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Energy spectroscopy of positrons re-emitted from polycrystalline tungsten

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Abstract. Energy spectra of positrons re-emitted from the surface of polycrystalline tungsten foil have been measured as a function of incident positron energy E in the range 100 eV–3 keV. The measurements were made using an electrostatic positron-beam apparatus incorporating a hemispherical energy analyser. At high E values the re-emitted spectra tend to a characteristic shape, dominated by a work-function peak at 2.7 eV with a low-energy shoulder comprising approximately 25% of the total spectrum. As E decreases a high-energy tail attributed to epithermal positrons increases steadily in relative intensity and extent. Below $E \approx 500$ eV the contribution of the low-energy distribution to the spectra increases significantly, and at 100 eV is dominant. These observations are interpreted in terms of elastic and inelastic processes at the tungsten surface.

1. Introduction

The phenomenon of re-emission of slow positrons from the surfaces of a wide range of materials bombarded with energetic positrons has enabled the development of monoenergetic positron beams over the past two decades for a rich variety of experimental investigations in surface, solid-state, and atomic physics [1]. In 1972 Tong [2] proposed that the emission of electronvolt positrons from a gold surface, observed by Costello *et al* [3], was associated with the existence of a negative positron work function ϕ^+ for gold. Only two years after the publication of this pioneering work Pendyala *et al* [4] measured the energy distribution of slow positrons re-emitted from chemically cleaned and partially outgassed polycrystalline tungsten, molybdenum, and chromium using a hemispherical electrostatic analyser. The measurements, using beta positron bombardment, were performed in high vacuum, and the samples were not atomically clean. The recorded spectra were seen to consist of two principal parts: a broad asymmetric peak at 0.12 ± 0.1 eV with a width of ~ 2 eV, and a second, sharp, usually symmetric peak with an energy that depended on the material under study. These observations may now be reinterpreted in the light of extensive research performed since 1974, including the work described below.

Early observations suggested that positrons were ejected essentially elastically at angles close to the surface normal from negative-ϕ^+ surfaces (a feature which later made these materials ideal for use as moderators in the formation of parallel slow-positron beams). The measurements of Murray and Mills [5] qualitatively support a simple model in which thermalized positrons from the bulk cross a simple one-dimensional step of height ϕ^+. The data of Gullikson *et al* [6] indicate that the positron yield from metals increases as ϕ^+ becomes more negative. Huttunen *et al* [7] observed that the emission yield from Cu(111) tends to zero as temperature tends to 0 K, when the positron wave is reflected

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from the surface potential. It was suggested that earlier experiments [8] failed to exhibit this dependence because of the dominance of inelastic escape processes [9] or because the positron had many encounters with the surface [10, 11].

With the acquisition of data for a wide variety of surfaces it was noticeable that the observation of re-emitted positrons with measured energies less than ϕ^+ was common. Murray and Mills [5] observed small low-energy tails on their measured distributions, but it could not be determined whether this was an indication of energy loss or elastic deflection during the emission process due to the method of measurement. Wilson [9] observed that after oxygenation re-emission from the W(111) surface was almost all at measured energies below ϕ^+. Pendry [12] attempted to explain the observed large spread in angle and/or energy. He suggested that the positron surface state could be thought of as a mixed positron/positronium (Ps)-like system, and that positrons are ejected with a large spread of energies and/or angles following its dissociation.

Fischer *et al* [13] measured the differential energy distribution of re-emitted positrons for Ni, W, and Cu surfaces, eliminating the ambiguity between inelastic re-emission and re-emission over large angles. The re-emission spectra of essentially clean samples consisted almost entirely of an elastic component. The incident positrons were implanted at 3 keV so that the numbers of epithermal positrons were negligible. These results, together with angular-distribution measurements, could be explained in terms of Maxwell–Boltzmann statistics—i.e., exhibiting only thermal broadening. The results are supported by those of Gullikson *et al* [14], who employed a varying magnetic field in the sample region; they concluded that the broad distribution of the perpendicular component of energy was due to a wide angular distribution of essentially monoenergetic positrons. These results were in apparent disagreement with theories explaining positron trapping into a surface state via electron–hole formation [15]. Also, the persistence of emission to low temperatures [8, 16] and the small positron reflection coefficient [9] at incident energies below the barrier, due to the negative work function, imply that strong inelastic processes occur at the metal surface [17]. Large energy losses due to electron–hole excitation were also predicted by Neilson *et al* [18] and Pendry [19].

