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Positron Annihilation Induced Auger Electron Emission

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ABSTRACT

We report on measurements of Auger electron emission from Cu and Fe due to core hole excitations produced by the removal of core electrons by matter-antimatter annihilation. Estimates are developed of the probability of positrons annihilating with a 3p electron in these materials. Several important advantages of Positron annihilation induced Auger Electron Spectroscopy (PAES) for surface analysis are suggested.

Conventional Electron induced Auger Electron Spectroscopy (EAES) is a powerful method for the elemental analysis of surfaces, but it is not without limitations.[1] The energetic electron beam used in conventional AES causes a large secondary electron background, damage to organic systems, charging problems in insulators, and desorption of adsorbed layers. Further, an electron beam with sufficient energy to produce the core hole excitations necessary for EAES penetrates deep below the surface. This fact limits the surface specificity of EAES as the signal represents an average over a depth corresponding to the 10-30Å inelastic mean free path of the Auger electrons.

Recently, Weiss et al.[2] demonstrated a fundamentally new mechanism for the excitation of Auger electrons. In this process a core-hole excitation is created by the removal of an inner shell electron via matter - antimatter annihilation (not by collisional ionization). The atom then relaxes by emitting an Auger electron. This mechanism makes possible a new surface analytic technique: Positron annihilation induced Auger Electron Spectroscopy (PAES). PAES permits the use of low incident beam energies and therefore eliminates the problems of beam damage and the large background (energy conservation forbids the production of collisionally excited secondaries with energies larger than the incident beam energy). In addition, in systems in which positrons can be localized at the surface in a surface state[3], PAES may provide a way of obtaining an Auger electron spectrum from the topmost atomic layer.

PAES measurements were performed on Cu and Fe at the University of Texas at Arlington using a magnetically guided positron beam.[4] A trochoidal energy spectrometer equipped with a 25mm diameter microchannel plate (MCP) was used to energy analyze and detect electrons emitted from the sample. A Nd-Fe-B magnet was placed behind the sample to reduce the angular spread of the Auger electrons at the spectrometer[5]. A NaI(Tl) detector was used to measure annihilation gamma rays emitted in coincidence with the Auger electrons. The polycrystalline Cu and Fe samples were cleaned by ion bombardment prior to the measurement.

Energy spectra of electrons leaving the copper surface are shown in figure 1. The signal was obtained from the detection of electrons in coincidence with annihilation gamma rays resulting from the removal of core electrons. In figure 1a, the sample was biased at -15 V. with respect to ground (the reference of the energy spectrometer), the positrons were incident on the surface with a kinetic energy of 25eV and the ratio of the magnetic field at the spectrometer to that at the sample[5], B_0/B_1, was 0.32. The peak at ~60 eV corresponds to the $M_{23}M_{45}M_{45}$ Auger transition. In figure 1b the voltage on the sample was -5V. The peak position can be seen to be ~ 10eV lower than in 1a demonstrating that the peak is due to electrons leaving the sample surface. The data shown in figure 1c was obtained with a ratio B_0/B_1 field of 0.03. A substantial reduction in the width of the peak can be seen (The contribution to the width of the peak due to the angular spread of the Auger electrons[6] scales like $E(B_0/B_1)$.

The measured energy distribution of electrons leaving the Fe surface is shown in figure 2. The signal plotted in figure 2a is obtained directly from the MCP detector. Figure 2b show the same spectrum taken with the $e^- - \gamma$ coincidence signal. The positrons were incident on the surface with a kinetic energy of 15 eV. The peak at ~40 eV corresponds to the Fe $M_{23}VV$ Auger transition.

The solid curves shown in figure 1 and 2 represents fits to a function[6] obtained by convoluting the

