MASTER

Analysis of light elements in thin films using high energy ion scattering techniques

van Dijk, P.W.L.

Award date:
1992

Link to publication
Analysis of Light Elements
in Thin Films using
High Energy Ion Scattering Techniques

P.W.L van Dijk

VDF/NK 92-02
Eindhoven, February 1992
## CONTENTS

Abstract

1. Introduction
   1.1 The Used Scattering Techniques

2. Theory of Collision Kinematics and Interaction of Ions with Matter
   2.1 Introduction
   2.2 Kinematics of Elastic Scattering
   2.3 Inelastic Scattering with Energy Loss $Q$
   2.4 Differential Scattering Cross Section $d\sigma/d\Omega$
      2.4.1 Resonant Scattering
      2.4.2 The Optical Model
   2.5 Stopping of Ions in Matter
   2.6 Energy Straggling
   2.7 Multiple Scattering
   2.8 Other Contributions to Energy Spread

3. The Atomic Hydrogen to Carbon Ratio in Amorphous Carbon Layers
   3.1 Experimental Setup
   3.2 Evaluation of the RBS Spectrum
   3.3 Determination of the Incident Proton Energy
   3.4 Evaluation of the $45^\circ$ Spectrum
   3.5 The Standard, Mylar on an Aluminum Carrier Foil
   3.6 Energy Spectra Obtained from an Aluminum Foil
   3.7 The Background Subtraction Procedure
   3.8 Background Subtraction Procedure for the Mylar Standard
   3.9 Results
      3.9.1 Reproducibility for an Aluminum Clean Foil
      3.9.2 Reproducibility for the Standard
      3.9.3 Reproducibility of the Experiments
      3.9.4 Composition Changes of the Samples
      3.9.5 Analysis of Different Samples
      3.9.6 16 MeV Incident Protons used for Analysis of the Samples
   3.10 Discussion of the Technique
   3.11 Conclusion

---

1  2  3  4  5  6  7  8  9  10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35

4.1 Energy Measurement at a Forward Angle 35
4.2 Coincident Elastic Recoil Detection Analysis (CERDA) 36
4.3 CERDA Technique Complemented with Time Of Flight Measurement 39
  4.3.1 The Geometry of a CERDA-Time-Of-Flight Setup 39
  4.3.2 The Timing of the Scattered Projectile and the Ejected Recoil Atom 40
  4.3.3 The Width of the CERDA-TOF Spectrum 40
  4.3.4 The CERDA-TOF Experiment 42
  4.3.5 Tilting of the Investigated Target 43
4.4 Depth Resolution at the Fast Time Leading Edge 45
  4.4.1 The System Time Resolution 45
  4.4.2 Time Spread Due to Energy Spread in the Detected Particles 46
  4.4.3 Energy Spread of the Incident Particles 46
  4.4.4 Energy Spread Resulting from Angular Spread 46
  4.4.5 Compensation of the Time Spread caused by Detector Acceptance Angles 48
  4.4.6 The Beam Spot Size 50
  4.4.7 The Divergence of the Incident Beam 51
  4.4.8 Depth Resolution as a Function of the Beam Divergence 51
4.5 Depth Resolution at the Slow Time Edge 54
  4.5.1 Energy Straggling 54
  4.5.2 Multiple Scattering 55
4.6 Carbon Recoil Atoms Traversing an Aluminum Layer 57
  4.6.1 The Energy Spread Caused by the Aluminum Layers 58
  4.6.2 Growth of Carbon on the Aluminum During the Experiment 59
4.7 CERDA-TOF at the 45° Scattering Geometry 60
  4.7.1 Depth Profiling of a Carbon Layer on an Aluminum Carrier Foil 61
  4.7.2 Analysis of Self-Supporting Foils 64
  4.7.3 Thin Layer CERDA-TOF with a Glancing Exit Angle 65
  4.7.4 Discussion of the 45° CERDA-TOF Setup 66
4.8 The CERDA-TOF Multi Mass Spectrometer 68
  4.8.1 Depth Profiling of Si₃N₄ 68
4.9 Conclusion and Recommendations 71
## Contents

References

Appendix

| Appendix A: Kinematic Factors | A1 |
| Appendix B: Detector Tilt Angles | B1 |
| B.1 The Recoil Detector Tilt Angle | B1 |
| B.2 The Start Detector Tilt Angle | B1 |
| B.3 Derivation of $\theta + \tau_a = \theta_c$ | B2 |
ABSTRACT

Three ion beam analysis techniques were used in order to analyze carbon self-supporting foils and carbon layers on a thick carrier. The aim was to attain improved depth resolution in depth profiling of light elements in a heavy matrix with CERDA-TOF and to determine the hydrogen and oxygen content in "diamond like" carbon films, deposited on an aluminum carrier, with use of simultaneously measurement of the energy of particles at forward and backward angle.

A method to determine the hydrogen, carbon and oxygen content in a, on aluminum deposited, "diamond like" carbon layer was developed. The composition of the deposited layers was determined from energy spectra obtained from a simultaneously ERDA and RBS measurement. The obtained results were compared to a mylar standard. The accuracy in the determined composition was about 3% for the mylar standard.

A better depth resolution is obtained in the CERDA-TOF experiment. The contributions that limit good depth resolution are the energy spread of the incident particles, divergence of the incident beam, energy straggling and multiple scattering. The measured depth resolution for carbon depth profiling was below 10 nm. A possibility for depth profiling of other elements, nitrogen, oxygen and silicon was demonstrated. Furthermore a geometry was found which gives possibilities for depth profiling of thin layers on a thick 20 μm aluminum foil with the same depth resolution.

However CERDA-TOF was developed to select only one element of interest for depth profiling, it seems to be possible to depth profile more than one element simultaneously (Multi Mass Spectrometer).
CHAPTER 1

Introduction

The experiments, described in this report, were carried out at the Physics Department of the Eindhoven University of Technology in The Netherlands. All experiments concern the investigation of a target by means of Ion Beam Analysis (IBA). Some well-known techniques in IBA are: Rutherford Back Scattering (RBS), Elastic Recoil Detection Analysis (ERDA), Proton Induced X-Ray Emission (PIXE) and Channeling. A new technique, CERDA-TOF, was developed for depth profiling of light elements in a heavy matrix [Rij 91]. All mentioned techniques use accelerated ions impinging on the investigated target. The ion accelerator was a 30 MeV AVF cyclotron. Both alpha particles and protons can be accelerated to final energies between 3 MeV and 30 MeV. The accelerated particles are transported towards the target chamber, where analysis of the samples takes place, by a beam transport system. Figure 1.1 shows a map of the location of all experimental facilities at the Eindhoven Physics Department. The major components of the nuclear beam system are denoted in the figure.

![Figure 1.1: The nuclear beam system at the Eindhoven Physics Department.](image)

The beam transport system consists of quadrupoles which focus the beam and dipoles in order to bend the beam. A dispersive system consisting of two slits with two 45° bending magnets inbetween selects ions on energy and reduces the energy spread of the beam. The beam enters a switcher magnet and is transported to the target chamber involved in the experiment.

There are four locations where different IBA techniques are practiced. The PIXE setup and the Scanning Proton microbeam setup (SPIXE). The Channeling setup,
which is being developed but already denoted in the figure, and the RBS and
CERDA-TOF techniques, described in this report, were carried out in the RBS
scattering chamber. The Time Of Flight pipe is also indicated in figure 1.1.

The slits located between the switcher magnet and the RBS target chamber
limit the beam divergence and are mentioned in section 4.4, where the beam
divergence will be discussed.

1.1 The Used Scattering Techniques

The experiments that were carried out concern RBS, ERDA and CERDA-TOF.
Rutherford Backscattering Spectrometry has been carried out for almost 70 years. In
RBS the elastically, from the target material, scattered particles are detected at a
backward angle. The energy of the backscattered particles depends on the atomic
mass of the target atom where the particle scattered from. Equations for both elastically
and inelastically scattering are derived in chapter 2. The RBS is extremely powerful for
analysis of targets consisting of heavy elements (Z>20) compared to the target atoms.
RBS becomes increasingly difficult when one seeks to detect lighter elements because
of the ion scattering cross sections which decrease with atomic number. Especially
when heavier substrate material obscures the already small signal from the ion mass
material. Therefore other techniques were considered, namely ERDA and CERDA-
TOF.

The ERDA technique makes use of heavy incident ions and is used for depth
profiling of light elements and is powerful for hydrogen depth profiling. In ERDA
the detector is located at a forward angle so that ejected recoil atoms originated from
the target can be detected and the energy determined. In order to select the recoils
from the more heavy forward scattered projectiles, stopper foils in front of the
detector are used which stop the projectiles and let the recoils through. When the
recoils are ejected with high energy alpha particles, the recoils can not be
discriminated from the forward scattered alpha particles when a stopper foil is used.
An alternative to overcome this problem is Coincident Elastic Recoil Detection
Analysis (CERDA) [Kle 86] where both the elastically scattered alpha particle and the
ejected recoil atoms are detected.

CERDA is complemented with a Time Of Flight measurement of the ejected
recoil atom (CERDA-TOF) and is extremely powerful when depth resolution when
depth profiling of light elements in a heavy matrix is requested.

This report consists of two main parts. One part will concern the carbon and
hydrogen determination in "diamond like" carbon films, deposited on an aluminum
carrier, and is presented in chapter 3. The second part concerns depth profiling of
carbon and nitrogen with the CERDA-TOF technique. A Multi Mass Spectrometer
with improved depth resolution is presented and CERDA-TOF is further developed
for the depth profiling if thin films positioned on a thick carrier.
CHAPTER 2

Theory of Collision Kinematics and Interaction of Ions with Matter

2.1 Introduction

Materials are often analyzed with ion beams. The advantage of ion beam analysis over chemical techniques is that the material can be investigated without destructing the sample. This in contrast with chemical analysis. The Ion Beam Analysis can be described by scattering of ions from a target material. Other possible interactions, besides scattering, are excitation or inelastic scattering or nuclear reactions. This chapter focuses on resonant scattering, elastic and inelastic scattering.

A beam of charged particles with known mass and energy impinges on a target. Ions are scattered from the medium and detected under a chosen angle. The composition of the medium involved is derived from the final energy of the detected ions. The interaction of ions with matter is described by the interaction of particles with a repulsive potential originating from target nuclei at which interaction takes place. For low energy ions, below 3 MeV, the Coulomb potential is used as the interaction potential. Scattering at these energies is called Coulomb scattering or Rutherford scattering. Ions with higher energy, beyond 3 MeV for alpha particles, are able to penetrate the nuclei of the target atoms and nuclear reactions can take place or compound nuclei can be formed. The description of these interaction processes needs a different approach.

In this chapter the major interactions of ions with matter are summarized starting with elastic scattering. Inelastic scattering will be treated in section 2.3. The differential scattering cross section is discussed in section 2.4. This is a measure for the scattering probability and it changes with angle and energy. Finally expressions for stopping of ions in matter, straggling and multiple scattering are given.

2.2 Kinematics of Elastic Scattering

The interaction of two masses, denoted as $M_1$ and $M_2$, is considered. Mass $M_1$ has a kinetic energy $E_0$ and mass $M_2$ is assumed to be at rest. In case of a collision between two masses $M_1$ and $M_2$ both conservation of momentum, parallel and perpendicular to the direction of the projectile, and energy conservation have to be satisfied. Elastic scattering is simplified because kinetic energy is considered and no mass is transferred. Two angles $\theta$ and $\phi$ are defined in the scattering geometry shown in figure 2.1.
The scattering geometry. Angles \( \theta \) and \( \phi \) are scattering angles of masses \( M_1 \) respectively \( M_2 \) in the laboratory system. Corresponding angles in the centre of mass system, \( \theta_{cm} \) and \( \phi_{cm} \) are also shown. (From [Fel 86]).

The angle \( \theta \) under which the scattered projectile leaves the target after interaction with a target atom, is defined with respect to the incident beam direction. The kinematic factor \( K_1 \) for the projectile equals the ratio of final and incident projectile energy. The projectile energy after the collision is represented by \( E_1 \). The kinematic factor \( K_1 \) is given by:

\[
K_1 = \frac{E_1}{E_0} = \left\{ \frac{\cos \theta + \sqrt{\mu^2 - \sin^2 \theta}}{(1 + \mu)} \right\}^2
\]  

(2.1)

where \( \mu \) equals the mass ratio \( M_2 / M_1 \). The plus sign in the numerator of equation 2.1 is considered when \( M_2 > M_1 \) or \( \mu > 1 \). In the case where \( M_2 < M_1 \), which implies \( \mu < 1 \), an additional solution should be taken into account, namely with a minus sign in the numerator.

The recoil atom obtains an energy \( E_2 \) in the collision and leaves the sample under an angle \( \phi \) with respect to the impinging beam. A similar formula is given for \( K_2 \). This is the fraction of the kinetic energy of the incident projectile, \( 1 - K_1 \), obtained by the target atom in the elastic collision. An expression for the kinematic factor \( K_2 \) is:

\[
K_2 = \frac{E_2}{E_0} = \frac{4\mu}{(1 + \mu)^2} \cos^2 \phi
\]  

(2.2)

The relation between \( \theta \) and \( \phi \), in case of the elastic collision, is [Fel 86]:

\[
\theta_{cm} = \arctan \left( \frac{-2\mu \sin \phi}{1 + \mu^2 \cos^2 \phi} \right)
\]
\[ \tan \theta = \frac{\mu \sin(2\phi)}{1-\mu \cos(2\phi)} \]  

(2.3)

The previously defined angles \( \theta \) and \( \phi \) were both considered in the laboratory frame of reference. However it is also possible, and sometimes more convenient, to define the collision in the centre-of-mass coordinates (CM). The corresponding angles in the centre-of-mass frame of reference are \( \theta_{\text{cm}} \) and \( \phi_{\text{cm}} \) and are also shown in figure 2.1. A relation between \( \theta_{\text{cm}} \) and \( \phi \), in the laboratory frame, is [Fel 86]:

\[ \theta_{\text{cm}} = 180^\circ - 2\phi \]  

(2.4)

The energy of the centre-of-mass frame (\( E_{\text{cm}} \)) is related to the energy \( E_0 \), which is the total kinetic energy of the interacting masses in the laboratory frame of reference by:

\[ E_{\text{cm}} = \frac{M_1}{M_1 + M_2} E_0 \]  

(2.5)

As a consequence an energy of \( E_0 - E_{\text{cm}} \) is left for the energy of the relative motion in the centre-of-mass frame denoted as \( E_{0\text{cm}} \). A formula for \( E_{0\text{cm}} \) is:

\[ E_{0\text{cm}} = \frac{M_2}{M_1 + M_2} E_0 \]  

(2.6)

The foregoing considered only elastic collisions. The next section presents the description of the kinematic factors when inelastic scattering takes place.

2.3 Inelastic Scattering with Energy Loss \( Q \)

An inelastic collision is characterized by a change of kinetic energy. These nuclear scattering effects appear at energies where the interacting nuclei approach each other very closely, with the distance of the order of the size of the nucleus. An energy exchange of the projectile with the target occurs, leaving the target nucleus in an excited state. The excited nucleus decays rapidly generally with the emission of \( \gamma \) rays. Energy and angular distribution of the two masses \( M_1 \) and \( M_2 \) after the interaction is therefore different compared to the elastic scattering.

