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Temperature dependence of the fraction of re-emitted positrons and of the positron work function for Cu(111) + S

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A beam of 1-keV positrons incident on a Cu(111) + S surface has been used to study the dependence on temperature of the positron work function (ϕ_+), the yield of re-emitted positrons (Y) and of the positronium (Ps) fraction. A positive dependence of the slow-positron yield on temperature is found which is attributed in part to a reduction in the magnitude of ϕ_+ ($\sim$25%) at 50 K relative to its value at 300 K. A similar, though weaker, positive dependence on temperature was seen for the Ps fraction down to 40 K. When positrons leave the sample inelastic processes such as particle-hole excitations are thought to be responsible for the absence of a much stronger dependence on temperature that had been predicted previously by Nieminen and Oliva.

INTRODUCTION

When a low-energy positron ($E < 5$ keV) impinges on a metal surface, it will thermalize by interacting with electrons and phonons at mean depths of about 100 Å ($\sim$1 keV) to 1000 Å ($\sim$5 keV). After being implanted, it can annihilate within the bulk material or diffuse back to the surface. The potential for a thermalized positron inside the metal relative to the vacuum level is a combination of the positron bulk chemical potential μ_+ and the surface dipole D, yielding a value for the positron work function ($\phi_+ = -D - \mu_+$) which is analogous to that for the electron work function ($\phi_- = D - \mu_-$).[1] Both the chemical potential and the surface dipole terms change continuously as the positron diffuses through the surface to match with the image potential felt by the positron on the vacuum side of the metal surface. A minimum occurs directly outside the metal which can localize positrons in a two-dimensional well.[1,2] A positron which diffuses back to the surface before annihilation may become trapped in the surface state, be re-emitted into the vacuum as a free positron (if ϕ_+ is negative), or re-emitted as a positronium (Ps) atom. Other escape mechanisms do exist but will not be discussed in this paper. The possibility of reflection of the positron by the potential step formed at the surface[3] can be examined by measurements of the influence of temperature on these above processes.

In this paper we present the first low-temperature measurements (down to 40 K) of the fraction of slow positrons re-emitted into the vacuum, and of the positron work function ϕ_+. A decrease in $|\phi_+|$ with decreasing temperature is found which is suggested to produce (at least in part) the observed positive dependence of the slow-positron yield on temperature. This positive dependence on temperature of the yield can simply be associated with inelastic processes being more important the longer the positron spends in the region of the surface state. These results complement measurements of the Ps fraction previously reported[4] for the same specimen.

EXPERIMENTAL

The slow-positron beam and ultra-high-vacuum system used in these experiments have been described in detail elsewhere.[5] The low-temperature manipulator has been designed to permit the sample to be positioned for low-energy electron diffraction and Auger electron spectroscopy (AES) studies. Prior to each set of measurements, the sample was sputtered clean with 1-keV Ar^+ ions at 300 K and subsequently annealed *in situ* to approximately 850 K for 1 h. A partial overlayer of sulfur ($< \frac{1}{2}$ monolayer) migrates to the surface of the Cu during annealing which enhances the fraction of re-emitted positrons. A base pressure of approximately 5×10^{-10} Torr or better was maintained in the vacuum system during the experiment.

The measurements of slow-positron yield and ϕ_+ were made using a retarding field analyzer which is shown in Fig. 1. It is basically a pair of grids with a fixed retarding bias, and a specimen of interest

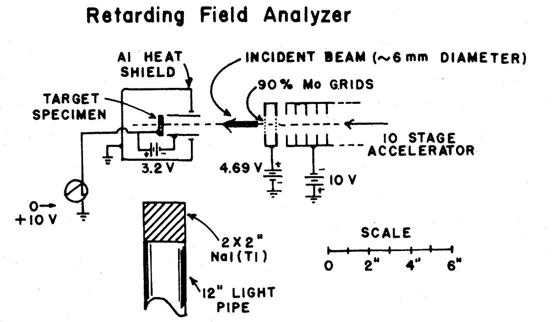

FIG. 1. Schematic diagram of the retarding field analyzer.

