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Measurement of the positronium formation potential for Ag(100)

M R Poulsen†, M Charlton and G Laricchia

Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK

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Abstract. Using a time-of-flight technique the velocity of positronium (Ps) emitted from Ag(100) has been studied. The maximum velocity of Ps, arising from implanted positrons which have thermalized and diffused back to the emitting surface, has been determined yielding a formation potential for Ps of $\epsilon_{Ps} = -1.5 \pm 0.2$ eV.

1. Introduction

Positronium (Ps) can be produced in vacuum by bombarding a metal surface with low-energy positrons (e^+) (Canter *et al* 1974). Its formation may be enhanced by increasing the temperature of the metal or by suitably treating the surface (Gidley *et al* 1988, Mills *et al* 1991). The excess Ps is associated with the desorption of e^+ trapped at the surface potential and has been characterized, for various metals, by studies of yields of such thermally activated Ps as a function of temperature (e.g. Mills 1979, Lynn 1979, Lynn and Welch 1980) together with velocity distributions perpendicular to the metal surface (Mills and Pfeiffer 1979, 1985, Poulsen *et al* 1991). These distributions have been found to be near-thermal and to depend on the temperature of the metal surface.

On encountering the surface region a thermalized e^+ may capture an electron and be emitted as Ps into vacuum if its formation potential, ϵ_{Ps}, is negative. The latter is simply determined by energy balance as

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

where ϕ_+ and ϕ_- are the positron and electron workfunctions, respectively, and 6.8 eV is the Ps ground state binding energy in vacuum. Measurements of the velocity distributions for various metals (e.g. Mills *et al* 1983, 1989, Howell *et al* 1987) have shown that Ps 'promptly' emitted perpendicularly to the surface does so with a distribution of energies extending from zero to $-\epsilon_{Ps}$. It has thus been suggested (Mills *et al* 1983) that this route to Ps formation is non-adiabatic with the metal being left in a single electron–hole excited state in the conduction band. The model, which calculates the energy distribution of Ps emitted into a cone of emission angles has been found to explain reasonably well the kinetic energy distributions measured by Mills *et al* (1983, 1989), Chen *et al* (1987) and Howell *et al* (1987).

Lang and Kohn (1970) have shown that the workfunction can be separated into contributions from the bulk (chemical potential) and the surface (dipole potential). The latter, however, is equal in magnitude but opposite in sign for the positron and electron

† Present address: Institute of Physics and Astronomy, Aarhus University, DK-8000 Aarhus C., Denmark.

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(Tong 1972). Hence ϵ_{Ps} is independent of the metal surface conditions and determined only by bulk properties. Since the surface dipole is more difficult to evaluate theoretically than the bulk chemical potential, ϵ_{Ps} is more amenable to calculations than either ϕ_+ or ϕ_- indicating that comparisons between experiment and theory may be more reliable in the case of Ps formation potentials.

In the present study prompt emission of Ps from a Ag(100) sample has been investigated. There are three theoretical estimates of ϵ_{Ps} for silver, namely the early studies of Hodges and Stott (1973) and Nieminen and Hodges (1976) which, when using $\phi_- = 4.64$ eV (Dweydari and Mee 1975), predicted values of $\epsilon_{Ps} = -1.89$ eV and $\epsilon_{Ps} = 0.24$ eV, respectively and a later calculation by Boev *et al* (1987) which found $\epsilon_{Ps} = -1.44$ eV. The latter is believed to be more accurate as this was determined by calculating the bulk chemical potentials for both the positron and the electron while the two earlier studies both calculated ϕ_+ where ϵ_{Ps} can then be extracted by known values of ϕ_-. Evidence that ϵ_{Ps} is indeed negative as implied by Hodges and Stott (1973) and Boev *et al* (1987) has come from a study of Ps yields from a Ag(100) sample as a function of temperature and e^+ implantation energy (Lynn and Welch 1980). This work found a Ps yield of $\simeq$13% at room temperature for 5 keV incident e^+ which cannot be accounted for by epithermal Ps emission since the e^+ backscattering coefficient at this energy is measured for silver to be $\simeq$22% (Coleman 1992) and, from the work of Howell *et al* (1986), less than 5% of the backscattered e^+ would be expected to leave the surface as Ps.