Conservation of energy, in the case of excitation of the target nucleus, leads to
the energy balance: $E_0 = E_1 + E_2 + Q$, where $Q$ the kinetic energy that is lost in the inelastic collision. Excitation of the nucleus is an endothermic reaction, which implies a negative quantity for $Q$. However the Q-value for this reaction is taken as a positive quantity in the above energy balance and the following equations. The ratio of projectile energies before and after the inelastic scattering, $E_{iL}/E_{0r}$ at an angle $\theta$, is given by:

$$K_{iL} = \frac{E_{iL}}{E_{0r}} = \left(\frac{\cos \theta + \sqrt{\cos^2 \theta - (1+\mu)(1-\mu + \mu Q/E_0)}}{(1+\mu)}\right)^2 \quad (2.7)$$

where the indices $i$ refer to the inelastic scattering. The energy gained by the recoil in the inelastic collision is smaller than in the case of elastic scattering. The expression for the kinetic energy of the recoil $E_{2r}$ leaving the target at an angle $\phi$ with respect to the incident beam direction is given by:

$$K_{2r} = \frac{E_{2r}}{E_0} = \frac{\mu \cos^2 \phi}{(1+\mu)^2} \left\{1 \pm \frac{1 - (\mu+1)Q}{\mu E_0 \cos^2 \phi} \right\}^2 \quad (2.8)$$

The energy of the incident particles is $E_0$. Note that there are two possible energies for the inelastical recoil atom at an angle $\phi$. The recoil angle $\phi$ is limited for the inelastic collision of two particles with mass ratio $\mu$ and incident energy $E_0$. It is a direct consequence of the minus sign of the term under the square root of equation 2.8 where the inelastically transduced energy $Q$ is limited: $Q \leq \mu E_0 \cos^2 \phi/(1+\mu)$. An example of inelastic scattering is the interaction of a 12.6 MeV alpha particle with a carbon target nucleus.

The carbon nucleus is excited to its first excited state $Q = 4.43$ MeV. Kinematics for this process can be derived from previously mentioned equations. A closer view to the equations is given in appendix A. Figure 2.2 shows a representation of the energy of the elastically and inelastically scattered alpha projectile and the ejected recoil as a function of the scattering angle.

Note that the energy of the recoils equals the energy of inelastically scattered alpha near 30° scattering angle. The energy of the inelastically scattered alpha is always smaller than the energy of the elastically scattered alpha. This plot is used for the analysis of energy spectra obtained from mylar and carbon samples that were investigated in experiments described in chapter 4.
The energy of elastically and inelastically scattered alpha particles and the ejected recoil as a function of detection angle $\theta$ and $\phi$. Angles are in degrees. The energy of the incident alpha particles is $E_0 = 12.6$ MeV.

2.4 Differential Scattering Cross Section $d\sigma/d\Omega$

The differential scattering cross section $d\sigma/d\Omega$ is the probability per unit solid angle that an incident particle is scattered into the solid angle $d\Omega$. To calculate the differential cross section for an elastic collision, the principles of conservation of energy and momentum must be taken into account as well as a specific interaction during the collision between the projectile $M_1$ and a target mass $M_2$. Taking the Coulomb repulsion for the force between the two interacting nuclei, the following equation, valid for the laboratory frame of reference, can be found for the scattering cross section [Chu 78]:

$$
\frac{d\sigma}{d\Omega} = \left( \frac{Z_1 Z_2 e^2}{4\pi\varepsilon_0 E_0^2} \right)^2 \frac{4}{\sin^4 \theta} \frac{\left\{ [1-((M_1/M_2)\sin\theta)^2]^{1/2} + \cos \theta \right\}^2}{[1-((M_1/M_2)\sin\theta)^2]^{1/2}}
$$

(2.9)

where $(e^2/4\pi\varepsilon_0 E_0^2)^2 = 0.02$ barn for $E_0 = 1$ MeV. The order of magnitude of this differential scattering cross section is predominantly given by the first factor. Note that the differential scattering cross section for Rutherford scattering is inversely proportional with the incident particle energy squared. It is decreasing for increasing scattering angle $\theta$. 
The Coulomb repulsion should only be used for low energy projectiles, that is as long as the distance of closest approach is large compared to nuclear dimensions, but small compared to the Bohr radius $a_0 = 0.53\text{Å}$. At large energies a departure from the classical Coulomb scattering has to be taken into account. The decrease of the scattering cross section is with increasing $\alpha$-particle energy, however the cross section does not decrease monotonously but shows strong fluctuations, due to nuclear resonances as discussed below.

2.4.1 Resonant Scattering

Maxima and minima of the elastic scattering cross section were found as a function of energy by early experiments of Chadwick and Bieler (1921) and Rutherford and Chadwick (1927). Results and theories based on these results are due to resonance phenomena of interacting nuclei. When the approach of the projectile to the scattering nucleus is near the nuclear dimensions an interaction function different from the Coulomb repulsion should be taken. In the case of resonance scattering wave functions of incident projectile and target nucleus have to be considered. A nuclear reaction may involve a direct transfer of particles or lead to the formation of a compound nucleus. The nucleus can be excited into an unbound or a bound nuclear state. The discrete states with definite wave functions are treated differently from the regime with continuum states, so numerous that their spacing is much less than the width of the individual states. Between these two regimes is the resonance region. Figure 2.3 shows the differential scattering cross section for elastic scattering of protons from aluminum as a function of energy of the incident proton beam.

![Figure 2.3](image)

**Figure 2.3:** The differential scattering cross section as a function of energy for the elastic scattering of protons from an aluminum target. The charged particle yields were taken at an angle of 164° with respect to the incident proton beam.

A resonance is a consequence of the existence of discrete levels in the compound nucleus region. These levels have a high probability of formation and the scattering cross section decreases dramatically at that energy. In order to obtain qualitative understanding of the formation of resonances a nuclear potential is
introduced, which has in first approximation a square well form. Projectile and nucleus are described by their wave functions. The oscillatory wave functions inside and outside the well must be matched smoothly. If amplitudes of the exterior and interior wave functions match exactly we have a resonance; the cross section for capture has a maximum and a compound nucleus is formed. When the energy of the incident particle is varied; the relative phase, the location of the matching point and the relative amplitudes of inner and outer wave function vary accordingly. Figure 2.4 shows three different cases for scattering of a projectile from a nucleus. A resonance corresponds to a greatly enhanced transmission of the incident particle across the nuclear boundary into the interior of the nucleus with radius, \( r = a \). The nuclear radius \( a \) is denoted in Figure 2.4. Note that the probability to find the particle inside the nucleus is given by the square of the wave function (amplitude).

The incident projectile is first captured by the target nucleus to form a quasi-stationary state of the compound nucleus. This state subsequently decays by the re-emission of the particle or by \( \gamma \)-radiation. The probability of the capture of the incident particle varies in a characteristic manner with the difference between the energy \( E_{\text{com}} \) of the colliding system and the energy \( E_R \) of the resonance level. The total cross section of the process is proportional to the capture probability:

\[
\sigma_{\text{res}} = \pi \bar{\lambda} (2l + 1) \frac{\Gamma_{\text{tot}}^2}{(E - E_R)^2 + (\Gamma/2)^2} \tag{2.10}
\]

where \( l \) the specific angular momentum of the considered partial wave and \( \bar{\lambda} = \)
\( \lambda/2\pi \), the reduced de Broglie wavelength. The characteristic width \( \Gamma = \hbar / \tau \) is related to the lifetime \( \tau \) of the compound nucleus state by the Heisenberg uncertainty principle. Equation 2.10 represents a Lorentzian shape line broadening. The above equation is well known as the Breit-Wigner resonance formula. It has a maximum value at the energy \( E_R \), the resonance energy, and falls to half maximum at \( E = E_R \pm \Gamma / 2 \).

For a single isolated resonance, the elastic scattering cross section has a remarkable shape. This originates from another contribution to the reaction amplitude namely from direct scattering of the incident particle by the nuclear potential, without forming the resonant state. This alternative process is called potential scattering or shape elastic scattering. An expression for the total elastic scattering cross section taking into account both processes is therefore:

\[
\sigma_{el} = \frac{\pi}{k^2} \left[ \exp(2ika) - 1 \right] + \frac{i\Gamma}{(E - E_R) + \frac{i\Gamma}{2}} \]  

where the first term in brackets is due to direct scattering without formation of an intermediate compound nucleus, the potential or shape-elastic scattering amplitude. The second term is the resonance or compound-elastic scattering amplitude. In equation 2.11, \( k \) is the wave vector of the incident particle and \( a \) is the nuclear radius for an impenetrable sphere. If the bombarding energy is off-resonance so that \( |E - E_R| >> \Gamma \), then the resonant elastic cross section is negligible compared to the non-resonant term and the behaviour of the nucleus is nearly classical.

Both potential scattering and resonant scattering contribute to the elastic scattering amplitude, and interference between the two processes causes variation in cross section. Near the resonance there is an interference between the two terms which produces the characteristic shape shown in figure 2.5.

**Figure 2.5:** Interference between resonance and potential amplitudes at an isolated resonance produces resonances with this characteristic shape for elastic scattering. (From [Sat 90]).
According to this model an interference dip is expected on the low energy side of the resonance. The resonance height should decrease roughly as $k^2$ (that is $E^{-1}$) with increasing incident energy. The cross sections for scattering and reactions involving charged particles may exhibit resonances. The repulsive Coulomb potential affects the value of $\sigma_{el}$. The Rutherford scattering amplitude must be added to the reaction amplitude for the elastic scattering. In case of a continuum of nuclear excited states, where different states can not be distinguished separately, a broad resonance region will occur.

2.4.2 The Optical Model

The scattering cross section for elastic scattering is also a function of scattering angle in case of high energy scattering. The angular distribution shows the oscillatory nature of the differential cross section for alpha particles elastically scattered from light nuclei. The elastic nuclear scattering bears strong resemblance to a familiar problem from optics, the diffraction of light. The optical model gives a description of the elastic scattering in the presence of absorptive effects. Fluctuations in the cross-section which correspond to interference minima and maxima were found and it is natural to describe these fluctuations by representing the effect of the target nucleus on the incident projectile by a potential well.

The optical model starts with the Plane Wave Born Approximation (PWBA) for the scattering from a square well. In this model the scattering is represented in terms of a complex potential. The real part of the potential is responsible for elastic scattering, the imaginary part for absorption, or rather formation of the compound nucleus with subsequent disintegration. The potential holds within the nucleus; for radii $r<R$, where $R$ is the interaction radius. The potential form is chosen as:

$$U(r) = \frac{-V_0}{1+\exp((r-R)/a)} + iW(r)$$

(2.12)

where $W(r)$ is often chosen as proportional to $dV/dr$, which has the proper shape being large only near the surface. Finally a Coulomb term $V_c(r)$ must be included if the incident particle is charged. For the chosen potential the Schrödinger equation can be solved. The Born approximation is expanded to the Distorted Wave Born Approximation, usually abbreviated to DWBA. As the name implies, the relative motions before and after the non-elastic event are described not by plane waves but by waves distorted by elastic scattering and its accompanying absorption. Hence this approximation is likely to be valid if the most important single event to take place is elastic scattering. Then other reactions can be treated as perturbations, as weak transitions between elastic states.
The relative motion of two interacting nuclei, before and after the collision, has been approximated by plane waves. A simple scattering process familiar from classical physics is the diffraction of waves at the edges of opaque objects. One approximation for the minima in the diffraction pattern is given by the Fraunhöfer diffraction. The minima of the differential scattering cross section for elastic scattering is then given by:

\[
\frac{d\sigma}{d\Omega} = k^2R^4 \left[ \frac{J_1(2qR \sin(\theta_{cm}/2))}{2qR \sin(\theta_{cm}/2)} \right]^2
\]  

(2.13)

where, \( J_1 \) is the spherical Bessel function of first order, \( q \) the magnitude of the difference between the initial and final wave vectors, \( k \) and \( k' \) respectively. The magnitude \( q \) follows from cosine rule: \( q^2 = k^2 + k'^2 - 2kk'\cos\theta_{cm} \) or \( q = 2ksin(\theta/2) = k\theta \) if \( \theta \) is small. The interaction radius is \( R \). The scattering angle in the centre of mass system is \( \theta_{cm} \). For light elements, the elastically scattered alpha particles are always accompanied by alpha groups due to inelastic scattering [Sei 58]. Figure 2.6 shows the angular dependence of the scattering cross section for 18 MeV and 18.5 MeV alpha particles, elastically scattered from carbon. A diffraction-like pattern is evidently visible. Note that the angle on the x-axis is the centre of mass angle \( \theta_{cm} \).

\[ C^{12}(\alpha,\alpha)C^{12} \]

\(-\cdot-\ E_\alpha = 18.0\ MeV\)
\(-\circ-\ E_\alpha = 18.5\ MeV\)

**Figure 2.6:** The differential scattering cross section versus the centre of mass angle \( \theta_{cm} \). Elastic scattering of 18 MeV and 18.5 MeV alpha particles by carbon. (From [Cor 59]).
2.5 Stopping of Ions in Matter

An energetic particle that impinges on a target will penetrate into it and might scatter from the target nuclei. It will slow down as a consequence of interaction with the target material on its passing through the electron clouds surrounding each target nucleus and is therefore called electronic stopping. The nuclear stopping originates from the multitude of small angle scattering collisions of the projectile with the atomic nuclei of the target and can be neglected for ion energies beyond the maximum of the stopping power curve [Chu 78]. The contribution of nuclear stopping to the total stopping power is smaller than one percent in that energy region.

The stopping power, \( S \), is defined as the energy loss \( \Delta E \) per distance \( \Delta x \) and depends on the density of matter and the charge of the incident particle. Measurements of energy loss per unit length have been done for many projectile atoms. Usually the Ziegler tables for stopping in matter are used [Zie 85]. Recent results on the determination of the stopping of carbon ions in a carbon target were presented by Abdesselam et al. [Abd 91]. Figure 2.7 shows the electronic stopping power for carbon ions in a carbon medium. Different theories and experimental results are represented.

![Figure 2.7: The stopping of carbon ions in a carbon target. (From [Abd 91]).](image)

From the experimental data the stopping power for ions with a kinetic energy \( E \) is known for different particles. Generally the energy at depth \( x \), \( E(x) \), can be calculated
from these data by:

\[ E(x) = E_0 - \int_0^x (dE/dx) dx \]  

(2.14)

However it is more convenient to use an approximation for the energy loss in matter. The first order approximation is the surface approximation. A particle with incident on a target with an energy \( E_0 \) has a stopping power \( S(E_0) \). After penetrating a depth \( x \), the energy has decreased till: \( E(x) = E_0 - xS(E_0) \).

Note that the energy loss has a positive quantity. The derivative, with respect to the energy, of the stopping power \( (dS/dE) \) is negative for energies beyond the maximum of the stopping curve and positive for lower energies. This can be concluded from the general shape of the stopping as a function of energy according to figure 2.7. At the value of approximately 0.2 MeV/amu the stopping power attains a maximum. The general form of the electronic stopping can be approximated by the Bethe Bloch formula, which is valid in the energy region beyond the maximum of the \( dE/dx \) curve:

\[ (dE/dx)_e = N Z_2 [4\pi Z_2^2 e^4 / m_e v_1^2] L \]  

(2.15)

where \( N \) is the atom density in the target, the electron mass is \( m_e \), the velocity of the incident projectile \( v_1 \) and the atomic numbers \( Z_2 \) of the target atoms and \( Z_1 \) of the incident particles. The stopping number \( L \) is defined as \( L = \ln(2m_e v_1^2 / I) \) according to quantum-mechanical calculations of Bethe, where \( I \) is an average over the various excitations and ionizations of the electrons in the target atom. Exact calculations of this mean excitation potential are difficult to perform. Bloch showed that \( I \) is approximately proportional to \( Z_2 \), that is \( I = K Z_2 \) where \( K \) equals 10 eV.

