Efficient emission of positronium negative ions from Cs deposited W(100) surfaces

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Abstract. The emission of positronium negative ions from Cs deposited W(100) surfaces has been studied. A dramatic change in the emission efficiency was observed upon coating a W(100) surface with Cs. The conversion efficiency (the fraction of incident slow positrons yielding the ions) of the W(100) target with a $2.2 \times 10^{14}$ atoms cm$^{-2}$ Cs coverage, measured over a time interval of $3 \times 10^3$ s immediately after deposition, was found to be 1.25%, which is two orders of magnitude higher than that obtained for the clean, uncoated W(100) surface and 45 times greater than the highest efficiency reported thus far.

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The existence of several kinds of exotic systems that are composed of three particles with equal masses and bound through Coulomb interaction, has been proposed theoretically [1]. The positronium negative ion (e\(^{-}\)e\(^{+}\)e\(^{-}\), Ps\(^{-}\)) is the only system that has been observed so far. After the first observation by Mills [2] in 1981, where a conversion efficiency (the fraction of incident slow positrons yielding Ps\(^{-}\)) of 0.028% was obtained using the beam–foil method, no experimental studies have been performed on this system except for only a few measurements of the decay rate [3]–[5]. In order to accomplish more extensive investigations into the fundamental processes, formation methods producing higher efficiencies must be developed.

Recently, we have observed the spontaneous emission of Ps\(^{-}\) from polycrystalline tungsten and tungsten(100) surfaces bombarded with slow positrons [6]–[9]. The conversion efficiencies were in the range 0.006–0.012%, and were lower than that obtained using the beam–foil method of Mills [2]. In the present work, we observed a dramatic change in the efficiency upon depositing Cs onto a W(100) surface.

When slow positrons impinge on metal targets, positronium (Ps) atoms are produced from thermalized positrons and emitted from the surfaces spontaneously because the Ps formation potential, \(\phi_{Ps}\), expressed as
\[
\phi_{Ps} = \phi_{+} + \phi_{-} - 6.8 \text{ eV},
\]
is negative, where \(\phi_{+}\) and \(\phi_{-}\) are the positron and electron work functions, respectively. Ps\(^{-}\) ions are also produced and emitted if the Ps\(^{-}\) formation potential, \(\phi_{Ps^{-}}\), given by
\[
\phi_{Ps^{-}} = \phi_{+} + 2\phi_{-} - E_B,
\]
is negative [6, 10], where \(E_B\) is the binding energy of Ps\(^{-}\), the energy required to break it up into an isolated positron and two electrons (\(~7.13 \text{ eV}\)), which has been calculated precisely [11, 12]. The estimated values of \(\phi_{Ps^{-}}\) for W(100) and (111) surfaces, as well as polycrystalline W surfaces, are negative; studies have confirmed the emission of Ps\(^{-}\) ions from polycrystalline W and W(100) surfaces [6]–[9].

The positron and electron work functions can be written using the corresponding chemical potentials, \(\mu_{+}\) and \(\mu_{-}\), as follows
\[
\phi_{+} = -\mu_{+} - \Delta, \quad \phi_{-} = -\mu_{-} + \Delta,
\]
respectively, where \(\Delta\) is the surface dipole barrier [13]. Therefore, the Ps\(^{-}\) formation potential can be written as
\[
\phi_{Ps^{-}} = -\mu_{+} - 2\mu_{-} - E_B + \Delta.
\]
This equation indicates that \(\phi_{Ps^{-}}\) is dependent on \(\Delta\), i.e. the emission of Ps\(^{-}\) from metal surfaces is dependent on the adsorbate coverage on the target surfaces [6]. An oxygen overlayer on a tungsten target increases the value of \(\Delta\) and therefore suppresses Ps\(^{-}\) emission. On the other hand, an alkali-metal coverage decreases \(\Delta\) because the surface dipole is depolarized by the adsorbate dipoles created [14]–[16], and thus the Ps\(^{-}\) emission efficiency is likely to increase. In the present study, we have successfully observed the emission of Ps\(^{-}\) from a Cs coated W(100) surface with an efficiency of 1.25%.
Figure 1. Schematic diagram of the target chamber used for observing the Ps\(^-\) emitted from Cs deposited W(100) surfaces.

2. Experimental procedure

The experimental system used was a magnetically guided slow positron beam apparatus with a trochoidal \( E \times B \) filter. The beam intensity was \( 1.3 \times 10^5 \) e\(^+\) s\(^{-1}\) and the transport energy was 0.1 keV. The beam diameter was about 5 mm.

