MONTE CARLO STUDIES OF POSITRON IMPLANTATION IN ELEMENTAL
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ABSTRACT

We have used a Monte Carlo computer code developed at Brookhaven^{1,2} to study the implantation profiles of 1-10 keV positrons incident on a wide range of semi-infinite metals and multilayer systems. Our Monte Carlo program accounts for elastic scattering as well as inelastic scattering from core and valence electrons, and includes the excitation of plasmons. The implantation profiles of positrons in many metals as well as Pd/Al, and Al/Co/Si multilayers are presented. Scaling relations and closed-form expressions representing the implantation profiles are also discussed.

INTRODUCTION

The motivation for the development of Monte Carlo methods to study positron slowing down in solids has been discussed before¹⁻⁴ (and references therein), and will not be discussed here, except to stress that the implantation profiles generated by Monte Carlo programs are needed in the analysis of data obtained by the positron beam study of solids and surfaces, in particular for defect profiling studies. Here, we will discuss, briefly, some results obtained by using a Monte Carlo program developed at BNL.^{1,2}

A brief description of this Monte Carlo program is given in Section II and the results for simple metallic systems, the variation of mean depth with energy as well as the implantation profiles are discussed in Section III. A scaling relation which reduces profiles of different energies to the same functional form is proposed. Section IV contains the Monte Carlo results for multilayer systems composed of layers of metallic elements. Trends followed by the backscattered fraction, mean depth, and the implantation profiles are discussed and a scaling procedure to reduce the complicated implantation profiles for multilayers to a simple functional form is described. Section V contains a brief conclusion and suggestions for extensions of the present work which will lead to a better understanding of the positron implantation process.

DESCRIPTION OF MONTE CARLO PROGRAM

The Monte Carlo code developed at Brookhaven (BNLMC) is very similar to that code of Valkealahti and Nieminen^{3,4} and is described in detail elsewhere.^{1,2} The positron or electron implantation is modeled by following the trajectories of a large number of particles of a given starting energy E_0 . The particle trajectory is changed by 'collisions' with the atoms of the host material. The collisions are either elastic, where the particle loses no energy, or inelastic, where there is a certain energy loss ΔE . During the collision the particle is also deflected through a scattering angle $\Omega(\theta, \phi)$. For each collision, the type of collision, the energy loss, the scattering angle, and the distance the particle travels between collisions are determined from a set of random numbers. The trajectory of the particle is followed until it either gets

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backscattered out of the material or until its energy is reduced below a 'cutoff energy' when it is considered to be implanted in the material.

The physics of the positron-atom or electron-atom interactions is incorporated via the cross-sections which describe the scattering processes. The incident particles undergo elastic Coulomb scattering from a single atom potential which has been modified to account for the effects of the lattice on the electronic structure.⁵ Such a potential, however, only partially accounts for crystalline order, and structural effects such as channeling are not included. The differential scattering cross-section is calculated using the method of partial waves⁶ for scattering from a central potential, for which the phase shifts are obtained by solving the radial Schrodinger equation using Milne's method.⁷

Inelastic scattering cross-sections and energy-loss functions are determined separately for core and valence electrons. The total inelastic cross-section for relativistic scattering by K-shell ionization includes contributions from processes at both large and small impact parameters. For large impact parameters the cross-sections of Kolbensvedt⁸ are used while for small impact parameters, Möller cross-sections⁹ are used for electrons and Bhabha cross-sections¹⁰ for positrons. For core electrons other than the K-shell electrons Gryzinski excitation cross-sections¹¹ are used. For all core inelastic scattering the method of Rohrlich and Carlson¹² is used to determine the energy loss functions and the Gryzinski formula relates the energy loss to the scattering angle.¹¹

Inelastic scattering from valence electrons can proceed either by single particle processes or by the collective plasmon excitation of the electron gas. For the single particle processes, Penn mean free paths,^{13,14} random-phase approximation energy loss functions,¹⁵ and DeSalvo, Parisini and Rosa scattering angle formulae¹⁶ are used. For the collective plasmon processes Quinn mean free paths,¹⁷ Isaacson energy-loss tables¹⁸ and the DeSalvo, Parisini and Rosa scattering angle¹⁶ formulae are used.

