Measurement of the positron diffusion constants in polycrystalline molybdenum by the observation of positronium negative ions

Takuji Suzuki, Simpei Iida, Takashi Yamashita and Yasuyuki Nagashima

Department of Physics, Tokyo University of Science, 1-3 Kagurazaka, Shinjuku, Tokyo, 162-8601, Japan
E-mail: 1213705@ed.tus.ac.jp and ynaga@rs.kagu.tus.ac.jp

Abstract. We have measured the positron diffusion constants in polycrystalline molybdenum by the observation of positronium negative ions ($\text{Ps}^-$). The $\text{Ps}^-$ ions emitted from the sample surface coated with Na were accelerated. The $\gamma$-rays from the accelerated $\text{Ps}^-$ ions were Doppler-shifted and thus the signals of self-annihilation of the $\text{Ps}^-$ ions were isolated from those of self-annihilation of para-positronium ($\text{p-Ps}$) or pair-annihilation of positrons in the bulk. Clear and reliable values of the diffusion constants have been obtained.

1. Introduction

Recently, we developed a method to investigate positron diffusion in metals by the observation of positronium negative ions ($\text{Ps}^-$) [1]. This method has been enabled by the recent studies of efficient emission of $\text{Ps}^-$ ions from alkali-metal coated surfaces [2–5]. The $\gamma$-rays emitted from self-annihilation of accelerated $\text{Ps}^-$ ions are Doppler-shifted [2] and the $\text{Ps}^-$ component can be isolated easily in the $\gamma$-ray energy spectrum. Thus reliable studies of positron diffusion are available. In the present work, we have applied this method for the measurement of the positron diffusion constants in polycrystalline molybdenum.

2. Experiment

The experimental system used was the same as that in the previous work [1]. The base pressure of the sample chamber was $4 \times 10^{-8}$ Pa.

The sample was a polycrystalline molybdenum foil of 25 $\mu$m thickness. Its purity was 99.95%. It was annealed in situ at 1500 K or 1800 K for 30 min by the passage of an electric current. After the sample was cooled down to room temperature, the surface was coated with Na to increase $\text{Ps}^-$ emission efficiency. The deposition source was an alkali-metal dispenser purchased from SAES Getters S.p.A. The thickness measured by the quartz crystal oscillation deposition monitor was $9 \times 10^{14}$ atoms cm$^{-2}$.

The sample was biased at $-1$ kV. Slow positrons were incident onto the sample after passing through a grounded grid located 1 mm in front of the sample. The energy of the positrons incident onto the sample, $E_{\text{e}^+}$, was varied from 1 keV to 30 keV. The $\text{Ps}^-$ ions emitted from the surface of the sample were accelerated by an electric field between the sample and the grid.
Figure 1. $\gamma$-ray energy spectra at incident positron energies of 2 keV and 6 keV after annealing of the molybdenum sample at 1800 K.

Figure 2. $\text{Ps}^-$ fractions $f_{\text{Ps}^-}$ plotted against incident positron energy $E_{e^+}$.

The $\gamma$-rays from self-annihilation of the accelerated $\text{Ps}^-$ ions were detected by a Ge detector in coincidence with signals from a NaI(Tl) scintillation detector placed behind the sample to reduce the background.

At each value of $E_{e^+}$ the data were accumulated for short time intervals (150 s) and the measurements were repeated 16 times.

3. Result and discussion
The annihilation $\gamma$-ray energy spectra for $E_{e^+} = 2$ keV and 6 keV are shown in figure 1. These spectra have been normalized to the measurement time. The blue-shifted peaks due to the self-annihilation of the $\text{Ps}^-$ ions are clearly isolated from those of positron pair-annihilation in the bulk or self-annihilation of $p$-$\text{Ps}$.

The yields of $\text{Ps}^-$ ions were obtained by fitting the annihilation $\gamma$-ray spectra with the sum of a Gaussian function and a background. The yields were normalized to the number of incident positrons at each $E_{e^+}$ obtained from the count of the 511 keV-peak for the unannealed sample in order to calculate the $\text{Ps}^-$ emission efficiency, $f_{\text{Ps}^-}$.

Figure 2 shows the experimental values of $f_{\text{Ps}^-}$ as a function of $E_{e^+}$. The $\text{Ps}^-$ emission
Table 1. Positron diffusion constants and diffusion lengths for the molybdenum sample annealed at 1500 K and 1800 K.

| $T_a$ (K) | $E_0$ (keV) | $D_+$ (cm$^2$·s$^{-1}$) | $L_+$ (nm) |
|----------|-------------|-------------------------|----------|
| 1500     | 3.8±0.5     | 0.13±0.09               | 40±14    |
| 1800     | 6.0±0.5     | 0.6 ±0.4                | 87±29    |

efficiency is expressed as follows:

$$f_{\text{Ps}^-\text{(e+)}} = \frac{f_{0\text{Ps}^-}}{1 + (E_{\text{e+}}/E_0)^n},$$

where $E_0$ is the incident energy at which half of the positrons return to the surface via diffusion motion, $n$ is the power dependence coefficient of the mean positron stopping depth and $f_{0\text{Ps}^-}$ is the Ps$^-$ emission fraction. The relationship between the positron diffusion length $L_+ = \sqrt{D_+ \tau}$ and $E_0$ is expressed as follows:

$$L_+ = AE_0^n.$$

Here, $D_+$ is the positron diffusion constant, $\tau$ is the positron lifetime in the sample (121 ps) [6] and $A$ is a constant value which depends on the mass density of the sample, $\rho$, as follows [7–11]:

$$A = \frac{40 \text{ nm} \cdot \text{keV}^{-n} \cdot \text{g} \cdot \text{cm}^{-3}}{\rho}.$$

We fitted equation (1) to the experimental values for the annealing temperature, $T_a = 1500$ K and 1800 K, simultaneously. The optimized value of $n$ was 1.7±0.1. The values of $E_0$, $D_+$ and $L_+$ are listed in table 1. The dependence of $D_+$ and $L_+$ on the annealing temperature indicates that more vacancies were removed by the annealing at higher temperature. The diffusion constant for molybdenum single crystal was measured by Huomo et al. from the energy dependence of the positronium fraction emitted from the surface [12]. However the present values cannot be compared with it because the annealing temperatures were low and the sample was polycrystalline.

In the future, we are planning to measure the positron diffusion constants in single crystals of tungsten, molybdenum and other metals where efficient emission of Ps$^-$ ions has been confirmed [13, 14].

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