Conceptual study of a heavy-ion-ERDA spectrometer for energies below 6 MeV

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Abstract

Elastic recoil detection analysis (ERDA) is a well established technique and it offers unique capabilities in thin film analysis. Simultaneous detection and depth profiling of all elements, including hydrogen, is possible only with time-of-flight ERDA. Bragg ionization chambers or $\Delta E - E$ detectors can also be used to identify the recoiling element if sufficiently high energies are used. The chief limitations of time-of-flight ERDA are the beam induced sample damage and the requirement of a relatively large accelerator.

In this paper we propose a detector setup, which could be used with 3 MeV to 6 MeV medium heavy beams from either a single ended accelerator (^{40}Ar) or from a tandem accelerator (^{39}K). The detector setup consists of two timing detectors and a gas ionization chamber energy detector. Compared to use of very heavy low energy ions the hydrogen recoils with this beam have sufficient energy to be detected with current gas ionization chamber energy detector. To reduce the beam induced damage the proposed detector setup covers a solid angle larger than 1 msr, roughly an order of magnitude improvement over most time-of-flight ERDA setups. The setup could be used together with a small accelerator to be used for light element analysis of approximately 50 nm films.

The concept is tested with ^{39}K beam from a 1.7 MV Pelletron tandem accelerator with the Jyväskylä ToF-ERDA setup. In addition to the measurements effects related to low energies and increase in the solid angle are simulated with Monte Carlo methods.

Keywords: ERDA, time-of-flight

1. Introduction

Time-of-flight ERDA has been used since 1976 [1] for light element analysis. Typical heavy ion with energies of approximately 1 MeV u^{-1} also allow medium heavy ions to be analyzed with time-of-flight techniques or ionization chambers [2]. The accelerator required for these beams are out of reach of many ion beam analysis (IBA) labs.

Low energy ERDA has been proposed and studied by others [3–6]. These systems are used with 2 MV or smaller tandem accelerators. The main limitations of these instruments compared to a higher energy system are reduced probing depth and mass resolution. When low energy very heavy ions such as ^{127}I or ^{197}Au are used the quantification of hydrogen is difficult because of the low recoiling energy.

Large solid angle $\Delta E - E$ gas ionization telescopes or Bragg ionization chambers do not offer the elemental resolving power required for low energy measurements, so it seems the time-of-flight–energy spectrometer is the only practical choice. Moreover this avoids complicated calibrations of the spectrometer, as only the time-of-flight calibration and detection efficiency for hydrogen and some of the lightest elements must be accounted for.

We propose a compact large solid angle spectrometer design, backed up by Monte Carlo simulations and experiments with existing detectors. The design is intended to be used with incident beams with masses between 35 and 40, (e.g. ^{35}Cl, ^{39}K or ^{40}Ar) with energies around 5 MeV.

The design uses previously reported [7, 8] detector constructions, which are used successfully in routine analysis. The requirements for the accelerator system are lessened, enabling many labs access to a high resolution light element analysis tool.

Hydrogen detection and quantification is possible in the very low energy range of 70 keV to 300 keV. The larger solid angle than with previous designs is intended to mitigate the increase in sample damage due to use of lighter ions, while the position sensitive energy detector reduces the effects of kinematic broadening.

2. Proposed spectrometer design

The proposal relies on achieving sufficient mass resolving power, the ability to detect hydrogen and a depth resolution similar to existing ToF-ERDA. The solid angle of the proposal is then maximized. The significance of timing resolution on depth resolution is reduced at lower energies, therefore the distance between the timing detectors can be reduced, increasing the solid angle. A time-of-flight resolution of 200 ps and a time-of-flight length of 50 cm to 100 cm are typical with current spectrometers. These typically use beams with 10 MeV or

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more energy. The design presented here should be able to offer similar depth resolution with lower energy beams. Even after accounting for grids reducing the geometric transmission in the timing detectors the solid angle should be much greater than 1 msr if the distance between the timing detectors is reduced to 30 cm.

2.1. Accelerator requirements

The required beam can be provided by a variety of accelerators with different ion source designs. A single ended 3 MV accelerator can provide much more than 1 pnA of $^{40}Ar^{2+}$ to meet the upper range of energies considered in this study with the necessary flux even with conventional RF ion sources. Sufficient beam currents of $^{40}Ar^{3+}$ should also be possible with minor modifications [9], so that existing 2 MV accelerator systems are also usable. A miniaturized accelerator system could be built around a single ended system with 500 kV to 1000 kV terminal voltage if such a system is equipped with an ion source capable of producing higher charge state ions, such as Penning or electron cyclotron resonance (ECR) ion source.

