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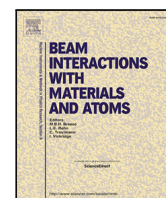
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Full length article

Elastic recoil and scattering yields measured in low energy heavy ion ERD

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ABSTRACT

Experimental yields of low energy recoils and scattered beams in ToF-ERD have been measured and compared against theory. A significant discrepancy between Rutherford or Andersen cross-section predicted yield vs the experimental results is now demonstrated. Scale of the discrepancy is up to 50% compared to yields predicted by theory for low energy Au recoils. MCERD simulations were used to study the carbon foil scattering in the timing detectors to explain the observed discrepancy. Simulations indicate that a major part of the discrepancy occurs due to the scattering of low energy, heavy mass particles in the timing detector foils. The yield discrepancy can be narrowed down by taking into account the reduction of recoil yields caused by the carbon foil scattering. Further studies are in progress to study carbon foil scattering, aiming to further improve the quantitativity of ToF-ERD for the heavy elements.

1. Introduction

Elastic Recoil Detection (ERD) as an analytical method was developed by L'Ecuyer [1] in the 1970s. ERD made it possible to detect light elements down to hydrogen and its isotopes, not just heavier elements on a light substrate like in Rutherford backscattering spectrometry (RBS).

Time-of-Flight ERD [2–4] gained some popularity immediately at the larger, 5 MV or larger tandem laboratories [5–8] during the 1990s but then experienced a slower popularity period after that. New ToF-ERD spectrometers have become operational more frequently during the recent decade and today the ToF-ERD method with low energy incident beams (<20 MeV) is used regularly in several laboratories for thin film analysis [9–14].

Low energy ToF-ERD offers benefits that have increased the popularity of the method in the past decade. Low energy heavy ion beam offers increased recoil and scattering cross-sections and increased stopping force of the sample, thus providing a better signal of the thin film to substrate-ratio [11,15]. Increased cross-sections provide also faster data collection, which limits the beam fluence on the target and offers an important possibility to decrease the irradiation damage [11,15,16]. At the same time, low energy ion beams allow the increase of the detector solid angle due to shorter required time-of-flight detectors for

the same timing resolution [17]. Additionally, infrastructure costs are cut down as reduced beam energies allow the use of small ≤ 3 MV accelerators for the ToF-ERD.

However, the use of low energy heavy ion beams does not come without drawbacks. One of the drawbacks is decreased mass resolution at low energy incident ion beams. This resolution decrease, however, can be compensated by using better gas ionization detectors as energy detectors [10,18]. More importantly, for heavy target elements, there is a discrepancy between the measured yields compared to the theoretical predictions or if low energy heavy recoil ToF-ERD data is compared against other methods like RBS. Different explanations for this discrepancy exist. One contributing factor for the discrepancy in the low energy heavy recoils is their known tendency to experience more multiple scattering in a sample and in the detector foils. It is also debated if this discrepancy is caused by incorrect theoretical cross-section models or only due to the scattering happening in the sample or in the carbon foils in the ToF-detector.

In ERD analysis recoil cross-sections used are based on Rutherford cross-sections. Typically Rutherford cross-sections are modified using for example Andersen screening correction [19]. It is also debated [20] whether known screening correction models for Rutherford cross-sections apply for recoils [11,15], as originally only scattered

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beam data of light beam species was used to verify the Andersen correction. On the other hand, Kim et al. [21] have studied ERD cross-sections using inverse kinematics experiments, and their measured cross-sections for elements in Li – F range agree with Rutherford cross-sections.

Carbon foil scattering is a known issue in low energy ToF-ERD [11, 17], but has been usually neglected as a minor effect in the analysis. There are a few papers about low energy heavy ion carbon foil scattering, mostly in keV range. In Refs. [22–25] foil scattering is measured and results are compared to the plural scattering model by Meyer [26]. Their results show clearly that scattering reduces detected yields in this type of ToF applications and Meyer's theoretical scattering model seems to agree relatively well with their data. An extensive review by Amsel et al. [27] discusses the nature of multiple scattering and that can be applied to carbon foil scattering. Monte Carlo simulation tools, like MCERD [28] and Corteo [29], have been developed to model the multiple scattering in ion beam analysis applications. In MC simulations scattering distributions are based on the potential the code uses, in the case of MCERD universal potentials are used [30]. Although MCERD is a powerful analysis tool, in heavy recoil analysis simulated yields do not always match what has been measured. Simulations indicate that yields are enhanced in non-mirror geometry due to the multiple scattering in the sample film [31] and reduced due to timing detector foil scattering [11].

In a typical ToF-ERD analysis performed in the Jyväskylä accelerator laboratory, heavier-than-beam recoils are generally not considered reliable, and heavy elements are analyzed using forward scattering of the Heavy ion ERD (HIERD) incident ion beam. The issue with low energy heavy recoils in ToF-ERD is nowadays more profound as modern heavy materials, like CdTe, InSb, and GaAs, are becoming more important to the thin film industry and novel heavy element semiconductor materials are being developed. Quantification of heavy films becomes an issue due to the discrepancy of detected yields as heavier materials induce more multiple scattering in the sample and scattering in the detector. Although other methods like RBS can be used for heavy element thin film characterization for light substrates, the benefit of the ToF-ERD is that independently from the substrate, it can probe all elements, including hydrogen, simultaneously.

In the present work, discrepancies between theoretical, simulated, and experimental ERD yields of heavy elements are studied. Focus is set to study the discrepancy from the point of view of cross-sections and carbon foil scattering. Stopping force effects of the yields are not considered here, although taken into account as a potential source of uncertainty, and samples are chosen so that energy losses are minimized in the films of interest. Multiple scattering processes in the sample and their contributions to yield discrepancy have been referenced by earlier publications from the field.

2. Experimental setup and methods

Series of ToF-ERD measurements were done using University of Jyväskylä [12] and University of Helsinki [32] ToF-ERD setups. Several different incident beams (^{35}Cl , ^{63}Cu , ^{79}Br and ^{127}I) were used with energy range of 6.8–17 MeV in Jyväskylä and 10–50 MeV in Helsinki. Thus two independent ToF-ERD measurement systems were used for validation and to extend the incident ions' energy range. A basic schematic of ToF-ERD setup and geometry is presented in Fig. 1. Timing detector carbon foil thicknesses in Jyväskylä ToF-detector are 3 $\mu\text{g}/\text{cm}^2$ and 10 $\mu\text{g}/\text{cm}^2$ and in Helsinki 3 $\mu\text{g}/\text{cm}^2$ and 9 $\mu\text{g}/\text{cm}^2$ for first and second timing detector respectively. Additionally Jyväskylä ToF-detector carbon foils are coated with 1 nm atomic layer deposited Al_2O_3 .