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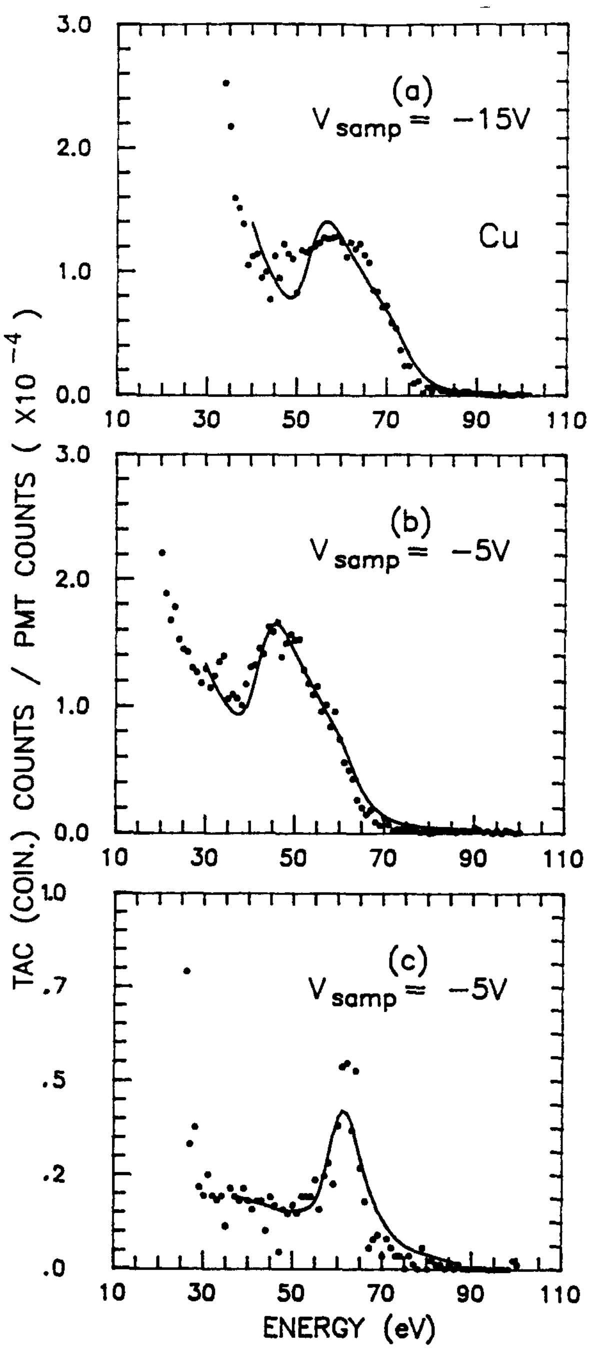

Figure 1. The energy spectra of electrons leaving polycrystalline Cu surface. The kinetic energy of the positions was 25ev, 15eV and 25eV in figs. 1a, 1b, and 1c respectively. TAC counts represent the detection of electrons in coincidence with annihilation gamma rays from the sample. PMT counts represent detected γ-rays. Solid lines represent a fit to a function obtained by convoluting the energy distribution for the 60eV (nominal) $M_{23}M_{45}M_{45}$ Auger transition for Cu measured using EAES with the instrument response function.

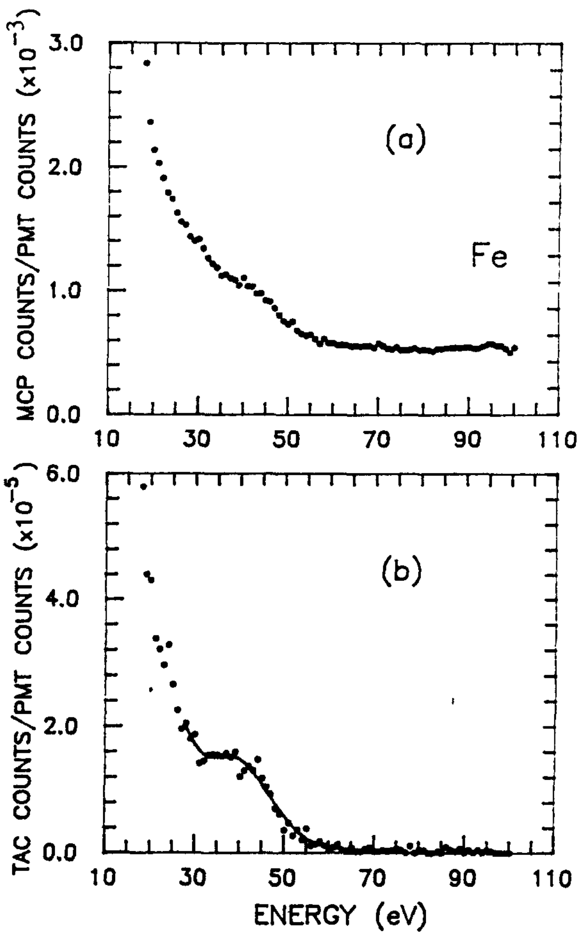

Figure 2. Energy spectra of electrons leaving polycrystalline Fe surface irradiated by 15 eV. positrons. In Fig. 2a The signal in fig. 2a was obtained from MCP singles counts. The signal in 2b was obtained from γ-e$^-$ coincidence.

instrumental response function for our spectrometer with the energy distribution for Cu $M_{23}M_{45}M_{45}$ Auger transition and the Fe $M_{23}VV$ Auger transition respectively taken from EAES measurements.[7,8] The width of the peak and the centroid position came directly from our instrumental response function and the previous EAES measurement and were not adjustable parameters. The number of Auger electrons per incident positron emitted into the solid angle subtended by the detector per incident positron, A(Ω), was determined by integrating under the Auger peaks in the electron energy spectra giving[6]: $A(\Omega) = 9.8(4) \times 10^{-4}$ $M_{23}M_{45}M_{45}$ Auger electrons/e^+-2π sterad. for Cu and $A(\Omega) = 2.0(7) \times 10^{-4}$ $M_{23}VV$ Auger electrons/e^+-2π sterad. for Fe.

