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Positron Implantation Profiles in Elemental and Multilayer Systems

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Abstract

The different Monte Carlo schemes for modeling positron and electron implantation in solids have been briefly reviewed. The main results (mean depth, backscattered fraction, profile shapes) of the different Monte Carlo schemes are compared with experimental data and with each other. Qualitatively, the results of the different Monte Carlo schemes are the same, the most significant difference is in the value of the mean implantation depth. The scaled profiles (for positron implantation in any material) generated by the different Monte Carlo schemes can be superimposed on a universal curve which can be appropriately parameterized. The shapes of the scaled profiles, and the mean depth parameters A and n are found to be material-dependent.

I. Introduction.

The calculation of positron implantation profiles is necessary for the analysis of positron annihilation (Doppler broadening) experiments [1]. This analysis is carried out in two separate parts, one modeling the thermalization or implantation of the positrons, and the other the diffusion and subsequent annihilation of the thermalized positrons [2,3,4]. When a slow positron (with an implantation energy between 1 and 40 keV) enters the host or target material it loses its kinetic energy by electron- or phonon-assisted energy-loss processes until it achieves thermal equilibrium with its environment. This process typically takes less than 10 picoseconds. The depth distribution of the thermalized positrons before diffusion is called the implantation or stopping profile.

The thermalized positrons then diffuse in the material until they annihilate with a lifetime of 100-500 picoseconds, depending on the annihilation site. The depth distribution of the time-averaged positron density after diffusion/annihilation can be obtained by solving the steady-state diffusion equation using the positron implantation profile as a source term. The extraction of defect profiles from the Doppler broadening data has been described in detail in Refs. 1, 3, 5 and 6, and programs have already been developed for the diffusion analysis. The most often used programs are VEPFIT [5], and POSTRAP [6].

Both POSTRAP and VEPFIT require as input the positron implantation profile $p(z,E)$, where $p(z,E)dz$ represents the probability of finding a positron (of initial energy E) at a depth between z and $z+dz$ from the surface of the material. These implantation profiles are usually generated by Monte Carlo simulations of the implantation process.

The Monte Carlo procedure and the Monte Carlo programs currently being used will be briefly reviewed in Section II. Monte Carlo results for the median implantation depth and the backscattered fraction will be compared with experimental data in Section III. The main Monte Carlo results [7] for the Jensen and Walker [8,9] and the BNL [2,10] schemes will be presented in Section IV. If the backscattered positrons are reimplanted into the sample the positron density near the surface increases. The effect of reimplanting positrons on the mean depth and the implantation profile will be discussed in Section V. Results on multilayer implantation profiles are discussed in Section VI. A brief summary is included in Section VII.

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II. The Monte Carlo Procedure and Programs.

The Monte Carlo programs used to model positron implantation were developed by Valkealahti and Nieminen [11], Jensen and Walker [8,9], and Lynn and McKeown [2, 10], (hereafter called the BNL Monte Carlo). All these Monte Carlo programs have the same structure. They follow the trajectories of a large number of positrons (typically 10^5 - 10^6) as they traverse the target material. For each positron the program repeats a basic series of steps:

1. It selects the incident energy of the particle.
2. Determines the type of collision and the distance traveled after each collision.
3. Determines the energy loss. If the collision is elastic, the energy loss is zero. If the collision is inelastic, the non zero energy loss can be as large as the energy of the positron.
4. Determines the scattering angle in the positron frame of reference.
5. Transform the scattering angle back into the sample frame of reference, and calculates the position of the positron.
6. Determines the energy of the positron after the collision.
7. Checks a predefined set of criteria to determine if the trajectory should be continued. These criteria are
 - a. if the positron position $z < 0$, the positron is considered to be backscattered
 - b. if $z > T$, where T is the thickness of the sample, then the positron is considered to be transmitted, and
 - c. if the positron energy E is less than a cutoff energy E_{cut} then the positron is considered to be implanted or stopped.