The present study is not a test of the model of Mills *et al* (1983) as this requires the ability to measure the kinetic energy distribution of the Ps emitted perpendicularly to the surface from the data. This can be done either by inserting a collimator in order to limit the angle of Ps emission towards normality (Mills *et al* 1983, 1989, Howell *et al* 1987) or by the deconvolution technique of 2D ACAR (Chen *et al* 1987). The aim of this work was, in conjunction with a previous study of thermally activated Ps (Poulsen *et al* 1991), to give a more complete characterization of the Ps produced at a Ag(100) surface since this has been suggested as the host metal for conversion of e^+ into a Ps gas target to be interfaced with an antiproton beam for the production of antihydrogen (Deutch *et al* 1988). As the target density depends on the atomic velocity, the distribution perpendicular to the sample surface of all Ps emitted was investigated.

2. Apparatus

The present study was performed with a low-energy e^+ beam in a UHV system with a base pressure of 2–4 $\times$ 10^{-10} Torr. Approximately 10^4 slow e^+ per second were obtained from a 4 mCi ^{22}Na source in conjunction with a tungsten mesh moderator. The moderated e^+ were accelerated to 1.5 keV and transported to the interaction region in a magnetic confinement field of 50 G provided by an array of Helmholtz coils, as shown in figure 1. A fraction of the e^+ were timed by detecting, using a channel electron multiplier (CEM), the secondary electrons liberated by the e^+ upon striking the sample. The CEM was positioned off the beam axis, perpendicular to and in front of the sample (see inset of figure 1) and was mounted in an earthed stainless steel box (hereafter termed timing-box). The timing-box had an enclosure in front of the cone of the CEM through which the incoming e^+ beam and the emitted Ps atoms had access obstructed only by a 95% transmission Cu mesh. The purpose of the Cu mesh was to confine the field lines produced by the potential applied to the cone of the CEM (+500 V) to the enclosed volume in front of the CEM cone and the sample.

The sample, a $\simeq$100 μm thick, 8 $\times$ 10 mm Ag(100) crystal, was mounted in a holder

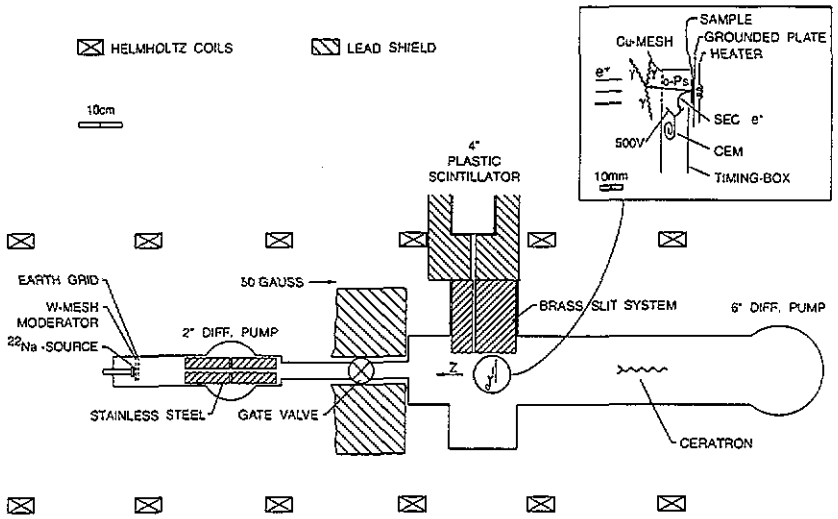

Figure 1. Schematic arrangement of the magnetically guided e^+ beam. Inset: timing-box containing a channel electron multiplier used for detecting secondary electrons liberated by incoming positrons.

attached to a 3D manipulator. The holder contained a tungsten filament used for annealing the sample and thermocouples to measure the annealing temperature. Also attached to the sample holder was a plate with a 10 mm aperture which could be inserted into the beam to ensure correct alignment of the beam with respect to the sample. In this case the beam was monitored by a ceramic electron multiplier located, as shown in figure 1, further down the beam line. During both the beam location and annealing, the timing-box, which was attached to a separate rotary drive manipulator, was removed.