### 2.6 Energy Straggling

Energy straggling is the statistical fluctuation in the energy loss of an energetic particle that passes through a medium with electronic stopping power \( S \). As a result, identical energetic particles, which all have the same initial velocity, do not have exactly the same energy after passing through a homogeneous medium of thickness \( x \). A formula for energy straggling was derived by Bohr (1915). Bohr's theory is based on fully stripped fast ions and the free electron gas model. For a layer of thickness \( x \), Bohr straggling produces a variance of:

\[ \Omega_b^2 = 4\pi e^4 Z_1^2 Z_2 N x \]  

(2.16)

where \( Z_1 \) is the atomic number of the projectile, \( Z_2 \) that of the target element, \( N \) the
atomic density, x the thickness of the target and e the elementary charge.

In the practical case of a Gaussian distribution the Full Width at Half Maximum (FWHM) is $2.35 \Delta B$. The assumed Gaussian distribution is a consequence of the assumption that the number of collisions is large and follows a Poisson distribution. For small energies the contribution of the real straggling is smaller than the Bohr straggling. Empirical formulae for energy loss straggling of ions in matter have recently been published by Yang and O'Connor [Yan 91]. The experimentally obtained data confirm the Bohr straggling as a good approximation for the energy region beyond 0.2 MeV/amu in a carbon target.

2.7 Multiple Scattering

Multiple scattering leads to an energy spread via small angle scattering from target nuclei. An ion beam will obtain an angular spread when the ions traverse a trajectory in the target. The effect should be distinguished from straggling of ions in matter. In case of a Thomas-Fermi-type screened Coulomb interaction of the ions with the target atoms the differential cross sections for small angle scattering are calculated by Lindhard et al [Lin 68]. In the theory of multiple scattering the following assumptions that are made:
- random homogeneous distribution of scattering in space,
- binary collision events with azimuthal symmetry of scattering,
- neglect of all energy loss,
- small angles.

The Half Width at Half Maximum HWHM in the angular spread arising from multiple scattering after traversing a trajectory through a medium is:

$$\alpha_{1/2} = \frac{180^\circ}{\pi} \Gamma g_1(\tau)$$  \hspace{1cm} \text{(2.17)}

where $\Gamma$ in rad, the values of $g_1(\tau)$ are tabulated by Meyer [Mey 71] and Sigmund and Winterbon [Sig 74] and $\tau$ is the reduced path length $\tau = \pi a^2 N x$, which is an effective area per unit of area. The atomic density $N$ of the target material in at/cm$^3$. The Thomas Fermi radius $a$, and trajectory length $x$, are the variables in the equation for the reduced path length and have both the dimension cm. Thomas and Fermi derived a formula for the effective interaction radius for small angular scattering:

$$a = 0.885 \frac{a_0}{(Z_{\text{pro}}^{2/3} + Z_{\text{targ}}^{2/3})^{1/2}}$$  \hspace{1cm} \text{(2.18)}

where $a_0$ the Bohr radius ($a_0=0.529*10^{-8}$ cm), $Z_{\text{pro}}$ and $Z_{\text{targ}}$ are atomic numbers of the
projectile and target atom respectively.

The $\Gamma$ in equation 2.17 is calculated from:

$$\Gamma = 2Z_{\text{pro}} Z_{\text{targ}} e^2 / E_a$$  \hspace{1cm} (2.19)

where $E$ is the projectile energy in ergs ($1 \text{ J} = 10^7 \text{ ergs}$) and $e$ the elementary charge in Statcoul ($e = 4.8 \times 10^{-10} \text{ Statcoul}$). The magnitude of gamma is inversely proportional with the projectile energy.

The Thomas Fermi radius $a$ is only dependent on the atomic numbers of the ions and the target atoms. A formula for $\tau$ in SI units is:

$$\tau = 13.20 \pi \rho^*/(M(Z_{\text{pro}}^{2/3} + Z_{\text{targ}}^{2/3}))$$  \hspace{1cm} (2.20)

where $\rho^*$ is the distance in $\mu g/cm^2$ and $M$ the atomic mass of the target atoms in amu. Note that $\rho^* = \rho x$, where $\rho$ the target density ($\mu g/cm^2$) and $x$ the distance in cm. Equation 2.20 can be rewritten to an equation with the parameters in convenient dimensions:

$$\Gamma = 6.15 \times 10^{-5} Z_{\text{pro}} Z_{\text{targ}} \frac{\sqrt{Z_{\text{pro}}^{2/3} Z_{\text{targ}}^{2/3}}}{E}$$  \hspace{1cm} (2.21)

where $\Gamma$ is in rad and $E$ is the energy of the projectile in MeV.

### 2.8 Other Contributions to Energy Spread

If both energy straggling and multiple scattering are taken into account it should be noticed that the variation in film thickness or surface roughness gives another significant contribution to the variance in ion energy. Energy straggling and multiple scattering are fundamental contribution to the energy variance resulting from interaction with the target. On the contrary variation of target thickness and surface roughness are sample dependent parameters. The total measured energy variance squared, $dE^2$, is the sum of various contributions squared according to Poisson statistics. The contribution in the energy variance $\Delta E$, due to target surface roughness $\Delta x$ is:

$$\Delta E = \Delta x S(E)$$  \hspace{1cm} (2.22)

where $S(E)$ is the stopping power in the medium.
CHAPTER 3

The Atomic Hydrogen to Carbon Ratio in Amorphous Carbon Layers

In this chapter a method to determine the hydrogen and carbon content in an amorphous carbon film deposited on a 10.6 μm aluminum foil will be described. These amorphous carbon films are produced by means of plasma deposition. The composition of the obtained "diamond like" carbon film depends on the plasma parameters during deposition. The used plasma contains about 10% electrons and argon ions used as a carrier for the monomer CH₄. The argon ions dissociate and ionize the hydro-carbon through charge transfer and subsequent dissociative recombination reactions [Beu 92]. Both the produced carbon containing radicals and ions are transported towards a substrate, by means of expansion, where they are deposited on an aluminum carrier. Different samples were made using different reactor settings. Plasma parameters that were varied are the ratio of the flow of hydrocarbon to the argon flow, the kind of organic gas, C₂H₂ or CH₄, and the admixture of oxygen and hydrogen to the plasma. During the deposition single wavelength ellipsometry was performed to determine the diffractive index of the growing film which is a quantity that is related with the amount of hydrogen in the deposited film.

A combination of two different ion beam analysis techniques was used for the analysis, Rutherford Backscattering Spectrometry (RBS) to determine both the carbon and the oxygen content and Elastic Recoil Detection Analysis (ERDA) to determine the hydrogen content. RBS was performed because of a good separation of the peaks due to elastic collisions from carbon and oxygen. Protons were used as incident projectiles because they penetrate the aluminum carrier more easy than alpha particles. Spectra were simultaneously measured with two detectors positioned at a 165° and 45° with respect to the incident beam direction.

In order to obtain a comparison of the samples with a sample of known composition a standard was needed. A mylar foil (C₁₀H₈O₄) located on an aluminum carrier foil was used as a standard for the hydrogen to carbon ratio. The thickness of both the mylar and the aluminum foil were obtained from an experiment with an AmCm alpha source. The thicknesses were determined from the energy loss of the alpha particles with use of the electronic stopping powers obtained from TRIM. The determined thicknesses were 10.6 ± 0.2 μm for the aluminum carrier foil and 3.3 ± 0.1 μm for the mylar foil. From the mylar standard a hydrogen to carbon ratio H/C was obtained but also a quantity for the absolute number of carbon and hydrogen and oxygen atoms in the target. The deposited carbon layers that were analyzed had a thickness of 0.2 - 1 μm depending on the used deposition parameters. In order to
make corrections to the peak contents, a background subtraction procedure was executed. These background spectra were obtained from the 10.6 µm aluminum clean film and is described in section 3.7.

### 3.1 Experimental Setup

The experiments were carried out in a 60 cm diameter scattering chamber with the sample located in the centre. The vacuum during the experiment was better than $5 \times 10^{-5}$ mbar. A 3 MeV proton beam impinges on the target. The detectors used for measurement of the energy of scattered particles are Canberra Planar Implanted Passivated Silicon (PIPS) detectors with an energy resolution of 15 keV. The detectors are positioned at a 15 cm distance, with respect to the centre of the chamber. The RBS detector is located at a 165° angle, the ERD detector at 45°. Both angles are defined with respect to the incident beam direction. The detector acceptance angles are 20 mrad in the scattering plane. Detector signals are amplified by Silena pre-amplifiers and ORTEC main amplifiers. The signals are converted in a Canberra ADC. The digitized data is offered to an MCA. Figure 3.1 shows the scattering geometry.

**Figure 3.1:** The scattering geometry for combined RBS and ERDA. A 3 MeV proton beam impinges on the sample. The forward and backscattering angles are 45° and 165° with respect to the incident beam. Detector distances are 15 cm and acceptance angles are 20 mrad. The hydrogen content of the layer positioned at the back side of the aluminum is determined.

The orientation of the deposited sample is chosen in such a way that the beam hits the aluminum foil first. This geometry is favourable because the peaks in the RBS spectrum due to elastic scattering from carbon and oxygen are separated from the broad spectrum due to elastic scattering of protons from the aluminum carrier. The incident protons lose 240 keV in the aluminum foil due to electronic stopping. The carbon and oxygen content are derived from the C(p,p)C and O(p,p)O peak content in the RBS spectrum. The hydrogen content of the "diamond like" carbon film is derived from the peak content of the H(p,p)H peak in the 45° spectrum.
3.2 Evaluation of the RBS Spectrum

An example of the RBS spectrum is given in this section. The experiment is carried out with a sample consisting of a "diamond like" carbon film on an aluminum carrier foil. The broad peak in the 165° spectrum, denoted as Al(p,p)Al is the elastic proton backscattering distribution, showing a distinct resonant structure. The peak occurring from elastic scattering from carbon is denoted as C(p,p)C and is located at an energy of 1500 keV.

![RBS Spectrum](image)

**Figure 3.2:** The RBS spectrum obtained from a "diamond like" carbon film on a 10.6 μm aluminum carrier.

The incident 3 MeV protons scatter from a target nucleus at a certain depth. A proton, scattered from an aluminum target atom on the front surface, loses an energy in the aluminum due to electronic stopping (22.6 keV/μm). Protons scattered from the reverse side of the aluminum already lost 240 keV of kinetic energy due to electronic stopping even before scattering. Elastically from aluminum scattered protons lose another 287 keV due to electronic stopping. From the RBS spectrum is seen that the low energy edge of the Al(p,p)Al edge is located at an energy of 2100 keV. The variation in the yield of the aluminum peak is a consequence of the energy dependence of the differential scattering cross section for elastic scattering of protons from aluminum. Section 3.3 will focus on this peak shape.

A second remarkable peak in the 165° RBS spectrum is the one with a high energy edge of approximately 1500 keV. This peak is due to elastically from carbon scattered protons denoted as C(p,p)C. This total peak content is a measure for the
amount of carbon in the "diamond like" film on the aluminum layer. However there are problems when the peak content is determined because the C(p,p)C peak is founded on a background which is due to inelastically scattered protons from aluminum, hydrogen in the aluminum or impurities in the sample. There is only one way to determine this contribution of inelastically scattered protons. The proper way to do this is to obtain the same spectra from an aluminum clean foil, without the deposited carbon film, and to subtract that spectrum from the spectrum of the sample. This method is described in section 3.8.

3.3 Determination of the Incident Proton Energy

The scattering of 3 MeV protons from aluminum is non-Rutherford as has previously been mentioned in chapter 2. This implies that the differential scattering cross section for elastic scattering varies strongly with energy and shows a resonant structure for different energies. The differential scattering cross section was determined for elastically from aluminum, under 165° scattered, protons with incident energies between 1.4 and 4 MeV by Shoemaker [Sho 51]. A figure of the scattering cross section of protons scattered from aluminum has previously been shown in chapter 2. The resonant structure of the Al(p,p)Al peak in the obtained 165° spectrum agrees with the resonant structure found by Shoemaker.

The analysis of the different samples was done on different days and the RBS spectra obtained on these different days show a different peak shape for the elastically from aluminum scattered protons. This is a consequence of the slightly different energy of the incident protons. Figure 3.3 shows two, on a different day, obtained spectra.

Figure 3.3: Two RBS spectra obtained on a different day. Note that the peak shape, due to elastic scattering of protons from aluminum Al(p,p)Al differ.

The thickness of the aluminum foil is 10.6 ± 0.2μm. The highest peak in the resonant structure is due to the elastic scattering of a 2874 keV proton, over 165°, from aluminum, according to Shoemaker. With use of the electronic stopping power for
protons in aluminum [TRIM 89] it is possible to calculate the incident beam energy as well as to obtain a calibration for the channel to energy conversion.

The stopping power for 2.8 MeV protons in aluminum is 23.8 keV/μm. The thickness at which elastic scattering took place is 4.5 ± 0.2 μm for spectrum A and 4.1 ± 0.2 μm for spectrum B according to the spectra in figure 3.3. The energy $E_0$ of the incident protons is $E_0 = E_{res} + Sx$, where $E_{res}$ the resonant energy, $S$ the mean stopping power at the trajectory $x$ which is the covered distance of the incident proton before the resonant scattering.

Hence it follows that the incident proton energy is $3.0 \pm 0.3$ MeV. The mean stopping power on the trajectory $x$ is $22.9 \pm 0.2$ keV, according to TRIM. It is concluded that the incident beam energy was $9.2 \pm 0.1$ keV lower on the second day.

3.4 Evaluation of the 45° Spectrum

The dominating feature in the 45° spectrum, shown in figure 3.4, is the elastically from aluminum scattered proton distribution located at 2350 keV. The yield in the region between 500 keV and 1900 keV is blown up with a factor of 20. It shows the elastically from hydrogen scattered protons, denoted as $H(p,p)H$, and is therefore a measure for the hydrogen content of the "diamond like" film. This peak also contains inelastically from aluminum scattered protons and protons scattered from eventually present hydrogen in the aluminum. Therefore a background subtraction procedure is carried out in order to determine the proper hydrogen content of the carbon film. The energy of the protons scattered at 45° is smaller then half of the incident beam energy because the incident protons lose 240 keV of kinetic energy, due to electronic stopping in the aluminum foil.

Figure 3.4: The 45° spectrum obtained from a carbon layer on a 10.6 μm aluminum carrier.
The observed energy spread in the elastically scattered protons denoted as H(p,p)H, contains several contributions; detector energy resolution, energy spread occurring from finite beam spot size, the detector acceptance angle, energy spread of the incident protons, divergence of the incident proton beam, energy spread occurring from energy straggling and multiple scattering of protons in the target. The detector energy resolution was 15 keV. The contribution of an acceptance angle in the scattering plane is 53 keV/° for protons scattered at the 45° scattering angle. The energy resolution of the incoming beam is approximately 9 keV which is 0.3% of the incident beam energy. The divergence of the incident beam had already a magnitude of 1°, and gives a contribution of 53 keV. Energy straggling of the beam in the aluminum foil is 35 keV according to Bohr straggling and the contribution. Multiple scattering causes an extra beam divergence in the aluminum of 2°. This results in an energy broadening of 105 keV. The total energy resolution is found by adding the variances of different contributions and equals 140 keV. With this technique it is impossible to obtain a hydrogen depth profile of the carbon film on the aluminum target because the stopping power for 1.4 MeV protons in a carbon is 42 keV/μm which implies a depth resolution of 3 μm when the previously calculated energy spread of forward scattered protons is assumed.

