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# Crossed beam measurement of binding energies of positronium compounds: a preliminary study

D.M. SCHRADER, G. LARICCHIA* and T.N. HORSKY**

*Chemistry Department, Marquette University, Milwaukee, WI 53233, U.S.A.*
* *Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, U.K.*
** *Balzers High Vacuum Products, 8 Sagamore Park Road, Hudson, NH 03051, U.S.A.*

**Abstract**

We describe an experimental method which will enable one to establish the chemical stability and measure the binding energies of compounds containing positronium atoms. The method consists of crossing a monoenergetic beam of very low energy (1 – 25 eV) positrons with a supersonic expansion molecular beam, and detecting a signature ion while measuring its kinetic energy. Except for the use of a positron beam instead of an electron beam, this is conventional MS/IKES (mass spectrometry/ion kinetic energy spectrometry), and the process involved is dissociative attachment.

## 1 Introduction

The reaction to be studied is:

$$e^+ + AB \rightarrow \{e^+,AB\} \rightarrow PsA + B^+ \tag{1}$$

The positron, with a well-defined energy, approaches a nearly stationary molecule AB (not necessarily diatomic), forming a resonance which subsequently dissociates to the indicated product. Simple energy balance considerations, which can be easily read from Fig. 1, yield the desired quantity, the Ps–A bond energy, from the A–B bond energy, the ionization potential of B, and the kinetic energy of the products:

$$E(e^+) - KE(PsA + B^+) = BE(AB) + IP(B) - 6.8eV - BE(PsA) \tag{2}$$

$E(e^+)$ is the energy of the positron beam, 6.8 eV is the binding energy of the positronium atom, Ps, and $BE$ and $IP$ denote respectively the bond energy and ionization potential, both presumed known. $KE(PsA + B^+)$ is the kinetic energy of the products, and this is related to that of $B^+$, which is to be measured, by

$$KE(PsA + B^+) = \frac{M_{AB}}{M_A} KE(B^+), \tag{3}$$

where the $M$'s are masses.

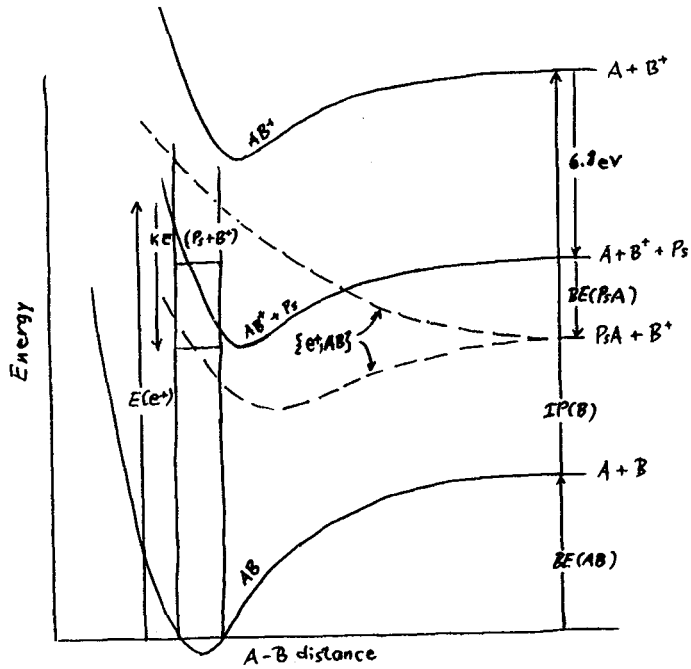


Figure 1: Potential energy curves relevant to the discussion. Two possible resonances are shown as dashed lines.

## 1.1 Logic of the Experiment

An observation of the appearance potential of $B^+$ and its kinetic energy is sufficient to establish the binding energy of PsA, providing the identity of the ion is established. This depends upon thermodynamic threshold for the process (1) being lower than that for all other processes which produce the species $B^+$. While this ideal is never fully realized in practice, an energy window between the thresholds for PsA + $B^+$ and A + $B^+$ + Ps in which interfering reactions give manageable complications. Thus, within this window, the detection of the signature ion $B^+$ is proof of the formation of the compound PsA.

