Shower approach in the simulation of ion scattering from solids

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An efficient approach for the simulation of ion scattering from solids is proposed. For every encountered atom, we take multiple samples of its thermal displacements among those which result in scattering with high probability to finally reach the detector. As a result, the detector is illuminated by intensive "showers," where each event of detection must be weighted according to the actual probability of the atom displacement. The computational cost of such simulation is orders of magnitude lower than in the direct approach, and a comprehensive analysis of multiple and plural scattering effects becomes possible. We use this method for two purposes. First, the accuracy of the approximate approaches, developed mainly for ion-beam structural analysis, is verified. Second, the possibility to reproduce a wide class of experimental conditions is used to analyze some basic features of ion-solid collisions: the role of double violent collisions in low-energy ion scattering; the origin of the "surface peak" in scattering from amorphous samples; the low-energy tail in the energy spectra of scattered medium-energy ions due to plural scattering; and the degradation of blocking patterns in two-dimensional angular distributions with increasing depth of scattering. As an example of simulation for ions of MeV energies, we verify the time reversibility for channeling and blocking of 1-MeV protons in a W crystal. The possibilities of analysis that our approach offers may be very useful for various applications, in particular, for structural analysis with atomic resolution.

I. INTRODUCTION

The classical binary collision approximation is well applicable for the description of ion-solid interactions in the energy range above ~ 1 keV (in each ion-atom collision, the interaction with nearby atoms is weak enough to allow it to be treated as a perturbation if required). This provides the possibility of an efficient reproduction of many experimental conditions playing with the parameters of the interaction model. Due to the importance of this subject, big efforts have been directed toward the development of efficient simulation programs [1]. To illustrate the central role that computer simulations have played in the field, it will suffice to mention that one of the most prominent phenomena, the channeling of ions in crystals, was discovered in simulation results [2]. In this context, one should refer also to the paper of Barrett [3], which provides an important supplement to the theory of ion-crystal interaction allowing us to address some aspects of the phenomena that are difficult to treat theoretically. Currently, with ion beams being widely used as a precision tool for the analysis and modification of materials, there has been an increase in the level of demands to simulation algorithms. One example is the use of low-energy ion beams for surface structure determinations. The classical picture of scattering together with the effects of blocking of scattering on atomic pairs form the basis for obtaining detailed information about the atom locations. The only way to extract this information is by comparison of intensities measured for different scattering geometries with the results of simulations for many trial structures.

However, a fundamental problem occurs here due to the insufficient efficiency of existing algorithms. Due to the small scattering cross sections, the direct simulation by calculation of individual ion trajectories (the program MARLOWE [4] is the most developed code of this type) is often not practicable. To understand this, one has to keep in mind that the experimental procedure typically consists of the measurement of angular scans with a small-aperture energy-resolving detector. To acquire the necessary statistics in a measured spectrum, the required number of ions in the beam amounts to $\sim 10^9$ for low-energy (keV range) scattering of heavy ions such as Ne or Ar and to $\sim 10^{13}$ for scattering of ions with energies in the MeV range. It is clear that the direct simulation of such a large number of ion trajectories is impossible, especially in the latter case. For this reason, it is concluded (see, for example, Ref. [1]) that even the power of supercomputers is by far not enough to perform such simulations.

An overview of the existing approaches to this problem shows that two main ideas are used, depending on the ion energy. In the simulation of backscattering of high-energy channeled ions, one can rely on the single-scattering model, assuming that the motion of the ion in the outgoing path can be described by a straight-line trajectory. This allows us to avoid a precise description of this segment of the ion trajectory. As a result, each ion contributes to the statistics according to the probability of close collisions along the ingoing path. The inverse (blocking) condition can be treated analogously (see Ref. [5], for example). This model is well tractable and serves as the basis for a large number of algorithms that

have been developed (see Refs. [6–8] for the most widely used).

This algorithm does not work, however, if one is interested in the simulation of channeling-blocking conditions or in the scattering of low-energy or heavy ions. To tackle this problem, Tromp *et al.* [9] proposed to use the property of ion motion known as the Lindhard time-reversibility rule [10]. According to this property, the scattering yield can be obtained by a proper convolution of the flux of impinging ions with the time-reversed flux of ions imagined as being scattered into the detector aperture when these two fluxes meet in the sample volume. In general, the use of this feature seems to not provide a possibility to facilitate the simulation; the convolution of fluxes in a six-dimensional phase space which, even at small depths, can have complicated distributions, is also a hardly solvable problem. The procedure (further referred to as the "reversing" procedure) is, however, heuristic in the sense that, in contrast with the direct simulation, it admits the use of certain approximations in the description of ion trajectories, and this can be used to design much more efficient schemes of simulation. In other words, one has here the possibility to dramatically boost the efficiency, although this is achieved at the cost of partly sacrificing the accuracy of the description of the phenomenon, when this is admissible. In particular, a simplification of the description is applied in the algorithm of the program VEGAS [9]: the energy and angular variables are not considered in the flux convolution. This simplified procedure can not reproduce all the details of multiple and plural scattering, as well as some specific features of energy losses. Fortunately, this turns out not to be a serious obstacle for the use of this program in the important application of structure analysis using medium-energy (≈ 100 keV) light ion scattering (MEIS).

On the other hand, the full version of the reversing approach is feasible for the simulation of scattering of low-energy heavy ions (LEIS). In this case, only a few layers at the surface of the sample contribute to the scattering yield (this feature provides sensitivity to the surface structure) and the structure of fluxes is not strongly developed. The latter circumstance permits us to use a coarse-grained representation of the fluxes and, as a result, their convolution becomes a tractable problem. Such a full version of the reversing procedure is implemented in the program MATCH [11], where the convolution of fluxes is performed using a specific method: for two sets of precalculated ingoing and outgoing trajectories, those pairs are matched that can be connected as a result of the scattering on one target atom.

All the indirect methods of simulation mentioned above have rather restricted regions of applicability. As a result, for many conditions where the multiple and plural scattering effects are significant, simulation results are now not available. The development of efficient methods of simulation, if possible, is important for basic studies of ion-solid interaction and for the improvement of methods of analysis of materials based on the use of ion beams.

In this paper, we describe a simulation method that uses the advanced possibility of the Monte Carlo method, i.e., the strategy of importance sampling. This strategy is used in sampling of thermal displacements of atoms met on the ion path. This approach provides a possibility to increase the efficiency of the direct method by several orders without the necessity to sacrifice the exact treatment of the binary collisions. Section II describes how the importance sampling strategy can be implemented in the simulation. We give the name TRIC (transport of ions in crystals) to the developed computer program. In all other respects, we employ the standard version of the binary collision model as used in the code MARLOWE. Therefore, for all features of this model, readers are referred to Ref. [4]. In Sec. III, some results of the application of the program TRIC are presented to demonstrate the efficiency of the developed approach for the simulation of different experimental conditions. In Sec. IV, we discuss the advantages of our approach over the simulation algorithms proposed hitherto. Some conclusions are formulated in Sec. V.

II. THE SIMULATION PROCEDURE

The ion velocity is assumed to be much higher than the thermal velocities of crystal atoms, a condition which is usually well satisfied. This means that the configuration of thermal displacements of all atoms can be predetermined before the simulation of scattering of an individual ion. The configurations must be chosen randomly according to the statistics of thermal vibrations. However, it is a more convenient procedure, and therefore it is commonly used, to choose the displacement of an atom met along the ion trajectory just before the treatment of the collision. Note that this procedure must take into account the correlation of thermal displacements of different atoms as it is present in the lattice dynamics. Note also that, for any atom met by the ion, not only a single but also multiple samples of its displacement can be taken, resulting in different trajectories after the collision. It can be argued that the simulation will correctly represent the statistics of atom thermal vibrations if the contribution of each ion in the Monte Carlo sum is determined as the average of the results obtained in such multiple trials [12]. This simple conclusion plays an important role in the following consideration.

The direct simulation of histories of collisions with the actual distribution of atom displacements is very inefficient because the number of successful scattering events in all trials is minute (as in a real beam irradiation experiment). The main idea that allows us to increase the efficiency is a separate treatment of those displacements that result in scattering events of the ion in the direction of the detector because this fraction of ion trajectories has a much higher probability to actually end up there. Moreover, by multiple sampling of such displacements for each atom, the scattering flux can be increased even more.

To implement this simple idea, we use the following procedure. First, the impact parameter b_i, which corresponds to scattering into the direction to the center of the detector (see the left-hand side of Fig. 1), is determined (i denotes the number of the current lattice site). Then, assuming that the interesting scattering directions lie within a cone of width $\Delta\Theta$, we can define the corresponding region of impact parameters, with half-widths in the scattering plane $\Delta b_\parallel$ and in the perpendicular direction $\Delta b_\perp$ that can be estimated as

$$\Delta b_\parallel \approx |d\Theta/db|^{-1}\Delta\Theta, \quad \Delta b_\perp \approx (b/\sin\Theta)\,\Delta\Theta \qquad (1)$$

(variation of $b_\perp$ means actually a rotation of the scattering plane by an angle $\Delta\varphi \approx \Delta b/b$ with the corresponding variation of the scattering angle $\Delta\Theta = \sin\Theta\Delta\varphi$). These conditions separate a region close to the ion trajectory, a tube aligned with its velocity vector, and the associated fraction of the (Gaussian) distribution of atom displacements. Then, one or several (n) atom positions can be drawn according to the distribution with such a cutoff. As a result, a "shower" of ion trajectories is directed toward the detector. The remaining part of the distribution is used to continue the trajectory of the "primary" ion. In such a way, all possible displacements of the atom are sampled. Notice that the width of the shower $\Delta\Theta$ is assumed to be significantly larger than the width of the detector $\delta\Theta$.

In the accumulated statistics, events due to ions in the showers ("secondary" ions) must contribute with weights w_i given by the probability for the atom to be displaced into the "hot" region. To calculate the weights, we note first that a weight should be ascribed also to the primary ion itself (W_i before the ith collision). In every collision, this weight is decremented by the already considered probability to scatter into the shower cone (trajectory "degradation"). Then, the weights w_i and W_{i+1} after the collision are updated as

$$w_i = P_i W_i/n, \qquad W_{i+1} = (1-P_i)W_i, \qquad (2)$$

where P_i is the integral probability of atom displacement into the "hot" region. As it should be, the sum of probabilities is conserved: $W_{i+1} + nw_i = W_i$. It is worthwhile to note also that this approach provides the correct absolute value of the scattering yield: the expectation value for a certain energy-angular range is equal to that obtained in a direct simulation in which the same number of ions is sent to the sample.

