Simulation of Multiple Scattering Effects on Coincidence

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Abstract. The Monte Carlo ion beam analysis simulation program Corteo was adapted to the case of coincidence spectrometry of identical ions. Since multiple scattering (MS) is properly simulated, it reproduces very well the shape of a spectrum, especially the depth-dependent detection efficiency, provided that the actual experimental conditions are taken into account in the simulation. Assumptions such as an energy difference discrimination equivalent to an angular restriction on the detector cannot be made for thick targets because multiple scattering decorrelates the angle at which an ion is scattered or recoiled from the one at which it emerges from the target. It is also shown that in thick targets, the efficiency of detection depends on MS effects on incident ions. The consequence is that if a standard is used to estimate the detection efficiency, it should be made of material with a similar composition as the analysed sample.

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INTRODUCTION

Coincident detection of scattered and recoiled atoms in a collision is a powerful method to obtain the depth profile of light elements in relatively thick targets. It was initially developed by Cohen *et al.*[1] for hydrogen profiling and was further extended to micro-beams[2] and detection of heavier elements.[3]

A common feature of coincidence measurements is that as a result of multiple scattering (MS), the efficiency of detection depends on depth. In this paper, an adaptation of the Monte Carlo program Corteo for the simulation of coincidence spectra is presented. It is shown that it can reliably reproduce most features of an experimental spectrum, namely the depth-dependence. The effect of target composition on the efficiency of detection is also discussed.

MULTIPLE SCATTERING

In the idealised picture of coincidence spectrometry, an incident ion enters the target and slows down by electronic energy loss following a strait trajectory until it undergoes a single scattering event where it is directed towards one of the two detectors while the primary recoil aims at the other detector. If the scattered and recoiled atoms have the same mass, they will be scattered at 90° from each other, and if both sample surfaces are parallel and normal to the beam and the detectors are conveniently positioned at ±45° from the beam axis, both ions will leave the main collision site with the same energy, following straight trajectories. They will undergo the same electronic energy loss and will be detected with the same energy.

Obviously, many assumptions are made in this idealized picture, the most consequential being that the trajectories are straight. The incident, scattered or recoiled ions may undergo collisions that deflect their trajectory enough to escape detection. The probability of such MS increases with the distance traveled across the target. Samples measured by coincidence spectrometry are typically several micrometers thick in other to form a self-supporting film, so the effect is generally significant. It is more important for events occurring near the front of the sample (beam side), since either the scattered or recoiled ion may undergo significant MS leading to the loss of coincidence, than for events generated near the back of the sample (detectors side) for which only the incident ion suffers MS. The effect is thus depth-dependent, and a reliable evaluation requires a precise MS modeling.

SIMULATIONS

Corteo is a Monte Carlo simulation program intended to simulate ion beam analysis spectra. It computes the trajectory of ions in materials, based on binary collision and random phase approximations. Computations are significantly accelerated by extracting the scattering angle components from tables

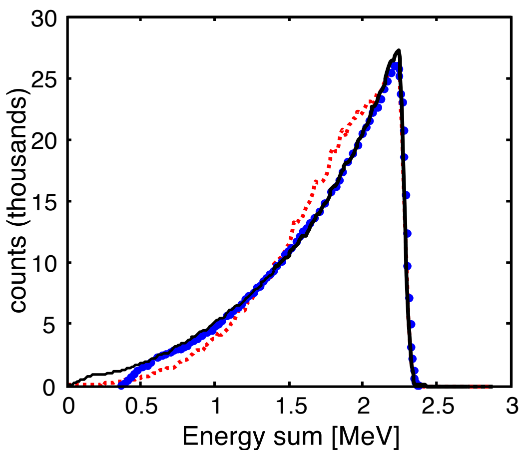

FIGURE 1. Coincidence spectrum of H in a Mylar film 29 µm thick, obtained with a 2.8 MeV proton beam. Symbols: experiment by Wegden et al.[8]; Solid line: simulation with ΔE<146.5 keV and full size detectors; Dotted line: simulation obtained considering a detector sensitive in an annulus that forms an angle between 43.5 and 46.5 degrees to the beam axis and no energy difference discrimination.

rather than computing them using the MAGIC algorithm used in TRIM.[4] After computing the trajectory of the incident ion to a certain depth, Corteo generates (during what will be called below the main collision) a scattered ion or a recoil. The trajectory of this ion is then computed until it emerges out of the target, and then its intersection with the detector is checked. An energy spectrum is accumulated by summing the cross-section of the main collisions. A detailed description of Corteo and its use for the simulation of Rutherford Backscattering Spectrometry and Elastic Recoil Detection can be found in Ref. [5].