3.5 The Standard, Mylar on an Aluminum Carrier Foil

A 3.3 μm mylar (C_{10}H_{8}O_{4}) foil on a 10.6 μm aluminum carrier foil was taken as a standard for the determination of the hydrogen to carbon ratio from the ratio of the H(p,p)H peak content in the ERDA spectrum and the C(p,p)C peak contents in the RBS spectrum. The same can be done with the oxygen to carbon ratio. However a background subtraction procedure should first be carried out. This is described in section 3.7. First a closer view is given to the mylar spectra. The RBS and ERDA spectra obtained from the mylar standard on the aluminum carrier foil are shown in figure 3.5.

![Figure 3.5: ERDA and RBS spectra from 3.3 μm mylar foil on a 10.6 μm aluminum carrier.](image-url)
The peak contents are determined after execution of the background subtraction procedure, which makes corrections for the background at the location of the peaks of interest. The obtained hydrogen to carbon ratio corresponds to an atomic ratio of 0.8. The same procedure is carried out for the determination of the oxygen to carbon ratio. That obtained peak ratio equals an atomic ratio of 0.4.

3.6 Energy Spectra Obtained from an Aluminum Foil

In order to improve the determination of both the carbon and hydrogen peak contents, spectra were obtained from a clean, non-deposited aluminum foil. This aluminum foil is the same standard aluminum used as a carrier for the "diamond like" carbon films. The ERDA and RBS spectra are used for background subtraction for all spectra obtained from experiments on the samples with deposited carbon films. Figure 3.6 shows the ERDA and RBS spectra obtained from the aluminum foil.

![RBS and ERDA spectra obtained from a clean aluminum foil.](image)

The RBS spectrum shows the expected resonant structure of the Al(p,p)Al peak but even more important is the background in the region between 700 keV and 1550 keV is due to inelastic scattering of protons from aluminum nuclei. This reaction is denoted Al(p,p')Al. Protons that were scattered from inhomogeneities might also occur in this energy region. The ERDA spectrum shows the dominating Al(p,p)Al peak originating from elastically forward scattered protons from aluminum but it also shows a broad peak between 1100 keV and 1550 keV arising from elastic proton-hydrogen scattering or inelastic scattering of protons from aluminum and particles scattered from inhomogeneities.

3.7 The Background Subtraction Procedure

In order to normalize all spectra with respect to total transferred charge, the peak contents of the Al(p,p)Al peaks are determined from the RBS spectra obtained from all samples. The aluminum carrier foil is the only resemblance of all analyzed...
samples. The peak content is proportional to the number of incident protons and the differential scattering cross section. The scattering cross section for Al(p,p)Al does not vary during the experiment and the number of aluminum target atoms does not decrease. Therefore the total amount of elastically scattered protons from aluminum is a measure for the beam current and consequently for the number of protons that impinged on the sample.

A F-factor is introduced, equalling the ratio of the Al(p,p)Al peak contents of the sample and the Al(p,p)Al peak content in the background. The remaining RBS spectrum after background subtraction shows the, from the carbon film scattered, protons. The remaining subtracted spectrum is represented by:

$$\text{Sample'} = \text{Sample} - F \cdot \text{Bckgnd}$$

The subtraction is done to both ERDA and RBS spectra of the sample using the same F-factor which is allowed because the RBS and ERDA spectra were simultaneously taken. After the background subtraction procedure is carried out, peak contents of the remaining spectra were determined with use of a computer program. This program calculated the peak contents in a given region of interest. The margin of error in the peak content is defined as the square root of the total amount of counts. The variance in the peak ratio $dN_{\text{SUBTR}}$ is determined by:

$$dN_{\text{SUBTR}}^2 = dN_{\text{PEAK}}^2 + 2dN_{\text{BACK}}^2$$

where $dN_{\text{PEAK}}$ is the square root of the total peak content and $dN_{\text{BACK}}$ the variance in the background.

The next step is to determine the peak ratios H/C and O/C, the hydrogen to carbon ratio and oxygen to carbon ratio, respectively from the remaining ERDA and RBS spectrum. The obtained peak ratios do not equal the atomic ratios. The conversion factors from peak ratio to atomic ratio must be determined when these are desired.

In order to determine the atomic ratios from peak contents two ways are available. One is to divide by cross section ratios determined from literature, the other to compare with a sample of known composition. Literature cross sections, determined in nuclear physics experiments, are sensitive to small energy variations and the accuracies of about 5 - 20% due to insufficient sample standardization. It was judged better not to compare to some one else's sample at a probably different energy and use fresh standards at the identical energy. The comparison to a standard should be done on every day when new spectra are obtained because the beam energy might differ for a different day (section 3.3) which might result in different conversion factors due to different scattering cross sections for these different energies. One demand for the standard is that it does not suffer from composition changes during the experiment.
3.8 Background Subtraction Procedure for the Mylar Standard

The standard that was taken in the experiments was mylar \( (C_{10}H_{8}O_4) \) on an aluminum carrier. The mylar thickness was determined in an experiment with use of a AmCm alpha source. From the electronic stopping of the alpha particles was derived that the thickness equals 3.3 \( \mu \text{m} \). From the density of mylar, 1.397 g/cm\(^2\), and the thickness, the number of hydrogen, carbon and oxygen atoms is calculated. The number of carbon, hydrogen and oxygen are: \( 14.5 \times 10^{18} \) at/cm\(^2\), \( 11.6 \times 10^{18} \) at/cm\(^2\) and \( 5.8 \times 10^{18} \) at/cm\(^2\).

With the use of the standard an absolute determination of the present atoms can be obtained. The Al(p,p)Al peak is an indicator of the total current and the peak contents of the carbon, hydrogen and oxygen originate from scattering from a known amount of atoms.

Figure 3.7 shows the spectra involved in the determination of the hydrogen, carbon, and oxygen content in the mylar standard. The beam current during each experiment, which took 250 seconds, was approximately 15 nA. The peak ratios are also determined from the obtained spectra. Both the ERDA and RBS spectra from the mylar on the aluminum carrier as well as the background spectra of the clean, non deposited, aluminum are shown. The ERDA spectra are located at the left and the RBS spectra at the right side. The aluminum background spectrum is multiplied by a F-factor equalling 1.01, which is the correction factor for the amount of impinged protons.

Note that the Al(p,p)Al peak in the 45° ERDA mylar spectrum is shifted to lower energy, compared to the Al(p,p)Al peak in the aluminum spectrum. This is due to electronic stopping of the protons in the mylar and causes an energy shift to lower energy. The extra energy loss of the protons is a measure for the thickness of the mylar. The low energy part of this peak also contains the elastically from carbon and oxygen scattered protons which explains the peak being broader and higher than the
Al\((p,p)\)Al peak in the ERDA spectrum obtained from the clean aluminum carrier. The background subtraction procedure results in the spectra shown in figure 3.8.

**Figure 3.8:** The RBS and ERDA spectra, after background subtraction, for the mylar standard.

In the subtracted ERDA spectrum the hydrogen peak due to elastic scattering, \(H(p,p)H\), is clearly visible and should be free from unwanted background. The hydrogen peak has a negative yield at the right side of the remained peak. This results from subtraction of shifted peaks. These negative values should be taken into account when the peak ratio is determined. Therefore a computer program is written to take these negative values into account because the background subtraction procedure of the Canberra MCA, used in the experiment, can not handle negative yield values. The oscillation in the high energy peak is of course due to the subtraction of two shifted peaks in figure 3.7.

The 165° RBS spectrum remaining after background subtraction shows the two peaks due to elastic scattering from carbon and oxygen, denoted as \(C(p,p)C\) and \(O(p,p)O\) respectively. The energy region where the peak due to elastic scattering from aluminum was located shows an oscillating structure which is due to statistical fluctuations in the count rate during the experiment.

The determined peak contents correspond to a known amount of target atoms, because the thickness of the mylar foil is known. Therefore the concentrations of carbon, oxygen and hydrogen are known in the samples.
3.9 Results

In this section the results of the analysis of the carbon deposited aluminum samples will be presented. The reproducibility of the aluminum background, mylar standard and a carbon deposited sample (sample 4) are discussed and the accuracy of the technique is evaluated. The thicknesses of the aluminum foil, used as a carrier, and the mylar foil were determined in a previous experiment with use of an AmCm alpha source. The obtained thicknesses were 10.6 ± 0.2 µm for the aluminum foil and 3.3 ± 0.1 µm for the mylar foil.

3.9.1 Reproducibility for an Aluminum Clean Foil

Spectra obtained from the clean aluminum carrier foil were several times repeated in order to prove the reproducibility of both the RBS and ERDA spectra. An indication for the total impinged charge, and therefore the total amount of incident particles, is the Al(p,p)Al peak in the 165° RBS spectrum. The high energy edge of this Al(p,p)Al peak was taken as an indicator for the stability of the detector and electronics. A shifted peak indicates instable or drifting electronics. These high energy edges are expressed by the channel number of the MCA and were the same for each experiment which indicates reliable electronics. Each spectrum was collected for 250 seconds. The beam current was approximately 15 nA. Figure 3.9 shows the Al(p,p)Al peak content as a function of the time of the day.

![Figure 3.9](image_url)  

Figure 3.9: Determination of the Al(p,p)Al peak contents in the 165° RBS spectra for different time of the day.

Figure 3.9 shows a significant increase in the beam current at a certain time resulting from an optimization of the cyclotron settings.
3.9.2 Reproducibility for the Standard

In order to obtain a measure for the reproducibility of spectra acquired from the mylar standard, seven measurements were carried out on two different days and compared. The peak contents of the elastically backscattered protons from aluminum, Al(p,p)Al, were determined for every measurement. Furthermore the background subtraction procedure was carried out for all measurements. For every measurement the peak contents after background subtraction were determined and the amount of carbon, oxygen and hydrogen atoms was calculated. The results are shown in table 3.1.

Table 3.1: The number of atoms in the mylar foil calculated from the RBS and ERDA spectra. All values are in $10^{18}$ at/cm$^2$. The tabulated accuracies originate from the experimental statistics. The measurements indicated with an asterisk (*) are obtained on a different day than the other results.

<table>
<thead>
<tr>
<th># carbon</th>
<th># hydrogen</th>
<th># oxygen</th>
<th>H/C</th>
<th>O/C</th>
</tr>
</thead>
<tbody>
<tr>
<td>14.3 ± 0.2</td>
<td>5.7 ± 0.1</td>
<td>11.36 ± 0.1</td>
<td>0.79 ± 0.01</td>
<td>0.40 ± 0.01</td>
</tr>
<tr>
<td>14.4 ± 0.2</td>
<td>5.7 ± 0.1</td>
<td>11.56 ± 0.1</td>
<td>0.80 ± 0.01</td>
<td>0.40 ± 0.01</td>
</tr>
<tr>
<td>14.3 ± 0.2</td>
<td>5.7 ± 0.1</td>
<td>11.46 ± 0.1</td>
<td>0.80 ± 0.01</td>
<td>0.40 ± 0.01</td>
</tr>
<tr>
<td>14.3 ± 0.2'</td>
<td>6.0 ± 0.2</td>
<td>11.45 ± 0.1</td>
<td>0.80 ± 0.01</td>
<td>0.42 ± 0.02</td>
</tr>
<tr>
<td>14.2 ± 0.2'</td>
<td>5.6 ± 0.2</td>
<td>11.38 ± 0.1</td>
<td>0.80 ± 0.01</td>
<td>0.40 ± 0.02</td>
</tr>
<tr>
<td>14.4 ± 0.2'</td>
<td>5.8 ± 0.2</td>
<td>11.51 ± 0.1</td>
<td>0.80 ± 0.01</td>
<td>0.40 ± 0.02</td>
</tr>
<tr>
<td>14.4 ± 0.2'</td>
<td>5.5 ± 0.2</td>
<td>11.50 ± 0.1</td>
<td>0.80 ± 0.01</td>
<td>0.38 ± 0.02</td>
</tr>
</tbody>
</table>

The results show a very good reproducibility. Even results obtained on different days are comparable. It follows that the accuracy of the used method is better than 3% for mylar on aluminum. This is the accuracy with which the ratio H/C and O/C can be given with this technique.

However the determination of the absolute amount of target atoms should be provided with an extra inaccuracy, resulting from the determination of the thickness of the mylar foil which equals 3%. Therefore the conclusion is that the determination of atomic concentrations can be determined with an accuracy of 4%.

It is noticed that the atomic densities do not increase nor decrease during the day and even the ratios stay constant. It may be concluded that the mylar does not disintegrate nor changes composition under used beam conditions which were a beam current of 15 nA of 3 MeV protons and a beam spot of 1 mm$^2$ which
corresponds to an irradiated energy of 45 Js/m². The reason for not damaging the mylar might be found in the fact that incident protons are liable to multiple scattering in the aluminum foil before they reach the mylar. Multiple scattering results in an extra divergence of the incident protons of 2°. The irradiated energy on the mylar foil, located at the back side of the aluminum carrier foil is therefore spread out over a larger area. The heat that may occur from irradiation of the protons is also absorbed by the aluminum foil. This saves the mylar foil from heating and disintegration.

3.9.3 Reproducibility of the Experiments

The analysis of all samples was carried out with the background subtraction procedure which was previously described. Table 3.2 shows the results of the analysis of one sample. Measurements were carried out during one day.

Table 3.2: Atomic densities of carbon, hydrogen and oxygen in one analyzed sample. All values are in 10¹⁸at/cm². The atomic densities are calculated from comparison with the spectra obtained from the mylar standard.

<table>
<thead>
<tr>
<th># carbon</th>
<th># oxygen</th>
<th># hydrogen</th>
<th>H/C</th>
<th>O/C</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.86 ± 0.11</td>
<td>0.66 ± 0.06</td>
<td>3.83 ± 0.05</td>
<td>0.99 ± 0.03</td>
<td>0.17 ± 0.02</td>
</tr>
<tr>
<td>3.80 ± 0.11</td>
<td>0.72 ± 0.06</td>
<td>3.66 ± 0.05</td>
<td>0.99 ± 0.03</td>
<td>0.19 ± 0.02</td>
</tr>
<tr>
<td>4.10 ± 0.11</td>
<td>0.76 ± 0.06</td>
<td>3.65 ± 0.05</td>
<td>0.89 ± 0.03</td>
<td>0.19 ± 0.02</td>
</tr>
<tr>
<td>3.77 ± 0.11</td>
<td>0.76 ± 0.07</td>
<td>3.57 ± 0.05</td>
<td>0.95 ± 0.03</td>
<td>0.20 ± 0.02</td>
</tr>
<tr>
<td>3.88 ± 0.10</td>
<td>0.72 ± 0.06</td>
<td>3.55 ± 0.05</td>
<td>0.92 ± 0.03</td>
<td>0.19 ± 0.02</td>
</tr>
<tr>
<td>3.94 ± 0.12</td>
<td>0.72 ± 0.07</td>
<td>3.53 ± 0.05</td>
<td>0.89 ± 0.03</td>
<td>0.18 ± 0.02</td>
</tr>
<tr>
<td>4.07 ± 0.12</td>
<td>0.80 ± 0.07</td>
<td>3.69 ± 0.05</td>
<td>0.91 ± 0.03</td>
<td>0.20 ± 0.02</td>
</tr>
<tr>
<td>3.94 ± 0.12</td>
<td>0.73 ± 0.07</td>
<td>3.55 ± 0.05</td>
<td>0.90 ± 0.03</td>
<td>0.19 ± 0.02</td>
</tr>
<tr>
<td>3.92 ± 0.04</td>
<td>0.73 ± 0.02</td>
<td>3.63 ± 0.04</td>
<td>0.93 ± 0.01</td>
<td>0.19 ± 0.02</td>
</tr>
</tbody>
</table>

The average hydrogen to carbon ratio for the sample equals 0.93 ± 0.01 and follows from averaging over all measurements. The oxygen to carbon ratio is 0.19 ± 0.02. The variance is large because of inaccuracy of the measurements.

