Multiple scattering of MeV ions: Comparison between the analytical theory and Monte-Carlo and molecular dynamics simulations

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Abstract

Angular and energy distributions due to multiple small angle scattering were calculated with different models, namely from the analytical Szilágyi theory, the Monte Carlo code MCERD in binary collision approximation, and the molecular dynamics code MDRANGE, for 2 MeV ^{4}He in Au at backscattering geometry and for 20 MeV ^{127}I recoil analysis of carbon. The widths and detailed shapes of the distributions are compared, and reasons for deviations between the different models are discussed.

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1 Introduction

The trajectories of MeV ions in matter are usually approximated by straight lines. This is, however, only a crude approximation due to many small angle deflections by collisions with large impact parameters. This has been called multiple scattering. The combined effect of these collisions results in angular and energy straggling, leading to a degradation of the achievable depth resolution. An analytical theory describing these spreads was developed by Szilágyi et al. [1,2,3,4], and is employed in several widely used computer simulation codes for ion beam analysis, such as DEPTH [1], SIMNRA [5,6], and WiNDF [7].

However, as experimental data for multiple scattering distributions at typical IBA geometries are almost non-existent, the analytical theory was never seriously validated. Monte-Carlo and molecular dynamics simulations include multiple scattering naturally, but require much longer computing times. Additional energy broadening effects, such as energy loss straggling or surface roughness effects, can be switched off in simulations, thus allowing an even clearer comparison than with experimental data.

In this work we compare predictions of the analytical theory with Monte-Carlo and molecular dynamics simulations for typical RBS and ERDA applications.

2 Analytical theory and computer codes

2.1 Analytical multiple scattering theory

Angular spread distributions of ions in mono-elemental targets without stopping were calculated analytically by Sigmund and Winterbon (SW) [8]. The SW theory was expanded to multi-elemental targets with stopping by Szilágyi *et al.* [1] and Amsel *et al.* [3]. Energy spread distributions are derived from angular and lateral spread distributions, taking the correlation between angular and energy spread into account [3, Section 4].

2.2 DEPTH and SIMNRA codes

The analytical Szilágyi theory is implemented in the DEPTH code [9], which is also used by WiNDF [7].

SIMNRA is a simulation program for RBS, ERD and NRA energy spectra

[5,6,10], and contains an independent implementation of the Szilágyi theory [1] .

SIMNRA and WiNDF utilize only the widths of the multiple scattering energy distributions. Their shape is approximated by Gaussian functions.

2.3 MCERD Monte-Carlo code

MCERD is a TRIM-like Monte Carlo (MC) code in binary collision approximation [11,12]. ZBL electronic stopping powers [13] and the universal interaction potential were used. Electronic energy loss straggling was switched off.

2.4 MDRANGE molecular dynamics code

MDRANGE is a molecular dynamics (MD) code for calculation of ion ranges and deposited energies [14,15]. ZBL electronic stopping powers [13] were used. Electronic energy loss straggling was switched off. The targets were assumed to be

- Single crystalline gold at random orientation. Ions channeled for distances larger than 100 nm were rejected, corresponding approximately to polycrystalline material with a maximum grain size of 100 nm.
- Two different kinds of amorphous carbon: a tetrahedral amorphous network obtained from density-functional theory calculations [16], and a placement of atoms in 3 dimensions which is completely random, except that the minimum separation between the atoms is set to slightly less than the covalent bond distance.

3 Results and discussion

3.1 Backscattering

Fig. 1 shows the widths of the angular and energy spread distributions of 2 MeV ^{4}He penetrating through Au. As already pointed out in [1,3], angular and energy distributions are non-Gaussian. The distributions obtained from MCERD and MDRANGE were fitted with Pearson VII distributions in order

[1] Previous versions of SIMNRA treated the correlation between energy and angular spread only approximately, the complete Szilágyi theory is implemented in SIMNRA 5.76 and higher.