If none of these conditions is satisfied then the particle simulation is continued until a maximum number of collisions has taken place. Usually the cutoff energy is 100 eV for JW Monte Carlo and 25 eV for the BNL Monte Carlo. When all the trajectories have been generated, statistical information on the mean or median implantation depth, and the angle- or energy-resolved backscattered and transmitted fractions can be extracted. The choice of the particular set of scattering mechanisms is introduced via the scattering cross-sections or the mean free paths. This obviously results in somewhat different results for different Monte Carlo schemes.

The VN [11], BNL [10], and JW1 [8] schemes are quite similar. In all three schemes the elastic scattering cross-sections are calculated by the method of partial waves, and the results are in good agreement. For inelastic scattering cross-sections the electrons of the host atom are divided into two categories, core and valence. The core electron mean free paths are calculated using the Gryzinski model [12]. In the VN model the valence electrons are given a modified binding energy and then treated by the Gryzinski approach. BNL and JW1 codes use a dielectric energy-loss function for a free-electron gas when valence-electron scattering is considered. JW1 use Gryzinski energy loss functions for the core electron scattering, BNL uses the Rohrlich and Carlson energy-loss functions, which approximate to the Möller cross-sections for electrons and the Bhabha cross-sections for positrons. The Bhabha and Möller cross-sections, however, represent scattering of positron and electrons, respectively, from free electrons and not bound electrons, which is what we are attempting to model.

The use of two distinctly different energy-loss mechanisms for core and valence electrons causes the implantation profiles to be dependent on how this delineation is made. Though this division is straightforward for simple metals, it is somewhat arbitrary for transition metals, leading to rather large uncertainties in the values of the mean depths and the backscattered fractions. For the BNL Monte Carlo the experimental value of the plasmon energy is used to calculate the number of valence electrons [14]. The rest of the electrons are designated as cores.

However, this prescription does not seem to work well for Cu and Ni, in both cases it seems to underestimate the mean depth, and for Cu, it also underestimates the backscattered fraction.

Core and valence electrons are given equivalent treatment in the second Monte Carlo option provided by Jensen and Walker [9]. This method (denoted JW2) is based on Penn's dielectric energy-loss functions which use optical data to calculate the inelastic mean free paths [15]. The scattering angle is calculated from the energy-momentum conservation equations.

III. Comparisons with Experimental Data.

Median implantation depths in Al and Au are calculated using the JW2 and BNL Monte Carlo schemes and are compared with the data of Baker et al. [16] and the transmission experiments of Mills and Wilson [17] in Fig. 1. We note that for Al the JW2 scheme overestimates the median depths whereas the BNL scheme under-estimates it. For Au, however, the JW2 results agree fairly well with the data.

Backscattered fractions calculated using the two schemes are compared (in Fig. 2) with the experimental data of Coleman et al. [18] for Al, Cu, Ag, Au, and the results of Makinen et al. for Au [19]. The JW2 results agree well with the experimental data for Al, Cu, and Ag. For Au the two sets of experimental data show considerable difference below 15 keV, the difference is not as large at higher energies. The results of JW2 agree well with the higher backscattered fraction data and the BNL values lie in between the two data sets. The BNL backscattered fractions show reasonable agreement with the data except for the Cu.

The lack of more experimental data prevents us from deciding which Monte Carlo scheme agrees better with experiment. The JW2 scheme certainly appears easier to use as it does not require the division of electrons into core and valence. The only limitation appears to be that optical data are not available for all materials.

IV. Monte Carlo Results

Implantation profiles have been generated for many materials using the BNL and JW Monte Carlo codes, and the results have been presented in Ref. 7. The implantation energies range from 1-10 keV for the BNL scheme and from 1-25 keV for the JW scheme. Implantation profiles for 1 and 10 keV positrons in Al, Cu, Ag, and Au are shown in Fig. 3. It is clear that the mean implantation depths obtained from the JW scheme are larger than those predicted by the BNL scheme. Profiles for a particular energy get narrower as the atomic number of the host material increases. The mean implantation depths increase with energy. The energy dependence of the mean depth has been fitted with the Katz and Penfold expression [20]

$$\langle z(E) \rangle = \frac{A}{\rho} E^{\alpha + \beta \ln E} \quad (1)$$

where ρ is the material density, and A , α , and β are found to be material-dependent as well as model-dependent parameters. Values of A , α , β for the two schemes are given in Table 1. The BNL mean depths (except for energies <1 keV) can be fitted reasonably well with the simplified expression

$$\langle z(E) \rangle = \frac{A}{\rho} E^{\alpha} \quad (2)$$

The values of A and n are listed in Table 2 for both schemes even though the simplified expression is not a good fit to the JW results. Plots of A and n as a function of the atomic number are shown in Fig 4.

