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McChasy2: New Monte Carlo RBS/C simulation code designed for use with large crystalline structures

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ABSTRACT

A new tool for Monte Carlo simulation of ion channeling in crystals is described. The recently developed McChasy2 code follows the algorithm used previously in the McChasy program, but is well suited for simulations in large crystalline samples. The state of works on the validation of the code, and the possibilities of the new code for supplementing molecular dynamics with a tool supporting the experimental analysis of crystalline structures are presented and discussed.

1. Introduction

Monte Carlo (MC) simulations of the Rutherford Backscattering spectra recorded in channeling conditions (RBS/C) have demonstrated already their potential in the analysis of the channeling data. Ability to extract quantitative information about structural disorder in multi-elemental targets, thick damaged layers and structures containing dislocations are only some of the advantages of this approach. One of the first fully operational MC packages used for simulations of the RBS/C spectra was McChasy, the code developed in National Centre for Nuclear Research (NCBJ), Świerk, Poland. Today's research trends are much more focused on the massive use of computer simulations in material design (see, e.g., Ref [1]) leading to modelling of large atomic structures, such as heterostructures, complex defect structures or multiatomic solids. One of the main tools used for the prediction of these structures is simulation made by Molecular Dynamics (MD) method. It came out, thus, that a new generation of MC simulation codes is needed to treat channeling data to ensure an interchangeability of data between Molecular Dynamics and Rutherford Backscattering/Channeling. This idea is the main reason for the development of a simulation tool able to reproduce RBS/C spectra from structures containing hundreds of millions of atoms, i.e. the cells used in MD simulations.

The first generation of programs used for MC simulations of backscattering process used rather small simulation cells, slightly larger than the unit cell of the crystal. This was the case of, e.g. McChasy (Monte Carlo CHAnelling SYmulations) code developed in NCBJ [2]. The code has been based on numerical solutions developed by Barrett, who used Nuclear Encounter Probability (NEP) notion[3]. Several options were progressively introduced into a code, among them the most important was the possibility to simulate structures with dislocations. Ones of the matchless options of the code was the ability to quantitatively determine depth distributions of dislocations [4–6] or damage distributions in crystals irradiated with swift heavy ions [7–8]. The code has been successfully used by several groups, mainly NCBJ from Poland and CSNSM from France, what led to about one hundred of articles presenting data analysis based on its use. During the development of the new version of the code its original version was also massively modified [9,10]. This version was marked as "McChasy1" to distinguish it from the current version named "McChasy2". The McChasy1 version is still limited to the rather small number of atoms, generally slightly larger than number of atoms in a unit cell, and serves the purpose of routine analysis of the RBS/C spectra.

Current trends in material science clearly point to the importance and huge potential of the holistic approach (also known as integrated approach) in material design and analysis [11]. This method is based on the use of a combination of different methods, from atomistic simulations of material structure to prediction of functional properties and including various analytical techniques for model validation. Since

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Molecular Dynamic technique is one of the most commonly used methods of atomistic simulations, it became clear, that the further development of MC simulations of RBS/C data should be based on the capability to reproduce channeling spectra from MD-like structures.

The aim of this work is to communicate that a new tool for ion-channeling simulations has been built, to show the principles behind it, and to document that it adequately addresses nanometer-sized extended structural defects. We want to show that using modern PCs it is possible to depart from the simplified description of structures, as used in McChasy1.

Key features of McChasy2 code are outlined below. After a description of large crystalline sample generation, and the simulation algorithm, specific examples of using McChasy code were shown and discussed. The conclusions end the work.

2. Crystalline samples

To perform channeling simulation with McChasy2 a sufficiently large set of atoms should be prepared in a computer memory; further called a sample. Typically, the atoms that form a sample belong to a cuboid with the base from 10 nm × 10 nm up to 50 nm × 50 nm and with the height of 500 nm to 2000 nm. Such samples contain from millions to a few hundred millions of atoms. Atoms in the sample are presented by their spatial coordinates and a variable denoting the chemical element.