## 1.2 Design Goals

We take the following as our design goals:

- We want to determine $BE$(PsA) in Eq. (1), the binding energy of the compound, to within 0.1 eV.
- We want to determine the mass of the signature ion to be less than 1 amu, so that we can tell whether a given ion has lost a hydrogen atom.

- We want the count rate of the signature ion (when its kinetic energy is *not* being measured) to be statistically equivalent to at least 1/sec with no background. [KAJC90a] While an experiment is possible with count rate substantially less than this, we take this figure to be our nominal design goal.

In the prototype experiment, [SCHR92a] our ion count rates were $\sim 2/\mathrm{hr}$, the mass resolution was about 10%, and the energy of the signature ion was not measured. Compared to the earlier experiment, what we are proposing here is a qualitatively upgraded experiment.

# 2 Feasibility of the Proposed Experiments

In this section we use three target molecules as examples in the interest of concreteness: $H_2$, HI, and $I_2$. These molecules represent the mass range of the targets of interest, an important consideration (see Eq. (3)), and their chemical simplicity aids quantitative analysis.

## 2.1 Quantum Mechanics of the Process

Some of the quantum mechanical aspects of the process have already been discussed, especially the formation, autoionization, and dissociation of the resonance state. [SCHR92c] In the present work, we limit the discussion to topics which have not yet been considered.

For the three example target molecules, there are four possible products:

$$
\begin{array}{lcccccc}
e^+ + & \underset{{}^1\Sigma_g^+}{H_2} & \rightarrow & \underset{{}^{2,1}\Sigma_g^+,\ {}^{2,1}\Sigma_u^+}{\{e^+, H_2\}} & \rightarrow & \underset{{}^{2,1}S^e}{PsH} & + \underset{{}^1S^e}{H^+} \\
e^+ + & \underset{{}^1\Sigma^+}{HI} & \rightarrow & \underset{{}^{2,1}\Sigma^+}{\{e^+, HI\}} & \rightarrow & \underset{{}^{2,1}S^e}{PsI} & + \underset{{}^1S^e}{H^+} \\
e^+ + & \underset{{}^1\Sigma^+}{HI} & \rightarrow & \underset{{}^{2,3}\Sigma^-,\ {}^{4,3}\Sigma^-,\ {}^{2,3}\Pi,\ {}^{4,3}\Pi}{\{e^+, HI\}} & \rightarrow & \underset{{}^{2,1}S^e}{PsH} & + \underset{{}^3P_2^e}{I^+} \\
e^+ + & \underset{{}^1\Sigma_g^+}{I_2} & \rightarrow & \underset{\substack{{}^{2,3}\Sigma_g^-,\ {}^{4,3}\Sigma_g^-,\ {}^{2,3}\Pi_g,\ {}^{4,3}\Pi_g,\\ {}^{2,3}\Sigma_u^-,\ {}^{4,3}\Sigma_u^-,\ {}^{2,3}\Pi_u,\ {}^{4,3}\Pi_u}}{\{e^+, I_2\}} & \rightarrow & \underset{{}^{2,1}S^e}{PsI} & + \underset{{}^3P_2^e}{I^+}
\end{array}
\tag{4}
$$

Term symbols are given beneath each species. The super-pre-scripts of the term symbols are the total spin quantum number $S_{tot}$, and the total for the electrons, $S_e$. [LEBE69a] The term symbols of the resonances are the result of the coupling of those of the two product species in each case, by a slight extension of the Wigner-Witmer rules. [WIGN28a, MULL32a] In general, there is a one-to-many correspondence between product states and resonance states. Each resonance has its own potential energy curve, all of which are unknown at the present except at their common asymptote.

These resonances are all the Feshbach-type; i.e.; they form when the incident positron excites the target ground state molecule to an excited state which supports an attractive potential sufficiently strong to bind the positron.

