Formation of Positronium Hydride

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We report the formation of positronium hydride (PsH) in collisions between positrons and methane. A preliminary value of 1.1 ± 0.2 eV for the binding energy of PsH is obtained.

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Since the existence of the positronium atom (Ps) was predicted [1] and later experimentally discovered [2], there has been much theoretical and experimental interest in studying the possibilities of forming Ps-containing molecules. The simplest of these are the polyleptons Ps_2 and Ps^-. Beside these, PsH, PsF, PsCl, PsBr, and PsI have all been predicted to be chemically stable from quantal calculations [3–6]. Many other, more complicated, Ps-containing molecules are thought to be stable [7].

Ps^- has been observed by Mills [8] whereas none of the other simple molecules have been observed in vacuum. However, several of the halogen compounds have been produced and identified in aqueous solutions [9] and in graphite [10]. Much less convincing experimental results suggest the formation of PsH in various kinds of condensed matter [11,12]. Below, we report an experiment in which PsH is formed in positron collisions with CH_4.

The stability of the PsH molecule was first established by Ore [5] who found it to be bound by at least 0.0683 eV. Later, many other theoretical studies of PsH have confirmed its stability with the most accurate calculation by Ho [13] yielding a binding energy, B_{PsH}, of 1.0598 eV. Whereas the stability of PsH is well known from a theoretical point of view, no calculations exist for its formation cross section in positron-molecule scattering. The simplest collision system in which PsH can be formed is obviously $e^+ + H_2$. However, as we shall see below, it is not possible by the present experimental method for this system to yield an unambiguous signature proving the formation of PsH. Therefore, CH_4 is chosen as the gas target. Below, we list some of the important products which may result from $e^+ + CH_4$ collisions together with their threshold energies:

$$e^+ + CH_4 \rightarrow CH_4^+ + Ps, \quad 6.18\ \text{eV}, \tag{1a}$$

$$CH_3^+ + PsH, \quad 7.55\ \text{eV} - B_{PsH}, \tag{1b}$$

$$CH_3^+ + H + Ps, \quad 7.55\ \text{eV}, \tag{1c}$$

$$PsCH_3 + H^+, \quad 11.31\ \text{eV} - B_{PsCH_3}, \tag{1d}$$

$$Ps + CH_3 + H^+, \quad 11.31\ \text{eV}. \tag{1e}$$

A positive signature for the formation of PsH is the detection of the CH_3^+ ion, providing it takes place below the threshold for the production of $CH_3^+ + H + Ps$.

Basically, the experimental machinery used in the present work is similar to that applied at Aarhus for mea-

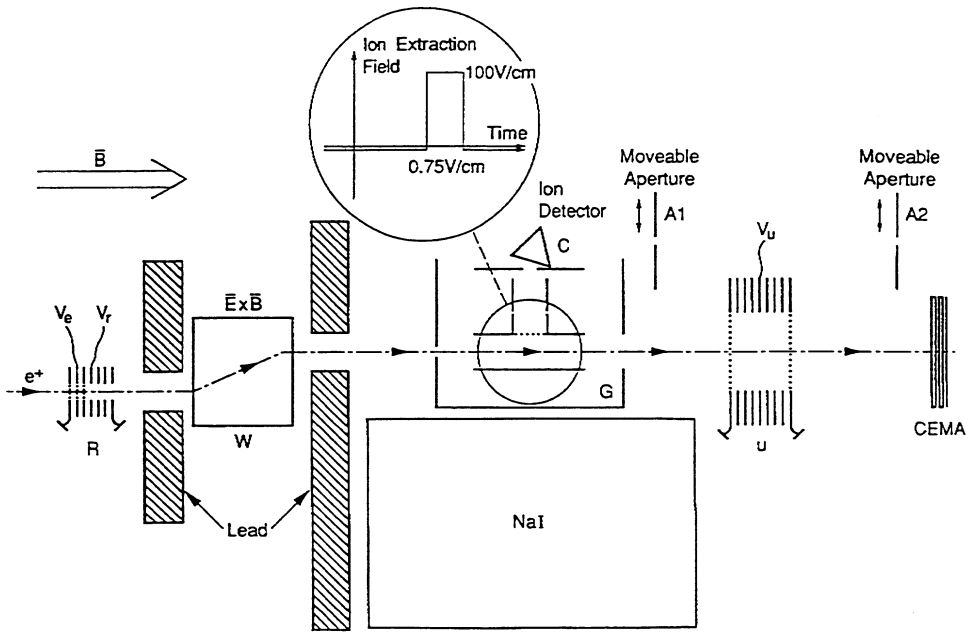

FIG. 1. Experimental setup (see text).

