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Evidence of very low-energy positron reflection off tungsten surfaces

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Abstract. Two different methods of measuring the reflection coefficient of positrons impinging on tungsten foils with incident energies below the positron workfunction for tungsten have been employed to study reflection. One involves using a 1 keV incident positron energy and monitoring the build-up in annihilation signal as a function of time after a grid in front of the sample is switched to a positive potential to return re-emitted positrons to the sample. The other method involves using very low-energy incident positrons (20 eV) and observing the dying out of the annihilation signal over time after the grid is switched to a potential in excess of +20 V to reflect re-emitted positrons and switch off the primary beam. A reflection coefficient of approximately 0.55 (±0.05) is found from the first experiment and a value of 0.58 (±0.04) is derived from the second experiment.

1. Introduction

When positrons encounter a metal–vacuum interface, a proportion of the positrons may undergo reflection and be repelled away from the interface. The quantum mechanical reflection of positrons approaching the surface of a metal from within the solid is caused by the rapidly changing potentials near the surface and has been the subject of numerous studies and experiments [1–7]. Most of these studies have dealt with the positron emission yield as a function of temperature since this type of reflection increases with lower temperature. The aim of these studies was to get a better understanding of the details of the surface potential as well as to investigate the possibilities of producing brighter monoenergetic positron beams by reducing the thermal component of the energy and angular spread of the positrons.

Most metals, and particularly those of interest as moderator materials, exhibit a negative positron workfunction, φ_+ [8]. Those positrons that approach such surfaces from the bulk side and are not quantum mechanically reflected will be emitted from the surface with a perpendicular velocity corresponding to a kinetic energy of $-\varphi_+$ and thermal velocities parallel to the surface. Since the absolute value of the workfunction for many materials is several orders of magnitude larger than the thermal energy of a few tens of meV, the positrons are essentially emitted perpendicularly to the surface of the metal. This forms the basis of most moderator systems. However, when positrons approach the surface of such negative-workfunction materials from the vacuum side with an energy smaller than the absolute value of the positron workfunction for the material, a reflection off the surface potential might occur (figure 1). However, as the positron encounters the surface potential well it may get trapped there and eventually annihilate there. A fast annihilation event from a pick-off process might also occur if the positron wavefunction overlaps momentarily with

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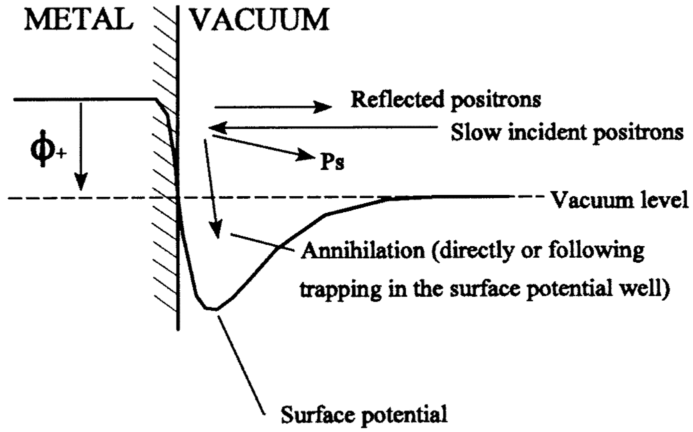

Figure 1. A potential energy diagram for slow positrons approaching a metallic surface. Also shown are the branching options open to such positrons.

that of one of the electrons making up the surface dipole. Finally the positron may bind to an electron and form positronium.

Very little research has been done to investigate this phenomenon. This is mostly due to the difficulty in producing a good positron beam with these very low energies. Wilson [9] using a low-energy incident beam and slowly raising the bias on his samples of W(111), Cr(100) and Al(100) measured the emission and possible reflection of slow positrons but found no evidence of elastic reflection of very low-energy incident positrons.

Walker *et al* [10] in a series of calculations of positron surface sticking rates, i.e. trapping at the surface potential well, found for Al(100) that the probability for trapping at the surface as a function of incident positron energy peaked around the workfunction energy at a value for Al(100) of about 0.25 eV. Such relatively large trapping rates would obviously be a strong competing channel to reflection and limit the maximum attainable reflection probability.