A 1 MV tandem normally used with proton and He beams can in principle be used to produce 5 MeV to 6 MeV $^{39}K^{4+/5+}$, however the characteristics of such a system for heavy ion focusing etc. remain untested. Additionally the accelerator system should be equipped with a heavy ion source, such as a caesium sputtering ion source.

2.2. Geometry

In this design the time-of-flight length is 300 mm, much shorter than in any current published designs. The solid angle increases dramatically if the first timing detector is placed only 150 mm from the sample, allowing the energy detector to be placed approximately 500 mm from the sample. See Fig. 1.

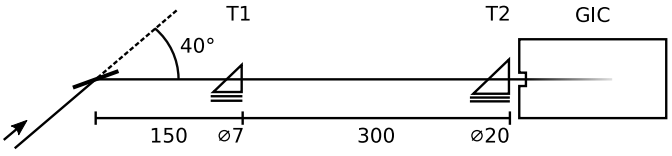

Fig. 1. Schematic of the proposed detector setup. The timing detectors T1 and T2 are placed close to each other and the sample, maximizing the detector solid angle.

The choice of the detector angle (θ) is a compromise between sensitivity, and mass and depth resolutions. Sensitivity improves at large angles, due to increasing cross sections. Mass resolution improves with recoil energy, which is increases at low angles. Depth resolution improves with longer effective ion and recoil path lengths at glancing angles. The sample inhomogeneity and multiple scattering also play a role in the choice of ideal geometry.

Most ERDA-telescopes are placed at an angle of 30° to 45° in respect to the beam direction. Quite often a fixed angle is preferred since the angle must be known well. Here an angle of 40° has been chosen so that the simulated and calculated results could be compared with the measurements performed with the existing spectrometer installed at 41.3° angle.

2.3. Timing detectors

The proposed timing detector design is based on the design by Busch et al [10], since these offer excellent timing resolution and simplicity of construction. The only drawback is the large number of various grids, which reduce the solid angle and increase the probability of background events.

T1 foil should be as thin as possible, but still offer adequate hydrogen detection efficiency. As thin as 0.5 $\mu g\,cm^{-2}$ diamond-like carbon (DLC) foils have been used for the start detector [4, 11]. The secondary electron yield can be enhanced by coating the foil with LiF [11] or Al_2O_3 [7]. Between 70 keV to 300 keV energies the detection efficiency with an Al_2O_3 coated 2 $\mu g\,cm^{-2}$ foil should be larger than 50%, depending on the MCP gain and electronics. The scattering of heavy recoils by the T1 carbon foil may limit the quantification of heavier recoils, this is discussed in Sect. 4.3.

T2 size is ultimately limited by the practical difficulties in mounting very large thin foils. Based on previous experience 5 $\mu g\,cm^{-2}$ carbon foil can be mounted on a frame with a 20 mm hole if supporting wires are used.

2.4. Gas ionization chamber

The gas ionization chamber should be designed to stop all recoils in the active volume. This means that either the length of the electrodes or the pressure of a GIC designed for higher energies can be reduced. The current Jyväskylä design [8] with an electrode length of 150 mm would be directly usable with a pressure of 10 mbar to 15 mbar.

At this pressure up to 400 mm^2 square entrance window is possible without supports, if 100 nm thick silicon nitride window is used. Alternatively patterned windows can be used, enabling the use of thinner silicon nitride. A thin entrance window will improve the mass resolution of the spectrometer and also reduce background [12].

3. Experimental studies

Experiments to study the feasibility of low energy ERDA were performed with the 1.7 MV Pelletron 5SDH-2 in Jyväskylä Accelerator Laboratory.

In the time-of-flight spectrometer [7, 8] the time-of-flight length is 623 mm and the solid angle is limited by a 200 mm^2 energy detector window approximately 100 cm from the sample. The T1 detector has a 3 $\mu g\,cm^{-2}$ carbon foil.

3.1. Mass and depth resolution

All light elements H, Li, C, N and O and their isotopes can be separated with ease even with 4 MeV beam, see Fig. 2. Even the small Cl impurity in the Li_2CO_3 sample can be detected, although the quantification is affected by background from scattered ^{39}K. Depth profiles from measurement with 5.1 MeV beam of the same sample are presented in Fig. 3.

