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Relative probabilities of work-function and epithermal positron re-emission from silver

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Abstract. Experimentally determined values for the relative probabilities for work-function and epithermal positron re-emission from silver are reported as a function of incident positron energy E. The method involves the comparison of absolute slow-positron yields from a polycrystalline Ag sample with a very small negative work function with the epithermal yields from an Ag(100) sample having a small positive work function. The results show that the epithermal fraction increases steeply from almost zero as E decreases below 2 keV, tending to almost 100% as E approaches 0.

1. Introduction

The phenomenon of slow-positron re-emission from the surfaces of solids bombarded with energetic positrons lies at the core of a plethora of experimental techniques employing positron beams. Since 1972, when the idea of a negative positron work function ϕ^+ was first proposed [1] to explain the emission of low-energy positrons from a gold surface [2], there have been numerous measurements of negative work functions and slow-positron yields for a range of solid surfaces [3].

It was first shown by Mills [4] that as the incident-positron energy (and therefore the mean implantation depth) decreases, an increasing fraction of the positrons reaching the surface from within the target material do so before being thermalized. These non-thermalized, or epithermal, positrons leave the surface with energies up to several eV in excess of the work function [5].

An understanding of the relative probabilities for work-function and epithermal positron re-emission is important if researchers are to interpret correctly data from a variety of experimental studies which employ positron implantation as a method for probing the surface, subsurface and interface properties of materials. Fischer *et al* [6] observed epithermal positron emission from Ni(100)+C and concluded that the epithermal fraction falls to a negligible level at incident-positron energies above about 3 keV. More recently, Goodyear *et al* [7] studied the epithermal component of positron re-emission spectra from a polycrystalline tungsten surface. At an incident energy of 2 keV they detected no appreciable signal 2 eV above the work function ϕ^+, whereas for an incident energy of 100 eV a detectable tail was observed to extend up to 20 eV above ϕ^+. Overton *et al* [8] estimated the epithermal contribution to the spectrum of positrons re-emitted from polycrystalline copper by fitting a normalized secondary-electron spectrum to the tail of the positron spectrum. They found a rapid increase in the relative intensity of epithermal positrons for incident

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energies below 500 eV. Although this result was qualitatively similar to the theoretical result of Kong and Lynn [9], there were differences in detail which merit further investigation.

In this paper, a different approach to that of Overton *et al* is taken to the study of work function and epithermal positron emission from silver, based on measured, absolute total yields and not on the shapes of the energy spectra. This was made possible by the fact that positron emission associated with a small negative work function was observed from a piece of cleaned polycrystalline silver, whereas only epithermal emission was observed for Ag(100), which is known to have a positive ϕ^+ of 0.72 eV [10]. A comparison of the total slow-positron yields from the two samples—in essence 'turning on and off' the work-function component—allows an estimation of the epithermal positron contribution to the total yield measured from the polycrystalline sample.

2. Experimental apparatus

The measurements were performed using the UEA magnetic transport positron beam which is described in detail elsewhere [11]. The experimental arrangement was described by Coleman *et al* [12]. Positron annihilation radiation from the silver targets was detected by a 72%-efficiency HPGe system, which enabled signal counts to be accumulated at an acceptable rate when the detector was placed behind a lead slit designed so that only annihilation radiation from the target was detected. Only counts in the 0.51 MeV photopeak of the annihilation gamma ray energy spectrum were used as these offered the best signal:background ratio. The sample chamber was capable of being evacuated to $\sim 10^{-8}$ Pa.

The shape and position of the beam was monitored before each measurement by raising the sample and observing the beam profile with a CEMA/phosphor screen assembly at the end of the beam line. The beam was ~ 4 mm in diameter compared to the sample size of 25 mm square. The position of the sample holder was adjusted so that the beam would hit the sample centrally by viewing the shadow of the corner of the sample holder in the image of the beam.

Backscattered positrons were prevented from returning to the sample by the use of $E \times B$ deflector plates positioned in front of the target, as described by Coleman *et al* [12] and Knights and Coleman [13].

The sample holder allowed *in situ* heating to 700 °C and electrical isolation of the silver samples. A 25 × 25 × 1 mm piece of beryllium was suspended below the sample in good electrical but poor thermal contact.