The relatively high impact energy of 1.5 keV was chosen firstly to avoid the beam being significantly deflected by the cone potential and hence not striking the sample, and secondly to ensure that the majority of the e^+ returning back to the surface, where the Ps formation occurs, were thermalized. This avoided substantial emission of energetic Ps arising from epithermal e^+ (Howell *et al* 1986) whilst ensuring that a large fraction of the implanted e^+ could still diffuse back to the surface. The fraction of e^+ emitted as Ps into vacuum could be monitored by a 125 mm $\times$ 100 mm plastic annihilation γ-ray detector. The signal from this detector was routed in a conventional delayed coincidence circuit with that from the CEM, which, by registering the arrival of a e^+ to the sample, signalled the possible birth of a Ps atom. A lifetime spectrum was thus obtained. An example is shown in figure 2 which exhibits a narrow peak and a delayed exponential component with a mean lifetime of 124 ns. The narrow peak arises from e^+ annihilating in the bulk of the sample (mean lifetime of 138 ps (MacKenzie *et al* 1975)) or from a surface state (expected mean lifetime roughly 500 ps (Lynn *et al* 1984, Nieminen and Puska 1983)) and from para-Ps, the singlet ground state of Ps (para-Ps has a mean vacuum lifetime of 125 ps). The width of the peak (12 ns) gives the overall resolution involved in timing the e^+ and detecting a prompt γ-ray whilst the position of the peak gives the location of time zero. The long-lived component in the spectrum is due to the triplet ground state of Ps (ortho-Ps) annihilating in vacuum. Collisions with the chamber and/or timing-box walls would decrease the mean free vacuum lifetime of 142 ns. The presence of ortho-Ps was also monitored by measurements of the energy of the annihilation γ-rays with a NaI(Tl) detector using the peak-to-valley parameter

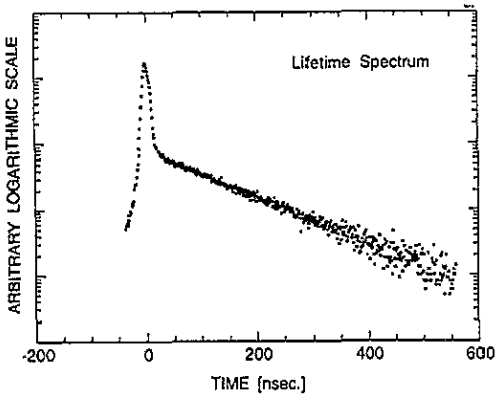

Figure 2. Decay of ortho-Ps yielding a lifetime of 124 ns (see text for details).

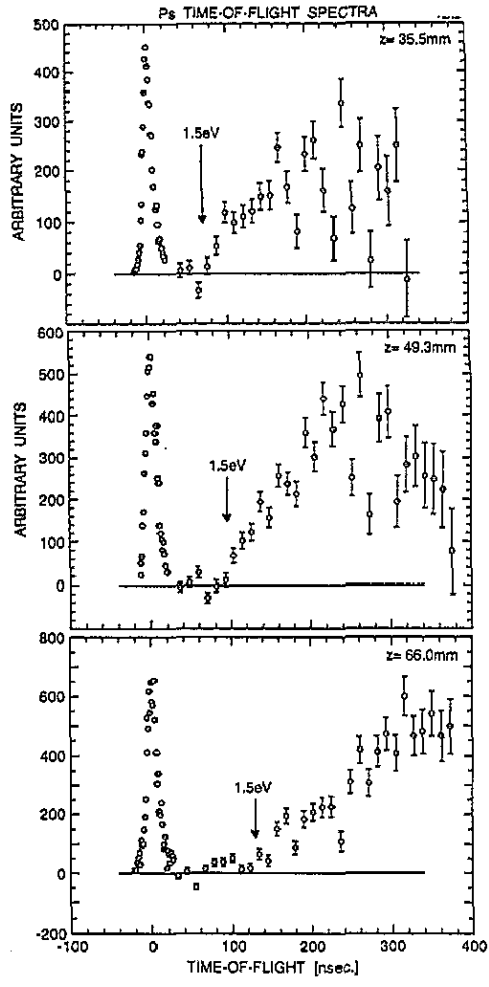

Figure 3. Time-of-flight spectra of ortho-Ps obtained for three (indicated) distances from the sample to the opening of the slit. A kinetic energy of 1.5 eV is indicated in each spectrum.

(Marder *et al* 1956). This allowed the stability of the Ps yield to be checked routinely. It was found to be constant for at least 24 h following cleaning of the sample by heating it to $\simeq$550 °C for a period of $\simeq$20 min. This procedure was repeated daily during a sequence of measurements, each of which could take several days.

To measure ϵ_{Ps} a slit system was incorporated in front of the γ-ray detector with the purpose of defining a volume of view for the detector into the vacuum chamber at a known distance from the sample as shown in figure 1. The slit system consisted of an array of 155 mm long brass shutters followed by 4 in of collimating lead bricks. The position of the sample with respect to the slit system was measured by mapping the coincidence count rate between the CEM and the γ-ray detector through a 1.5 mm slit opening as a function of the position of this. The resolution of the slit system was determined for a 4.8 mm slit opening by measuring the count rate of 511 keV γ-rays in the γ-detector while moving the sample across the gap. This yielded a width of 6.4 mm FWHM.