Jyväskylä ToF-ERD setup uses 1.7 MV NEC Pelletron accelerator and NEC MC-SNICS ion source. Energy calibration was done using 3038 keV $^{16}\text{O}(\alpha, \alpha)^{16}\text{O}$ resonance [33]. Scattering angle was determined using laser alignment and alignment telescopes. Scattering angle is

$(40.6 \pm 0.2)^\circ$. All measurements were done in mirror geometry, in Fig. 1 angles α and β are equal at 69.7° .

Measurements done in Helsinki use TAMIA 5 MV EGP-10-II tandem accelerator and MC-SNICS ion source. Beam energy of the Helsinki setup has been calibrated using proton beam resonances of multiple elements. Scattering angle was calibrated using optical telescopes and scattering angle is $(39.8 \pm 0.5)^\circ$. Like in Jyväskylä measurements, the sample was set to mirror geometry. The purpose of high energy ERD measurements in Helsinki was to collect data with overlapping energy region and extend the incident beam energy much higher than is currently available in Jyväskylä. Another reason was to minimize the screening effect at the higher incident ion beam energies.

Sample films were chosen to be monoisotopic metal elemental or metal oxide films. Film materials used were Co, Y_2O_3 and Au. Thicknesses of the films were 5, 2 and 5 nm respectively. Very thin films were chosen to minimize energy loss, straggling, stopping force effects, and multiple scattering in the film. Films were deposited on top of amorphous wet thermal grown 1 μm SiO_2 substrate on Si wafer supplied by University Wafer. Amorphous SiO_2 substrate was used for beam fluence normalization as well as to avoid channeling and to provide a uniform matrix for both light, O, and medium mass, Si, element, which ratio is the same as the nominal ratio. Thus both Si or O recoils can be used for fluence normalization in the analysis also independently.

Co and Au films were deposited by sputtering at VTT Micronova facilities. Sample films were imaged with Atom Force Microscopy (AFM) using Bruker Dimension Icon and SCANASYST-AIR tip. AFM verified that films are rough with RMS roughness of 3.50 nm and 1.85 nm respectively and RBS fitting is in agreement with these roughnesses. RBS was used to analyze not just a single sample from the deposited 150 mm wafers but also to confirm the required uniformity for samples selected for the ToF-ERD analysis. Co and Au films are not smooth on top of the substrate and the substrate is visible in between metal film grains. This is not seen to have any effect on the purpose of the study. Atomic layer deposited Y_2O_3 films [34] were smooth with RMS roughness of 0.25 nm. AFM images can be found in Fig. 2 and additionally from Appendix A.

From ToF-ERD data the recoil and scattering events were selected in Potku software [35]. In Potku software, ToF-E histograms, the selected coincidence events affected by multiple scattering were included. When compared against the simulations, the data from simulations also includes multiple scattering induced tails. Example 2D-histograms from both Jyväskylä and Helsinki laboratories are shown in Fig. 3. In Helsinki ToF-ERD setup recoil energy is measured with a silicon solid state detector (SSD), which are known for relatively poor energy resolution for heavy ions [36]. Jyväskylä measurement setup uses a gas ionization chamber detector for energy detection [18].

3. Results and discussion

Both recoil and scattering yields have been measured by the two mentioned ToF-ERD systems and compared against theoretical yields. To calculate theoretical yield information of sample film areal density is needed. Areal densities were determined with 2 MeV He-RBS beforehand and analyzed using SIMNRA 7.02 [37]. Detected yields are available in Appendix B and RBS example spectra in Appendix C. Beam fluence of the ToF-ERD analysis is determined by fitting it to the amorphous SiO_2 substrate energy spectrum.

In Section 1 there was discussion about the role of multiple scattering in the sample film in the context of yield enhancement. We assume that multiple scattering is net zero effect on yields and this is backed up by Ref. [31], when we work in mirror geometry. SIMNRA was used for ToF-ERD fluence fitting with Simnra's dual and multiple scattering options on.

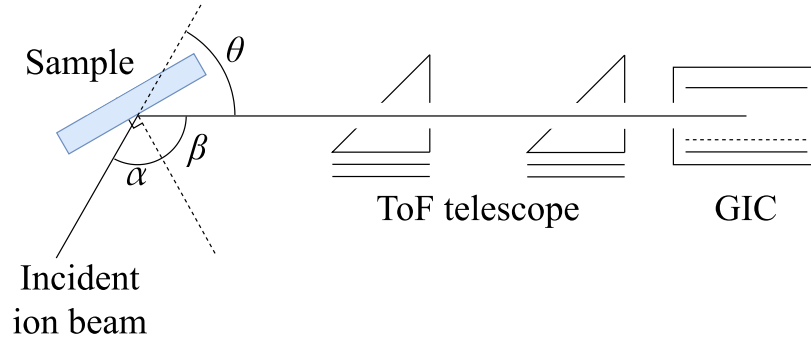


Fig. 1. ToF-ERD setup schematic. Scattering angle θ was calibrated in both systems before the measurements and in mirror geometry angles α and β were equal and measured from sample normal. Time-of-flight telescope and gas ionization chamber detector (GIC) were used in coincidence to detect particle velocity and energy that are used to determine the mass of a detected particle. ToF lengths in Jyväskylä and Helsinki are 623 and 684 mm respectively. In Helsinki, a silicon detector was used for the energy measurement.

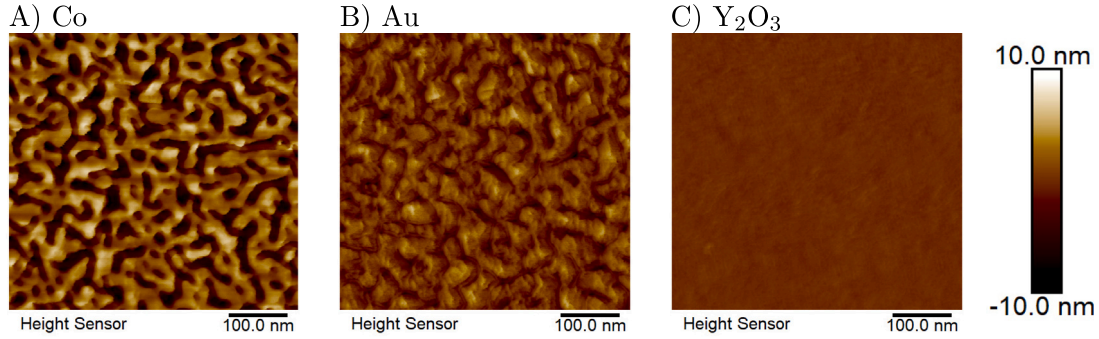


Fig. 2. AFM images of samples: (A) Co 5 nm, (B) Au 5 nm and (C) Y_2O_3 2 nm thin films. Sputtering deposited Co and Au films are rough and substrate can be seen in between film element grains. The Y_2O_3 film was deposited by ALD and the film was smooth and uniform.

