Measurement of He and H depth profiles in tungsten using ERDA with medium heavy ion beams

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Abstract

Elastic recoil detection analysis method (ERDA) with medium heavy analysing ion beam and its application for the simultaneous measurement of light elements in a very heavy substrate is presented. The availability of cross section data and the method of cross section calculation for recoiled particles are discussed. Different ion species for analysing beam are discussed with respect to the cross-section data availability, sensitivity of the method, and the depth resolution. Calculations of depth resolution for each element and maximum depth of analysis for tungsten substrate are presented. The influence of the detector geometry and multiple scattering effects on the depth resolution is discussed. An example spectrum measured on tungsten implanted with He seeded D plasma is shown.

1. Introduction

Elastic recoil detection analysis (ERDA) with an incident He beam is widely used for determining the concentration or depth profile of hydrogen (H - includes 1H, 2H) in materials. However, an incident beam of heavier ions is required when simultaneous measurements of H and He concentrations or depth profiles are needed. The use of heavier ions has the advantage of larger stopping powers, increased sensitivity, and higher mass resolution in comparison to conventional He-ERDA. The measurements can be performed on different types of ERDA systems: 1) Time-of-flight ERDA (TOF-ERDA) or 2) conventional ERDA with a stopping foil in front of the detector. TOF-ERDA has the advantage of detecting and separating several elements lighter than the incident beam. However, in the case of very heavy substrates with low concentrations of light elements (e.g. H and He in tungsten), the use of TOF-ERDA is impractical due to very high yield of backscattered primary ions. For such cases, the use of conventional foil-ERDA, where the backscattered primary ions are filtered by a stopper foil, is advantageous.

The use of medium heavy ions like C or O has already been proposed for simultaneous depth profiling of H and He [2, 3]. We refer to this method as medium heavy ion ERDA (MI-ERDA). Medium heavy ions can be easily generated in tandem accelerators with negative sputter ion sources. However, the lack of cross-section data for ERDA in the past has limited the application of MI-ERDA for quantitative depth profiling of H and He. The present development of the IBANDL database and the availability of evaluated backscattering cross-section data at any angle by SigmaCalc now make MI-ERDA a practically applicable method. Still, the incident ion beam has to be selected based on availability of cross-section data, as cross-section data is still scarce or unavailable for many projectile-target combination. This paper discusses the application of MI-ERDA with respect to cross-section data availability, mass resolution, achievable depth resolution, and maximum analyzable depth.

2. Experimental

The measurements were performed at the Garching tandem accelerator (Max-Plank-Institut für Plasmaphysik, Garching, Germany) in the RKS analysis station [7] using a solid state detector with a nominal resolution of 10 keV FWHM for 5.5 MeV alpha. O ions were created by sputtering an aluminum oxide target. The samples were hit with 15 MeV O^{5+} ions under an incident angle of 75 ° to the sample normal. During the measurement the current on the target was kept between 5 and $10*10^{-9}$ A in order to avoid thermal damages of the target. Recoiled particles were collected at 30 ° recoil angle. A 5 µm Ni foil was placed in front of the detector. The precise foil thickness was determined before installation with proton-RBS at 165 ° and was $4.66x10^{19}$ atoms/cm^2. The aperture in front of the detector is 0.35 mm wide and 1.65 mm high at a distance to the sample of 24.8 mm, corresponding to a geometrical solid angle of 0.967 msr. This geometrical solid angle was checked by He-ERDA and a calibrated implanted target [10] containing H and was experimentally reproduced within 5 %, which is the inaccuracy of the measurement.. A set of Si samples with different aC:H and aC:D coatings was used as a reference targets.

