Increase in the positronium emission yield from polycrystalline tungsten surfaces by sodium coating

Hiroki Terabe a,*, Shimpei lida a, Takashi Yamashita a, Takayuki Tachibana b, Bernardo Barbrellini c, Ken Wada d, Izumi Mochizukid, Akira Yagishitad, Toshio Hyodod, Yasuyuki Nagashimaa, and Hiroki Terabeb

a Department of Physics, Tokyo University of Science, 1-3 Kagurazaka, Shinjuku, Tokyo 162-8601, Japan
b Department of Physics, Rikkyo University, 3-34-1 Nishi-Ikebukuro, Toshima, Tokyo 171-8501, Japan
c Department of Physics, Northeastern University, Boston, MA 02115, USA
d Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK), 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan

* Corresponding authors.
E-mail addresses: 1211706@alumni.tus.ac.jp (H. Terabe), ynaga@rs.kagu.tus.ac.jp (Y. Nagashima).

ARTICLE INFO

Article history:
Received 25 March 2015
Accepted 13 May 2015
Available online 22 May 2015

Keywords:
Positron
Positronium
Time-of-flight
Sodium coated tungsten

ABSTRACT

The study of positronium emission from metal surfaces bombarded by slow positrons provides information on the topmost layer of the metals such as electron and positron energy levels because positronium atoms are formed as a result of the interactions between the positrons and the electrons there. In the present work, time-of-flight spectra of ortho-positronium atoms emitted from polycrystalline tungsten surfaces with and without a sodium coating have been measured. The data shows a significant increase on coating in the yield of the 5 eV component due to positronium formed from thermalized positrons and conduction electrons. An attempt is made to explain the increase by an emission model based on the formation of positronium in a low electron density surface layer extended by the coating.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

When low-energy positrons impinge on metals, they penetrate into the bulk, lose their energy until thermalized, and then some diffuse back to the surface [1,2]. If the positron work function, $\phi_p$, is negative, a proportion of the positrons is emitted with the characteristic energy $|\phi_p|$. Emission as positronium (Ps) is also an allowed process from most metals since $\phi_p$, which is expresses as

$$\phi_{Ps} = \phi_p + \phi_e - E_{Ps} \quad (1)$$

and referred to as the Ps affinity in this paper, is negative in most cases. Here $\phi_e$ is the electron work function and $E_{Ps}$ is the Ps binding energy in vacuum (5.8 eV). The energy distribution of the Ps emitted via this process reflects that of the conduction band of the metals and has a maximum energy at $|\phi_{Ps}|$.

Some of the positrons are also trapped in the surface potential well and annihilate there. On application of heat to the metal, Ps yields increase due to the thermal desorption of Ps atoms formed from such positrons [3]. The activation energy of this process, $E_a$, is written as

$$E_a = E_b + \phi_e - E_{Ps} \quad (2)$$

where $E_b$ is the binding energy of the positron at the surface. An increase in the yield of Ps atoms emitted from a Si surface has also been observed when the surface was coated with Cs [1]. This increase was interpreted as the desorption of Ps formed from positrons trapped in the surface potential well with electrons whose work function had been lowered by the coating [1]. Thermal desorption of Ps from Cs coated Ni surfaces was reported for temperatures down to 325 K and cryogenic desorption of Ps by this technique was predicted [4]. However, direct measurements of Ps energy distributions from surfaces with and without a coating have not been performed.

In the present work, we have measured the time-of-flight (TOF) spectra of ortho-Ps emitted from a Na coated tungsten surface in order to investigate the effect of Na coating on the Ps emission. The obtained data showed an increase in the Ps component governed by Eq. (1). We have developed a model to explain the increase based on the change in the electron density distribution at the surface.

2. Experimental procedures

TOF measurements for the Ps emitted from a polycrystalline tungsten surface with and without a Na coating were performed using a pulsed slow positron beam at the Slow Positron Facility (SPF) in the Institute of Materials Structure Science (IMSS), High Energy Accelerator Research Organization (KEK) [9]. Positrons were generated at a beam dump of a 55 MeV electron linac and moderated using a tungsten...
The pulse width was 10 ns and the beam intensity was $5 \times 10^6$ e+/s. The repetition rate of the linac was 50 Hz. The positrons were transported to the Ps–TOF measurement chamber following acceleration to an energy of 4.2 keV, which was high enough to prevent pulse widening. The beam was collimated using an aluminum plate with an aperture of 9 mm in diameter before entering the Ps–TOF chamber.