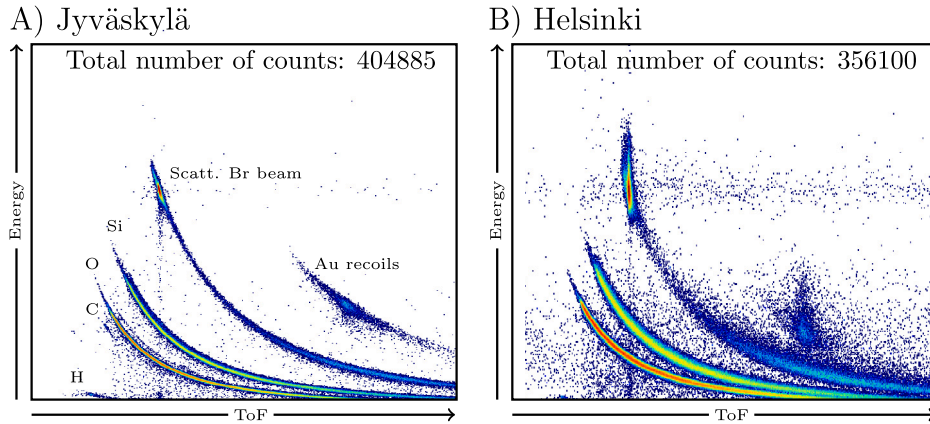


Fig. 3. Comparison between Jyväskylä and Helsinki ToF-ERD setups. Both measurements are from Au sample film using ^{79}Br beam with incident energy of 10.2/10.0 MeV in Jyväskylä/Helsinki measurements.

3.1. Measured ToF-ERD recoil and scattered particle yields

ERD yields of different elements are measured and compared to theory predictions of yields, shown in Fig. 4. Theoretical yields are calculated using Rutherford cross-sections, fitted number of incident particles times solid angle and experimental parameters of sample tilt angle, and sample film areal density, determined by RBS. Beam fluence is determined by fitting to SiO_2 signal using SIMNRA version 7.02 [37], and is discussed more in Section 3.2. As a side result, Si to O ratio was determined to be coherently 1:2 within less than 5% variation in all

measurements with different beam species and incident energies. Fig. 4 also includes theoretical yields for Andersen screened cross-sections for comparison.

Detected recoil yields deviate from the expected theoretical yields. Co, being the lightest of the sample recoils of interest, agrees somewhat with Andersen screened yields, but Y (in Y_2O_3 film) and Au yields show significant deviation from what the theory predicts. This is evident in both Jyväskylä and Helsinki results, so it is not a systematic feature of a single ToF-ERD setup. Also, similar discrepancies have been reported in Refs. [11,15].

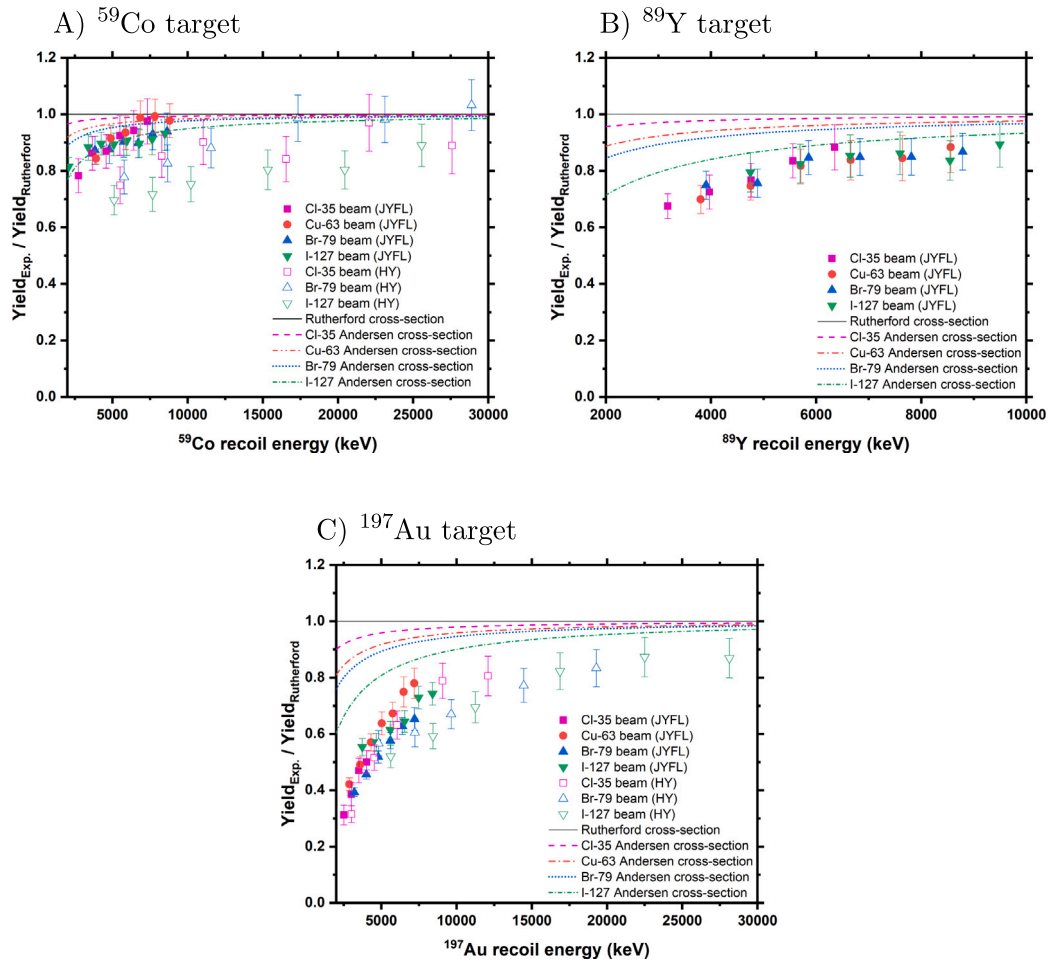


Fig. 4. Experimentally determined yields of (A) Co, (B) Y (Y_2O_3), and (C) Au recoils normalized with theoretical yields calculated with Rutherford cross-sections. Measurements are done in the University of Jyväskylä (JYFL) and University of Helsinki (HY) accelerator laboratories. Andersen screening correction factors are presented with dashed lines. With heavier recoils experimental results deviate strongly from theoretical predictions.