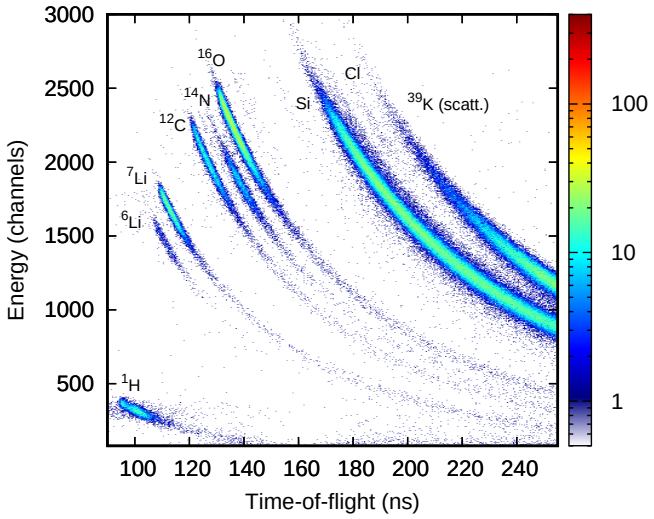

Fig. 2. 57 nm thick Li_2CO_3 film on Si measured with 4 MeV ^{39}K beam. The mass resolution is limited by the energy resolution of the GIC.

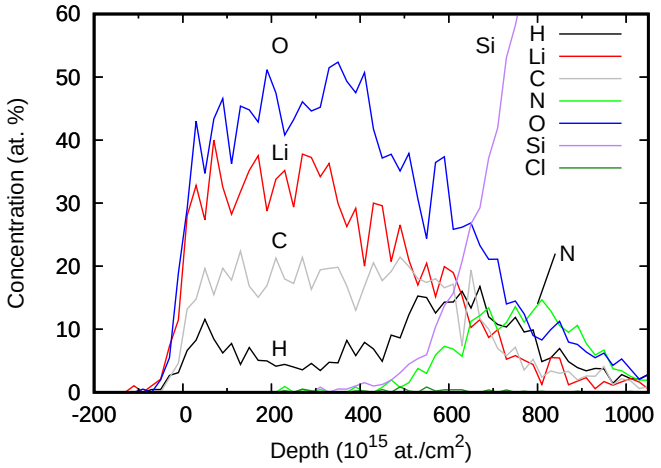

Fig. 3. 57 nm thick Li_2CO_3 film on Si measured with 5.1 MeV ^{39}K beam and 10° tilt between the beam and the sample surface. The depth resolution is affected by the roughness of the sample.

3.2. *Hydrogen detection and analysis*

Hydrogen down to 80 keV can be detected with existing ionization chambers, but the energy calibration will not be perfectly linear. Also the resolution of the GIC will limit the resolution of the hydrogen efficiency calibration. Since the detection efficiency near the hydrogen stopping maximum is strongly energy dependent [13], the uncertainties associated with the energy resolution and calibration will be large.

The efficiency calibration of the spectrometer involves measuring the fraction of recoils detected by all the detectors to recoils detected by the energy detector [13], which is assumed to have a 100% efficiency above a certain threshold energy. For this purpose the energy calibration of the GIC must be monotonous, i.e. similar pulse heights may not be created by recoils with two different energies.

If another detector, such as a silicon detector, is placed after the GIC it is possible to calibrate the detection efficiency of the time-of-flight detectors over a broad energy range if the GIC detector gas is removed. The solid angle of such a silicon detector would not match the GIC solid angle, so deviations due to pinholes at the edges of timing detector carbon foils etc. may cause deviations in the calibration. Such differences would however be evident in the common energy range where either the silicon detector and the GIC can be used to determine the detection efficiency.

Certified hydrogen reference material with 13.9% hydrogen in silicon, deposited using chemical vapor deposition (CVD) [14] was measured with beams of ^{39}K with various energies. The hydrogen concentration was determined using Potku [15] using cross section corrections by Andersen et al. [16]. The results are plotted in Fig. 4. The hydrogen concentration from the same depth interval can be seen to increase when measured using lower incident energy. The results may vary due to inaccuracies in the stopping forces in silicon for H and Si recoils. Other reasons for this are discussed in Sec. 4.3.

