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Molecular dynamics simulation of ion ranges at keV energies

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We present an efficient molecular dynamics method for calculating ion ranges and deposited energies in the recoil energy region 100 eV to 100 keV. The method overcomes some of the drawbacks of the binary collision approximation methods conventionally used to calculate ion ranges. We describe principles by which one can simulate implantation into polycrystalline materials, and study the effect of the crystal structure on the ion range. Application of the simulation method to practical cases is demonstrated by analyzing experimental range results of 50 keV silicon self-ion-implantation measured at our laboratory.

I. INTRODUCTION

Ion implantation is an important technique commonly used in several research and industrial applications [1–3]. The damage produced during implantation affects the final properties of the specimen strongly [4]. The first step in determining the distribution and nature of the damage produced consists of calculating the range and deposited energy distributions of the implanted ions.

To obtain detailed information of range and damage distributions in the low energy range (namely below about 100 keV) where elastic collisions dominate the slowing down process, computer simulation methods have been developed [5]. Binary collision approximation (BCA) methods provide a fairly efficient means for calculating ion ranges, and have been used much since the 1960's [5–7]. Molecular dynamics (MD) methods describe the interactions involved in ion implantation more realistically, but require much larger amounts of computer capacity than BCA methods [8]. As computers have become more powerful, interest has started to shift towards MD methods [9, 10]

In this study, we present an efficient MD method for calculating ion ranges at keV energies. We discuss the advantages offered by our method compared to conventional BCA simulations, and apply the method to study the damaged structure of 50 keV self-ion-implanted silicon.

II. MOLECULAR DYNAMICS ION RANGE CALCULATIONS

In molecular dynamics simulations the time evolution of a system of atoms is calculated by solving the equations of motion numerically. In the Newtonian formalism the force $\bar{F}_i$ acting on an atom i in the system is calculated as

$$\bar{F}_i(\bar{r}_i) = \sum_{j \neq i} \bar{F}_{ij}(r_{ij}) = -\sum_{j \neq i} \nabla V_{ij}(r_{ij}), \qquad (1)$$

where $\bar{F}_{ij}$ is the force acting between atoms i and j and $V_{ij}(r_{ij})$ is a potential energy function describing the interaction between atoms. The sum over j is taken over all atoms whose interaction with atom i is stronger than a threshold value $V_{\rm min}$ [11].

When the forces F_i have been calculated for all moving atoms in the system, the equations of motion for the system are solved using a suitable algorithm [12]. The solution yields the change in the atom positions, velocities and accelerations over a finite time step Δt. The smaller the time step is, the more accurate is the solution of the equations of motion.

The MD simulation principles outlined above offer a more realistic method to calculate the movement of atoms than the BCA simulation methods conventionally used to calculate ion ranges. In BCA calculations the movement of atoms is usually treated as a succession of individual collisions between the implanted ion and atoms in the sample [7]. The selection process of the next colliding atom always involves several unphysical parameters, i.e. simulation program parameters (like the maximum impact parameter and bulk binding energy [13]) that can not be directly determined from any physical quantity. Multiple collisions can be taken into account simultaneously in BCA simulations, but this typically complicates the calculations further [6, 14]. The choice of the unphysical parameters may affect the BCA simulation results by 10-20 % even for quite reasonable-seeming choices of the parameter values [6, 13].

In MD simulations multiple collisions are taken into account inherently in the simulations. Also, by making the unphysical parameters involved in the MD algorithms (Δt and $V_{\rm min}$) small enough one can ensure that they do not affect the simulation results.

Despite their wide use in other fields of physics and principal advantages over BCA methods, MD simulations have been very little used for calculating ion ranges at keV and higher energies. The main reason is that the MD methods used so far require very much larger amounts of computer capacity than BCA calculations. One reason to this is that the BCA approach of numerically solving the scattering integral requires less calculation steps than

solving the equations of motion.

Another reason is that when calculating ion ranges with BCA simulations, it is sufficient to calculate the motion of the recoil (incident) atom. In conventional MD simulation methods (see e.g. [8, 15–19]) the movement of all energetic ions have been calculated.

We have very recently developed MD algorithms which can be used to efficiently calculate ion ranges, while still maintaining the advantages offered by the MD approach over BCA methods [20,21]. The basis for our algorithms is formed by an ordinary microcanonical MD method using the Newtonian formalism and a Verlet neighbour list [11,22]. The electronic stopping is taken into account as a frictional force. However, our method differs from conventional MD simulations on a few counts. The basic aspects of our simulation method has been presented elsewhere [21,23]; in this paper we concentrate on the novel properties of our method.

