Positronium formation cross sections in He and H_2 at intermediate energies

L. M. Diana, P. G. Coleman,* D. L. Brooks, P. K. Pendleton, and D. M. Norman
Center for Positron Studies, Department of Physics, The University of Texas at Arlington, Arlington, Texas 76019
(Received 24 April 1986)

Measurements of cross sections for positronium formation in collisions of positrons of energies up to 251.4 eV with helium atoms and hydrogen molecules are reported. In both gases the cross sections are more than twice as large as the only other experimental values published to date. At low energies the measured values agree quite well with available calculations.

I. INTRODUCTION

Recent measurements of the positronium (Ps) formation cross section Q_{Ps} for atoms and molecules[1-3] have demonstrated that the Ps formation channel is very important, even dominant, at energies a few tens of eV above threshold (E_{Ps}). Knowledge of Q_{Ps}, up to energies where it becomes a negligible contributor to the total cross section Q_{tot}, is not only of interest for the sake of completeness, but also enables a more comprehensive interpretation of astrophysical data on the characteristics of annihilation radiation from the direction of the galactic center[4,5] and laboratory simulations of these observations.[6] Complete knowledge of Q_{Ps} is also necessary in the comparison of positron and electron total cross sections, which is of special interest in those gases, including hydrogen, for which $(Q_{tot})_{e^+}$ exceeds $(Q_{tot})_{e^-}$ over certain energy ranges.

A further element of interest in Q_{Ps} at intermediate positron energies lies in the investigation of whether the large differences, in magnitude and energy dependence, between the experimental results of Refs. 1 and 2 at energies below 75 eV persist to higher energies.

The experimental method employed for the current measurements is in principle the same as that of Fornari *et al.*,[1] i.e., we measure the fraction f of positrons in the incident beam which is not transmitted through a gas cell because of Ps formation, together with the fraction F which are scattered via any channel; then in the thin-target limit $Q_{Ps}=fQ_{tot}/F$.

Measurements at intermediate energies are more difficult than at low energies because (a) it becomes more difficult to discriminate against positrons elastically scattered through small forward angles with a consequent underestimation of F, and (b) a high axial magnetic field which is required to insure that no scattered positron is lost from the beam increases the background count rate. (a) is combated by the modification of experimental technique described in Sec. II, and (b) is not severe enough to have deleterious effects on the measurements.

On the positive side, the fraction of positrons backscattered following collisions is very small and positron losses through the reflector element (the moderator) near the source are negligible. Finally, the predominantly small-angle forward scattering of the positrons means that the probability of double or multiple scattering, which can increase the number of Ps-formation collisions and hence the measured Q_{Ps}, is very small and can be estimated straightforwardly.

II. EXPERIMENTAL TECHNIQUE

A. Apparatus

A schematic diagram of the apparatus, illustrating the modifications to that employed in the low-energy measurements of Ref. 1, is shown in Fig. 1. The basic features remain unchanged, i.e., slow positrons from an annealed tungsten mesh bombarded by β positrons are accelerated to the desired mean energy and traverse a 2.3-m-long solenoid-surrounded flight tube to a channel-electron-multiplier (CEM) detector. However, major changes have been made at the source and detector ends of the system.

In Ref. 1 the total attenuation of the beam F was measured by recording time-of-flight (TOF) spectra for the positrons with the flight tube evacuated and with gas admitted, following the procedure of earlier experiments.[7]

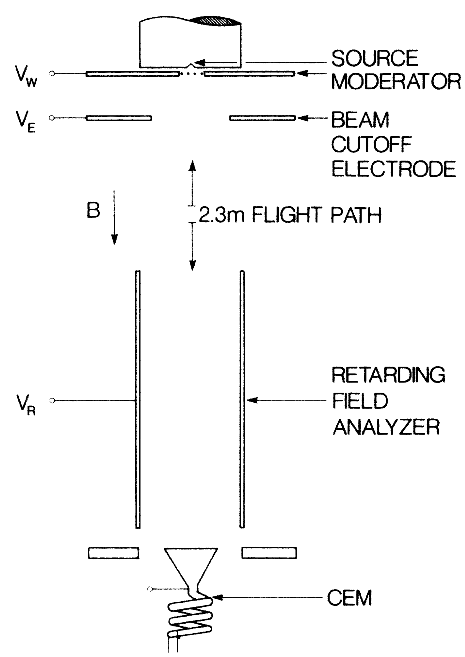

FIG. 1. Schematic diagram of experimental apparatus.