Figure 1 shows schematically the target chamber used in this experiment. The chamber was similar to that used in the previous works \[6\]–[9], with the exception of a change in the direction of the Ge detector axis and the addition of a Cs deposition system. The head of the detector was placed in a re-entrant tube of wall thickness 1 mm, which was positioned at an angle of 107° to the beam line. The \( \gamma \)-ray detection efficiency was found to improve by 70% over the previous setup \[6\]. The base pressure of the target chamber was \( 2.4 \times 10^{-8} \) Pa.

The target employed was a W(100) foil, purchased from Aarhus University, of dimensions 10 mm \( \times \) 10 mm \( \times \) 2 \( \mu \)m. It was supported on a backing of polycrystalline W foil 25 \( \mu \)m thick and annealed in situ at 1800 K for 30 min by the passage of an electric current. Heating in oxygen, which is known to be effective in reducing surface carbon \[17\], was not performed in the present experiment because a thick oxide layer is formed which inhibits Ps\(^-\) emission \[9\].

The Cs layer was produced using a Cs dispenser purchased from SAES Getter S.p.A.. The thickness of the layer was set to be \( 1.7 \times 10^{14} \) atoms cm\(^{-2}\), \( 2.2 \times 10^{14} \) atoms cm\(^{-2}\) and \( 1.0 \times 10^{15} \) atoms cm\(^{-2}\). (One physical monolayer of Cs coverage on a W(100) surface corresponds to \( 2.8 \times 10^{14} \) atoms cm\(^{-2}\) \[15\].) Just prior to the Cs deposition, the W sample was resistively heated at 1800 K for 5 min in order to clean the surface. Then, Cs coverage of the W(100) surface was performed where the thickness of the layer on the target was controlled by the deposition time. The relationship between the coverage and the deposition time was calibrated using a quartz crystal dispenser monitor at the target position before and after the Ps\(^-\) measurements. The uncertainty in the thickness of the Cs layer was estimated to be about 30%.
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. An earthed grid with 78% transmission was located in front of the target so that an electric field could be applied to accelerate the emitted $\text{Ps}^-$. The grid was mounted on a linear transfer in order to change the distance between it and the target, which, in this experiment, was set at 3 mm. The target potential applied, $-W$, was $-3$ kV.

The Doppler-shifted $\gamma$-rays emitted from $\text{Ps}^-$ self-annihilation were monitored by the Ge detector in coincidence with the signal from the NaI scintillation detector placed behind the target. The measurements were performed at room temperature.

3. Results and discussion

Figure 2 shows the annihilation photon energy spectra for the uncoated W(100) target accumulated for $1.9 \times 10^5$ s before and after annealing, and those accumulated for $4 \times 10^4$ s after Cs deposition. The energy of the $\text{Ps}^-$ annihilation $\gamma$-rays emitted at an angle $\theta$ from the direction of the $\text{Ps}^-$ momentum, $E$, is given by

$$E = \frac{1}{1 + \frac{\lambda}{\sqrt{2\lambda + \lambda^2}} \cos \theta} mc^2,$$

where $\lambda = eW/3mc^2$, $m$ is the rest mass of the positron and $c$ the speed of light. The value of $E$ for the present experimental condition, calculated using equation (5) is 542 keV.

For the uncoated W(100) after annealing, a small peak due to $\text{Ps}^-$ is observed at this energy. The conversion efficiency, $f$, was obtained by fitting the peak using a Gaussian function with corrections applied to account for annihilation in the acceleration region [6] and the transmission efficiency of the earthed grid in front of the target. The value of $f$ for the uncoated W(100) after annealing was 0.005%, which is lower than that obtained using a different W(100) sample in a previous experiment [9].

For the targets with Cs coverage, the peak intensity increased dramatically. The efficiencies, $f$, measured over a time interval of $4 \times 10^4$ s for the $1.7 \times 10^{14}$ atoms cm$^{-2}$, $2.2 \times 10^{14}$ atoms cm$^{-2}$ and $1.0 \times 10^{15}$ atoms cm$^{-2}$ covered targets were 0.064%, 0.83% and 0.085%, respectively. The highest value was obtained for a coverage of $2.2 \times 10^{14}$ atoms cm$^{-2}$, where the value of $\phi^+$ has been reported to be the lowest [16]. It is two orders of magnitude higher than that obtained for the annealed, uncoated W(100) sample.

