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Dependence of the positron reemission probability on the positron work function of a metal surface

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We have measured the probability y of positron reemission from Ni(100) and Cu(111) surfaces as the positron work function ϕ_+ is varied by depositing a fraction of a monolayer of K. We find that y increases monotonically with $-\phi_+$. The measurements agree with either of two possible models: an "ion-neutralization" mechanism that causes slower positrons to form positronium more effectively, or a positron emission rate proportional to the density of final states of the emitted positron.

It is now well established that the positron work function for certain metals with high electron density can be negative.[1] A positron implanted into such a metal thermalizes, and then can diffuse to the surface where it may become stuck in a surface bound state or be reemitted as a slow positron or positronium atom (Ps). Although the positron-metal surface interaction has been the subject of much experimental and theoretical work, the dynamics of the emission process and the mechanism that determines the branching ratio between positron emission and Ps emission or surface state formation are only partly understood. It has recently been shown that positrons reemitted from a metal with a negative positron work function ($\phi_+ < 0$) have a narrow Maxwell-Boltzmann energy distribution peaked at $(-\phi_+ + kT)$ with essentially no shake-up or energy loss tail.[2] It must be mentioned that the apparent energy loss reported by Wilson and Mills[3] was actually a loss of energy in the normal direction due to elastic scattering. On the other hand, the Ps emission spectrum exhibits a step at the negative Ps work function ($\phi_{\mathrm{Ps}} < 0$) and a tail extending to low energies. From these observations, one concludes that while positron emission occurs adiabatically, the Ps emission process is non-adiabatic and leaves the metal in a one-hole excited state.[4,5] In order to learn more about the emission processes, we would like to find out what determines the relative probabilities of positron emission, Ps emission, and Ps surface state formation. To this end we report measurements of the slow positron yield as a function of ϕ_+. The observed dependence of the positron yield on the emission energy is similar to what is observed for ion neutralization.[6,7] The data is thus consistent with a model in which an emitted positron must pass through a region outside the surface where Ps formation may occur. The data can also be explained by the positron emission rate being proportional to the density of final states of the emitted positron.

In an earlier study[8] the positron yield was observed for Al and Cu samples as ϕ_+ was varied by changing the orientation of the crystal face and the temperature of the sample. For the Cu samples, different values of ϕ_+ were also obtained by bringing S impurities to the surface by heat treatment. The possibility that systematic differences due to changes in bulk trapping and diffusion rates occur in data taken under such different conditions prompted us to perform new measurements in an improved manner. In the present experiment, the slow positron yield and work function of a Ni(100) or Cu(111) sample were measured in a single sweep while K was being deposited continuously on the initially clean surfaces. Adsorbed K is partially ionized and thus reduces the surface dipole layer,[9] causing the electron work function (ϕ_-) to decrease and ϕ_+ to increase by equal and opposite amounts ($\Delta\phi_- = -\Delta\phi_+$).[10] The positron emission energy ($-\phi_+$) changed smoothly from 1.4 eV (0.4 eV) for Ni(100) [Cu(111)] to zero with only a very dilute coverage of K, approximately 0.05 ML (0.02 ML), where a monolayer (ML) is defined to be one adsorbate atom per substrate surface atom. The repulsive mutual interaction between the K ions prevents the formation of islands that would create patches of varying work function.[9] Since the coverage is dilute and uniform, we conclude that the measurements have been made under essentially constant sample conditions as the work function is varied.

The experiments were performed with a magnetically guided positron beam in an ultrahigh vacuum chamber at a pressure of 2×10^{-10} torr. The samples were prepared by Ar-ion bombardment followed by annealing. The surface order of the samples was examined using low energy electron diffraction (LEED) and the surface contamination was measured with a double-pass cylindrical mirror Auger electron spectrometer. After the surface preparation, the Ni surface was contaminated with 0.15 ML of C, 0.02 ML of S and <0.002 ML of O. Similarly, the Cu surface was contaminated with 0.05 ML of C, 0.04 ML of S and <0.002 ML of O. Potassium was evaporated from a SAES getter source. The source was allowed to warm up behind a shutter before K was deposited on the sample in order to achieve a constant evaporation rate. Figure 1 shows the change in the electron work function during a typical evaporation. The reflected positron yield was measured with a channeltron as in Ref. 11 using a multiscaler to obtain a continuous sequence of 32 channel spectra. Each point in Fig. 1 was deduced from a separate spectrum by determining the sample bias required to turn around the positron beam. Since the ini-