This is an excellent technique at positron energies below about 30 eV, but then, as the mean elastic scattering angle becomes small, the resolution of elastically scattered and unscattered positrons requires careful analysis of the shapes of the TOF peaks, following the practice of Coleman *et al.*[8] This method of analysis is satisfactory for positron energies up to 76 eV (the highest incident energy of the earlier $Q_{\rm Ps}$ measurements), but above 76 eV in an extended gas-cell system it has been shown that complete discrimination against scattered positrons may not be achieved, and F is underestimated.[9]

To overcome this problem retarding-field analysis was employed and particle timing abandoned. The retarding potential V_R was applied to a cylindrical element 50 mm long and 19 mm in diameter held on axis directly in front of the CEM. The operation of this electrode is described in Sec. II B below.

To obtain timing pulses from the source end of the flight tube in earlier experiments, the ^{22}Na deposit was covered by a 0.25-mm-thick disk of plastic scintillator. Over 50% of the β positrons were absorbed by the plastic; in addition, it was apparent from the TOF spectra that approximately 50% of the slow positrons entering the flight tube did not have associated timing signals. Furthermore, the nuclide activity was limited to about 150 μCi because of the high pulse rate from the scintillator ($\approx 2\times 10^6$ s^{-1}) and its adverse effects both on photomultiplier tube performance and on the time spectra.[10] Removal of the need for timing and, hence, for the scintillator cover, improved the slow positron yield from 150 to 1800 s^{-1}. This latter figure was achieved with about 460 μCi of ^{22}NaCl deposited on an insulin-wetting agent[11] coating a 60° half-angle cone in the surface of a brass rod. Backscattering of β positrons from the substrate was enhanced by electroplating 40 μm of gold on the surface of the cone prior to source deposition.[12] The annealed tungsten mesh moderator is held 1 mm in front of the source behind a 5-mm-diameter hole, which determines the beam diameter, in the center of an otherwise solid disk. The accelerating potential $V_{\rm W}$ is applied to the moderator so that the mean positron energy $\bar{E}=(V_{\rm W}+1.3)\pm 0.6$ eV. The slow positron generation efficiency has been remarkably stable over periods when the moderator has remained undisturbed in the apparatus, decreasing by less than 1.5% per month.

The moderator mesh serves also to turn backscattered positrons back to the CEM. A ring electrode was installed at the source end of the apparatus to cut off the positron beam, i.e., to prevent slow positrons from entering the flight tube by the application of a potential V_E as discussed in Sec. II B below.

B. Measurement of beam attenuation

A representative integral spectrum, or profile, of the number of positrons reaching the CEM with the flight tube both evacuated and filled with low-density gas, as a function of the retarding potential V_R, is shown in Fig. 2. The gas pressures were controlled in nearly all of the data runs to keep the total attenuation F to $\leq 20\%$, thereby minimizing multiple scattering effects. However, the few runs in which F exceeded 20% reproduced results made at the same energies with lower F, a fact which may indicate that the double scattering correction employed is sufficiently detailed for these measurements. An interesting feature of profiles such as that of Fig. 2 is the information they provide on the energy losses of the scattered positrons, evidently a maximum of about 45 eV in the run depicted.