Samples for McChasy simulations can originate from two sources. First, McChasy is suited to use sets of atoms produced in MD simulations performed with LAMMPs (Large-scale Atomic/Molecular Massively Parallel Simulator) [12]. The second option allows one to construct a sample within the McChasy code on the basis of crystallographic structural data. These two possibilities are marked on McChasy2 general scheme (Fig. 1) with arrows leading through ADJUSTEMENT and CONSTRUCTION blocks, respectively. Both procedures produce samples that consist of sets of parallel atomic layers lying perpendicular to the required channeling axis.

The result of CONSTRUCTION procedure is a set of atomic planes (*hkl*), where [*hkl*] defines the required channeling direction. Sample' production based on crystallographic data (unit cell parameters, space group, crystallographic positions of atoms) is a complex but a purely

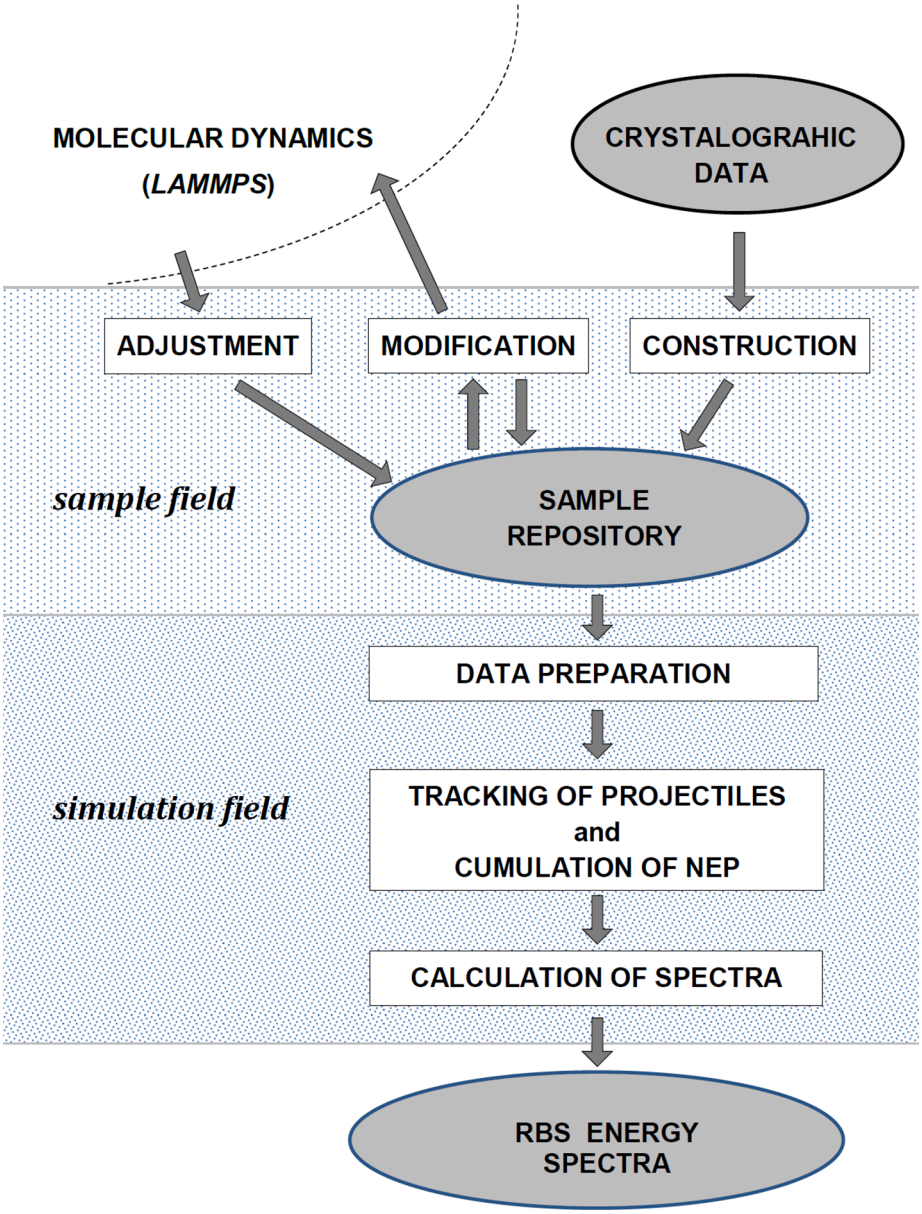

Fig. 1. General scheme of the McChasy2 code.

crystallographic task. It is worth to point out that wide range of *hkl* indices is available (e.g. [1 1 1], [0 1 2]).