3. Experimental results and discussion

3.1. Cross-section calculations

Various analyzing ion beams can be used for the simultaneous measurement of hydrogen isotopes and helium: lithium, carbon, nitrogen, oxygen, and silicon are all easily obtainable ion species. However, the evaluation of H and He depth profiles or concentrations from the measured spectra requires the recoil cross-sections of H and He with each different incident beam. Experimental

recoil cross-sections for incident ions heavier than lithium are still very scarce. Fortunately, the H and He recoil cross-sections in the forward direction can be calculated through inverse kinematics from the backscattering cross-sections at backward angles [2]. But this still requires the knowledge of the H and He backscattering cross-sections which is not always available. Due to this fact oxygen and carbon ion beams were chosen, because for both of these elements evaluated backscattering cross-sections of H and He are available. The following calculations of the depth resolution (section 3.3) showed that the use of an oxygen beam gives a better depth resolution and no overlap between deuterium and helium peaks.

A schematic view of the scattering and recoil geometry during MI-ERDA in the laboratory and center of mass frame is given in Fig. 1. The recoil angle φ_1 and incident energy $E_{1,1}$ are related to the scattering angle Θ_2 and incident energy $E_{1,2}$ in the inverse scattering reaction case by equations (1) and (2) below [1, 2]: .

$$\tan\Theta_2 = \frac{\sin 2\varphi_1}{M_2/M_1 - \cos 2\varphi_1}, \tag{1}$$

$$E_{1,2} = \frac{M_2}{M_1} E_{1,1} \tag{2}$$

where the $E_{1,2}$ is the energy of incident ions in the inverse case and θ_2 is the scattering angle in the inverse case. The calculated inverse scattering reaction values of Θ_2 and $E_{1,2}$ for hydrogen, deuterium and helium with O at the energy of 15 MeV and $\varphi_1 = 30$ ° is tabulated in Table 1. Using these values the scattering cross-sections for the inverse reactions O_{16} (H_1 , H_1) O_{16}; O_{16} (H_2 , H_2) O_{16}; O_{16} (He_4, He_4) O_{16} were calculated using SigmaCalc available from the IBANDL web site. The cross-section data for ERD reactions were then converted from the data obtained from SigmaCalc using the following equation:

$$\frac{\sigma(\Theta')}{\sigma(\Theta_2)} = \frac{\sin^2\Theta_2}{\sin^2\Theta'}\cos(\Theta' - \Theta_2), \tag{3}$$

The calculated recoil cross-sections in the energy range from 8 MeV to 17 MeV are shown in Fig.2. At beam energies above about 7 MeV the cross-sections become non-Rutherford. The deviation from Rutherford value increases with increasing beam energy, and at 15 MeV the difference (for H recoils) is already about 30%. It should be mentioned that for 2H and He the calculated cross-sections have complicated shapes, and especially in the case of He the cross-section has a number of resonances. Consequently, the incident beam energy has to be selected with care.

3.2 Mass resolution and peak overlap

One of the main purposes of the MI-ERDA method is to measure all light elements presented in the sample in one spectrum. In this respect the peaks from different elements should not overlap. In Figure 3 the energy of recoils at the different incident ion energies is plotted. At the energies of 14 MeV and 20 MeV respectively carbon and oxygen ions are already penetrating the stopper foil. At low energies both for carbon and oxygen beams the overlap between deuterium and helium peaks is expected. However in the case of carbon beam the energy range, where the overlap is observed, is wider as for oxygen. In this respect it is preferred to use the oxygen ions for analysing beam.

3.3. Depth resolution and maximum depth of analysis

In Figure 4, the depth resolution for ERD analysis with O and C beams is plotted calculated using the ResolNRA program [8] for the tungsten substrate case. The stopping powers and foil straggling were calculated with SIMNRA [8] using SRIM 2010 stopping power data. The oxygen beam provides a better depth resolution than the carbon beam for all three elements; the difference at the surface is about 20%. In Figure 5, the contribution of geometric straggling and multiple scattering

on depth resolution is plotted for the O beam. The depth resolution at the surface is dominated by geometrical straggling due to the width of the used detector aperture, and a somewhat better resolution could be achieved with a smaller aperture. However, as this would also decrease the detector solid angle and consequently the sensitivity, a reasonable compromise has to be made between resolution and sensitivity, resulting in the selected aperture size. At larger depths, the depth resolution deteriorates mainly due to multiple scattering on the outgoing path: The influence of the multiple scattering in the case of a very heavy substrate like tungsten is rather large.

