procedures

In the application of Eq. (1) to determine the total cross sections, several key precautions need to be included in the data analysis procedures. The entrance and exit apertures of the scattering cell to allow the transmission of the positron beam are both 1.5 mm in diameter. End effects, due to the target gas leaking from the scattering cell through these apertures, causing the effective length to be longer than the geometric length, were considered in this study. In addition, the effect of the pressure being a little lower immediately inside the cell near the apertures, effectively decreasing the length of the scattering cell, was also considered. It has been demonstrated [19] that for entrance and exit apertures of the same diameter, these two effects almost cancel, so that the uncertainty in the scattering cell length due to these factors is most likely to be less than 1% of the geometrical length. A correction to the scattering cell length has also been applied to account for the gyration of the positrons in the focusing magnetic field within the scattering cell. This correction increases the effective length by less than 5% over the incident energy range up to 35 eV, where the magnetic field was 11 G. At the three highest energies of our study, the magnetic field was decreased to ~ 6 G and, therefore, a correction of just 2.5% was applied in those cases. Note that the gyration of the positrons can potentially increase the correction that should be applied to account for the system angular resolution. No correction for angular resolution has been possible here as no experimental differential cross sections are known to us at this time. We can only state that if the angular resolution correction were applied to the current data then the corrected TCSs would be somewhat larger than those reported in this paper.

A thermal transpiration correction to the measured pressures has also been made before they are used in Eq. (1). The

pressures within the scattering cell were measured by a MKS Baratron capacitance manometer (Model 627BX, 1 mbar full scale), which operates at 45 °C. As the scattering chamber is at room temperature (24 ± 2 °C), a thermal transpiration correction needs to be applied to the pressure reading in order to determine the actual pressure within the scattering cell. This correction has been made according to the model of Takaishi and Sensui [20], with a H₂ molecular diameter of 2.9 Å [21] being used for this calculation. The corrections in this case were always in the order of 2%.

Finally, an energy scale calibration is made to the data. The energy zero position has been periodically checked through the use of a retarding potential analysis of the positron beam [22]. We have found that the energy zero position is remarkably stable in our spectrometer, being reproducible to within less than 0.05 eV on the time scale of months. We note, however, that the energy zero is different for the Ni versus the tungsten moderator, so that a new calibration was necessary in this case. Taking into account both the 0.05 eV calibration uncertainty and the energy width of our positron beam, we append a probable error of 0.2 eV to the present energy zero calibration.

III. RESULTS AND DISCUSSION

In Table I, we present the energy dependence of the total cross sections for positron scattering from H₂. These values are also plotted in Figs. 1 and 2, where they are compared with previous experimental measurements (Fig. 1) and a selection of the theoretical total cross-section data (Fig. 2) available within the literature. Considering Fig. 1 in more detail, we see that the present TCS data qualitatively exhibit two distinct forms of behavior depending on the incident positron energy. For energies between 0.1 and about 9 eV, the magnitude of the TCS decreases largely monotonically in value with increasing energy. Notwithstanding this general observation, there is a suspicion for some very small structure in our TCS data associated with the opening of the lower-lying vibrational sublevels of the ground electronic state ($v'=0 \rightarrow 1, 2, 3$). The threshold energy of the first vibrational level in H₂ is at about 0.52 eV and due to the harmonic nature of this system, each higher level, to first order, opens at energies that are integer multiples of that value. However, this effect on the TCS is rather small here and so, in the absence of theoretical guidance, we cannot be more definitive in our discussion at this time. As H₂ is a homonuclear diatomic, it has no permanent dipole moment. Under these circumstances, the observed general behavior of the TCS in this first energy region is likely to be due to its small ($6.74 \text{ a.u.} \equiv \sim 1 \times 10^{-24} \text{ cm}^3$ [23]) but not insignificant dipole polarizability. For energies between 9 and 50 eV, we initially see that the TCS increases sharply in value until it reaches a secondary maximum at around 25 eV, before once again falling in magnitude as the energy increases further. This initial sharp rise in the value of the TCS, in this second energy region, can be associated with the opening of the positronium formation channel (see below) followed by the opening of the higher-lying singlet electronic states in H₂ (e.g., the B $^1\Sigma_u^+$ and C $^1\Pi_u$ states) and finally by the opening of the direct ionization channel.

TABLE I. The present total-cross section data (10^{-16} cm^2) for positron scattering from H₂. Statistical errors are typically on the order of 3% and 4% in the entire energy range.