The increase in the efficiency upon Cs deposition can be interpreted from the viewpoint of work function changes as follows. If we assume that the $\text{Ps}^-$ is formed from positrons in the bottom of the positron band, then

$$-\phi^+ + \epsilon_1 + \epsilon_2 + E_B > 0$$

holds, where $\epsilon_1$ and $\epsilon_2$ are the energy levels of two electrons which contribute to the formation of $\text{Ps}^-$ measured from the vacuum level. Hence, $f$ can be written as

$$f \propto \int_{\phi^+ - \epsilon_1 - E_B}^{0} D(\epsilon_1) d\epsilon_1 \int_{\phi^+ - \epsilon_2 - E_B}^{0} D(\epsilon_2) d\epsilon_2,$$

where $D(\epsilon)$ is the electron density of states. If we assume that the $D(\epsilon)$ is constant below the Fermi energy, then

$$f \propto (-\phi^+ - 2\phi^- + E_B)^2.$$

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The values of $\phi_+$ and $\phi_-$ for a clean, uncoated W(100) surface are reported to be $-3.0 \text{ eV}$ [18] and $4.63 \text{ eV}$ [19], respectively. For a W(100) surface with $2.2 \times 10^{14} \text{ atoms cm}^{-2}$ Cs coverage, the value of $\phi_-$ is shifted by $-3.10 \text{ eV}$ [15] and $\phi_+$ is shifted by $3.10 \text{ eV}$ according to equation (3). The estimated value of $f$ for the Cs deposited W(100) is 21 times as high as that for the clean, uncoated W(100). This interpretation gives not only a rough estimate but indicates that the effect of Cs deposition is large because two electrons are involved. However, this estimate is still too small to explain the experimental results. The Ps$^-$ formation mechanism should also be taken into account. The formation depends on the overlap of a
Figure 3. Ps\textsuperscript{−} emission efficiency from the target with a $2.2 \times 10^{14}$ atoms cm\textsuperscript{−2} Cs coverage plotted against time after Cs deposition.

The positron wave function and two electron wave functions just outside the surface, which, in turn, depend on their corresponding work functions, as in the case of Ps formation [20]. Therefore, the Ps\textsuperscript{−} formation mechanism should be affected by the Cs deposition. The electron work function for Cs deposited W(100) surface is reported to decrease with Cs coverage steeply in the region below $2.2 \times 10^{14}$ atoms cm\textsuperscript{−2} and then increase gradually [16]. The fact that the efficiency for the $2.2 \times 10^{14}$ atoms cm\textsuperscript{−2} coverage is one order of magnitude higher than those for the $1.7 \times 10^{14}$ atoms cm\textsuperscript{−2} and $1.0 \times 10^{15}$ atoms cm\textsuperscript{−2} coverages is consistent with the work function changes qualitatively.

The background levels on the lower energy side of the Ps\textsuperscript{−} peaks for the Cs deposited targets are higher than those on the higher energy side. The difference is attributed to the Ps\textsuperscript{−} that have self-annihilated into $2\gamma$-rays in the space between the target and the grid. It is also to be noted that the shape of the 511 keV peak is asymmetrical for the annealed W(100) and Cs deposited targets while that for the unannealed target is symmetrical. The asymmetry is due to the self-annihilation of para-Ps emitted from the target with energies of a few electronvolt, governed by equation (1).

The values of $f$ at each point of time after Cs deposition for the $2.2 \times 10^{14}$ atoms cm\textsuperscript{−2} coverage is shown in figure 3. The mean efficiency, over a $3 \times 10^{3}$ s interval after the Cs deposition, was 1.25%, which is 45 times as high as the conversion efficiency reported thus far [2]. The value decreased and was almost constant at about 0.1% efficiency after $6 \times 10^{4}$ s, which is still higher than that obtained using the beam–foil method. The decrease might be due to the accumulation of residual oxygen in the target chamber onto the target surface and the structural change of the Cs layer on the W(100) surface. If the vacuum in the target chamber is improved, the high efficiency may remain for longer. In order to investigate in more detail, the Ps\textsuperscript{−} measurement should be combined with other analytical methods for the target surface, for example, positron annihilation induced Auger electron spectroscopy (PAES) [21].

The efficiency obtained in the present work might be sufficient to enable more thorough investigations of, for example, precision measurements of the Ps\textsuperscript{−} decay rate and the

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Ps− binding energy. We are now planning to obtain the Ps− binding energy by measuring the values of $\phi_+$, $\phi_-$ and $\phi_{Ps−}$ simultaneously. Additionally, the fact that the emission process is affected by the surface condition means that Ps− ions might be used in future applications as a sensitive probe of metal surfaces.

4. Conclusions

In conclusion, we have found that the Ps− emission efficiency has been greatly improved by the deposition of Cs onto a W(100) surface. The method might be applied in order to realize more extensive investigations of the fundamental processes of Ps− but also as a useful tool in studies of metal surfaces.

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