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Range of slow positrons in metal overlayers on Al

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Polycrystalline Pd and amorphous PdTa films on Al substrates were studied by a variable energy positron beam and by Rutherford backscattering. Since positron diffusion in the overlayers is limited, the range follows directly from the Doppler broadening as a function of incident positron energy. To observe possible effects of positron backscattering, a sandwich of Al/Pd/Al was studied as well. It was found that the mean penetration depth is not described well by $\bar{z}(E) = A(\mu\text{g}/\text{cm}^2) \times E^{n(E)}$, if A and n are assumed to be material and energy independent.

Slow positron beams are utilized to characterize thin-film interfaces,^{1,2} solid-state reactions,³ and implantation damage.⁴ Crucial to such experiments is a proper description of the positron implantation profile, since that determines the accuracy of derived depth scales, and, therefore, location of interfaces and defects in the interface and overlayer regions. Generally a Makhovian distribution⁵ is assumed which still leaves the choice of a general parameter usually designated m . Vehanen *et al.*⁶ demonstrated that good results are obtained for $m = 2$, meaning that a Gaussian derivative profile is applied; see also the recent review article by Schultz and Lynn.⁷ In this study we used the Gaussian derivative implantation profile as well. The dependence of the mean penetration depth $\bar{z}$ on the incident positron energy E is assumed to be a power law

$$\bar{z} = A \times E^n, \quad (1)$$

A is a constant with dimension $\mu\text{g}/\text{cm}^2$ and n is a dimensionless constant independent of energy. Uncertainties are whether A and n are really material independent and constant with energy. Recent tables released by the International Commission on Radiation Units and Measures⁸ point out that in the keV energy range the power n is slightly larger for positrons than for electrons, meaning that one has to be reluctant in applying electron ranges to positron depth profiling. Few measurements on positron range exist. Mills and Wilson⁹ measured the transmitted positron flux through thin wedge-shaped foils of Al and Cu supported by a 10-nm-thick carbon foil and mounted close to an Al foil used as an annihilation target. Parameters they found were $A = 3.32^{+0.84}_{-0.70} \mu\text{g}/\text{cm}^2$ and $n = 1.60^{+0.15}_{-0.08}$ for Al and $n = 1.43^{+0.07}_{-0.11}$ for Cu. Measurements by Vehanen *et al.*⁶ on a multilayer structure of $\text{Al}_2\text{O}_3/\text{ZnS}/\text{Al}_2\text{O}_3$ on glass yielded an energy-dependent Doppler broadening which best reproduced for $A = 4.0 \pm 0.3 \mu\text{g}/\text{cm}^2$ and $n = 1.62 \pm 0.05$. The same relation yielded good fits for the thickness of SiO_2 layers on Si (110); see Ref. 10. However, in previous experiments we found that thicknesses of PdTa films as determined by Rutherford backscattering spectrometry (RBS) were poorly reproduced by slow positron depth profiling, utilizing the implantation profiles and parameters mentioned above.³ To investigate this further, the currently described experiments were performed.

Al was chosen as the substrate material. Being a good conductor, potentially disturbing influences from electric fields, as can be present for Si substrates, are avoided. Positron diffusion in the substrate should be minimal, so that the initial implantation profile is not broadened by diffusion of thermal positrons. Generally, the diffusion length is inversely proportional to the defect density and, therefore, the substrates should have a high defect density. To achieve this, the substrates were made by cold deformation of a piece of 99.999% pure Al; the thickness was reduced from 20 to 0.8 mm in this process. Suitably sized targets with a diameter of 14 mm were cut from the sheet. Both sides were first flattened by a lathe, after which the film side was machined by a precision optical cutter. The Al substrates were degreased and mounted in an ultrahigh vacuum deposition chamber. Pure Pd and $\text{Pd}_{0.5}\text{Ta}_{0.5}$ were chosen as overlayer material. Vapor-deposited Pd films have been shown to have a high defect density,³ so that the positron diffusion length is short. The PdTa films were amorphous, as confirmed by x-ray diffraction of films grown under similar conditions.³ Vehanen *et al.*¹¹ observed that the average positron diffusion length in amorphous binary metal alloys is of the order of 1 to 2 nm, which is too short to broaden the initial implantation profile. Furthermore, the lack of long-range order will rule out possible channeling effects. The substrate was kept at room temperature during deposition. Since the melting temperature of both Pd and Ta is rather high, hardly any mobility leading to pronounced surface roughness is expected. Indeed, no density or thickness fluctuations were observed by transmission electron microscopy on 30-nm-thick unsupported PdTa films. Various specimens were prepared with different overlayer thicknesses. The growth process was monitored by quartz crystal oscillators; however, the thicknesses quoted in this letter were all determined from RBS spectra. The spectra were taken with the specimens tilted 7° relative to the incoming 2 MeV He^+ ion beam to avoid any possible channeling effects in the substrates; the scattering angle was 170° . The spectra were analyzed using the simulation code described in Ref. 12. No high mass impurities were detected in the metal overlayer. The random height of the Pd and PdTa signals did not indicate low mass impurities (O,N,C) throughout the layer. However, at the overlayer/substrate interface an oxygen signal could be detected probably corre-

sponding to ~ 3 nm of Al oxide. There were no indications of any roughness; the back slope of the peak emerging from He backscattered by the overlayer was perfectly fit assuming the usual detector resolution (15 keV).