In calculating ion ranges and deposited energies at high energies ($\gtrsim$ 1 keV), the interactions between the recoil ion and its nearest neighbours are much stronger than the interactions between lattice atoms. Hence the amount of interactions calculated can be significantly reduced by treating only the recoil ion interactions [24]. This approximation is hereafter called the recoil interaction approximation (RIA). We have shown that use of the RIA does not significantly affect final range results at energies higher than a few hundred eV [21].

During the actual range ("recoil event") calculation, when drastic changes occur in the maximum atom velocity in the system, a variable time step Δt is employed to speed up the simulations. Three criteria are used to determine Δt. Firstly, the time step is made inversely proportional to the recoil velocity v using a proportionality constant k_t. This approach is used e.g. in ref. [12]. Detailed analysis of very strong collisions in the keV energy range showed that the solution of the equations of motion does not describe strong collisions realistically if the time step is calculated from k_t alone. To obtain smaller time steps in strong collisions, the time step is also made inversely proportional to the product of the total force F the recoil atom experiences and its velocity v using a proportionality constant E_t. Since large increases of the time step make the solution of the equations of motion inexact [12], the value of the time step is never allowed to increase more than 10% from its previous value.

The final time step is determined from the criterium yielding the smallest value, that is

$$\Delta t_{\mathrm{new}} = \min\left(\frac{k_t}{v}, \frac{E_t}{Fv}, 1.1\Delta t_{\mathrm{old}}\right). \qquad (2)$$

With smaller values of k_t and E_t the error in the solution of the equations of motion gets smaller.

Because of efficiency considerations it is advantageous to keep the simulation cell as small as possible. On the other hand it is quite clear that a small cell containing a few hundred of atoms cannot contain the entire path of an implanted ion in the keV energy range. Therefore, a mechanism for ensuring that the recoil atom is always surrounded by lattice atoms is needed. We have developed a translation method for ensuring that the recoil atom moves in a structure unaffected by its previous motion. The method is similar to translation techniques used in BCA simulations [6], but has to our knowledge not been used earlier in MD simulations together with the RIA.

In our approach critical distances R_{S_i} are defined for the three space coordinates $i = 1, 2, 3$. If the recoil atom during the simulation gets nearer than R_{S_i} to the simulation cell border, for any coordinate i, then the recoil atom and all atoms surrounding it are moved this distance R_{S_i} away from the simulation cell border. The moved atoms retain their previous velocities and accelerations. The moving of atoms creates an empty region in the simulation cell "in front of" the recoil atom, which is filled with atoms whose initial positions correspond to the undisturbed sample structure used in the simulation.

In this way movement of the recoil atom can be simulated in an arbitrarily big sample without including more than a few hundred atoms in the simulations.

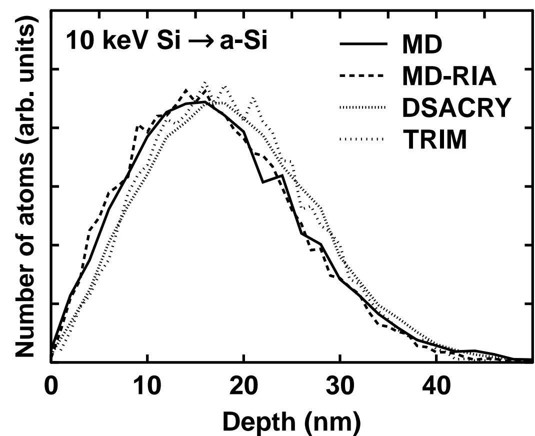

FIG. 1. MD and BCA range profiles of 10 keV Si implantation in a-Si. The legend MD denotes an MD calculation where all interactions are taken into account, MD-RIA an RIA calculation, DSACRY a calculation with a BCA code which takes account of the crystal structure and TRIM a very popular Monte Carlo BCA simulation code.

The size of R_{S_i} must obviously be less than half the size of the simulation cell. In simulating crystalline materials we usually select R_{S_i} to equal the size of the unit cell or some integer fraction of the unit cell size.

The ion range is determined in the simulations by following the movement of an implanted ion in a sample until it has slowed down to an energy of 5 eV, after which it no longer moves distances significant for the ion range. To obtain a representative statistics for the range distribution typically at least 1000 recoil events are simulated. The nuclear deposited energy is determined by tabulating the amount of energy transferred to primary knock-on atoms.

Our method has been implemented in the computer code MDRANGE [25] written in the C language for Unix environments. The method has been successfully compared to experimental results for several different ion-substrate combinations at ion energies between 10 keV [20] and 10 MeV [26].