The backscattered fraction for most materials displays a weak energy dependence which can be fitted with the functional form

$$B(E) = b_1 - b_2 \exp(-b_3 E) \quad (3)$$

where b_1, b_2 , and b_3 are material and model dependent parameters. Values of b_1, b_2 and b_3 for the two schemes are listed in Table 3, and are plotted as a function of the atomic number in Fig 5.

The positron implantation profiles are fitted to the expression

$$p(z, E) = \frac{N_{\ell m}}{\langle z(E) \rangle} \left(\frac{z}{C_{\ell m} \langle z(E) \rangle} \right)^\ell \exp \left[- \left(\frac{z}{C_{\ell m} \langle z(E) \rangle} \right)^m \right] \quad (4)$$

where $\ell, m, C_{\ell m}$ are fitting parameters. The normalizing constant $N_{\ell m}$ is determined by requiring that

$$\int_0^\infty p(z, E) dz = 1 \quad (5)$$

New variables $u = z / \langle z(E) \rangle$, and $P(u) = \langle z(E) \rangle p(z, E)$ are defined so that the implantation profiles can be written as

$$P(u) = N_{\ell m} \left(\frac{u}{C_{\ell m}} \right)^\ell \exp \left[- \left(\frac{u}{C_{\ell m}} \right)^m \right] \quad (6)$$

These scaled profiles were found to be energy-independent for the BNL model over the limited energy range 1-10 keV [21]. The JW1 and JW2 Monte Carlo results have confirmed that they are energy-independent over a larger energy range, 1-25 keV [7]. Comparison of the scaled profiles from the two different models show that these profiles are model-independent as well. The scaled profiles for a number of materials are shown in Fig. 6. The Monte Carlo profiles are fitted to Eq. (6), and the fitting parameters are listed in Table 4. The fitted profiles are plotted in Fig. [7] as a function of the atomic number. The fitted profiles for Be, Al, Cu, Ag, and Au are plotted in Fig. [8]. As the atomic number increases there is a small systematic change in the shape of the profile. Profiles for elements close to each other in the periodic Table, like Co, Ni, and Cu, are found to collapse on same curve, as do the profiles of Ag and Au in spite of the large difference in atomic numbers. The profile fitting parameters reflect this effect, when the atomic number goes above a large value the profile parameters become material-independent.

V. Reimplantation of Backscattered Positrons.

When backscattered positrons are reimplanted into the material, the positron density near the surface increases and the mean implantation depth decreases. Mean implantation depths in Al and Au with and without reimplanted positrons are compared in Table 5. The energy dependence of the mean depth can still be represented by Eq. (2), and the parameters A and n are material- and model-dependent. The values of A and n are listed in Table 6, and it is obvious

that the change in the mean depth is largely reflected in the parameter A. The implantation profiles follow the scaling relationship that results in an energy-independent profile. Implantation profiles in Al with all positrons reimplanted are shown in Fig. 9. These profiles can be fitted with the functional form

$$P(u) = \left[A_{\ell m} \left(\frac{u}{C_{\ell m}} \right)^{\ell} + B_{\ell m} \right] \exp \left[- \left(\frac{u}{C_{\ell m}} \right)^m \right] \quad (7)$$

Fitting parameters for some materials are listed in Table 7.

VII. Implantation Profiles in Metallic Multilayers.

Implantation profiles in metallic multilayers are fairly complicated and energy-dependent. As an example see Fig. 10(a). These profiles can be scaled to an energy-dependent form (see Fig. 10(b)) using the scaling relationships discussed in Ref. 20. Although this scaling prescription works well at energies below 10 keV, some correction factors are needed at higher energies to account for the difference in the backscattered fraction on either side of the interface. A modified scaling formula has been discussed by G. C Aers [22,23]. Multilayer profiles have been extracted from one-material profiles and compared to multilayer Monte Carlo simulations for a large number of layered systems [23].