A similar procedure can be used for the simulation of the yield of recoiled atoms. When the ion crosses a lattice site occupied by an atom of the considered species, a "shower" of recoiled atoms is emitted. The "hot" region (see the right-hand side of Fig. 1) selects now those atom displacements that result in emission of recoiled atoms within the chosen cone. To account for all possibilities of recoiling, one additional recoil atom is emitted by sampling the remaining part of the Gaussian distribution. The resulting recoil is allowed to produce new recoils, analogously as the ion itself, and also scattering showers. Then, the trajectory of the ion is followed further with the atom displacement drawn according to the total Gaussian distribution. To describe the whole cascade, we consider also recoils produced by the recoiled atoms in showers. The result of these many possibilities is a strongly developed tree of cascade. Since all particles in the cascades have similar histories of collisions, the treatment using a recursive algorithm turns out to be very efficient.

One can easily recognize the similarity of this approach with the strategy of "importance sampling" used in the Monte Carlo numerical integration [13], where the values of the integration variable are sampled more densely in the regions that give the highest contribution to the integral. This similarity is not accidental because the scattering yield, as an average over the ion initial conditions and the thermal displacement of atoms, is, in fact, determined by an integral over a multidimensional space. Due to the inherent complexity
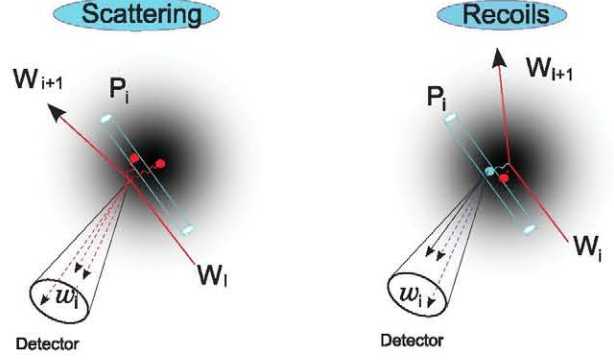

FIG. 1. (Color online) Illustration of the principle of selection of the "hot" region of atomic displacements used in the shower generation. The density of the Gaussian distribution of atomic displacements is represented by the gray cloud, while the "hot" region is indicated as the tube enclosing the region of impact parameters for scattering into the chosen angular cone (left panel). Analogously, the picture at the right panel illustrates the generation of showers of recoiled atoms. W_i and w_i are the weights ascribed to the trajectories of primary and secondary ions, respectively, while P_i designates the integral probability of an atom displacement into the "hot" region [see the explanations following Eq. (2)].

of this integral, the importance-sampling strategy can be used only on an intuitive basis as described above. In fact, we assume here that, even though the subsequent rescattering of ions in the showers tends to diminish their intensities, the final effect will be a dramatic increase of the number of detection events. It is clear that this strategy should be effective at high energies where the rescattering events are of little importance. However, the shower approach turns out to be efficient also at low energies where rescattering strongly modifies the flux of outgoing ions. The reason is that, fortunately, this adverse circumstance is well compensated by the fact that the primary ions themselves can leave with large probability the sample volume, and the numerous accompanying showers from the top layers will produce again an intensive flux in the direction of the detector. We can say that, in this case, the showers are used to improve the direct simulation at its last stage. These arguments show that the most serious difficulties in the application of the shower approach can be met in simulations for intermediate ion energies where the rare plural-scattering events result in violent fluctuations (see below how this problem can be eliminated).

Violent fluctuations in the accumulation of scattering events can have different causes. A first circumstance that should be noted in this respect is that the detection of the primary ion itself (with large weight W_i) is completely excluded if the showers are generated at every lattice site met along the ion path. Thus, the displayed fluctuations are entirely due to the dispersion of the values of w_i.

If the number of ions in one shower n is fixed, the value of the weight w_i for a certain scattering angle Θ depends on the position of the "hot" region with respect to the center of the Gaussian distribution of thermal displacements. Since the width of this distribution is rather small compared to interatomic distances, variations of several orders of magnitude in the values of w_i are possible. To improve upon this situation, it

is reasonable, instead of fixing n, to fix the value of the weights $w_i = w_0$, treating instead the number of ions in a shower as variable $n_i = P_i W_i / w_0$ (the fractional part is treated as the probability for sending one additional ion). A reasonable value for w_0 can be estimated by defining a maximum value for the number of ions n_i in the most intense shower. When $w_i \gg w_0$, the resulting effect is expressed as an increase of the number of detected events with a smaller weight w_0 from intense showers instead of one event with a large weight. An advantage in the inverse case $w_i \ll w_0$ is that the load on the computer due to the simulation of nonsignificant events is avoided.

An additional advantage of the above approach is that the discrete counts of such "quanta of probability" closely mimic the aspect of experimental data. The fluctuations of these counts are also the same as in the experiment. Basically, the simulation performs two types of averaging: over the initial conditions of impinging ions (the diversity of unperturbed trajectories of the ions) and over the thermal displacement of target atoms that perturb the ion motion. By variation of the value of the weight w_0, and consequently, the number n of ions in the showers, we can separately control the fluctuations due to the finite statistics of atom displacements.

The fluctuations due to the finite number of impinging ions can be controlled independently. The simplest way for doing this is just to increase the number of ions (with the associated proportional enhancement of the computation effort). However, in the case of simulation for a sample with crystalline structure, a more efficient approach can be proposed where the same strategy of "importance sampling" is applied. The contribution to the scattering yield from ions entering at different points of the crystal surface can be widely different. Therefore, it would be decisive to distribute the initial coordinates at the surface nonuniformly, giving preference to those that result in an increased scattering yield. Additionally, we must modify the initial values of the weight W_0, taking them as inversely proportional to the density of the distribution. This can be organized as a self-adapting procedure; such an approach is implemented in our computer code.

Another source of fluctuations is plural scattering. To illustrate how these fluctuations arise, we should mention the following. A reasonable criterion for the choice of the width of the showers $\Delta\Theta$ is that the showers should be wide enough to encompass the whole profile of multiple scattering on the outgoing path. One may believe in this case that the transport into, and the transport out of, the central region (of width $\delta\Theta$) are properly balanced in the showers. However, at low energies and/or for heavy ions, this condition is difficult to fulfill because the showers should be taken very wide and, as a result, only a small fraction of the ions in the showers reaches our small-aperture detector. But, if the above condition is not fulfilled, the reduction of ion flux in the shower cone due to diffusion outward is to be compensated by the plurally scattered primary ions. As this takes place, the main contribution comes from primary ions scattered accidentally into directions close to the cone mantle. Since the probability of further scattering into the cone is large for these ions [small $|d\Theta/db|$ in (1)], they produce rare but very intense showers of secondary ions with almost equal energies. As a result, the accumulated energy spectrum is disturbed by splashes in some channels.

We solve this problem in a similar way as described above: the plural scattering events are sampled more frequently than they happen in reality. For this goal, a second cone is considered, coaxial with the first and significantly wider. Wide showers are produced by the primary ion inside this cone in addition to the showers in the internal cone. Then, ions of the outer cone are allowed to send showers into the internal cone, similar to the primary ion. As a result, our goal is achieved because the weights of ions in the outer cone are small compared with the weight of the primary ion W_i, and the number of showers sent to the internal cone is now large enough to sufficiently smooth the accumulated energy spectrum. In principle, a whole hierarchy of such nested cones or even some smooth deformation of the density of sampling of atom displacements could be organized (the strategy of stratified sampling [14]). It turns out, however, that in many cases the implementation of the above-mentioned two-step approach solves practically all problems. This is illustrated in the next section [Fig. 7(a)].

The idea of stratified sampling has already been used in the simulation of ion scattering [15], although in a different form. The stratification was applied as an adaptive procedure in the sampling of random numbers. The author used this method for the simulation of scattering from amorphous samples using a Poissonian distribution for the intercollision distances. The application of the strategy of stratified sampling as described above relies on physical arguments and, therefore, results in an efficient procedure, which can be applied in general.

The last point that should be noted concerning the shower approach is that the emission of showers aiming to the detector mainly results in misses. This unavoidable drawback, when one is interested only in the yield on a small-aperture detector, turns out to be an advantage if one needs to simulate the two-dimensional (2D) angular distribution of the scattered ions. Such possibility is illustrated in the following section. In fact, we are capable of calculating with the present approach three-dimensional (3D) distributions including the energy scale.

The implementation of this algorithm as a FORTRAN program incorporates all important elements of the binary collision model [1]. A wide possibility to choose the type of the interaction potential is provided. The scattering integrals for binary collisions as functions of impact parameter and energy are tabulated at a preliminary stage of the calculation and used afterward by interpolating with splines. Additionally, the impact-parameter dependence of inelastic energy losses is considered. To account for the simultaneous interaction with two or more atoms, we sum the deviations of ion motion due to the interaction with individual atoms. In principle, all this provides the possibility to perform simulations for energies in a wide range.

At high energies, the procedure of generation of showers can encounter difficulties due to restrictions in the accuracy of computer calculations. The reason is that the width of the "hot" region in the transverse plane becomes so small that an accurate transformation of impact parameter to scattering angle may be practically impossible. For this reason, if this width becomes smaller than 0.01 of the thermal vibration amplitude u_1, the procedure of shower generation is inverted: the scattering angle is sampled within the shower cone and then, in reverse order, the impact parameter is calculated.

Materials of any crystal structure, including compounds, with rectangular unit cells can be described in the input. On a lower level, however, the program works with a description in terms of Wigner-Seitz cells. In principle, this provides the possibility to also treat wider classes of structures. Amorphous structures can be simulated by a random rotation of the crystal lattice after each collision. The surface of the sample is defined as an imaginary plane appropriately located with respect to the crystal lattice. If the detector position is defined to be at the back side of the sample, a simulation of transmission through a crystal slab of a given thickness is performed. As output data, depth profiles of close collisions, energy spectra, and 2D angular distributions of scattered ions or recoiled atoms of a certain species are calculated. Finally, the procedure is automated to calculate angular scans for a stepwise rotation of the target, the detector, or the beam direction around a given axis. The FORTRAN code of the program TRIC is supplied with a graphical user interface, allowing us to comfortably supply the input data, to run the calculations, and to inspect the simulation results. The software is available on the Internet and all technical details together with the description of the underlying physical model are described in the supplied instructions.

