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LETTER TO THE EDITOR

Median penetration depths and implantation profiles for low energy positrons in Al

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Abstract. New measurements and Monte Carlo simulations for 1–30 keV positron implantation in Al are reported. The annihilation lineshape was measured as a function of positron energy E and the thickness of Al overlayers on a glass substrate. Median implantation depths vary as $117(6)\ E^{1.64(2)}$ Å, with E in keV. The implantation profiles differ significantly from the widely accepted Gaussian derivative form. The excellent agreement between experiment and simulation confirms the power of the Monte Carlo method in positron (and by implication electron) spectroscopies.

The analytical effectiveness of many surface and subsurface spectroscopies is ultimately governed by our understanding of energy loss mechanisms within a given material. For example, the electron inelastic mean free path is a vital parameter in quantitative AES and XPS; a description of primary electron penetration is also important, particularly in electron probe microanalysis. Monte Carlo simulations are commonly used to describe these processes. Measurements of electron inelastic mean free paths and penetration profiles are fraught with difficulties [1] and data interpretation is often ambiguous [2]. For this reason only qualitative comparisons between experiment and theory have been possible.

The situation for positrons, however, is potentially much better than for electrons; because of the positrons' distinguishability, trajectories can, in principle, be traced and an implantation profile obtained. Comparisons between Makhovian fits to the earlier Monte Carlo simulations of Valkealahti and Nieminen [3] (hereafter referred to as VN) and the only direct measurements of implantation profiles to date, by Mills and Wilson [4], (hereafter referred to as MW), show substantial disagreement, although median depths are acceptably reproduced. Furthermore, the MW results and the VN simulations only extend to 6 and 10 keV, respectively, whereas incident energies of up to 40 keV are used in positron implantation spectroscopy, a method which has recently emerged as a powerful depth-sensitive probe of subsurface defects [5]. Thus, studies using this method have to date had to rely on extrapolation much beyond the range of the data of VN and MW. For an adequate description of positron implantation and hence a reliable deconvolution of the experimental data, fresh approaches in both experimental technique and the simulation cross sections used to obtain the profiles are clearly required.

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In this letter we report measurements, using a new technique, and Monte Carlo simulations of positron implantation in Al at energies of up to 30 keV. The results suggest an implantation profile different from the widely used Gaussian derivative form.

The excellent agreement between theory and experiment that we observe is gratifying, in view of the fact that this is the first time to the authors' knowledge that Monte Carlo simulations of charged-particle slowing down have utilized conduction electron cross sections calculated from a fully first-principles theory *without adjustable parameters*, i.e. the random phase approximation (RPA). Since this method and these, or similar, cross sections are widely used to study electron transport [6–10], the present positron results also have important implications for studies of solids using electron spectroscopies.

MW, using a thin-foil transmission technique, obtained broadly peaked implantation profiles $P(z, E)$ for Al and Cu (z being the depth and E the incident energy). It has been argued that their non-zero values for $P(z, E)$ at $z = 0$, at variance with expectations for a perfectly absorbing surface, may have been rooted in an incorrect allowance for backscattering [10]. It is, however, important to note that fitting of the majority of positron subsurface data, although highly dependent on median penetration depths, is likely to be only weakly dependent on the exact profile shape. For this reason the median penetration depths $z_{1/2}$, measured by MW as a function of incident positron energy E between 1 and 6 keV, shown to satisfy the empirical law

$$z_{1/2} = (A/\rho)E^n \tag{1}$$

(in Å) where $A/\rho = 125(30)$ Å keV$^{-1.6}$ and $n = 1.6(1)$ for Al (ρ is the material density), have been used to fit 1–30 keV positron subsurface data in conjunction with the Gaussian derivative approximation to the Makhovian form for $P(z, E)$ first suggested by VN. The exception to the general success of this procedure has been in the modelling of positron fractions back-diffusing to a surface to form positronium [11]; in these measurements only the low-z part of $P(z, E)$ is sampled and its detailed form is thus of importance.

The method used in the Monte Carlo simulation is similar to that used by VN but with improved description of the inelastic cross sections. The scattering of positrons with Al 3s valence electrons is calculated assuming a free-electron gas using the dielectric formalism of Tung and Ritchie [6]. The full energy loss and angular dependences of the cross sections for electron–hole pair and plasmon excitations from [6] are included in the simulations. The only approximation we make to the exact random phase approximation formalism [6] is to assume that the positron cross sections are identical to those for electrons, which amounts to neglecting scattering events in which the final positron state lies below the Fermi energy. Since we are concerned with energies above 50 eV this is a valid approximation [12]. For scattering with the Al 1s, 2d and 2p core electrons we use the cross sections of Salvat *et al* [13]. The elastic scattering is described, as in VN, by the scattering of a plane wave off an atom in the crystal using a partial wave expansion. The simulations were performed for semi-infinite Al with a planar surface and normal incidence of the positrons. More than 10000 particle trajectories were calculated in each simulation. The positrons were followed until they either came back to the surface (backscattered positrons) or slowed down to an energy below 50 eV (implanted positrons).

