Auger-Mediated Sticking of Positrons to Surfaces: Evidence for a Single-Step Transition from a Scattering State to a Surface Image Potential Bound State

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We present the observation of an efficient mechanism for positron sticking to surfaces termed here Auger-mediated sticking. In this process the energy associated with the positrons transition from an unbound scattering state to a bound image potential state is coupled to a valence electron which may then have sufficient energy to leave the surface. Compelling evidence for this mechanism is found in a narrow secondary electron peak observed at incident positron kinetic energies well below the electron work function.

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Recently, positrons [1] have been shown to be very effective in probing surfaces and reduced dimensional systems such as nanoparticles, which possess high surface-to-volume ratios. If positrons become trapped in image potential surface states before annihilation, they can provide a means of selectively sampling the top most layer of a material or nanostructure due to the fact that such states typically extend about one atomic layer below the surface. Subsequent annihilation of surface trapped positrons with core or valence electrons results in signals (e.g., annihilation induced Auger electrons [2] or annihilation gamma rays [3]) containing crucial information about the composition of the outermost regions of nanomaterials.

In this Letter, we present experimental evidence for an efficient quantum mechanism for depositing positrons directly into surface states through a single step. In this process, the energy associated with the positron transition from an unbound scattering state to a bound surface state is coupled to a valence electron which may then have sufficient energy to leave the surface. Because of its similarity with the Auger transition in solids, this process has been termed Auger-mediated sticking (AMS). The quantum nature of the AMS follows from the fact that the de Broglie wavelength of a 1 eV positron (about 12 Å) is an order of magnitude more than the width of the surface potential well (about 1 Å) [4]. Similar ideas have been suggested in theoretical models [5–7]; however, no experimental evidence of this mechanism was available until now. The AMS process schematized in Fig. 1 is related closely to the Auger deexcitation of atoms [8] or molecules [9] near surfaces, which has been studied for decades in various fields.

Here we provide direct experimental confirmation of the AMS process through measurements of electron energy spectra resulting from very low energy positron bombardment (1.5–7 eV). The strongest evidence for the AMS is found in a narrow electron peak observed at incident positron kinetic energies well below the electron work function value. The present experiment also allowed us to determine the positron sticking probability as a function of incident particle energy and to obtain an independent measurement of the positron binding energy at the surface [10]. The fact that this new mechanism has an efficiency exceeding 10% at positron energies ~1 eV proves that it will be possible to use low energy positron beams to selectively probe the surfaces of fragile systems such as nanoparticles and biomaterials and to obtain Auger signals which are completely free of secondary electron background.

The experiments were carried out using time of flight (TOF) positron annihilation induced Auger electron spec-
trometer (PAES) [11], which uses a magnetic bottle analyzer [12]. Positrons from a Na-22 source are guided to the sample using $E$ and $B$ field. The emitted electrons are detected by a multichannel plate. The energy of the emitted electron was calculated from the electron TOF, which was determined from the time difference between the electron and annihilation gamma ray detections (see Ref. [11]). An Au sample (a 99.985% pure polycrystalline foil, 0.025 mm thickness) was sputter cleaned every 12 h while a Cu(100) sample (a 99.9% pure single crystal 10 mm diameter × 1 mm thickness) was sputter cleaned followed by annealing at 740 °C every 12 h. The incident beam profile at 0 V sample bias was fitted with a Gaussian of 0.4 eV FWHM and maximum at 0.65 eV. Ninety-nine percent of the positrons have energy less than 1 eV, which is referred to as the beam energy. The incident positron beam energy was increased by negatively biasing the sample.

The primary evidence for the AMS process in Cu is shown in Fig. 2(a), where the normalized energy spectrum taken at different positron beam energy $E$ (1.5–7 eV) is plotted. In each spectrum the large peak at low energies (<10 eV) corresponds to electrons that are emitted as a result of positron impact at the sample surface. The much smaller peak at about 60 eV is the PAES peak [2].

AMS can be distinguished from another process in which the final state of the incident positron is the bulk state. In the latter process, the maximum kinetic energy of the outgoing electron (as measured outside the sample surface) is given by

$$E_{\text{max}} = E - \phi^- + \phi^+, \quad (1)$$

where $E$ is the incident positron energy (measured from the vacuum level) and $\phi^- = 4.65$ (4.8) eV is the electron work function in Cu (Au) and $\phi^+ = -0.02$ (+0.9) eV is the positron work function [13–16]. Both work functions are measured from the vacuum level with positive sign below the vacuum level. The electrons can escape the sample if $E_{\text{max}} > 0$ eV, which implies that $E$ should be greater than $\phi^- - \phi^+$. Hence, for incident positron kinetic energies of less than 4.7 (3.9) eV there should be no secondary electron emission according to this mechanism. In the case of AMS, the positron excites an electron-hole pair while dropping to the surface state. The energy to dissipate is the initial positron kinetic energy plus the positron binding energy to the surface; thus, we have

$$E_{\text{max}} = E + E_{ss} - \phi^- , \quad (2)$$

where $E_{ss}$ is the surface binding energy of the positron measured from the vacuum level (with positive sign below the vacuum). Annihilation induced processes including Auger transitions and $\gamma$-ray emission can lead to the emission of electrons with energies as high as the Auger transition energy and 511 keV, respectively. However, such processes would lead to the formation of broad electron peaks. In our experiments, we found a narrow electron peak even when the incident kinetic energies of the positrons were less than 3 eV. This can be explained from Eq. (2) by considering the process in which the electron excited from the Fermi sea escapes from the surface if the positron incident energy is greater than a certain threshold of about 2 eV since $E_{ss}$ is of the order of 3 eV in most metals [15].

