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Thermal activation of positronium from thin Ag(100) films in backscattering and transmission geometries

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Abstract. Emission of positronium (Ps) from thin Ag(100) foils (1500–1900 Å) has been studied in reflection and transmission geometries. Positron transmission probabilities of the 1900 Å foil have been extracted from the data and fitted to give an effective positron diffusion length of 425–475 Å. Furthermore, the one-dimensional velocity distribution perpendicular to the foil surface has been estimated to be non-Maxwellian. The conversion efficiency of incident positrons to Ps in transmission is estimated to be ~ 12% for a 1900 Å foil at 800 K.

1. Introduction

The positronium atom is a hydrogen-like bound state of an electron–positron pair and there have been many studies of its properties (Rich 1981) mainly as tests of QED. Recently techniques have been developed (Schultz and Lynn 1988) which allow the controlled production of low energy Ps in vacuum using low energy positron (e^+) beams. Such developments have led to the measurement of the 1^3S_1–2^3S_1 Ps transition frequency (Chu and Mills 1982, Chu *et al* 1984) and the first production of Ps in a Rydberg state (Ziock *et al* 1990). It has also been pointed out that collisions of antiprotons with Ps can be used to form antihydrogen in the laboratory (Deutch *et al* 1986, 1988) and it is this application which has motivated the study reported here.

It was found a number of years ago that Ps may be formed at the surface of a metal target by bombarding it with positrons and that the yield was enhanced when the target temperature was raised (Canter *et al* 1974). A positron may form Ps by picking up an electron at the outer surface of an unheated metal. The Ps is then emitted into vacuum and the formation potential, ϵ_{Ps}, for this process is determined by energy balance

$$\epsilon_{Ps} = \phi_+ + \phi_- - 6.8 \text{ eV} \tag{1}$$

where ϕ_+ and ϕ_- are the positron and electron workfunctions, respectively, and 6.8 eV is the groundstate Ps binding energy. The enhancement of the Ps yield with temperature has been described in terms of surface traps in which the positrons reside

but from which they may be desorbed as Ps due to thermal fluctuations within the trap (Mills 1979). The activation energy, E_a, needed for desorption is given by

$$E_a = E_b + \phi_- - 6.8 \text{ eV} \tag{2}$$

with E_b the binding energy of the positron in the trap.

The yields of such thermally activated Ps from various metals have been studied in the backscattering (reflection) geometry at different temperatures (e.g. Mills 1979, Lynn 1979, Lynn and Welch 1980) and in some cases (Mills and Pfeiffer 1979, 1985) the velocity distribution of the desorbed Ps perpendicular to the sample surface has been determined. Whilst the description of the former is well established (e.g. Lynn and Welch 1980) the latter is not. In the first study of Mills and Pfeiffer (1979) of Ps emission from Cu(111) the initial assumption of a Maxwellian distribution of the desorbed Ps had to be modified by the inclusion of a pre-exponential factor in order to fit the data. This pre-exponential factor was determined empirically to be proportional to $1 + (v_z/v_0)^2$, where v_z is the velocity perpendicular to the sample surface and v_0 is a fitting parameter found to be $8.7(1.3)\times10^4$ m s^{-1}. In a later study (Mills and Pfeiffer 1985) the pre-exponential factor was not needed in order to fit the data obtained from an Al(111) surface. The physical significance of the form of these distributions is not well understood.

To date the thermal activation of Ps has only been studied from thick crystals—and consequently in the backscattering geometry. However in some cases it may be advantageous to have the Ps formed on the transmission side of a thin metal foil (hereafter termed the Ps converter) to allow, for instance, electrical isolation between the incoming e^+ beam and the region in which the Ps interaction is studied (Sferlazzo 1985). Formation of Ps has been observed in the transmission geometry from MgO (Curry and Schawlow 1971) bombarded with β^+ particles and Mills *et al* (1989) have recently studied Ps emission from a pressed pellet of SiO_2 powder at various temperatures in the backscattering geometry. This latter work could be extended to the transmission case (Mills and Leventhal 1989) and used either with a β^+ source or a slow e^+ beam. However the use of these insulators may not always be suitable if, for instance, the experiments involve charged-particle–Ps collisions.