We estimate[2] $\sigma(M_{23})$, the probability that a positron trapped at the surface will result in the annihilation of electrons in the M_2 or M_3 levels, from our results using equation 1:

1) $\sigma(M_{23}) = A(\Omega)(4\pi/\Omega)(1/f_s)(1/b)(1/SM_{23})$

where: $A(\Omega)$) is defined above; ($4\pi/\Omega$) takes into account the solid angle of the detector; f_s is the fraction of incident positrons that get trapped at the surface; b is the fraction of Auger electrons (resulting from annihilations of positrons) that escape into the solid angle, Ω , without suffering significant energy loss; and SM_{23} is the probability that an $M_{2,3}$ core hole results in a $M_{23}M_{45}M_{45}$ Auger transition. As in reference 2 we use $\Omega/4\pi = 0.5$, $SM_{23} = 1$, $f_s = 0.5$ and $b = 0.86$ Inserting these values into equation 1 we calculate $\sigma(M_{23}) = 0.46(2) \times 10^{-2}$ for Cu and $\sigma(M_{23})$) $= 0.96(3) \times 10^{-3}$ for Fe. A similar calculation produced an estimate of $\sigma(M_{23}) = 3.7(7) \times 10^{-2}$ for Ni(110) based on experimental measurements carried out at Brookhaven National Laboratory[2] on a well annealed single crystal sample.

The estimates of $\sigma(M_{23})$ from the Ni(110) measurement is within a factor of 2 of theoretical estimate of $\sigma(M_{23}) = 7 \times 10^{-2}$ for Ni obtained from the calculations of Nieminen and Jensen of the fraction of positrons trapped in a Ni surface state that annihilate with core electrons[9] An upper bound of 6×10^{-2} can be placed on the value for $\sigma(M_{23})$ in Cu from the calculations of the annihilation of positrons with 3p electrons in bulk Cu.[10] We would expect that $\sigma(M_{23})$ would be larger in Fe than in Cu and Ni based on the lower binding energy of the Fe M_{23} core levels. The fact that values of $\sigma(M_{23})$ for Cu and Fe estimated from our measurements are lower than the theoretical values probably reflects our overestimation of b, because we did not take into account subsurface defect trapping or the effect of surface impurities. Surface impurities may also lower $\sigma(M_{23})$ by reducing the annihilation rate of positrons in a surface state with substrate atoms. Future experiments will examine the affects of defects and overlayers on the PAES signal as well as the degree of surface specificity that is possible with PAES.

The coincidence measurements shown in figure 1c indicate that there is almost no background on the high energy side of the peak. The "background" on the low energy side of the peak is due to Auger electrons that have lost energy on the way out of the sample and Auger electrons with large transverse components of momentum. The onset of collisionally induced secondaries can be seen on the left at <30eV. This demonstrates the ability of PAES to eliminate the secondary electron background which plagues EAES. In the limit where the signal is much smaller than the background, the ratio, R, of charge dose, Q^+,needed for PAES to the charge dose , Q^-,needed for EAES for the same signal to noise ratio and elemental concentration can be written:

$$(2) \quad R = Q^+/Q^- = [(Y_o^-)^2 B^-] / [(Y_o^+)^2 B^+]$$

where Y_o^+ (Y_o^-) is the signal to background ratio obtained using PAES (EAES) for the pure element and B^+ (B^-) is the ratio of background current to incident current. Using values for $B^\pm$ and $Y_o^\pm$ obtained from our coincidence data and from A. Joshi et al.[1] we obtain $R = 3 \times 10^{-2}$. This thirty fold reduction in charge dose combined with a two order of magnitude reduction in beam energy, would result in a reduction in the total energy dose to the surface of more than three orders of magnitude using PAES as compared to EAES.

The implementation of PAES using intense positron beams[3] of $\sim 10^7$ positrons/sec which have recently become available (compared with the $\sim 10^4$ positrons/sec used in our measurements) should enable Auger analysis to be performed on fragile adsorbed layers, chemically unstable systems, and insulators, where conventional electron excitation methods cannot be used because of beam damage, electron stimulated desorption, or charging problems. Further, PAES should offer a high degree of surface specificity in systems in which positrons can become trapped in a surface state (metals and semiconductors).[3] In these materials the Auger electron signal should originate almost exclusively from the first atomic layer due to the fact that the positron surface state wavefunction dies off rapidly as a function of depth. Other possible applications of the positron annihilation induced Auger process include: 1. studies of the positron surface state 2. studies of Auger induced desorption, 3. identification of near surface defects containing impurity atoms, and 4. elemental mapping of surfaces when used in conjunction with scanning and reemission positron microscopes.

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