The maximum depth of analysis was estimated by taking the depth at which the energy of the recoiling particles reaching the detector was close to zero. The maximum depth of analysis was calculated to be $3200*10^{15}$ atoms/cm ^{2}, corresponding to a thickness of ~500 nm in tungsten. In reality, this value cannot be realized due to contributions from electronic noise at small signal amplitudes and overlap of the signals from the detected recoils.

3.4. Examples

The calculated cross sections from Section 3.2 are now used to fit experimentally measured spectra from tungsten samples implanted with He seeded D plasma ($1.4*10^{20}$ D^+/m^2s; 2% He; t_{impl} = 1 hour; 500 K) at the permeation experiment in Osaka University, Japan [9]. Before the irradiation the mirror polished pure tungsten samples were kept at 1000 K for about 12 hours in order to remove surface impurities and surface damage produced by the polishing process. Several ERDA measurements with different samples were performed in order to compare the spectra from the samples implanted with He seeded and with pure D plasma. An example spectrum is presented in Figure 6. The small number of counts in the measured spectrum could be explained by the value of the total charge collected during the measurement. For this spectrum this value was kept by 10 μC in order to avoid the radiation damages of the sample and the loss of H due to the

bombardment with relatively heavy ions of analysing beam. The simulation of the spectrum was done with the SIMNRA program [8].The simulated curve is in very good agreement with measured data. As it was mentioned before the low statistics is caused by the collected charge number, though for H one more effect can play a role. The implantation was done at the temperature of 500 K, after the implantation the specimen was cooled down to the room temperature and then kept at the atmosphere. During the cooling process significant amount of implanted D can come out of the specimen, which results in a smaller statistics in ERD measurement. Nevertheless one point, which is mostly important for He data evaluation, should be mentioned. For the depth profile data evaluation from the experimental spectrum a good energy calibration of the ERD detector is required. In this work the detector calibration was performed with a set of Si samples coated with a:C-H and a:C-D films, thus providing a reasonable energy calibration for both hydrogen isotopes. The same calibration was also used for He, An accurate calibration for He is generally difficult, because targets containing both He isotopes in sufficient amounts and at (or at least very close to) the surface are difficult to obtain. The lack of an accurate calibration for He has some minor influence on the accuracy of He depth profile calculations.

4. Conclusions

The classical foil ERDA method with a medium heavy ion beam (MI-ERDA) is a simple and convenient method for simultaneous measurement of light elements (hydrogen isotopes, helium) in very heavy substrates (tungsten). Due to the recent development of IBANDL cross-section database the quantitative evaluation of the depth profiles and total amount of elements from the measured spectra is now possible. The cross-sections for recoiled particles can be calculated from the backscattered cross-sections available from IBANDL through the inverse kinematics. However for many projectiles the available data is still scarce. The use of oxygen beam is advantageous due to

availability of the cross-section data, the better depth resolution and less overlap of signals. The achievable depth resolution in the case of oxygen beam for helium is 20 nm. The maximum depth of analysis for tungsten using 15 MeV oxygen beam is 500 nm (regardless to the parameters of the detector). In the case of tungsten substrate the influence of multiple scattering on the depth resolution is rather strong. As this effect is caused by the material of the sample it can not be avoided.

List of references

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Table 1 – 1 column

Parameters used for the kinematic conversion.