Our interest in very low-energy positron reflection from surfaces was piqued by some unexpected results from test experiments on the reactor-based positron beamline, POSH, currently under construction at our institute [11]. A part of the tests was studying the voltage needed to extract workfunction positrons emitted from the inside surface of tungsten foil cylinders. The tungsten foils were 7 μm thick and placed inside copper cylinders of length 12 mm and diameter 9.5 mm. Grids were placed at the ends of the cylinders and bias was applied to these grids to extract the positrons from inside the cylinders. On the basis of simulations it was expected that an extraction bias of at least 30 V would be needed to get any significant amount of positrons out. However, even with no bias applied to the grids, copious amounts of slow positrons emerged from the cylinders. With a grid potential of 0 V, about 60% of the eventual total positron yield was observed. Furthermore, retarding-field measurements showed that a grid bias of +2.5 V was needed to stop these positrons. Further simulations indicated that these results could only be explained by low-energy positrons being reflected by the opposite cylinder wall with a reflection coefficient in excess of 0.5. The experiments presented here were done to study this phenomenon of low-energy positron reflection in more detail.

2. Experimental details

The experiments were performed using the variable-energy positron beamline (VEP) at Delft. This magnetically guided beamline delivers 2×10^4 positrons per second in a 7 mm spot. The energy of the positrons is tunable in the range from 0 to 30 keV with an energy resolution of 1.7 eV FWHM. A grid placed 1.15 m in front of the sample was used to either remove low-energy positrons from the sample region or return them to the sample. The experiments fell in two parts, both of which used a sample consisting of a 7 μm thick tungsten foil similar to the ones used for the POSH. The foil was baked to 1250 °C in a separate vacuum vessel for several hours prior to installation in the UHV sample chamber. This mimicked the procedure for the foils used in the POSH set-up. We did not have any means of monitoring the surface conditions of the sample after installation.

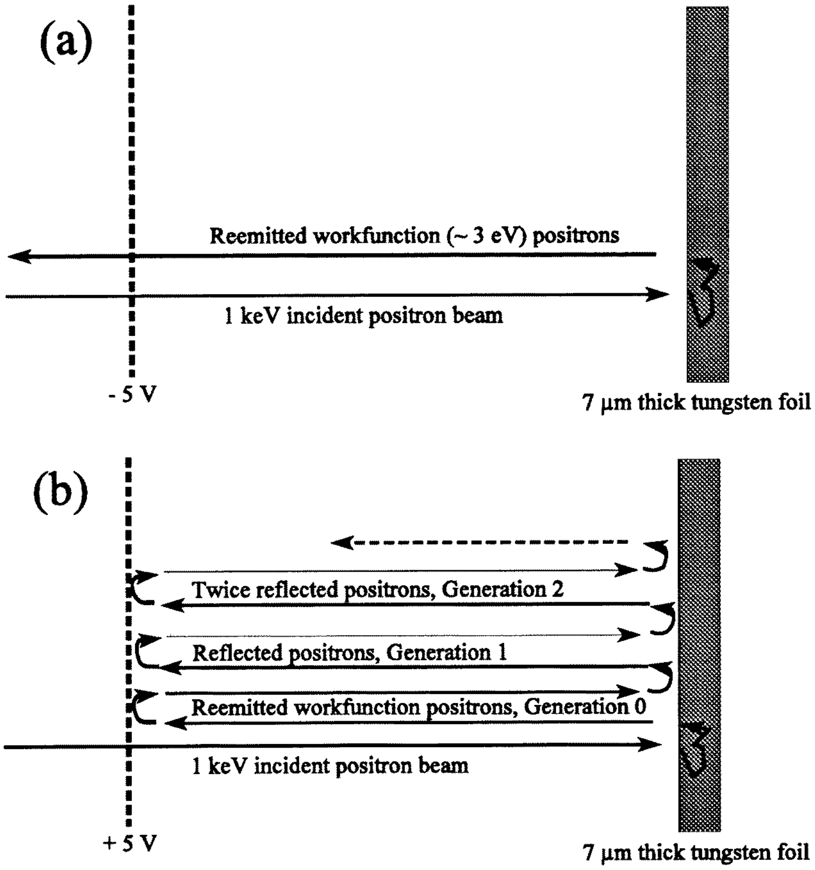

Figure 2. A schematic diagram of the experiment for 1 keV incident energy positrons with a bias on the grid in front of the sample of (a) −5 V and (b) +5 V, respectively.