In figure 1 range results are shown for 10 keV silicon self-ion-implantation. An amorphous sample structure and the ZBL interatomic potential and electronic stopping power was employed in all the simulations. The two MD curves are in very good agreement with each other, demonstrating the validity of the RIA approximation. The BCA calculations with the DSACRY [27] and TRIM [28] programs differ somewhat from the MD results.

III. APPLICATION TO 50 KEV SILICON SELF-ION-IMPLANTATION

Ion implantation of silicon is an important processing step in semiconductor technology. The need to understand the nature of the damage produced during ion implantation is emphasized by the desire to reduce device sizes of integrated circuits.

MD simulations of full collision cascades can be used to study the damage production mechanisms produced during implantation [15, 17, 18]. However, they are limited by the computer capacity to study only small portions of a sample. Ion range calculations encompass the whole implanted sample region, but can not be used to predict the damaged sample structure. However, it is possible to examine the damaged sample structure with range calculations by calculating ion range profiles in different sample structures, and comparing the results with experimental range profiles.

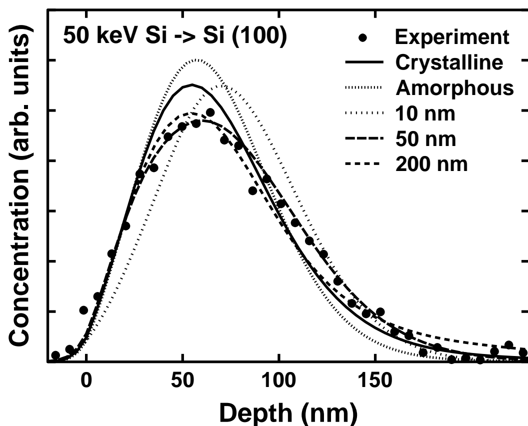

FIG. 2. Experimental and simulated range distributions for 50-keV Si self-ion-implantation. The simulated range results are for amorphous, crystalline and polycrystalline (grain sizes 10, 50 and 200 nm) silicon.

The MDRANGE program can be used to simulate implantation into many different types of samples, like crystalline, amorphous, polycrystalline and multi-layered sample structures [21]. Polycrystallinity is modeled by letting the recoil ion travel through a number of grains during the simulation of the slowing down. When the recoiling ion has traveled through one grain, it is injected into a new grain with the velocity it had when it left the former. The incident position of the ion entering in the new grain is selected randomly. The orientation of the new grain is either selected randomly or relative to the orientation of the previous grain.

Some simulated polycrystalline range profiles are shown in fig. 2 along with the profiles for amorphous and polycrystalline silicon. In fig. 3 the dependence of the range profile width and mean range as a function of the grain size is illustrated

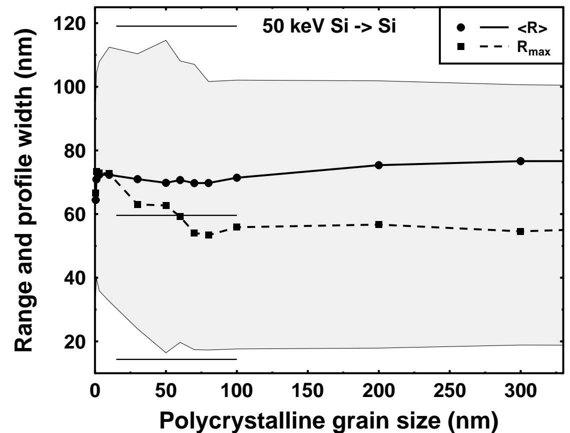

FIG. 3. Weighted means (< R >) and locations of the profile maximum (R_{max}) of simulated polycrystalline range profiles as a function of the grain size. The limits of the shaded area indicate the lower and upper half maximum R values of the range distributions. The thin horizontal lines show the location of the maximum and lower and upper half maximum of the experimental range distribution.

The small grain size (10 nm) range profile has an almost Gaussian form, similar to that of an amorphous profile. However, comparison with the amorphous profile shows that the front edge of the polycrystalline profile is significantly deeper in the sample than the amorphous profile. This can be attributed to the possibility of channeling in the first grain. For grain sizes larger than about 30 nm the range profile has a visible tail at large z values, which can be readily understood to be due to the possibility of long-range channeling in the grains deep in the sample. This tail causes $< R >$ to increase at large z values despite the fact that $R_{\max}$ remains constant (fig. 3).