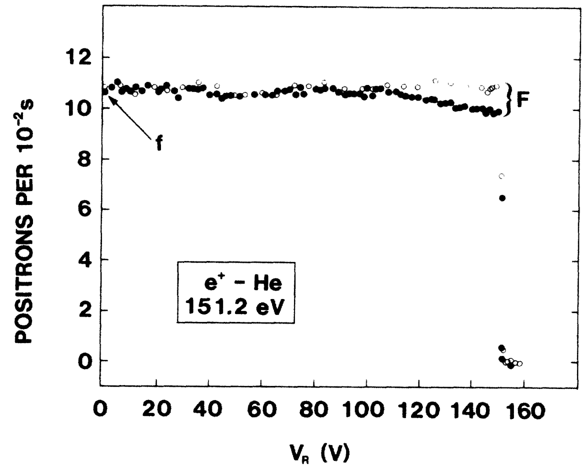

FIG. 2. Integral profile of positrons ($\bar{E}=151.2$ eV) detected by the CEM vs retarding potential V_R. Background counts ($\sim$27 per 10^{-2} s) have been subtracted.

Clearly, one can determine the scattered fractions f and F by measuring the decrease in the beam intensity at $V_R=0$ and at $V_R\approx V_W$, respectively. Two obstacles to the extraction of reliable f and F values, however, are the uncertainties in (a) the background levels, especially at $V_R=0$, and (b) the location of the profile "edge" when determining F. (a) is solved by using the cutoff potential V_E. To measure signal + background count rates, V_E is set so that all the positrons are just allowed to pass through the electrode into the flight tube, and to measure the background count rate V_E is increased so that the positrons are just all stopped (e.g., $V_E=158$ and 190 V for 141-eV positrons). The two values of V_E are kept as close to each other as possible so that the background count rate, evidently due to energetic secondary electrons, is essentially unchanged between the two measurements. Background values obtained by this method were consistent with, and more reliable than, those obtained by extrapolation from the high V_R region.

Locating the correct edge of the profile (in the vicinity of $V_{\rm W}$) is important, as this determines how well one achieves discrimination against small-angle elastically scattered positrons. Initially the profiles were differentiated in the region of the edge, or step, to yield positron-energy spectra whose shapes were examined.[13] Low-energy tails containing elastically scattered positrons, similar to those seen in the earlier TOF spectra, were observed, and the true value of F was determined following the same procedure as that of Ref. 8. This method of analysis is lengthy and requires a large number of data points in the vicinity of the profile step. An alternative technique was adopted for some of the measurements reported herein; the total attenuation F_{41} was measured for 41.3-eV positrons, using the same gas density as for the high-energy measurements. With V_R set at 39.3 V essentially all of the scattered positrons are turned away from the CEM, and a reliable value for F_{41} is obtained. Then, assuming knowledge of Q_{tot} values at 41.3 eV and at the energy of interest E (Q_{41} and Q_E, respectively) F at E, F_E, is calculated from

$$F_E = 1 - \exp\{[\ln(1-F_{41})]Q_E/Q_{41}\} \ . \tag{1}$$

Uncertainties in Q_E and Q_{41} are about 5%,[9] and the additional uncertainty in F_E resulting from the use of this procedure is therefore less than 5%. However, it was found that F measured directly from the profile at $V_R = V_W - 0.7$ V agreed to within statistical uncertainties with the values obtained using the 41.3-eV technique, indicating that discrimination against scattered positrons was excellent. This is discussed further in Sec. II C.

Full V_R profiles of the type shown in Fig. 2 were, therefore, unnecessary, and subsequently measurements of total and background counts were made (using V_E) only at $V_R = 0$ and $V_W - 0.7$ V.

C. Possible sources of systematic error

In this section we list and discuss a number of possible sources of systematic error in the measurements, all of which have been considered and for which appropriate corrections have been applied, if necessary, to the Q_{Ps} values prior to their tabulation below.