Our Monte Carlo simulations are limited to electrons and positrons with initial energies of 1 to 10 keV. The particles are considered implanted when their energy reduces to 25 eV, which is well above the energy of thermalized positrons. Thus the BNLMC does not incorporate the final stages of thermalization. As the positron energy decreases from 25 eV to <1 eV, conduction electron scattering becomes less and less efficient and is replaced by phonon scattering as the major energy-loss mechanism. Phonon scattering is included in a low-energy Monte Carlo algorithm being developed by Ritley, Lynn, Ghosh and Welch.¹⁹

RESULTS FOR SEMI-INFINITE ELEMENTAL METALS

A. Mean Depth

Monte Carlo simulations were done for many semi-infinite metals: Al, Si (ignoring the effects of band gaps), Co, Ni, Cu, Pd, Ag, W, Pt, and Au. Initial positron energies were varied from 1 to 10 keV and the trajectories of 2,000 to 10,000 particles were studied. The mean implantation depth (as a function of incident energy) was calculated from the Monte Carlo data. The mean depths scale, albeit imperfectly, with the density of the host material, and it has been customary to assume that the density dependence could be factored out so that the mean depths would lie on a universal curve;

$$\langle z \rangle = \frac{A}{\rho} E^n \quad (1)$$

where E is the energy of the positrons (in keV), $\langle z \rangle$ the mean depth, ρ the density of the host material and A and n are constants. The values of these constants were found to be $n=1.6$ and $A=400 \mu\text{g}/\text{cm}^2 (\text{keV})^{1.6}$, when the mean depth is expressed in angstroms.²⁰

Fig. 1. Density scaled mean depths as a function of the implantation energy for Al $\diamond$, Pd $\square$, Cu $\times$, Si $+$, Pt $\times$, W $\ast$, Au $\ast$, Ag $\ast$, Ni $\circ$. The straight line represents the universal curve.

The density-scaled results of the mean depth from our simulations are plotted in Fig. 1. Our Monte Carlo results show that the fitted values of A and n are material dependent and can be correlated to the material density as is shown in Figs. 2(a) and 2(b), i.e. a simple inverse density dependence still leaves some residual density effects.

Fig. 2. (a) Variation of A with density z .
(b) Variation of n with density z .

B. Implantation Profiles

The Monte Carlo studies of Valkealahti and Nieminen^{3,4} showed that the positron implantation profiles in a semi-infinite material were best represented by the derivative of a Gaussian. The derivative of a Gaussian profile is a special case of a more general class of profiles, the so-called Makhovian distribution 2b, where as a function of z , the probability $P(z)$ that a given positron is implanted between z and $z+dz$ is:

$$P(z) = N_0 m \frac{z^{m-1}}{z_0^m} \exp \left[- \left(\frac{z}{z_0} \right)^m \right] \quad (2)$$

with $m = 2$ and $z_0 = \langle z \rangle / \Gamma(1+1/m)$. N_0 is a normalization constant. For $m = 2$ the gamma function = 0.8862. Generally z_0 and m are used as adjustable parameters to fit the experimental data.

Fig. 3. Implantation profiles of positrons in Al for implantation energies of 1 keV (—), 2 keV (....), 3 keV (---) and 4 keV (-.-).

The Monte Carlo profiles of positrons implanted in Al are plotted in Fig. 3 with 2000 positrons implanted at each energy. Since the mean depth $\langle z \rangle$ increases with implantation energy E_0 , the profiles rapidly broaden with increasing E_0 . However, this effect can be accounted for by a simple scaling procedure. The prescription for scaling profiles of different energies can be obtained by rewriting Eq. (2) in the form

$$\langle z \rangle P(z) = \frac{N_0}{c_m} \left(\frac{z}{c_m \langle z \rangle} \right)^{m-1} \exp \left[- \left(\frac{z}{c_m \langle z \rangle} \right)^m \right] \quad (3)$$

where, $z_0 = c_m \langle z \rangle$, with c_m a numerical constant which depends on m .