The efficiency of the proposed approach depends on the choice of the type of stratification, the width of the shower cones, and the numbers of ions emitted in each shower. It is hardly possible to give a "universal" recipe for the choice of these parameters for a given condition. Our experience gained by the use of the program shows that, by estimating the possible role of plural and multiple scattering in the considered conditions, one can easily guess values for the parameters that are close to optimal.

As a measure of efficiency, the flux directed to the detector should be estimated. In the case wherein the flux of incoming ions is assumed to be uniform, the yield of scattering from one atomic layer is easy to estimate. For example, the yield of scattering of 100-keV protons from silicon within a cone of $10°$ width amounts to $\sim 10^{-8}$ per incoming ion. Precisely the same yield is reproduced in the direct simulation. On the other hand, it is realistic for the shower approach to obtain, on average, one ion from each layer within a shower of the same width. Of course, the volume of calculation in this case is larger; the same computations, together with the calculation of the trajectory of the incoming ion, need to be performed for each ion in the showers. For lower energies, the relative efficiency decreases. For example, the yield of scattering of 10-keV Ar ions from one layer of copper is $\sim 10^{-3}$. This gain in efficiency is less expressive; however, the problem itself is also much easier. The increase in efficiency in the treatment of the plural scattering is more significant; in the double-cone approach, the procedure of shower generation is performed twice. By applying the stratification of initial conditions in the simulation of channeling, we artificially increase the ion flux near the atom locations. As our experience shows, this can result in an additional increase in efficiency by one order of magnitude.

In general, the shower approach solves the main problem; simulations of ion-solid collisions become possible, even using an ordinary PC. In this way, all the results shown in the next section were obtained. The required CPU times ranged from several minutes, as for the simulations of low-energy

ion scattering, to several hours, as for the data shown in Fig. 8. In cases as in this example, wherein whole histories of binary collisions need to be calculated up to large depths, the simulation becomes a rather difficult problem.

III. PROGRAM TESTS AND APPLICATIONS

In this section, we present several examples of applications of our program to simulations of different experimental conditions. The calculations were performed using simple and commonly used models of ion-atom interaction. At low energies, as in the cases shown in Figs. 2–4, the angle of deflection Θ and the elastic energy loss in each collision were calculated using the Ziegler-Biersack-Littmark (ZBL) potential [16]. For determining the inelastic energy loss as a function of the impact parameter $\Delta E(b)$, the Oen-Robinson model [2] was applied. At higher energies, the Moliere potential was used and $\Delta E(b)$ was taken to be proportional to the electron density along the ion trajectory. In order to check the sensitivity of our results to the shape of $\Delta E(b)$, we varied the screening parameter a in the potential while determining the electron density from the Poisson equation. For the type of data we simulated, we found a negligible sensitivity and we chose for a a value twice higher than its standard value. In all cases, $\Delta E(b)$ was normalized to the Ziegler stopping cross section [16]. To account for the energy loss straggling, we chose in each collision the value for the actual energy loss T randomly according to the distribution for free electrons: $dP(T) = k/T^2$ $(T_{min} < T < 2mv^2)$, where
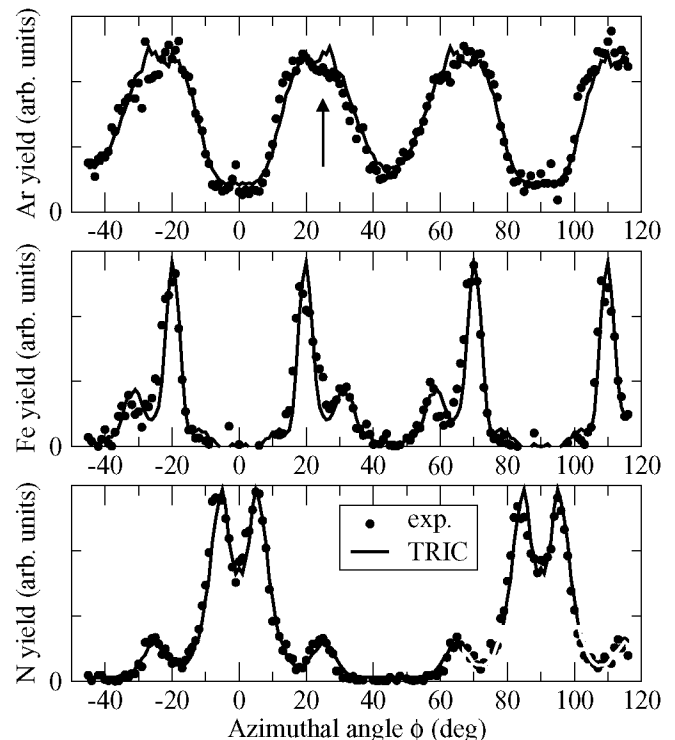

FIG. 2. (Color online) Comparison of simulated angular scans for the scattering of 5-keV Ar^+ ions from the surface of a $Fe_4N(100)$ crystal with the experimental results from Ref. [18] (top panel). The yield of recoiled Fe and N atoms is shown in the center and lower panels, respectively. See text for more details.
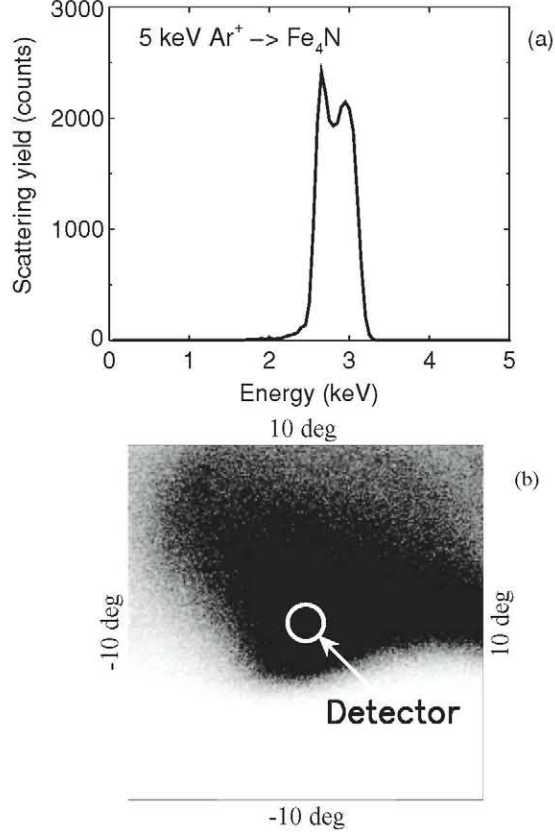
3000

Scattering yield (counts)

5 keV Ar$^+$ -> Fe$_4$N

(a)

2000

1000

0

0   1   2   3   4   5

Energy (keV)

10 deg

(b)

-10 deg

10 deg

Detector

-10 deg

FIG. 3. Energy spectrum of Ar$^+$ ions scattered from the (100) surface of a Fe$_4$N crystal for the orientation indicated in Fig. 2 by an arrow. The lower part of the figure shows the angular distribution of scattered ions. Results are shown using a gray scale where darker represents higher intensity.

$k = 2\pi Z_1^2 e^4 n/mv^2$, Z_1 and v are the atomic number and velocity of the ion, respectively, and n is the integral of the electron density along the ion trajectory. The minimal value $T_{\min}$ was determined by the condition that the mean value of T must be equal to ΔE. Such a form of energy loss distribution has been used previously (see Ref. [17]). Finally, the multiple scattering on electrons was accounted for by a broadening of the deflection angle Θ with a Gaussian distribution of width $\Delta\Theta = \sqrt{(m\Delta E)/(M_1 E)}$. Here, M_1 and E are the mass and energy of the projectile, respectively.

The first example concerns the scattering of 5-keV Ar$^+$ ions from the (100) surface of a Fe$_4$N crystal as studied experimentally in Ref. [18]. The structure of this crystal can be considered as face-centered-cubic (fcc) Fe with an additional N atom located at the center of the unit cell. The top layer of the stable (100) surface is the layer containing both Fe and N atoms. Depending on the growth conditions, the surface can exist in two types of reconstructions, $c4$ and $4pg$. Figure 2 shows simulated angular scans for the $c4$ surface, which differs from the bulk-terminated one only by an outward displacement (of 0.23 Å) of the nitrogen atoms. In these scans, the crystal is rotated around the surface normal, which is coplanar with the beam and the detector directed, respectively, at 42° and 12° from the surface at opposite sides of the normal. The yields of scattered Ar as well as of Fe and N recoils were calculated.
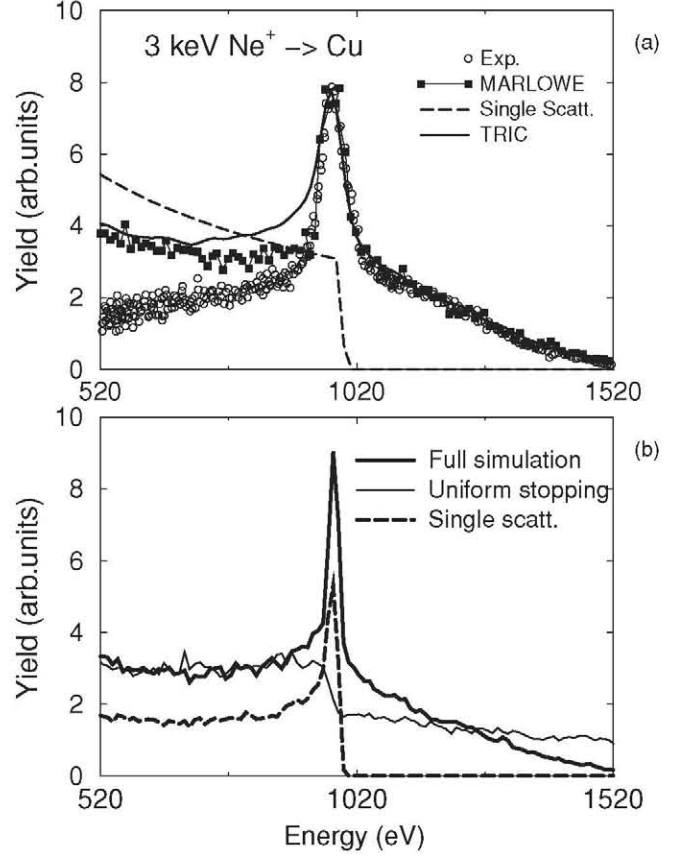

10

Yield (arb.units)

3 keV Ne$^+$ -> Cu

(a)

○ Exp.
■ MARLOWE
--- Single Scatt.
— TRIC

8

6

4

2

0

520   1020   1520

10

Yield (arb.units)

(b)

— Full simulation
— Uniform stopping
--- Single scatt.