VII. Summary

Monte Carlo simulations have been done for a large number of materials using the BNL and the JW Monte Carlo schemes. Functional forms and fitting parameters for the energy dependence of the mean depths and the backscattered fractions have been listed, and plotted as a function of the atomic number. These parameters are found to be material and model dependent. The implantation profiles have been scaled to a parameterizable functional form which is both energy and model independent. It has been shown that multilayer profiles can also be scaled to an energy-independent form. Conversely, multilayer profiles can be extracted from one-material profiles, and they agree well with multilayer simulations

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Table 1. Fits of $(A/\rho)E^{\alpha+\beta\ln E}$ to mean important depth $\langle z(E) \rangle$ in Å (incident energy E in keV) from Monte Carlo calculations where ρ is the density in gm/cm³.
(BNL using Rohrlich and Carlson energy loss: fitted over energy range 1-10 keV)

Element	A	α	β
Be	295.8	1.840	-0.049×10^{-2}
Al	353.7	1.462	2.58×10^{-2}
Si	313.0	1.601	-1.212×10^{-2}
Co	398.1	0.400	-1.33×10^{-2}
Ni	384.7	1.363	-1.863×10^{-2}
Cu	360.1	1.358	3.306×10^{-2}
Mo	640.2	1.201	5.077×10^{-2}
Pd	679.7	1.267	4.403×10^{-2}
Ag	620.7	1.213	4.248×10^{-2}
W	724.5	1.251	2.691×10^{-2}
Pt	781.7	1.253	0.922×10^{-2}
Au	830.3	1.180	3.327×10^{-2}

(J.W Gryzinski/ROA: fitted over energy range 1-25 keV)

Element	A	α	β
Be	387.2	1.591	3.986×10^{-2}
C	333.9	1.575	4.01×10^{-2}
Mg	397.5	1.440	5.544×10^{-2}
Al	430.7	1.411	5.949×10^{-2}
Si	418.7	1.402	5.961×10^{-2}
K	453.7	1.378	5.703×10^{-2}
Ti	491.6	1.363	5.609×10^{-2}
Co	572.4	1.306	6.263×10^{-2}
Ni	514.8	1.291	6.503×10^{-2}
Cu	446.8	1.340	5.512×10^{-2}
Ge	566.6	1.311	5.722×10^{-2}
Mo	716.3	1.240	5.996×10^{-2}
Ag	631.8	1.318	4600×10^{-2}
Pt	1001	1.178	5.724×10^{-2}
Au	952.4	1.215	5.113×10^{-2}

(JW Penn: fitted over energy range 1-25 keV)

Element	A	α	β
Al	462.8	1.340	7.011×10^{-2}
Cu	658.4	1.201	7.214×10^{-2}
Ag	668.9	1.200	6.383×10^{-2}
Au	1148	1.103	6.469×10^{-2}

Table 2. Fits of $(A/\rho)E^n$ to mean implantation depth $\langle z(E) \rangle$ in Å (incident energy E in keV) from BNL Monte Carlo calculations where ρ is the density in gm/cm³.
(BNL using Rohrich and Carlson energy loss: fitted over energy range 1-10 keV)

Element	A	n	ρ
Be	349.95	1.656	1.85
Al	324.41	1.558	2.701
Si	326.36	1.555	2.33
Co	415.17	1.352	8.90
Ni	406.84	1.297	8.90
Cu	322.30	1.481	8.90
Mo	543.42	1.388	10.2
Pd	590.56	1.427	12.0
Ag	541.60	1.369	10.5
Ta	725.77	1.334	16.7
W	664.66	1.350	19.4
Pt	759.50	1.287	21.5
Au	750.34	1.299	19.3

(JW Gry/RPA: fitted over energy range 1-25 keV)