surements of ionization cross sections by particle impact. Figure 1 shows a sketch of our method. Low-energy positrons are obtained from a ^{22}Na isotope in combination with a tungsten β^+ moderator. The low-energy positrons are transported by an axial magnetic field of 50 G. The beam traverses a retarding element, R, whereafter it is deflected 4 cm by a Wien filter, W, and then it enters the gas cell, G, through a 1-cm-diam aperture. The beam leaves the gas cell through a 1.8-cm-diam aperture and continues downstream, passing an accelerator structure, U, whereafter it is dumped onto a channel plate detector. Two mobile 0.5-cm-diam apertures (A1, A2) are used prior to each measurement in order to ensure that the beam is on the geometrical axis of the apparatus.

The gas cell is viewed by a NaI detector and when a gamma event is recorded a 3-μsec pulse of 100 V/cm is applied across the gas cell in order to extract and transport a possibly created ion to a ceatron detector, C. The effective length of the gas cell is approximately 1 cm. Our signal is obtained by the time-of-flight method with C supplying the start signal and NaI the stop signals to produce an inverted time-of-flight spectrum [Fig. 2(a)].

Ideally, our signal originates from the formation of Ps either free or bound. However, it is possible that an ion is created by some unintended means and then extracted by a random pulse from the NaI detector. The agents for producing such ions could be high-energy electrons or positrons contaminating the beam. In order to reduce such false signals, the ion lifetime in the gas cell is limited to 5 μsec by a weak electric field of 0.76 V/cm permanently applied across the gas cell and pointing away (to avoid background) from the ion detector. Furthermore, to prevent electrons created at the channel plate detector from entering the gas cell, -1 kV is applied to the accelerator structure U whereas electrons coming from the moderator region are suppressed by applying -100 V to V_e.

One can think of other mechanisms by which random signals may be created. In this respect it is important to emphasize that any significant random contribution is visible in the time-of-flight spectra as it results in a broadening of the peaks since these ions are collected over a larger volume. This point is illustrated in Fig. 2(b) which shows an ion time-of-flight spectrum for which the ions were created by an electron beam and extracted by the noise counts of the NaI detector.

The energy spread, $\delta\varepsilon$, of the positrons is 3 eV (full width) when they leave the tungsten moderator. In order to reduce $\delta\varepsilon$, only 25% of the beam is allowed to pass the retarder R (by applying a sufficiently high positive potential to V_r), resulting in an energy spread $\delta\varepsilon < 0.5$ eV and with a FWHM approximately equal to a 0.3 eV. To this quantity, however, must be added about 0.7 eV due to deceleration or acceleration of the e^+ caused by the static weak field applied across the gas cell. Hence, the effective energy spread $\delta\varepsilon$ is close to 1 eV.