Energy (eV)	Total cross section (10^{-16} cm^2)	Energy (eV)	Total cross section (10^{-16} cm^2)
0.10	7.32	8.95	1.43
0.15	6.29	9.45	1.42
0.20	6.21	9.95	1.61
0.25	5.22	10.45	1.87
0.35	4.14	10.95	2.19
0.45	3.71	11.95	2.44
0.55	3.50	12.95	2.87
0.65	3.30	13.95	3.43
0.75	2.93	14.95	3.89
0.85	2.79	15.95	3.99
0.95	2.52	16.95	4.44
1.05	2.17	17.95	4.54
1.25	2.06	18.95	4.54
1.45	1.96	19.95	4.93
1.85	1.76	20.95	5.29
1.95	1.87	21.95	4.97
2.05	1.64	22.95	4.82
2.25	1.61	23.95	5.03
2.45	1.55	24.95	5.17
2.95	1.46	25.95	5.16
3.95	1.48	26.85	4.66
4.95	1.38	29.95	5.02
5.95	1.36	31.95	4.59
6.95	1.25	34.85	4.62
7.95	1.33	39.95	4.45
8.25	1.26	44.95	3.78
8.65	1.31	49.95	4.14

The cross-section values for positron scattering from H₂ are quite low compared to the TCSs of all other molecules previously measured with the present spectrometer [16,24–27]. We believe this is due to one or more of the following reasons. First, H₂ has no permanent dipole moment, whereas many of our previous targets did. Second, the dipole polarizability of H₂ is a lot smaller than those for any of those earlier targets [16,24–27] and, finally, from a semi-classical point of view, molecular hydrogen is by far the smallest species we have studied to date.

Total cross-section measurements can be used to determine the positronium formation threshold energy (E_{Ps}). Using the technique outlined in previous papers [16,24–27], our best estimate for this energy is 8.4 ± 0.2 eV. Furthermore, as a general rule [28] the positronium formation threshold for a given species is given by

$$E_{Ps} = V_i - 6.8 \text{ (eV)}. \quad (2)$$

Using an ionization potential (V_i) of 15.4 eV [29] for H₂, Eq. (2) gives $E_{Ps} = 8.6$ eV. This value is consistent with the pos-

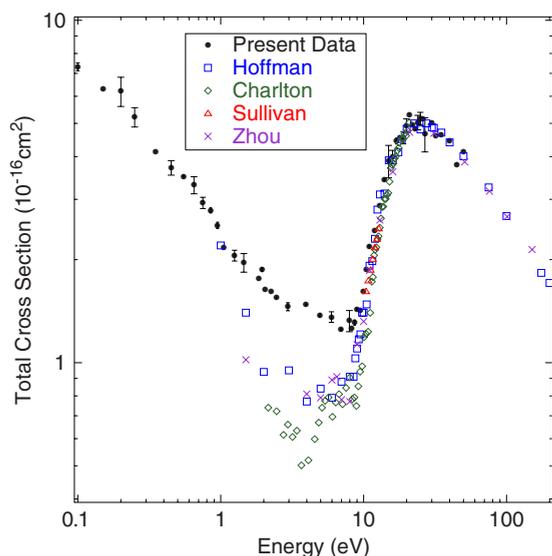


FIG. 1. (Color online) The present total cross-section data (\bullet) for positron scattering from H_2 in comparison with previous experimental measurements: (\square) Hoffman *et al.* [4], (\times) Zhou *et al.* (1997) [2], (\diamond) Charlton *et al.* (1983) [3], and (\triangle) Sullivan *et al.* (2001) [1]. Note that only a fraction of the points from Sullivan *et al.* have been included for clarity. Errors bars are marked at each point; where the error bars are not visible, they are smaller than the symbol size.

ironium threshold energy we obtain from the TCS data of the present study.

In Fig. 1 we also compare the present TCSs with those from previous experimental measurements on H_2 . As can be seen from this figure, the current scattering data include the