2.1. The experiment with 1 keV incident energy

The first set of experiments attempted to study the build-up of the count rate when re-emitted positrons were sent back to the sample again. The idea was that the build-up would take an infinitely long time when the reflection is unity and a very short time for zero reflection. The large distance to the grid was intended to make any reflection more easily discernible. In this first experiment a 1 keV positron beam was directed at the sample. The energy was chosen to maximize the number of re-emitted thermalized positrons as opposed to epithermal ones, i.e. positrons emitted before thermalization, typically with energies up to about 10 eV. The grid was switched between +5 V and −5 V. The positrons, being re-emitted with workfunction energy, could then either be returned to the sample by applying +5 V to the grid (figure 2(b)) or dumped away from view of the detector by applying −5 V (figure 2(a)). The aim of the experiment was then to study the temporal development in the count rate as the grid was switched from −5 V to +5 V over a period of tens of microseconds.

A Philips PM 5134 Function Generator (FG) was used to generate a duty-cycle-skewed

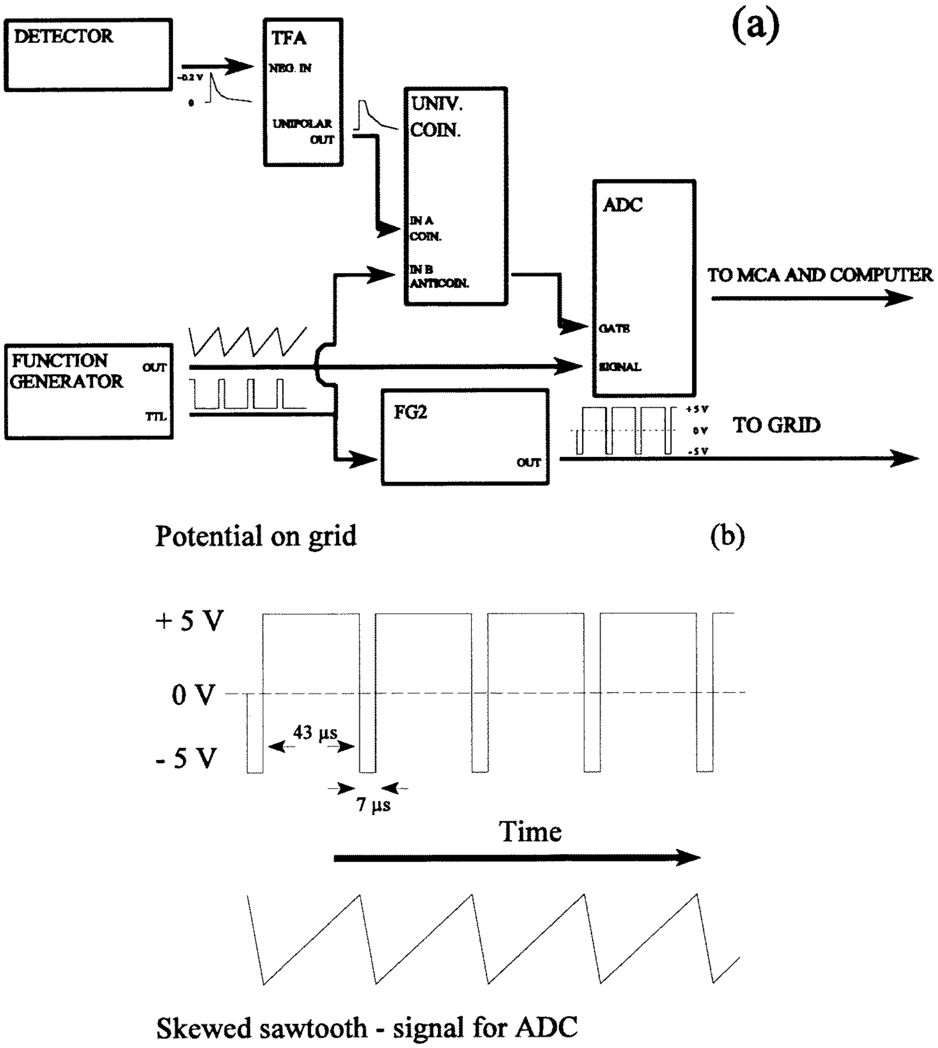

Figure 3. (a) A schematic diagram of the electronic set-up for the 1 keV incident energy experiment. (b) Detail of the time structure of the signals to the grid and computer.