8

6

4

2

0

520   1020   1520

Energy (eV)

FIG. 4. (a) Comparison of our simulation results for 3-keV Ne$^+$ ions scattering from amorphous copper with experimental data [20] and results of simulation with the MARLOWE code. The simulation results were folded with a Gaussian distribution in order to simulate the experimental energy resolution of 40 eV. The dashed curve corresponds to a calculation in the single-scattering approximation. Panel (b) shows simulations performed by applying additional approximations. The dashed curve corresponds to a simulation using the single-scattering approximation with impact-parameter-dependent energy losses, while the thin solid curve was calculated considering the energy losses as the result of a uniform stopping force.

In Fig. 3, the energy spectrum of scattered Ar is shown for the orientation indicated in Fig. 2 by an arrow. Also shown is the angular distribution of scattered ions. The latter illustrates the blocking of scattering from the top-layer Fe atoms by rescattering from nearby atoms; this largely explains the strong variation of the yield in the angular scan. These variations are strongest when either the beam or the detector are located close to the scattering "horizon," which is formed by reflection from the surface as a whole (very small intensity at the bottom part of the angular distribution). In the case shown in Fig. 3, the detector is located at the edge of the shadow cone. The shape of the energy spectrum as an isolated peak is due to the blocking of the particles scattered from atoms of deep atomic layers by atoms of the top layer. This shape of the spectra favors the choice of the energy window for the performance of angular scans. The double-humped structure is explained by the effective competition of single and double scattering from Fe atoms of the top layer [19].

As seen in Fig. 2, the experimental results are well reproduced in the simulation both for scattered ions and for recoils of the two atomic species. Note that, in order to achieve this agreement, the screening radii in the used ZBL potentials were properly reduced, as commonly done for the description of scattering of heavy ions at low energies. With the same potential, the data for the more complex reconstruction 4pg are also reproduced with the same quality. In general, these results demonstrate that the quality of the description of the experimental data is similar to that provided by the code MATCH [18]; in fact, the results of both simulations can hardly be distinguished when plotted together.

The second example, shown in Fig. 4(a), corresponds also to a LEIS experiment, now of 3 keV Ne$^+$ scattering from a polycrystalline Cu sample. Under these conditions, an intriguing peak is observed [20] in the experimental spectrum at the energy corresponding to a single Ne-Cu collision. Here, the beam incidence was along the surface normal and the scattering angle was 129°. For these strong-scattering conditions, the code MARLOWE is capable of reproducing the general shape of the spectrum, including the surface peak. Our simulations shown in the same picture also reproduce the spectrum shape; some differences with the MARLOWE simulation at low energies may be due to a difference in the potentials used for the description of the ion-atom interaction or in the treatment of inelastic energy losses. In general, the shape of the spectrum under these conditions differs considerably from that predicted by the single-scattering model (also shown in the figure). The authors of Ref. [20] associated the appearance of the peak with the onset of plural and multiple scattering in the deeper layers. Here, by using our advanced possibilities for simulation, we are able to come to more detailed conclusions about this striking phenomenon.

From general considerations, one can conjecture the following two reasons for the formation of the surface peak. First, due to the large variations of kinematic energy losses, the plural collisions yield a broad energy spectrum in scattering (in particular, this explains the presence of the high-energy tail), while ions that undergo only one close collision with atoms (single-scattering fraction) have a principally different energy distribution. It can be assumed in the considered case that, due to the strong scattering, only the ions coming from shallow depths can leave the sample without additional rescattering. Thus, all these ions have almost equal energies and form a peak in the energy spectrum, which is superimposed on the plural-scattering background. On the other hand, the surface peak can appear even in the single-scattering model if the impact-parameter dependence of the energy loss is taken into account. Indeed, in the case of sharp localization of energy loss at small impact parameters, ions scattered at shallow depths often do not experience significant energy losses on both ingoing and outgoing paths. So, again, many scattered ions have almost the same energies near the high-energy edge of the energy spectrum.

To establish the relative role of these two effects, we performed simulations with certain modifications of the model of interaction. Figure 4(b) shows the results of a simulation where both nuclear and electronic energy losses are replaced by equivalent continuous stopping forces (thin solid curve). As seen, by using such a description of energy losses, the spectrum changes drastically. First, the surface peak disappears since ions scattered from different depths now have different energies. And second, the variation of kinematic energy loss in plural scattering is now not effective and, as a result, the shape of the spectrum on both sides of the surface peak is also strongly modified. On the other hand, in the shower approach, we can also check the role of plural and multiple scattering. For this purpose, a simulation was performed using a special procedure in which in all collisions before and after the emission of the shower, the ion deflection was canceled. The difference with the ordinary single-collision model is that here, in all collisions, the energy loss is treated as a function of the impact parameter. These results are also shown in Fig. 4(b) (dashed curve) and demonstrate that the multiple and plural scattering events themselves are of minor importance for the formation of the surface peak. In summary, we can conclude that the surface peak reflects mainly the correlation between scattering and energy loss: at these low energies, ions scattered in deeper layers have more chances to exit from the sample if they do not rescatter strongly on atoms of the upper layers and, consequently, the energy losses in the passage through these layers are also small. At energies below the surface peak, both the present and MARLOWE simulations differ significantly from the experimental results. A discussion of the difference is out of question, however, because the shape of the experimental spectrum was not reliably determined due to the uncertainty in the efficiency of the detector employed. On the other hand, there are also significant differences between the results of both types of simulations. Assuming that both simulation codes perform a reliable treatment of the problem, the difference must be ascribed to the use of different models of ion-atom interaction (interaction potentials or inelastic energy losses).

It is worthwhile to note that the program TRIC passes here a serious test: the simulation with shower generation produces results identical to those obtained when running the program in the direct simulation mode.

Turning now to medium-energy ion scattering (MEIS), we show in Fig. 5 the angular distribution of 100-keV He$^+$ ions scattered on a Si(100) crystal. The geometry (the ⟨112⟩ axis is in the center of the position-sensitive detector) and the two depth ranges chosen for collection of the scattering events are equivalent to the experimental conditions used by Kobayashi [21]. The results of the simulations are also very similar to the experimental results: even at such relatively small depths, the washing out of the blocking pattern is well seen, first of all for the narrow channels. Although this effect of rechanneling is very predictable, its detailed demonstration in the referred experiment is rather interesting and our simulations support these results. Note that, with 10^5 ions being sent to the crystal in the simulation, the total yield over the detector amounts here to ∼0.01 only (recall that, in the direct simulation, this would be an expected number of counts). In this sense, the results of our simulation are unique, and it is hardly possible to obtain them using other known methods.

In Fig. 6, we show MEIS angular scans for the yield of 100-keV protons scattered from Y atoms of a YSi$_2$ monolayer epitaxially grown on Si(111) (a side view of the structure is shown in the picture). The results of the measurements and of the simulations with the program VEGAS are taken from Ref. [22]. The dips in the scans are due to blocking of the
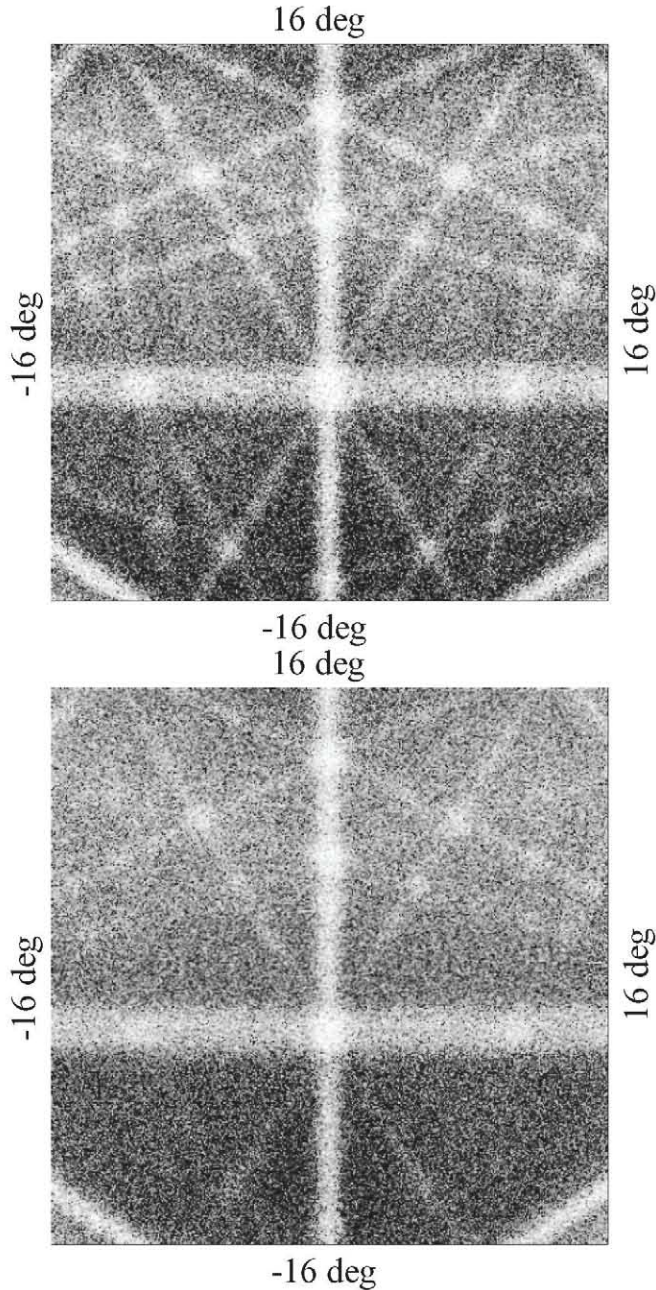
FIG. 5. Results of the simulation of 100-keV He$^+$ ions scattering from a Si crystal. The scattering yield is accumulated from depth ranges of (a) 5–30 nm and (b) 30–55 nm. The displayed angular range lies around the $\langle 112 \rangle$ crystal direction.