We chose the termination energy of 50 eV since the distance travelled below this energy prior to thermalization is very short. Below 50 eV the mean positron energy E varies with time t approximately as $E(t) = 0.1t^{-1/2}$ eV ps$^{1/2}$ [14, 15]. Assuming a constant inelastic mean free path of 25 Å [16] and a thermalization time of 3 ps [15], both at

300 K, one can estimate the average diffusion length for positrons between 50 eV and thermalization to be approximately 200 Å by using the diffusion model for non-thermalized positrons of McMullen and Stott [16, 17]. The distance travelled between 50 eV and 1 eV can similarly be estimated to be only about 30 Å.

The experiments were performed in the UEA slow positron beam system. The samples consisted of an 0–8000 Å thick Al wedge evaporated on to a glass microscope slide and a series of thin (~7770 Å) Al foils set down on to similar substrates. The 99.99% Al was evaporated at ~40 Å s^{-1} at a vapour pressure of 10^{-3} Pa from a resistively-heated boron nitride-(BN) crucible (base pressure of 10^{-4} Pa). This procedure has been shown to produce homogeneous growth within ~1000 Å of the substrate [18]. The use of BN is believed to reduce oxygen contamination greatly [19]. During evaporation a linearly driven shutter passed over the substrate to produce the wedge, whose profile was measured by Michelson interferometry to an accuracy of ±100 Å. Electron micrographs have confirmed the uniformity of the film along its length. The density of the glass was measured to be 2.53(5) g cm^{-3} and the density of bulk and evaporated Al are 2.70 and 2.65 g cm^{-3} respectively. This matching of substrate and overlayer densities means that the correction for backscattering effects is expected to be negligible; the ratio of measured backscatter coefficients for the glass and Al used in this study was found to be 0.9(2), a result confirmed by our Monte Carlo simulations. The overlayer geometry is also common to samples studied by slow positron implanation spectroscopy.

The annihilation lineshape parameter S is used to deduce the position of annihilation within the sample. This parameter, defined as the ratio of a central section to the total area of the photopeak as measured by a Ge detector, measures the Doppler broadening of the annihilation linewidth, and reflects the momentum of the positron–electron pair at the moment of annihilation. For a system with overlayer thickness ζ, and with negligible thermalized positron diffusion, the measured S at a given incident energy E will consist of the sum of the fractional contributions from the overlayer F_A and substrate F_B ($=1 - F_A$):

$$S(E) = F_A(E)\, S_A + F_B(E)\, S_B. \tag{2}$$

Clearly at a particular incident energy half the positrons will annihilate from each state. At this energy the measured S will simply be the average of the overlayer (S_A) and substrate (S_B) values, and $\zeta = z_{1/2}$; in practice $z_{1/2}$ is deduced by measuring S as a function of E for a known ζ. Furthermore $P(z, E)$ for the overlayer material can be generated by taking the negative derivative of the substrate fraction with respect to overlayer thickness:

$$P(z, E) = -\,\mathrm{d}F_B(\zeta, E)/\mathrm{d}\zeta = [1/(S_B - S_A)]\,\mathrm{d}S/\mathrm{d}\zeta. \tag{3}$$

Figure 1 shows both experimental and Monte Carlo median penetration depths against incident positron energy on logarithmic axes. The results at 10 keV and below were obtained using the wedge sample. The low-energy data of MW are included in the figure and used in subsequent fits. A straight line fit to the whole data set yields a power law (equation (1) with $A/\rho = 117(6)$ Å $\mathrm{keV}^{-1.64}$ and $n = 1.64(2)$), in good agreement with the MW result. A fit to the UEA data only provides a slightly steeper gradient and lower A; however, a positron diffusion length of 300(50) Å is obtained for both the Al wedge overlayer and the glass substrate, measured by applying first iterative fitting procedures to $S(E)$ data using the Gaussian derivative form. This means that a significant fraction F_I(~21% at 5 keV and ~7% at 10 keV) of the incident positrons will diffuse to the interface and be annihilated with characteristic S parameter S_I. While this has proved