Prior to installation the silver samples were chemically etched by immersion in 50% HNO_3 for 30 s. To achieve a clean surface, the samples were heated *in situ* to 600 °C for 2700 s in an atmosphere of 10^{-6} Torr O_2. Auger electron spectroscopy was used to monitor surface condition, which was found to be essentially contamination-free following cleaning.

3. Experimental method

Epithermal positrons are here defined as those re-emitted with momentum components normal to the surface corresponding to energies below 15 eV, a criterion discussed and adopted by Knights and Coleman [13]. At higher energies the positrons are deemed to have been backscattered.

With −15 V and +10 V applied to the samples the respective measured count rates N_{sam} and N'_{sam} are

$$N_{sam} = N_b + N_s + N_p[1 - \beta] \tag{1}$$

and

$$N'_{sam} = N_b + N_s \tag{2}$$

where N_b and N_s are the number of counts resulting from annihilations in the bulk and at the surface of the target respectively, N_p is the fraction of positrons re-emitted from the sample and β refers to the fraction of re-emitted positrons that eventually form ortho-positronium (o-Ps) after being returned to the sample by the applied potential. Note that in equations (1) and (2), and hereafter, only those incident positrons which are not backscattered are considered. The short-lived para-positronium (p-Ps) atoms decay within the viewed area of the detector and are therefore treated as surface annihilations for the purpose of this analysis. From (1) and (2)

$$N_p = \left(N_{sam} - N'_{sam}\right)/[1 - \beta]. \tag{3}$$

It is a straightforward matter to show that the incident positron count rate N_0 is given by

$$N_0 = \left(N_p\beta + N_{sam}\right)/(1 - F) \tag{4}$$

where F is th fraction of the incident flux which forms o-Ps. The re-emitted fraction of slow positrons J_s is then simply

$$J_s = N_p/N_0. \tag{5}$$

Values of β = 15% and 19%, both ±5%, were obtained for polycrystalline silver and Ag(100), respectively, by comparison of the count rates at an incident energy of 100 eV and with −15 V applied to the silver and beryllium samples, the latter having a known value of β of 19% [13]. These values of β for silver are in reasonable agreement with the value of 20–25% which can be deduced from the study by Lynn and Welch [14] on single-crystal silver surfaces. Values for F can be calculated using the formula suggested by Mills *et al* [4]:

$$F = F_0\left(E/E_0 + 1\right)^{-1} \tag{6}$$

where F_0 is the positronium formation fraction at an incident energy of $E = 0$, and E_0 is taken to be 5000 eV [14].

Values of the slow-positron re-emission fraction J_s were obtained for incident energies ranging from 1 keV to 10 keV. In addition, an integral 'perpendicular' energy spectrum of positrons re-emitted from polycrystalline silver for E = 1 keV was obtained by ramping the potential applied to the sample from +10 V to −15 V. Perpendicular energy is here defined as $P_p^2/2m$, where P_p is the component of positron momentum normal to the sample surface.

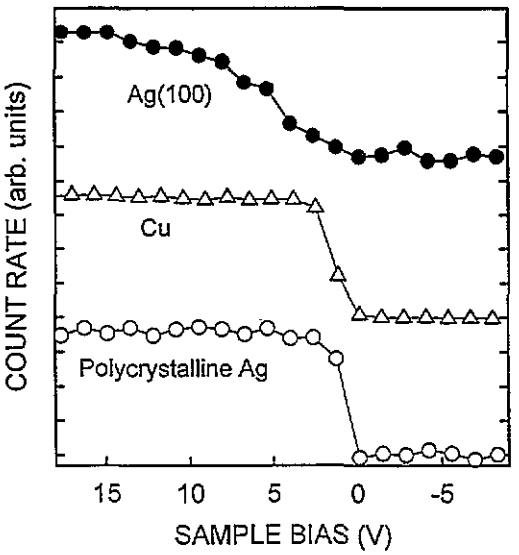

Figure 1. Signal count rate versus sample bias for polycrystalline silver (open circles), Ag(100) (solid circles) and copper (triangles).