The schematic diagram of the Ps–TOF chamber is illustrated in Fig. 1. The beam diameter at the target was measured to be about 10 mm using a microchannel plate with a phosphor screen. The base pressure of the chamber was $6 \times 10^{-8}$ Pa.

The target was a polycrystalline tungsten foil of 25 μm thick with dimension of about 12 mm × 12 mm. It was annealed in situ at 1800 K for 30 min by the passage of an electric current and, after cooling to room temperature, Na was deposited onto it using dispensers purchased from SAES Getters S.p.A. The thickness of the layer was estimated by a deposition monitor system using a quartz crystal sensor. The deposition was set at $6 \times 10^{14}$ atoms/cm$^2$, where the electron work function has been reported to be at its lowest (2.1 eV)\[10,11\]. The target was grounded electrically during the TOF measurements and hence the positron incident energy onto the sample was 4.2 keV.

Time intervals between the linac pulse signals and the detection of $\gamma$-rays from in-flight self-annihilation of the emitted ortho-Ps in view of the narrow slits were measured. Two identical plastic scintillation detectors were used of size 100 mm × 100 mm × 10 mm, and these were coupled to photomultiplier tubes (HAMAMATSU H6614) via light pipes. They were encased in lead shielding with narrow slits to allow the passage of the annihilation $\gamma$-rays and were placed by the chamber. The distances between the sample and the slits along the positron beam line, d, were 40 mm and 120 mm, and the slit widths were 2 mm and 6 mm, respectively. The signals from these photomultiplier tubes were recorded with a digital oscilloscope (Lecroy WaveRunner 64Xi-A). The time range of the oscilloscope was set to be 1 μs and the sampling rate was 500 MS/s. The waveforms were digitized and stored with the trigger pulses from the linac. After acquisition, the data was analyzed off-line. The signals with pulse heights higher than a predetermined threshold were selected and the counts for two adjacent bins were summed and plotted as the TOF spectra.

### 3. Results

Fig. 2 shows the Ps–TOF spectra from the polycrystalline tungsten sample: unannealed, annealed, and Na-coated after annealing. The spectra were normalized to the measurement time. The prompt peaks at time zero are attributed to the annihilation of the positrons in the target and self-annihilation of para-Ps (singlet $^1S_0$ state, short-lived) and positronium negative ions ($e^- e^+ e^-$, Ps$^-$)\[12–15\].

For the surface after annealing, the counts of the ortho-Ps (triplet $^3S_1$ state, long-lived) component appeared after 40 ns and 130 ns for (a) $d = 40$ mm and (b) $d = 120$ mm, respectively, both of which correspond to a Ps emission energy of 5 eV. This energy agrees with the maximum Ps emission energy calculated of 5.2 eV using Eq. (1) together with reported values of $\phi^-$ and $\phi^+$ for polycrystalline tungsten\[16\]. The shapes are determined from the electron band structure of the sample, the angular distribution of Ps emission, and the geometrical resolution of the
detector system. The Ps emission angle may be broader than that of single crystal, where positrons are emitted with a narrow angular distribution [17,18]. Moreover, some of the emitted Ps may bounce off the chamber wall and contribute to the shape. However, the experimental resolution of the present experimental setup is not sufficient to clearly discern these factors.

After Na coating, the yield of the emitted Ps increased significantly. Furthermore, the Ps energy distribution was observed to be narrower than that for the uncoated surface.

4. Discussion

The present data shows that the maximum Ps emission energy is not affected by the Na coating and is consistent with the fact that the Ps affinity, $\phi_{ps}$, should be unchanged upon coating. The electron and positron work functions for the coated surface, $\phi_e$ and $\phi_p$, respectively, are written as

$$\phi_e' = \phi_e - \Delta$$

and

$$\phi_p' = \phi_p + \Delta,$$  

(3)

(4)

where $\Delta$ is the effect of the coating [1,2]. Hence $\phi_{ps}$ calculated using Eq. (1) to be unchanged. These relations have been studied using two-component density functional theory simulations for Li coated Al(100) recently [19].

The data also shows an increase upon Na coating in the intensity of this component. The Ps emission model suggested for Si surfaces [1], where Ps atoms formed from positrons trapped in a potential well at the surface and electrons whose work function is lowered by the coating are desorbed, thus does not account for the energy distribution observed in the present work.

The value of the negative positron work function approaches zero upon coating and thus the positron re-emission probability becomes lower. This may contribute to the relative increase in the Ps emission probability. It is also possible that the increase of the surface region with a low conduction electron density favors Ps formation.