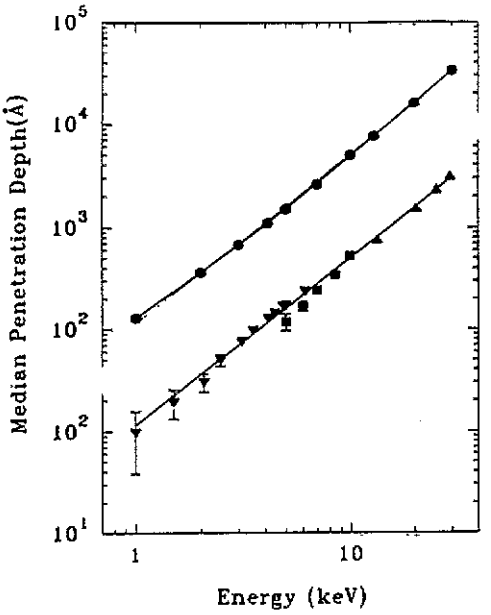

Figure 1. Measured median penetration depths in Al, $z_{1/2}$, for positrons of incident energy E. The symbols denote: ■, Al wedge; ▲, thin foils; ▼, MW (reference [4]); ●, Monte Carlo results. The full curve through the Monte Carlo data is the fit $z_{1/2} = 129\ E^{1.45+0.053\ln(E)}$ (Å). Full and dotted lines are straight line fits through the experimental and Monte Carlo data; both are well represented by $z_{1/2} = 116\ E^{1.64}$ (Å).

to be a limitation in the measurement of $z_{1/2}$ by this method at low energies, the high-energy data, for which F_I is negligible, can be accepted with confidence. The MW low-energy data are valid since diffusion within their sample did not have any serious effects on their (transmission) measurements. Also shown in figure 1 are the results of the UEA simulations (equation (1) with $A/\rho = 115(5)$, $n = 1.64(2)$), in remarkably good agreement with experiment.

On close inspection the Monte Carlo data exhibit a small positive curvature. This type of behaviour has been observed in electron measurements [20]. Following the approach of Katz and Penfold [21] the Monte Carlo results have been fitted using an energy dependent n given by

$$n(E) = \alpha + \beta \ln(E) \tag{4}$$

with $A/\rho = 129(1)$ Å keV^{-n}, $\alpha = 1.45(1)$ and $\beta = 0.053(3)$; this fit is also shown in figure 1.

To deduce implantation profiles for 7 keV positrons in Al from the raw wedge data a first-iterative correction for diffusion effects can be made. Full details of the correction procedure will be given in a later paper. In summary, the correction $\Delta S(z_{1/2})$ is first deduced such that $z_{1/2}$ becomes equal to the value predicted at 7 keV by the linear fit in figure 1. Independently we have calculated approximate interface fractions $F_I(\zeta)$ using a Makhovian profile and the measured positron diffusion lengths in Al and glass. We then deduce a value for the interface S parameter S_I from $\Delta S(z_{1/2}) = F_I(z_{1/2})S_I$ and thus obtain $\Delta S(\zeta)(=F_I(\zeta)S_I)$ for all ζ. These corrections are then applied to all the measured S values; the corrected data are shown in figures 2(a) and (3a) combined with cubic spline and Padé fits, the latter consisting of the ratio of two quadratics.

Differentiated curves (implantation profiles) are shown in figures 2(b) and 3(b) with the results of the Monte Carlo simulations. Padé and cubic spline fits are included to illustrate the magnitude of fluctuations arising as artefacts of the fitting procedure.