1. *CEM efficiency*

As reported in Ref. 1, it was noted during the course of those measurements that the positron (and electron) detection efficiency ϵ of the CEM appeared to be slightly lower when gas was present in the flight tube. Similar reductions in ϵ, of the order of 1%, were seen for both positrons and electrons and for different gases at the same density; an apparent independence of projectile energy was also noted. All these observations were made by measuring the fractional decrease f in the transmitted beam intensity below E_{Ps}, where f should be zero. These conclusions about ϵ have been confirmed in the current set of measurements by the consistent observation of a reduction in the background counts, both at $V_R = 0$ and $V_W - 0.7$ V, typically by about 0.5%, on the admission of helium or hydrogen into the gas cell. (We recall here that the background counts are evidently almost wholly attributable to energetic secondary electrons.) The positron signal count rates in the "gas" runs were accordingly increased by a factor evaluated for each run (i.e., the ratio of background levels, ≈ 1.005) prior to evaluating f and F. Such corrections have negligible effect on the final Q_{Ps} in the large peak at < 75 eV in both gases but becomes increasingly important at higher energies where 0.5% is no longer negligible compared with f. One is encouraged to find that at the highest energies studied f tends to zero after the correction is made rather than, for example, -0.5%, as would be possible if the fall in measured background levels were not due to the change in ϵ.

2. *Positron backscattering*

The possibility that a positron scattered by a gas atom or molecule into the backward hemisphere travels back to the source end of the flight tube and is absorbed there, and not counted by the CEM, has to be considered. The problem is combated by reflector electrodes; in the current apparatus both the moderator mesh and the beam-cutoff electrode play this role. As stated above, V_E is set so that incident positrons from the source, traveling along paths essentially parallel to the tube axis, all just pass through the electrode. Therefore, any backscattered positron would have to have suffered almost 180° scattering in order not to be reflected. The preceding statement is true, also, for reflection by the moderator mesh. Because the mean energy eV_0 of the incident positrons is ≈ 1.3 eV above that of the slowest which have to overcome the potential in the center of the reflector electrode, an estimate of the maximum scattering angle θ_{max} suffered by positrons which are still reflected is obtained simply from

$$\theta_{max} = \pi - \arccos(1 - 1.3V/V_0)^{1/2} \tag{2}$$

so that, for example, $\theta_{max} \approx 175°$ for $V_0 = 150$ V. At intermediate energies the differential cross section for elastic scattering at angles greater than 150° in helium and hydrogen is so small[14] that it can be asserted, with confidence, that the contributions of backscattering losses to f are negligible.

3. *Multiple scattering*

Ps formation in a second or subsequent interaction increases f and, therefore, the measured Q_{Ps} values. This is especially troublesome if the scattering angle is close to 90°, when the path length through the gas is increased considerably. Corrections to the lower-energy cross sections, via Monte Carlo simulations using estimated angular distributions, were discussed in Ref. 1 and were typically about 7%. At intermediate energies, however, the analysis can be simplified if one makes the reasonable assumption that the scattering angles for both elastic and inelastic scattering $\sim 0°$. Let us first assume that more than two scattering events experienced by a positron passing through the gas cell is an unlikely occurrence, especially as gas pressures were typically kept low to effect $F = 0.20$ or less. We are then concerned with the total probability p for Ps formation following elastic scattering or ionization or excitation,

$$p = p_e + p_{ex+i} \ , \tag{3}$$

where p_e is the probability for Ps formation after elastic scattering and p_{ex+i} is the probability for the scattering sequences excitation followed by positronium formation and ionization followed by positronium formation. Straightforward algebraic manipulations lead to

$$p_e = Q_e Q_{Ps}[1-(1+nLQ_{tot})\exp(-nLQ_{tot})]/Q_{tot}^2 \tag{4}$$

and

$$p_{ex+i}=Q_{ex+i}Q^*_{Ps}\{Q_{tot}[1-\exp(-nLQ^*_{tot})]-Q^*_{tot}[1-\exp(-nLQ_{tot})]\}\times[Q_{tot}Q^*_{tot}(Q_{tot}-Q^*_{tot})]^{-1}\ . \quad (5)$$