Ps formation in materials is explained using the electron gas density parameter, $r_s$, expressed as

$$r_s = \left( \frac{3}{4\pi n} \right)^{1/3} \alpha_0,$$  

(5)

where $n$ is the conduction electron density and $\alpha_0$ is the Bohr radius. Held and Kahana [20] made a variational calculation for an electron–positron pair in electron gases and found that Ps appears when $r_s > 8.5$. Kanazawa et al [21] calculated the bound state including short range positron–electron interaction and concluded that Ps is not formed in the range of $r_s$ corresponding to all the actual metals. Weisberg and Berk [22] measured the positron annihilation rates in metals and plotted them against $r_s$. They found that the rates for the nearly free electron metals are on a smooth curve which approaches the spin-averaged Ps self-annihilation rate, $\lambda_{pos}$. All the rates are larger than $\lambda_{pos}$ even in the case of alkali metals, where the values of $r_s$ are larger than other metals. According to the calculation of the electron–positron bound states in electron gases by Lowy and Jackson [23], there can be no Ps for $r_s > 6.2$. Furthermore, Barbiellini and Platzman [24] determined the Ps binding energy in electron gases, $E_{Ps}$, to be

$$E_{Ps}^{calc} = E_{Ps}^{calc} \frac{r_s^2}{r_s^2 + 12.5}.$$  

(6)

When $r_s > 6$, $E_{Ps}^{calc}$ is larger than the electron–positron correlation energy of the free positrons and hence Ps states are stable, as in the case of Ps lying on surfaces [25] or in some insulators [26,27].

In the case of metal surfaces, $r_s$ is dependent on $x$, the coordinate of the axis perpendicular to the surface. As positrons, thermalized in the bulk, approach the surface, they carry with them correlated cloud of electrons and Ps atoms appear at $x > x_{rs} = 6$. If we assume that the Ps emission yield, $Y_{Ps}$, is dependent on the electron capture cross section $\sigma(n(x))$ which depends on the conduction electron density $n(x)$, then

$$Y_{Ps} = \int_{-\infty}^{\infty} \sigma(n(x)) n(x) dx.$$  

(7)

Further assuming that $\sigma(n(x))$ is a step function, i.e.,

$$\sigma(n(x)) = \begin{cases} 0 & x < x_{rs} = 6, \\ \alpha_0 & x > x_{rs} = 6, \end{cases}$$

where $\alpha_0$ is a constant, then the Ps yield can be described by

$$Y_{Ps} = \int_{x_{rs} = 6}^{\infty} n(x) dx.$$  

(8)

We applied this model to the case of bare tungsten and fully Na coated tungsten surfaces, using the relevant values of $n(x)$ from Fig. 1 of [28] although the Na layer of this figure is thicker than the present experimental setup. The estimated increase in the Ps yield was found to be 1.4 times, which is too small to account for the observations of the present experiment. We have also calculated $n(x)$ for the present thickness using an analytical model [29] and obtained the increase to be 1.6 times. If we assume that the value of $\alpha$ is proportional to the Ps binding energy in the electron gas given by Eq. (6), the estimated increase is 2.2 times, which is still smaller than the experimental result. These discrepancies could be attributed to a possible lack of accuracy in the tail part of the electron density distribution; we actually compared the area below $n(x)/n_{sub} = 1/27$, where $n_{sub}$ is the electron density in the bulk. The above assumption may be too simplistic and more detailed calculations for $n(x)$ and $\sigma$ are needed. The present approach should also clarify the mechanism of Ps emission from clean metal surfaces, which is not yet satisfactorily understood. An increase in the yield of Ps emitted from metal surfaces upon alkali-metal coating observed recently [12–15] may also be explained by a similar process.

5. Conclusion

In conclusion, we have investigated Ps emission from Na coated polycrystalline tungsten surfaces. The data shows that the maximum Ps emission energy is independent of the coating and the intensity of the Ps formed from thermalized positrons and conduction electrons predominantly increases upon coating. It is thought that this increase reflects the change in the conduction electron density near the surfaces.

Acknowledgments

We thank K. Michishio, K. Watanabe and S. Hagiwara for valuable discussions. We also thank T. Shidara, S. Osawa, M. Ikeda, and other KEK staff for their generous support for the positron beam line. This work has been performed under the approval of the Photon Factory Program Advisory Committee (Proposal No. 2012G666 and 2013S-005). It was supported by JSPS KAKENHI Grant Number 24221006 and a Grant-in-Aid for JSPS Fellows (No. 25-8190). B.B. was supported by the U.S. Department of Energy (USDOE) Contract No. DE-FG0207ER46352.
References