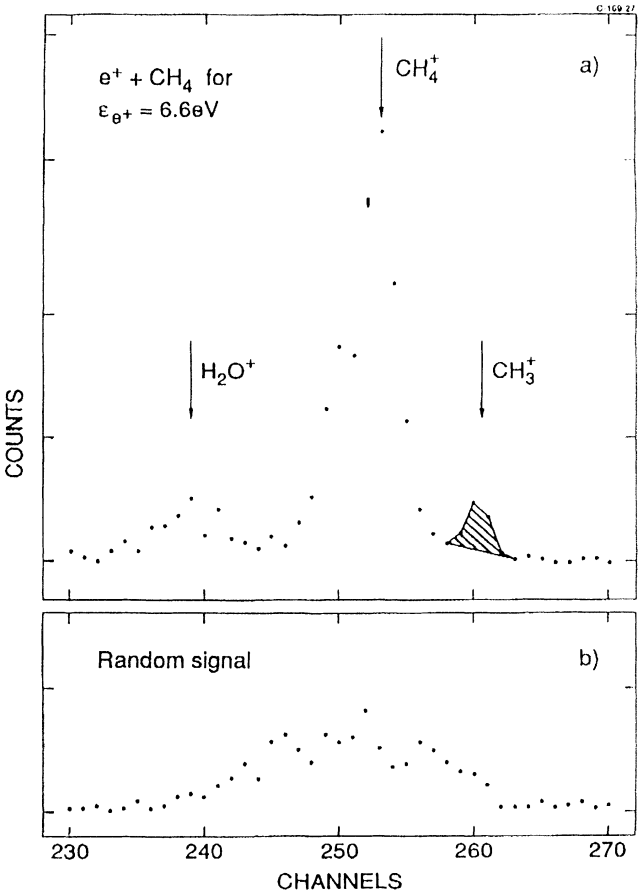

FIG. 2. (a) A normal time-of-flight spectrum where the start signal is supplied by the ion detector and the stop signal by the gamma detector. (b) A time-of-flight spectrum where the ions were created by electrons and extracted by the noise counts of the gamma detector. Note the broadening of the random signal. For both spectra the ion flight time increases from right to left.

The cross sections σ_i for the production of the various ions are related to the gas density, n (mtorr), and the number of positrons, N_+, entering the gas cell as

$$\sigma_i = kA_i/nN_+ , \tag{2}$$

where A_i is the number of ions of type i and k is a simple constant which in principle can be determined by a suitable normalization procedure. In Fig. 3 we display the results for CH_4 as σ_i/k versus the average positron impact energy.

The measurements of the yields of CH_4^+ do not quite reflect the behavior of the ordinary Ps formation cross section, σ_{Ps}, as the detection efficiency of the o-Ps depends on its kinetic energy. The lifetime of o-Ps is about 140 nsec, resulting in a flight distance of approximately $6.5[\varepsilon_{Ps}(\mathrm{eV})]^{1/2}$ cm, a distance which is, in most cases, long in comparison to the size of the NaI detector. For p-Ps and PsH this problem does not exist because of their

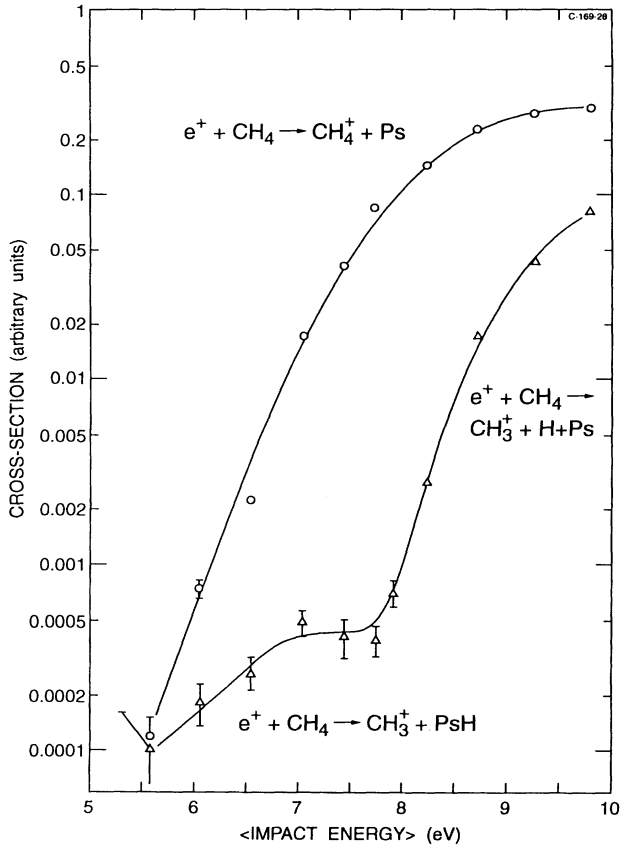

FIG. 3. Cross sections for the production of CH_4^+ and CH_3^+ ions in positron collisions with CH_4.