Figure 4 shows a plot of $\langle z \rangle P(z)$ as a function of $z/\langle z \rangle$ for 10,000 positrons in Ni. This scaling has the advantage that the implantation profiles at different energies can be plotted on the same curve and one set of fitting parameters can describe all the profiles. This form of scaling also applied to more general forms of the profile than the Makhovian, as discussed below.

Fig. 4. Scaled implantation profiles for 1-10 keV positrons in nickel with 10,000 positrons implanted at each energy.

This universal scaling relationship, (i.e. $\langle z \rangle P(z)$ is the same function of $z/\langle z \rangle$, independent of the implantation energy,) was found to apply to all the materials that we studied (Al, Si, Co, Ni, Cu, Pd, Ag, W, Pt, and Au) for profiles with implantation energies of 1-10 keV.

Although our Monte Carlo profiles can be fit fairly well with a Makhovian using m and z_0 as adjustable parameters, we found, however, that even the best fit Makhovian underestimates the positron density at the surface. Monte Carlo results of Jensen and Walker²¹ also showed similar results and led them to propose a multiplying factor which would adequately model the profile near the surface. We find that a good fit to our Monte Carlo data can be obtained with a function of the form

$$\langle z \rangle P(z) = N_{\ell m} \left(\frac{u}{c_{\ell m}} \right)^{\ell} \exp \left[- \left(\frac{u}{c_{\ell m}} \right)^m \right] \quad (4)$$

where $u = z/\langle z \rangle$, $z_0 = c_{\ell m} \langle z \rangle$ and $P(u) = \langle z \rangle P(z)$. $N_{\ell m}$ is the normalization constant and ℓ , and m are fitting parameters. $c_{\ell m}$ is a numerical constant obtained from the condition that $\langle z \rangle$ obtained from the distribution function equals that obtained from the Monte Carlo "data." (When $\ell=m-1$ this reduces to the Makhovian form.)

From the form of the fitting function Eq. (4) it is obvious that near the surface (i.e. for $u < 1$) the shape of the profile is determined by the value of ℓ , while the shape of the tail ($u > 1$) is dependent on the value of m . By not restricting $\ell=m-1$ we can model the shape of the profile with greater accuracy. The results of the fitting using Eq. (4) are listed in Table I. The fitting parameter m showed a systematic decrease

from a value of 2.7 for Al to 1.78 for Pt. Values of ℓ ranged from 0.4 to 0.5 but there was no systematic z dependence and the values of c_0 varied from 1.30 to 1.55.

Element	Density (g/cc)	m	ℓ	$c_{m\ell}$	$N_{m\ell}$
Al	2.698	2.70	0.47	1.53	1.64
Si	2.329	2.52	0.54	1.46	1.71
Cu	8.96	2.55	0.40	1.56	1.57
Ni	8.96	2.53	0.43	1.53	1.60
Co	8.96	2.34	0.49	1.46	1.65
Ag	1.50	2.03	0.43	1.41	1.57
Pd	11.99	2.11	0.40	1.45	1.54
W	19.25	2.31	0.31	1.55	1.46
Pt	21.45	1.78	0.43	1.30	1.53

Table I. The values of m , ℓ , $c_{m\ell}$, and $N_{m\ell}$ for different elements.