Q_e is Q_{tot} at E_{Ps} obtained by smooth extrapolation of the Q_{tot} values just below threshold. Q^*_{tot} and Q^*_{Ps} are the values at the positron energy remaining after the initial inelastic scattering event (taken to be ionization in the application of this correction). Q_{ex+i} is the sum of the excitation and ionization cross sections, and nL is the mean gas-number-density—path-length product. To arrive at these expressions a linearly decreasing gas density along the flight tube was assumed, but assumption of constant density does not affect the result. As an example, for 176.5-eV positrons incident on He, $F=0.144=1-\exp(-nLQ_{tot})$, uncorrected $Q_{Ps}=0.065\pi a_0^2$, and $Q^*_{Ps}=0.16\pi a_0^2$, and we obtain from values in the literature[9,15] $Q_e=0.22\pi a_0^2$, $Q_{tot}=0.87\pi a_0^2$, and $Q^*_{tot}=0.96\pi a_0^2$. Subtraction from Q_{tot} yields $Q_{ex+i}=0.6\pi a_0^2$. The result, $p=0.00161$, represents a correction to Q_{Ps} of 14.9%. Corrections such as this have been applied to the Q_{Ps} values of Table I.

TABLE I. Total positronium formation cross sections in He and H_2 (statistical uncertainties in parentheses).

Positron energy (eV)	Q_{Ps} (πa_0^2) He	H_2
16.2		3.23(0.10)[a]
46.0	0.57(0.04)	
51.2	0.52(0.03)[a]	
61.0	0.414(0.027)[a]	
76.1		0.43(0.03)[a]
76.3	0.28(0.07)	
86.4		0.30(0.09)
88.6	0.15(0.03)	
101.3	0.200(0.025)[a]	0.23(0.07)
113.7		0.26(0.10)
113.8	0.23(0.04)	
125.3		0.18(0.07)
126.4	0.19(0.04)	
140.9	0.119(0.027)[a]	
141.3		0.27(0.07)
151.2	0.16(0.04)[a]	
158.7		0.20(0.07)
161.4	0.13(0.04)	
171.3		0.03(0.05)
176.5	0.06(0.04)	
191.3	0.04(0.03)[a]	
201.2		0.12(0.06)
201.3	0.041(0.023)[a]	
213.3	0.12(0.04)	
213.8	0.125(0.021)	
225.3	0.116(0.016)[a]	
226.3		0.07(0.08)
226.4	0.07(0.03)[a]	
236.4	0.119(0.015)[a]	
239.1	0.06(0.04)	
251.4	0.017(0.019)[a]	

[a]Weighted average of two or more measurements.

4. Ps dissociation

Positronium atoms formed in the gas cell with appreciable kinetic energy could conceivably be dissociated in an interaction with a gas atom or molecule, the resulting positron being detected by the CEM. This would reduce f and lead to an underestimation of Q_{Ps}. It is difficult to treat this problem at present in any more than a qualitative fashion; however, it seems likely that its effect is very small. Let us assume that the total cross section for the scattering of the energetic neutral Ps atoms is approximately $1\pi a_0^2$; the dissociation cross section will thus be less than this. Q_{tot} for positrons in both He and H_2 are $\gtrsim 1\pi a_0^2$ at intermediate energies so that the Ps dissociation probability would at most be comparable to p for double scattering, discussed above. However, the Ps atoms are not constrained by the magnetic field, and their mean free path will be many times the 2.3-m length of the gas cell. One can conclude from the geometry of the system that on the average a Ps atom would have to leave the scattering center at an angle of less than about 0.5° in order to dissociate with probability p and yield a positron which could then be guided to the CEM. This angle is in all likelihood smaller than the angular spread of the incident positron beam, and in the absence of more precise information it has to be assumed that the effect of Ps dissociation on f and Q_{Ps} is negligible.