Table 1: Angular Dependence of the Products with Respect to the Positron Beam

| Reaction | Resonance Symmetry/Angular Dependence | | | |
|---|---|---|---|---|
| $H_2 \rightarrow PsH$ | ${}^{2,1}\Sigma_g^+$ isotropic | ${}^{2,1}\Sigma_u^+$ parallel | | |
| $HI \rightarrow PsI$ | ${}^{1}\Sigma^+$ isotropic | | | |
| $HI \rightarrow PsH$ | ${}^{2,3}\Sigma^-$, ${}^{4,3}\Sigma^-$ forbidden | ${}^{2,3}\Pi$, ${}^{4,3}\Pi$ perpendicular | | |
| $I_2 \rightarrow PsI$ | ${}^{2,3}\Sigma_g^-$, ${}^{4,3}\Sigma_g^-$ forbidden | ${}^{2,3}\Pi_g$, ${}^{4,3}\Pi_g$ forbidden | ${}^{2,3}\Sigma_u^-$, ${}^{4,3}\Sigma_u^-$ forbidden | ${}^{2,3}\Pi_u$, ${}^{4,3}\Pi_u$ perpendicular |

### 2.1.1 Dissociation of the Resonance State

#### 2.1.1.1 Angular Dependence of the Dissociation Products

A study of the symmetry of the dissociative attachment scattering event reveals constraints on the direction which the products may take when leaving the scattering site. [DUNN62a] For diatomic targets, this takes the role of a $\sin^2\theta$ or $\cos^2\theta$ dependence of the scattering probability, where $\theta$ is the angle between the direction of the incident particle beam and the internuclear axis of the target molecule. Two other possibilities are: an isotropic distribution, and a null distribution (i.e., the dissociation is forbidden at all angles). For the three example targets in the Reactions (4), the results are given in Table 1.

Such considerations, which can be extended to more complicated targets, are important, for they tell us where to put our detectors for a given target and a particular desired product. For the "forbidden" cases, dissociation will not occur. The resonances are there, but they cannot be created by the collision process considered.

#### 2.1.1.2 The Influence of Spin on the Dissociation Products

For the last two reactions in (4), the signature ion is $I^+$, which has a ${}^3P_2^e$ ground state. Since the total electron spin $S_e$ is a good quantum number throughout the collision process, and $S_e = 0$ for the reactants, it follows that resonance states with $S_e = 1$ cannot be formed by the process. Thus, the bottom half of Table 1 is forbidden on account of spin conservation.

## 2.2 Meeting the Design Goals

### 2.2.1 Mass Resolution

The start signal for the MS/IKES detection can in principle come from any of several sources. The final remoderation of the positron beam produces secondary electrons, which can be detected and made to provide a start signal with an uncertainty of $\sim 10^{-9}$ sec. [LARI88b]

For a LINAC-based beam, the pulse rate can be up to $\sim 10^3$ Hz, with a pulse width of $\sim 10^{-9}$ sec. [HOWE85a] Since the pulsing is an intrinsic part of the operation of the LINAC, it can be used to provide a start signal for our MS/IKES detectors. For either type beam, the annihilation gammas from PsA can also be used to provide a start signal with a time uncertainty of $\sim 1$ nsec.

The flight time for the signature ion $B^+$ depends upon its energy and mass. If $M_B \ll M_{AB}$, then most of the kinetic energy of the products will be deposited in $B^+$ (Eq. (3)). If the energy of the incident positron is 1 eV above the threshold for the formation of PsA, and if $B^+$ is a proton, so that more than 0.5 eV is imparted to it, then less than $10^{-4}$ sec is required for its traverse of a 1 m flight path of an MS/IKES detector. Thus, for a LINAC-based positron beam, the highest pulse rate is satisfactory. A mass resolution of 1 amu requires the uncertainty of the start signal to be not greater than 1% of the time of flight for a target molecule with a mass of 100 amu. We see that any of the detection devices will provide this. Another necessary condition is that the dimension of the scattering region along the ion flight path be not more than 1 cm, which is 1% of the flight path.

### 2.2.2 Ion Kinetic Energy Resolution

Uncertainty in the measurement of the ion kinetic energy comes from the thermal motion of the target gas, and from the variation of flight times from the scattering site to the MS/IKES detectors on account of the thickness of the scattering region and the acceptance angle of the detectors.