that is ramped with an applied bias $V_{app}=0\rightarrow+10$ V. Since positrons which pass through the grids are not detected, the total count rate as measured with the NaI(Tl) detector is proportional to the yield of slow positrons (Y) with insufficient energy to pass the grids. A typical distribution (inset of Fig. 2) has been shown[6] to fall off with a maximum slope when the net target bias ($V=V_{app}-4.69$ V) is $\phi_+ + \phi_- - \phi_g$, where $\phi_- - \phi_g$ is the difference between the electron-contact potentials for the sample and retarding grids. In practice, this point is defined by the centroid of a Gaussian that is fitted to the slope of the curve ($\partial Y/\partial V$) as illustrated in the inset of Fig. 2. The distribution flattens out when $V=\phi_- - \phi_g$, which is defined as the intersection of two straight lines fitted (visually, or least squares) to the data. Following the analysis of Murray *et al.*,[6] the difference between these points is interpreted as a measure of the positron work function ϕ_+. The slow-positron yield is determined (after appropriate corrections for annihilations of both incident and re-emitted positrons in the retarding grids) by taking the normalized difference in count rate at low (region *A*) and high (region *B*) sample bias.

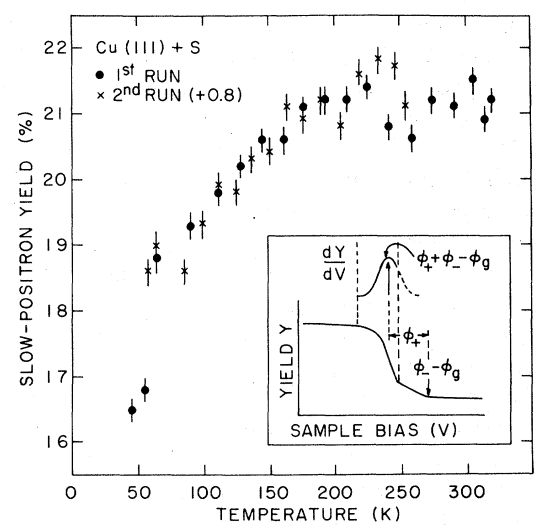

FIG. 2. Re-emitted slow-positron yield is shown. The inset shows a typical distribution of raw data, as described in the text.

DISCUSSION OF RESULTS

Figure 2 (note expanded scale) demonstrates that the yield of slow positrons re-emitted from the surface of Cu(111)+S exhibits a slight positive dependence on temperature below about 200 K, slightly larger but not unlike that found for the fraction of incident positrons re-emitted as Ps.[4] The relatively small change in the absolute yield ($<5\%$ from 50 to 200 K) supports our previous conclusion that internal reflection of the positron by a potential change at the surface does not play a significant role in this temperature region.[4] Since inelastic processes are now thought to play an important role in positron-surface interactions,[7,8] it is likely that they also are responsible for this discrepancy. Estimates for surface-state binding energies lead to positron trapping rates like 10^{14} sec^{-1}, which is of the same order of magnitude as the inverse time a re-emitted positron would take to traverse the surface state. A loss of energy at the surface would suppress any consideration of elastic reflection.[3] Concurrent with the decrease in sample temperature, it was also observed that the linear portion of the yield-versus-bias distribution was reduced in magnitude. This would be consistent with the slight positive dependence on temperature of the yield seen in Fig. 2 being attributable to elastic reflection.

Figure 2 shows the results of two separate runs, after annealing, as a function of descending temperature. The second data set was normalized to the first by adding 0.8% to each point. The fact that the two runs were different probably indicates that different amounts of sulfur diffused to the surface during each heat treatment, although the difference was not detectable with AES.

It is possible that the discrepancy between our data and the predictions of Nieminen and Oliva's model[3] arises from positrons which are not fully thermalized before reaching the sample surface and escaping into the vacuum. The "athermal" fraction would, presumably, increase at lower temperatures as the positron diffusion length and the thermalization time increase. This possibility requires that the relative probability of forming Ps and positron reemission is increased for positrons escaping from the sample with more than thermal energy. The observation (presented in Ref. 4) that the Ps fraction is even more independent of temperature when incident positron energy is increased to 5000 eV would not be consistent with the premise that athermals lead to the observed discrepancy with the model based on internal reflection.[3]

Figure 3(a) shows the temperature dependence of $\phi_- - \phi_g$ and 3(b) represents $\phi_+ + \phi_- - \phi_g$ (see inset, Fig. 2). The scatter in 3(a) is due to the fact that the low-temperature manipulator used for this experiment does not allow crystal rotation about an axis perpendicular to the incident positron beam and B field. As mentioned this reduces the precision with which $\phi_- - \phi_g$ can be obtained, as was demonstrated by Murray *et al.*[6] Figure 3(c) indicates the results for ϕ_+, which is the point-by-point subtraction between 3(a) and 3(b). It is reasonable to conclude that the observed decrease in the magnitude of ϕ_+ is partly responsible for the lower yield of re-emitted positrons at low temperatures.[6] As already mentioned, the data also indicate a reduction of the fraction of positrons scattered at low sample temperatures.