Inverse scattering parameters, 15 MeV O+, recoil angle 30°		
projectile	Incident ion energy in inverse case (keV)	Scattering angle in inverse case (°)
1H	945.11	116.78
2H	1888.82	113.4
4He	3753.63	106.1

List of figure captions

1. schematic view of scattering geometry in laboratory and center of mass systems
2. recoil cross section for ^{1,2}H and ^{4}He reactions in 8 – 17 MeV energy range
3. depth resolution calculated for hydrogen, deuterium and helium. On the left: carbon ion beam 12.5 MeV; on the right: oxygen ion beam 15 MeV
4. energy of recoils as a function of the incident ion energy. On the left: oxygen beam in the energy range from 10 to 20 MeV; on the right: carbon analyzing beam in the energy range from 8 to 14 MeV.
5. depth resolution for the following reaction - 4He(16O, 4He)16O. the solid line is the total depth resolution, the other lines show the contribution of geometrical straggling, multiple scattering and other effects to the final depth resolution
6. ERD spectrum for tungsten sample irradiated with H-D-He mixed beam.

Figure 1

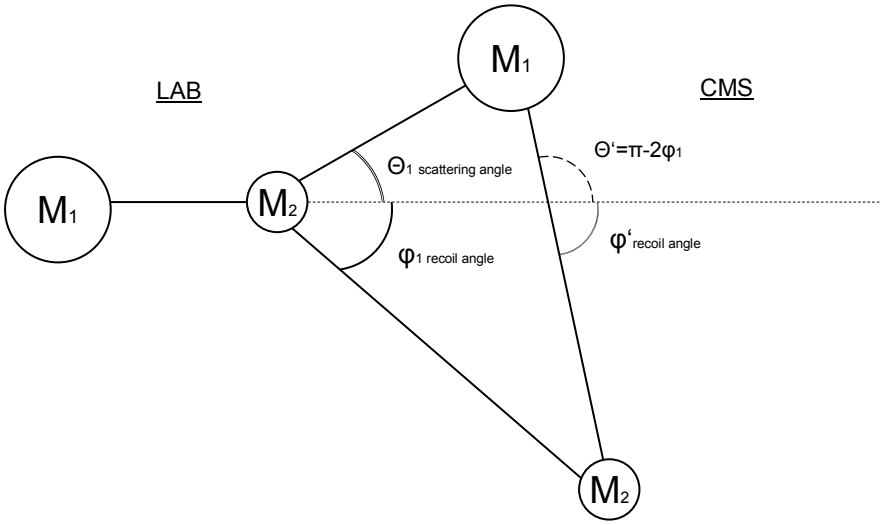

LAB
CMS
M1
M2
M1
M2
Θ1 scattering angle
Θ'=π-2φ1
φ1 recoil angle
φ' recoil angle

Figure 2

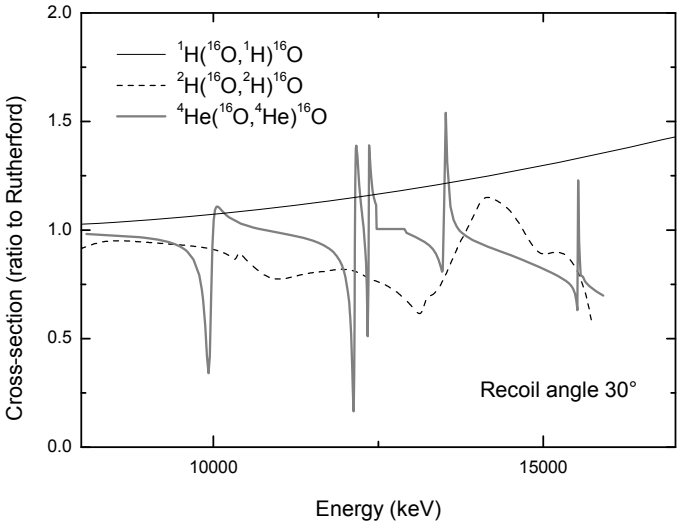
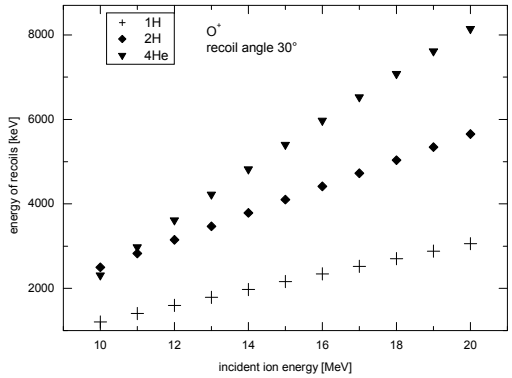