#### 2.2.2.1 Thermal Motion of the Target Gas Molecules

In order to determine $KE(\text{PsA}+\text{B}^+)$ to within 0.1 eV, we must measure the ion kinetic energy to within an uncertainty of (Eq. (3)):

$$\delta KE(B^+) \leq \frac{M_A}{M_{AB}} \times 0.1eV \tag{5}$$

It is easy to derive another expression for this quantity: [ZIPF84a, for example]

$$\delta KE(B^+) \sim 2\sqrt{\frac{3}{2}kT \times KE_0}. \tag{6}$$

Combining Eqs. (5) and (6) with Eq. (3) gives a useful expression,

$$\sqrt{\frac{3}{2}kT_{max}} \sim \left(\frac{M_A}{M_{AB}}\right)^{\frac{1}{2}} \frac{0.05\,eV}{\sqrt{KE(PsA+B^+)}}, \tag{7}$$

from which the maximum temperature of the gas consistent with our design goal may be determined. For example, for the four examples in Reaction (1), we find $T_{max}$ = 9K, 19K, 0.15K, and 9K, resp.

Clearly, a supersonic adiabatic expansion molecular beam is the only suitable target, as such temperatures are routinely achieved. Rotational energy is largely subsumed into kinetic

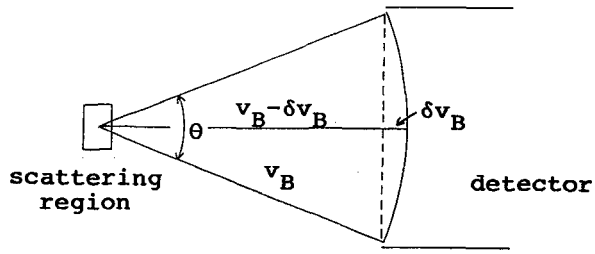


Figure 2: Relationships between the acceptance angle $\theta$, $v_B$, and $\delta v_B$.

motion, but the conclusion is the same. Only the third of the reactions (4) won't work, but that is spin-forbidden anyway.

2.2.2.2 Acceptance Angle of the Detectors

Clearly

$$\delta KE(B^+) \sim M_B\, v_B\, \delta v_B = 2KE(B^+)\, \frac{\delta v_B}{v_B}. \tag{8}$$

From Fig. 2(a), $\delta v_B$ is given in terms of the acceptance angle $\theta$ as

$$\cos\frac{\theta}{2} = \frac{v_B - \delta v_B}{v_B} = 1 - \frac{1}{2}\frac{\delta KE(PsA + B^+)}{KE(PsA + B^+)}, \tag{9}$$

For measuring a kinetic energy of 1 eV to within 0.1 eV, we find:

$$\theta \leq 36^\circ \tag{10}$$

### 2.2.3 Estimation of the Minimum Positron Beam Intensity

The count rate of the signature ion may be written as

$$i_{B^+} = \varepsilon\, i_{e^+}\, \sigma_{PsA}\, n(z)\, L = \varepsilon\, i_{e^+} \times 10^{-5}, \tag{11}$$

where we have used the following numerical values: $10^{-18}$ cm$^2$ for the cross section $\sigma_{PsA}$, $10^{13}$ cm$^{-3}$ for the number density $n(z)$, and 1 cm for the path length for the interaction, $L$. $\varepsilon$ is the combined efficiencies of the ion collection, transmission down the flight path, and detection at the end, and $i_{e^+}$ is the positron beam intensity.

If we take 36° as the acceptance angle, then, for an isotropic distribution of products, the fraction that will reach the input end of the detector assembly is just the area on a unit sphere subtended by a solid angle of 36°, expressed in units of $4\pi$. This is easily calculated

to be $2.4 \times 10^{-2}$.[1] This quantity is a factor of $\varepsilon$ for zero-field IKES measurements, but not for high-field MS measurements. We take 0.5 as the combined efficiency of the rest of the MS/IKES assembly: i.e., when only the mass of $B^+$, not its kinetic energy, is measured.

Our design goal for the minimum acceptable ion count rate $i_{B^+}$ is 1/sec without kinetic energy measurement. Substituting values into Eq. (11) gives the minimum acceptable positron beam intensity:

$$i_{e^+} \geq 2 \times 10^5 \text{ sec}^{-1} \tag{12}$$

For an isotope-based beam with a solid neon moderator and a transmission foil for remoderation and timing, a reasonable overall efficiency is about $2 \times 10^{-5}$. Thus a source strength of about 250 mCi is sufficient.

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[1] This quantity changes only slightly for nonisotropic product distributions (Table 1), as long as the detector is sampling a favorable part of the distribution.