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BACKSCATTERING, TRAPPING AND DEPTH
PROFILES IN C AND S

BY

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A MODEL FOR HYDROGEN ISOTOPE BACKSCATTERING,
TRAPPING AND DEPTH PROFILES IN C AND a-Si

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ABSTRACT

A model of low energy hydrogen trapping and backscattering in carbon and a-silicon is described. Depth profiles are calculated and numerical results presented for various incident angular and energy distributions. The calculations yield a relation between depth profiles and the incident ion energy distribution. The use of this model for tokamak plasma diagnosis is discussed.

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1. Introduction

Amorphous Si(a-Si) and C are members of a class of materials that has high hydrogen solubility, up to 10's of atomic percent, and low room temperature diffusivity, less than 10^{-20} cm²/s. Backscattering, trapping and depth profiles of hydrogen ions bombarding these materials are important for such diverse applications as solar cell lifetimes [1], amorphous semi-conductor properties [2], plasma diagnosis [3,4,5], and tritium loading [4,6]. At present there are limited experimental data [7,8,9,], on these processes. A salient feature observed in the experiments however is that saturation occurs at high fluences and the saturation fluence depends only on the energy of the incident ion. The data available have been taken for normally incident, monoenergetic beams. In most cases depth profiles have not been measured.

A variety of approaches have been made to the problem of calculating particle backscattering coefficients. The first attempts used single scattering analytical models which are valid at high energies where the reflection coefficient is small. Recent single scattering theories [10] have been shown to be reliable for reduced energies [11] $\epsilon > 3$. At lower energies where multiple scattering is important, reflection coefficients have been calculated using transport theory [12,13]. These calculations start by obtaining range profiles and then integrate that part of the profile which exists outside the surface. A correction then has to be made for the fact that ions crossing back into the vacuum cannot be scattered again. This correction is difficult to estimate reliably as it requires a knowledge of

the angular and energy distribution of the backscattered atoms. A third approach is to use Monte Carlo calculations and this technique has been used extensively [14,15,16]. Inherently such an approach takes a great deal of computational time and it is difficult to obtain a feel for the individual effect of the various parameters of interest. Moreover it does not readily lend itself to the calculation of the effect of fluxes which have broad energy and angular distributions, nor to the effects of saturation of the target.

In the present work we describe a model of hydrogen back-scattering and trapping. Saturation in trapping is explicitly included. Energy loss mechanisms are taken into account by using range and straggling parameters. A solution for the depth profiles in a finite target is made which is accurate at the low energies considered. Results are presented for normal incidence and $\cos \theta$ incidence angular distributions, and for monoenergetic and Maxwellian energy distributions. The results for normally incident monoenergetic beams have been compared with recent experimental measurements presented in a companion paper. The agreement in general is very satisfactory. Though the model can be used for arbitrary distributions, emphasis is placed on Maxwellian energy and cosine angular distribution since they are close to those expected in tokamak plasmas.

2 Model

As energetic deuterons penetrate solid matter they are scattered by the lattice atoms and lose energy by interactions with the electrons and the lattice. Once sufficiently slowed down they may become bound in the lattice or may continue to move in a diffusive fashion. Experimental data on room temperature hydrogen implants in α -silicon and carbon show that for low fluences and keV energies the depth profile matches the calculated range and straggling [9,17]. This means that the hydrogen is tightly bound in these lattices. The trapping may occur at beam-induced damage, naturally occurring damage, impurities, or even at undamaged sites. Diffusivity [18] and out-gassing [7,19] experiments indicate that tight binding occurs in undamaged carbon. Thus, in the model, we ignore beam-induced damage; trapping is taken to occur where a particle has slowed down to thermal energy.