The experimental range profile shown in figures 2 and 3 was determined with nuclear resonance broadening (NRB) measurements of high-dose ($> 2 \times 10^{16}$ ions/cm^2) 50 keV self-ion-implanted silicon [29]. The form of the profile did not change significantly in the dose

range measured. RBS measurements of the same samples showed that the sample structure was neither crystalline nor fully amorphous. From the figures it is evident that the experimental range profile is well reproduced by range calculations in a polycrystalline sample structure with a grain size of roughly 50 nm.

IV. CONCLUSIONS

We have presented an efficient method based on molecular dynamics simulations to calculate ion ranges in the keV energy range. The method combines cell translation techniques and the RIA within the framework of molecular dynamics simulations.

The usefulness of the method was demonstrated by studying the effect of the grain size in polycrystalline samples on the range profile, and by comparing simulated range profiles to experiment.

We conclude that our method offers the most realistic practical method for calculating the distribution of primary damage in keV ion implantations.

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[1] J. Asher, Nucl. Instr. Meth. Phys. Res. B 89 (1994) 315.
[2] D. M. Rück, D. Boos, and I. G. Brown, Nucl. Instr. Meth. Phys. Res. B 80/81 (1993) 233.
[3] B. Holländer, S. Mantl, W. Michelsen, S. Mesters, A. Hartmann, and L. Vescan, Nucl. Instr. Meth. Phys. Res. B 84 (1993) 218.
[4] J. Narayan, D. Fath, O. S. Oen, and O. W. Holland, J. Vac. Sci. Technol. A 2 (1984) 1303.
[5] J. F. Ziegler, J. P. Biersack, and U. Littmark, The Stopping and Range of Ions in Matter, Pergamon, New York, 1985.
[6] M. T. Robinson and I. M. Torrens, Phys. Rev. B 9 (1974) 5008.
[7] O. S. Oen, D. K. Holmes, and M. T. Robinson, J. Appl. Phys. 34 (1963) 302.
[8] M. Hou and Z. Pan, Nucl. Instr. Meth. Phys. Res. B 90 (1994) 469.
[9] D. J. Bacon and T. D. de la Rubia, J. Nucl. Mat. 216 (1994) 275.
[10] R. S. Averback, J. Nucl. Mat. 216 (1994) 49.
[11] D. W. Heermann, Computer Simulation Methods in Theoretical Physics, Springer, Berlin, 1986.
[12] R. Smith and D. E. Harrison, Jr., Computers in Physics Sep/Oct 1989 (1989) 68.
[13] J. Likonen and M. Hautala, J. Phys.: Condens. Matter 1 (1989) 4697.
[14] K. Gärtner, M. Nitschke, and W. Eckstein, Nucl. Instr. Meth. Phys. Res. B 83 (1993) 87.
[15] M. Sayed, J. H. Jefferson, A. B. Walker, and A. G. Gullis, to be published in Nucl. Instr. Meth. Phys. Res. B (1994).
[16] A. M. Mazzone, Nucl. Instr. Meth. Phys. Res. B 83 (1993).
[17] T. D. de la Rubia and M. W. Guinan, Phys. Rev. Lett. 66 (1991) 2766.
[18] K. Nordlund, J. Keinonen, and A. Kuronen, Physica Scripta T54 (1994) 34.
[19] H. Zhu and R. S. Averbach, Nucl. Instr. Meth. Phys. Res. B 83 (1993) 334.
[20] J. Keinonen, A. Kuronen, K. Nordlund, R. M. Nieminen, and A. P. Seitsonen, Nucl. Instr. Meth. Phys. Res. B 88 (1994) 382.
[21] K. Nordlund, Comp. Mat. Sci. 3 (1995) 448.
[22] L. Verlet, Phys. Rev. 159 (1967) 98.
[23] J. Keinonen, A. Kuronen, P. Tikkanen, H. G. Börner, J. Jolie, S. Ulbig, E. G. Kessler, R. M. Nieminen, M. J. Puska, and A. P. Seitsonen, Phys. Rev. Lett. 67 (1991) 3692.
[24] D. S. Karpuzov and V. E. Yurasova, Phys. Stat. Sol. 47 (1971) 41.
[25] A presentation of the program MDRANGE is available on the World Wide Web in http://beam.helsinki.fi/~knordlun/mdh/mdh_program.html.
[26] K. Nordlund, T. Ahlgren, and E. Rauhala, 6 - 10 MeV nitrogen range profiles in silicon, unpublished.
[27] M. Hautala and I. Koponen, Defect and Diffusion Forum 57-58 (1988) 61.
[28] J. P. Biersack and L. G. Haggmark, Nucl. Instr. Meth. 174 (1980) 257.
[29] K. Nordlund, J. Keinonen, E. Rauhala, and T. Ahlgren, Accepted for publication in Phys. Rev. B (1995).