2.0
1.5
1.0
0.5
0.0
Cross-section (ratio to Rutherford)
$^{1}H(^{16}O,^{1}H)^{16}O$
$^{2}H(^{16}O,^{2}H)^{16}O$
$^{4}He(^{16}O,^{4}He)^{16}O$
Recoil angle 30°
10000
15000
Energy (keV)

Figure 3


1H
2H
4He
O+
recoil angle 30°
energy of recoils [keV]
incident ion energy [MeV]
8000
6000
4000
2000
10
12
14
16
18
20

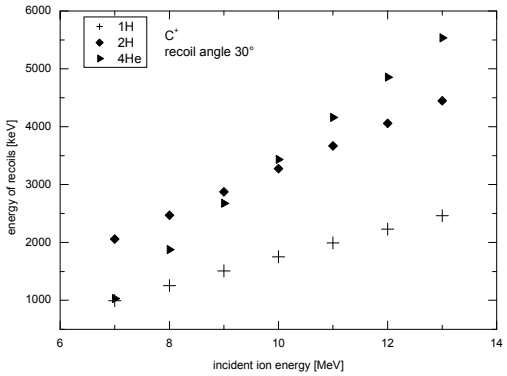

1H
2H
4He
C+
recoil angle 30°
energy of recoils [keV]
incident ion energy [MeV]
6000
5000
4000
3000
2000
1000
6
8
10
12
14

Figure 4

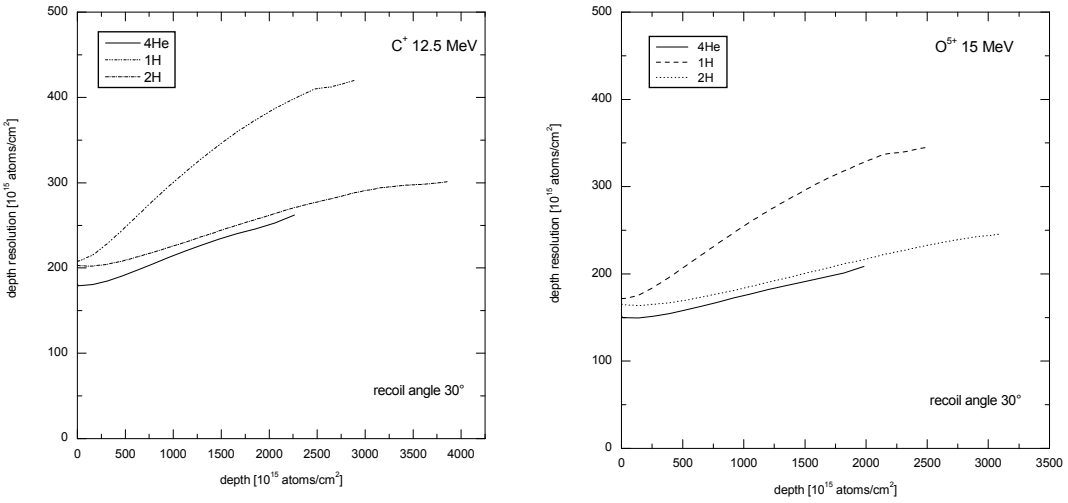

4He
1H
2H
C+ 12.5 MeV
recoil angle 30°
depth resolution [10^15 atoms/cm^2]
depth [10^15 atoms/cm^2]
O5+ 15 MeV