FIG. 6. Comparison of our simulations (solid curves) of angular scans for 100-keV p scattering from a monolayer-thick YSi$_2$ film on Si(111) with the result of a VEGAS simulation (dashed curves) and experimental data [22] (dots). The direction of the incident beam is parallel to (a) the $\langle \bar{1}00 \rangle$ and (b) $\langle 0\bar{1}\bar{1} \rangle$ directions of the Si substrate, while the detector lies in the plane $(01\bar{1})$. The structure of surface layers is shown at the top. The yield is normalized to the Rutherford cross section. The dashed-dotted curve in the lower graph shows results for a trial structure with Y atoms located at the top layer.

scattering from Y atoms by Si atoms of the upper layers. All parameters in the two simulations are taken to be identical. The achievement of a perfect agreement by optimization of the parameters of crystal structure and lattice dynamics is, in fact, the actual goal of the MEIS analysis [22]. Here, we emphasize only that results of our simulation coincide well with the results of the VEGAS simulation. In principle, this is predictable for such thin layers (see the Introduction). As an additional test of our simulation procedure, we performed simulations also for the case of the top layer being terminated by Y atoms instead of Si atoms. As seen in the figure, the
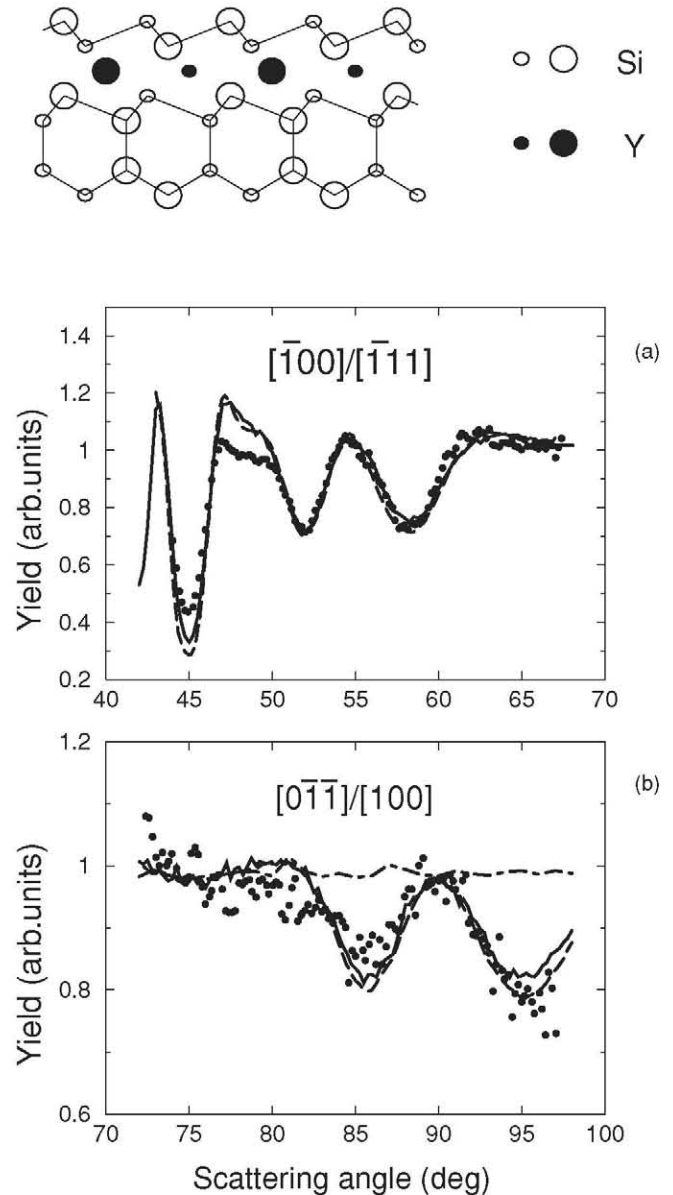
scattering yield in this case simply reproduces the Rutherford cross section.

The most difficult problem for simulations is the description of scattering in conditions where the contribution from plural scattering is significant. Aside from low energies, this happens also in the medium- and high-energy cases where heavy-ion beams and/or heavy-atom targets are considered. For amorphous samples, a very valid approach is to consider the sample as a continuous media and to draw the path lengths between close ion-atom collisions according to a Poisson
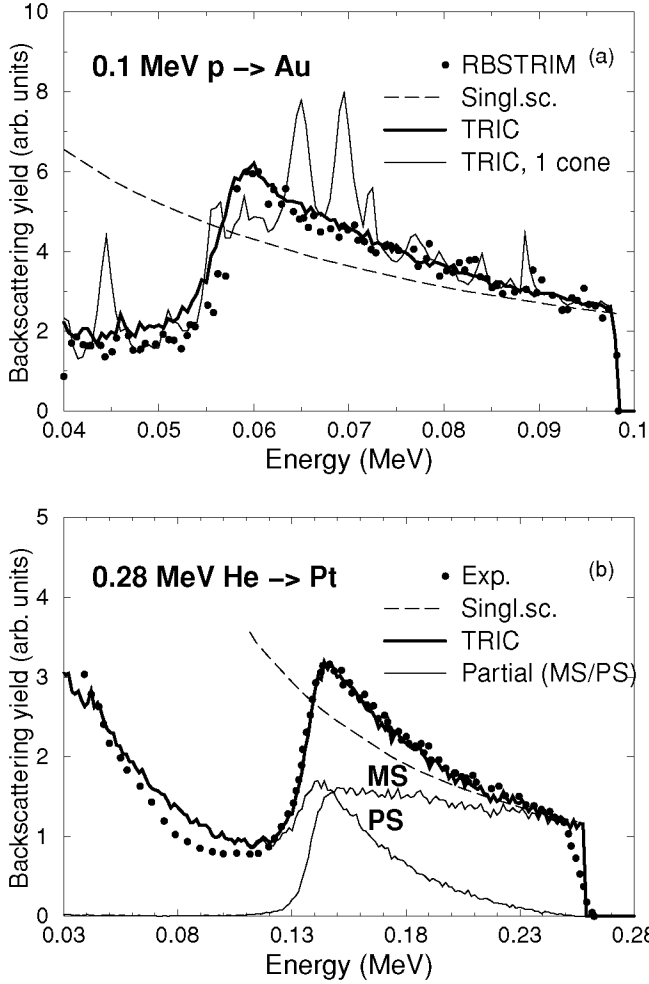
FIG. 7. Backscattering energy spectra of 0.1-MeV p scattered from a 1000-Å Au film (a) and of 0.28-MeV He ion scattering from a 1130-Å Pt foil (b). The simulation results and the calculations in the single-scattering approximation are compared with the RBSTRIM simulation [25] and with experimental data [26]. Results of simulation in the single-cone approximation are shown in the top panel. The lower graph shows the partial contributions of ions from the internal (MS) and outer (PS) cones.

distribution. Many simulation programs of such type have been developed (see Ref. [23] for a review), which include different possibilities to treat multiple and plural scattering. The most developed model of interaction is used in the widely known code TRIM [16], but its efficiency is not sufficient for simulation of backscattering. Biersack *et al.* [24] found a possibility to increase the efficiency of this code up to an appropriate level without a significant loss of accuracy of the description. Figure 7(a) shows their results [25] for scattering of 100-keV protons from a 1000-Å-thick gold foil, together with the results of simulations done with the shower approach. As seen, the two approaches produce, in fact, identical results, also with similar computation efforts. The only visible difference is at the low-energy tail, where the yield is entirely due to plural scattering. Therefore, these results can be regarded as a confirmation of the accuracy of the TRIM approach by comparison with an exact treatment of the model with a similar structure of the target.

Figure 7(b) shows the simulation results compared with results of an experiment [26] of 280-keV α particles scattering from a 1130-Å-thick Pt foil. In general, the agreement is also satisfactory here. In the two cases shown in Fig. 7, the shapes of the spectra differ significantly from those predicted by the single-collision model. The yield is higher for the main part of the spectra; also, the low-energy tail, which is entirely due to the plural scattering, can not be predicted at all by the single-collision model. To reproduce all these features with an accuracy above the fluctuation level, the showers must be generated in wide cones. In the cases considered, the double-cone approach (see Sec. II) was used with a half-width of 40° for the internal cone and of 160° for the outer cone. In this way, at least the double scattering is treated with special efforts. The effect of the use of the two cones is illustrated in Fig. 7(a) where, for comparison, the simulation results obtained in the single-cone approach are included. The level of fluctuations in this case suggests that much larger efforts are necessary to achieve the same level of precision of simulation as in the double-cone approach. To demonstrate the role of plural scattering in more detail, we show [Fig. 7(b)] separately the contribution of showers emitted by ions moving in the outer cone. It is seen that these histories of collisions completely explain the nature of the low-energy tail and, in general, contain mainly the effect of plural scattering. On the other hand, the partial spectrum of ions in the showers produced directly by the primary ion is influenced by multiple scattering. It looks surprising at a first glance that, in contrast with the predicted increase of the yield compared to the single-scattering spectrum [27,28], this partial spectrum shows the inverse ordering due to multiple scattering. However, this is natural because, in this partial spectrum, we do not consider the compensation of the transport out of the internal cone by the counter transport from the external region.
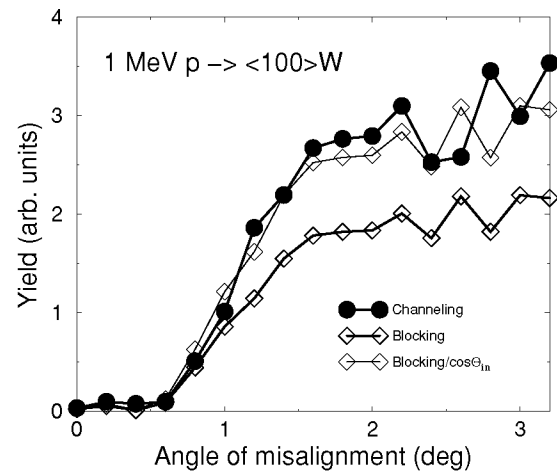

FIG. 8. Channeling and blocking dips calculated for the case of irradiation with 1-MeV protons of a tungsten crystal along the ⟨100⟩ axial direction with detection at a 135° scattering angle and in the inverse geometry. The yield is calculated in the energy window corresponding to scattering from a depth range between 2500 and 3500 Å. The blocking dip normalized to the thickness of the layer along the beam direction is also shown.