Figure 2(b) shows a typical electron spectrum of Cu. The large peak centered at 3 eV corresponds to AMS induced electrons. Figure 2(c) shows the spectrum similar to AMS induced electrons. Figure 2(d) except the beam energy is below the threshold for electron emission; thus, one can notice the absence of the low energy (AMS) peak: the sticking is still taking place, but electron emission outside the sample is energetically prohibited. The same broad background between 5–30 eV can be seen in both Figs. 2(b) and 2(c). This feature is presumably the low energy electron tail associ-
The AMS peak integrated intensity was also used to estimate the positron sticking probability \( S(E) \). It has been assumed that the transition of the positron to the surface state is always associated with an electron-hole pair excitation. Hence, the sticking probability can be written as

\[
S(E) = \frac{N_{\text{AMS}}/N_{\text{\(e+inc\)}}}{P(E)r(E)},
\]

where \( N_{\text{AMS}} \) is the integrated intensity of the AMS peak, \( N_{\text{\(e+inc\)}} \) is the number of incident positrons, \( P(E) \) is the escape probability for the excited electrons [17], and \( r(E) \) is that fraction of the excited electrons which have enough energy to escape by overcoming the work function \( \phi \). The ratio \( r(E) \) is therefore given by

\[
r(E) = \frac{\int_{E_E}^{E_F} g(E)dE}{\int_{E_E}^{E_F} g(E)dE},
\]

where \( E_F \) is the Fermi energy of the metal, \( E_a = E_F - (E + E_{ss} - \phi) \), \( E_b = E_F - (E + E_{ss}) \), and \( g(E) \) is the density of states [18]. The number of incident positrons \( N_{\text{\(e+inc\)}} \) was estimated using

\[
N_{\text{\(e+inc\)}} = N_{ss} + N_{\text{Ps}} + N_{\text{ref}},
\]

where \( N_{ss} \) is the number of positrons trapped in the surface state, \( N_{\text{Ps}} \) is the number of positrons that form positronium, and \( N_{\text{ref}} \) is the number of positrons that are reflected from the surface. \( N_{\text{Ps}} \) and \( N_{ss} \) are related by \( N_{\text{Ps}} = f(N_{ss} + N_{\text{ref}})/(1 - f) \), where \( f \) is the fraction of incident positrons that form positronium (\( \sim 0.5 \)), determined as in Ref. [10]), while \( N_{\text{PAES}} = C N_{ss} \) where \( N_{\text{PAES}} \) is the integrated intensity of PAES peak and \( C \) is the probability that a positron trapped in the surface state will annihilate with a core electron (4.6%) [14]. Taking 0.2 as the upper limit of \( R = N_{\text{ref}}/N_{\text{\(e+inc\)}} \) [19], the total number of incident positron can be written as

\[
N_{\text{\(e+inc\)}} = \frac{N_{\text{PAES}}}{0.8(1 - f - R)CT_{\text{PAES}}},
\]

where \( T_{\text{PAES}} = 0.45 \) (0.58) is the fraction of Auger electrons for Cu (Au) that are transmitted to and detected by our analyzer. The sticking probability \( S(E) \) estimated in this way is plotted in Fig. 4(c), and its trend is consistent with the calculations by Walker et al. [7], especially for the Cu data. The reduced probability \( S(E) \) observed for the Au data can be explained by a stronger screening. Walker et al. have used a screening parameter \( \mu = 0.6\mu_{TF} \) (where \( \mu_{TF} \) is the Thomas-Fermi screening parameter) in their calculation; however, in gold this parameter is bigger [20]. The prediction of Walker et al. that \( S(E) \) will vanish as \( E \to 0 \) cannot be resolved by the present experiment. Here we are only interested in high positron surface sticking rates for nonzero positron energies.

We have reported experiments that provide strong evidence for a sticking process in which a low energy positron, incident on a surface, makes a direct transition from...
an unbound scattering state into an image potential surface state, resulting in the emission of a secondary electron. We have termed this process Auger-mediated sticking (AMS) because the energy lost as the positron makes a transition to the bound state is given to an outgoing electron. Measurements of the incident beam energy at which the secondary peak first appears indicate a threshold almost 3 eV lower than the value that would be expected if the positron were making a transition to a bulk state. These measurements were used to obtain the first estimates of the surface state binding energy at room temperature. The AMS peak integrated intensity was used to estimate the sticking probability of positrons to surface and it is found to be in qualitative agreement with the theory [7].

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[17] $P(E)$ is from Ref. [8] with $\alpha = 0.7$ and $\beta = 6$.
[18] We have assumed a constant density of states near $E_F$.
[24] The error bars are predominantly due to the uncertainty in the intercept as measured in Figs. 4(a) and 4(b).