First observation of positronium negative ions emitted from tungsten surfaces

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Abstract. Spontaneous emission of positronium negative ions from polycrystalline tungsten surfaces has been observed for the first time. It is a new channel for the fate of thermalized positrons near the surface. The obtained formation probability is \((7 \pm 2) \times 10^{-5}\). This method provides a new source of monoenergetic positronium negative ions for future applications.

When low-energy positrons impinge on metal surfaces, there is a significant probability that they will penetrate into the bulk, lose their energy until thermalized, and then diffuse back to the surface where there are several channels for the fate of the positrons [1]–[4]. The positrons may be trapped in the surface potential well and annihilate there. If the positron work function \(\phi_+\) is negative, the positrons may be emitted with a characteristic energy \(|\phi_+|\). The positrons may be also emitted as positronium (Ps) atoms with a maximum energy governed by the Ps formation potential \(\phi_{Ps} = \phi_+ + \phi_- - 6.8\,\text{eV}\), where \(\phi_-\) is the electron work function. Furthermore, the emission of excited Ps has also been observed [5]. In the present study, we have successfully observed another channel, that of emission as a Ps negative ion (Ps\(^-\)), which is a bound state of a positron and two electrons.

Ps\(^-\) were first observed by Mills [6] in a study using the beam-foil method. In this experiment, a beam of slow positrons was guided to a thin carbon target whereupon some travelled through the target and removed two electrons from the surface to produce Ps\(^-\). A grid, located downstream of the target, was biased positively with respect to the target in order to accelerate the Ps\(^-\) which were subsequently detected by their Doppler-shifted annihilation lines.

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Table 1. Experimental values of $\phi_+$ and $\phi_-$. Values of $\phi_{Ps^-}$ estimated using equation (1) are also listed.

| Element                  | $\phi_+$ (eV) | $\phi_-$ (eV) | $\phi_{Ps^-}$ (eV) |
|--------------------------|----------------|---------------|-------------------|
| W (1 0 0)                | -3.0 [17]      | 4.63 [18]     | -0.9              |
| W (1 1 0)                | -3.0 [19]      | 5.22 [18]     | 0.3               |
| W (1 1 1)                | -2.59 [10]     | 4.47 [10]     | -0.79 [10]        |
| W (polycrystalline)      | -3 [2]         | 4.55 [18]     | -1                |
| Mo (1 0 0)               | -1.7 [2]       | 4.53 [18]     | 0.2               |
| Mo (polycrystalline)     | -2.2 [20]      | 4.6 [18]      | -0.1              |

The conversion efficiency of the Ps$^-$ production was found to be $2.8 \times 10^{-4}$. Mills also measured the Ps$^-$ decay rate using the same method [7]. Recently, Fleischer et al [8, 9] produced Ps$^-$ using a diamond-like carbon foil and measured the Ps$^-$ decay rate more precisely.

Ps$^-$ may be produced from thermalized positrons at metal surfaces and emitted if the Ps$^-$ formation potential, $\phi_{Ps^-}$, expressed as

$$\phi_{Ps^-} = \phi_+ + 2\phi_- - E_B,$$

is negative, where $E_B$ is the binding energy of Ps$^-$, the energy required for it to break-up into an isolated positron and two electrons [10]. The value of $E_B$ has been calculated theoretically by several authors [11]–[16] to be 7.13 eV. The experimental values of $\phi_+$ and $\phi_-$ for tungsten and molybdenum are listed in table 1 together with the estimated values of $\phi_{Ps^-}$ using equation (1). Table 1 shows the process of Ps$^-$ emission is energetically allowed from tungsten (1 0 0), (1 1 1) and polycrystalline surfaces and from molybdenum polycrystalline surface. Wilson and Mills [10] have attempted to observe the emission of Ps$^-$ from tungsten (1 1 1) surfaces and set an upper limit of 0.1% for the Ps$^-$ branching ratio. In the present study, we have successfully detected Ps$^-$ emission to confirm the existence of this channel for polycrystalline tungsten surfaces.