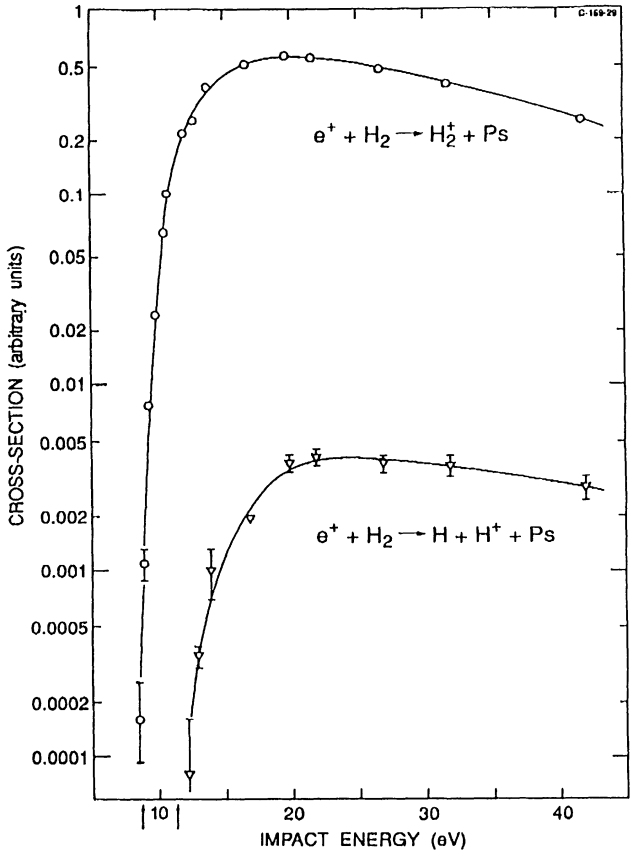

FIG. 4. Cross sections for the production of H_2^+ and H^+ ions in positron collisions with H_2.

short lifetimes (0.125 and 0.407 nsec [13]) and further because of the heavy mass of PsH.

To contrast the results for $e^+ + CH_4$ we display the data for the $e^+ + H_2$ system in Fig. 4. Here, the signature ion for the formation of the PsH molecule is H^+. However, the first appearance of the H^+ signal occurs at an impact energy comparable to the threshold for the $Ps + H + H^+$ channel. This means that either σ_{PsH} is very small for this collision system or the potential curve of $PsH + H^+$ in the Franck-Condon region is repulsive, implying that the production threshold for PsH is higher than its thermodynamical threshold. If the latter is true the H^+ as well as PsH will be produced with kinetic energy, and providing this quantity is measured it will be possible to separate the $PsH + H^+$ channel from that of $Ps + H + H^+$.

Let us now return to the $e^+ + CH_4$ system (Fig. 3). From the simplest theoretical point of view, it is expected that σ_{PsH} should rise sharply at the threshold and then fall off quickly with increasing impact energy. The reason is that the positron has almost to come to a stop in order to form PsH. In contrast, we observe (Fig. 3) a gradual increase over approximately 1.5 eV. However, this is mainly due to the large energy spread $\delta\varepsilon$ of the e^+ beam. From a comparison between the onsets of $e^+ + CH_4 \rightarrow CH_4^+ + Ps$ and $e^+ + CH_4 \rightarrow CH_3^+ + PsH$ and Eqs. (1), we can deduce a binding energy of PsH to be $B_{PsH} = 1.1 \pm 0.2$ eV.

In the present study we also look for the appearance of the H^+ ion below the threshold for the channel leading to $CH_3 + H^+ + Ps$, however, with a negative result.

In this Letter, we have demonstrated that PsH can be formed *in vacuo*. Although the binding energy of PsH as obtained in the $e^+ + CH_4$ experiment is comparable to the theoretical one given by Ho [13], the accuracy of the present work is not high enough to challenge the theory.

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