As the data in Fig. 4 are presented as a function of the energy of the recoiled particle, the measured relative yields seem to line up in every sample material measured within the error bars. Andersen screened cross-sections predict significantly different yields depending on the incident ion species, as seen in Fig. 4 dashed curves. Normalized recoil yields form a curve relative to their energy in the ToF-E system, which might indicate that the incident ion species do not play a major role and the discrepancy is more pronounced for the smaller energy and heavier species in the ToF-E telescope. Thus, recoils, and not the incident ion beam, mass and energy seem to be among the determining factors reducing the yield, indicating that this discrepancy is caused by multiple scattering in the sample or by the detector carbon foils.

Scattering yields were analyzed from the same measurement data as recoil yields. Scattering yield results are shown in Fig. 5. According to our data using forward scattering in thin film analysis works when the incident beam species is lighter than the target element. This has been also the common practice of the Jyväskylä analysis, as stated above in Section 1. Scattering of heavier-than-sample ion beam produced reduced yields compared to what would be expected from theory. As with the recoils, this observation suggests that sample or carbon foil multiple scattering is the main contributor to the yield discrepancy due to low energies of the heavy scattered particles.

For more evidence towards the detector-related origin of the discrepancy, yields of Au recoils and scattered Au beam with similar energy in the timing detectors has been measured. Experimental parameters were tuned so that in the ToF-detector Au particles with

5.7–5.8 MeV surface energy were obtained. Au recoils were measured with ^{63}Cu , ^{79}Br and ^{127}I incident beams, and ^{197}Au beam was scattered from 5 nm thick Ta sample film — Ta being again one more different parameter than in earlier presented cases. Measurement results are presented in Table 1. There seems to be very little, if any, difference between the recoiled or scattered 5.7–5.8 MeV Au particles at the detectors when ^{63}Cu recoiled Au measurement is considered as an outlier, since ^{63}Cu incident beam Y_E/Y_R has been taken from a different data set as others, and stands out from the rest of the measurements. Currently, there is no other explanation for the outlier, but as seen in Fig. 4C target more variation exists between different incident ion beams compared to the yttrium target in Fig. 4B. This result indicates that the main effect is not the multiple scattering in the sample or recoil cross-sections. The yield reduction phenomenon is related to the detectors, and results indicate that carbon foil scattering is the main contributing effect behind the yield discrepancy between the theory and the experimental data.

The 5.7–5.8 MeV Au experiment shows that yields are reduced independently from incident ion species or the target element. Typically used screening corrections would differ depending on the incident ion nuclear charge, but evidence of that is not found in our ToF-ERD data. This finding indicates that the leading correction is carbon foil scattering and HIERD cross-section corrections have less influence. The search for the discrepancy must be concentrated on the carbon foils and not on the cross-sections, which, according to Fig. 4 and Table 1 seems not to be the main suspect.

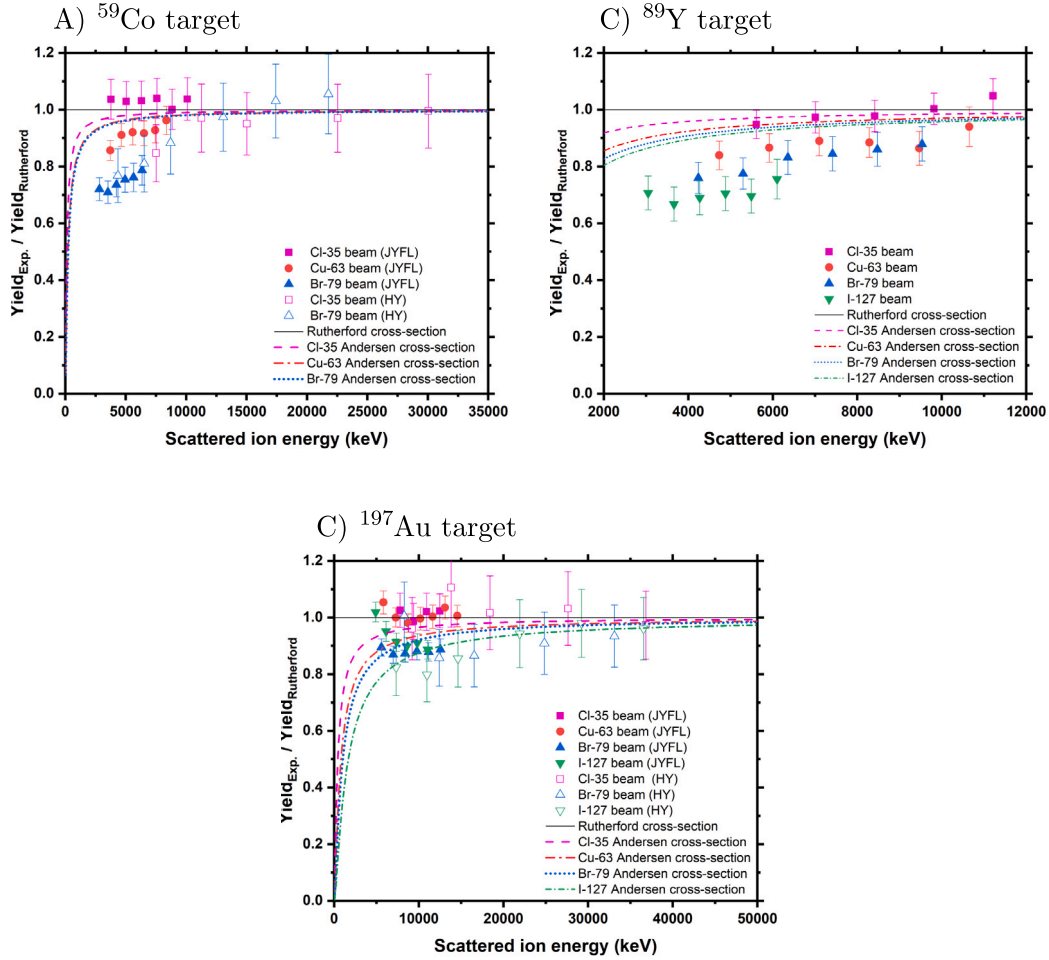


Fig. 5. Scattering yields from target films (A) Co, (B) Y (Y_2O_3), and (C) Au. Scattering yields were normalized with theoretical yields calculated with Rutherford cross-sections. Measurements are done in the University of Jyväskylä (JYFL) and University of Helsinki (HY) accelerator laboratories. Andersen screening correction factors are presented with dashed lines.