5. Other sources of uncertainty

Discrimination against forward-scattered positrons has already been discussed in Sec. II B. An estimate of the minimum resolvable scattering angle θ_{min} for elastically scattered positrons of mean energy $e(V_W+1.3\text{ V})$ is obtained from

$$(V_W+1.3\text{ V})\cos^2\theta_{min}=V_R=V_W-0.7\text{ V}$$

i.e.,

$$\theta_{min}=\arccos[(V_W-0.7\text{ V})/(V_W+1.3\text{ V})]^{1/2}\ . \quad (6)$$

Therefore, for $V_W=100$ V, $\theta_{min}\approx 8°$, and for $V_W=200$ V, $\theta_{min}\approx 6°$. It is important to note that the θ_{min} values quoted here are similar in magnitude to those of Kauppila *et al.*,[9] whose Q_{tot} we are adopting in our calculations of Q_{Ps}. For a gas cell of length L filled with gas at atomic or molecular number density n,

$$Q_{tot}=-[\ln(1-F_1)]/nL\ ,$$

and

$$Q_{Ps}=-f[\ln(1-F_1)]/nLF_2$$
$$=fF_1/nLF_2+\text{ higher-order terms.} \quad (7)$$

Here F_1 and F_2 are the total attenuations measured by Kauppila *et al.* and the authors, respectively, for the same nL product. If F_1 and F_2 are both low but are equal through being subject to the same θ_{min}, then to first order (a good approximation for small F_1) $Q_{Ps}=f/nL$, independent of F. An experimental verification of the null effect on f of the detection by the CEM of slow positive ions produced in Ps formation and ionization col-

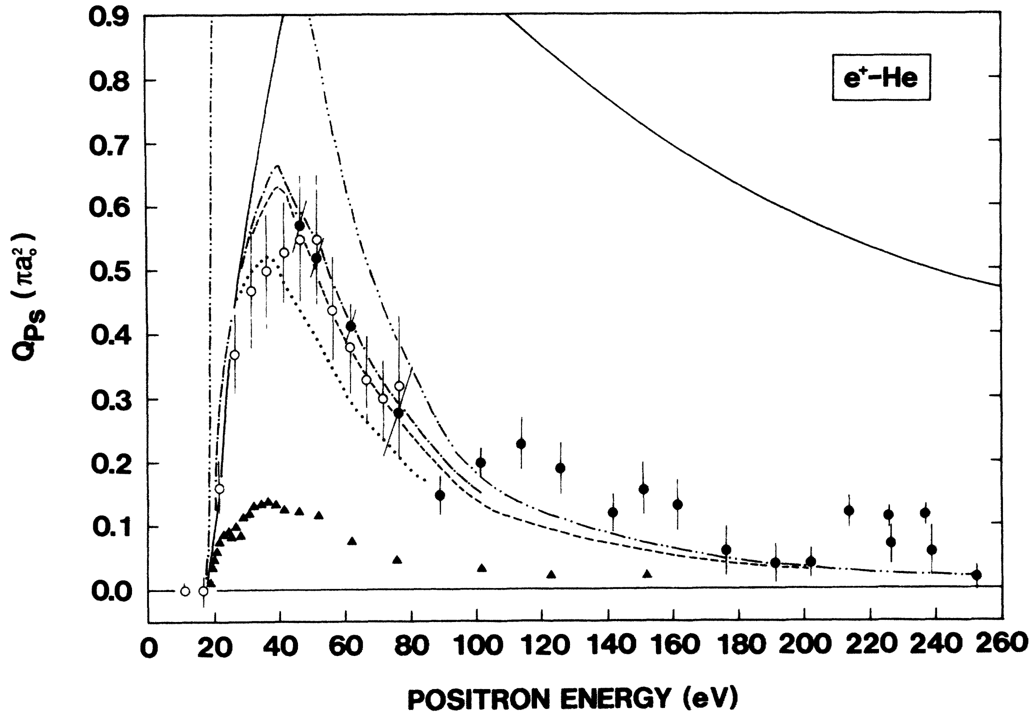

FIG. 3. Total Ps formation cross sections in He: solid circles, present results; open circles, Ref. 1; triangles, Ref. 2; solid line, Q_{inel} (see text); dash—double-dotted line, first Born Approximation (Ref. 19); dash-dotted line, distorted wave $\times 1.20$ (assuming n^{-3} rule for Ps* formation) (Ref. 18); dashed line, first-order exchange (Ref. 19); dotted line, distorted wave-polarized orbital (Ref. 20).

lisions was described in Ref. 1.