The projected range and straggling of deuterons has not been measured at all energies of interest. Hence we rely on values calculated using the Brice [20] code. For deuterons incident at oblique angles, the depth penetrated is assumed to be the range reduced by $\cos \theta$, where θ is the angle from the normal. (See Figure 1). For the case of light ions slowing down in solids it has been shown [21] that the range distribution is well approximated by a Gaussian for reduced energies ϵ less than 1. In the model the Gaussian distribution is assumed to be spherically symmetric. This is accurate to a few percent in the energy range of interest [20]. Thus, aside from boundary conditions, the depth profile of a line beam of deuterons is approximated by

$$(1) \quad \rho(r) = \frac{F \, dA}{(1.44\pi S^2)^{3/2}} \exp(-r^2/1.44S^2)$$

where F is the deuteron fluence,

dA is the area element,

S is the (energy dependent) straggling parameter defined as the half-width at half-height, and

r is the distance from the end of mean projected range.

For a plane parallel beam, equation (1) can be integrated to give

$$(2) \quad \rho(Z) = \frac{F}{1.28\sqrt{\pi}} \exp \left[- \left(\frac{Z - R \cos \theta}{1.28S} \right)^2 \right]$$

where R is the (energy dependent) projected mean range, and Z is the distance to the surface.

Since the deuterons passing through the solid undergo multiple scatterings, some may pass back through the front surface and be lost. To account for this boundary condition in a simple manner we make the assumption that the spreading of the beam as it proceeds through the solid occurs mainly at the end of the range. This allows the depth profile problem to be formulated as a diffusion problem, with the exact result that the net depth profile is given by the Gaussian centered at the mean projected range, $+R \cos \theta$, minus its mirror image, a Gaussian centered at $-R \cos \theta$ [22]. For unit fluence the depth profile is given by

$$(3) \quad p_o(Z, E, \theta) = \frac{1}{1.2S\sqrt{\pi}} \left\{ \exp \left[-\left(\frac{Z-R\cos\theta}{1.2S} \right)^2 \right] - \exp \left[-\left(\frac{Z+R\cos\theta}{1.2S} \right)^2 \right] \right\}$$

It is in this fashion that backscattering is included in the model.

It is experimentally observed that as fluences of mono-energetic deuteron beams exceed a certain value, saturation occurs and complete reemission from the surface results. In one of these experiments [9] simultaneous depth profile measurements showed that as the fluence increased the profile extended back towards the surface, and not further into the solid. These two facts suggest that there are saturation concentrations for D in C and Si at which all tight binding sites are filled. The excess implanted deuterons appear to be mobile at room temperature. Apparently there is a preference for motion towards the near surface. Thus in the model, after saturation levels are reached, the implanted deuterons are assumed to move back towards the surface, filling vacant tight binding sites. If there are no vacant sites the particles reach the surface and are assumed to thermally desorb.

This empirical model is consistent with the isotopic exchange observed in experiments where a carbon surface is bombarded by one isotope (hydrogen, say) after it has previously been implanted to saturation with deuterium [23]. We note that from diffusion data [18] the normal residence time of a hydrogen atom trapped in a site is essentially infinite at room temperature. Thus the incident beam must be responsible for the isotopic mixing. This may occur by energy transfer releasing the previously trapped implanted atoms from these traps or by atom-atom exchange in the

modified potential of the lattice. We only have to assume that the vacant sites are filled in proportion to the local concentration in the mobile state and that the mobile atoms left after this game of musical chairs move towards the surface where they are released. One prediction of this model is that if deuterium is implanted to below saturation, and the target is then bombarded with hydrogen there will be a delay in deuterium re-emission until the net fluence, deuterium plus hydrogen, exceeds saturation.

The depth profile obtained from the model will, of course, depend on the energy, $f(E)$, and angular, $g(\theta, \phi)$ distributions of the impacting particles. For arbitrary $f(E)$ and $g(\theta, \phi)$ the depth profile, for fluences below saturation, is given by

$$(4) \quad \rho_o(z) = \int_0^{2\pi} d\phi \int_0^{\pi/2} d\theta \int_0^{\infty} dE \sin \theta f(E) g(\theta, \phi) \rho_o(z, E, \theta)$$

and the areal concentration of D, N/cm^2 , is given by

$$(5) \quad N = \int_0^{\infty} dz \rho_o(z)$$

The one free parameter in the code is the saturation level of hydrogen in α -Si and C. This was determined by comparison with monoenergetic normal incidence D implants. Values of $3.5 \times 10^{22}/\text{cm}^3$ for C and $1.55 \times 10^{22}/\text{cm}^3$ for Si were found