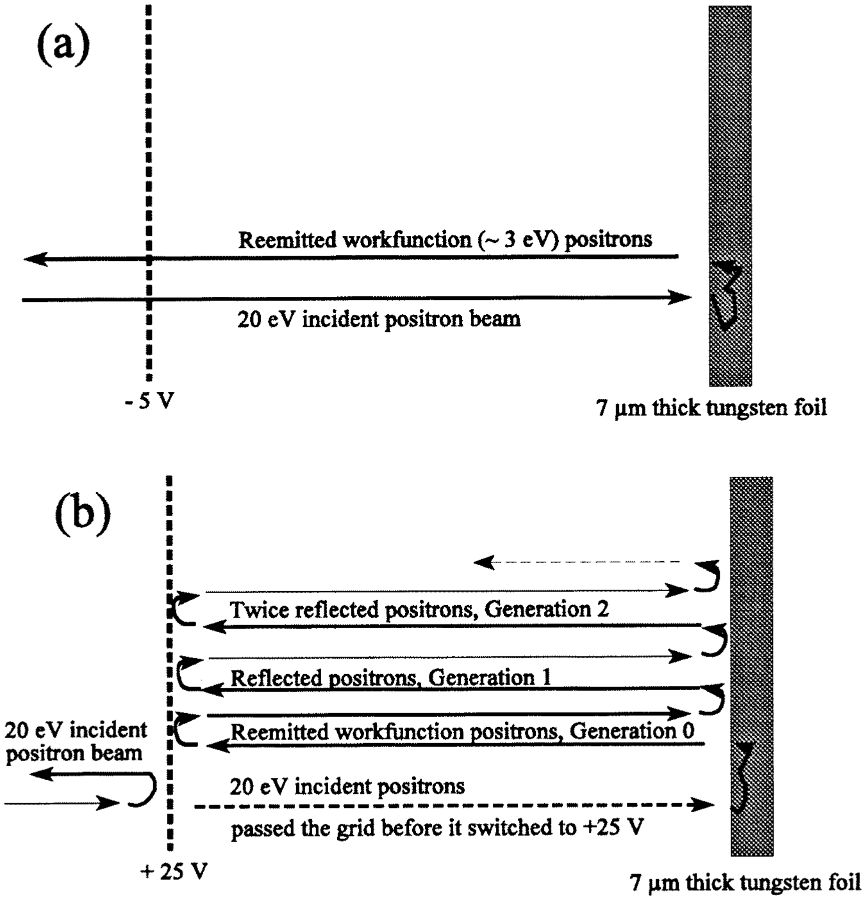

Figure 4. A schematic diagram of the experiment for 20 eV incident energy positrons with a bias on the grid in front of the sample of (a) −5 V and (b) +25 V, respectively.

sawtooth signal as a time ramp. The FG was set to a frequency of 20 kHz (=50 μs total duration of one cycle) and the duty cycle set so that the positive-slope part of the sawtooth signal had a duration of 43 μs and the negative-slope part a duration of 7 μs. The TTL 'sync.' output of the function generator was used to both switch the grid and provide a coincidence signal. The signal from the ORTEC Germanium γ-counter was fed into an ORTEC 454 Timing Filter Amplifier (TFA). The signal from the TFA was sent directly to an ORTEC 418A Universal Coincidence Unit where it was set to be in anticoincidence with the TTL signal from the FG (see figure 3(a)). The coincidence unit needs a signal of more than +2 V for at least 50 ns to trigger. The output of the coincidence unit was used as a gate in the ADC with the sawtooth signal from the FG as the ADC signal. Since the TTL signal from the FG was only 2.8 V, it could not feed the grid directly. Instead it was used to trigger a second function generator (FG2) which sent a rectangular signal of ±5 V to the grid. Figure 3(b) shows the time structure of the bias to the grid and the skewed sawtooth signal in more detail. Data were accumulated when the bias was positive. An annihilation event in the gamma counter was used as a gate in the ADC if it happened while the grid was positive. This gate would trigger the ADC to record the pulse height of the sawtooth signal at that time. This height corresponded to the time elapsed since the grid was switched to positive.