Theories of positron emission not involving intermediate Ps formation are based on transition rates for the various surface scattering mechanisms; here the barrier height ϕ^+ and the dimensions of the image potential well are important. More recently theoretical models, employing an adjustable screening parameter, have shown reasonable agreement with measured energy and angular distributions of re-emitted positrons [20, 21].

When positrons impinge upon a surface at energies below a few kiloelectronvolts, they may suffer enough large-angle scattering events just below the surface that they can be re-emitted into the vacuum before they have been thermalized. For much of the work already discussed above non-thermal positron re-emission has been noted; in the first paper on positron re-emission from clean metallic surfaces Mills *et al* [22] noted that the epithermal contribution to the re-emitted positron spectrum decreased as a fraction of the total yield, with increasing incident energy. However, in contrast to work-function emission, epithermal re-emission was seen to be temperature independent. Another demonstration of epithermal positron re-emission was given by Fischer *et al* in differential energy-distribution measurements of low-energy positrons re-emitted from a Ni(100)+CO surface [13]. Nielsen *et al* [23] measured the epithermal energy distribution of positrons re-emitted from the surface of Al(111) after being implanted at low energies, and the presence of epithermal positrons was indirectly confirmed by Howell *et al* [24], who observed energetic Ps leaving metallic surfaces.

This paper reports new measurements of the energy spectra of positrons re-emitted

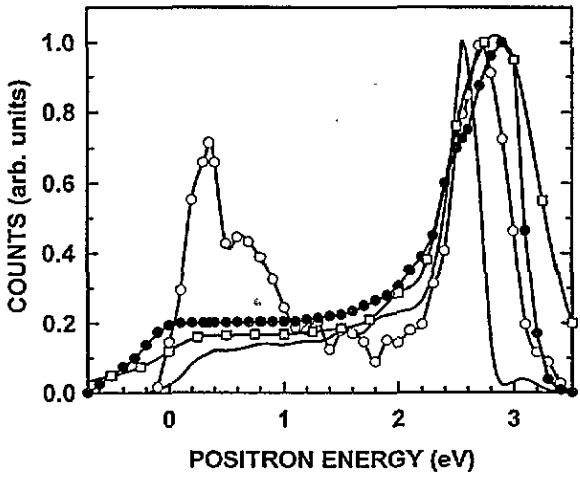

Figure 1. Energy spectra measured in earlier work for positrons re-emitted from tungsten. —— parallel-energy spectrum for positrons re-emitted from clean W(111) [28] (E = 1 keV); ○, polycrystalline W heated to 475 K [4] (E = beta spectrum); ●, parallel-energy spectrum for 2500 Å W(100) film [25] (E = 12 keV); □, parallel-energy spectrum for W(100) [27] (E = 2 keV). Here 'parallel energy' refers to $p_p^2/2m$, where p_p is the positrons' momentum perpendicular to the exit surface of the sample; this is the quantity measured in magnetic-transport positron-beam systems.

from the surface of polycrystalline tungsten foil. This surface was chosen because of the wide application of tungsten foils, crystals, and meshes as moderators in high-vacuum systems, often annealed by resistive heating or electron bombardment in an external vessel and transported through air to the positron-beam system. Earlier measurements of positron emission from tungsten, both single-crystal and polycrystalline, suggested the existence of low-energy tails, shoulders, or peaks of varying shapes and relative intensities [25–29]. Four examples of differential positron energy spectra for tungsten samples, all obtained with relatively high-energy incident positrons, are illustrated in figure 1. The primary motivation for the present study was to endeavour to interpret the nature of this part of the observed energy distribution by recording re-emitted positron energy spectra for the first time as a function of incident positron energy.