A simple case for a normally incident beam with a Maxwellian energy distribution can be considered if we neglect straggling and take a single valued range given by the power law

$$(6) \quad R = AE^n ; \quad S=0$$

A further simplification is to consider the concentration distribution sufficiently far from the surface so that the effects of backscattering and saturation can be neglected. We then have

$$(7) \quad \rho(Z) \propto \frac{f(E)}{(kT)^{-3/2}} (Z/A)^{1/n} \exp \left\{ -\frac{(Z/A)^{1/n}}{kT} \right\}$$

For carbon it has been shown that $n=1.0$ over the energy range 0.05 - 10keV

$$(8) \quad \text{Hence } \rho(Z) \propto (kT)^{-3/2} (Z/A) \exp \left\{ -\frac{Z}{kT} \right\}$$

Thus plotting the concentration vs depth in the form $\ln (\rho/Z)$ vs Z gives the temperature of the incident ions directly. We will show that a similar relationship holds even when straggling and the angular distributions of the incident ions are included in the model. However the calculations also show that the temperature calculated in this manner is sensitive to the value of n and thus it is necessary to carry out the numerical solution with accurate values of the range to obtain reliable results.

3. Distributions of Impacting Particles

In this section we summarize the angular and energy distributions expected for exposure at the edge of tokamak plasmas. At the plasma boundary there are four hydrogen populations of concern: thermal and supra-thermal distributions of both neutrals and ions. By thermal we mean, having a Maxwellian energy distribution characteristic of the local source. In tokamak plasmas this ranges from $T \sim 10\text{eV}$ at the periphery to $T \sim 10\text{keV}$ in the core. Supra-thermal distributions are non-Maxwellian distributions with high average energy. They occur due to the plasma heating mechanisms.

Neutrals originate by either charge exchange processes within the plasma or reflection of ions from surfaces. The angular and energy dependent thermal charge exchange out-flux has been calculated by numerous authors [24]. It strongly depends on plasma density and temperature. However, we may note that the angular distribution is not far different from a $\cos \theta$ Maxwellian distribution [25]. The temperature ranges from $T \sim 200\text{eV}$ to $T \sim 5\text{keV}$. We stress that this is an outflux from the plasma.

Ions which are reflected from material surfaces represent an influx into the plasma. These should come off the surfaces with a near $\cos \theta$ distribution [14] and an energy distribution which is shifted to lower energy than the impacting distribution [14]. The sources of these particles are localized to the limiter region. Charge exchange particles leaving the plasma may also impact on surfaces and be reflected back into the plasma. This would be a nearly uniform source around the torus.

The incident distribution for ions is expected to have a Maxwellian energy distribution of 20 to 200eV at the plasma edge. Future experiments may have a 10 times higher value. The distribution of impact angles would be a cosine one, if there is equipartition of energy between parallel and perpendicular motion. If there is a gradient in the local potential, as might be caused by sheaths, the angular distribution can be altered. For supra-thermal ions, such as those formed by neutral beam injection, the angular deposition can be strongly peaked in the direction parallel to injection. Supra-thermal ions formed by ICRH would initially have high perpendicular energy. The energy distribution will have a time dependent behavior, starting as monoenergetic, degrading towards a plateau and ending as a Maxwellian [26]. Pitch angle scattering could make the velocity distribution isotropic before it becomes Maxwellian.

In the following section we consider both normal incidence and $\cos \theta$ incidence angular distributions and monoenergetic and Maxwellian energy distributions.