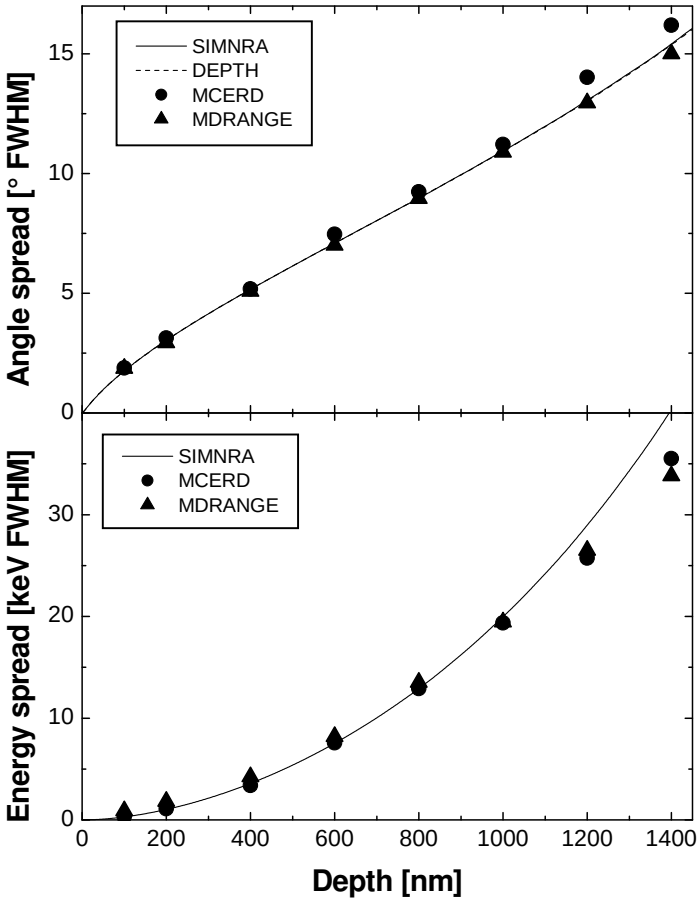

Fig. 1. Penetration of 2 MeV ^{4}He through Au at an incident angle of 15° (to surface normal). Top: FWHM of the angular spread distribution as a function of sample depth. The results from the DEPTH and SIMNRA codes are almost identical and cannot be distinguished. Bottom: FWHM of the multiple scattering induced energy spread distribution as a function of sample depth.

to obtain the full width at half maximum (FWHM). The distributions get asymmetric at larger depths (see Section 3.3), and the approximation with Pearson VII distributions gets increasingly inaccurate. Angular distributions are generally more symmetric than energy distributions.

The widths of the distributions from the analytical theory agree with the MC and MD results within the error bars of the fit until a depth of about 1000 nm. At that depth the incident particles have lost 35% of their initial energy.

The multiple scattering induced energy spread at the target surface is shown in Fig. 2. While Fig. 1 compares angular end energy spread on incident or exit paths, distributions at the surface also take the correlation between angular and energy spread into account. Analytical theory and MC agree within about 2 keV until a depth of about 600 nm, at which the particle energy at the surface has decreased to about 1000 keV. At larger depths the disagreement increases

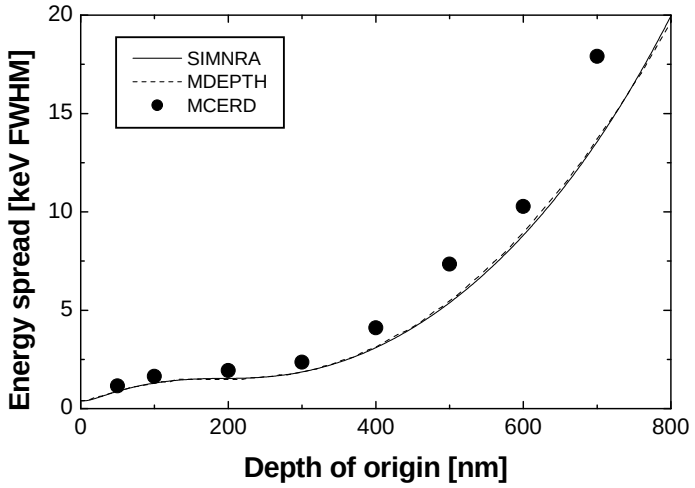

Fig. 2. FWHM of the energy spread distribution at the target surface as a function of the depth of origin. 2 MeV ^{4}He in Au, incident angle 15°, exit angle 15°, scattering angle 150°. The results from the DEPTH and SIMNRA codes are almost identical and can hardly be distinguished.

due to progressing asymmetry of the energy distributions (see Section 3.3).

We also found, that for multi-elemental targets (^{4}He and ^{127}I in WC) the same level of agreement between analytical theory and Monte-Carlo simulation is obtained as already shown in Figs. 1 and 2.

3.2 Recoil detection

Fig. 3 (top) shows the FWHM of the angular spread distribution of 20 MeV ^{127}I penetrating through C. The small difference between SIMNRA and DEPTH is due to the use of slightly different stopping powers. The agreement with the MC and MD results is good. Fig. 3 (bottom) compares the widths of the energy spread distributions. Two different results are shown for MCERD: With energy transfer to target atoms (i.e. with nuclear stopping) and without. Energy transfer to target atoms is not included in the analytical theory, see Section 3.3. As can be seen in Fig. 3 (bottom), the FWHM of the energy spread distribution for depths smaller than 300 nm is only slightly changed by including energy transfer to target atoms. The agreement between the Monte Carlo result and the analytical Szilágyi theory is excellent close to the surface, while at larger depths the energy spread is overestimated by the analytical theory. The two different amorphous carbon MD models were identical within the error bars. But we also observed, that good agreement with MD is obtained only for amorphous carbon: For crystalline carbon the distributions differ markedly due to channeling effects, which play an important role at these low energies.