3.2. Fluence fitting and effect of stopping force

Beam fluence was determined by fitting it to experimental data. Fitting was done to the substrate signal and mainly to the first ~ 100 nm of it. This type of fluence normalization can be prone to errors caused by incorrect stopping values at film and substrate. It is possible to achieve a similar trend for Fig. 4 and Table 1 if the fluence or stopping force of the substrate, which was used for the normalization of the data, is determined wrong.

The significance of the stopping effect was investigated using ToF-ERD on 4 nm TaN on top of 100 nm SiO_2 layer on a Si substrate sample. 100 nm SiO_2 is easily penetrated by the low energy beam so full layer thickness of the SiO_2 was detected in the stopping force measurements. More importantly, the oxygen signal lies on the clean background, and thus all yielded O counts are affected by the potential changes or uncertainties in the stopping force of the SiO_2 substrate, as seen in Fig. 6A. Selected energies available in the Jyväskylä accelerator laboratory are compared to each other and two different beams that are important for this experiment. Measurements were done in three sessions and the sample was every time loaded in again and set at the same angle each time. In Fig. 6B analyzed SiO_2 areal density of the same sample results in a maximum of 2.5% discrepancy between measurements in the surface stopping force range from 470–700 eV/(10^{15} at./cm 2). Thus normalization to the bulk-like SiO_2 substrate is sufficient and the results between different energies are not affected by variations in the stopping force for SiO_2 . Thus maximum stopping force related uncertainty for the recoil yields with used beams and energy range is

Table 1

Measured yields Y_E of 5.7–5.8 MeV Au particles from different target-ion beam combinations, normalized with the Rutherford yields Y_R . The same energy and mass in the ToF-detector results in a similar yield ratio for all beam/target pairs. ^{63}Cu , ^{79}Br , and ^{127}I beams were used to recoil Au target atoms, and the Au beam was scattered by 5 nm Ta film. The average yield ratio of all data points is 0.56.

^{197}Au	Detected energy [MeV]	Y_E/Y_R
Recoiled by		
^{63}Cu	5.76	0.67 ± 0.04^a
^{79}Br	5.72	0.54 ± 0.06
^{127}I	5.83	0.51 ± 0.03
Scattered from		
^{181}Ta	5.84	0.54 ± 0.04

^a ^{63}Cu , experimental data is considered as a minor outlier in this set.

within 2.5%, while the largest differences are more than ten times to Andersen corrected cross-sections (Fig. 4).

3.3. Carbon foil scattering in simulations

The effect of the scattering from the carbon foils in the ToF telescope was simulated using MCERD [28]. Simulations were done with and without carbon foils, supplied by The Arizona Carbon Foil Co., with a thin 1 nm Al_2O_3 layer as present in the real detection setup in Jyväskylä. Carbon foil thicknesses used in the simulations correspond

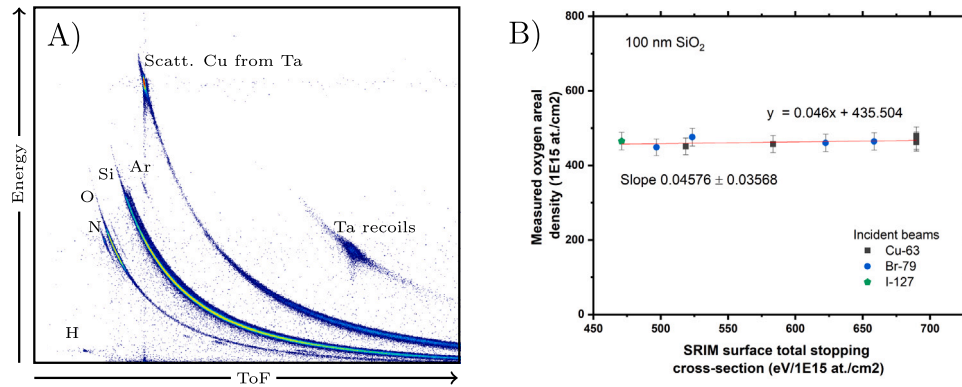


Fig. 6. (A) Example 2D-histogram from stopping cross-section measurements using 13.6 MeV ^{63}Cu beam and TaN/SiO₂/Si target. (B) Measured O areal densities in 100 nm SiO₂ film are plotted in function of surface stopping force by SRIM. Measurements were done with multiple beam species and energies: 8.5–13.6 MeV ^{63}Cu , 8.5–13.6 MeV ^{79}Br , and 10.75 MeV ^{127}I . Oxygen areal density used to normalize the fluence changes maximum of 2.5% in this stopping force range of multiple energies and ion species.

to the nominal foil thicknesses in the Jyväskylä ToF-ERD setup. The number of events simulated to end up inside the energy detector entrance window was counted in both cases and compared. MCERD has been validated by comparing them against SRIM [38]. Ion transport through a carbon foil has been simulated and the transmitted ion angular distributions are compared against each other with various ion energies and masses. MCERD agrees with SRIM well, as the distribution widths were within a 5% margin.

Simulation results produce a ratio between scattered (foils included) and non-scattered (no foils) cases. Simulated data matches, or at least produces a similar trend, when compared to the experimental results as shown in Fig. 7. Although simulation results do not reproduce experimental yield ratios exactly for the Au, the majority of the discrepancy in yields can be explained by the carbon foil scattering (see Fig. 8).

In Ref. [11] simulations results have a similar trend compared to our data. However, the carbon foil scattering effect is larger in our data than in Ref. [11]. This is most likely due to the fact that simulations in Ref. [11] consider only the first timing detector transparency and not the whole system. Similarly, our MCERD simulation results show more scattering than what has been previously measured in Refs. [22,24].