4. Results

Results of numerical integration of equations [4] and [5] are presented in this section. The impacting particles are deuterons and the target is either C or Si. Deuterium, rather than hydrogen, has been used because of applications to tokamak experiments, and because of lower background levels in solids. The range and straggling parameters used in these calculations are shown in Figures 2 and 3. [20]

The depth profile for a normally incident, monoenergetic deuterium implant ($E=500\text{eV}$) as a function of fluence is shown in Figure 4. The asymmetry due to the boundary conditions is evident even at low fluence. At a dose of $4 \times 10^{16}/\text{cm}^2$, saturation occurs at a depth of $80\text{\AA}$. As the dose is increased the saturated region moves rapidly toward the front surface. Because of straggling the saturated region expands to greater depths, but at a decreasing rate for high fluences. Depth profiles for normal incidence monoenergetic beams $100 < E < 1000\text{eV}$ are shown in figure 5. In all cases the fluence is 10^{15} D/cm^2 .

The depth profiles for normally incident Maxwellian distributions of 20 to 500eV temperature are shown in Figure 6. The tail of the depth profiles, due to the Maxwellian exponential, is nearly linear. Its $1/e$ fall-off, λ , is plotted as a function of temperature for deuterium ions incident on carbon and silicon, in Fig. 7. There is little difference in λ between normal and cosine incident distributions. Experiments are needed to check the calculated range because of the dependence of λ on it. At high fluence saturation occurs. Because of the exponential tail, for high fluence the deuterium content increases proportionally to $\log F$.

The loss of particles from the front surface due to multiple

scattering depends strongly on angle of incidence and incident energy. This is shown in Figure 8 for monoenergetic D beams, impacting on C at low fluence. The normal incidence results are systematically ~20% below those of the Oen and Robinson calculation [14]. The angular dependence at fixed energy is also in similar agreement for $E < 3\text{keV}$.

Because of the uncertainty in the theoretical estimates of the straggling parameter S and the fact that there is no direct experimental check on it we have investigated the effect of varying straggling in the calculation. A change of $\pm 50\%$ varies the reflection-coefficient by ~30% at 20eV and more than a factor of 2 at 500eV as shown in Fig. 9. However the good agreement with Monte Carlo calculations, [14] and better agreement with experimental data plotted versus reduced energy [27], gives confidence that the values used in the present calculation are reasonable.

Trapping vs. fluence predicted by the code for monoenergetic, normal incidence deuterium implants in carbon and silicon are shown in figures 10 and 11. They are in good agreement with experimental data presented in the companion paper [28]. Discrepancies could be due to experimental uncertainties, topographical effects, surface impurities, or errors in the range and straggling parameters. The agreement of the code with the silicon data is within $\pm 5\%$ for $E \geq 300\text{eV}$, but only $\pm 20\%$ for $E < 300\text{eV}$. This may be due to the crystalline form of the Si sample.

The retained deuterium as a function of incident fluence is shown for Maxwellian distributions in Fig. 12. Including a $\cos \theta$ impact angle distribution in the model lowers the retained D by ~3 times for $F \leq 10^{16}/\text{cm}^2$ and by ~1.3 for $F \geq 10^{18}/\text{cm}^2$ as shown

in figure 13. Trapping versus fluence for Maxwellian and $\cos \theta$ distributions of D impacting on Si is shown in figure 14.

Summary & Conclusions

A model for describing backscattering, trapping and depth profiles of hydrogen isotope ions incident on carbon and silicon has been described. The slowing down of the incident ions in the solid has been taken into account through the range and straggling predicted by another code [20]. Reflection is taken into account by equating the spreading of the ions to a diffusion problem and adopting the standard solution of subtracting the image Gaussian. This assumption is justified by the good agreement between the calculated reflection coefficients and the measured ones. On the basis of experimental observations it is assumed that saturation occurs in the solid at any point when the implanted concentration exceeds a certain critical level. The absolute value of this level is the only free parameter in the model and is determined by fitting the model predictions to experiment. When saturation occurs further incident ions are assumed to move to the next nearest traps in the direction towards the surface.

The model does not attempt to give a physical explanation of the mode of transport of ions to the surface, merely to account for the fact that it occurs. The model does not make any assumption about whether it is the incident or previously trapped atom which is released when saturation occurs. However experiment indicates that the probability of release depends only on the proportion of a particular isotope present.