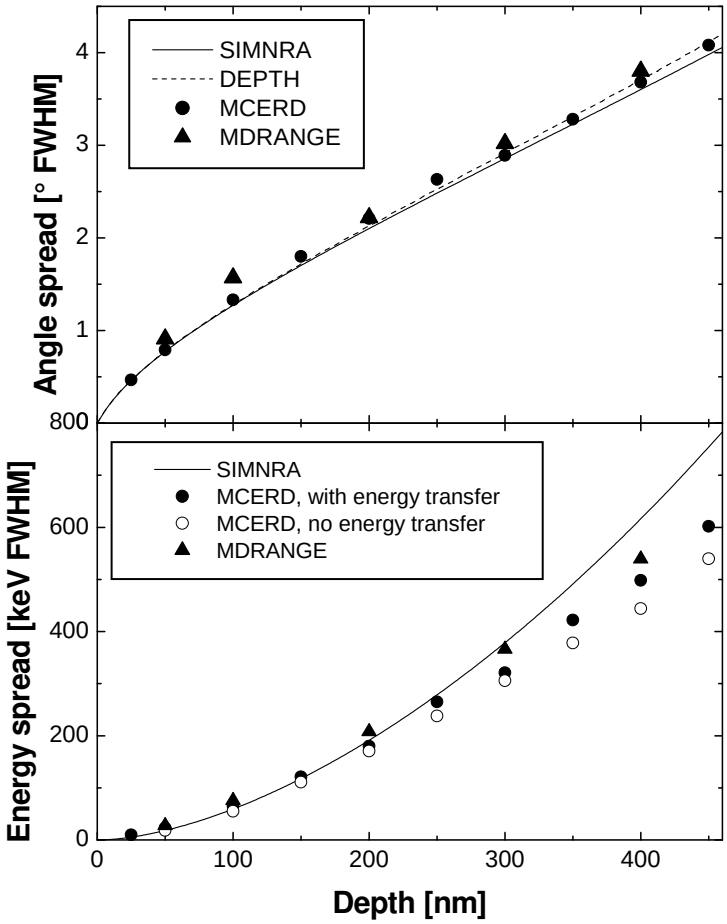

Fig. 3. Penetration of 20 MeV ^{127}I through C at an incident angle of 70° (to surface normal). Top: FWHM of the angular spread distribution as a function of sample depth. Bottom: FWHM of the multiple scattering induced energy spread distribution as a function of sample depth.

The energy spread of ^{12}C recoils at the target surface is shown in Fig. 4. The overall shape of the depth-dependent energy spread is well reproduced by the analytical theory. Close to the surface the agreement between the Monte Carlo result and the analytical theory is excellent. At depths between 100 nm and 300 nm the analytical theory deviates from the Monte Carlo results, but less than 15%. At larger depths the angular spread distributions get so broad, that the analytical theory is not valid any more. The break-down of the theory is handled differently by DEPTH and SIMNRA, resulting in different results at depths larger than 400 nm.

3.3 Limitations of the analytical theory

The analytical Szilágyi theory makes the following approximations:

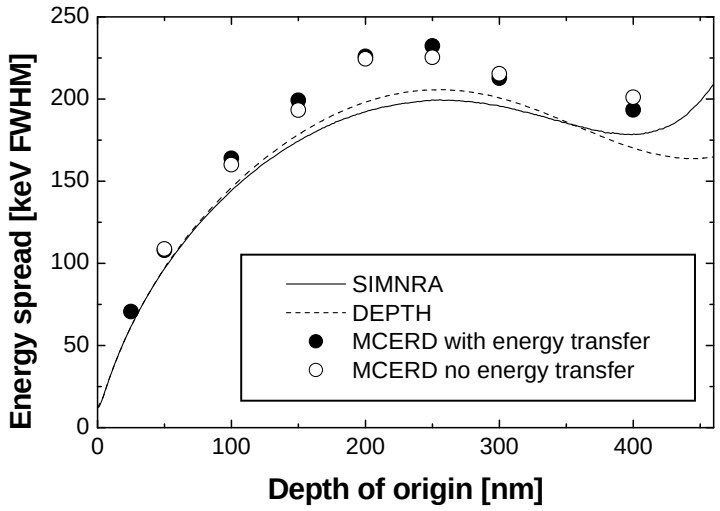

Fig. 4. FWHM of the energy spread distribution of ^{12}C recoils at the target surface as a function of the depth of origin. 20 MeV ^{127}I in C, incident angle 70°, exit angle 70°, scattering angle 40°.