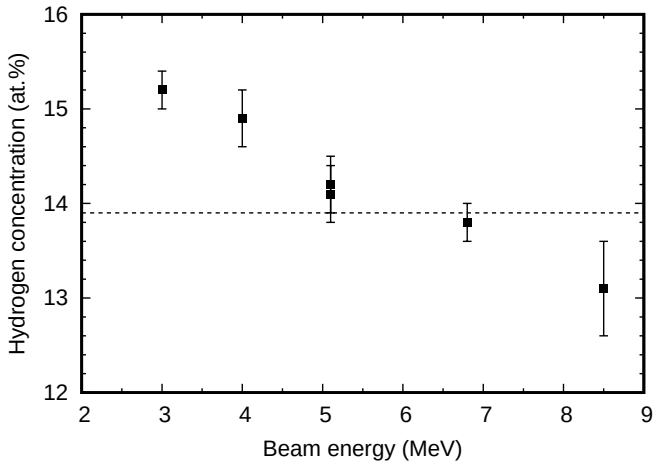

Fig. 4. Hydrogen concentration of the reference sample [14] measured with beams of ^{39}K with different energies. The error bars indicate the statistical uncertainty. The dashed line represents the nominal H concentration of the sample, (13.9 ± 1.1) at.%. The 5.1 MeV point has been measured twice, both as the first and the last measurement in order to monitor possible changes in the sample by the ion beam bombardment.

4. Simulations

4.1. *Geometrical effects*

Path length differences due to detector geometry $\frac{\Delta L}{L}$ affects velocity resolution of the spectrometer. If T1 and T2 foils are assumed to be parallel and $L = 300$ mm the largest deviation assuming a point-like beam spot is neglible, $\frac{\delta L}{L} \approx 1.00025$. This figure does not include scattering from the carbon foils. The perfect correction for different path lengths requires two 2D position sensitive detectors, and in practice also very careful alignment of the detectors.

More importantly large solid angle geometry causes kinematic broadening due to scattering angle variations. This contribution can be reduced with a position sensitive detector. When we consider a monoenergetic beam to recoil surface

atoms with a nominal centerline time-of-flight of 100 ns the actual time-of-flight distribution as a function of T2 lateral position is plotted in Fig. 5. This distribution includes variations due to different recoiling angles (θ, ϕ), and time-of-flight length differences. The beam was assumed to be collimated to a 1 mm (horizontal) by 2 mm (vertical) rectangle, which projected on to a sample placed 20° relative to the beam creates a spot of 3 mm by 2 mm on the sample.

The 1D position x at T2 can be used as a measure of the true recoil angle θ around the centerline ideal recoil angle $\theta_{\text{center}} = 40°$ with simple geometry:

$$\theta = \theta_{\text{center}} + \arctan\left(\frac{x}{d_{\text{T2}}}\right), \quad (1)$$

where d_{T2} is the distance of the T2 detector from the sample.

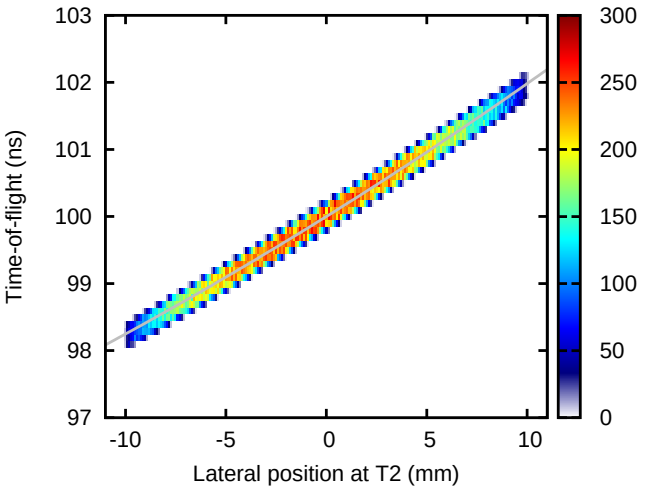

Fig. 5. Calculated time-of-flight of surface recoils created by a monoenergetic incident beam as a function of the lateral (scattering plane) coordinate of the second timing detector (T2). The spectrometer centerline time-of-flight corresponds to 100 ns. The kinematic broadening can be compensated well with one dimensional position sensitivity, i.e. the measured lateral position x is assumed to affect the recoiling angle directly, see Eq. 1. The plotted line is the expected time-of-flight given one dimensional corrections.

These geometrical effects scale linearly with time-of-flight. For recoils originating beneath the surface of the sample the geometry affects in a more complicated manner due to path length differences in the sample. The fundamental limit to depth resolution will eventually be energy straggling. The effective timing resolution (standard deviation) at surface with the specified spot size and geometry can be predicted using formula

$$\delta t = \sqrt{\left(\frac{200\,\text{ps}}{2.35}\right)^2 + (0.00136 \cdot t)^2}, \quad (2)$$

for a time-of-flight t, assuming the position sensitivity is 1 mm at T2. While the kinematic broadening after corrections is non-negligible for the time-of-flight range of interest (30 ns to 150 ns), the total effective resolution is comparable to or better than in many setups where position sensitive detectors are not used.