Figure 5

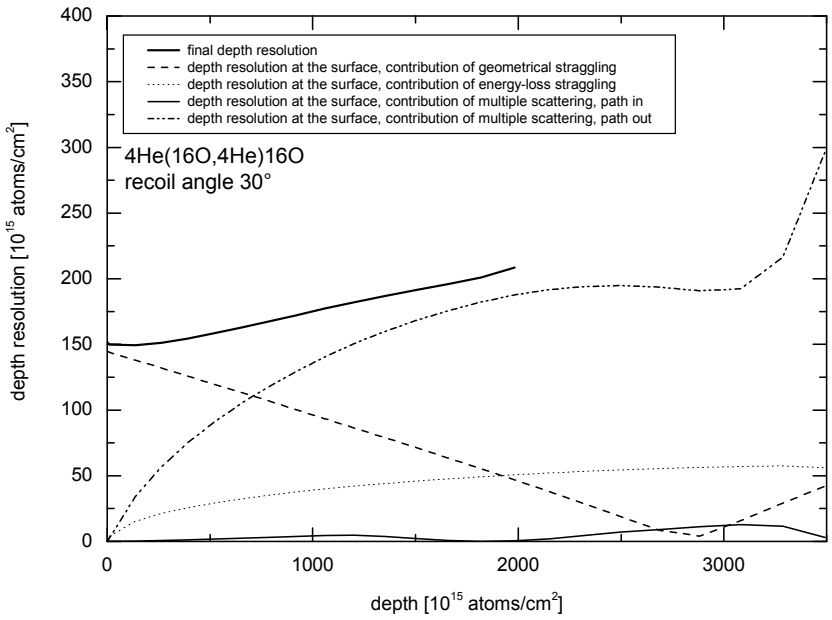
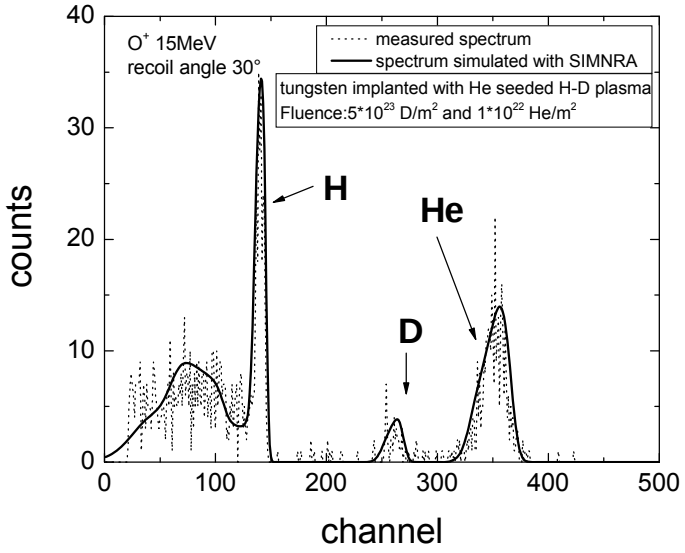

final depth resolution
depth resolution at the surface, contribution of geometrical straggling
depth resolution at the surface, contribution of energy-loss straggling
depth resolution at the surface, contribution of multiple scattering, path in
depth resolution at the surface, contribution of multiple scattering, path out
4He(16O,4He)16O
recoil angle 30°
depth resolution [10^15 atoms/cm^2]
depth [10^15 atoms/cm^2]
0
50
100
150
200
250
300
350
400
0
1000
2000
3000

Figure 6


O+ 15MeV
recoil angle 30°
measured spectrum
spectrum simulated with SIMNRA
tungsten implanted with He seeded H-D plasma
Fluence:5*10^23 D/m^2 and 1*10^22 He/m^2
H
He
D
counts
channel
0
10
20
30
40
0
100
200
300
400
500