Each series of measurements consisted of four experiments:

(1) the actual coincidence measurement;
(2) a measurement with +5 V static on the grid;
(3) one with −5 V static on the grid; and
(4) a measurement where the polarity of the signal to the grid was switched.

The polarity-switched measurement worked as a kind of anticoincidence measurement where only counts accumulated when the grid bias was negative were collected. Only the bias on the grid was changed between each of the above measurements. All of the settings of the electronics stayed the same.

2.2. The experiment with 20 eV incident energy

The second half of the experimental procedure was done with a primary positron energy of 20 eV and switching the grid between −5 V and +25 V (figure 4). This was done to try to avoid the large background due to the primary positron beam. In this case we would thus try to observe the decrease in the number of annihilation events in the field of view of the detector as a function of time over a period of tens of μs. The grid bias was supplied by setting the second function generator (FG2) to its maximum output of ±15 V, giving it a DC offset of +1 V (the maximum offset was ±5 V) and adding a 9 V battery.

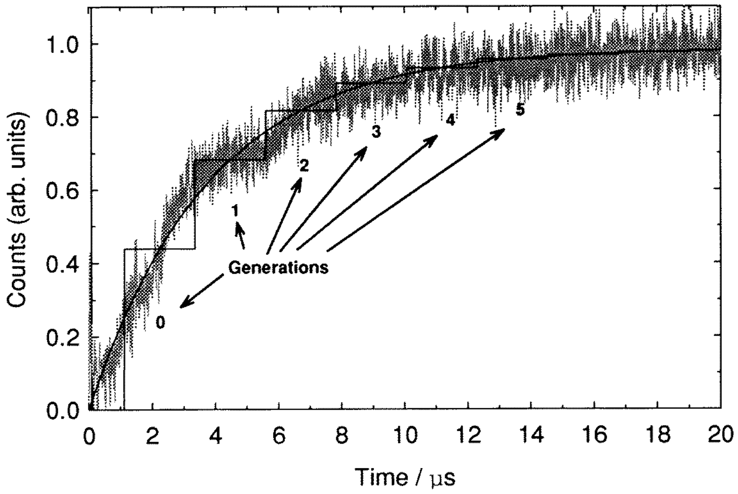

Figure 5. The time of annihilation for re-emitted positrons after activating a potential barrier (+5 V) at a distance of 1.15 m from the target. The incident energy of the primary positron beam was 1 keV, giving rise to a continuous background (the zero level). The smooth line through the data is the fit to the data. The step-function line is for the ideal case of 2.5 eV monoenergetically emitted positrons with no energy loss during reflection.

3. Results and discussion

The results of the 1 keV incident positron energy experiment are shown in figure 5. The background obtained by having −5 V static on the grid has been subtracted from the data.

If we assume a monoenergetic ‘train’ of positrons, the relative annihilation frequency after the nth reflection is given by

$$a_n = 1 - R^n$$

where R is the reflection coefficient. The step length is determined by τ, the characteristic time of the system, indicating the average time of flight for a re-emitted/reflected positron to travel the distance from the grid to the sample. A round trip would thus take 2τ. A plot of a_n is shown in figure 5. Also shown in figure 5 is the result of a fit to the data using the function

$$f(t) = 1 - \exp\left(\frac{\ln(R)}{2\tau}t\right) \tag{1}$$

where again R is the reflection coefficient and τ is the characteristic time of the system. This is a continuous function approximating equation (1). The best fit was obtained for a reflection coefficient, $R = 0.55$ (±0.05) and $\tau = 1.2$ μs. The value for τ thus corresponds to an average kinetic energy of the re-emitted positrons of about 2.5 eV travelling over a distance of 1.15 m. This is the value for τ that was also used to plot a_n. If one assumes that all re-emitted positrons have an energy of exactly 2.5 eV and no energy spread occurs, the expected result for $R = 0.55$ and $\tau = 1.2$ μs would be exactly the step-function plot of a_n shown in figure 5, where each step indicates a different ‘generation’ of reflected positrons (see figure 2). The smoother result is thus most probably caused by energy- and/or angular spread of the positrons.