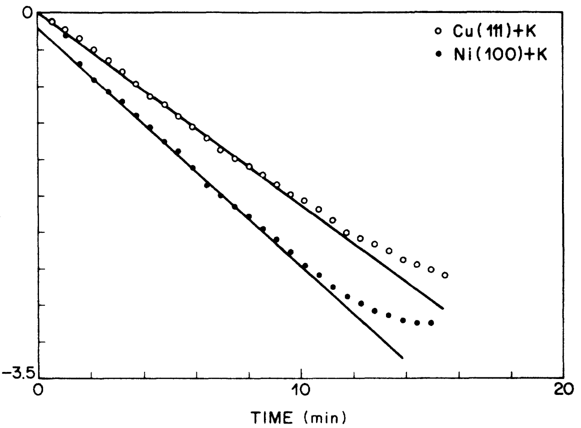

FIG. 1. The change in the electron work function for Ni(100) and Cu(111) as a function of K evaporation time.

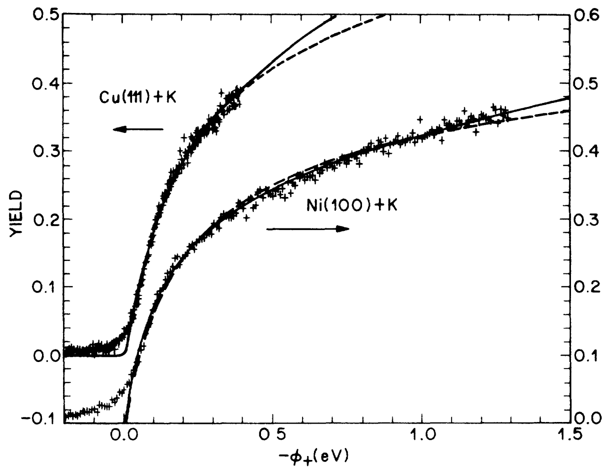

FIG. 2. The slow positron yield for Ni(100) and Cu(111) versus the positron work function. The curves through the data points are fits to the models discussed in the text: solid — resonance tunneling model of Yu and Lang with $\gamma=1.1a_0^{-1}$ [Eq. (1)]; dashed — positronium production proportional to the density of final states [Eq. (3)]. Other parameters used in the fits are listed in Table I.

tial linear decrease of ϕ_- with K coverage is well known,[9] we conclude from the data that the K deposition rate and $\partial\phi_+/\partial t$ are quite uniform in time over the small range of work function change relevant to our measurements. Presumably the line through the Ni data does not intersect the origin because the evaporator had not quite reached equilibrium when the shutter was opened. On the other hand we believe that equilibrium was established before the start of the runs presented in Figs. 2 and 3.

The slow positron yield for Ni(100) and Cu(111) is shown versus the positron work function in Fig. 2. The yield was measured with the channeltron and corrected for background by alternately biasing the sample positive and negative with respect to an electrode in front of the sample. The channeltron counts were recorded by the multiscaler as the K was being evaporated. The initial value of ϕ_+ was obtained from the positron emission spectrum taken prior to the K deposition. The zero of ϕ_+ was taken to be the point where the yield extrapolated to zero.[10] The absolute yield was determined for the clean surface using a NaI(Tl) detector to measure the γ-rays resulting from positron annihilation at the sample. To take into account the annihilation of positrons in the bulk material the yield was corrected by increasing it 25% for Ni (positron implantation energy 2.7 keV) and 50% for Cu (positrons implanted at 4.7 keV). The clean surface slow positron emission probabilities extrapolated to zero implantation energy are 0.45 ± 0.05 for Ni(100) and 0.39 ± 0.05 for Cu(111).