Our scope is on carbon foil scattering happening in timing detectors. However, the gas ionization chamber detector, used as an energy detector, also has a silicon nitride window that can scatter incoming particles. Scattering of the particles in the energy detector silicon nitride window has been studied before. According to Ref. [39], silicon nitride window does not induce major energy loss. Also, those incoming ions that recoil window atoms into the gas do not reduce the total energy measured. Energy loss and scattering in the gas detector entrance window do cause some particles to be detected in lower energy than average and in our analysis, these were included in the selections of recoils.

4. Conclusions

A significant discrepancy between measured and theoretical yields in the low energy ToF-ERD has been demonstrated. With the results presented in this paper, measured data is compared to the theoretical predictions by Rutherford cross-sections and Anderssen screened cross-sections. Discrepancies in yields studied are not affected by the uncertainties in the stopping force. Sample films selected for this study had practically zero composition changes during the measurements, confirmed by time-stamped data acquisition.

In very low energy incident ion beam ToF-ERD, the discrepancy of yields is the largest and for the very heavy target materials, the detected yield discrepancy can be over 50%. When measurements were done at the higher incident beam energies, detected yields approach theoretical yields as expected as the foil scattering cross-sections are reduced.

As a function of recoil energy, recoiled yields from a sample line up regardless of the incident beam Z number. Earlier simulations show that for the mirror measurement geometry, the sample scattering does not cause a reduction of recoil yield. This suggests that the discrepancy would be originating mainly from the carbon foil scattering effect.

Looking for further evidence for the carbon foil scattering effect, the 5.7–5.8 MeV Au recoil and scattered particle yields were reduced by about the same amount compared to the theory prediction independently from the target configuration. In this experiment, the low velocity Au particles in the detector are identical Au ions with identical energy. There is no difference if the particle was a scattered beam particle or a recoil from the sample. This suggests that either screening corrections for both scattering and recoiling particles are the same in this energy, or the vast majority of the reduction of the yield comes from the carbon foil scattering in the detector setup.

Our simulations confirm that the scattering from ToF-telescope carbon foils is a significant factor in the detected reduction of the yield. However, in the measurements, some yields are reduced more than is obtained from the MCERD simulations. The experimentally demonstrated discrepancy in Fig. 7 can be mostly explained by carbon foil scattering, and the remaining discrepancy can be due to the carbon foil thickness being thicker than nominal.

As a main conclusion for every day ToF-ERD measurements, relatively light, like ^{35}Cl and ^{63}Cu , incident beam scattering can be used reliably for heavy target element analysis. According to the data presented in this paper scattering analysis is a viable method with light to middle mass, for example, Cl and Cu, incident beam energies over 5.7–5.8 MeV. The pitfall of using the scattered beam in analysis is that more than one heavier-than-beam target elements are hard or impossible to separate in the spectrum. Recoil yields of middle mass, for example, Co and Ti, reproduce expected yields with incident beam energies over 10 MeV. Detected low energy heavy, for example, Hf and Au, recoil yields are far from the theory expectations and might have significant discrepancies even with relatively highly energetic incident beams.

As mentioned in the introduction, it is considered that at least heavier than incident beam recoils should not be used in the analysis. This consideration was now confirmed as at the moment, there does not exist an analytical model to calculate the detected reduction of the yield due to the carbon foil scattering. There are simulation tools, for example MCERD, to model multiple scattering happening in the detector foils and in the sample itself. It is possible to measure yield correction curves for each setup or to do simulations and use these results as detection efficiency, in this case meaning how many recoils are lost due to the foil scattering. Results show that carbon foils used in ToF-detectors should be characterized and the effects of foil scattering need to be systematically studied. In this work, the thicknesses of

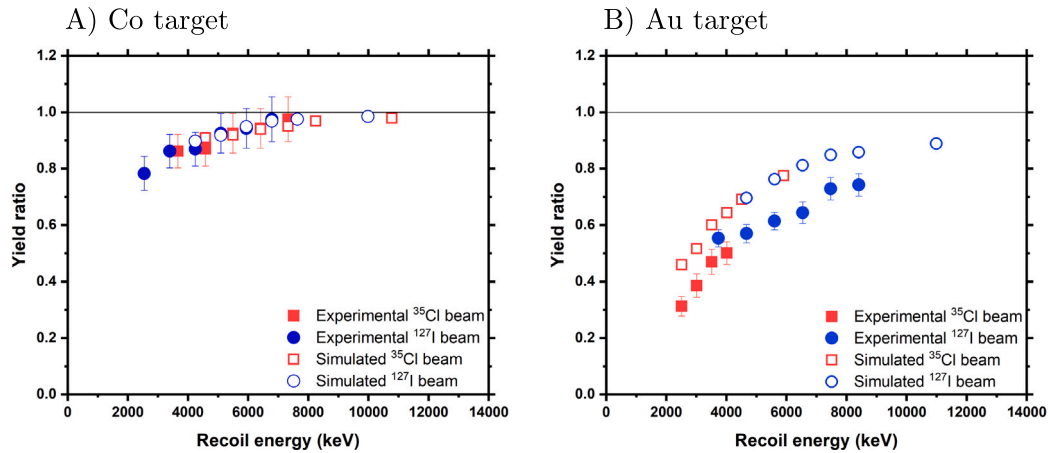


Fig. 7. Simulated yields compared to experimental results shown in Fig. 4 for selected beams for (A) Co recoils and (B) Au recoils. In simulations, yields are reduced by carbon foil scattering and yield ratios produce a similar trend to experimental yield ratios.

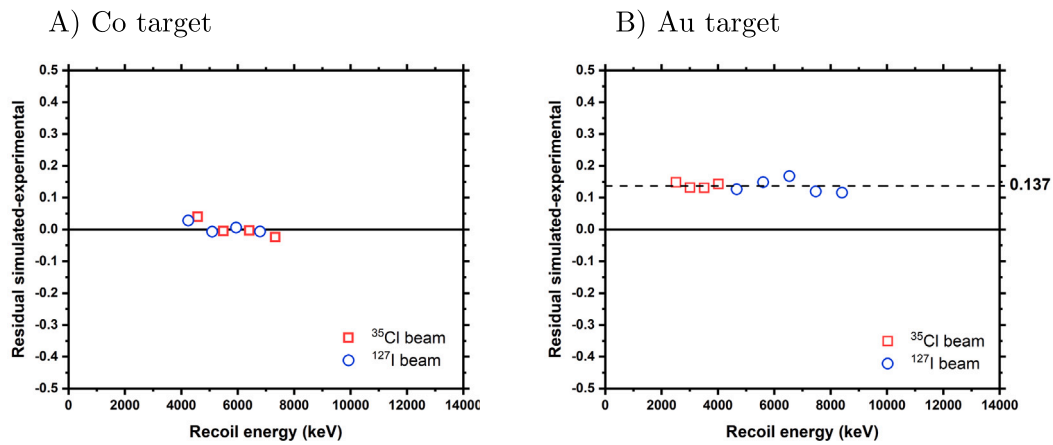


Fig. 8. Simulation residuals for (A) Co and (B) Au thin films compared to the experimental result. From the simulated yield ratio, the experimental yield ratio has been subtracted. In (B) residuals are around a constant line of 0.137. Simulations have been done using nominal carbon foil thicknesses of 3 and 10 $\mu\text{g}/\text{cm}^2$ in first and second timing detectors respectively. In (B) the residual would be reduced close to zero when the first timing detector foil thickness was set to 5 $\mu\text{g}/\text{cm}^2$.