MONTE CARLO STUDY OF MULTILAYERS

The two multilayer systems studied in detail were palladium-aluminum bilayers or overlayers of Al and Co on a Si substrate. For palladium-aluminum Monte Carlo studies were done on the following bilayers:

- i) semi-infinite aluminum
- ii) 100 Å layer of Al on Pd
- iii) 300 Å layers of Al on Pd
- iv) 500 Å layers of Al on Pd
- v) semi-infinite Pd
- vi) 100 Å Pd on Al
- vii) 300 Å Pd on Al
- viii) 500 Å Pd on Al

The variation of mean depth with energy for these bilayers is plotted in Fig. 5. The dashed and solid lines have been drawn to guide the eye and are not fits to any data. The top dashed line represents the mean depth for semi-infinite Al calculated from the Monte Carlo data; the bottom solid line represent semi-infinite Pd. The Monte Carlo results show that for low energies the multilayer system has the mean depth of the overlayer and that at higher energies the mean depth is close to that of the bulk material. The backscattered fraction of the Pd-Al system also showed a similar transition from overlayer to bulk properties with an increase in energy.

A 3-metal system consisting of 1000 Å of Al on 968 Å of Co on a silicon substrate was also modeled using the Monte Carlo code. X-ray reflectivity and variable-energy positron studies on this and similar systems have been done by Heald and Nielsen.²² The values of the mean depth and backscattered fraction of the multilayer system were similar to those of Al at 1-3 keV, those of Co at 4-6 keV, and those of Si at 7-10 keV. The implantation profiles are plotted in Fig. 6 for incident energies of 1, 3, 5, 7 and 9 keV. At 1 keV the positrons are implanted primarily in the aluminum overlayer. At 3 and 5 keV some positrons are planted in the Al and some in Co, the fraction in Co going up at 5 keV. At 7 and 9 keV there is positron implantation in the Si, too. In fact, the profiles show the same trend as the back-scattered fraction and the mean depth, showing that the results are quite consistent.

Fig. 5. Variation of mean depth with energy for the different palladium-aluminum bilayer systems.

Fig. 6. Implantation profiles of positrons in Al/Co/Si. Solid lines represent 1, 3, 7 keV and dashed lines represent 5 and 9 keV profiles.

The differences in the density of the substrate and the overlayers have previously been modeled by a density-normalization procedure.²³ Density-normalized profiles of the Al/Co/Si system are plotted in Fig. 7. When density normalization was tried

Fig. 7. Density normalized implantation profiles in Al/Co/Si. Solid lines represent 1, 3, 7 keV and dashed lines represent 5 and 9 keV profiles.

for copper overlayers on tungsten the procedure seemed to work fairly well and it should, in general, work fairly well when the densities of overlayer and substrate are not too different. However, in the case of palladium-aluminum, (Pd density = 11.95 g/cc., Al density = 2.698 g/cc) we found that the density normalization did not adequately describe the implantation profiles. We note that in the case of Al/Co/Si also density normalization leaves some residual multilayer effects.

Since the mean-depth scaling had worked so well for semi-infinite elements we tried a similar scaling for multilayers as well. Let us consider a system of two overlayers, one of material 1 and thickness T_1 , and another of material 2 and thickness T_2 on a semi-infinite substrate of material 3. Now define two new variables u_{eff} and $[\langle z \rangle P(z)]_{\text{eff}}$ such that

$$\begin{aligned}
 &\text{for } z \leq T_1 \\
 &\quad u_{\text{eff}} = z / \langle z_1 \rangle \\
 &\quad [\langle z \rangle P(z)]_{\text{eff}} = \langle z_1 \rangle P(z) \\
 &\text{for } T_1 < z \leq T_2 \\
 &\quad u_{\text{eff}} = (z - T_1) / \langle z_2 \rangle + T_1 / \langle z_1 \rangle \\
 &\quad [\langle z \rangle P(z)]_{\text{eff}} = \langle z_2 \rangle P(z) \\
 &\text{and for } z > T_2 \\
 &\quad u_{\text{eff}} = (z - T_2) / \langle z_3 \rangle + T_2 - T_1 / \langle z_2 \rangle + T_1 / \langle z_1 \rangle \\
 &\quad [\langle z \rangle P(z)]_{\text{eff}} = \langle z_3 \rangle P(z)
 \end{aligned} \tag{5}$$