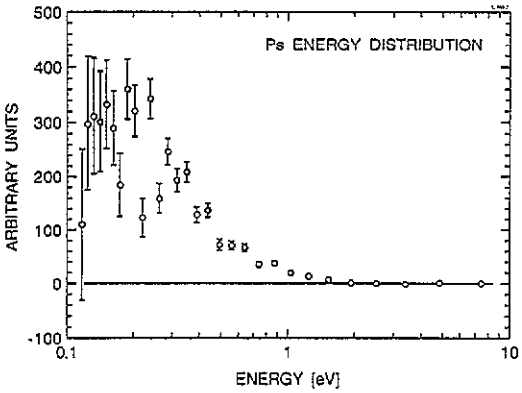

Figure 4. Energy distribution of the emitted ortho-Ps extracted from the measurement with $z = 49.3$ mm.

3. Results and discussion

Time-of-flight data have been obtained for three different slit openings with distances, z, from the sample to the entrance of the gaps of 35.5 mm, 49.3 mm and 66.0 mm and widths of 4.8 mm, 9.4 mm and 13.2 mm, respectively. The data were recorded in channels of 1.14 ns which were later added into 10 channel bins for improved statistics. The spectra all exhibit a narrow peak at zero time. These are, as mentioned above, due to e^+ and para-Ps annihilating in or at the sample surface, though now the annihilation γ-rays are scattered through the slit opening as the sample is not seen directly by the γ-detector. At later times the detection of ortho-Ps annihilating in front of the slit window is seen as an increase in the coincidence count rate.

With reference to figure 3, the spectra shown have undergone the following corrections. A time-independent, random background evaluated at long times, has been subtracted from all the data points in each spectra. Then the data, apart from the peak at zero time, were corrected for the decay of ortho-Ps by multiplying each channel by $\exp(t/(142\text{ ns}))$, where t is the time evaluated from the peak at time zero, thus correcting for the decrease in apparent Ps signal with time. This results, along with a distribution arising from Ps atoms annihilating in front of the slit, in a flat background due to ortho-Ps annihilating outside the view of the detector but where one of the γ-rays scatters through the slit opening. This background, evaluated between the time zero peak and the Ps time-of-flight distribution, was subtracted in the spectra shown in figure 3.

All Ps time-of-flight spectra exhibit an onset at short times which moves towards longer times with increasing distance to the entrance to the slit opening. This onset is correlated to the maximum kinetic energy of the emitted Ps arising from thermalized e^+. From all the spectra the maximum kinetic energy, or $-\epsilon_{Ps}$, is evaluated according to $E = m_e z^2/t^2$, where m_e is the electron rest mass. This yields an estimate of the formation potential of $\epsilon_{Ps} = -1.5 \pm 0.2$ eV, where the main uncertainty is that incurred in determining the onset. In each case the uncertainty is taken to be ± 11.4 ns, the size of a 10 channel bin, which yields an overall combined error of 0.2 eV. This estimate agrees with the value of -1.44 eV found from the calculation of Boev *et al* (1987).

As mentioned previously ϵ_{Ps} is only dependent on the bulk properties of the metal and therefore independent of the surface conditions such as direction and small doses of contaminants. The latter may however, in each individual case, alter both the Ps yield and the Ps velocity distribution. So although the Ps yield, ensured by regular cleaning of the

surface, was found to be stable throughout the measurements, ϵ_{Ps} is the only information intrinsic to the host metal to be extracted from the data since no tools for surface analysis were available for this study. With the only extractable information being ϵ_{Ps} and the poor statistics in some of the data we also chose to present the spectra in figure 3 without a l/t correction which compensates for the velocity-dependent time spent by the Ps atoms in front of the slit opening. This correction, however, was applied before the data were transformed, for illustration, into an energy distribution. The transformation was done by multiplying the data by t^3 and converting to an energy scale as shown in figure 4 for the spectrum taken with $z = 49.3$ mm. The value of z used to calculate the energy was 49.3 mm + (9.4 mm)/2 = 54.0 mm as this is the centre of the slit opening.

4. Conclusion

Prompt emission of Ps from Ag(100) has been studied using a time-of-flight technique. The positronium formation potential has been determined to be $\epsilon_{Ps} = -1.5 \pm 0.2$ eV, in agreement with the calculation of Boev *et al* (1987).

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