Element	A	n	ρ
Be	288.8	1.809	1.85
C	246.8	1.797	3.52
Mg	261.9	1.746	1.74
Al	283.8	1.727	2.701
Si	272.4	1.725	2.33
K	293.8	1.696	0.86
Ti	319.1	1.677	4.5
Co	326.8	1.654	8.90
Ni	309.7	1.657	8.90
Cu	334.7	1.651	8.90
Ge	363.2	1.633	5.35
Mo	462.2	1.567	10.2
Ag	481.8	1.547	10.5
W	656.6	1.492	19.4
Au	659.3	1.494	19.3

(JW Penn: fitted over energy range 1-25 keV)

Al	278.4	1.721
Cu	381.1	1.601
Ag	397.6	1.568
Au	694.8	1.467

Table 3. Fits of $b_1 - b_2/\exp(-b_3E)$ to positron backscattered fraction $B(E)$ (incident energy E in keV) from Monte Carlo calculations.

(BNL using Rohrlich and Carlson energy loss: fitted over energy range 1-10 keV)

Element	b_1	b_2	b_3
Be	0.3920	0.3443	0.0098
Al	0.1655	0.0552	0.1342
Si	0.1754	0.0738	0.1593
Co	0.1615	0.0550	0.4399
Ni	0.1419	0.0598	0.4113
Cu	0.1782	0.0712	0.2205
Mo	0.3073	0.1503	0.2326
Pd	0.2679	0.1265	0.3688
Ag	0.2967	0.1405	0.2365
Pt	0.3192	0.1774	0.2754
Au	0.3413	0.2001	0.2121

(JW Gryzinski/RPA: fitted over energy range 1-25 keV)

Element	b_1	b_2	b_3
Be	3.18×10^{-2}	-7.634×10^{-3}	0.1830
C	4.19×10^{-2}	-1.767×10^{-2}	2.437×10^{-3}
Mg	0.1360	6.461×10^{-2}	0.3601
Al	0.1535	7.168×10^{-2}	0.3619
Si	0.1648	7.518×10^{-2}	0.2359
K	0.2165	0.1180	0.2768
Ti	0.2354	0.1313	0.2304
Co	0.2546	0.1518	0.1878
Ni	0.2581	0.1525	0.1824
Cu	0.2745	0.1910	0.1939
Ge	0.2819	0.2056	0.2110
Mo	0.3378	0.2162	0.1796
Ag	0.3480	0.2557	0.1710
Pt	0.4074	0.3013	0.1607
Au	0.4077	0.3172	0.1746

(JW Penn: fitted over energy range 1-25 keV)

Element	b_1	b_2	b_3
Al	0.1277	3.20×10^{-2}	0.3074
Cu	0.2522	0.1256	0.2257
Ag	0.2928	0.1909	0.1605
Au	0.3891	0.2674	0.1776

Table 4. Parameters from best fits of scaled Monte Carlo data to Eq. (6).

(BNL using Rohrlich and Carlson energy loss: fitted over energy range 1-10- keV)

Element	m	l	C _{lm}	N _{lm}
Be	4.145	0.8237	1.511	1.349
Al	2.603	0.5330	1.485	1.152
Si	2.729	0.4671	1.548	1.064
Co	2.344	0.4874	1.457	1.133
Ni	2.521	0.4305	1.529	1.046
Cu	2.452	0.5311	1.463	1.161
Mo	2.020	0.4831	1.367	1.182
Pd	2.309	0.4921	1.450	1.142
Ag	2.040	0.4782	1.378	1.170
W	2.106	0.4395	1.421	1.113
Pt	2.058	0.4690	1.383	1.160
Au	1.994	0.4760	1.365	1.177

(JW Gryzinski/RPZ: fitted over energy range 1-25 keV)

Element	m	l	C _{lm}	N _{lm}
Be	4.634	1.0365	1.467	1.567
C	4.244	0.7619	1.542	1.288
Mg	3.201	0.5370	1.572	1.104
Al	2.915	0.5415	1.534	1.132
Si	2.915	0.5245	1.544	1.113
K	2.566	0.5049	1.500	1.124
Ti	2.497	0.5130	1.483	1.141
Co	2.531	0.5101	1.490	1.135
Ni	2.524	0.5206	1.483	1.147
Cu	2.603	0.5198	1.402	1.215
Ge	2.463	0.5076	1.476	1.141
Mo	2.145	0.5292	1.384	1.212
Ag	2.292	0.5013	1.438	1.158
Pt	2.087	0.502	1.370	1.214
Au	2.101	0.5204	1.372	1.213