Atomic sets resulting from LAMMPS should be adequately prepared in this code and adjusted for channeling simulations. In LAMMPs the MD-simulated models (samples) must be relaxed; in all studied cases we applied room temperature relaxation. Optimal number of steps of this relaxation should be chosen by numerical experiments. Typically the relaxation includes ca. 10 ps of simulation-time (10 000 steps with 10 fs per step). Next, instantaneous atomic positions should be averaged to obtain thermodynamic equilibrium positions of atoms. Satisfactory statistic for this averaging is usually obtained with ca. 250 frames (steps) taken every 10 fs. Further, in ADJUSTMENT procedure, the set of atoms is sliced into 10-picometer-thick wafers, and, finally, the sample is assembled from all wafers containing at least one atom. In practice MD-simulation supplemented by averaging and slicing procedures results in formation of samples with larger amount of atomic layers than the virgin crystal, but with a smaller mean number of atoms in the layers than in the original crystalline planes. This change is due to movements of atoms during MD-simulations.

The McChasy2 program is designed to be able to consider structural defects. Apart from small, localized defects, like random displaced atoms (RDA), or Frenkel pairs, the code is ready to include defects involving wide regions of the crystal, comprising dislocations, dislocation loops, stacking faults, gas bubbles, etc. The sample-modification procedure (MODIFICATION block in Fig. 1) is applied for this purpose. The slicing procedure used here is ready to cut any structure on thin 10-picometer wafers and construct samples usable by McChasy2. In parallel, the atomic data file (boxes) for LAMMPs can be also produced.

The procedures used to create or modify samples, as well the simulation procedure is suitable not only for cuboid samples, but also for the cylindrical ones.

3. Monte Carlo simulations

Mono-energetic He ions with energies belonging to 500–3500 keV range are randomly generated on the sample's surface. To ensure quick convergence of probabilistic estimates a low-discrepancy sequences of initial points is applied [13]. Initial velocities of the projectiles are directed along the crystalline axis. A single projectile, during its flight in the sample, collides with atoms, hence its trajectory is no longer a straight line. The binary collision approximation is applied to determine the interaction of the projectile with atoms. A threshold of 10^{-7} rad is used for a single collision; deflections of trajectories by angle smaller than this limit are neglected. Since the sample is treated as a set of separate layers, the performed projectile tracking results in approximation of the real (curved) trajectory by a broken line with vertices located at the layers.

The Barret's approach [3] of nuclear encounter probability is applied. This method relies on statistical sampling of actual locations of atoms. Thermal vibrations of atoms are taken into account and a Gaussian distribution of instantaneous atomic positions is assumed according to theoretical substantiation formulated by Willis and Prior [14]. Nuclear encounter probability calculated for subsequent collisions are cumulated in tables separately to each nuclide present in the target. This table, obtained from simulation with large number of trajectories is a base to calculate the RBS energy spectrum in channeling conditions (after trajectory tracking was done). Ziegler data on stopping power are used [15] in a procedure transforming cumulated NEP tables into channeling spectra.

The projectile tracking complemented with NEP calculations constitutes the core of ion-channeling procedure. This part of McChasy2 program is common for both types of samples: these constructed on the basis of crystallographic data and those produced basing on LAMMPS results. Such a solution enables the program' user to apply the same channeling simulation procedure to samples relaxed with molecular dynamics, and to the samples which are not relaxed, containing pure geometrically defined structures (with defects or ideal ones).