The present investigations were undertaken with the purpose of producing an efficient Ps converter to be positioned in one of the electrodes of a radio-frequency-quadrupole ion trap in order to make antihydrogen in collisions between antiprotons and Ps (Deutch *et al* 1988). In order to maintain the characteristic quadrupole field within the trap, metal converters were preferred. Thin Ag foils were chosen since they are relatively easy to fabricate, not very brittle and because their temperature can be raised (up to $\sim$ 800 K) to get a high Ps yield without increasing positron trapping at vacancies (Lynn and Welch 1980). Also since the positron workfunction is believed to be positive (Nieminen and Hodges 1976, Boev *et al* 1987) Ps formation is the only escape route into the vacuum for thermalized positrons which reach the surface.

In this paper we present the following results of thermal activation of Ps from thin Ag(100) films: (i) transmission probabilities compared with incident positron energy for a 1900 Å Ag(100) foil and (ii) velocity measurements of the thermally desorbed Ps in both backscattering and transmission geometries.

2. Apparatus

The present studies were performed using a low energy e^+ beam. The slow positrons

were obtained from a 10 mCi ^{22}Na source positioned in front of a W mesh moderator (see left insert of figure 1(*a*)) in which some β^+-particles thermalize, diffuse to the surface and are ejected into vacuum with eV energies. This yielded a beam intensity of $2\text{–}3\times10^4$ e^+s^{-1}. The moderated e^+ were accelerated to the desired beam energy and magnetically confined and transported up to the interaction region (right-hand side of figure 1(*a*)) by an array of Helmholtz coils providing an axial magnetic field of approximately 50 G. The interaction chamber contained the sample holder, a ceramic electron multiplier (ceratron) and a halogen lamp with its associated Au-coated reflector. The chamber pressure was $\sim 10^{-9}$ Torr.

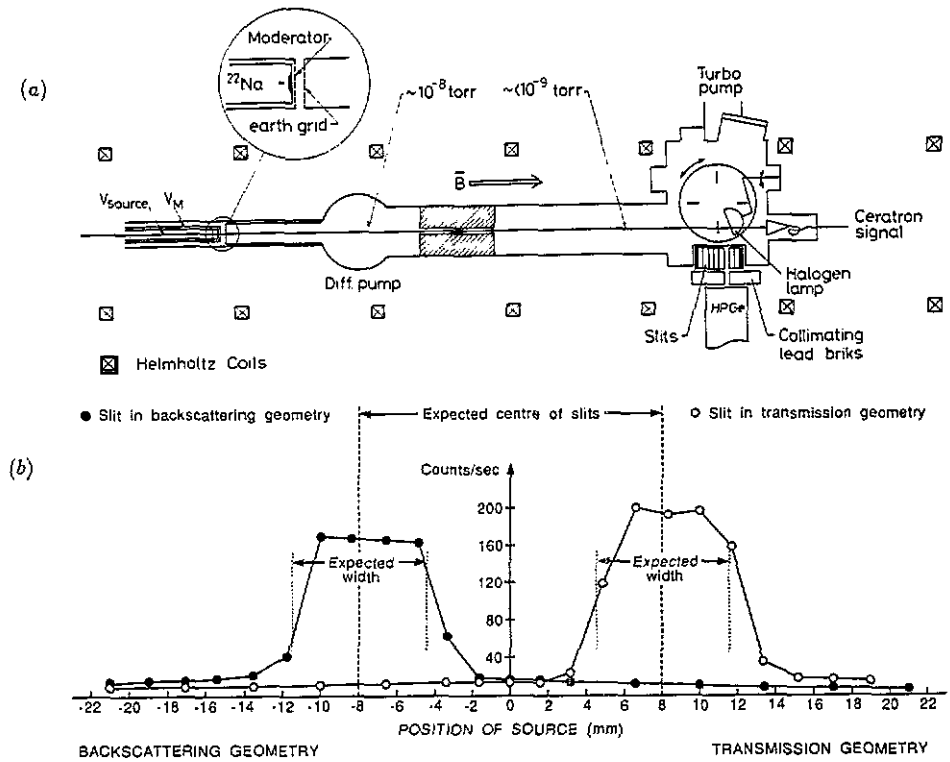

Figure 1. (*a*) Schematic experimental setup of the magnetically guided e^+ beam with the interaction region on the right-hand side. (*b*) The resolution of the slit system measured with a small ^{22}Na source (see text for details).