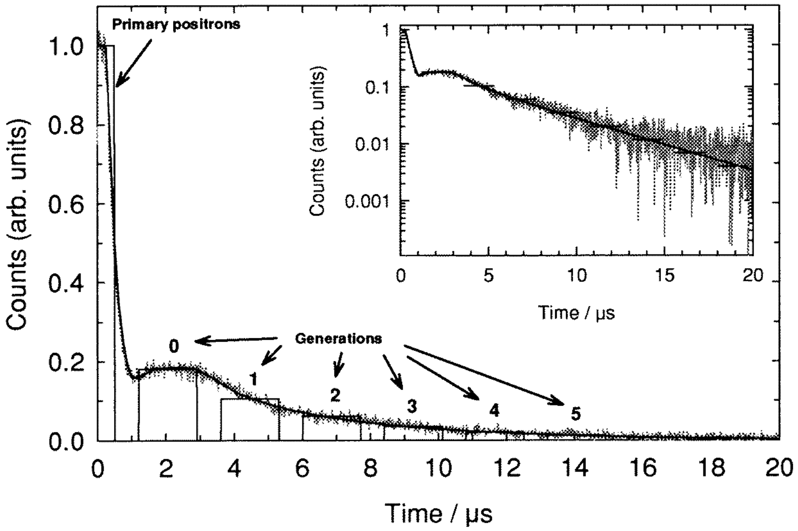

Figure 6. Time of annihilation when using an incident energy for the primary positrons of 20 eV and a potential barrier of +25 V. Now the primary beam is also cut off by the barrier but can still be seen for very short times. The smooth line through the data is the fit to the data (see the text for details). The step function line is for the ideal case of 2.5 eV monoenergetically emitted positrons with no energy loss during reflection. The inset shows the same data on a logarithmic scale.

The result of the 20 eV incident positron energy experiment is shown in figure 6. Again the data were fitted with exponentials in the different time regions and the best fit to the reflection time region (above about 3 μs; see below) was obtained with $R = 0.58$ (±0.04)

and $\tau = 1.3$ μs. This fit, as well as a step diagram assuming monoenergetic positrons, are also shown in figure 6. The smaller uncertainty on the value of R in this experiment than in the previous experiment (figure 5) is due to better statistics. The data in this experiment were accumulated over 18 hours, while the data in figure 5 were accumulated over one hour.

When the grid switches from -5 V to $+25$ V, there is still a 'train' of primary beam positrons that have passed the grid but have not yet reached the sample. That means that these positrons reach the sample within 0.5 μs ($E_{e+} = 20$ eV) after the grid has switched to positive, though with a possible tail up to 1 to 1.5 μs due to backscattering of these primary positrons. The annihilation of these positrons can be seen in figure 6 as the high count rate at very short times. The backscattered tail should be a fast-decreasing exponential. Re-emitted positrons with an energy of 2.5 eV that are approaching the grid when the bias switches will be sent back and reach the sample about 1.2 μs after the grid has switched to positive. The last positron in this 'train' of re-emitted generation-0 positrons should then reach the sample 1.7 μs later, if they all have an energy of 2.5 eV. Thus if all re-emitted positrons were monoenergetic, there would then be 0.7 μs after each generation with no positrons arriving at the sample. This is not visible in the data, except the dip between the primary beam positrons and generation 0, thus indicating that the re-emitted positrons have a rather broad energy spectrum. This was confirmed by a later retarding-field measurement and was to be expected, given that the surface of the foil was undoubtedly contaminated. That the lack of dips between the generations is due to a spread in the positron energies, and not caused by a poor time resolution of the detection system, can be seen from the fact that typical time resolutions for Ge counters are of the order of 0.1 μs and thus much too small to explain the observed spread.