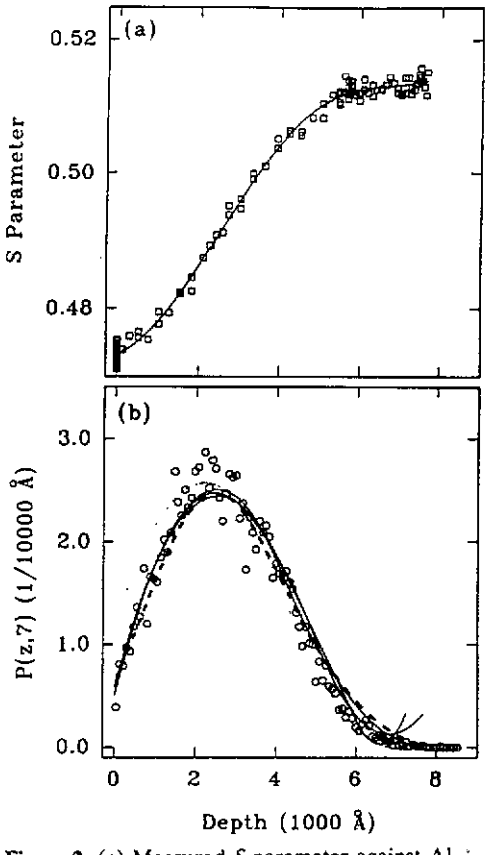

Figure 2. (*a*) Measured *S*-parameter against Al overlayer thickness ζ for 7 keV positrons; the data have been corrected for diffusion to the interface (see text). The full curve is a cubic spline fit through the data. (*b*) Implantation profiles $P(z, E)$ for 7 keV positrons in Al: O, Monte Carlo simulation; full curves are generated by differentiating cubic spline fits to the experimental data in (*a*) with one, two and three knots. The broken curve is produced by adopting a similar procedure to a Padé fit; the dotted curve shows a differentiated spline fit to the raw data before correction for diffusion to the interface.

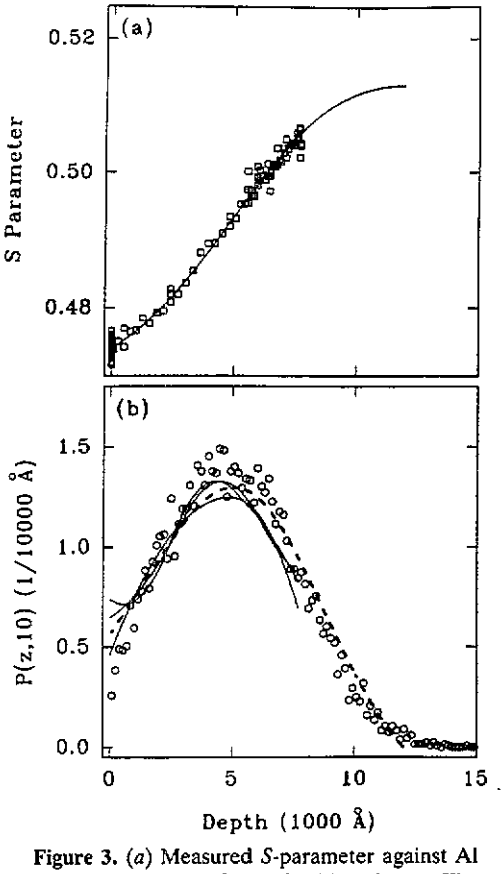

Figure 3. (*a*) Measured *S*-parameter against Al overlayer thickness ζ for 10 keV positrons. The full curve is a Padé fit through the data. (*b*) Implantation profiles $P(z, E)$ for 10 keV positrons in Al: O, Monte Carlo simulation; full curves are generated by differentiating cubic spline fits to the experimental data in (*a*) with one, two and three knots. The broken curve is produced by adopting a similar procedure to the Padé fit.

The dotted line for 7 keV (figure 2(*b*)) is obtained without performing the interface (diffusion) correction. The observed and calculated distributions show excellent agreement and demonstrate several common features. The abruptly ending high-z tail and low-z shoulder cannot be described by a Makhovian profile. In the experimental study the low-z shoulder is smeared by diffusion, giving rise to an apparent non-zero value at $z = 0$. These features are also apparent in the observations of MW. Moreover, the low-z behaviour supports the positronium observations of Nielsen *et al* [11]. The main experimental support for the Gaussian derivative profile is based on measurements of

integrated areas and is therefore unlikely to be particularly sensitive to the exact functional form of $P(z, E)$ [22]. In the light of the available experimental evidence we believe the parametrized fit

$$P(z, E) = -(\mathrm{d}/\mathrm{d}z)\exp[\![-\{(z/(z_0(E))[1 + z/z_0(E)]^2\}^{1.28}]\!] \tag{5}$$

where $z_0 = 2.58z_{1/2}$ (with $z_{1/2}$ given by equations (1) and (4)), which accurately reproduces our Monte Carlo simulations, gives a better representation of positron implantation in Al than the currently used Gaussian derivative form. Details of the parametrized fits for other materials will follow in a fuller paper; one should note, however, that the Al data are expected to describe adequately positron implantation in Si.

Several features of this work merit further investigation. The material independence of A/ρ in the power law relationship for $z_{1/2}$ (equation (1)), proposed by MW, seems unlikely. It is plausible that dependence on other parameters, particularly atomic number Z and E, exists. The authors are currently planning a series of measurements in high Z materials to test this hypothesis. Concurrently, the Monte Carlo codes are being extended to describe both positron and electron implantation in a wide range of materials.

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