The experimental system used was a magnetically guided slow positron beam apparatus with a trochoidal $E \times B$ filter. Positrons from a 20 mCi $^{22}$Na positron source were moderated in an electropolished tungsten mesh moderator [21] and transported to the target chamber. The beam intensity was $9 \times 10^4$ e$^+$ s$^{-1}$ and the transport energy was 0.1 keV.

Figure 1 shows schematically the target chamber used in this experiment. The target was biased at a voltage $-W$, so that the positrons were incident on the target with an energy of $eW + 0.1$ keV, where $e$ is the charge of the positron. A grid maintained at earth was placed at a distance $d$ in front of the target so that Ps$^-$ emitted from the surface would be accelerated by the potential difference $W$. The distance $d$ was set at 3 mm. A Ge detector was used to monitor the annihilation $\gamma$-rays of the oncoming Ps$^-$ which would be blue-shifted. As the count rate from Ps$^-$ emission events was low, the background was reduced by using the Ge detector signal in coincidence with that from the NaI scintillation detector placed behind the target. The $\gamma$-ray energy was calibrated using annihilation $\gamma$-rays emitted from positron annihilation in the bulk of the target measured for a positron transport energy of 30 keV. The energy resolution of the Ge detector was measured using 512 keV $\gamma$-rays from a $^{106}$Ru source and was found to be 1.1 keV. The base pressure of the target chamber was $7 \times 10^{-8}$ Pa which was attained using a turbo-molecular pump and a getter pump.

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The targets used were commercially obtained polycrystalline tungsten of thickness 15 µm (99.95% purity). They were annealed in situ at 1500°C by passing electric current through them in order to reduce bulk defects, which act as positron traps, and to clean the target surfaces [22, 23]. The temperature was raised, maintained for 30 min then lowered to room temperature over a period of 10 min. A spectrum was also taken using an unannealed target. All measurements were performed at room temperature.

Figure 2 shows the annihilation photon energy spectra for \( W = 1 \) and 3 kV. The spectrum for the unannealed target was obtained over a duration of 20 h. Those for the annealed targets were taken for a period of 35 h after the annealing process.

No significant peak of blue-shifted photons was observed for the unannealed target. For the annealed targets, a low-intensity peak was seen which appeared to move with \( W \).

The energy shift of the Ps\(^-\) annihilation photons, \( \Delta E \), emitted at an angle \( \theta \) from the direction of the Ps\(^-\) velocity is given by

\[
\Delta E = \frac{-\lambda + \sqrt{2\lambda + \lambda^2 \cos \theta}}{1 + \lambda - \sqrt{2\lambda + \lambda^2 \cos \theta}} mc^2,
\]

(2)

where \( \lambda = eW/3mc^2 \), \( m \) is the mass of the positron and \( c \) is the speed of light. This equation is different from that of equation (1) in [6], where the angle \( \theta \) is defined in the centre-of-mass system of the positron. When \( \theta = 0 \), as in the case of [6], then both equations are equal. The angle \( \theta \) of the present experiment was estimated to be 18° from the geometry of the target and the detectors. The arrows in figure 2 indicate the energies corresponding to the Doppler shift predicted by equation (2). The good agreement between the observed and predicted energy shifts shows it is likely that the shifted peaks can be attributed to the detection of Doppler-shifted \( \gamma \)-rays from the 2-\( \gamma \) annihilation of Ps\(^-\) which was formed at the target surface.
Figure 2. The annihilation photon energy spectra for unannealed and annealed tungsten polycrystalline targets: (a) unannealed target with \( W = 1 \) kV, (b) annealed target with \( W = 1 \) kV, (c) annealed target with \( W = 3 \) kV and (d) the logarithmic plot of (c). The arrows indicate the energies of the Ps\(^-\) annihilation \( \gamma \)-rays estimated using equation (2).

The annihilation photon energy spectra from a molybdenum polycrystalline target, annealed at 1500\(^\circ\), was also taken. However, no significant peak of the energy corresponding to Ps\(^-\) was detected.