carbon foils in simulations were the nominal values of foils in the Jyväskylä ToF-detector but we are in the process of investigating the carbon foil scattering with a range of foil thicknesses to further improve the quantitativeness of the low energy heavy ion ToF-ERD method.

CRediT authorship contribution statement

Mikko Kivekäs: Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Kenichiro Mizohata:** Investigation. **Jaakko Julin:** Writing – review & editing. **Markku Kainlauri:** Resources. **Mika Prunnila:** Resources. **Laura Keränen:** Resources. **Matti Putkonen:** Resources. **Tatu Korkiamäki:** Investigation. **Mikko Laitinen:** Writing – review & editing, Supervision, Investigation, Conceptualization.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.nimb.2024.165542>.

Data availability

Data will be made available on request.

References

- [1] J. L'Ecuyer, C. Brassard, C. Cardinal, J. Chabbal, L. Deschênes, J.P. Labrie, B. Terreault, J.G. Martel, R. St-Jacques, An accurate and sensitive method for the determination of the depth distribution of light elements in heavy materials, *J. Appl. Phys.* 47 (1) (1976) 381–382.
- [2] R. Groleau, S. Gujrathi, J. Martin, Time-of-flight system for profiling recoiled light elements, *Nucl. Instrum. Methods Phys. Res.* 218 (1) (1983) 11–15.
- [3] J. Thomas, M. Fallavier, D. Ramdane, N. Chevarier, A. Chevarier, High resolution depth profiling of light elements in high atomic mass materials, *Nucl. Instrum. Methods Phys. Res.* 218 (1) (1983) 125–128.