The program is now adapted to follow both scattered ion and recoil in order to simulate a coincidence spectrum. It also includes provisions for two arbitrary positioned rectangular or elliptical detectors, which can be annular. For coincidence, an energy difference discrimination can also be specified. The current version is limited to the coincident detection of identical particles, and the virtual detector strategy[5,6] cannot be applied in that case. The program is now parallelised to run on multi-core processors and comes with a user interface, both available for download with their source code under the terms of the Gnu General Public Licence.[7]

In order to compare to experiments, recent data published by Wegden *et al.*[8] where simulated. Using a 2.8 MeV proton beam normal to the target, they obtained the H spectrum of a 29 µm Mylar foil, used as a standard, and other geological samples. They used an annular detector divided in two sensitive parts and positioned at 9 mm behind the sample and centered on the beam axis. The diameter of the sensitive zone was approximately 13.4 mm (deduced) and the annulus radius was 6 mm. The detector energy resolution was 25 keV. They also included an energy difference discrimination ΔE of about 146.5 keV (deduced), inferred to correspond to an angular restriction of 45±1.5 degrees from beam axis on the detector.

For the simulations presented here, the mean free path was set to 10 nm, and the scattered particles where thrown in a randomly selected direction within a half sphere towards the detector, the recoil being correspondingly directed at 90 degrees from the scattered ion in the same collision plane. The spot size was assumed to be 1 mm in diameter. For Mylar, a compound correction factor of 0.957 was applied to the stopping power computed using the Bragg's rule. In order to get smooth spectra, 50 million incident ions where simulated in each case, the simulation lasting about 50 minutes on an Intel processor featuring four 2.4 GHz cores. Depending on the conditions, between 6×10^4 and 3×10^6 events where detected.

COMPARISON TO EXPERIMENT

Figure 1 presents the comparison of two simulations to the results obtained by Wegden *et al.* for a 29 µm Mylar foil.[8] The simulated spectrum represented by a solid line is obtained using the (deduced) ΔE<146.5 keV. It is seen that it reproduces very well the part of the experimental spectrum above 0.5 MeV. For this simulation, an incident charge of 4.5 µC was considered. Wegden et al. indicate that the spectrum was obtained in about 1.5 hour with a 200 pA beam, which implies an incident charge of about 1 µC. The discrepancy can be due to different factors, namely whether one or both halves of the detector can trigger events, how the solid angle is computed, or the use of the Rutherford cross-section for the simulation. It is also seen from Fig. 1 that the simulated spectrum extends below 0.5 MeV. Events below a certain energy threshold where probably rejected in the experiment.

Wegden *et al.* surmise in their paper that the ΔE discrimination they applied is equivalent to limiting the detection to an angle range of 45±1.5 degrees. A simulation was carried out considering an annular detector with an interior radius of 8.540 mm and an exterior radius of 9.484 mm to represent this condition, and no ΔE discrimination. The result is presented as a dashed curve in Fig. 1. (An incident charge of 18 µC was considered so the maximum of the simulation fits that of the experimental spectrum.) Clearly, the shape of this simulated spectrum does not reproduce the experiment: an energy difference discrimination cannot be assumed to be equivalent to an angular restriction, at least for targets thicker than a few micrometers.

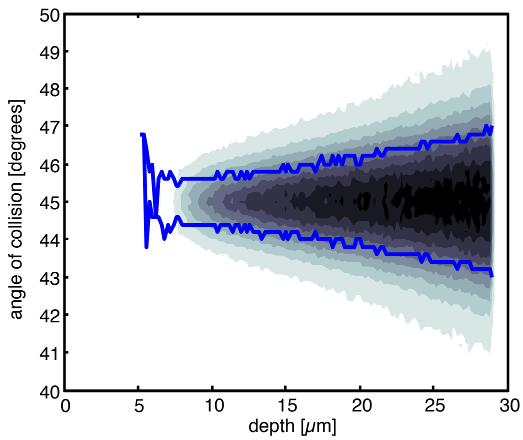

FIGURE 2. Contour bands: distribution of simulated coincident events from Mylar (with ΔE<146.5 keV) as a function of the main collision depth and the angle to the beam axis of the scattered ion or recoil. The solid line contour delimits the angle range for which the distribution reaches 50% of its maximum value at this depth.