2. Experimental method

The energy spectrometer employed in the present study has been described fully by Goodyear and Coleman [30]. Positrons from a ^{22}Na source are moderated in an annealed 5 μm tungsten foil, transported electrostatically and focused to a millimetre-diameter spot on the surface of the sample at an angle of incidence of 45°. The sample is held at +150 V directly behind a cover plate at +100 V, so that only those positrons leaving the tungsten can contribute to the measured spectra. A 90% transmission tungsten mesh, also at +100 V, is held a few millimetres in front of the sample to ensure a normal electrostatic field. Positrons re-emitted within 2° of the sample normal are accelerated into the transport optics of the hemispherical analyser system, which is ramped synchronously with a PC-based multichannel scaler. The tungsten sample used was in the form of a 25 μm thick foil. It was initially annealed in a separate vacuum chamber evacuated to a pressure of 10^{-5} Pa; a strip of the foil

(40 mm×20 mm) was clamped at each end between two copper plates and a current of 50 A passed through it for several minutes. The foil glowed white hot and the chamber pressure temporarily increased to 10^{-3} Pa. The foil was then transported in air and the central 12 mm×20 mm portion cut and mounted on to a sample manipulator. After installation the vacuum system was pumped and baked at 140 °C for 24 h. The vacuum pressure for the duration of the experiments was below 10^{-7} Pa. Chemical analysis of the sample surface was performed using Auger electron spectroscopy. The resulting spectrum was dominated by the 273 eV peak corresponding to carbon contamination, and repeated measurements showed no measurable change in sample condition throughout the data-collection period. No further cleaning was performed at this stage so as to reproduce conditions common to those in the majority of laboratory-based positron-beam systems.

Re-emitted positron energy spectra were recorded for incident beam energies of 100, 200, 500, 1000, 1500, 2000, and 3000 eV. The incident-beam transport lenses were re-tuned for each energy to produce a uniform beam whose quality and intensity were monitored using a channel-electron-multiplier (CEM) detector mounted below the sample, before lowering the sample into position.

A fixed (hemispherical) analyser transmission energy of 22 eV (FAT-22) was chosen for the measurements, being a compromise between the need for acceptable resolution (measured to be 550 meV FWHM) and signal count rate. Spectra were recorded with the incident beam turned on and off, the latter (background) being subtracted from the former. The background spectrum was observed to be constant in intensity and shape for the duration of the experiment; the absolute background pulse count rate was about 0.25 s^{-1}.

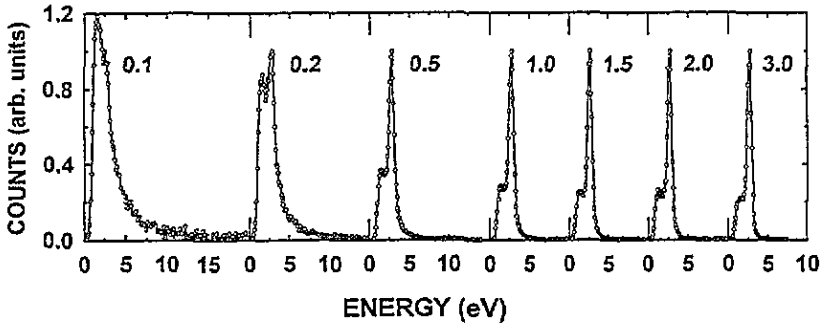

Figure 2. Differential energy spectra for positrons re-emitted from polycrystalline tungsten foil. The background has been subtracted. Next to each spectrum is shown the incident positron energy in kiloelectronvolts.

The measured signal spectra are shown in figure 2. Reproducibility of applied potentials and analyser settings resulted in an uncertainty in the energy setting of no more than ±0.05 eV. The spectra are displayed normalized to the maximum count rate recorded in the vicinity of 2.75 eV, the positron work function for tungsten [31]. Some of the displayed spectra consist of data averaged from several sweeps across the energy range; all component spectra were compared and confirmed to be identical before combination.

3. Interpretation of spectra

Figure 3 shows the spectra of figure 2 overlaid to give a clearer comparison of trends in the shapes of the re-emitted positron energy spectra as the incident energy is varied.

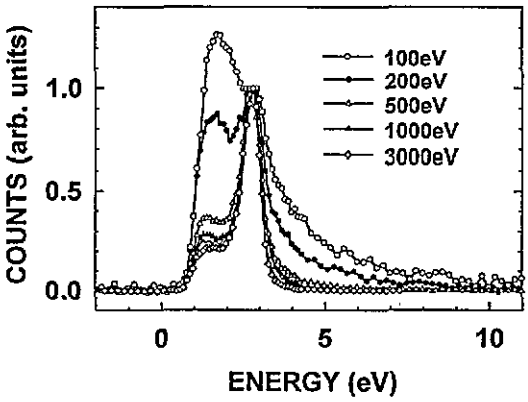

Figure 3. The data of figure 2 normalized to the peak counts at 2.75 eV. Data at $E = 1.5$ and 2 keV lie between the 1 and 3 keV curves, but are left as solid lines without symbols for clarity.