4.2. Depth resolution

In order to get an understanding of the significance of multiple scattering, kinematic broadening and how well it could be compensated in practice simulated were made using a modified version of MCERD [12, 17].

This simulated data was fed to an analysis program Potku [15] to extract the depth profiles shown in Fig. 6, which correspond to the depth profiles used in the simulations, but with many effects contributing to the depth resolution. The sample was tilted 10° in respect to the incoming beam. A 5.1 MeV ^{39}K beam with a dose of 10^{11} particles (16 pnC) was used in the simulations, corresponding to approximately 28 000 oxygen counts. If the energy detector count rate is kept below 2 kHz this measurement is possible in 10 minutes with a beam current below 30 ppA.

4.3. Scattering

Low energy heavy ions have issues with multiple scattering in the sample and large angle scattering in the detector foils [6]. Scattering in the T1 carbon foil leads to reduction in detection efficiency, since some recoils miss the T2 or energy detector. The distance between the timing detectors increases this effect. The apparent enhancement of H concentration at lower energies seen in Fig. 4 is partially due to this, since the silicon is more likely to scatter in the T1 carbon foil than hydrogen. In order to quantify this effect MCERD simulations were performed. The scattering from carbon foils did not enhance the H concentration results more than 0.2 percentage points when 4 MeV or higher energies were used, with the proposed 2 μg cm^{-1} T1 foil the effect will be even smaller.

5. Discussion and conclusions

The presented setup can be used to give a complete elemental depth profiles of thin film samples containing mostly light elements. Light elements including H, C, N, and O can be unambiguously identified.

The high solid angle combined with high cross sections provides means to study sensitive materials. Quantitative depth profiling of approximately 50 nm thick films is possible, although at lowest energies some corrections might be necessary. Quantification of heavy elements is not recommended, because of the multiple scattering and scattering in the detector foils.

The low energy and low beam current requirements combined with a compact design open possibilities to use this spectrometer design as a part of a low-cost time-of-flight ERDA setup.

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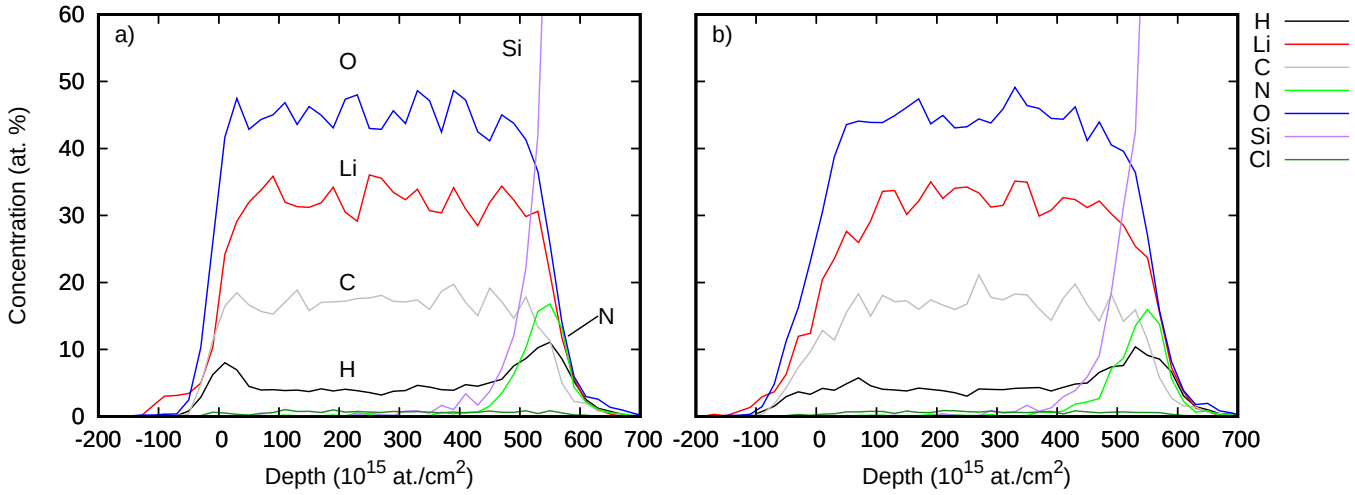

Fig. 6. Depth profiles from simulated data with 5.1 MeV ^{39}K beam. MCERD and gas ionization chamber simulations were used to model the proposed setup. One dimensional position sensitivity was used in the analysis in (a), while no position sensitivity was used in (b). The Li_2CO_3 sample was assumed to be smooth and homogeneous and contain some H at the surface and H and N at the interface, similarly to depth profiles in Fig. 3. With the position sensitivity the depth resolution near the surface is comparable to the existing setup.

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