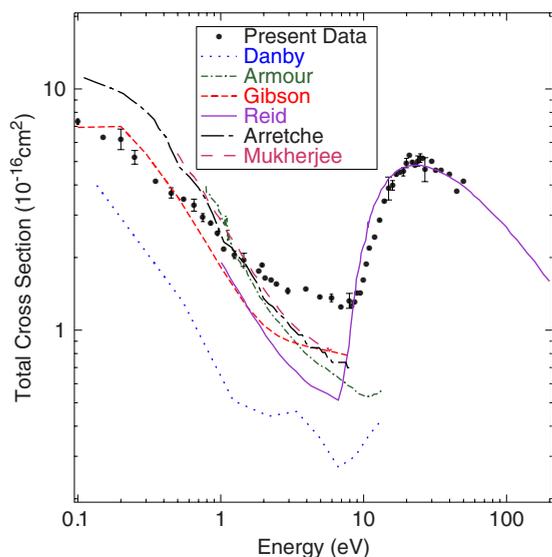


FIG. 2. (Color online) The present total cross-section data (\bullet) for positron scattering from H_2 in comparison with results from previous theoretical models: (\cdots) Danby and Tennyson [7], ($- \cdot -$) Armour *et al.* [5], ($- - -$) Gibson [8], ($- - -$) Reid *et al.* [10], ($- \cdot -$) Arretche *et al.* [6], and ($- - -$) Mukherjee and Sarkar [9]. Errors bars are marked at each point; where the error bars are not visible, they are smaller than the symbol size.

first very low-energy measurements of positron scattering from H_2 , i.e., for energies below 1 eV. The data measured at Detroit by Hoffman *et al.* [4] together with those at University College by Charlton *et al.* [3] were among the first measurements of the positron scattering era, where measurement difficulties were of increasing importance when going toward low energies. Hence, there is no surprise in finding that the lowest-energy cross sections measured were to ~ 1 eV. We reiterate that we believe that the large scatter in their data [3,4] below 10 eV is due to these difficulties. Above 10 eV, the agreement between the present data and all the previous experimental measurements is rather good, indicating, at least experimentally, that this kinematic region is well characterized. In the energy range between ~ 1 and 10 eV, a large inconsistency is present between our TCS values and the other experimental results (see Fig. 1). This discrepancy between the present data and the earlier ones may be explained, at least in part, in terms of the better experimental reliability of the measurements performed with the current apparatus and because of the need for only relatively small corrections to the measured data due to thermal transpiration and the effective scattering length. Indeed, it looks very probable that the angular resolution correction is smaller in the present configuration, so that a larger factor for the observed discrepancy in the respective TCSs is the superior angular resolution in the present apparatus. Note that support for this proposition can be found in electron-tetrahydrofuran TCS scattering experiments, where measurements from Mozejko *et al.* [30], performed on an electron spectrometer with a higher angular resolution than the present apparatus found TCS that were somewhat larger in magnitude than the corresponding data of Zecca *et al.* [27] measured with the current spectrometer. A very advanced positron spectrometer has recently been commissioned at the Australian National University (ANU) [31]. From comparison of preliminary data in water and formic acid [32], we believe that the angular resolution of this apparatus and the present one are possibly on the same order of magnitude. Therefore, a comparison between measurements of positron scattering from H_2 from ANU and Trento could be used to confirm the angular resolution as the dominant source of the observed discrepancy between the present results and previous data in the 1–10 eV energy region.

Figure 2 shows our TCSs compared with results from various theoretical models, which aim to describe the physical mechanism for positron scattering from H_2 . These include the R -matrix calculation of Danby and Tennyson [7], the KVM calculation of Armour *et al.* [5], Gibson's [8] DPM, and Reid *et al.*'s [10] calculation employing GAUSSIAN to generate the molecular electronic charge density and using a complex model potential to describe the scattering. Integral elastic cross sections from Arretche *et al.* [6], using the Schwinger multichannel method, and those of Mukherjee and Sarkar [9], which includes the rotational and vibrational motion of the target nuclei, are also included in this figure. It is quite clear from Fig. 2 that while none of the present theories are able to quantitatively reproduce our TCS, over the entire energy range of the measurements, most at least are qualitatively agreeing with the trend in the measured data at energies below the positronium formation threshold. Clearly though some more theoretical work is still needed on

this most fundamental of positron-molecule scattering systems, before a good level of quantitative accord between calculation and measurement might be achieved. In this respect, we note the recent theoretical work from Cooper *et al.* [13], who demonstrated the importance of using an accurate target wave function in variational calculations for positron-H₂ scattering. Notwithstanding this, we highlight that the computation of Reid *et al.* [10] is doing a very good job in reproducing the TCS data above 10 eV.

IV. CONCLUSIONS

We have reported experimental TCS values of positron scattering from H₂ in the energy range between 0.1 and 50 eV. This paper includes measurements at energies below about 1 eV, which allows comparison with theoretical results already available in the literature in that energy range. The discrepancy between the present data and previous experimental measurements, at lower energies, suggests that the

present apparatus has a superior angular resolution compared to those used to obtain the earlier experimental data. Agreement between all the measured data above 10 eV is, however, very good. A comparison between the present experimental data and available theoretical models indicates that a further improvement in theories trying to understand the positron collision dynamics is required.

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