The measured spectrum can be broadly divided into three main features. The larger, sharp peak was determined to be due to elastic re-emission from the tungsten with an energy corresponding to the positron function ϕ^+ for tungsten (2.75 eV). The higher-energy tails correspond to epithermal, or non-thermalized, positron emission. At energies below ϕ^+ there is a broadly-peaked distribution, which dominates the spectrum at low E. Each of these three components will now be considered individually.

3.1. Work-function positron emission

With the analyser set at this peak energy, slight rotation of the sample ($\sim \pm 10°$) caused a dramatic decrease in the measured count rate, confirming that the elastically re-emitted positrons are ejected in a small angular cone about the surface normal.

Gidley and Frieze [32] state that the elastic-peak position is invariant and depends only on the sum of the positron and electron bulk chemical potentials $\mu^+ + \mu^- = -(\phi^+ + \phi^-)$. They also demonstrated that the zero cut-off of a differential energy spectrum is determined by the ϕ^- of the system, this being sensitive to the conditions of the sample. For a clean single material this is simply equal to ϕ^- for that material. For a multilayer structure, ϕ^- has a single value equal to ϕ^- for the top layer of the system, due to the equalization of the electron Fermi levels of the various layers. The carbon overlayer, with an electron work function of 4.6 eV [33], should determine the zero-cut-off energy on the present spectra. For the purposes of this analysis the tungsten electron work function of 4.55 eV [34] is indistinguishable from that of carbon given the uncertainties in these values (± 0.1 eV) and analysis of the spectra (± 0.1 eV). Assuming an electron work function of 4.6 eV for the sample, the difference in energy between the tungsten elastic peak and the zero cut-off should be about 1.9 eV. Allowing for instrumental resolution, this energy difference was observed in all the present measurements.

For all incident energies work-function emission from tungsten was observed, although at the lowest incident energy (100 eV) the signal is very weak. The signature of re-emission of thermalized positrons from the tungsten is a clear indication of positrons penetrating the carbon overlayer elastically. It cannot be determined from the data whether these incident positrons penetrate directly into the tungsten and complete their thermalization there, or whether they lose most of their energy within the carbon overlayer and diffuse into the tungsten. They would of course require in excess of 2.75 eV to overcome the barrier

presented by the positron work function on entering the tungsten. It has been stated by *Nielsen et al* [23] that positrons with close-to-thermal energies have long mean free paths due to the strong energy-loss processes (creation of electron–hole pairs on bulk or surface plasmons) being energetically impossible.

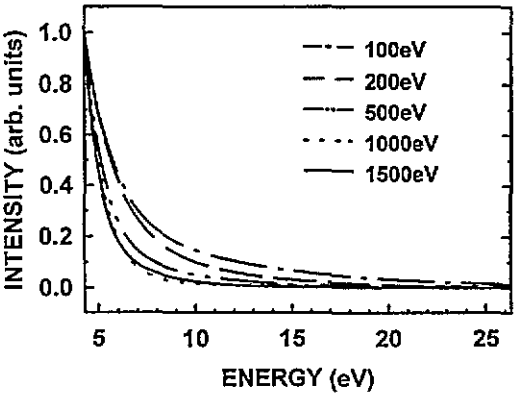

Figure 4. Fits to the epithermal positron tails for a range of incident positron energies. The curves are normalized to unity at 4.3 eV.