The solid curves through the data points shown in Fig. 2 are derived from the resonant electron transfer ion-neutralization model of Yu and Lang.[7] In this model, as an ion travels away from the surface normal with constant velocity v_p along z, resonant tunneling takes place between its valence level and the metal states at the same energy. The ion is assumed to move along a classical trajectory and to have a nondegenerate valence state of energy ε_p lying in the metal conduction band. The metal is treated as a noninteracting Fermi gas (jellium) of work function ϕ_-. The width $\Delta(z)$ of the valence state due to its interaction with the metal is assumed to fall off exponentially with the distance of the ion from its equilibrium position on the surface at $z=0$: $\Delta(z)=\Delta_0\exp(-\gamma z)$. Far from the surface the difference between the valence energy and the Fermi level is given by $\varepsilon_p(\infty)-\varepsilon_F(\infty)=\phi_- - I$, where I is the ionization potential of the atom. Closer to the surface, the energy is lowered by the image interaction of the positive ion so that $\varepsilon_p(z)-\varepsilon_F(z)=\phi_- - I+e^2/[4(z-z_{\mathrm{im}})]$, where z_{im} is the image plane of the metal. Resonant tunneling becomes very important at the level crossing point z_c outside the surface: $\varepsilon_p(z_c)-\varepsilon_F=0$. The probability of the atom's escape without being neutralized is given to good approximation[7] by $P^+\simeq\exp[-2\Delta(z_c)/\hbar\gamma v_p]$. For the case of slow positron emission, the classical trajectory approximation is crude, as the positron has such a small mass. Nevertheless, slow positrons are emitted from negative work function metals with a narrow energy distribution peaked about the surface normal,[2] so that the velocity $v_p=(-2\phi_+/m)^{1/2}$ is a well defined parameter. Since the positronium ionization potential 6.8 eV is larger than the Fermi level of Ni or Cu (ϕ_- of 5.22 and 4.94 eV, respectively, for the clean surfaces[12]) the neutral valence level ε_p always faces filled metal states as the work function of the metal is varied. Lowering the electron work function of the metal has two effects: it moves the level crossing point z_c closer to the surface and lowers the positron exit velocity, thus increasing the possibility for electron tunneling from the metal, and decreasing the slow positron yield.

The data in Fig. 2 have been fitted with the expression

$$y=y_0\exp\left[-(-b/\phi_+)^{1/2}\exp\left[-\frac{\gamma e^2}{4(I-\phi_-)}\right]\right], \tag{1}$$

where $b = 2m[\Delta_0 \exp(-\gamma z_{\rm im})/\gamma\hbar]^2$. Values of $z_{\rm im} = 1.5a_0$ and $\gamma = 1.1a_0^{-1}$ were used in the fits in Figs. 2 and 3, where a_0 is a Bohr radius. The value of $z_{\rm im}$ corresponds to $r_s = 2$ jellium and the value of γ to a metal with $\phi_- \sim 4$ eV. The data can also be fit with γ between zero and $2a_0^{-1}$. The fitting parameter y_0 can be viewed as the branching ratio for the emitted positrons that are able to reach a low enough electron density outside the metal to form positronium or escape compared to those that are immediately trapped into the surface state.

If we set $\gamma = 0$ in the expression above, the fitting expression becomes

$$y = y_0 \exp[-(-b/\phi_+)^{1/2}]. \tag{2}$$

This is the form the yield would take[6,8,13,14] if as the positron leaves the surface, it passes through a region of thickness δz in which it can either pick up an electron to form positronium or emit an electron-hole pair and fall into the image potential well at the surface. We assume that (1) the time δt spent by the positron in the region δz is just what one would calculate assuming the velocity of the positron is $(-2\phi_+/m)^{1/2}$; and (2) the positron gets captured at a constant rate Γ while it is in the region δz. The probability that the positron escapes as a free particle is $\exp(-\Gamma\delta t)$, or $b = \frac{1}{2}m\Gamma^2\delta z^2$. This model gives a reasonable fit to the data with $b \simeq 0.2$ eV. For $\delta z = a_0$, the positron capture rate is then $\Gamma \simeq 10^{-15}\ \text{sec}^{-1}$.