(1) Symmetry of angular and energy distributions around a mean value.
(2) Angular independence of the scattering cross section during the main scattering event. The main scattering event is scattering with large scattering angle in RBS, or recoil creation in ERDA.
(3) No energy transfer to target atoms during small angle deflections.

The first two approximations are valid as long as the width of the angular spread distributions is not too large. The approximation of symmetric energy distributions is most strongly violated at grazing incidence or exit angles, and at normal incidence: At normal incidence deflections to either side lead to lower, but never higher, energies, resulting in asymmetric energy distributions. The angular dependence of the scattering cross section during the main scattering event plays especially a role in forward scattering due to the strong angular dependence of the forward scattering cross section, and may be also important at small recoil angles.

Energy transfer to target atoms is the source of nuclear stopping, and fluctuations in this energy transfer are called nuclear straggling. The variance of the nuclear straggling distribution can be calculated for realistic potentials [17], but this result has little practical relevance: As was already pointed out by Schmelmer *et al.* [18] and was shown recently by Glazov and Sigmund [19], the nuclear straggling distributions are asymmetric and strongly non-Gaussian with long low-energy tails. These tails lead to large variances of the distributions, but change the FWHM only slightly [18]. This can be seen in Figs. 3 and 4 by comparing the MCERD results with energy transfer to target atoms (i.e. with nuclear stopping and straggling) and without it. The FWHM is only slightly changed. Nevertheless, despite the small influence on the FWHM's,

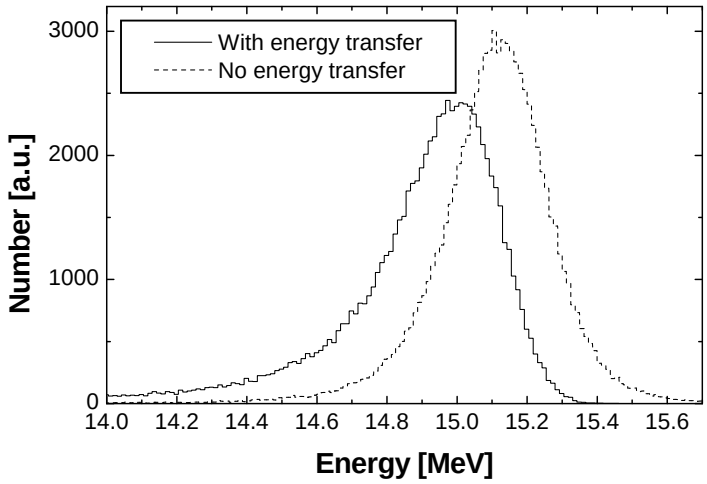

Fig. 5. Energy distributions of 20 MeV ^{127}I in a depth of 300 nm C. Calculations with and without energy transfer to target atoms, incident angle 70°. Both curves are normalized to the same area.

nuclear straggling changes the mean and the shape of the energy distribution considerably. This is shown in Fig. 5 for 20 MeV ^{127}I in C at a depth of 300 nm. The dashed line shows the energy distribution without nuclear energy loss: The distribution is almost symmetric. The FWHM of the angular spread is about 3° in this depth (see Fig. 3), and the small asymmetry is due to path length differences between particles deflected towards the surface and away from the surface. With nuclear energy loss (solid line) the particle energies are lower, and the distribution gets more strongly asymmetric with a long tail towards lower energies. The shapes of these asymmetric distributions are difficult to describe analytically.

4 Conclusions

Angular and energy distributions due to multiple small-angle scattering, as calculated from the analytical Szilágyi theory by the DEPTH and SIMNRA code, Monte Carlo (MC) simulation in binary collision approximation using the MCERD code, and molecular dynamics (MD) using MDRANGE, were compared for typical light ion backscattering and heavy ion recoil detection analysis applications. The agreement between MC and MD is good. The widths (characterized by their FWHM) of the angular distributions are accurately described by the analytical theory until large depths. For not too large depths, the analytical theory also predicts the FWHM of the energy distributions accurately, while at larger depths deviations up to 20% are observed. The energy distributions are asymmetric with long tails towards lower energies due to energy transfer to target atoms, i.e. nuclear straggling. The tails and asymme-

tries are not correctly described by the analytical theory, but they change the FWHM of the distributions only slightly. These limitations of the analytical theory have to be kept in mind when using simulation codes which rely on it, such as DEPTH, WiNDF and SIMNRA. If necessary, a more accurate description can be obtained from Monte Carlo simulations, at the cost of longer computing times.

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