3.2. Epithermal positron emission

The work-function peak is broadened on the high-energy side by instrumental resolution and also by the presence of a tail attributed to positrons that are re-emitted into the vacuum before thermalizing completely inside the sample. (Mean implantation depths of kiloelectronvolt positrons are several atomic layers—remembering that they are implanted at 45° to the surface normal—so it is not surprising that a fraction of the implanted positrons will be re-emitted before complete thermalization.) Because of relatively poor counting statistics in the epithermal tail region, particularly at low incident energies, the raw data of figure 2 have been fitted; figure 4 shows the resulting fits to spectra for $E = 100$–1500 eV, above which range the very small signal levels and the resulting large uncertainties made meaningful fits impossible. The fits have been performed only at energies greater than 6 eV above the applied sample bias, so that interference from the high-energy side of the work-function peaks is negligible. To compare the shapes of the tails the fits are normalized at 4.3 eV.

Although the uncertainties in the fits are quite large, particularly at high incident energies, it is apparent that at lower incident energies the distribution spreads to higher energies. Above $E = 2000$ eV no appreciable signal is detected 2 eV above the work-function energy. However, for 100 eV incident positrons a detectable tail is observed to extend up to 20 eV above $-\phi^+$. One must be careful in stating absolute extents for tails as the recorded spectra are a representation of not only the true emission but also the collection efficiency of the analyser. When investigating this feature of the apparatus the bias potential applied to the sample must also be considered. The angular acceptance of the entrance to the hemispherical analyser is 2° so the viewed area of the sample is essentially defined by the analyser slits. It is clear that the number of re-emitted epithermal positrons per unit solid angle entering the analyser slit will depend on their intrinsic energy of emission, because of the +150 V bias applied to the sample (which, with the 2.75 eV work function, accelerates the positrons normally to the surface). For example, 100% of the positrons

approaching the surface isotropically from within the energies up to 0.2 eV will essentially have trajectories at angles less than 2° to the normal, and be detected, whereas this is true for only 10% of positrons approaching with 1 eV and 5% with 2 eV. As a consequence the recorded energy spectra are distorted, with the systemic attenuation increasing with increasing energy. The analyser collection efficiency, discussed in detail by Goodyear and Coleman [30], would have to be folded into any future theoretical simulation with which the data could be compared.

3.3. Inelastic positron emission

The third main feature on the spectra is a broad peak with recorded energy less than the work-function energy. One can discount the source of this distribution as the detection of positrons elastically re-emitted at large angles because of the ±2° angular acceptance of the entrance optics to the hemispherical analyser. The possibility of the feature being due to elastic re-emission from surface contamination with a negative positron work function ϕ^+ less than that for tungsten is unlikely for two reasons. Firstly, the Auger-electron-spectroscopy analysis suggested the majority of surface contamination to be due to carbon, which is known not to re-emit work-function positrons [35, 36]. Secondly, the low-energy distribution appears to be broader than that expected for a work-function peak, even after allowing for distortion due to the proximity of the tungsten work-function peak. One can only conclude that this feature is due to positrons that have lost energy in the process of re-emission, either from the tungsten, or the surface contamination, or both.

Previous direct differential energy spectra measurements such as those depicted in figure 1 have almost all been confined to the use of relatively high-incident-energy beams—i.e., high enough that epithermal positrons form a negligible part of the distribution. For the results shown here this would indicate an incident energy of 1500 eV or higher for the tungsten sample. Fischer *et al* [13] made high-resolution angle-resolved measurements of positron re-emission energy spectra from metal surfaces using a 3 keV incident beam, and observed a low-level energy-loss tail for W(110) plus an ordered carbon overlayer. Whether the inelastic shoulder is wholly due to sample contamination or whether a small residual tail remains for a clean surface is still open to question.

3.4. The dependence of the spectra on incident positron energy

An attempt was made to separate the three main components of the re-emitted positron energy spectra, i.e. work-function, epithermal, and energy-loss emissions, in order to extract the relative fractions of the measured spectra associated with each component and their dependence on E.

It was first assumed that the data obtained for a 3000 eV incident beam were representative of 'work-function' emission for the sample under study, being the high-E asymptotic limit. This standard spectrum could then be scaled and subtracted from the other spectra to leave the residual energy-loss and epithermal distributions. However, since the measured spectra could not be normalized absolutely it was not known what scaling factor should be applied to the 'work-function' spectrum prior to subtraction; a range of arbitrarily chosen factors yielded acceptable, but markedly different, residuals. Given the ambiguities arising from this procedure, a more approximate method was adopted. The spectra were divided into three regions at energies indicated in the inset in figure 5 and the integral counts calculated for each region, giving estimates of energy-loss, work-function, and epithermal emission fractions. Figure 5 shows the relative sizes of the energy-loss and