We will see the reason in Fig. 3, but in order to understand it, let's first look at Fig. 2. The coloured contour bands represent the distribution of the number of detected events as a function of the depth of the main collision and the scattering or recoiling angle after this collision. The distribution is centered at 45°, and as for the energy spectrum, the largest number of events originates from the back of the target (detector side). It is also seen that very few collisions originate from a depth of 5 to 7 μm, and no events originate from below 5 μm. This is due to the fact that an incident proton at 2.8 MeV produces upon collision scattered and recoil protons that, if they are symmetrically scattered at 45° from the beam axis, have an energy of 1.4 MeV. A 1.4 MeV proton at 45° of incidence has a projected range of about 25 μm in Mylar, which is smaller than the target thickness.

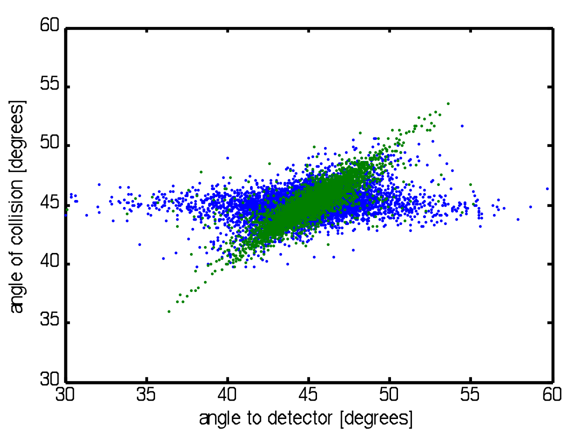

FIGURE 3. Angle of ions as they emerge from the main collision locus as a function of the angle they make with the detector when they are detected. Green, mostly diagonal dots: events originating from a depth between 21 and 29 μm. Blue, mostly horizontal dots: events originating from a depth interval between 7 and 21 μm.

The contour plot is influenced both by the efficiency of detection (i.e. deflection of initially well oriented ions) and the collision angle distribution as a function of depth. The solid line on Fig. 2 delimits for each depth the angle interval for which the distribution reaches 50% of its maximum. Considering these limits, it is seen that the angle interval of events originating from the back of the sample (29 μm) is 45±1.9°, a bit larger than the inferred angle restriction based on the ΔE discrimination, but narrows down by a factor of more than 3 to 45±0.6° at a depth of 8 to 10 μm. The effect of MS is thus to filter out events not directed at 45°, and the filtering narrows as the distance the pair of ions has to travel increases.

Figure 3 shows the relation between the collision angle and the impact angle on the detector. Since the annular detector is symmetrically facing the beam, their axes are the same. Events mostly following the diagonal (plotted in green) are those generated in the depth interval between 21 and 29 μm. For them, a clear correlation exists between the angle at which the ion is emitted after the main collision and the angle it has as it reaches the detector: trajectories are mostly straight. However, for the main collisions occurring at depth between 7 and 21 μm, no such correlation exists and whatever is their detection angle, they are emitted from the main collision at an angle close to 45°, while reaching the detector with a wide range of angles. This explains why for thick targets a ΔE discrimination is not equivalent to an angle restriction on the detector. As a result of MS, events generated in certain direction may end up in any region of the detector while fulfilling the ΔE condition, and this effect changes with depth, influencing the shape of the whole spectrum in a non-linear way.

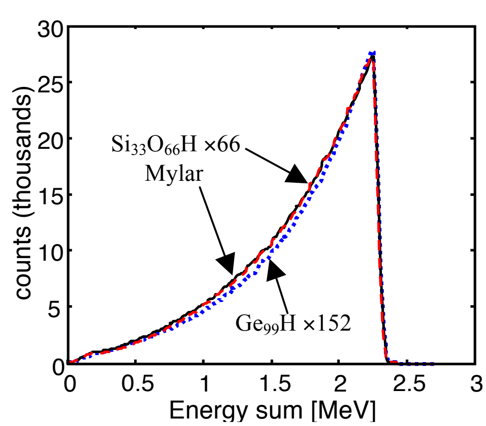

FIGURE 4. Simulated H coincidence spectrum, including the condition ΔE<146.5 keV. Solid line: 29 μm of Mylar. Dashed line: 18.5 μm of $Si_{33}O_{66}H$. Dotted line: 14.4 μm of $Ge_{99}H$.

TABLE 1. Main contributions to the spectral ratio of the different simulations plotted in Fig. 4. The total stopping $(dE/dx)_{total}$ represents the spectral energy difference per unit depth (at/cm^2), including angular factors, near the back of the sample (detector side). The fraction detected is the fraction of incident ions that reach the back of the sample and lead to a detected event, compared to Mylar. The ratio A/B was used as multiplying factor in Fig. 4.