The 20 eV experiment was fitted using an exponential for the drop off from the primary beam (0.3–1.0 μs). In a manner similar to the 1 keV experiment (equation (2)), the reflection time regimen (3–20 μs) was fitted using two exponential functions of the form

$$f(t) = A \exp\left(\frac{\ln(R)}{2\tau}(t-3)\right) + B \exp\left(\frac{\ln(R')}{2\tau'}(t-3)\right). \qquad (2)$$

Of these two exponentials, the dominant one had the values $R = 0.58$ (± 0.04) and $\tau = 1.3$ μs as mentioned earlier. The other exponential was fitted to values of R' and τ' of 0.40 (± 0.20) and 0.68 μs respectively. This last exponential had an amplitude of about 30% of the total number of re-emitted positrons. That this exponential is needed in order to reproduce the data can be seen from the inset in figure 6 where the data, when plotted using a logarithmic scale, only fit a straight line for times larger than about 6 μs. The value for τ' corresponds to an average positron kinetic energy of about 8 eV. We therefore conclude that the parts of the spectrum described by R' and τ' are caused by epithermally emitted positrons. The fact that such a contribution was not seen in the 1 keV experiment is ascribed to the fact that the fraction of epithermal positrons increases with lower incident energy, as the positrons are implanted closer to the surface and thus have a greater chance of being re-emitted from the surface before they are fully thermalized. The short characteristic time, τ', and low reflection coefficient, R', for the epithermal positrons make that contribution die off very quickly, so above 6 μs the reflection of the thermally emitted positrons is dominant.

The fact that the reflection did not disappear over time even after several days of experiments and the fact that it was also so clearly present in the POSH experiments, with the poorer vacuum conditions of that set-up, shows that the effect is very rugged. The heat treatment that the foils received prior to installation would cause the surface to be predominantly covered with oxide, since this would only leave the surface at a temperature of about 1600 °C [12]. There would probably also be traces of C diffusing to the surface from the bulk, and maybe N, but the main surface contaminant is thought to be oxide. Such a coverage combined with a possible physisorption of, for example, water, would be in agreement with our results. It is noteworthy that oxygen coverage on W has been shown to increase the positron workfunction for the surface to about 4 eV, depending on the surface orientation [9, 13]. This might also explain the broad energy spectrum of the re-emitted positrons, as observed in the retarding-field experiment. The extra oxide layer, possibly with minor amounts of physisorbed water, would give the emitted positrons ample opportunity to lose energy on the way out through this layer by inelastic scattering or by elastic scattering that causes the positron to be emitted into vacuum at an angle other than 90°. Since it is only the perpendicular velocity that is important for the observed characteristic τ-value, emittance at a different angle would also be observed as a spread in the energy. The average energy of 2.5 eV and large energy spread would be consistent with this, but clearly more knowledge of the details of the process could be obtained by being able to monitor and alter the surface conditions *in situ*.

Energy loss may also occur repeatedly during multiple reflection. Since the positrons during reflection would approach the surface very closely, this could possibly also lead to some scattering events and loss of energy. This should lead to a gradually lower average energy for each new generation of positrons, but this has not been observed in the data. This suggests that either the positrons are reflected with negligible or no loss of energy or they are lost due to annihilation after the close encounter with the surface. This annihilation can follow instantaneously after an initial trapping at the surface potential well, or after the formation of positronium.

4. Conclusion and final remarks

We have observed reflection of very low-energy positrons from the surface of tungsten, where the surface is most probably covered with oxygen. This was done by monitoring the time of annihilation of re-emitted positrons after implanting 1 keV positrons into such a W sample. A reflection coefficient of 0.55 (±0.04) was observed. In a second experiment where 20 eV positrons impinged on the same surface, a reflection coefficient of 0.58 (±0.03) was observed.

We should note that our results are also in perfect agreement with the results from test experiments of our new reactor-based beamline. Here the best fit to the measurements on similar foils yielded a reflection coefficient of about 0.6.

The effect demonstrated here of positron reflection off oxygen-covered tungsten surfaces could be utilized to improve the overall efficiency of moderators by allowing geometries not previously considered. It could also be used to increase the slow-positron density in, for example, atomic scattering experiments by making the positrons bounce back and forth. The effect of reflection shows good promise as an additional tool to increase the overall efficiency of positron moderators.

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