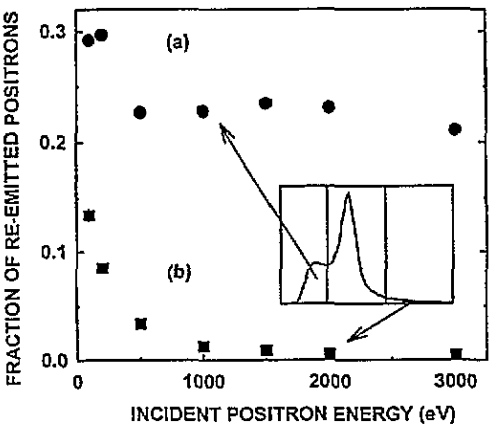

Figure 5. Fractions of re-emitted positrons in the inelastic (circles) and epithermal (squares) sections of the spectra of figures 2 and 3. The sections are defined by two fixed values of E, as illustrated by the inset, which shows the 500 eV spectrum. Statistical uncertainties lie within the data points.

epithermal emission distributions as fractions of total recorded emission at each incident energy.

Let us first consider the variation of the relative intensity of the low-energy peak as the incident energy E is varied. At low E it dominates the spectrum, but as E increases its relative contribution to the total spectrum diminishes—it tends towards an asymptotic size in relation to the now-dominant work-function peak, as shown in figures 2 and 5. The interpretation of the behaviour is open to some question but several points may be considered.

One may postulate with some confidence that at high incident energies the majority, if not all, of the implanted positrons pass through the surface contamination, penetrate into the tungsten, and are thermalized. The majority of those re-emitted are ejected elastically from the tungsten into the vacuum with the signature of the positron work function. A small fraction of the work-function positrons may have lost energy on passing through the tungsten surface and/or the overlayer. This fraction is seen to be approximately constant at energies above about 500 eV. There may also be a contribution, again approximately constant for E of the order of kiloelectronvolts, from positrons backscattered from the tungsten and undergoing inelastic interactions with the overlayer before reaching the vacuum.

At low incident energies it may be that some of the positrons do not reach the tungsten at all but remain in the non-re-emitting overlayer. A fraction of these positrons are able to leave the overlayer with energies ranging from the zero cut-off up to the incident-beam energy. Thus, the results for the 100 eV incident beam could be interpreted as the energy distribution of positrons leaving a positive-work-function material with a very small peak due to the few positrons that reach the tungsten and are re-emitted elastically with the work-function energy.

An alternative explanation of the dependence of the fractional intensity of the low-energy distribution of E, based simply on the E dependence of the positron implantation profile, may be made as follows. At low E the majority of the implanted positrons are stopped in the overlayer, as assumed above. As E increases above 200 eV the implantation profile starts to extend into the tungsten and the fraction stopping in the overlayer decreases rapidly. At kiloelectronvolt energies a smaller fraction of the implantation profile—now

varying much more slowly with E—spans the shallow overlayer.

Let us now consider the possibility that the two fractions shown in figure 5 are directly correlated. Nieminen and co-workers proposed that inelastically emitted positrons lose most of their energy to the creation of electron–hole pairs, with other processes—such as bulk or surface plasmons and phonons—being less important [10, 20]. It may be proposed that higher-energy epithermal positrons have more scattering mechanisms available, and/or have a greater probability of suffering large energy losses, thus placing them in the low-energy inelastic region. Certainly it is clear from figure 5 that both fractions are significantly larger at low E; the difference in the detail of their E dependence may arise from the fact that the low-energy region is associated with the overlayer alone, whereas the epithermal emission may occur from both epilayer and substrate.

3.5. The effect of surface cleaning

The tungsten sample was heated several times and the resulting changes in the spectra observed after each heating cycle. The sample surface was monitored throughout using Auger electron spectroscopy. Only the work-function and energy-loss peaks were monitored, with short runs between the heating cycles to keep the data collection times to a minimum. Details of the heating cycles are listed below with the resulting changes in the Auger electron spectra.

(1) The sample was heated to 600 °C for 15 min followed by flash heating to 800 °C for a few seconds. The carbon Auger peak remained dominant, with small oxygen and sulphur peaks; a very small tungsten Auger peak at 179 eV was just visible above the background.