- [4] H.J. Whitlow, G. Possnert, C.S. Petersson, Quantitative mass and energy dispersive elastic recoil spectrometry: Resolution and efficiency considerations, *Nucl. Instrum. Methods Phys. Res. B* 27 (3) (1987) 448–457.
- [5] W. Assmann, P. Hartung, H. Huber, P. Staat, H. Steffens, C. Steinhausen, Setup for materials analysis with heavy ion beams at the Munich MP tandem, *Nucl. Instrum. Methods Phys. Res. B* 85 (1) (1994) 726–731, [http://dx.doi.org/10.1016/0168-583X\(94\)95911-0](http://dx.doi.org/10.1016/0168-583X(94)95911-0).
- [6] J. Forster, P. Currie, J. Davies, R. Siegle, S. Wallace, D. Zelenitsky, Elastic recoil detection (ERD) with extremely heavy ions, *Nucl. Instrum. Methods Phys. Res. B* 113 (1) (1996) 308–311, [http://dx.doi.org/10.1016/0168-583X\(95\)01392-X](http://dx.doi.org/10.1016/0168-583X(95)01392-X).
- [7] J. Stoquert, G. Guillaume, M. Hage-Ali, J. Grob, C. Ganter, P. Siffert, Determination of concentration profiles by elastic recoil detection with a ΔE -E gas telescope and high energy incident heavy ions, *Nucl. Instrum. Methods Phys. Res. B* 44 (2) (1989) 184–194, [http://dx.doi.org/10.1016/0168-583X\(89\)90426-6](http://dx.doi.org/10.1016/0168-583X(89)90426-6).
- [8] H. Timmers, R. Elliman, G. Palmer, T. Ophel, D. O'Connor, The development of a facility for heavy-ion elastic recoil detection analysis at the Australian national university, *Nucl. Instrum. Methods Phys. Res. B* 136–138 (1998) 611–615, [http://dx.doi.org/10.1016/S0168-583X\(97\)00811-2](http://dx.doi.org/10.1016/S0168-583X(97)00811-2).
- [9] A. Razpet, P. Pelicon, Z. Rupnik, M. Budnar, Development of a time-of-flight telescope for ERDA at the jožef stefan institute, *Nucl. Instrum. Methods Phys. Res. B* 201 (3) (2003) 535–542.
- [10] C. Kottler, M. Döbeli, F. Glaus, M. Suter, A spectrometer for low energy heavy ion ERDA, *Nucl. Instrum. Methods Phys. Res. B* 248 (1) (2006) 155–162, <http://dx.doi.org/10.1016/j.nimb.2006.02.013>.
- [11] S. Giangrandi, T. Sajavaara, B. Brijs, K. Arstila, A. Vantomme, W. Vandervorst, Low-energy heavy-ion TOF-ERDA setup for quantitative depth profiling of thin films, *Nucl. Instrum. Methods Phys. Res. B* 266 (24) (2008) 5144–5150.
- [12] M. Laitinen, M. Rossi, J. Julin, T. Sajavaara, Time-of-flight – Energy spectrometer for elemental depth profiling – Jyväskylä design, *Nucl. Instrum. Methods Phys. Res. B* 337 (2014) 55–61.
- [13] M. Msimanga, D. Wamwangi, C. Comrie, C. Pineda-Vargas, M. Nkosi, T. Hlatshwayo, The new heavy ion ERDA set up at ithemba LABS gauteng: Multilayer thin film depth profiling using direct calculation and Monte Carlo simulation codes, *Nucl. Instrum. Methods Phys. Res. B* 296 (2013) 54–60.
- [14] Y. Sugisawa, I. Harayama, Y. Takimoto, T. Wakasugi, Y. Hirose, T. Hasegawa, S. Ishii, D. Sekiba, Installation of TOF-e telescope ERDA in UTTAC at the University of Tsukuba: Analysis of metal-nitride-based multi-layer coatings on glasses, *Nucl. Instrum. Methods Phys. Res. B* 503 (2021) 68–74.
- [15] M. Döbeli, C. Kottler, F. Glaus, M. Suter, ERDA at the low energy limit, *Nucl. Instrum. Methods Phys. Res. B* 241 (1) (2005) 428–435.
- [16] S. Giangrandi, B. Brijs, T. Sajavaara, H. Bender, F. Iacopi, A. Vantomme, W. Vandervorst, Irradiation-induced damage in porous low-k materials during low-energy heavy-ion elastic recoil detection analysis, *Nucl. Instrum. Methods Phys. Res. B* 249 (1) (2006) 189–192.
- [17] J. Julin, T. Sajavaara, Conceptual study of a heavy-ion-ERDA spectrometer for energies below 6MeV, *Nucl. Instrum. Methods Phys. Res. B* 406 (2017) 61–65.
- [18] J. Julin, M. Laitinen, T. Sajavaara, Time-of-flight ERD with a 200mm² Si₃N₄ window gas ionization chamber energy detector, *Nucl. Instrum. Methods Phys. Res. B* 332 (2014) 271–274, <http://dx.doi.org/10.1016/j.nimb.2014.02.076>, 21st International Conference on Ion Beam Analysis.
- [19] H.H. Andersen, F. Besenbacher, P. Loftager, W. Möller, Large-angle scattering of light ions in the weakly screened Rutherford region, *Phys. Rev. A* 21 (1980) 1891–1901.
- [20] M. Laitinen, J. Meererschaut, M. Döbeli, I. Bogdanović Radović, Private communication.
- [21] J. Kim, Y. Kim, H. Choi, G. Kim, H. Woo, C. Whang, Measurement of elastic recoil cross sections of light nuclei by ³⁵Cl using inverse scattering, *Nucl. Instrum. Methods Phys. Res. B* 129 (3) (1997) 323–326, [http://dx.doi.org/10.1016/S0168-583X\(97\)00312-1](http://dx.doi.org/10.1016/S0168-583X(97)00312-1).
- [22] W. Stüdemann, B. Wilken, Detection efficiency of a heavy ion time-of-flight spectrometer with thin carbon foils in the start detector, *Rev. Sci. Instrum.* 51 (2) (1982) 175–180.
- [23] H.H. Andersen, J. Böttiger, Multiple scattering of heavy ions of keV energies transmitted through thin carbon films, *Phys. Rev. B* 4 (1971) 2105–2111.
- [24] H.H. Andersen, J. Böttiger, H. Knudsen, W. Möller Petersen, T. Wohlenberg, Multiple scattering of heavy ions of keV energies transmitted through thin films, *Phys. Rev. A* 10 (1974) 1568–1577.
- [25] H. Knudsen, H. Andersen, Multiple scattering of MeV gold and carbon ions in carbon and gold targets, *Nucl. Instrum. Methods* 136 (1) (1976) 199–201.
- [26] L. Meyer, Plural and multiple scattering of low-energy heavy particles in solids, *Phys. Status Solidi (b)* 44 (1) (1971) 253–268.
- [27] G. Amsel, G. Battistig, A. L'Hoir, Small angle multiple scattering of fast ions, physics, stochastic theory and numerical calculations, *Nucl. Instrum. Methods Phys. Res. B* 201 (2) (2003) 325–388.
- [28] K. Arstila, T. Sajavaara, J. Keinonen, Monte Carlo simulation of multiple and plural scattering in elastic recoil detection, *Nucl. Instrum. Methods Phys. Res. B* 174 (1) (2001) 163–172.
- [29] F. Schiettekatte, Fast Monte Carlo for ion beam analysis simulations, *Nucl. Instrum. Methods Phys. Res. B* 266 (8) (2008) 1880–1885, <http://dx.doi.org/10.1016/j.nimb.2007.11.075>.
- [30] M. Mayer, K. Arstila, K. Nordlund, E. Edelmann, J. Keinonen, Multiple scattering of MeV ions: Comparison between the analytical theory and Monte-Carlo and molecular dynamics simulations, *Nucl. Instrum. Methods Phys. Res. B* 249 (1) (2006) 823–827, <http://dx.doi.org/10.1016/j.nimb.2006.03.148>.
- [31] S. Giangrandi, K. Arstila, B. Brijs, T. Sajavaara, A. Vantomme, W. Vandervorst, Considerations about multiple and plural scattering in heavy-ion low-energy ERDA, *Nucl. Instrum. Methods Phys. Res. B* 267 (11) (2009) 1936–1941.
- [32] J. Jokinen, J. Keinonen, P. Tikkanen, A. Kuronen, T. Ahlgren, K. Nordlund, Comparison of TOF-ERDA and nuclear resonance reaction techniques for range profile measurements of keV energy implants, *Nucl. Instrum. Methods Phys. Res. B* 119 (4) (1996) 533–542.
- [33] J. Colaax, G. Terwagne, C. Jeynes, On the traceably accurate voltage calibration of electrostatic accelerators, *Nucl. Instrum. Methods Phys. Res. B* 349 (2015) 173–183.
- [34] M. Putkonen, T. Sajavaara, L.-S. Johansson, L. Niinistö, Low-temperature ALE deposition of Y₂O₃ thin films from β -diketonate precursors, *Chem. Vapor Depos.* 7 (1) (2001) 44–50, [http://dx.doi.org/10.1002/1521-3862\(200101\)7:1<44::AID-CVDE44>3.0.CO;2-Q](http://dx.doi.org/10.1002/1521-3862(200101)7:1<44::AID-CVDE44>3.0.CO;2-Q).
- [35] K. Arstila, J. Julin, M. Laitinen, J. Aalto, T. Konu, S. Kärkkäinen, S. Rahkonen, M. Raunio, J. Itkonen, J.-P. Santanen, T. Tuovinen, T. Sajavaara, Potku – New analysis software for heavy ion elastic recoil detection analysis, *Nucl. Instrum. Methods Phys. Res. B* 331 (2014) 34–41.
- [36] G. Knoll, *Radiation Detection and Measurement*, Wiley, 2010.
- [37] M. Mayer, *Simnra user's guide*, 1997.
- [38] J. Ziegler, J. Biersack, U. Littmark, *The Stopping and Range of Ions in Matter*, Pergamon Press, 1985.
- [39] J. Julin, K. Arstila, T. Sajavaara, Simulations on time-of-flight ERDA spectrometer performance, *Rev. Sci. Instrum.* 87 (8) (2016) 083309, <http://dx.doi.org/10.1063/1.4961577>.