In order to obtain the fraction \( f \) of positrons yielding Ps\(^-\) at the tungsten surfaces, the shifted peaks in the spectra were fitted using a Gaussian function with a correction applied to
account for annihilation in the acceleration region. The fraction of the Ps$^-$ emerging from the acceleration region before annihilation, $\zeta$, is given by [6]

$$\zeta \approx \exp\left(-\frac{3}{\Gamma_1} \frac{\Delta E d}{\epsilon W c}\right),$$

where $\Gamma$ is the Ps$^-$ annihilation rate, which has been determined experimentally to be 2.09 ns$^{-1}$ [9]. The corrected values of $f$ obtained over a period of 35 h following annealing were $(7 \pm 2) \times 10^{-5}$ and $(6.1 \pm 0.9) \times 10^{-5}$ for $W = 1$ and 3 kV, respectively.

We have measured the fractions of $f$ for several targets of polycrystalline tungsten and have found that the values depended on the targets largely. This may be due to the surface condition of the targets or crystal directions of the grains which consisted the polycrystal. Carbon contamination accumulating on the surface of the target [24] may kill the Ps$^-$ emission [10]. If we use carbon-free surfaces obtained by heating in oxygen, the yield might be improved. Using tungsten single crystals might also improve the yields since these have higher positron diffusion constants than polycrystalline tungsten.

The observed Ps$^-$ ions were formed from thermalized positrons or epithermal positrons. If the Ps$^-$ were formed from epithermal positrons, the formation probability would depend on the positron incident energy since more epithermal positrons would reach the surface at lower energies. However, the obtained fractions $f$ were found not be dependent on the target bias. This indicates that most of the Ps$^-$ were formed from thermalized positrons. The full-width-at-half-maximum (FWHM) of the Gaussian function fitted to the Ps$^-$ peak for $W = 3$ kV was $(1.5 \pm 0.2)$ keV, which is narrower than that of the Ps$^-$ formed by the beam-foil method of Mills [6, 7]. The narrowness of the Ps$^-$ peak too might indicate that the Ps$^-$ was formed from thermalized positrons. It is to be noted that the present method will provide a ‘monoenergetic’ Ps$^-$ beam source, which can be used for future Ps$^-$ studies such as the measurement of the Doppler broadening of annihilation $\gamma$-rays from Ps$^-$, which has been estimated theoretically [25], or the study of Ps$^-$ photo-detachment [6, 26].

The spectrum for $W = 3$ keV was taken for a period of 63 h after annealing and divided into four time regions in order to study time dependence. Figure 3 shows that the values of $f$
decreased with time after annealing. This is due to adsorbate coverage of the target surface by residual molecules in the target chamber.

The positron and electron work functions can be written using the corresponding chemical potentials $\mu_+$ and $\mu_-$ as follows

$$\phi_+ = -D - \mu_+,$$
$$\phi_- = D - \mu_-,$$  \hspace{2cm} (4)

where $D$ is the surface dipole barrier [27]. Therefore, the $\text{Ps}^-$ formation potential can be written as

$$\phi_{\text{Ps}^-} = -\mu_+ - 2\mu_- - E_B + D.$$  \hspace{2cm} (5)

This equation indicates that $\phi_{\text{Ps}^-}$ is dependent on $D$, i.e., the $\text{Ps}^-$ formation on metal surfaces depends on the coverage on the target surfaces. Wilson and Mills [10] suggested that the $\text{Ps}^-$ formation on the tungsten surface is more energetically favourable if the electron work function is reduced through the coverage by residual molecules in the target chamber. We expect that it should be possible to reduce the electron work function by using an alkali coverage and thereby increase the $\text{Ps}^-$ emission probability.

In conclusion, we have successfully observed the spontaneous $\text{Ps}^-$ emission from polycrystalline tungsten surfaces. This method will provide a new source of monoenergetic $\text{Ps}^-$ beam for future $\text{Ps}^-$ studies.

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