A nearly identical fit results when we use a density-of-final-states (DOFS) model (dashed curves in Fig. 2). We assume that the emission of positrons and positronium and the formation of the positron surface state all proceed via some interaction V as discussed in Ref 4. The rate for each process will be given by Fermi's golden rule and thus will be proportional to the square of a matrix element times a density of final states. Under the assumption that the matrix elements are unaffected by changes in the surface dipole, each rate will be proportional to its density of final states alone. Since the emission energy of positronium is independent of changes in the surface dipole potential,[1,4] the density of final states for positronium emission and therefore the positronium formation rate should be constant. If we adopt the Platzman and Tzoar picture[15] of the positronium surface state, we will be led to the same conclusion concerning the surface state formation rate because of the relative insensitivity to the surface dipole of the positronium binding energy.[16] The only variable rate is thus the positron emission rate which will be proportional to its density of final states $\sim(-\phi_+)^{1/2}$. The positron emission probability is then

$$y = \frac{y_0(-\phi_+)^{1/2}}{(-\phi_+)^{1/2} + b}, \tag{3}$$

where b is a constant taking into account the rates for positronium emission and surface state formation, and y_0 is the positron yield extrapolated to infinite emission energy.

Table I gives the parameters for the fits to the data using the three models. In the case of the DOFS model, it is remarkable that the parameter y_0 is approximately unity, and the parameter b is close to the same value for Cu and Ni. The parameter b for the ion neutralization model with $\gamma = 0$ is close to the earlier value found for Al and Cu, 0.27 eV, reported in Ref. 1.

TABLE I. Fitting parameters y_0 and b for the models discussed in the text.

	Ni(100)		Cu(111)	
	y_0	b	y_0	b
DOFS model	0.92	1.16	1.07	1.12
Yu-Lang model, $\gamma = 0$	0.71	0.28	0.83	0.21
Yu-Lang model, $\gamma = 1.1$	0.46	25	0.51	69

The models do not fit well for small positron emission energies because we need to take into account the thermal distribution of positrons in the solid and the fraction of positrons that do not thermalize. In Fig. 3 we show the improved agreement that results for Yu and Lang's model when we include a positron temperature of $kT^* = 0.032$ eV and a nonthermal fraction 0.4 having an energy distribution derived from our measurement shown in the inset.

Our new experimental results are in excellent agreement with our earlier measurements,[8] but despite the improved accuracy and reliability of the new data, we are unable to distinguish between three different models that predict the positron yield versus positron work function. All of the models are rather naive since they ignore recoil effects and many body interactions. Nevertheless, it is clear directly from the data that the yield has a simple dependence on the square root of the negative affinity, i.e., the positron emission velocity. Independent of the details of a model for the positron emission process at a surface, it would seem that when positrons are able to reach the low electron density region at the metal surface they should form some positronium. Thus as the sign of

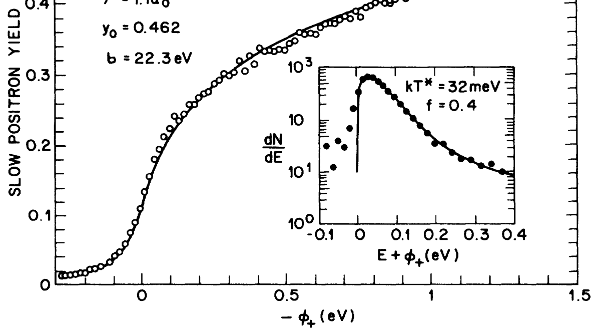

FIG. 3. The slow positron yield for Ni(100) with the fit to Yu and Lang's resonance tunneling model (solid line in Fig. 2) convolved with the actual measured energy distribution of emitted positrons depicted in the inset.

the positron work function changes from positive to negative a new channel for positronium production should appear. The momentum dependence of the matrix element for the direct positronium production that occurs for $\phi_+ > 0$ could differ from that associated with this new channel due to positrons in the low electron density region. It might be interesting to look for such an effect in the angle-resolved positronium momentum density.[5]

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