To illustrate the practice of using McChasy2 program two short calculation runs are depicted below. The CONSTRUCTION procedure was used to build a crystalline silicon brick-shaped sample. The sample was 30 nm wide and 500 nm thick, and contained 22,485,691 atoms constituting 3683 (0 0 1) atomic planes. During the simulation with ten thousand 2000-keV He ions the analysis of 224,773,952,066 ion-atoms collisions was accomplished. From among them 53,621,935 collisions influenced the projectile trajectories, while remaining 2,240,720,330,131 (i.e. more than 99.976%) did not. These collisions were barren due to large ion-atom distances. 604,319 collisions provided contributions to NEP. 9 projectiles did not reach the back of the sample; they were eliminated from simulations when their trajectories traversed the sample' walls. Similar simulation was launched with 20% of RDA; then number of lost projectiles increased to 28. Both samples were constructed in a few seconds, while simulations longed 16 min and 19 min. The runs were executed with 4-thread computing on a modern PC with 8 GB RAM.

4. Examples of simulations

The first, mandatory test of the new simulation procedure of RBS/C spectra is obviously checking that the results obtained by a new version of the code are identical as those produced by using the procedures that are already well established and tested. This is the case of the McChasy1 code which has been validated against Two Beam Approximation (TBA) method [16]. This comparison has been described in Ref [17] and proved, that the results obtained using McChasy and TBA method were identical.

Verification that the quantitative analysis of defect distribution made by McChasy2 code is correct has been done by comparing the results generated by McChasy1 and McChasy2 codes. A test case of $MgAl_2O_4$ magnesium aluminate spinel irradiated with 160 keV Ar^+ ions up to a fluence of $5x10^{15}$ cm^{-2} has been used for this purpose. The results are shown in Fig. 2 showing RBS/C spectra (Fig. 2a) and damage profiles extracted from these data by using McChasy1 and McChasy2 codes (Fig. 2b). In simulations the defects were assumed to be RDA only. One can note that both codes allowed for a good reproduction of the shape of the RBS/C spectra including random, virgin aligned and irradiated aligned ones. The damage profiles, shown in Fig. 2b, are also essentially identical since they differ due to the probabilistic character of the simulations. This confirms that McChasy2 code correctly simulates the distribution of RDA (or equivalent scattering centers). Taking into account that McChasy1 has been validated versus the TBA method the same defect distributions obtained for McChasy1 and McChasy2 confirms that also McChasy2 produces defect distributions identical to the TBA approach.

In the next step of testing, channeling simulations with samples containing edge dislocations was performed. A [0 0 1] nickel sample, created by CONSTRUCTION procedure, was modified by MODIFICATION procedure to include edge dislocations. We assumed that dislocation edges run along [0 1 0], and [1 0 0] directions, being perpendicular to the channeling axis. Each single edge dislocation was included by elimination of a half-plane followed by displacements of sample atoms. These displacements recreated the characteristic arctan-shaped planes around the dislocation edge following a Peierls-Nabarro dislocation model [18]. It was assumed that dislocations are randomly distributed in the crystal. Several samples were produced for various dislocation densities.

Fig. 3a shows the results of the simulation. As expected distortion of channels influenced the RBS signal. The greater the number of dislocations, the stronger the impact. Unexpectedly, we observed that channeling spectra simulated for samples with dislocations are not smooth, they show waving. This effect is stable, i.e. it does not vanish with increasing number of projectiles used in the simulations. The reason of the effect relies in a too low number of dislocations as objects tossed in