The absolute amount of target atoms follow direct from the experiment when a thickness of the mylar foil of 3.3 µm is assumed. The determination of the mylar thickness was 3%. This correction leads to the results for the average number of atoms. The average number of carbon atoms is 3.9 ± 0.1 10¹⁸at/cm². The number of oxygen is 0.73 ± 0.03 10¹⁸at/cm² and the number of hydrogen is 3.6 ± 0.1 10¹⁸at/cm².
3.9.4 Composition Changes of the Samples

Three samples were analyzed on one day. The experiment was repeated after four weeks and the sample composition was redetermined. Table 3.3 shows the results of the experiments.

Table 3.3: A comparison of the obtained results from analysis of three carbon deposited aluminum foils denoted as sample 4, 5 and 6. The samples were reanalysed after four weeks. All values must be multiplied with \(10^{18}\) at/cm².

### 3.3 A: Analysis of three samples.

<table>
<thead>
<tr>
<th>Sample</th>
<th># carbon</th>
<th># oxygen</th>
<th># hydrogen</th>
<th>H/C</th>
<th>O/C</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>4.18 ± 0.10</td>
<td>0.51 ± 0.05</td>
<td>3.77 ± 0.04</td>
<td>0.90 ± 0.02</td>
<td>0.12 ± 0.01</td>
</tr>
<tr>
<td>5</td>
<td>3.85 ± 0.10</td>
<td>0.44 ± 0.05</td>
<td>3.77 ± 0.04</td>
<td>0.98 ± 0.03</td>
<td>0.12 ± 0.01</td>
</tr>
<tr>
<td>6</td>
<td>2.41 ± 0.09</td>
<td>0.24 ± 0.04</td>
<td>2.19 ± 0.03</td>
<td>0.91 ± 0.04</td>
<td>0.10 ± 0.02</td>
</tr>
</tbody>
</table>

### 3.3 B: The results obtained after four weeks.

<table>
<thead>
<tr>
<th>Sample</th>
<th># carbon</th>
<th># oxygen</th>
<th># hydrogen</th>
<th>H/C</th>
<th>O/C</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>3.90 ± 0.12</td>
<td>0.75 ± 0.07</td>
<td>3.73 ± 0.06</td>
<td>0.95 ± 0.03</td>
<td>0.19 ± 0.02</td>
</tr>
<tr>
<td>5</td>
<td>3.83 ± 0.12</td>
<td>0.72 ± 0.07</td>
<td>3.77 ± 0.05</td>
<td>1.00 ± 0.03</td>
<td>0.19 ± 0.02</td>
</tr>
<tr>
<td>6</td>
<td>2.16 ± 0.10</td>
<td>0.51 ± 0.06</td>
<td>2.16 ± 0.04</td>
<td>1.00 ± 0.05</td>
<td>0.23 ± 0.03</td>
</tr>
</tbody>
</table>

The results show that sample 4 and 5 have a comparable amount of carbon oxygen and hydrogen but sample 6 has a significant lower amount of these elements. The ratios in the last columns are comparable. The oxygen content in all samples has dramatically increased during the four weeks. The relative increment of oxygen is higher for the sample with a small carbon coverage and the detected oxygen might therefore originate from the oxidized aluminum carrier foil. The hydrogen content remains constant which indicates that the hydrogen is bound in the carbon layer.

3.9.5 Analysis of Different Samples

The carbon oxygen and hydrogen amount were determined for 26 different "diamond like" carbon films which were deposited at the Eindhoven University of Technology. The accuracy in the determination of the amount of target atoms is however dependent on both the statistics of the measurement and the accuracy in the determination of the amount of atoms in the standard. The mylar foil on an aluminum carrier foil was used for comparison of the peak contents that were
obtained from the analysis of different samples. The ERDA and RBS spectra were obtained for 250 seconds using a beam current of 15 nA. Figure 3.10 shows a plot of the calculated number of atoms versus the relative error in the determined number of counts.

From the figure is concluded that the determination of the absolute amount of carbon in a "diamond like" deposited layer can be determined with an accuracy of less than 10% if the amount of carbon atoms is more than $10^{18}$ at/cm$^2$. The maximum accuracy is caused by statistics. The minimum value for hydrogen, when the same accuracy is requested is at $0.4\ 10^{18}$ at/cm$^2$. The accuracy in the determination of the oxygen content is comparable with the accuracy of the carbon content. The accuracy in the plot should be complemented with the accuracy in the determination of the mylar thickness.

3.9.6 16 MeV Incident Protons used for Analysis of the Samples

An experiment was done in order to determine the hydrogen, carbon and oxygen contents in the samples from one single energy spectrum. The target geometry, shown in figure 3.1, with the aluminum located at the front, is maintained but the energy of the incident protons is increased up to 16 MeV. At this high energy
a separation of the peaks, originating from elastic scattering from aluminum, oxygen and carbon, in the $45^\circ$ energy spectrum is expected. In order to determine the energy of the scattered protons, special detectors should be used. These detectors should have a large fully depleted depth region as the 16 MeV protons have a high range in the detector material. The range of a 16 MeV proton in silicon is for example 1600 $\mu$m (!!). This would demand a depletion depth of comparable thickness. The energy spectrum is shown in figure 3.10.

The expected separation of the peaks due to elastic scattering from carbon, oxygen and aluminum is visible. The carbon, oxygen and aluminum peaks are denoted in the spectrum, $C(p,p)$, $O(p,p)$ and $Al(p,p)$. The elastically scattered protons from hydrogen are located at 8 MeV in the spectrum. When the incident proton energy is increased, the energy of the relative motion of the two particle system is increased which results in a higher available energy for nuclear reactions and inelastic processes and therefore causes an increase of peaks in the energy spectrum. The extra peaks originate from inelastic scattering from the aluminum and carbon in the target. The inelastic carbon peak is denoted as $C(p,p')$. All other peaks are due to inelastic scattering of protons from aluminum. Energies of some states in aluminum excited in inelastic scattering are; $Q = 1.01$ MeV, $Q = 2.21$ MeV and $Q = 3.00$ MeV.

A disadvantage of high energy protons, used in an experiment, is the radiation background in the experimental room which will rise when nuclear reactions take place in the scattering chamber. A radiation background of 1.5mSv/h was measured, just after the experiment. This dose, sustained in one hour, is comparable with the dose experienced from natural radiation during one year in, for example, Holland.
It is therefore imprudent to expose a researcher to this high radiation. The radiation originates from the decay of $^{11}\text{C}$. The formation of the gamma emitter $^{11}\text{C}$ took place during the experiment. The instable carbon nucleus decays under emission of a gamma quant with a half-life is 20 minutes. The radiation is therefore long time after the exposure to the high energy protons still present.

Finally it should be noted that the differential scattering cross section for elastic scattering decreases strongly with higher proton energy. However a lower energy will result in a worsening of the peak separation of oxygen and carbon. A larger detection angle, beyond $45^\circ$, can lead to separation of the peaks but the scattering cross section will decrease with increasing angle. The conclusion is therefore that the use of high energy protons is inadvisable and that the use of 3 MeV protons with the simultaneous RBS and forward energy measurement is better.

### 3.10 Discussion of the Technique

The 3 MeV technique, with the use of two detectors and simultaneous measurement is advantageous over the ERDA technique with use of a 16 MeV proton beam, because the radiation level is much lower and the scattering cross section for elastic scattering is higher. The opportunity of the simultaneous acquisition of two energy spectra is advantageous because it guarantees equal beam conditions for both the spectra. It is therefore allowed to take the peak content of the elastically, from aluminum, backscattered protons, as a measure for the total amount of impinged protons, for the ERDA as well as for the RBS spectrum.

A condition which has to be considered is the beam current. There are two reasons for the beam current requested low in the experiment. One reason is that the ERDA detector might suffer when a high beam current is used and the number of impinging particles per second increases. The dead time of the ERDA detector will rise as a consequence which is disadvantageous for the experiment. The second reason is that in case that the beam current is too high the mylar foil, taken as a standard for the atomic hydrogen to carbon ratio, might change composition or sustain damage.

A suggestion for the depth profiling of the hydrogen in the carbon sample is the use of the Helium ERDA technique with use of a stopper foil in front of the detector. The stopper foil stops both the scattered alpha particles and heavier atoms released from the sample but it lets the proton recoils pass. However, this technique does not determine the amount of carbon in the film. This should be measured in a separated experiment or with use of Pulse Shape Discrimination (PSD). This technique has been described in detail in the masters thesis of Tolsma [Tol 91].
3.11 Conclusion

The conclusion can be drawn that the combination of simultaneous measurement of both an RBS and forward energy spectrum can be used to achieve a hydrogen to carbon ratio and an oxygen to carbon ratio for "diamond like" films which were deposited on a 10 μm aluminum carrier. With the use of a standard a calculation of the absolute amount of target atoms is obtained. In case of a 15 nA beam current and a 1 mm² target spot, a disintegration of the mylar is not seen and it can therefore be taken as a good standard.

The reproducibility of the different obtained data was within 3 % for the mylar foil. This result was obtained from experiments of 250 s with a beam current of 15 nA. Better statistics can be obtained when experiments take longer. The beam current should not be increased in order to save the forward detector.

It is noticed that the composition of the analyzed samples differs after four weeks from the earlier measured composition. The oxygen content increased dramatically. This oxygen increase is related with the thickness of the deposited carbon layer. It is higher in the case of a thin carbon layer, and might therefore originate from oxidation of the aluminum carrier foil. Further research should be done to prove this.
Chapter 4

Coincident Elastic Recoil Detection Analysis with Time Of Flight Measurement

In this chapter the CERDA-TOF experiments will be discussed. The CERDA experiment is an extension of the ERDA experiment, which was previously introduced in chapter 3. In an ERDA experiment, ejected recoil atoms are detected at a forward angle, with respect to the incident beam direction. ERDA is commonly used for depth profiling light elements H, C, N and O, with use of heavy ions, usually Si or Cl, and uses a stopper foil to discriminate between ejected recoils and scattered projectiles. In case of a light projectile (α or H) an other selection technique should be used in order to discriminate between recoils and scattered particles. Therefore CERDA was developed by Klein [Kle 86].

CERDA means Coincident Elastic Recoil Detection Analysis. In a CERDA experiment both the elastically scattered projectile and the ejected recoil atom are detected in coincidence. In case of an elastic collision between two particles with masses \( M_1 \) and \( M_2 \), the scattering angles of these particles are correlated which was derived in chapter 2, equation 2.3.

In order to obtain better depth resolution the CERDA experiment is complemented with a time of flight measurement (TOF) instead of direct energy measurement with semi-conductor detectors which will limit depth resolution till 50 nm [Rij 91]. The recoil detector is therefore positioned at a large distance so that the time of flight of the recoil atom is measured.

4.1 Energy Measurement at a Forward Angle

In an experiment the energy of the forward scattered particles are measured at an angle \( \phi \) with respect to the incident beam direction. Both scattered projectiles and recoil atoms hit the detector. It is first tried to obtain a good depth resolution for the depth profiling of carbon in a mylar foil. In order to have both a high electronic stopping power for carbon recoils and a high scattering cross section, 12.6 MeV alpha particles were used as projectiles. The dependence of the scattering cross section with scattering angle and energy is given in section 4.2.

Figure 4.1 shows the energy spectrum obtained from a Mylar foil \( (\text{C}_{10}\text{H}_{8}\text{O}_4) \). The energy spectrum shows different peaks, due to both the elastic and the inelastic scattering of alpha particles from target elements C and O (\( \text{C}(\alpha,\alpha), \text{C}(\alpha,\alpha'), \text{O}(\alpha,\alpha) \)) and peaks originating from the ejected recoils. The large number of peaks leads to superposition of peaks in the obtained energy spectrum. An example is the occurrence of the hydrogen recoils (H) arising at 6.1 MeV. This peak is superimposed on the broad peak arising from the ejected carbon recoils \( (\text{C}_{el}) \). The carbon recoil...
atoms, ejected at the front side of the mylar foil lose kinetic energy, due to the high electronic stopping power, on their trajectory through the mylar. Therefore detected carbon recoils have energies between 5.5 and 7.1 MeV.

Note that the ejected carbon recoils appear in the spectrum, but a peak due to the ejection of oxygen recoils from the mylar foil, expected at an energy of 4.9 MeV, does not appear. The absence is due to the lower magnitude of the non-Rutherford differential scattering cross section for the elastic collision of alpha particles with oxygen target atoms for $\phi = 30^\circ$.

### 4.2 Coincident Elastic Recoil Detection Analysis (CERDA)

Figure 4.2 shows an example of the detector geometry in a CERDA setup. In case of the elastic collision between an incident alpha and a carbon target atom, where the carbon is ejected at $30^\circ$ with respect to the incident beam direction, the coincident alpha particles will be scattered at $100.9^\circ$. The addition of the coincident gate in the previously described experiment would have given an energy spectrum showing only one broad peak due to detection of the carbon target atoms. Neither the hydrogen peak nor the peak due to inelastic scattering of alpha particles from carbon target atoms would appear in the CERDA spectrum.

A good efficiency of the CERDA technique is obtained when the differential scattering cross section for an elastic collision is large. Alpha particles, as projectiles,
Figure 4.2: The CERDA setup. Two detectors are used for the coincident detection of both elastically scattered alpha and ejected recoil atom. The detectors are positioned at an angle \( \theta \) and \( \phi \) respectively.

are used instead of protons because both the differential scattering cross section for alpha particles and the energy transfer to the recoil are higher, which is a consequence of the higher mass of the alpha particle. The differential scattering cross section as a function of energy are given in figure 4.3 for two scattering geometries, \( \theta_{cm} = 125.2^\circ \) and \( \theta_{cm} = 90^\circ \). Spectra are obtained from Carter et al. [Car 64].

Figure 4.3: Resonances in scattering cross section as a function of incident alpha energy. Two figures are shown, one with scattering geometry \( \theta_{cm} = 125.2^\circ \) and one at scattering geometry \( \theta_{cm}=90^\circ \).

A resonance in the \( \theta_{cm} = 125.2^\circ \) spectrum, located at 12.1 MeV, has a FWHM of 550 keV. In the case of elastic scattering at an angle \( \theta_{cm} = 90.0^\circ \) a resonance is located at 12.3 MeV and has a FWHM of 460 keV. The incident alpha particles are subject to
electronic stopping and their energy will decrease on their trajectory through the target material. In order to obtain depth profiling it is therefore profitable to have a high differential cross section for a certain depth range in the target. According to figure 4.2 the scattering cross section for 11.0 MeV incident alpha particles can even decrease dramatically. This implies a negligible magnitude for the differential scattering cross section at a certain depth in the target, and therefore few counts of elastic collisions at that depth. The target depth where the incident particles will have an energy of 11 MeV is 12 μm.

The scattering cross section not only depends on the energy of the incident alpha particle but is a function of the centre of mass scattering angle $\theta_{cm}$ as well. The angular dependence of the differential scattering cross section for elastic collisions originates from Fraunhöfer diffraction explained in chapter 2. Figure 4.3 shows the differential scattering cross section as a function of $\theta_{cm}$.

Two maxima in the differential scattering cross section are located at $\theta_{cm} = 120^\circ$ and $\theta_{cm} = 90^\circ$ in the case of an incident alpha energy of 12.1 MeV. Their magnitudes are 170 mb/sr and 155 mb/sr respectively. The recoil angles follow from equation 2.4, the correlation $\theta_{cm} = 180^\circ - 2\phi$. The results in corresponding detection angles for the elastically scattered alpha particles are derived from equation 2.6 and result in $\theta = 100.9^\circ$ and $\theta = 71.6^\circ$. The energy of the carbon recoils after the elastic collision with an incident alpha particle is 6.8 MeV in case of $\phi = 30^\circ$ and the stopping power is 6.65 keV/μg/cm² which is near the maximum stopping power.