As a last example, we reproduce in our simulations an experiment [29] performed in the early times of channeling studies with the special aim to test the Lindhard time-reversibility rule [10]. Here, a proton beam of high energy (1 MeV) is incident on a W crystal along the $\langle 100 \rangle$ direction (close to the surface normal) and the protons scattered by 135° into a random direction of the crystal lattice are detected. In the inverse situation, the beam and detection directions were interchanged. The angular spread in the beam and the aperture of the detector were approximately equal, 0.1°, and the yield was measured within an energy window corresponding to a layer of 1000 Å thickness at a depth of 3000 Å. The yield was measured as a function of the beam misorientation in the first case and as a function of the detection angle relative to the $\langle 100 \rangle$ channel in the second. The simulated channeling and blocking wells are shown in Fig. 8. Their widths are similar and coincide well with the experimental results. To achieve also an agreement between the absolute values, we had to normalize the yield to the path length of the incoming ions inside the considered layer (by multiplying the yield by $\cos \Theta_{in}$ in the second case, where Θ_{in} is the angle of beam incidence relative to the surface normal). In general, the time reversibility is confirmed by the simulation similarly as in the referred experiment.

IV. DISCUSSION

In this section, we discuss the advantages of the proposed approach in comparison with those used currently in simulations of swift ion-solid interaction. First, the shower approach is equivalent in accuracy with the direct simulation as performed by the code MARLOWE. The difference lies only in the much higher efficiency (lower level of statistical fluctuations) as illustrated in Secs. II and III. This is, in fact, of primary importance since, in many situations, extensive analysis of experimental data becomes feasible. Furthermore, for medium and higher ion energies, a proper simulation of multiple and plural scattering is not possible at all by other methods. For medium-energy ion scattering from amorphous samples, the accelerated version of the code TRIM [24] seems competitive, but this is achieved at the cost of additional approximations.

Barrett's approach [3], followed by later developments such as the programs FLUX [6] and UPIC [8], is based on the single-scattering model and has, consequently, its region of applicability restricted to the cases where multiple and plural scattering can be neglected. In principle, the calculation of the close-encounter probabilities in this approach bears some similarity with our procedure of shower generation. In this way, the picture of collisions with small impact parameters is well reproduced. However, only one of the two segments of the trajectory is described realistically, while the other is approximated by a straight line.

In fact, the only previously proposed method demonstrating, for the case of low-energy ions, an adequate and simultaneously efficient treatment is the program MATCH [11]. The time-reversing procedure used in this program is closely related with the shower approach and it is rather instructive to compare both strategies in detail. As found in simulations of LEIS (of the type shown in Fig. 2), the use of the reversing

approach is very competitive compared with the shower approach. It is not clear, however, if this is also the case in other conditions. In the rest of this section, we perform an analysis aiming to clarify whether there are some important differences in the basis of two approaches, allowing us in some cases to choose one of them as more convenient. Note in advance that, in practical application, one would most probably prefer the shower approach since, in its implementation, the reversing approach is much more cumbersome.

To clarify the above question, let us look closer at the basis of the reversing approach. In the case of pure potential scattering, the time reversibility of ion motion suggests that the probability for an ion of the beam to reach the detector and the probability of the inverse scattering are directly related. The yield of scattering is simply proportional to the phase volume of the detector, i.e., its acceptance. This property is a simple consequence of Liouville's theorem: The scattering yield is obtained as the overlap of the flux of all scattered ions with the detector acceptance and the volume of the region of overlap is invariant with respect to the time translation. In the simulation, this means that there is no preference for the time-reversed mode compared to the direct reproduction of scattering; the two methods require comparable efforts. The presence of energy losses does not affect this conclusion. The reversing approach for simulation proposed in Ref. [9] assumes a convolution of the beam profile with that of the detector at an intermediate position when ions reach a certain lattice site.

The procedure is illustrated in Fig. 9. We consider the scattering of a single ion of the beam on a "frozen" lattice additionally averaged over thermal displacements of one particular atom. In fact, the whole procedure of the simulation consists in the solution of such "elementary" problems (see the first paragraph of Sec. II). The region Φ'_d shows schematically the detection profile shifted backwards in time to the considered site. This region of phase space should be considered as actually five dimensional (the longitudinal coordinate is not relevant). The possible states of motion of the ion after collision with the atom of the considered site are distributed over a three-dimensional hypersurface. The dimension is determined by the dimension of the vector $\mathbf{R}$ of atom displacements. This lower-dimensional hypersurface is shown in Fig. 9 schematically as the thick solid curve. The probability of the interesting event of scattering can be determined by the convolution of the distribution $\delta\Phi'_b$ of ion states with Φ'_d.

In the simulation, we represent the shifted detection profile Φ'_d by a set of time-reversed trajectories ending up in the detector (shown in Fig. 9 by dots). Then, by selecting those trajectories that can be connected with the trajectory of the incoming ion, we can reconstruct specific examples of whole trajectories of detected ions. A connection takes place when some point in Fig. 9 lies on the hypersurface $\delta\Phi'_b$. The displacement of the atom $\mathbf{R}$ in the interesting collision is determined. It is clear, however, that exact connections are not probable. Thus, to obtain sufficient statistics in the simulation, we have to introduce a certain tolerance for the connections. In Fig. 9, this is illustrated as the shadowed area around the hypersurface $\delta\Phi'_b$: the points, states of outgoing ions, that fall within this region are associated with possible connections. Hence, this approach assumes from the outset
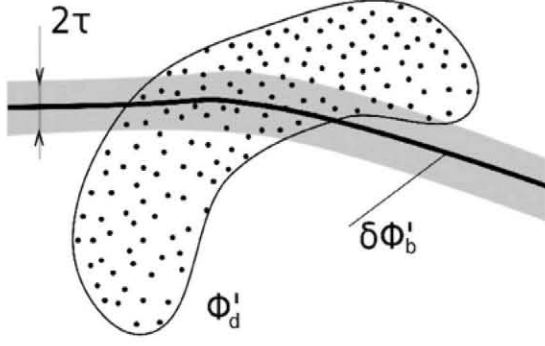
FIG. 9. Schematic illustration of the procedure of flux convolution used in the reversing approach (see the text for explanation of details).

a certain approximation. It is assumed that, in fact, the flux of ions before the collision is so smooth that its variations within the tolerance region can be neglected. Clearly, taking a weaker criterion for tolerance will increase the efficiency of the simulation. However, in the general case, it is difficult to estimate *a priori* how smooth the flux at a given site will be and this means that, strictly speaking, the resulting accuracy must be checked in each case by repeating the calculations with a tighter tolerance.

In the calculation of the scattering yield, the contributions of the found connections of trajectories are not equal but depend on the probability for the atom to be displaced to the required position **R** given by the distribution density $D(\mathbf{R})$. As derived in the Appendix, these contributions (the weights for the connections) are determined by the expression

$$w = \frac{S_0 \Delta \Omega \Delta E}{N_{\text{out}}} \cdot \frac{1}{\tau_y \tau_E \sin \Theta} \frac{d\sigma}{d\Omega} D(\mathbf{R}). \qquad (3)$$

Here, Θ is the angle of deflection in the ion-atom collision and $d\sigma/d\Omega$ is the differential cross section for ion scattering from the atom. The tolerance for trajectory connection enters here through two parameters: τ_y, the distance between trajectories along the direction perpendicular to the scattering plane, and τ_E, the discrepancy between energies. The first fractional factor in the previous expression includes the dependence on all other parameters and ensures the correct absolute value of the yield. These parameters include the area of the unit cell of the crystal surface S_0, the solid angle corresponding to the detector aperture $\Delta\Omega$, the range of energies of the detected ions ΔE (the simulation gives the yield integrated over this range), and the number of calculated outgoing trajectories N_{out}.

Equation (3) has a simple meaning. The first fractional factor in the right-hand side is the phase volume per outgoing trajectory as it is determined by the detector acceptance. The remainder in the right-hand side is the density of flux of the ion after the collision averaged over the distribution of atom thermal displacements **R**. With nonzero tolerances τ_y and τ_E, the flux is distributed within a layer of finite "thickness" around the original 3D hypersurface.

One can readily see that this procedure also uses a special sampling of atomic thermal displacements. Indeed, only those **R** are selected here that, with full certainty, result in interesting collisions. In this respect, the shower approach seems to be less efficient because, by far, not every sampling of **R**

results in a useful outcome. However, one should additionally consider the following two circumstances. First, to shift the detection profile to the considered site, we have to calculate a sufficiently large number of outgoing trajectories, and the computational cost for this could be comparable with that in the direct simulation. The second disadvantage in the reversing approach is the possibility of strong fluctuations of the values of the weight [Eq. (3)], first of all due to the strongly varying $D(\mathbf{R})$. The relatively rare events of plural scattering, where all except one of the close collisions are treated in the ordinary way, can also result in exceedingly large weights w. In such cases, additionally, the cross section $d\sigma/d\Omega$ can have large values. It is easy to verify that the same variations of the weights would be found in the the direct simulation if one were to use an inconvenient uniform distribution instead of the natural (Gaussian) distribution of **R**. Such coincidence is not accidental; when pairs of trajectories are connected, the corresponding displacement of the atom **R** is determined only by the kinematics of the binary collision and does not depend on anything else.

The yield of collisions with the atom, determined in this way, must be additionally averaged over the distribution of displacements of all other atoms. This means that one needs to repeat the procedure described above for a sufficiently large number of randomly chosen configurations of the displacements. Furthermore, one should be careful when the total yield of scattering is determined as the sum of the contributions of different atoms. It is easy to understand that considering all possible connections of trajectories will inevitably result in repeated account of the same histories of motion. In fact, the same problem is treated in the shower approach when we account for "beam degradation." An analogous scheme could be applied here also, although this would result in an exceedingly cumbersome computational algorithm. The problem is partially eliminated in the algorithm of the program MATCH, where only close collisions are treated by trajectory connection. This approach works well when each ion experiences only one close collision but could fail in the treatment of plural scattering.