4. Results and discussion

Figure 1 shows the integral energy spectrum of positrons re-emitted from polycrystalline silver for $E = 1$ keV, plotted together with equivalent spectra obtained from polycrystalline copper and Ag(100).

The spectra clearly indicate the work-function nature of the re-emission of positrons from this sample of polycrystalline silver, the sharp change in signal count rate as the sample potential is ramped through zero is consistent with a small negative work function, as for copper [3]. The resolution associated with this method does not allow an accurate measurement of the polycrystalline silver work function, but comparison of the silver and copper spectra suggest that it is close to zero. These two spectra are in contrast to that obtained from the Ag(100) sample, which shows a less sharp increase in count rate characteristic of the emission of epithermal positrons only. A negative work function for a silver surface contaminated with oxygen has been previously reported [14]; however, the polycrystalline silver surface in this study was observed to be essentially free of oxygen contamination by Auger electron spectroscopy, and the *in situ* treatments of the polycrystalline and single-crystal samples were identical.

Figure 2 shows the fractions of incident (non-backscattered) positrons (J_s) re-emitted from the polycrystalline silver and single-crystal Ag(100) samples as a fraction of incident-positron energy E. The yield curves have been fitted satisfactorily using the procedure commonly applied to the evaluation of diffusion lengths by slow-positron implantation spectroscopy. In this method the re-emitted fraction has the form

$$J_s(E) = \int_0^\infty P(z, E) \exp(-z/L)\, dz \tag{7}$$

where $P(z, E)$ is the positron implantation profile and L the effective positron diffusion length. Here the Gaussian derivative form of $P(z, E)$, $(2z/z_0^2)\exp(-z^2/z_0^2)$, is used, with

$z_0\Gamma(1.5) = \bar{z} = 56E$ Å (E in keV). For the polycrystalline sample $L = 1200 \pm 200$ Å, in good agreement with an earlier result of 1050 ± 100 Å for pure, well-annealed silver made in this laboratory by the more conventional Doppler broadening spectroscopy method, and with the value of 1100 ± 100 Å obtained for Ag(111) by Soininen *et al* [15]. L for the epithermal positrons from Ag(100), representing not the diffusion length but rather the length over which the positron energy in the sample is reduced from $\sim 10^1$ eV to a value below the positron work function (0.72 eV), was found to be 25 ± 2 Å. It is interesting to note that the functional form of the epithermal positron yield from Ag(100) can also be represented approximately by E^{-n}, where n is close to 1. This value of n is significantly lower than the 1.5 quoted by Kong and Lynn [9], but is in reasonable agreement with the result of Howell *et al* [16], who studied positronium formed at surfaces by epithermal positrons.

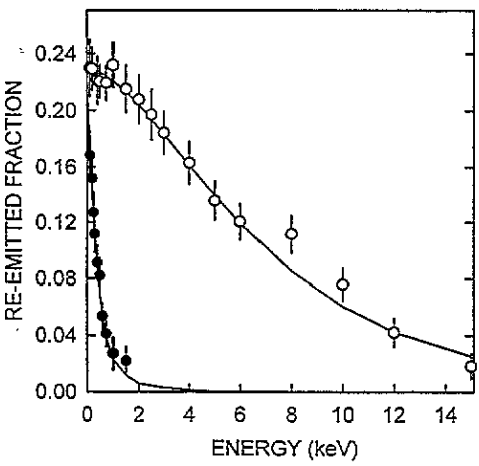

Figure 2. Fractions of positrons re-emitted from polycrystalline silver (open circles) and Ag(100) (solid circles). Solid lines are fits to equation (7).

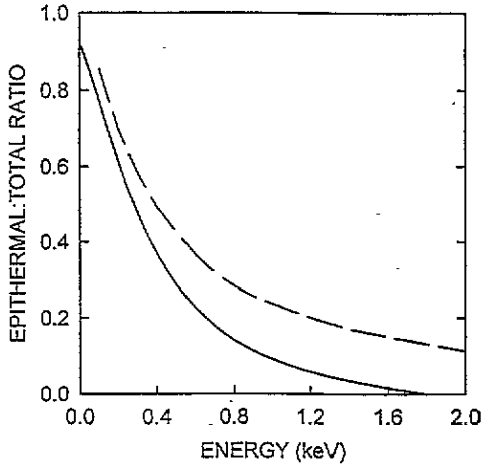

Figure 3. The ratio of epithermal to total positron reemission from polycrystalline silver deduced from figure 2 (solid line), and that obtained from the theoretical model of Kong and Lynn (broken line) [14].