(JW Penn: fitted over energy range 1-25 keV)

Element	m	l	C _{lm}	N _{lm}
Al	3.212	0.5601	1.559	1.130
Cu	2.412	0.5190	1.458	1.161
Ag	2.434	0.5121	1.466	1.151
Au	2.064	0.5373	1.352	1.239

Table 5. Comparison of mean implantation depths without and with reimplanted positrons.

(BNL Monte Carlo results).

Energy (keV)	Mean depth in Al (Å)		Mean depth in Au (Å)	
	without	with	without	with
1.	141.3	133.1	43.2	38.4
2.	369.2	346.9	99.2	86.4
3.	665.0	628.2	164.0	141.5
4.	1039.9	970.2	232.2	203.9
5.	1473.0	1388.8	315.7	272.1
6.	1957.5	1855.9	400.2	339.2
7.	2481.6	2342.9	481.2	411.8
8.	3067.6	2839.3	578.8	497.1
9.	3688.4	3486.2	674.6	575.8
10.	4347.7	4094.1	777.7	664.5

Table 6. Values of the mean depth fitting parameters A and n for BNL profiles without and with reimplanted positrons.

Element	A		n	
	without	with	without	with
Be	349.9	336.3	1.656	1.655
Al	324.4	304.5	1.558	1.558
Si	326.3	290.4	1.555	1.569
Pd	590.6	565.2	1.427	1.398
Au	750.3	658.2	1.299	1.287

Table 7. Values of the profile fitting parameters for scaled BNL profiles with all backscattered positrons reimplanted.

Element	m	l	C _{lm}	A _{lm}	B _{lm}
Be	3.327	1.608	1.405	1.358	0.254
Al	1.846	1.715	1.111	1.208	0.475
Si	2.081	1.634	1.305	0.836	0.460
Pd	1.639	1.8568	1.001	0.978	0.506
Au	1.356	1.734	0.847	0.855	0.599

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Figure Captions

1. Comparison of the median implantation depth data of Baker et al. ($\diamond$) and Mills and Wilson (x) with the Monte Carlo results of JW2(----) and the BNL (—) Monte Carlo scheme.
2. Comparison of the backscattered fraction data of Coleman et al. ($\diamond$) and Mäkinen (x) with the JW2 (---), JW1 (....) and BNL (—) schemes.
3. Implantation profiles of 1 and 10 keV positrons in Al, Cu, Ag, and Au using the BNL (—) and JW2 (---) schemes.
4. Plots of the mean depth fitting parameters A and n as a function of atomic number using the BNL ($\diamond$), JW1 ($\square$), JW2 ($\blacksquare$) schemes.
5. Plots of the backscattered fraction fitting parameters b_1 , b_2 , b_3 as a function of the atomic number using the BNL ($\diamond$), JW1 ($\square$), JW2 ($\blacksquare$) schemes.
6. Scaled implantation profiles for Al, Cu, Ag and Au for the JW2 ($\square$) and BNL ($\diamond$) Monte Carlos.
7. Fitting parameters ℓ , m, C_{tm} for the scaled profiles using the BNL ($\diamond$), JW1 ($\square$) and JW2 ($\blacksquare$) schemes.
8. Best fit profiles for Be (—), Al (---), Cu (....), Ag (-----), and Au (-----).
9. Scaled are the positron profile in Al with all backscattered positrons reimplanted into the sample. $\diamond$ Monte Carlo data, — the fitted results.
10. Multilayer Al/Co/Si profiles before (a) and after scaling (b).

Fig 1

Fig2

$p(z)$

Fig 3

fig 4

Fig 5

Fig 6

Fig 7

Fig 8

Fig 9

Fig 10a

Fig 106.

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