The use of this approach turns out to be advantageous in two cases important for applications: scattering of medium-energy light ions, MEIS, and low-energy heavy ion scattering, LEIS. In both cases, simplified descriptions of ion-solid interaction are possible without significant loss in accuracy. Both simplified versions of the reversing approach are sufficiently efficient and, in the absence of other alternatives, they are widely used in the analysis of experimental data. In MEIS (with a predominant use of beams of hydrogen ions), the picture of single close collision is very adequate. As a first approximation, the fluctuations of the angle of scattering in the main collision can be neglected. Also, the energy of ions is considered as directly related to the depth of scattering. As a result, the description of ion fluxes is reduced to the form of dependence on the transverse coordinates only. The convolution of such fluxes, taking into account additionally the probabilities of atom displacements and the scattering cross sections, is a well tractable problem. This is the basis of the widely used program VEGAS. A more general approach is applied in the program SILISH [30]. In this case, the trajectories are connected accurately neglecting only the energy relation.

The results in Ref. [30] show that such a minimal simplification turns out to be sufficient for the simulation to become possible, at least for the description of scattering from one monolayer.

The reversing approach is suitable to treat also LEIS of sufficiently heavy ions (the case of strong interaction). One example of results for such conditions is shown in Fig. 2. A favorable circumstance here is that only scattering from a few atomic layers is important. In addition, the large scattering cross section implies that the scattering on atoms results in strongly dispersed fluxes and, therefore, the result of their convolution is not very sensitive to the specific details of the flux distribution. The program MATCH was developed to simulate scattering with such conditions.

Compared to the reversing approach, the simulation algorithm proposed in this paper treats scattering exactly within the binary collision model. At the same time, it demonstrates an unprecedented efficiency. It is also less cumbersome; the only difference with the direct method lies in the way the atom displacements are sampled. This means, in particular, that one can easily incorporate, if necessary, any additional features of the binary collisions such as energy loss straggling or charge exchange. Additionally, the shower approach is capable of reproducing in one run the energy spectrum of scattered ions (or recoiled atoms) and also their angular distribution in a wide range. As demonstrated by the examples presented in Sec. III, these two features make the method exceptionally powerful. Compared to this, the reversing approach is dramatically nonefficient. In fact, to obtain such results, one has to repeat the calculations for each bin in the energy spectrum (with a width ΔE) and for each bin $\Delta \Omega$ of the 2D angular coordinates.

To finish this section, we make some remarks intended to clarify possible consequences of the reversibility rule for the interpretation of experimental results. It is seen in Eq. (A6) that, under the assumption of a pure potential motion of the ions (the Jacobians $J_{in} = J_{out} = 1$), the yield of scattering from one lattice site is symmetric under exchange of the beam and detector directions provided that the flux in the beam is also uniform and the respective phase-space volumes are equal. The latter condition is less relevant because the difference can be simply accounted for by a factor in the yield. Therefore, we can conclude that the yield of scattering from one atom for a given beam-detector configuration is determined if it is known for the inverse situation. However, in the measurements of the yield in a certain energy window, as ordinarily made, the effective number of contributing atoms can be different. This fact was taken into account in the transformation of the data shown in Fig. 8.

Rigorously speaking, the reversibility rule is justified only when the picture of potential motion of the ions is assumed (pure potential scattering on infinitely heavy ions). In a real experiment, this rule can be violated due to the recoiling of the atoms and due to the manifestation of their internal degrees of freedom, energy losses, and multiple scattering on electrons. However, in the performed simulation, these features were not considered and, in fact, this simulation is nothing more than a test of sensitivity to the round-off errors unavoidable in numerical calculations. In principle, it is not obvious from the outset that the trajectories of ions and, consequently, the final results are stable with respect to these errors. Thus, any attempts to study the physical effects capable of leading to a violation of the reversibility rule should be preceded by a simulation as that performed here.

V. CONCLUSIONS

The shower approach proposed in this paper effectively solves the main problem of simulation of ion scattering from solids within the binary collision model, which is the elimination of the violent statistical fluctuations in the Monte Carlo sum. This is achieved by specific improvements of the direct simulation approach: the use of the strategies of importance and stratified sampling. As a result, the computer power required for simulation is reduced by several orders of magnitude. This is, in fact, a decisive advantage allowing us to address simulation problems that can not be treated with other methods. As examples, we can mention the plural scattering of medium-energy ions and the simulations of 2D angular distributions. As discussed in Sec. IV, our method avoids also many shortcomings inherent to alternative approaches. It is argued, in particular, that this method allows a reliable treatment of the rare events of plural scattering. Such a possibility is especially important because the plural scattering is also not amenable to theoretical treatment.

We performed a detailed analysis of the approach based on the convolution of fluxes of incoming and outgoing ions, as performed by the programs VEGAS and MATCH. The latter program offers an alternative for an exact treatment of the binary collision model, including multiple and plural scattering. However, as follows from the arguments presented in Sec. IV, the main illness of the direct simulation, violent fluctuations in the accumulated statistics, is inherited by this method. In general, the proposed shower approach represents an effective replacement of widely used algorithms of simulation providing qualitatively new possibilities for the analysis of experimental results.

Simulations with the code TRIC can be performed for large classes of crystal structures and provide a detailed picture of scattering or recoil yields in the form of energy and angular distributions. All these qualities are promising for a wide use of the developed computer code both in basic research and in the analysis of materials. In particular, this provides the possibility to efficiently compare measurements with simulations made for many trial structures allowing in this way precise structural analysis. Currently, the alternative for analysis of MEIS results is the program VEGAS. It has, however, many restrictions in its application. The level of approximations used does not ensure a sufficient accuracy of the simulation for ions other than the lightest H and He. Also, this program does not provide energy spectra of scattered ions, an experimental result containing a large amount of information. In addition, it is very hard to account with this program for an intimate feature of lattice dynamics, the correlations in thermal vibrations, to which data such as those shown in Fig. 6 can be sensitive. VEGAS can not help at all in the analysis of complex data measured with the modern technique, 3D-MEIS [31], where energy and angular distributions are simultaneously measured. In contrast, the shower approach is free of such limitations.

This paper shows several examples of the use of the program although, of course, it is not possible to cover all potential applications (e.g., simulations of the sputtering or

total reflection yields, possible in this approach, are also interesting applications of the code TRIC). The illustration examples in Sec. III are chosen to demonstrate the capabilities to solve specific problems and to show the accuracy of this method in comparison with others. In particular, we show the capability to simulate the interaction of different ions of low and medium energies with solid matter including complex structures, to calculate the yield of scattered ions and recoils, and to reproduce their energy spectra and angular distributions. In addition, we address the interpretation of the time-reversibility rule and provide additional insight into the origin of the surface peak.

APPENDIX

In this appendix, the expression [Eq. (3)] for the weights assigned to trajectory crossings in the MATCH approach is derived.

Let ω_1 represent the phase variables of an ion impinging on the sample when it reaches the volume of the considered lattice site. The probability that, in the course of its further motion, the ion will end up in the detector is obtained as

$$P_i(\omega_1) = \int d\omega_2 \frac{dP_{sc}}{d\omega_2} P_{\text{out}}. \qquad (A1)$$

The integration is performed over the phase variables of the ion after the collision ω_2 and $(dP_{sc}/d\omega_2)d\omega_2$ is the probability of scattering into one of the states within $d\omega_2$; this probability is related to the probability that the atom is located at the appropriate position. Finally, $P_{\text{out}}(\omega_2)$ is the probability that, in the course of its further motion, the ion in a state within $d\omega_2$ will leave the crystal with an energy and in a direction within the detector acceptance.

The probability $dP_{sc}/d\omega_2$ is explicitly determined in the case that both the scattering angle $\Theta(\mathbf{b})$ and the energy loss $\Delta E(\mathbf{b})$ in the binary ion-atom collision are uniquely determined by the impact parameter $\mathbf{b}$. To derive the corresponding expression, we describe the states ω_1 and ω_2 in a local coordinate system where the z axis is aligned with the ion velocity before the collision. As a result,

$$\frac{dP_{sc}}{d\omega_2} = \delta(y_2 - y_1)\delta[E_2 - E_1 + \Delta E(b)] \left| \frac{\partial^3 \mathbf{R}}{\partial x_2 \partial^2 \mathbf{n}_2} \right| D(\mathbf{R}), \qquad (A2)$$

where x and y are the coordinates in the scattering plane and in the perpendicular direction, respectively. The first δ function satisfies the condition that the two trajectories must intersect and the second takes into account the relation between the energies before and after the collision. In the case of potential

scattering, the state of ion motion after the collision (given by the coordinate in the scattering plane x_2 and the velocity direction $\mathbf{n}_2$) is uniquely determined by the atom position $\mathbf{R}$. The Jacobian of this relation appearing in (A2) is expressed as

$$\left| \frac{\partial^3 \mathbf{R}}{\partial x_2 \partial^2 \mathbf{n}_2} \right| = \frac{b}{\sin^2 \Theta} \left| \frac{db}{d\Theta} \right| = \frac{1}{\sin \Theta} \frac{d\sigma}{d\Omega}, \qquad (A3)$$

where b is the impact parameter corresponding to the scattering angle Θ and $d\sigma/d\Omega$ is the scattering cross section. Finally, $D(\mathbf{R})$ in (A2) is the density of distribution of thermal displacements of the target atom. Note that $\mathbf{R}$, the atom position which results in the considered collision, is a function of ω_1 and ω_2.

To confirm the validity of Eqs. (A2) and (A3), let us calculate the angular dependence of the probability of scattering on a single atom (the variation due to the uncertainty of the atom position $\mathbf{R}$). First, we integrate both sides of Eq. (A2) over y_2 and E_2; this cancels the two δ functions. The coordinate x_2 is related to the coordinate z_c of the point of crossing of the scattering asymptotes in such a way that $dx_2 = \sin \Theta \, dz_c$. In addition, we take into account that, for a given scattering angle, z_c is directly related to the coordinate z of the atom: $dz_c = dz$. As a result, we arrive at the familiar expression

$$\frac{d^3 P_{sc}}{dz \, d^2 \mathbf{n}_2} = \frac{d\sigma}{d\Omega} D(\mathbf{R}), \qquad (A4)$$

which expresses nothing but the concept of differential cross section $d\sigma/d\Omega$.