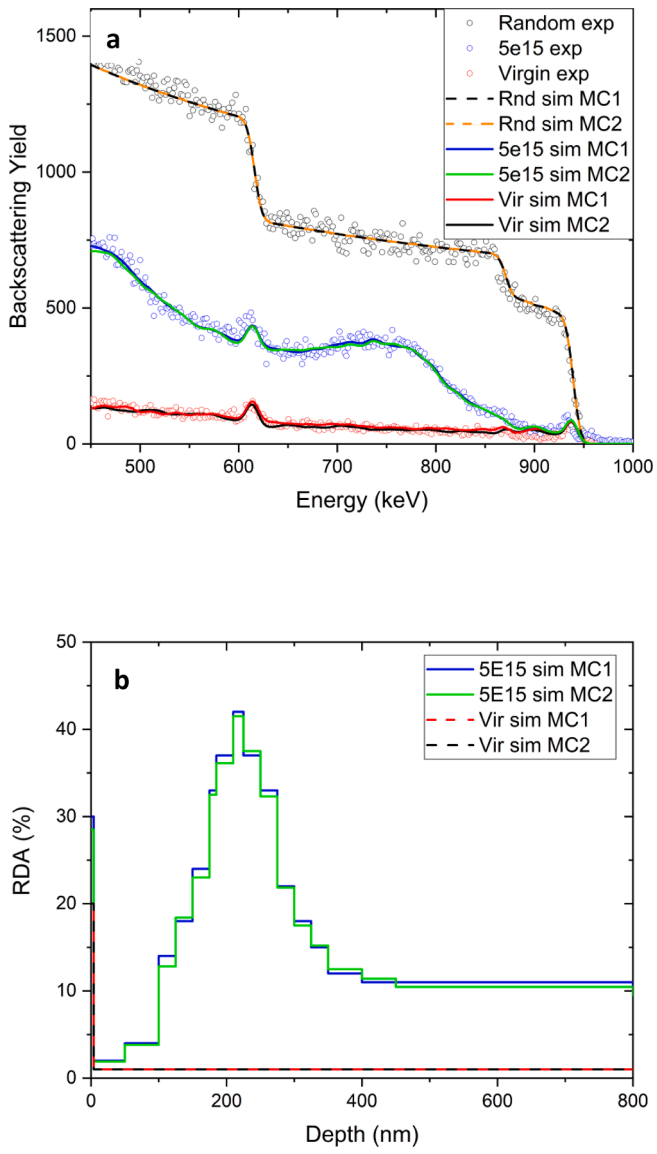

Fig. 2. RBS/C spectra recorded for irradiated MgAl2O4 sample irradiated with 160 keV Ar ions up to a fluence of $5x10^{15}$ cm^{-2} and fitted with McChasy1 and McChasy2 codes (a) as well as defect distributions extracted by using both MC procedures (b).