The sample holder supported one dummy sample and three Ag foils. These were grown by vacuum evaporation onto NaCl crystals covered with a thin layer of LiF. Each foil was held between two polycrystalline Ag rings (opening diameters of 6 mm), which were mounted in a more robust stainless steel holder. The purpose of the dummy sample (opening diameter of 6 mm) was to locate the incident positron beam which was monitored using the ceratron.

After measurements the Ag foils were taken out of the vacuum system and examined with a transmission electron microscope. According to the diffraction patterns, the samples were monocrystalline with extra spots corresponding to twins. The area of the twins represents about 10% of the total surface of the samples.

The lamp was positioned on the transmission side of the foil and its purpose was to heat the foil up to the desired temperature. The lamp (OSRAM model No HLX

64635) consisted of a halogen lamp positioned in a Au-coated glass reflector and had a maximum output of 150 W at 15 V. The lamp was removed from its original holder and placed in a special UHV compatible holder mounted on a manipulator. The latter made it easy to withdraw from the beam axis when locating the positron beam. The lamp was powered by a stabilized supply.

The presence of positronium in vacuum formed by e^+ interactions with the Ag films was monitored using a HPGe γ-ray detector located behind a slit system which was perpendicular to the beam axis and consisted of an array of 80 mm long stainless steel shutters separated by a plastic frame of effective length 12 mm and 6 mm wide. The purpose of the plastic frame was to define a view-area for the HPGe detector into the interaction region. Calculated values for the attenuation of the 511 keV annihilation γ-ray in the steel shutters and the plastic frame were 99.4% and 13.6%, respectively. The resolution of the slit system was improved by two additional collimating lead bricks of lengths of 50 mm placed between the slits and the detector. The resolution of the slit system was measured with a small ^{22}Na source mounted in one of the sample positions and the rate of 511 keV γ-rays through a 7 mm slit opening was counted as a function of source position with respect to the centre of the slit system on either side of that centre. The result is shown in figure 1(*b*) where it is seen that the measured widths of the slit openings are in agreement with the actual widths but that the measured positions of these are shifted by approximately half a millimetre towards the transmission side compared with expectations. This shift resulted in a small correction to the data which will be discussed.

The temperature of the silver foils was estimated by an indirect measurement. At the end of a run the slit system was removed and the detector moved close up to the sample in order to increase the detection efficiency. A relative measure of the fraction of 100 eV incident positrons converted into Ps was recorded as a function of the voltage supplied to the lamp. An Ag(100) crystal (~125 μm thick) was then placed at the foil position and similar measurements were performed, but this time as a function of the sample temperature measured with a thermocouple spotwelded onto the backscattering side of the crystal. By normalization to each other a temperature conversion was obtained of the former measurement.

In this study two temperatures of the foils were used, namely room temperature (cold) and the foils elevated to $\sim$ 800 K (hot) as deduced by the previously described method. Higher temperatures for the experiments were attempted but due to difficulties in maintaining sufficient stability to avoid evaporation of the foils the maximum temperature of $\sim$ 800 K was used.

3. Results and discussion

In the measurements discussed in this section γ-ray spectra were recorded through an opening in the slit system. A measure of the rate of ortho-Ps annihilating within the view of the slit opening was obtained by time-normalizing areas at energies below the 511 keV annihilation peak. These areas also contain a background contribution but by subtracting cold measurements from hot at the same incident energy this contribution cancels out as it is temperature independent.

In the cold measurements the possible contributions to the count rate of Ps are, if the positrons reach the surface of the foil and leave this into vacuum (i) prior to thermalization as energetic Ps and (ii) after thermalization as Ps (provided $\epsilon_{Ps} < 0$).

In the hot measurements there is an additional contribution namely (iii) thermal activation as Ps from the surface potential well in which the positrons may be trapped following thermalization and diffusion. Contributions (i) and (ii) are taken to be temperature independent, the latter since the diffusion length was expected to be unaltered by the temperature change (Lynn and Welch 1980). Therefore the difference between hot and cold measurements is solely due to the contribution (iii) from thermal activation of Ps which in turn depends upon the transport probabilities of thermalized positrons to an external surface.