An implicit result of the data analysis[6] is that changes in ϕ_+ and ϕ_- for a particular surface are correlated. Since the grids and their holder are assumed to remain at room temperature, there is no reason to expect the contact potential for the grids

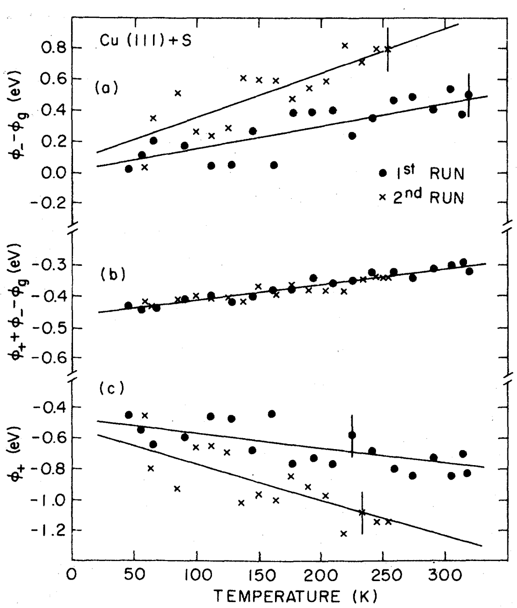

FIG. 3. Dependence on temperature is shown for the positron (ϕ_+) and electron (ϕ_-) work functions.

(ϕ_g) to change throughout the experiment. Owing to the constancy of ϕ_g, Fig. 3(a) should be a direct measurement of the reduction in ϕ_- with reducing temperature. Changes in both ϕ_- and ϕ_+ of opposite signs are consistent with a temperature-dependent surface dipole term. The extracted slopes in Fig. 3(a) for runs 1 and 2 are 150×10^{-5} and 290×10^{-5} eV/K, and for Fig. 3(c) are 100×10^{-5} and 240×10^{-5} eV/K, respectively. The difference in slope between the two runs may be associated with differing amounts of sulfur reaching the surface. The changes we have observed for $\phi_-(\partial\phi_-/\partial T)$ are much larger (and of opposite sign) than what would be expected for clean Cu on the basis of calculations and measurements at high temperatures.[9] This may be related to the S coverage (i.e., reconstruction occurring at low temperatures), or it may indicate an error in the interpretation of the data. There is, however, some evidence that favors the first of the two explanations. After a 7-h heat treatment of a Cu crystal (which presumably brings S to the surface), Gartland *et al.*[10] measured the electron work function as a function of temperature, finding a change of $+0.25$ eV from room temperature to 400 °C. This gives $\partial\phi_-/\partial T \sim 65\times10^{-5}$ eV/K, which is twice the magnitude of that observed for "clean" Cu, and of the same sign as the change we observed. On the other hand, it is possible that the inability to rotate the manipulator on axis combines with the low fraction of the linear tail (Fig. 2, inset) at lower temperatures and leads to an anomalously large change in both $|\phi_-|$ and $|\phi_+|$. This could be due simply to the error in determining the "zero-energy" point, $\phi_- - \phi_g$ (see Fig. 2). Pursuing this argument, if one were to assume that $\partial|\phi_-|/\partial T$ were approximately -10^{-5} (in agreement with expectations for *clean* copper), then $\partial|\phi_+|/\partial T$ would also be negative and much smaller than the straightforward analysis (Fig. 3) yields. This may provide one possible explanation for the reduction in the fraction of positrons inelastically scattered at low temperatures, since one would expect that the larger normal escape velocity associated with the larger negative work function would lead to a reduction in the trapping rate.

The negative change in $\phi_- - \phi_g$ [Fig. 3(a)] is larger than the positive change in ϕ_+ [Fig. 3(c)] by an amount which leads to the slope in Fig. 3(b). The reason for this dependence on temperature is not understood at present, but it could result from a real change between the bulk chemical potentials of the positron and electron, or from dynamic correlations affecting the dipole as the positron leaves the sample. A change in the bulk chemical potential (μ_+ or μ_-) could be partially associated with thermal expansion or contraction of the lattice.

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