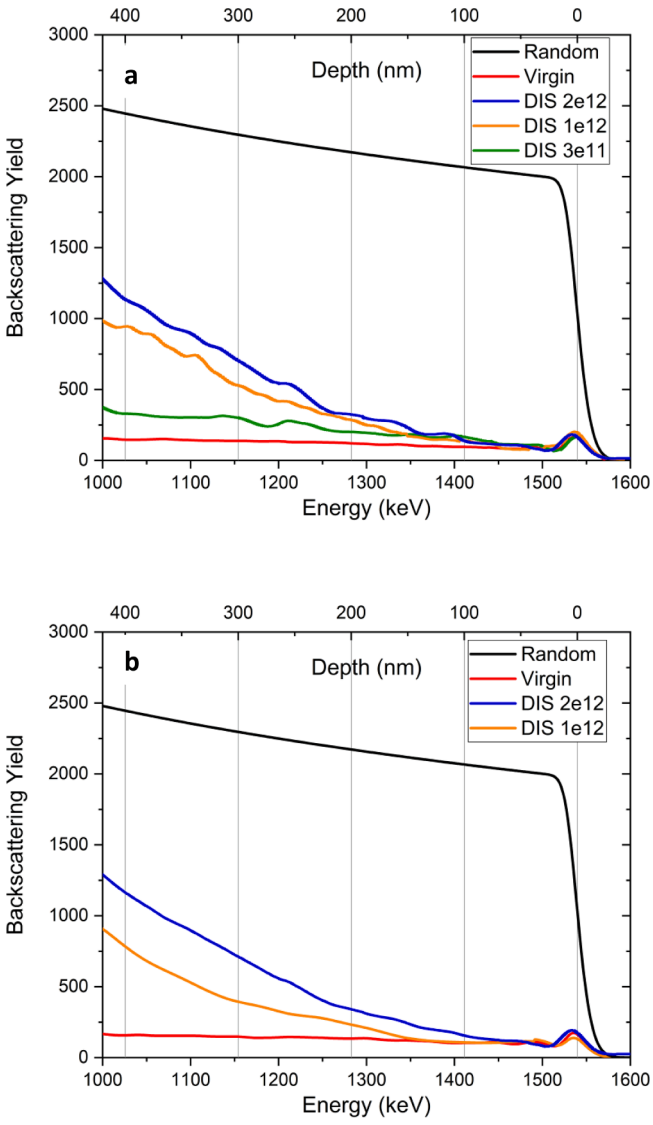

Fig. 3. MC simulations of RBS/C spectra for Ni single crystal containing edge dislocations, obtained with a single tossing of dislocations (a) and after improving the tossing procedure (b).

the sample. Really, to reproduce dislocation density in 10^{11} –10^{12} cm^{-2} range, we were obliged to toss from 5 to 50 dislocations in the sample. Tossing of such small number of objects produces non-uniformities: in some depth intervals number of dislocations was larger, in other ones smaller. Consequently, in some intervals the channels are stronger bent than in other ones. The non-uniformities yield hills and lows in the spectra that should be smooth.

Thus, tossing of large objects in the samples can lead to disturbances in the simulated spectra. To avoid them the manner of tossing was improved. We applied tossing of dislocation' depths in separate depth intervals (following the approach proposed by Sobol in ref. 13). Moreover, instead of using single sample for each concentration value, we created 10 samples, and averaged the spectra obtained for them. The applied solution could be replaced by using a wider sample to probe the crystal with ions at the larger surface. However, this method would be non-practical since it would require much larger computer memory, and would enhance fraction of barren collisions resulting in rise of computing time. As can be seen in Fig. 3b the applied improvements of dislocations' tossing eliminated the disturbances that had been observed.

The third step of McChasy2 testing was focused on the use of files generated by LAMMPS code. For this purpose a long cylindrical structure having 30 nm in diameter and 600 nm in length was created. Within this structure vacancy dislocation loops 6 nm in diameter were introduced every 30 nm and the structure was thermalized using LAMMPS code. After MD simulations the structure was transformed using ADJUSTMENT procedure to create a sample for Monte Carlo simulations. Fig. 4 contains the RBS/C spectra obtained as a result of Monte Carlo simulations on a structure used for MD simulations. The spectra produced have the shape expected for sample with periodic distribution of large structural defects and confirm the ability of the McChasy2 code to reproduce data obtained from large atomic structures used in MD calculations. We observed similar periodicity for heterostructures studied experimentally and simulated with McChasy1 [2].

5. Discussion

One of the main advantages of the McChasy1 code is the possibility to run simulations on PC computers. Ability to keep this feature for the McChasy2 code wasn't evident, as the new version uses six orders of

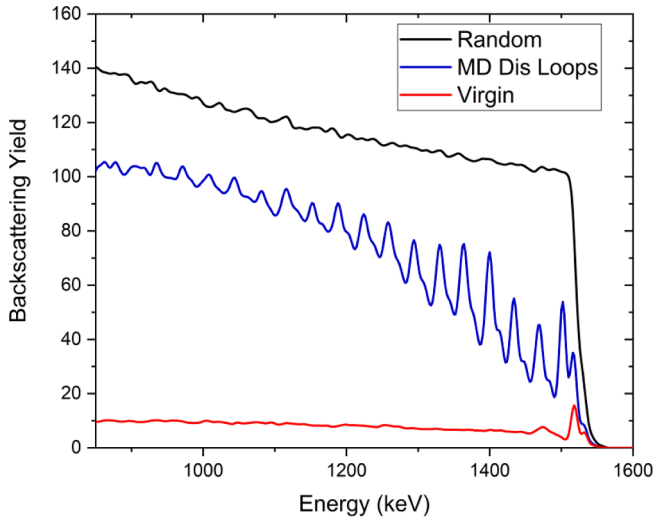

Fig. 4. MC simulations of RBS/C spectra from MD boxes produced by LAMMPS code. Structure assumed the presence of vacancy dislocation loops having 6 nm in diameter introduced into a structure every 30 nm.

magnitude larger structural files. However, the careful optimization of iterative procedures and the use of multithread calculations allowed us to run the code on PC computers and even to keep similar calculation times as it was the case of McChasy1. Typical calculation to reach a decent statistic takes usually about one hour using a modern PC-class computer.