3.1. Transmission probabilities

Count rates of thermally desorbed Ps (hot–cold) using a 6 mm slit opening located on the transmission side were taken for different energy positrons incident on a foil and compared with an identical measurement (reference point) on the backscattering side (mirror-like reversed slit system). To perform an appropriate comparison of the actual yield of desorbed Ps on the two sides the following corrections need to be applied: (i) since the beam is larger than the inner diameter of the Ag rings sandwiching the foils, a fraction of the incident positrons will impinge on the rings. In the backscattering geometry these positrons may still be thermally activated as Ps thus resulting in an overestimate of the reference point. Assuming equal desorption from the ring and the foil this can be corrected for using the ratio between the count rate of positrons which pass through the dummy sample to the ceratron and the full beam recorded with all sample holders out of its path. The effect of this is to multiply the measured transmission probabilities by 1.10. (ii) According to figure 1(*b*) the foil is off-centre with respect to the slit system by a half millimetre. In the next section an expression for the count rate of thermally activated Ps through a slit opening is made as a function of distance from the foil. Making use of this and taking the ratios between calculated count rates with $l = 4$ mm and $l = 5$ mm (l should ideally have been 4.5 mm), where l is the perpendicular distance from the foil to the entrance to the slit opening, the correction is to multiply the data by 1.09. Thus the total correction is a multiplicative factor of 1.20 and this has been taken into account in the data shown in figure 2. The data as such show the efficiency by which positrons can be converted into Ps by thermal activation in the transmission geometry versus incident energy, E, compared with what can be achieved in the backscattering geometry with a 100 eV incident beam.

When considering the fraction of incident positrons which are able to leave the surface as thermally activated Ps the possibility of emission as non-thermalized positrons or energetic Ps must be discussed. In the following it is assumed that the branching ratios of the transmission and backscattering surfaces are equal for all relevant emission processes. The measured ratios, $R(E)$, between transmission and backscattering geometries are then given by

$$R(E) = \frac{[1 - \beta_{e+}(E) - \beta_{Ps}(E)]}{[1 - \beta_{e+}(100\text{ eV}) - \beta_{Ps}(100\text{ eV})]} \frac{Y_T(E)}{Y_B(100\text{ eV})}. \tag{3}$$

Here β_{e+} and β_{Ps} are the fractions of incident positrons that are re-emitted into vacuum at the entrance surface prior to thermalization as either bare positrons or energetic Ps at the indicated incident energies, $Y_T(E)$ is the probability of thermalization and diffusion to the transmission surface of non-backscattered positrons in the foil versus energy, and $Y_B(100\text{ eV})$ is the probability of thermalized positrons returning to the

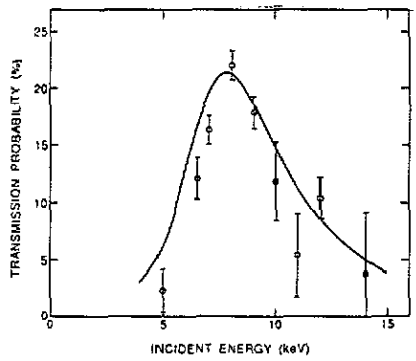

Figure 2. The measured energy dependence of the conversion efficiency of positrons into thermally activated Ps in transmission from a 1900 Å Ag(100) foil held at ~ 800 K normalized to that obtained under similar conditions in the backscattering geometry for 100 eV incident positrons. The error bars reflect statistical uncertainties only.

entrance surface when implanted with 100 eV incident energy. The latter parameter can be set equal to unity. As $\beta_{e+}(E)$ for Ag is measured to be nearly constant in the energy range of 0.5–30 keV, and as $\beta_{Ps}(E)$ in the same energy range is shown to be a small fraction of this (Baker and Coleman 1988), the assumption is made that the sum $[\beta_{e+}(E)+\beta_{Ps}(E)]$ is always a constant, and hence according to (3) $R(E) = Y_T(E)$. At energies below 500 eV Baker and Coleman (1988) have found $\beta_{e+}(E)$ falling gradually though this decrease will be offset by an increase in $\beta_{Ps}(E)$ (Howell 1988) such that this assumption is still realistic. Furthermore, over the incident e^+ energy range in which transmission Ps is observed from the 1900 Å foil (~ 5–15 keV) little error is introduced if $\beta_{e+}(E)$ is taken to be a constant and $\beta_{Ps}(E)$ is set to zero. Thus from equation (3) the pre-factor containing the β parameters is a constant and thus any error in our original assumption will not lead to the introduction of energy-dependent systematic errors in $R(E)$. It should also be emphasized that since the thermally activated yield is assessed by a (hot–cold) difference the 100 eV measurement is not sensitive to the presence of epithermal Ps. Thus the data shown in figure 2 can be considered as a measure of the variation of the transmission probability of thermalized positrons with incident positron energy at a given temperature (here ~ 800 K).