The scattering in (a) the source-electrode region and (b) within the retarding electrode does not significantly affect the measurements because the path-length—density product, and hence the probability of scattering, in these regions is negligible.

The background gas (presumably air) does scatter positrons, but at a base pressure $\sim 10^{-2} \times$(target gas pressure) and a scattering cross section $\lesssim 10 Q_{tot}$, the scattering is dominated by positron-target gas collisions. In profiles such as those in Fig. 2 it is only the (excess) attenuation due to positron collisions with He or H_2 that is measured.

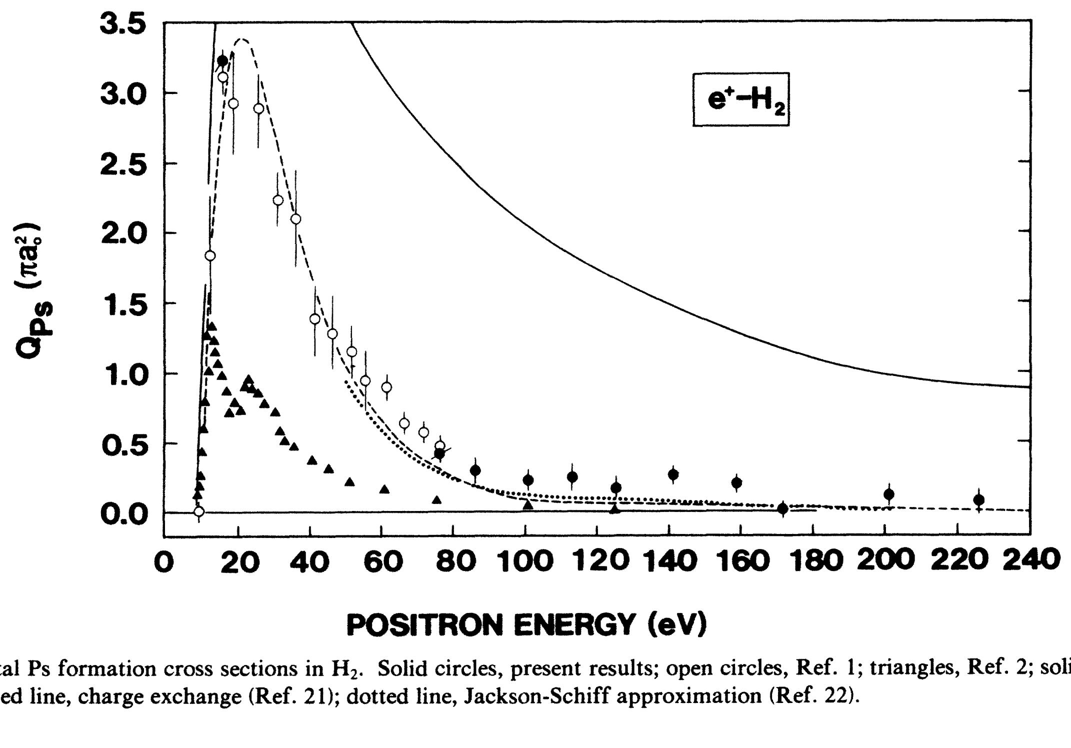

FIG. 4. Total Ps formation cross sections in H_2. Solid circles, present results; open circles, Ref. 1; triangles, Ref. 2; solid line, Q_{inel} (see text); dashed line, charge exchange (Ref. 21); dotted line, Jackson-Schiff approximation (Ref. 22).