Here $\langle z_1 \rangle$, $\langle z_2 \rangle$ and $\langle z_3 \rangle$ are the mean depths for positron implantation in semi-infinite materials 1, 2 and 3. For a particular energy E these values are calculated using Eq. (1) with values of A and n obtained by fitting the Monte Carlo mean depths of semi-infinite elements. The results of this normalization procedure for Al/Co/Si are plotted in Fig. 8. Not only does this normalization scheme adequately take

Fig. 8. Profiles for Al/Co/Si renormalized according to Eq. (5). The Monte Carlo data at 1 keV is represented by $\diamond$, 3 keV by $\times$, 5 keV by $\square$, 7 keV by $+$, and 9 keV by $*$. The solid line represents the fitted curve.

into account the effect of the different densities of different materials but the profiles for different energies which look so different from each other all fall on the same universal curve. The normalized data from 1 to 10 keV can all be fitted with an expression analogous to Eq. (4) where the parameters ℓ , m , $c_{\ell m}$, $N_{\ell m}$ are all obtained for the normalized data:

$$[\langle z \rangle P(z)]_{\text{eff}} = N_{\ell m} \left(\frac{u_{\text{eff}}}{c_{\ell m}} \right)^{\ell} \exp \left[- \left(\frac{u_{\text{eff}}}{c_{\ell m}} \right)^m \right] \quad (6)$$

where u_{eff} is given by Eq. (5). The fitted profile for this multilayer system is compared in Fig. 9 to the fitted curves for elemental Al, Co and Si [i.e. Eq. (5) with $T=c$]. The parameters ℓ , m , etc. for the various profiles are compared in Table II.

Element or Multilayer	m	ℓ	$c_{\ell m}$	$N_{\ell m}$
Al/Co/Si	2.59	0.43	1.51	1.66
Al	2.70	0.47	1.53	1.64
Co	2.34	0.49	1.46	1.65
Si	2.52	0.54	1.46	1.71

Table II. Comparison of the fitting parameters of the Al/Co/Si multilayer with those of semi-infinite Al, Co and Si.

This scaling procedure was also tested for the various palladium-aluminum multilayer systems for incident energies of 1 to 10 keV and was found to work just as well.

CONCLUSION

From the Monte Carlo study of simple metallics we find that the energy variation of the mean depth can be expressed by the functional form of Eq. (1). However, A and n are not universal constants but density-dependent parameters whose density dependence needs to be explored further.

We also find that the Makhovian distribution is not the best representation of the implantation profile and that it underestimates the positron density near the surface as found by Jensen et al.²¹ An alternate form, Eq. (4), similar to a Makhovian but somewhat more general, provides a better fit to our Monte Carlo results.

Fig. 9. Comparison of the renormalized Al/Co/Si profile (solid line) [Eq. (5)] with those of Al (---), Si (....), and Co (-.-.-) [Eq. (5) with $T_1 = \infty$].

The implantation profiles for different energies can be scaled in such a way that they all fall on a universal curve. This scaling procedure worked for positrons implanted at energies ranging from 1 to 10 keV in semi-infinite Al, Si, Co, Ni, Cu, Pd, Ag, W, and Au. It remains to be seen if these scaling relationships will still be valid for positrons implanted at higher energies and whether similar scaling can be done for electron implantation profiles.

The Monte Carlo studies of multilayer systems showed that the backscattered fraction and mean depth have values characteristic of the overlayer material at low energies and those of the substrate at higher energies. The implantation profiles also show the same trend; however, their detailed shapes are both complicated and energy-dependent. By suitably scaling these profiles, Eq. (5), we can get a profile independent of the implantation energy. This profile can be fitted with a Makhovian-like function [Eq. (6)] and it is very similar to the scaled profiles of the constituent

metals. Whether this scaling process is applicable at higher energies and for different metal combinations remains to be seen, but for Pd/Al and Al/Co/Si it works well.

These scaled energy-independent profiles can be used as the initial condition in defect profiling programs like VEPFIT²³ for the analysis of s-parameter experimental data.

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