Results of numerical calculations using the model agree well with experimental data for normally incident monoenergetic ions over a wide range of energies and fluences. However at energies above 10keV the choice of a Gaussian range distribution is a poor approximation and the agreement with experiment becomes less

satisfactory, as expected.

Calculated depth profiles for Maxwellian velocity distributions and $\cos \theta$ angular distributions are given. This is the first explicit calculation relating depth distributions to incident ion energy distributions. The results make it clear that much information can be derived from a careful analysis of the depth distribution of hydrogen ions implanted in surfaces. The use of these techniques should be valuable in the diagnosis of particle fluxes to surfaces in plasma confinement systems. Some preliminary results have already been reported [29]. The energy distribution of the incident ions can be deduced from the depth distribution. In particular, impacting Maxwellian distributions will result in exponentially decreasing depth distributions. Although there is an inherent smoothing due to the straggling, it should be possible to distinguish between several energy components provided the difference in their energies exceeds the straggling effect. At low fluences the incident fluence can be determined by integrating the depth distribution. At high fluences it can be obtained, at least to order of magnitude, from the depth of the saturated layer. The angular distribution cannot be obtained directly from the trapped hydrogen profiles but it can in principle be unfolded by use of apertures in front of the collector probe. The effect of increasing angle of incidence is important at low fluence where there is a difference of a factor of three between the backscattering of a normally incident beam and one with a $\cos \theta$ distribution. At high fluence the saturation level varies only by 30% between normal incidence and a $\cos \theta$ distribution. This is because ions with large angles of incidence contribute only to the already saturated surface

layer while only ions with near normal incidence have sufficient range to increase the concentration in the unsaturated tail of the distribution.

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FIGURE CAPTIONS

Fig. 1. Schematic of beam range and straggling in a solid. Also shown is the image Gaussian used in calculating boundary effects.

Fig. 2. Straggling, S , and mean projected range, R , for deuterons on carbon, taken from Brice code calculation [20].

Fig. 3. Straggling, S , and mean projected range, R , for deuterons on silicon, taken from Brice code calculation [20].

Fig. 4. Calculated depth profile versus fluence, F , for normally incident 500eV D on C. Saturation occurs at a concentration of $3.5 \times 10^{22}/\text{cm}^3$.

Fig. 5. Depth profiles for normally incident monoenergetic beams of deuterons bombarding carbon. The fluence, F , is $10^{15}/\text{cm}^2$.

Fig. 6. Calculated depth profiles versus fluence, F , for a normally incident 20-500eV Maxwellian D beam on C.

Fig. 7. Calculated $1/e$ fall-off in depth profile, λ , versus temperature for Maxwellian distribution deuteron implants in carbon and silicon.

Fig. 8. Calculated values of reflected fraction versus angle for various energies, (deuterons on carbon).

Fig. 9. Variation in the normalized reflection coefficient for deuterons on carbon with changes in the Brice straggling parameter, for various incident energies (a) Normal incidence (b) 45° incidence.

Fig. 10. Comparison of calculated trapped deuterons in carbon versus fluence with data of Staudenmaier et.al [28].

Fig. 11. Comparison of calculated trapped deuterons in silicon versus fluence with data of Staudenmaier et. al. [28].

Fig. 12. Calculated deuterons trapped in carbon versus fluence for normally incident Maxwellian distribution.

Fig. 13. Calculated deuterons trapped in carbon versus fluence for a $\cos \theta$ distribution of impact angles and a Maxwellian velocity distribution.

Fig. 14. Calculated deuterons trapped in α -silicon versus fluence for a $\cos \theta$ distribution of impact angles and a Maxwellian velocity distribution.

Fig. 1. 793119

FIG. 2. 793199

Fig. 3. 793113

Fig. 4. 793245

Fig. 5. 793243

Fig. 6. 793241

Fig. 7. 793190

FIG. 8. 798118

Fig. 9. 793244

Fig. 10. 793187

Fig. 11. 793123

Fig. 12. 793198

Fig. 13. 793197

Fig. 14. 793116