The speed tests of McChasy2 package have shown that time of calculations decreased about 25 times for multitasking mode based on four threads when compared to McChasy1 code. There is a linear dependence between the number of threads involved for computation and computation speed. It was observed that factor defined as a ratio between the number of threads to time reduction is equal to 1.6 (where 1 means the perfect parallelism). The use of large RAM memory is obligatory. The use of multicore processors is, obviously, strongly recommended. Apart from channeling spectrum the random spectrum is calculated in every simulation run in a standard (non-Monte Carlo) manner. The random spectrum can be also generated by Monte Carlo procedure, by neglecting deflections of ions during their flights in the sample.

As observed the simulations performed with McChasy2, with hundreds (?) millions of atoms, made on a typical modern PC, can be quite fast. However, possibility of simulations with large samples is limited by the computer memory. For simulations with samples with more than 400 million of atoms the usage of 32 GB RAM is recommended.

The essence of the Monte Carlo method consists in the statistical sampling of the probability distribution. In the case of ion channeling simulations, the probability distribution of the positions of the nuclei of atoms is sampled. Sampling is done by particles whose trajectories are determined on the basis of deviations in collisions with atoms moved from their equilibrium positions. In this way, we simulate the ion flux distribution in the channels of the structure. The nuclear encounter probability is calculated with Gaussian formula centered at the equilibrium position of the nucleus. Another approach to the statistical sampling, in which deviations from equilibrium positions determined by molecular dynamics are taken into account, seems to be useful also, but – we suppose that it will require much longer simulations.

When considering possible modifications of the algorithm used we remember that in a real ion-channelling measurement the ions hit an area of ca. 1 mm^2, i.e. 10^{12} nm^2. In the case of computer simulation, due to the obvious limitations of computer memory and calculation time, we limit ourselves to sampling several hundreds square nanometers, so the studies encompass regions that are still many orders of magnitude smaller than in reality.

It should be noted that the formula for considering a small working volume adopted in McChasy1 did not allow for a detailed assessment of the effects of the structural simplifications used in McChasy1. We plan (more detailed) McChasy2 – McChasy1 comparisons to study these effects.

McChasy2 is a program developed to link two well-established research techniques: RBS/c technique and molecular dynamics. The first steps in this direction were provided by Zhang et.al. [19,20]; these works were focused on the research of defects in [1–20] nickel and quartz crystals. McChasy 2 provides the ability to generate many possible crystal structures with symmetries classified into 12 symmetry groups. We plan to use MD to create structural models of defects, to relax them, and to use McChasy2 to verify existence of defects formed. Modelling of functional properties can be the next step on this path of material studies.

6. Summary

A new Monte Carlo simulation code, McChasy2, has been developed and initially tested. The code allows to reproduce RBS/C spectra from large atomic structures containing hundreds millions of atoms. Such structure can be generated by Molecular Dynamics LAMMPS code or can be built by the McChasy2 program on the basis of crystallographic data and models of crystalline defects. McChasy2 has been tested in basic cases, such as structures by introducing models of defects: RDA, edge dislocations and dislocation loops relaxed with MD LAMMPS calculations. Further testing and development of the code are needed to extend the areas of code validation.

7. Author Agreement

We certify that all authors have seen and approved the final version of the manuscript being submitted. They warrant that the article is the authors' original work, hasn't received prior publication and isn't under consideration for publication elsewhere.

8. Funding Source Declaration

The work has been financed by own funds coming from research institutions declared as affiliation of the authors.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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