	Mylar ($C_{10}H_8O_4$)	**$Si_{33}O_{66}H$**	**$Ge_{99}H$**
Hydrogen concentration [H]	36.4%	1%	1%
$(dE/dx)_{total}$ (10^{15} eV/[at/cm^2])	7.8	13.8	29.4
$(dE/dx)_{total}$/[H] ratio normalised to Mylar (A)	1	65	137
Fraction detected compared to Mylar (B)	1	0.986	0.905
A/B	1	66	152

TARGET ATOMIC MASS

MS depends on the collision cross-cross section resulting from the screened interatomic potential (here, the Universal potential) and increases in importance with the atomic weight of the target elements. The consequence is that it is not clear that a sample can be used as a standard to estimate the detection efficiency for another target made of elements with an atomic weight significantly different. In this section, we estimate the effect of target composition on the detection efficiency. In Fig. 4, two additional simulated spectra are compared to the simulated spectrum obtained for Mylar in Fig. 1, considering the same ΔE discrimination. One of them is a layer of 18.5 µm of $Si_{33}O_{66}H$ and the other is 14.4 µm of $Ge_{99}H$. Both thus contain 1 at% of hydrogen. The target thicknesses were adjusted so that the high energy edge of the simulations overlaps that of the Mylar. It is seen that the three simulations overlap quite well, except for a multiplicative factor. This spectral ratio is mainly due to the difference in H concentration and in stopping power. As seen from Tab. 1, these two factors account for a factor 65 between Mylar and $Si_{33}O_{66}H$, and for a factor 137 between Mylar and $Ge_{99}H$. While such a factor brings the $Si_{33}O_{66}H$ spectrum close enough to that of Mylar, this is not quite the case for $Ge_{99}H$. From the simulations, it is also found that due to MS, almost 10% of the incident ions reach the back of the sample at an angle that will not result in a detectable event compared to Mylar (factor B in Tab. 1). Taking this factor into account, the ratio between Mylar and $Ge_{99}H$ spectra is about 152 near the back of the sample (high energy edge). This is the factor applied in Fig. 4. MS has other non-linear effects that make the shape of the simulated spectrum for $Ge_{99}H$ not fully comparable to that of Mylar, as seen from the figure.

The main consequence is that if a standard is used to determine the detection efficiency, it must be made of elements with similar atomic weight than the sample to analyse. Alternatively, the spectrum may be simulated while accounting for experiment details.

CONCLUSION

It was shown that a detailed Monte Carlo simulation of the ion trajectory based on the binary collision and random phase approximations can effectively simulate a coincidence spectrum, namely its depth-dependent detection efficiency. Computation speed improvements brought by Corteo makes such simulations possible in a few minutes. However, the actual geometry has to be simulated in order to obtain a spectrum comparable to experiment. Assumptions such as a ΔE discrimination equivalent to an angular restriction cannot be made for thick samples because, as a result of MS, the correlation between the emission angle at the main scattering locus and the angle at which an ion emerges from the target is lost after crossing a few micrometers through the target. Consequently, an ion can hit the detector outside the assumed angular limit while satisfying ΔE condition. Also, in thick targets, the efficiency of detection depends of the MS effects on the incident ions, thus on target composition. As a consequence, if a standard is used to determine this efficiency, its composition must be relatively close to that of the analysed sample.

REFERENCE

1. B.L. Cohen, C.L. Fink, J.H. Degnan, J. Appl. Phys. **43** (1972) 19
2. D. Dujmić, M. Jakšić, N. Soić, T. Tadić and I. Bogdanović, Nucl. Instr. and Meth. **B111** (1996) 126
3. J. A. Moore, I. V. Mitchell, M. J. Hollis, J. A. Davies, L. M. Howe, J. Appl. Phys. **46** (1975) 52
4. J.F. Ziegler, J.P. Biersack, U. Littmark, *The Stopping and Range of Ions in Solids*, New York: Pergamon, 1985.
5. F. Schiettekatte, Nucl. Instr. Meth. **B266** (2008) 1880
6. K. Arstila, T. Sajavaara, J. Keinonen, Nucl. Instr. Meth. **B174** (2001) 163.
7. http://www.lps.umontreal.ca/~schiette
8. M. Wegden, P. Kristiansson, Z. Pastuovic, H. Skogby, V. Auzelyte, M. Elfman, K.G. Malmqvist, C. Nilsson, J. Pallon, A. Shariff, Nucl. Instrum. Meth. **B219–220** (2004) 550