For the slow positron beam experiments, the Doppler broadening of the 511 keV annihilation gamma peak was used to derive a single line shape parameter S .⁷ By measuring the value of S for various incident positron energies, the energy at which the transition of $S_{\text{overlayer}}$ to $S_{\text{substrate}}$ takes place is determined. In Fig. 1 S vs E spectra are given for two amorphous overlayers. In an attempt to fit the measured spectra using the Gaussian derivative implantation profile and the parameters $A = 4 \mu\text{g}/\text{cm}^2$ and $n = 1.6$, the assumed thicknesses were 20% less than those derived by RBS. Therefore, A and n were varied systematically with the overlayer thickness given the value as determined by RBS. In Fig. 2 the set of A and n values is given for all measured specimens. The error bars are smaller for the amorphous PdTa overlayers than for the polycrystalline Pd overlayers, since in the latter case there is still some profile broadening due to positron diffusion; the positron diffusion length was found to be of the order of 6 nm. Indicated in the figure are also the literature values mentioned above. The best fits (least-squares optimized) were obtained for $1.4 < n < 1.6$, whereby A could take values in between 4.5 and $8 \mu\text{g}/\text{cm}^2$.

Since in this experiment layers of heavy elements were deposited on a light element, differences in positron backscattering between substrate and overlayer should play a role. Linderoth *et al.*¹³ found that positron backscattering is 6% more for Mo than for Ni suggesting an effect of the atomic number. We estimated backscattered fractions from Pd and Al by positively biasing a grid in front of the specimen and monitoring the increase in count rate. It was found that the fraction of 2 keV positrons backscattered from Al was $\sim 8\%$, whereas from Pd it was $\sim 16\%$. An apparent reduction of thickness of the order of 8% may be expected for a Pd overlayer on Al, with the interface at a depth where the positrons still have an average energy of 2 keV. To examine the effect of backscattering, a specimen was made with a sandwich structure; Pd was deposited on an Al substrate, and then an Al layer was deposited on top of the Pd layer. The Al layer should appear thicker and the Pd layer thinner in the positron annihilation experiment relative to the RBS

FIG. 1. Line shape parameters S as a function of incident energy for amorphous $\text{Pd}_{0.5}\text{Ta}_{0.5}$ overlayers with thicknesses of 92.5 and 182 nm on deformed Al and calculated curve.

FIG. 2. Set of A and n parameters that reproduced the real average thickness for overlayers on Al: (a) 45 nm Pd, (b) 90.5 nm Pd, (c) 179.5 nm Pd, (d) 92.5 nm $\text{Pd}_{0.5}\text{Ta}_{0.5}$, and (e) 182 nm $\text{Pd}_{0.5}\text{Ta}_{0.5}$. Also indicated are A and n values found by Mills and Wilson for (f) Al, (g) Cu, and (h) by Vehanen *et al.* for a multilayer structure.

measurements. In Fig. 3 the S vs E curve is given. The Al film has a higher S value than the Al substrate, which points to a very high concentration of open volume defects as is expected for vapor-deposited films. The positron diffusion length in the Al overlayer is very short as can be seen from the steep rise of S at very low energies. Fitting yielded a diffusion length of $1\text{--}2$ nm. In calculations using the power law, a good fit was obtained for $n = 1.6$ and $A = 4.5 \mu\text{g}/\text{cm}^2$; a special effect from backscattering was not observed.

All A values found in this study are rather high; for electron ranges usually values of around 4 are found. By assuming an energy-dependent power $n(E)$, the value of A could be reduced and the least-squares fit was improved as well. It was found that the energy dependence of n as proposed by Katz and Penfold¹⁴ for the range of high-energy electrons in Al

$$n(E) = 1.923 - 0.095 \ln(E), \quad (2)$$

with E expressed in keV yielded good fits. The drawn lines in Fig. 1 are best fits using this expression for n and adapting A . Values of A are all observed specimens both for $n = 1.6$ and for the energy-dependent n are given in Table I. The value of A seems to be higher for the thinner specimens, suggesting

FIG. 3. Line shape parameter S as a function of incident energy for a multilayer structure of 174 nm Al on 175 nm Pd on a polycrystalline Al substrate. The drawn line was calculated using the thicknesses as derived by Rutherford backscattering spectrometry.

TABLE I. A parameters yielding the best least-squares optimized fit for $n = 1.6$ and for $n(E)$ of Eq. (2).

Overlayer	Thickness (nm)	A for $n = 1.6$ ($\mu\text{g}/\text{cm}^2$)	A for $n(E)$ ($\mu\text{g}/\text{cm}^2$)
Pd _{0.5} Ta _{0.5}	92.5 $\pm$ 2%	5.7 $\pm$ 0.1	4.3 $\pm$ 0.1
Pd _{0.5} Ta _{0.5}	182	5.0	3.9
Pd	45	5.7 $\pm$ 0.3	4.4 $\pm$ 0.3
Pd	90.5	4.9	3.7
Pd	179.5	4.6	3.6
Al/Pd	174/175	4.5	3.5

that the energy dependence of $n(E)$ is incorrect, especially at the lower energies. No attempt was made to improve $n(E)$ for the limited number of specimens used in this study, although preliminary results from new studies indicate a materials dependence of A , and an energy dependence on E somewhat weaker than given by Eq. (2).

In conclusion, the energy-independent parameters as determined for the system Al₂O₃ on ZnS (Ref. 6) do not describe very well the implantation of positrons in metal overlayers like Pd and PdTa on Al or Si. A values in between 4.5 and 5.7 $\mu\text{g}/\text{cm}^2$ have to be assumed when the power n is fixed at 1.6. Backscatter effects alone are too small to explain this discrepancy. Therefore, for high Z metal overlayers an energy-dependent n is proposed. We were not able to match all spectra with a single prefactor A . Deriving depth information for high Z overlayers on Al or Si from positron beam experiments should be carried out with caution; the depth scale may be out by more than 20% unless a calibration experiment is performed. Further experiments are necessary

on a large number of different specimens to improve the calculated profiles, as well as model calculations on positron stopping utilizing the Monte Carlo approach to relate the experimental results to theory.

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