The contribution of scattering from the considered atom to the yield at the detector is obtained as an integral of the probability (A1) multiplied by the flux $\Phi_{in}(\omega_1)$ of ions of the beam reaching this lattice site:

$$Y_i = \int d\omega_1 \Phi_{in}(\omega_1) P_i(\omega_1). \qquad (A5)$$

Furthermore, the variables ω_1 and ω_2 are uniquely related to ω_{in} and ω_{out}, respectively, the parameters of motion of the ion when it enters (exits) the sample volume. It is useful to refer directly to the latter parameters and, for this purpose, we replace the integrations over ω_1 and ω_2 in (A1) and (A5) by integrations over ω_{in} and ω_{out}, respectively. In this case, the Jacobian $J_2 = |d\omega_2/d\omega_{\text{out}}|$ must be included in the integrand, while the flux $\Phi_{\text{in}}(\omega_1)$ is replaced by the flux of ions in the beam $\Phi_b(\omega_{\text{in}})$ multiplied by the Jacobian $J_1 = |d\omega_1/d\omega_{\text{in}}|$. Note that the relation $\omega_1(\omega_{\text{in}})$ can not be defined for all ω_{in} (some initial conditions ω_{in} result in trajectories that never pass through the vicinity of the considered lattice site). The same is possible for the pair of variables ω_2 and ω_{out}. To account for this, we assume the value of the respective Jacobian in such cases to be zero. Now, combining the above equations, we arrive at the expression

$$Y_i = \int d\omega_{\text{out}} J_{\text{out}} P_{\text{out}} \int d\omega_{in} J_{in} \Phi_b \delta(y_2 - y_1)$$
$$\times \delta[E_2 - E_1 + \Delta E(b)] \frac{1}{\sin \Theta} \frac{d\sigma}{d\Omega} D(\mathbf{R}). \qquad (A6)$$

As a function of ω_{out}, the probability P_{out} is easily evaluated: $P_{\text{out}} = 1$ if ω_{out} is within the region of acceptance of the detector, otherwise $P_{\text{out}} = 0$.

One can interpret the integration over ω_{out} in (A6) as a projection of the outgoing flux on the set $|d\rangle$ of states determined by the detector acceptance; the "density of states" is here uniform. Denoting also the "state of beam" (the distribution over ω_{in} in the beam) as $|b\rangle$, we can formally write Eq. (A6) as

$$Y_i = \langle d|\mathcal{T}_2 \mathcal{S}_i \mathcal{T}_1|b\rangle, \qquad (A7)$$

where the operators of transformation of the fluxes $\mathcal{T}_1$ and $\mathcal{T}_2$ are represented as the Jacobians J_1 and J_2, respectively and the rest of the integrand in Eq. (A6) represents the operator of scattering on the atom $\mathcal{S}_i$. This interpretation shows how the integral Eq. (A6) can be evaluated using the Monte Carlo method. The distribution over ω_{in} for the impinging ions must be taken according to the beam profile. The transformation $\mathcal{T}_1|b\rangle$ is obtained by calculating the trajectories of the incoming ions. The detection profile $|d\rangle$ must be taken as a uniform distribution within the region of phase space restricted by the unit cell at the sample surface S_0, by the solid angle of detector acceptance $\Delta\Omega$, and by the considered energy range ΔE. The integral over ω_{out} is associated then with a Monte Carlo sum

$$\int d\omega_{\text{out}} \Rightarrow \frac{S_0 \Delta\Omega \Delta E}{N_{\text{out}}} \sum_{k=1}^{N_{\text{out}}}, \qquad (A8)$$

where N_{out} is the number of considered outgoing trajectories. The set of outgoing trajectories calculated in time-reversed mode represents the action of the operator inverse to $\mathcal{T}_2$. The fluxes of ingoing and outgoing ions are convoluted by the matching of crossing trajectories and the terms of the Monte Carlo sum (A8) are determined as the values of the integrand in Eq. (A6). To avoid the problems due to the presence of δ functions in the integrand, they are replaced by normalized pulse functions of finite width:

$$\delta(t) \Rightarrow \Pi(t) = \frac{1}{\tau} \begin{cases} 1 \text{ if } |t/\tau| < 1/2, \\ 0 \text{ if } |t/\tau| > 1/2. \end{cases} \qquad (A9)$$

This results immediately in the expression Eq. (3) of Sec. IV determining the terms w of the sum (A8), the weights of crossing of trajectories. The tolerances τ_y and τ_E must be chosen to provide sufficient accuracy of the simulation results.

Note that in Ref. [11], where this approach was proposed, the weight w was simply taken as the product of the atomic density $D(\mathbf{R})$ with the cross section $d\sigma/d\Omega$. The additional $\sin\Theta$ in the denominator of Eq. (3) accounts for the fact that the density of crossings of two sets of parallel trajectories is inversely proportional to $\sin\Theta$, merely a simple geometrical feature. Additionally, the first fractional factor provides the correct normalization of the fluxes.

[1] W. Ekstein, *Computer Simulation of Ion-Solid Interactions* (Springer, Berlin, 1991).

[2] M. T. Robinson and O. S. Oen, Phys. Rev. **132**, 2385 (1963).

[3] J. H. Barrett, Phys. Rev. B **3**, 1527 (1971).

[4] M. T. Robinson and I. M. Torrens, Phys. Rev. B **9**, 5008 (1974).

[5] S. A. Karamian, J. S. Forster, J. U. Andersen, W. Assmann, C. Broude, J. Chevallier, J. S. Geiger, F. Grüner, V. A. Khodyrev, F. Malaguti, and A. Uguzzoni, Eur. Phys. J. A **17**, 49 (2003).

[6] P. J. M. Smulders and D. O. Boerma, Nucl. Instrum. Methods Phys. Res., Sect. B **29**, 471 (1987).

[7] M. Posselt, B. Schmidt, C. S. Murthy, T. Feudel, and K. Suzuki, J. Electrochem. Soc. **144**, 1496 (1999); M. Posselt, B. Schmidt, T. Feudel, and N. Strecker, Materials Science and Engineering B **71**, 128 (2000).

[8] V. A. Khodyrev, V. Ya. Chumanov, K. K. Bourdelle, and G. P. Pokhil, Nucl. Instrum. Methods Phys. Res., Sect. B **94**, 523 (1994).

[9] R. M. Tromp and J. F. van der Veen, Surf. Sci. **133**, 159 (1983); J. W. M. Frenken, R. M. Tromp, and J. F. van der Veen, Nucl. Instrum. Methods Phys. Res., Sect. B **17**, 334 (1986).

[10] J. Lindhard, Mat. Fys. Medd. Dan. Vid. Selsk. **34**, 14 (1965).

[11] G. Dorenbos, M. Breeman, and D. O. Boerma, Nucl. Instrum. Methods Phys. Res., Sect. B **108**, 173 (1996); M. H. Langelaar, M. Breeman, A. V. Mijiritskii, and D. O. Boerma, *ibid.* **132**, 578 (1997).

[12] This statement directly follows from the proposition in probability theory known as the law of total expectation; see P. Billingsley, *Probability and Measure* (Wiley, New York, 1995).

[13] J. P. Lepage, J. Comput. Phys. **27**, 192 (1978).

[14] W. H. Press and G. R. Farrar, Computers in Physics **4**, 190 (1990).

[15] M. M. Jakas, Radiat. Eff. Defects Solids **141**, 23 (1997).

[16] J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Range of Ions in Solids* (Pergamon, New York, 1985).

[17] C. Tschalär, Phys. Rev. B **1**, 2863 (1970).

[18] S. Y. Grachev, J. M. Gallego, D. Écija, D. O. Boerma, R. Gonzalez-Arrabal, and R. Miranda, Nucl. Instrum. Methods Phys. Res., Sect. B **219**, 593 (2004).

[19] E. S. Mashkova, V. A. Molchanov, E. S. Parilis, and N. Yu. Turaev, Phys. Lett. **18**, 7 (1965); E. S. Mashkova and V. A. Molchanov, Radiat. Eff. Defects Solids **13**, 183 (1972).

[20] M. Draxler, R. Beikler, E. Taglauer, K. Schmid, R. Gruber, S. N. Ermolov, and P. Bauer, Phys. Rev. A **68**, 022901 (2003).

[21] T. Kobayashi, Nucl. Instrum. Methods Phys. Res., Sect. B **249**, 266 (2006).

[22] T. J. Wood, C. Bonet, T. C. Q. Noakes, P. Bailey, and S. P. Tear, Surf. Sci. **598**, 120 (2005).

[23] E. Rauhala, N. P. Barradas, S. Fazinic, M. Mayer, E. Szilágyi, and M. Thompson, Nucl. Instrum. Methods Phys. Res., Sect. B **244**, 436 (2006).

[24] J. P. Biersack, E. Steinbauer, and P. Bauer, Nucl. Instrum. Methods Phys. Res., Sect. B **61**, 77 (1991).

[25] P. Bauer, E. Steinbauer, and J. P. Biersack, Nucl. Instrum. Methods Phys. Res., Sect. B **64**, 711 (1992).

[26] W. K. Chu, J. W. Mayer, and M. A. Nicolet, *Backscattering Spectrometry* (Academic, New York, 1978).

[27] E. I. Sirotinin, A. F. Tulinov, V. A. Khodyrev, and V. N. Mizgulin, Nucl. Instrum. Methods Phys. Res., Sect. B **4**, 337 (1984).

[28] Z. Smit, Phys. Rev. A **48**, 2070 (1993).

[29] E. Bøgh and J. L. Whitton, Phys. Rev. Lett. **19**, 553 (1967).

[30] M. A. Muñoz-Márquez, G. S. Parkinson, D. P. Woodruff, A. Hentz, P. L. Grande, G. Schiwietz, T. J. Wood, C. Bonet, S. P. Tear, P. Bailey, and T. C. Q. Noakes, Phys. Rev. B **72**, 075415 (2005).

[31] T. Kobayashi, Phys. Rev. B **75**, 125401 (2007).