Figure 4.4: The Fraunhöfer diffraction pattern for elastic scattering of alpha particles from carbon at different scattering angles $\theta_{cm}$ for different incident alpha energies $E_a$ (From [Car 64]).
4.3 CERDA Technique Complemented with Time Of Flight Measurement

The CERDA technique is extended with a Time Of Flight (TOF) measurement in order to obtain a better energy resolution for the detected recoil atom and therefore a better depth resolution. The time of flight, determined in the CERDA-TOF experiment, is the difference in arrival of elastically scattered projectile and the ejected recoil atom. Therefore the recoil detector is placed at a large distance from the target so in effect the recoil time of flight is measured. The carbon recoil atom has a higher stopping power than the elastically scattered alpha particle and therefore the time of flight depends on the velocity of the recoil. A recoil with lower energy (velocity) will arrive later at the stop detector which results in a higher TOF. The improved depth resolution is obtained when special timing detectors and fast timing amplifiers with high time resolution are used for determination of the arrival of both projectile and recoil. The detector time resolution corresponds to an energy resolution and therefore with a depth resolution.

4.3.1 The Geometry of a CERDA-Time-Of-Flight Setup

In order to understand the TOF experiment some useful formulas will be derived in this chapter. Figure 4.5 shows a CERDA-TOF geometry for the elastic scattering of alpha particles from carbon. The geometry has a centre of mass angle $\theta_{cm} = 120^\circ$, corresponding to $\phi = 30^\circ$ and $\theta = 100.9^\circ$. All angles are defined with respect to the incident beam direction.

![Figure 4.5: The scattering geometry for a CERDA-TOF experiment. Detector distances are denoted as $L_1$ and $L_2$, for the alpha and the recoil detector respectively.](image)

In order to calculate a time of flight an expression for the velocity of a particle is
derived. A particle with mass \( M \) and a non-relativistic velocity \( v \) has a kinetic energy \( E \) with a magnitude of \( E = \frac{1}{2}Mv^2 \). From this equation follows an expression for the velocity, given by:

\[
v = 1.38 \times 10^7 \sqrt{\frac{E}{M}} \tag{4.1}
\]

where the kinetic energy is expressed in MeV, the mass \( M \) in atomic mass units (amu) and the velocity in m/s. The time necessary, for a particle with velocity \( v \), to traverse a distance \( L \) equals \( L/v \). An expression for the difference in arrival of the elastically scattered alpha projectile and the ejected recoil atom is given by:

\[
T_{\text{tof}} = 72.3 \left\{ L_2 \sqrt{\frac{M_2}{E_2}} - L_1 \sqrt{\frac{M_1}{E_1}} \right\} \tag{4.2}
\]

where \( T_{\text{tof}} \) in ns, \( E_1 \) is the kinetic energy of the elastically scattered alpha in MeV, \( E_2 \) kinetic energy of the ejected recoil atom in MeV and \( M_1 \) and \( M_2 \) the corresponding masses in amu. The detector distances, in m, with respect to the target are denoted as \( L_1 \) for the start detector and \( L_2 \) for the stop detector.

4.3.2 The Timing of the Scattered Projectile and the Ejected Recoil Atom

The derivative of equation 4.2 with respect to the energy \( E_1 \) and \( E_2 \) is given by:

\[
dT_{\text{tof}} = 36.15 L_1 M_1^{1/2} E_1^{-3/2} dE_1 - 36.15 L_2 M_2^{1/2} E_2^{-3/2} dE_2 \tag{4.3}
\]

where \( dE_1 \) and \( dE_2 \) are given in keV and \( dT_{\text{tof}} \) in ps.

The equation is a first approximation for the time shift resulting from the detection of particles with different energies and can be used when the energy difference is small compared to the energies of the detected particles.

Time spread, due to energy spread \( dE_1 \) and \( dE_2 \) in the kinetic energy of detected particles, occur in both start and stop detector. Moreover the timing resolution of a detector, dt_{det}, is consequently coupled to a limited energy resolution dE_{det}, according to equation 4.3.

4.3.3 The Width of the CERDA-TOF Spectrum

Equation 4.3 is a first approximation for the difference in arrival of a recoil atom with kinetic energy \( E_2 \) and another recoil atom with energy \( E_2 - \Delta E_2 \). The energy loss \( \Delta E_2 \) is due to the stopping of the recoil on a trajectory \( \Delta x \) through the target. Figure 4.6 shows the scattering geometry in the target.
Consider an ejected carbon recoil atom with an energy $E_{Cx}$ originated from a depth $x$ as is shown in figure 4.6. The energy, $E_{Cx}$, is calculated with respect to the energy of a carbon recoil that is ejected from the back of the carbon film, denoted as $E_{C_{back}}$. A formula for the calculation of $E_{Cx}$ is:

$$E_{Cx} = E_{C_{back}} + \left\{ \frac{K_2}{\cos \alpha_i} S_{ac}(E_0) - \frac{1}{\cos \rho_o} S_{cc}(K_2 E_0) \right\} x \quad (4.4)$$

where angles $\alpha_i$ and $\rho_o$ are denoted in figure 4.6. The first term takes into account the stopping of the incident alpha particle, the second term takes into account the stopping of the carbon recoils. The first term is much smaller than the second term and will therefore be neglected in further calculations. $K_2$ is the kinematic factor of the recoil atom, and $S_{ac}$ and $S_{cc}$ the stopping powers of the alpha particles in the carbon target and the carbon recoil atom in the carbon target. The recoil originated from the back of the target has an energy:

$$E_{C_{back}} = K_2 \left( E_0 - \frac{D S_{ac}(E_0)}{\cos \alpha_i} \right) \quad (4.5)$$

The second term is much smaller than $E_0$ and can therefore be neglected for convenience so that the energy equals $K_2 E_0$. The maximum energy difference for
recoils ejected from the sample is $E_{\text{Back}} - E_{\text{C}}(D)$. This energy difference will result in a different time of flight and can be calculated in a first approximation with equation 4.3. The major contribution to this time difference is the timing of the recoil, the second term in equation 4.3. The timing of the scattered alpha particle does slightly differ with the originated depth because the stopping power of alpha particles in matter is small, compared to the stopping power of carbon. Also the start detector was located at a 6 cm distance from the target, compared to 2.0 m or 3.5 m for the stop detector, which is as close as possible so that the time difference of the alpha particle can be neglected. The TOF difference will therefore only depend on the arrival of the recoil, originated from a certain depth. The expression for the maximum TOF difference is:

$$T_{\text{tof}}(0) - T_{\text{tof}}(D) = 36.15 \frac{L_2}{S_{\text{cc}}(E_2)}$$

where $T_{\text{tof}}(0) - T_{\text{tof}}(D)$ in ps, $M_2$ in amu, $L_2$ in m, and $S_{\text{cc}}(E_2)$ in keV cm$^2$ μg$^{-1}$. Note that the TOF difference increases when the stopping of recoils is at the maximum and the detector distance $L_2$ is increased. Also if the target is tilted in such a way that $\cos \theta_0$ decreases, the TOF difference will increase because the trajectory of the recoil atom in the target increases. Note that this approximation is a first approximation and therefore only valid in case of small energy losses $\Delta E_2$ with respect to the recoil energy $E_2$.

### 4.3.4 The CERDA-TOF Experiment

The first experiments concern the depth profiling of two thin self-supporting carbon films. The optimum conditions, a high differential scattering cross section for the elastic collision and high stopping power for the ejected recoils, are fulfilled when 12.6 MeV incident alpha particles are used. The scattering geometry is chosen at $\theta_{cm} = 120^\circ$ ($\phi = 30^\circ$, $\theta = 100.9^\circ$). The angles of the incident alpha particle $\alpha_i = 30^\circ$ and the scattered alpha particle $\alpha_o = 180^\circ - \alpha_i - \theta = 49.1^\circ$ are defined with respect to the target normal. The angle of the ejected recoil with the target normal is $\rho_o = 0^\circ$. The detector distances in the experiment were $L_1 = 0.06$ m and $L_2 = 2.00$ m. Figure 4.7 shows the measured TOF spectra. The analyzed carbon foils had thicknesses of 10 μg/cm$^2$ ± 5 μg/cm$^2$ (44 nm) and 20 μg/cm$^2$ ± 5 μg/cm$^2$, respectively, according to the specifications of the manufacturer.

The FWHM of the spectrum equals the time of flight difference, $T_{\text{tof}}$, in equation 4.6. The measured widths in the obtained spectra are 1.23 ns and 2.34 ns. These time differences correspond to energy differences of 92.8 keV and 176 keV. From these energy differences the target thicknesses of the investigated targets is derived with use of the stopping power for carbon recoils in a carbon target. The stopping power of 7 MeV carbon recoil atoms is $S_{\text{cc}}(7 \text{ MeV}) = 6.65 \text{ keV}/(\mu\text{g/cm}^2)$. 

---

42
The calculated target thicknesses are 14 $\mu$g/cm$^2$ and a 27 $\mu$g/cm$^2$, respectively.

The CERDA-TOF spectra do not have a step shape edges but they have smooth edges. The time spread $dT_{tot}$ at each edge corresponds to the difference in TOF at 12% and 88% of the maximum yield and is the time resolution at that edge when a Gaussian shape of the time resolution is assumed. This corresponds to an energy resolution $dE$ and a depth resolution $dx$. The fast time leading edge has a time spread of 450 ps which implies an energy resolution of 34 keV and a depth resolution of 5 $\mu$g/cm$^2$, corresponding to 22 nm. The slow time leading edges have time spreads of 640 ps and 860 ps which corresponds to depth resolutions of 7 $\mu$g/cm$^2$ (31 nm) and 10 $\mu$g/cm$^2$ (43 nm), respectively. The higher time spread is due to extra contributions to the energy spread due to energy straggling and multiple scattering of the recoil atoms in the target. These contributions will be discussed in section 4.5.

### 4.3.5 Tilting of the Investigated Target

The next experiment concerns the TOF-spectra obtained from tilted targets. The target tilt results in a more thick target "seen" by the recoil detector. Therefore an increased peak width in the CERDA-TOF spectrum is expected. Figure 4.8 shows two CERDA-TOF spectra obtained from the investigation of a target tilted in such a way that the exit angle of the recoils with the target normal, $\rho_0$, was 60°, and the same target where the ejected carbon recoil atoms left perpendicular from the target surface. The expected increase in peak width is therefore from D to D/cos$\rho_0$ which
implies a factor of two. The recoil detector distance was 3.50 m.

![Graph](image)

**Figure 4.8:** The CERDA-TOF spectra obtained from a 10 μg/cm² carbon foil. One with recoil angle parallel to the target normal, one with a 60° exit angle.

From figure 4.14 is seen that the expected doubling of thickness did occur. The thickness, D, of the tilted target is calculated and implies 15 μg/cm². The fast time leading edge remains the same for both cases with a corresponding energy resolution of 15 keV. In case of the target tilt the depth resolution at the surface is therefore improved with a factor of two, from 2.3 μg/cm² to 1.1 μg/cm² (5 nm). The resolution at the slow time edge is 7 μg/cm² in both cases. In case of a very glancing exit angle of the recoil atoms, the target flatness is very important.

It should be noted that the spectra in figure 4.14 are scaled so that both have the same height. This was done for convenience in order to compare both spectra. The total spectrum content corresponds with the number of coincidences. In case that the same counting statistics are demanded, the CERDA-TOF experiment with the tilted target should take two times longer because the incident ion beam sees the same amount of target atoms in both geometries, which was the case in this example. The contributions to the depth resolution at the fast time leading edge and at the slow time edge are summarized in section 4.4 and 4.5 respectively.
4.4 Depth Resolution at the Fast Time Leading Edge

In this section a summary is given of the different contributions to a good depth resolution in a CERDA-TOF experiment. The depth resolution at the surface is the depth resolution following from the time resolution at the fast time leading edge, $d_{\text{tof}}(0)$. The depth resolution at a target depth $D$, $d_{\text{tof}}(D)$, is derived from the time resolution at the slow time leading edge. A general expression for the time resolution is given by:

$$ (d_{\text{tof}})^2 = (d_{\text{tof,system}})^2 + (dT_2)^2 + (dT_1)^2 $$  \hspace{1cm} (4.7)

where $d_{\text{tof,system}}$ the system time resolution, $dT_2$ is the time spread arising from energy spread $dE_2$ of the detected recoils, and $dT_1$ is the time spread arising from energy spread $dE_1$ of the detected alpha particles. When the parameters of the previously described experiments are used, the expressions for the time spread in the recoil and the time spread in the detected alpha are $dT_2 = 13.3 \ dE_2$ in case of a detector distance of 2.00 m, and $dT_2 = 23.3 \ dE_2$ when the recoil detector distance is 3.50 m. In both cases is $dT_1 = 0.335 \ dE_1$. It is therefore allowed to neglect the contribution of the energy spread of the projectiles as long as $dE_1$ and $dE_2$ are comparable. The only contribution that is left to discuss is the time spread in the detected recoil atoms $dT_2$ due to an energy spread $dE_2$ and will be discussed in section 4.4.2. The system time resolution $d_{\text{tof,system}}$ will be discussed at first.

4.4.1 The System Time Resolution

The system time resolution $d_{\text{tof,system}}$ depends on the time resolution of the used detectors in the CERDA-TOF setup and the time resolution of the electronics processing the fast timing signals. The total time resolution of the TOF system is given by:

$$ (d_{\text{tof, system}})^2 = (d_{\text{det1}})^2 + (d_{\text{det2}})^2 + (d_{\text{amp}})^2 $$ \hspace{1cm} (4.8)

The CERDA-TOF experiment makes use of two fast timing detectors both having a detector time resolution of $d_{\text{det1}}$ and $d_{\text{det2}}$ respectively. The detector time signals are processed with Silena amplifiers fast timing output and amplified by fast amplifiers after which the signals are converted to pulses by a Constant Fraction Discriminator (CFD). The start and stop pulse, coming from the CFD, are the input of a Time to Amplitude Converter (TAC). The output of the TAC is the time difference between the arrival of the start and stop detector signals and is offered to a Multi Channel Analyzer (MCA). The time resolution of the amplifiers and electronics is denoted as $d_{\text{amp}}$. This contribution was determined with use of a test pulse offered to both detectors and resulted in $d_{\text{amp}} = 60\ \text{ps}$. The system time resolution for a combination
of two detectors has been determined before in experiments described in the master thesis of Teeuwen [Tee 91] and Rijken [Rij 91]. They showed that both the system time resolution and the TOF changes with the position of the detector. From a detector scan over the surface a mean system time resolution for a combination of two detectors is calculated (210 ps).

4.4.2 Time Spread Due To Energy Spread in the Detected Particles

One more terms of the general equation for the time resolution $dT_{\text{tof}}$, i.e. equation 4.7, is discussed. Contributions to the energy spread of the detected recoils have a different origin. The different contributions are:

- energy spread $dE_0$ of the incident projectiles that result in an energy spread $dE_2$ of the ejected recoils,
- the acceptance angle, $d\phi_{\text{det}}$ of the recoil detector,
- divergence of the incident beam which leads to an angular spread $\Delta$ in the scattering plane, and
- the beam spot size on the target which also leads to an angular spread in the scattering plane and contributes to the energy spread in the detected recoils.

The recoils, scattered at depth $x$, are due to two more contributions which result in an energy spread. The additional contributions are:

- multiple scattering, which results in an extra energy spread $dE_{\text{MS}}$ and
- energy straggling ($dE_{\text{ES}}$).