The transmission probabilities of themalized positrons, $Y_T(E)$, can be modelled using the solution for the one-dimensional (z) diffusion equation for a thin foil of thickness d (e.g. Chen *et al* 1985)

$$Y_T(E) = \frac{1}{\sinh(T/L_+)} \int_0^d \sinh(z/L_+)P(z)\,dz.$$

Here L_+ is the effective e^+ diffusion length and $P(z)$ is the implantation profile for a certain incident energy given by

$$P(z) = \frac{mz^{m-1}[\Gamma(1+m^{-1})]^m}{\bar{z}^m} \exp\left[-\left(\Gamma\left(1+\frac{1}{m}\right)\left(\frac{z}{\bar{z}}\right)\right)^m\right]$$

where m is the Makhovian shape parameter and $\bar{z}$, the mean implantation depth, is taken to be

$$\bar{z} = \frac{\alpha}{\rho}(E[\text{keV}])^n$$

where ρ is the density of the material, and α and n are fitting parameters. The full line in figure 2 is calculated using this solution with the fitted parameters $L_+ = 425$ Å, $n = 1.7$, $m = 2.4$ and $\alpha = 4.4$ μg cm^{-2}.

The value of $L_+ = 425$ Å is low compared to the bulk value measured by Soininen *et al* (1990) which, based on their measured temperature dependence, scales to $L_+ \approx 800$ Å at 800 K. We believe this can be due to defects in the foil produced as a result of thermal stress arising from a temperature gradient between the foil and the Ag rings sandwiching this. Also the low annealing temperature ($\sim$ 800 K) used in the present study when compared to higher temperatures feasible in bulk studies may not be sufficiently high to remove defects within the foil.

The extracted values of n and m differ from the more conventional values ($n = 1.6$, $m = 1.9$). However these latter values are based on studies of semi-infinite materials and do not as such apply to thin foils. A similar discrepancy with accepted values has been observed for thin foils by Chen *et al* (1985) and in new and substantial measurements and theoretical calculations on thin films by Baker *et al* (1991).

In the backscattering geometry at 800 K Lynn and Welch (1980) deduced a 43% conversion of 36 eV incident positrons into thermally activated Ps and this constituted 71% of the total Ps emission. This work also implied that $\epsilon_{Ps} < 0$. Our measurements, shown in figure 2, give the transmission probability of a 1900 Å foil to be 22% at 8 keV at 800 K foil temperature when compared to the backscattering geometry. Thus the absolute conversion efficiency in transmission, F, is

$$F = 0.22 \times 0.84 \times [0.43/(0.71 + f) + f] \qquad 0 \leq f \leq 0.29$$

$$\Downarrow$$

$$0.11 \leq F \leq 0.13$$

where the additional correction factor of 0.84 is taken from the work of Baker and Coleman (1988) and allows for the epithermal backscattering of e^+ from the foil, and f accounts for the Ps which can originate from the prompt emission of thermalized positrons.

An attempt was also made to estimate the effect on our measured transmission probabilities of different L_+ in the foil at the two different temperatures by using the temperature dependence of L_+ observed by Soininen *et al* (1990). By appropriately scaling the ratio of the thermally activated and promptly emitted Ps at 800 K from Lynn and Welch (1980) it was found that our estimated transmission probabilities should be multiplied by 1.10. This was due to an over subtraction of the promptly emitted transmission Ps in using the (hot–cold) method previously outlined. Due to the many uncertainties inherent in this argument, not least of which is the large difference in L_+ found for the foil and the bulk sample, this correction was not applied to our final values of $Y_T(E)$ (and F). It should be noted though that if the scaled values of $Y_T(E)$ are used the fitting parameters previously deduced are not altered except for L_+ which increases slightly to 475 Å.