(2) The sample was heated to 500 °C for several hours. The carbon peak greatly diminished and twin tungsten peaks were clearly visible at 169 and 179 eV. The oxygen and sulphur peaks were still present.

Figure 6 shows the spectra before heating and after the two heating cycles. Because essentially identical incident beams were used the spectra for any particular energy can be compared in absolute terms. The spectra appear incomplete because data were collected in the shortest possible time commensurate with acceptable statistics to avoid significant recontamination of the surface, therefore, only the salient features of the spectra—the two peaks—were scanned. For both the 200 and 500 eV spectra the size of the work-function peak increased dramatically. An increase in work-function emission is expected since more of the positrons within the tungsten are able to escape elastically as more of the tungsten surface is exposed. It may also be the case that more of the incident positrons reach the tungsten on the way into the sample and are made available for work-function emission. The low-energy peaks in the 200 and 500 eV data do not appear to change in absolute size, but there may be a decrease in inelastic emission masked by the resolution spread of the larger elastic peak. Consistent with this point is the fact that the inelastic peaks do appear to have become more of a shoulder on the low-energy elastic tail.

After flash heating to 1000 °C, when it was estimated that total contamination was reduced to approximately 0.25 monolayer, complete spectra were recorded for incident-beam energies of 100 and 2000 eV, as shown in figure 7. The spectrum obtained at 2000 eV is very similar to that obtained by Fischer *et al* [13] with the dominating work-function peak being slightly asymmetrical due to the low-energy inelastic shoulder on one side and a very small epithermal tail on the high-energy side. For the 100 eV data, work-function emission—hardly visible in figure 2—is clearly apparent and the energy-loss peak has greatly diminished.

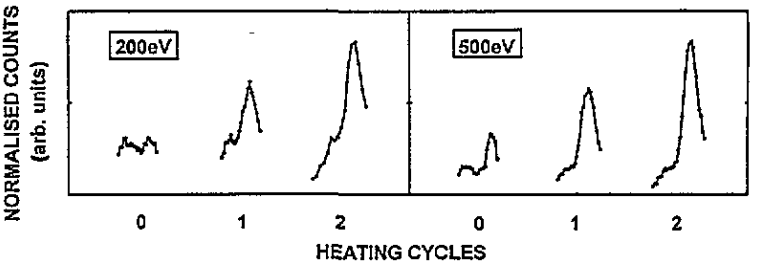

Figure 6. Re-emitted-positron energy spectra for polycrystalline tungsten before (0) and after the two heating cycles (1 and 2) described in the text. Incident energies are 200 and 500 eV. All conditions are identical for the three spectra at each energy, enabling direct comparison.

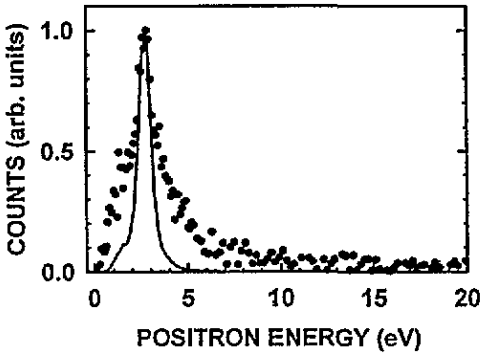

Figure 7. Re-emitted-positron energy spectra for cleaned polycrystalline tungsten for incident positron energies of 100 eV (circles) and 2 keV (line).

4. Conclusion

The energy spectra of positrons re-emitted from annealed polycrystalline tungsten foil have been measured for the first time for a wide range of incident positron energies E from 100 to 3000 eV. The spectra exhibit three major components: those associated with work-function and epithermal emission, and a lower-energy distribution associated predominantly with energy loss in the overlayer of contaminant on the foil surface. Whereas above $E = 200$ eV the low-energy peak is probably due to positrons re-emitted from the tungsten losing energy in the overlayer, at the lowest incident energies the significant increase in the size of the low-energy peak implies that many positrons are interacting primarily with the overlayer only. The value of cleaning the tungsten surface by heating *in situ* is demonstrated by the resulting large increase in work-function emission; this is of great importance in the use of such foils in laboratory-based positron-beam systems.

Acknowledgments

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