These additional contribute to the slow time edge in the CERDA-TOF spectrum and are discussed in section 4.5. The first four contributions limit the best obtainable depth resolution at the fast time leading edge in the CERDA-TOF spectrum and will be discussed separately in the next sections.

4.4.3 Energy Spread of the Incident Particles

The incident projectiles have an energy spread $dE_0$ of 3% due to the cyclotron specifications but was decreased till 2% with use of the dispersive system consisting of two slits located in the beam line (Figure 1.1). The scattered alpha particles are due to an energy spread $K_1 dE_0$, the detected recoil atoms are due to an energy spread $K_2 dE_0$.

4.4.4 Energy Spread Resulting from Angular Spread

Three of the considered contributions are related to an angular dependence of the energy of both the scattered alpha particle and the ejected recoil atom with a magnitude $dE_1/d\theta$ and $dE_2/d\phi$, respectively. Figure 4.9 shows the different angles
which are defined in the scattering geometry.

![Scattering geometry diagram](image)

**Figure 4.9:** Scattering geometry and denoted angles, the angular spread \( \Delta \) as a consequence of beam divergence, the finite beam spot size on the target, the acceptance angle \( d\phi_{\text{det}} \) of the recoil detector and the projectile detector acceptance angle \( d\theta_{\text{det}} \).

The energy of the projectile and the recoil have a angular dependency of \( dE_1(\theta) = (dK_1/d\theta)E_0 \) and \( dE_2(\phi) = (dK_2/d\phi)E_0 \) respectively, where \( K_1 \) and \( K_2 \) are the kinematic factors for projectile and recoil respectively. The expression for the derivative of the projectile energy with respect to the scattering angle is:

\[
\frac{dK_1}{d\theta} E_0 = -\frac{2\sin\theta}{(1+\mu)^2} \left( 2\cos\theta + \frac{\mu^2 + \cos(2\theta)}{\sqrt{\mu^2 - \sin^2\theta}} \right) \frac{\pi}{180^\circ} E_0 \tag{4.9}
\]

and is expressed in MeV per degree. A corresponding equation is derived for the recoil energy:

\[
\frac{dK_2}{d\phi} E_0 = -\frac{8\mu \sin\phi \cos\phi}{(1+\mu)^2} \frac{\pi}{180^\circ} E_0 \tag{4.10}
\]

These equations are used in the next sections to calculate the contributions of angular spread to energy spread for the detector acceptance angle \( d\phi_{\text{det}} \) and \( d\theta_{\text{det}} \), the angles due to beam divergence (\( \Delta \)) and beam spot size, denoted in figure 4.8, by multiplication of these angular spreads with the derivatives in equation 4.9 and 4.10. The calculations are only carried out for angles in the plane of incidence because the contribution of the acceptance angles perpendicular on the scattering plane are zero because the derivatives of the kinematic factors are zero.
In the case of the scattering geometry where \( \phi = 30^\circ \) and \( \theta = 100.9^\circ \) the expressions for the energy spread arising from acceptance angles \( d\theta \) and \( d\phi \) are given by \( dE_1 = 67 \ d\theta \), and \( dE_2 = 142 \ d\phi \), where the angles are in degrees and the energy spread in keV. The detector coincident acceptance angles for a CERDA scattering geometry where \( \phi = 30^\circ \), are correlated \( d\theta_{\text{det}} = 2.1 \ d\phi_{\text{det}} \) which follows from derivation of equation 4.9 and 4.10 when angles and particle and projectile masses are substituted.

4.4.5 Compensation of the Time Spread caused by Detector Acceptance Angles

Carbon atoms ejected at a slightly larger angle \( \phi + d\phi \) gain less energy in the elastic collision than atoms ejected at an angle \( \phi \), and will therefore arrive later at the stop detector. The difference in arrival time is compensated when the stop detector is tilted in such a way that the low energy recoils have a shorter flight path. The detector tilting is shown in figure 4.10.

![Diagram](image)

**Figure 4.10:** Elimination of the energy spread due to finite detector acceptance angles. The detector is located at a distance \( L_2 \) and is tilted on an angle \( \tau \), as denoted in the figure.

Consider a collision between an incident alpha particle with a kinetic energy \( E_0 \) and a carbon recoil in the target. The energy spread \( dE_2 \) in the detected recoil atoms resulting from the detector acceptance angle \( d\phi_{\text{det}} \) is:

\[
\frac{dE_2}{d\phi} = \frac{dK_2}{d\phi} \frac{E_0}{d\phi_{\text{det}}}
\]  

(4.11)

The path difference between recoils ejected at angles \( \phi \) and \( \phi + d\phi_{\text{det}} \), respectively is \( ds = v \ dt \), where \( v \) the velocity, according to equation 4.1, and \( dt \) the difference in arrival of a particle according to equation 4.3. Substitution of \( dT_2 \) and \( v_2 \) results in a general equation for the flight path difference of particles with slightly different energy \( dE_2 \) needed for compensation of the corresponding flight time difference \( dT_2 \):

\[
ds = \frac{-L_2 \ dE_2}{2 \ E_2}
\]  

(4.12)
where $L_2$ the distance between the target and the detector.

The flight path difference for the low energy recoil, ejected at an angle $\phi + \delta\phi_{\text{det}}$, follows from the geometry of the detector tilt. A formula for the flight path difference in case of a detector with diameter $D_{\text{det}}$ is given by:

$$ds = D_{\text{det}}\sin\tau_r$$

(4.13)

where $\tau_r$ the recoil detector tilt angle. The detector acceptance angle $\delta\phi_{\text{det}}$, can also be expressed as:

$$\delta\phi_{\text{det}} = \frac{D_{\text{det}}}{L_2} \cos\tau_r$$

(4.14)

according to the detector geometry which is valid for small detector acceptance angles $\delta\phi_{\text{det}}$.

Substitution of equations 4.11, 4.13 and 4.14 in equation 4.12 results in:

$$\tan\tau_r = \frac{1}{2} \frac{dK_2 E_0}{\delta\phi E_2}$$

(4.15)

This results in $\tau_r = \phi$, which is derived in appendix B, and does not depend on the energy nor on the detector distance.

The start detector can also be tilted in order to correct for the energy spread that results in a time spread. The tilt angle $\tau_\alpha$ can be derived in the same way and follows from:

$$\tan\tau_\alpha = \frac{1}{2} \frac{dK_1 E_0}{\delta\theta E_1}$$

(4.16)

It can be derived that for the detector, positioned at an angle $\theta$ the following equation for the detector tilt angle holds:

$$\tan\tau_\alpha = \frac{\sin\theta}{\sqrt{\mu^2 - \sin^2\theta}}$$

(4.17)

where $\mu$ the mass ratio $M_2/M_1$ of the colliding particles. The tilt angle does not depend on energy and detector distance. In the case of the alpha carbon scattering at the geometry where $\theta_{\text{cm}} = 120^\circ$, this results in $\tau_\alpha = 19.1^\circ$. The tilt angle of the recoil detector equals $\tau_r = 30^\circ$ in the chosen geometry. The energy difference that was compensated by detector tilting was 93 keV for both recoil and alpha particle detector...
in case of a recoil detector distance of 2.00 m. A convenient formula for the tilt angle of the alpha start detector is $\tau_a = \theta_{cm} - \theta$. This follows from equation 4.17 and is derived in appendix B.

### 4.4.6 The Beam Spot Size

A short description of the influence of the beam spot size, BS, on the obtained depth resolution is given in this section. The incident ion beam is focused through a 2 mm hole of a test target located in the centre of the scattering chamber. Both the current on this target and on the Faraday cup, behind the target, are measured. If 90% of the total beam passes the 2 mm hole, i.e impinges on the Faraday cup (Fig 4.12), 1.5 mm is taken as an approximation for the maximum beam spot size BS on the target. The distribution of the intensity of the beam in the hole is not known. Figure 4.11 shows the target geometry and the proportion of the beam spot on an investigated target.

![Figure 4.11: The beam spot size BS seen by the start and stop detector. Impinging alpha particles hit the target. The angle $\alpha_i$ and $\alpha_s$ are the angle of the incident and scattered alpha particles with respect to the sample normal. The angle $\rho_o$ is the angle of the ejected recoil with respect to the target normal.](image)

For the geometry of figure 4.11 the relations $180^\circ - \alpha_i - \theta = \alpha_o$ and $\phi - \alpha_i = \rho_o$ are valid. Expressions for the beam spot sizes, seen by the start and the stop detector are:

$$BS_1 = BS \frac{\cos \alpha_o}{\cos \alpha_i}$$

$$BS_2 = BS \frac{\cos \rho_o}{\cos \alpha_i}$$

(4.18)

The corresponding acceptance angles are $(BS_1/L_1)$ and $(BS_2/L_2)$, both in rad. Note that the magnitude of the two acceptance angles depends on the target orientation and the scattering geometry. The maximum energy spread due to the 1.5 mm beam spot is 6 keV in case of the 2.00 m CERDA-TOF experiment and therefore negligible compared to other contributions.
4.4.7 The Divergence of the Incident Beam

The angular spread $\Delta$ is due to divergence of the incident beam and is defined in the scattering plane. Figure 4.12 shows the geometry of the slit, used to decrease the divergence, the target position and location of the Faraday cup.

![Figure 4.12: The angular spread $\Delta$ in the scattering plane due to beam divergence. A slit is located at 1690 mm ahead of the target, denoted as $L_{\text{slit}}$. The incident beam is cut on both sides by the slit. Currents can be measured on both the target and the Faraday cup.](image)

The beam divergence, in the scattering plane, is decreased by cutting of the beam by slits located at the distance $L_{\text{slit}} = 169$ cm ahead of the target. The slits are positioned in such a way that the beam is cut on both sides. The contribution of the intensity of the beam as a function of the slit width $d$ is measured in a beam scan experiment where both the current on the slits and on the Faraday cup are measured. An expression for the angular spread $\Delta$, in the scattering plane, originating from the beam divergence is:

$$\Delta = \frac{180^\circ \cdot d}{\pi \cdot 1690}$$

where $d$ is the opening of the slit in mm.

4.4.8 Depth Resolution as a Function of the Beam Divergence

The next experiments show the contribution of the divergence of the incident beam to the depth resolution at the surface. The experiments were carried out in order to measure the contribution of the beam divergence to the obtained depth resolution in the CERDA-TOF experiment. The slit width $d$ is varied. These experiments were done on two different days. On both days the incident beam energy was 12.6 MeV. The recoil, or stop, detector was positioned at a $\phi = 30^\circ$ and the coincident alpha detector at $\theta = 100.9^\circ$. The recoil detector distance was 3.50 m on the first day and 2.00 m on the second day and was tilted at $\tau_r = 30^\circ$ so that time spread arising from the finite detector opening angle can be neglected which was mentioned in section 4.4.6. Four CERDA-TOF spectra with different divergences were obtained on each day and the energy spreads derived from the time spread of the fast time leading edges were determined. Figure 4.13 shows the calculated energy.
spreads, derived from the measured time spreads of the leading edges, as a function of the slit width d derived from the eight obtained spectra.

Figure 4.13: The contribution to the energy spread in the recoils originating from beam divergence. The graph shows two series of measurements that were taken on a different day.

From figure 4.13 is seen that the energy spread at the fast time leading edge increases with increasing slit width d. The depth resolution is improved when the width of the slits is decreased. The strong dependence of the energy spread with the slit width implies that the beam divergence has a major contribution to the obtained depth resolution at the surface when d is large. In case of the experiment with the recoil positioned at 3.50 m, and d = 2.5 mm, this is $dE_z = 17$ keV, in case of the 2.00 m experiment it is $dE_z = 27$ keV, also for d = 2.5 mm.

In the case that the slit width is larger than the beam width at the location of the slit, the local beam width should be taken instead of the slit width. This is done for the data points located at the right side in the figure. An estimate for the beam size at the slit was found in a scan measurement where the beam current on the faraday cup, located in the target chamber, is measured as a function of the slit position. The maximum beam size, measured at the slit position, was $14 \pm 1$ mm at the first and $11 \pm 1$ mm at the second day. The error is given as a horizontal error bar in the data points at the right side in figure 4.12.

A calculation of the contribution of the divergence is made, using equation 4.19 and 4.10. It follows that a formula for the maximum contribution to the energy spread, related to the slit width d, is $dE_z = 4.8 d$. This relation is drawn in the figure 4.12. From the figure is seen that this relation approximates the contribution of the beam divergence to the total energy spread at the leading edge in the CERDA-TOF
When \( d = 2.5 \) mm also energy spread of the incident particles and system time resolution contribute to the energy spread of the leading edges in the CERDA-TOF spectra. The system time resolution, for the detector combination used in these experiments, was 214 ps [Rij 91]. This results in an energy spread of 16 keV in the case of the 2.00 m experiment and 9.3 keV in the case of the 3.50 m experiment. These contributions are significantly different in the two experiments and therefore contribute to an increased energy spread for the 2.00 m experiment. Another cause to the shift is the different energy spread of the incident projectiles, \( dE_0 \). This arises from different beam conditions at a different day. In case of the 2.00 m experiment the energy resolution was \( 23 \pm 2 \) keV, in case of the 3.50 m experiment this was \( 21 \pm 1 \) keV. Table 4.1 shows a comparison of the calculated contributions and the measured energy spread.

<table>
<thead>
<tr>
<th>Contribution</th>
<th>( L_2 = 2.00 ) m</th>
<th>( L_2 = 3.50 ) m</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy spread ( dE_2 )</td>
<td>13 keV</td>
<td>12 keV</td>
</tr>
<tr>
<td>System time resolution (213 ps)</td>
<td>16 keV</td>
<td>9.3 keV</td>
</tr>
<tr>
<td>Beam divergence (( d = 2.5 ) mm)</td>
<td>12 keV</td>
<td>12 keV</td>
</tr>
<tr>
<td>Total:</td>
<td>24 keV</td>
<td>19 keV</td>
</tr>
<tr>
<td>Measured energy spread:</td>
<td>( 27 \pm 3 ) keV</td>
<td>( 17 \pm 2 ) keV</td>
</tr>
</tbody>
</table>

Note that the system time resolution limits good energy and depth resolution in case of the 2.00 m experiment. Therefore good timing detectors are requested. In case of the 3.50 m experiment this contribution is significant lower. However the detector acceptance angle and therefore the count rate will decrease with a factor of 3.

The error in the determination of the time spread at the fast timing edge is large because it is determined from a leading edge namely the time corresponding to 12% and 88% of the peak height. Therefore the peak height has to be determined which is subject to fluctuations that result from counting statistics.

A second error in the determination of the leading edge is the limited smoothness of the edge. Therefore an estimate for the total error in the determination of the leading edge is \( \pm 10\% \). The error in the calculated energy spread originates from the error in the determined time spread and is denoted in figure 4.12 as an error bar in each data point. Better statistics are obtained when the single measurement of the TOF-spectrum takes longer which results in better counting statistics. Another possibility is to increase the beam current but it was already at its maximum for all
spectra. This would imply that measurements with better counting statistics would take more than 2500 seconds which is an average of the time it took to obtain a CERDA-TOF spectrum.

It is concluded that the divergence of the incident beam is a major contribution to the time spread in the leading edge of the obtained CERDA-TOF spectra. However if the beam is cut by means of slits the beam current will decrease and the experiments will take longer when good counting statistics are demanded for the obtained CERDA-TOF spectra. The next section will describe the contributions that are due to depth resolution at the slow time edge.