3.2. Estimate of the velocity distribution of thermally activated positronium

The count rate of ortho-Ps was monitored through a 6 mm slit opening as a function of the distance from the silver foil. The difference between hot and cold spectra was taken as a relative measure of the amount of thermally activated Ps annihilating within

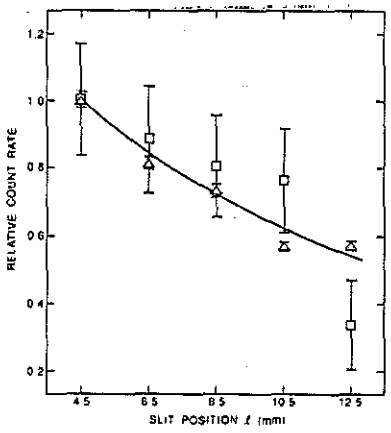

Figure 3. The spatial annihilation distribution of thermally activated Ps as a function of the perpendicular distance from the silver foil. The values are normalized to that obtained nearest the foil. Key: open triangle, 100 eV e^+ on a 1900 Å foil in backscattering; open square, 7 keV e^+ on a 1500 Å foil in transmission. The full line is a fit to the backscattering data based on (4) (see text for details).

the slit opening. Additionally the background at each slit position was measured by monitoring the HPGe count rate with the moderator at −100 V.

These measurements were done both in the backscattering geometry with a 100 eV incident beam on a 1900 Å foil and in the transmission geometry with a 7 keV beam on a 1500 Å foil. Figure 3 shows these results. Here the yield of thermally activated Ps is shown relative to the value for the slit position nearest the foil. The full line is a calculated fit to the backscattering data and will be described later. It should be noted that the experiment only yields information concerning the distribution of Ps velocities perpendicular to the surface, and therefore the measured spatial distribution of events is analysed by inferring a one-dimensional velocity distribution. The count rate, CR, in a slit opening, due to self-annihilation of ortho-Ps, is then

$$CR \propto \int_0^\infty \mathrm{d}v_z \int_{t_1}^{t_2} \mathrm{d}t\, \lambda \mathrm{e}^{-\lambda t} C(v_z). \tag{4}$$

Here t_1 and t_2 are, respectively, the times taken by a Ps atom of velocity v_z to reach the entrance and the exit of the slit, λ is the annihilation rate of ortho-Ps in vacuum and $C(v_z)$ is the velocity distribution perpendicular to the foil.

This distribution was taken to be of the Maxwell–Boltzmann form multiplied by a fitting parameter, namely

$$C(v_z) \propto A(v_z)\mathrm{e}^{-mv_z^2/kT} \tag{5}$$

where the factor, $A(v_z)$, can be expressed as a polynomial in v_z as suggested by Mills and Pfeiffer (1979), and m is the electron mass.

The full line in figure 3 is a calculation of (4) with the slit distance axis shifted 0.5 mm downwards as indicated in figure 1(*b*). The best fit of the data was found with $A(v_z)$ proportional to $1 + (v_z/v_0)^2$ (as it was in the study of Mills and Pfeiffer 1979) with $v_0 = 3(2) \times 10^4$ m s^{-1}. The temperature obtained by this fit was 800±100

K in agreement with the value measured in the manner previously described. The transmission data were not fitted because of their larger statistical and systematic uncertainties. The latter type of uncertainty was caused by the foil accidentally starting to evaporate during the measurements.

We note finally that the one-dimensional velocity distribution extracted from these data is in contradiction with the model of Platzman and Tzoar (1986), where the Ps is considered to be physisorbed to the metal surface prior to thermal activation though a more detailed study of the behaviour of positrons and positronium at surfaces is necessary before this model can be fully tested.

4. Conclusion

Thermal activation of Ps from thin Ag(100) films has been investigated, and it has been shown that conversion can be achieved in the transmission geometry to form a source of Ps atoms suitable for scattering experiments. The cost of this is the non-unity transmission probability shown in figure 2 and the non-unity Ps formation probability at the surface due to the lower maximum operating temperature of the foils compared to thick crystals. However these inefficiencies can be compensated for by easier interfacing with other experimental apparatus and, in particular, that to be used for the formation of antihydrogen in collisions of Ps with antiprotons (Deutch *et al* 1986, 1988). Furthermore the velocity distribution of the desorbed Ps has been fitted and found to be non-Maxwellian and of the same form as that suggested by Mills and Pfeiffer (1979).

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