III. RESULTS AND DISCUSSION

The results of the current measurements are listed in Table I and presented graphically in Figs. 3 and 4. It should be stressed that $f/F, = Q_{Ps}/Q_{tot}$, is measured here and that a Q_{tot} value at each energy is required to obtain the Q_{Ps} values in Table I. For helium the Q_{tot} of Kauppila *et al.*[9,15] and for molecular hydrogen those of Deuring *et al.*[16] were adopted. The (small) uncertainties in these Q_{tot} values were, of course, folded into the other uncertainties in the evaluation of Q_{Ps}. Clearly the Q_{Ps} values quoted herein may be normalized to any other set of Q_{tot} measurements, say Q'_{tot} by applying the multiplicative factor Q'_{tot}/Q_{tot}.

A small number of measurements below 76.3 eV, the upper limit of the earlier measurements of Ref. 1, were repeated in successful checks on reproducibility. In both H_2 and He the measured Q_{Ps} decreases at intermediate energies. It is faintly possible that there exists a broad, low-lying secondary peak in Q_{Ps} in H_2 between 100 and 175 eV or beyond. The case is somewhat stronger for such a secondary maximum in He between 90 and 200 eV and indeed for a tertiary maximum between 200 and 250 eV. We plan to make additional measurements of total ionization cross section Q_{ion}, in He (Ref. 17) to learn whether $(Q_{ion}+Q_{Ps})$ exhibits a smoothly decreasing energy dependence. It appears that measurements in He beyond 250 eV may well be fruitful, also.

On Figs. 3 and 4 the current values are added to the earlier results of Ref. 1. and are compared with those of Charlton *et al.*[2] and Griffith,[3] which are $\sim$90% smaller but which exhibit a similar energy dependence. Also shown are available theoretical results;[18–22] in general, the results agree remarkably well with theory. Note that we measure the total Ps formation cross section, including the excited Ps (Ps*) formation. Calculations at energies above 50 eV in He (Refs. 19 and 20) and H_2 (Ref. 22) estimate that $Q^*_{Ps}/Q_{Ps} \sim 0.12$, implying an approximate n^{-3} rule. The total inelastic cross sections Q_{inel} shown in Figs. 3 and 4 are the Q_{tot} values of Refs. 9 and 16 after subtraction of constant elastic cross sections extrapolated from below E_{Ps} (0.22 and $0.89\pi a_0^2$ for He and H_2, respectively). At lower energies Q_{Ps} is a large fraction of Q_{inel} whereas at intermediate energies it drops to a negligible fraction, probably leaving the ionization cross section Q_{ion} as the dominant inelastic channel.

Raith[23] has suggested that when comparing Q_{tot} for positron and electron scattering from the same target atoms one should first subtract Q_{Ps} from the positron cross section. This is particularly interesting in the case of H_2, in which the ratio of positron to electron Q_{tot} approaches unity from below (as is usually the case) but then overshoots in the region 50–200 eV and finally decreases asymptotically to unity from above.[23] Subtracting Q_{Ps} indeed keeps the cross section ratio below unity[24] and perhaps allows a more meaningful comparison to be made.

ACKNOWLEDGMENTS

We wish to thank Dr. L. S. Fornari, R. L. Chaplin, M. R. Murphy, R. C. Ashley, J. K. Chu, and B. E. Seay for assistance with taking and analyzing data and with apparatus maintenance and modification. It is a pleasure, also, to express appreciation to M. W. Lutes for assistance with the design, construction, and repair of the spectrometer and to Dr. R. N. Claytor and D. W. Coyne for assistance with the design, construction, and maintenance of the associated electronic equipment. We acknowledge the support of the National Science Foundation under Grants No. PHY-8306499 and No. PHY-8506933.

*Corresponding author. Present address: University of East Anglia, School of Mathematics and Physics, Norwich NR4 7TJ, United Kingdom.

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