Because both yield curves have been obtained from silver, the only difference between the two should be related to the different work-function values. If one should 'switch off' negative work function of the polycrystalline sample, the resulting positron yield would resemble that for Ag(100). The densities and elastic inelastic scattering rates and hence positron diffusion lengths should only be affected by the relative quality of the two samples. Both were at least 99.99% pure and received identical external and *in situ* treatments.

Figure 3 shows the ratio of epithermal to total positron emission for polycrystalline silver deduced from figure 2 and that obtained from the theoretical model of Kong and Lynn [9]. The ratio, obtained from the two fitted yield curves, indicates that the epithermal fraction increases dramatically for values of incident energy below 500 eV, in agreement with the conclusions of Overton *et al* for copper [8]. The difference between the present results and that of Kong and Lynn may have several sources. These may include uncertainties about the stopping profile for positrons at very low incident energies, and also the difference in energy at which the positron is taken to be thermalized. In the present study this value is the work function of Ag(100) whereas the thermalization length of 100 Å used by Kong

and Lynn implies that they considered positrons to be epithermal until their energy had been reduced to a value approaching kT.

Figure 3 also demonstrates that at low E the rapidly increasing numbers of epithermally emitted positrons, rather than adding to the total re-emitted flux, are compensated by a corresponding decrease in the numbers of work-function positrons. The measured low-energy asymptotic limit of the ratio is a little lower than the value of unity that one might expect; this is due to the fact that whereas all positrons, thermalized or epithermal, are re-emitted from the polycrystalline sample, a small fraction of epithermal positrons are prevented from leaving Ag(100) by the positive work function.

In summary, it has been shown that, by exploiting the small negative work function of polycrystalline silver, it has been possible to estimate the relative probabilities of epithermal and work-function positron emission by comparing the slow-positron yield from the polycrystalline sample with that from Ag(100). Similar comparisons may be possible for other materials whose crystal faces may incorporate both small negative and positive work functions.

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References

[1] Tong B Y 1972 *Phys. Rev.* B **5** 1436
[2] Costello D G, Groce D E, Herring D F and McGowan J W 1972 *Phys. Rev.* B **5** 1433
[3] Schultz P J and Lynn K G 1988 *Rev. Mod. Phys.* **60** 1988
[4] Mills A P Jr, Platzman P M and Brown B 1978 *Phys. Rev. Lett.* **41** 1076
[5] Nielsen B, Lynn K G and Chen Y-C 1986 *Phys. Rev. Lett.* **57** 1789
[6] Fischer D A, Lynn K G and Gidley D W 1986 *Phys. Rev.* B **46** 1687
[7] Goodyear A, Knights A P and Coleman P G 1994 *J. Phys.: Condens. Matter* **6** 9601
[8] Overton N, Knights A P, Goodyear A and Coleman P G 1995 *Appl. Surf. Sci.* **85** 54
[9] Kong Y and Lynn K G 1990 *Phys. Rev.* B **41** 6179
[10] Poulsen M R, Charlton M and Laricchia G 1993 *J. Phys.: Condens. Matter* **5** 5209
[11] Hutchins S M, Coleman P G, Alam M A and West R N 1985 *Positron Annihilation* ed P C Jain, R M Singu and P Gopinathan (Singapore: World Scientific)
[12] Coleman P G, Albrecht L, Jensen K O and Walker A B 1992 *J. Phys.: Condens. Matter* **4** 10311
[13] Knights A P and Coleman P G 1995 *J. Phys.: Condens. Matter* **7** 3485
[14] Lynn K G and Welch D O 1980 *Phys. Rev.* B **22** 99
[15] Soininen E, Huomo H, Huttunen P A, Mäkinen J, Vehanen A and Hautojärvi P 1990 *Phys. Rev.* B **41** 6227
[16] Howell R, Rosenberg I J and Fluss M J 1986 *Phys. Rev.* B **34** 3069