4.5 Depth Resolution at the Slow Time Edge

The contributions, considered so far, are limitations for the best obtainable depth resolution in a time of flight measurement and are calculated from the time spread of the fast time leading edge. An extra energy spread in the detected recoil atoms arises from energy straggling and multiple scattering of the recoil atoms in the target when recoils originate from the inside, at a certain depth \( x \) in the sample. The right edge, or slow time edge, in the CERDA-TOF spectrum corresponds to the arrival of low energy recoils originated from the target front. The contributions of energy straggling and multiple scattering are shortly reviewed.

4.5.1 Energy Straggling

Energy straggling is the statistical fluctuation in energy loss of ions in matter. The energy loss is a consequence of electronic stopping. The energy straggling of ions after traversing a distance \( x \) through a target material is estimated from the Bohr formula

\[
\Omega_{\text{Bohr}} = 4\pi e^4 Z_1^2 Z_2 N x
\]

where \( Z_1 \) is the atomic number of the ion and \( Z_2 \) the atomic number of the target material with atomic density \( N \), \( e \) is the elementary charge. The Full Width at Half Maximum (FWHM), of the energy straggling, is wider by a factor of 2.35, assuming Poisson statistics. The energy straggling is related to a depth or thickness \( \rho' \) (\( \mu g/cm^2 \)). Note that \( \rho' = \rho x \), where \( \rho \) the target density (\( \mu g/cm^3 \)) and \( x \) the distance in cm. A convenient equation for the straggling is:

\[
\Omega_{\text{Bohr,FWHM}} = 0.93 Z_1 \left( \frac{Z_2}{M_2} \right)^{1/2} \sqrt{\rho'}
\]

The Bohr formula for straggling is valid in case of fully stripped fast ions in a free electron gas. Empirical formulae for energy loss straggling of ions in matter as a function of the ion energy were found by Yang and O'Connor [Yan 91]. The Bohr straggling is a good estimate for the energy straggling in all experiments described in this report. In case of the carbon recoil atoms in a carbon target the contribution
of straggling is 12.5 keV for the 10 µg/cm² and 18 keV for the 20 µg/cm² target.

4.5.2 Multiple Scattering

Multiple scattering is caused by small angle scattering of recoil atoms from target nuclei. The angular spread of the recoil beam increases with the length of the covered trajectory in the target. The angular spread due to electronic interactions can be neglected compared to the angular spread arising from interaction with the target nuclei. The Half Width at Half Maximum (HWHM) in the angular spread arising from multiple scattering after traversing a trajectory in a medium is:

$$\alpha_{1/2} = \frac{180^\circ}{\pi} \Gamma g_1(\tau)$$  \hspace{1cm} (4.21)

where $\Gamma$ and $\tau$ depend on the atomic numbers of interacting nuclei and parameters of the medium. The angular spread is determined for calculated values of $\tau$ with use of the tabulated values for $g_1(\tau)$ obtained from Sigmund and Winterborn [Sig 74]. A formula for $\tau$ is:

$$\tau = 13.20 \pi \rho^*/(M(Z_{\text{pro}}^{2/3} + Z_{\text{tar}}^{2/3}))$$  \hspace{1cm} (4.22)

where $M$ the atomic mass of the target atoms in amu and $Z_{\text{pro}}$ the atomic number of the projectile and $Z_{\text{tar}}$ the atomic number of the target atoms. The formula for $\Gamma$, with the parameters in convenient dimensions, is:

$$\Gamma = 6.15 \times 10^{-5} Z_{\text{pro}} Z_{\text{tar}} \sqrt{\frac{Z_{\text{pro}}^{2/3} + Z_{\text{tar}}^{2/3}}{E}}$$  \hspace{1cm} (4.23)

where $\Gamma$ is expressed in rad and $E$ the energy of the ion traversing the medium, in MeV.

Figure 4.14 shows the obtained CERDA-TOF spectrum and the simulation that takes multiple scattering and energy straggling into account. The time spread at the slow time edge is determined from the CERDA-TOF spectrum in the same way as the time spread at the fast time edge. The thickness of the investigated target was determined from the FWHM of the spectrum and equals 15 µg/cm². The depth resolution is 3 µg/cm² (12 nm) at the fast time edge and 7 µg/cm² (28 nm) at the slow time edge. The contribution energy straggling for the carbon recoil atoms originated from the front side of the carbon target is calculated from equation 4.20 and equals 14.9 keV. Multiple scattering results in a contribution of 21.2 keV. The contribution of multiple scattering and energy straggling is therefore 25.9 keV. Table 4.2 shows the calculated contributions and measured values. The calculated energy
spread, which is the simulated slow time edge, is significantly lower than the direct measurement of the energy spread in the slow time edge of the obtained spectrum. The difference is 28 keV which might originate from an extra contribution of surface roughness and target thickness variation to the energy spread in the slow time edge.

The extra energy spread corresponds with a target thickness variation of 5 μg/cm² which corresponds with the specifications of the manufacturer.

The simulation program will be further developed in order to make further calculations and fits of obtained spectra so that both the contributions of target thickness variations and wrinkly surface structures can be taken into account.

Table 4.2: Contributions to the slow time leading edge.

<table>
<thead>
<tr>
<th>Contribution</th>
<th>Energy spread</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy Straggling (ES)</td>
<td>14.9 keV</td>
</tr>
<tr>
<td>Multiple Scattering (MS)</td>
<td>21.2 keV</td>
</tr>
<tr>
<td>Fast Time Leading Edge</td>
<td>20 ± 2 keV</td>
</tr>
<tr>
<td><strong>Total:</strong></td>
<td><strong>33 ± 3 keV</strong></td>
</tr>
<tr>
<td><strong>Measured Slow Time Edge:</strong></td>
<td><strong>44 ± 4 keV</strong></td>
</tr>
</tbody>
</table>

Figure 4.14: The obtained CERDA-TOF spectrum of a 10 μg/cm² carbon foil. The spectrum is simulated. The simulation takes contributions of energy straggling and multiple scattering into account.
4.6 Carbon Recoil Atoms Traversing an Aluminum Layer

The next experiment concerns the determination of the energy spread of the ejected recoil atoms as a function of the traversed thickness of an aluminum layer. Therefore samples were used consisting of a 20 $\mu$g/cm$^2$ carbon film with evaporated aluminum layers of different thicknesses of 25, 50, 75 and 100 $\mu$g/cm$^2$. The sample orientation is chosen in such a way that the incident 12.6 MeV alpha beam hits the carbon foil first. The carbon recoil atoms are ejected from the target and will traverse the aluminum layer. The energy spread of the traversing atoms will increase due to both multiple scattering and energy straggling of the carbon recoil atoms in the aluminum layer. Figure 4.15 shows the CERDA-TOF spectra obtained from different targets.

![CERDA-TOF spectra](image)

Figure 4.15: CERDA-TOF spectra obtained from aluminum layered carbon targets. The thickness of the carbon films is 20 $\mu$g/cm$^2$. The thickness of the evaporated aluminum is 0, 25 and 75 $\mu$g/cm$^2$ respectively, according to the manufacturer.

The spectra in figure 4.15 shift to a higher TOF, i.e. to the right, when the thickness of the aluminum layer is increased, because the carbon recoil will lose kinetic energy due to electronic stopping in the aluminum layer and will arrive later at the stop detector. It is also seen that the peak width increases for higher time of flight. This is a direct consequence of the energy dependence of the time of flight which is inversely proportional with the square root of the energy of the recoil atom. The $dT_{\text{tof}}$-dE correlation for a carbon foil, without aluminum, is $dt = 13.3$ dE. The correlation for recoils that are slowed down as a consequence of electronic stopping in a 100 $\mu$g/cm$^2$ Al foil is $dt = 14.7$ dE. Note that the difference is 10% and the first order
approximation should be adjusted for the analysis of each spectrum.

A remarkable peak in the measured spectra is the small peak occurring at 185 ns. This small peak is due to the arrival of fast recoil atoms, ejected from the aluminum surface, i.e. the back of the target. The time difference between this small peak and the fast time leading edge of the carbon spectrum is due to electronic stopping of the carbon recoil atoms in the aluminum. The thicknesses of the aluminum layers are checked from these time differences using equation 4.2. The TOF difference results in the thickness D. The calculations result in the thicknesses of 30.2 μg/cm², 49.8 μg/cm², 81.2 μg/cm² and 94.8 μg/cm² respectively.

4.6.1 The Energy Spread Caused by the Aluminum Layers

The time spread at the fast time leading edges of all obtained spectra are determined and the corresponding energy spreads are plotted as a function of the aluminum layer thickness in figure 4.16. Note that for the calculation of the energy spread from the time spread, the change of the recoil energy must be taken into account.

![Figure 4.16: The energy spread in the fast time leading edge as a function of the thickness of the aluminum layer. All values are obtained from the corresponding dt-dE correlations.](image)

The increase in energy spread with aluminum thickness is visible. The error bars indicate an accuracy in the calculated energy spread of ± 10%. The calculated contributions of energy straggling (ES), and multiple scattering (MS), are also plotted in the graph. The energy spread in the case of a measurement without aluminum is due to contributions of system time resolution, energy spread of the incident alpha
particles and beam divergence. From the results in figure 4.16 follows that these contributions result in a minimum energy spread of 30 keV. This minimum energy spread is added to the theoretical contributions of energy straggling and multiple scattering and the straight line in figure 4.16 is the obtained result.

In case that the investigated samples have wrinkly surfaces or thickness variations, an extra energy spread will contribute to the leading edge. This is the case for the carbon foil with the very thin evaporated aluminum layer according to figure 4.16.

The error in the measured data is still large and certainty about the exact fit can not be given. A subject of discussion is if it is allowed to take the fast time leading edge as a measure. The answer is that this is only allowed when the contribution of the energy spread, that cause the edge smoothing, should be small compared to the peak width and can therefore only be taken when the CERDA-TOF spectrum shows a flat top. Further measurements with use of simulations should therefore be carried out with thicker carbon layers so that a flat top remain even after the traversing of a more thick aluminum layer.

4.6.2 Growth of Carbon on the Aluminum During the Experiment

The occurrence of the small peaks in the CERDA-TOF spectra, arising from coincident detected recoil atoms from the back side of the investigated targets, is clearly seen in figure 4.15. This peak is due to carbon growth on the back side. Figure 4.17 shows the growth of the carbon.

![Carbon Growth Graph](image_url)

Figure 4.17: The growth of the carbon on the aluminum during a 3600 seconds experiment.
The peak content, as well as the peak content of the broad peak (20 μg/cm²), are measured during a 3600 seconds experiment. The beam current was 55 nA and vacuum conditions were 5 \times 10^{-5} \text{ torr}. A foil was used which had not previously been exposed to an incident ion beam and the determination of the peak growth was continued after another 3600 seconds using a beam current of 35 nA and a further growth of carbon on the aluminum was noticed. During the intermediate time another sample, closely located to the previously investigated target, was exposed to the beam. The indicated error bars are the variances of the counting statistics, calculated for each data point.

Note that the carbon growth on the aluminum layer did not stop in the intermediate time when the target was not exposed to the ion beam. The obtained data show a monotonous growth, stimulated by the first exposure to the incident beam. The explanation for the carbon growth on the aluminum is due to the presence of carbon ions and hydro-carbon molecules which will be dissociated and deposit on the target. No explanation is found yet for the continuous growth during the intermediate time. The growth of the carbon on the aluminum is derived from the figure and it is 2 monolayers in 1000 seconds. It may be concluded that in case of investigation where samples may not be subject to undesired deposition, the vacuum needs to be improved.

4.7 CERDA-TOF at the 45° Scattering Geometry

The previously treated time of flight spectra were obtained in a 30° - 100.9° geometry. A disadvantage of this geometry is that the scattered alpha particle has a low energy after the collision which might be insufficient to traverse a more thick (20 μm aluminum) target material in order to create the start trigger.

There are two possibilities to overcome this problem. The first is the increment of the energy of the incident alpha particles which results in an increased of the scattered alpha after the collision. This choice will also result in a higher recoil energy and therefore a slightly smaller stopping power according to figure 2.7. A more serious problem is that the higher energy will result in a smaller differential scattering cross section because in the previous experiments the advantages of the resonant cross section was used.

The second possibility is to change the scattering geometry in such a way that the alpha particle obtains a higher kinetic energy after the collision. This implies that the alpha particles should scatter more forward with respect to the incident beam direction. When a recoil atom is ejected at \( \phi = 45° \) (\( \theta_{\text{cm}} = 90° \)) with respect to the incident beam direction and the coincident alpha particle is detected at \( \theta = 71.6° \). The
energy of the incident alpha particles is again 12.6 MeV because the differential
scattering cross section for elastic scattering has a maximum which follows from
figure 4.4. Figure 4.18 shows the 45° scattering geometry.

![Image of the 45° scattering geometry. Carbon recoils are ejected at 45° and the coincident alpha
particles are detected at 71.6° with respect to the incident beam direction. The target is tilted
in such a way that the recoils leave from the target at a 15° glancing angle.]

4.7.1 Depth Profiling of a Carbon Layer on an Aluminum Carrier Foil

The investigated sample is a carbon 20 µg/cm² foil on a 20 µm (!) aluminum
carrier foil. Therefore the 45° geometry, described in this chapter, was used. The
energy of the scattered alpha is 7.9 MeV in case of 12.6 MeV incident alpha particles.
This is enough to penetrate the 20 µm aluminum carrier foil because the range in
aluminum is even 35 µm [TRIM 89]. The scattered alpha is detected at the start
detector, positioned at 6 cm from the target. A CERDA-TOF spectrum obtained from
a 20 µg/cm² carbon film on a 20 µm aluminum carrier is shown in figure 4.19.

![Image of the CERDA-TOF spectrum obtained from a 20 µg/cm² carbon film positioned on a 20 µm
aluminum carrier foil.]

Figure 4.18: The 45° scattering geometry. Carbon recoils are ejected at 45° and the coincident alpha
particles are detected at 71.6° with respect to the incident beam direction. The target is tilted
in such a way that the recoils leave from the target at a 15° glancing angle.

Figure 4.19: The CERDA-TOF spectrum obtained from a 20 µg/cm² carbon film positioned on a 20 µm
aluminum carrier foil.
The measured spectrum shows some bumps that were not seen before in CERDA-TOF spectra. These bumps are accidental coincidences and form an unwanted background. The accidental coincidences occur when a large number of start and stop triggers, arising from elastically, from the aluminum carrier foil, scattered particles, are detected in both the start and the stop detector. The number of start triggers in the start detector was 20000 sec\(^{-1}\). The observed bumps in the CERDA-TOF spectrum are periodic with a period of 133 ns. The periodicity occurs from the cyclotron frequency which was 7.5 MHz in case of this experiment. The cyclotron accelerates bunches of alpha particles with this frequency.

A method that leads to suppression of the accidental coincidences is the introduction of an extra trigger for a valid coincidence. As the energy of the elastically scattered alpha particle and recoil atom are known within a certain region, it is possible to introduce an extra demand for the energy of both the detected projectile and the recoil atom. Figure 4.20 shows the obtained energy spectra from the start and the stop detector.

![Energy spectra obtained from the start and the stop detector](image)

Figure 4.20: Energy spectra obtained from the start and the stop detector positioned at a 71.6° and a 45° angle respectively.

The energy spectrum measured at the start detector shows two broad peaks due to the elastically and inelastically scattered alpha particles from aluminum (Al(\(\alpha,\alpha\)), Al(\(\alpha,\alpha'\))). The alpha particles scattered from the carbon film can not be distinguished. The maximum energy of the alpha particles that are scattered from the aluminum is 7.8 MeV. The aluminum recoil atoms are detected in the start detector but have a very low energy. The obtained 45° spectrum shows the large number of elastically and inelastically scattered alpha particles from aluminum, forming a continuous background, and aluminum recoils (Al) expected at 2.8 MeV and lower.

The aim for background suppression in the TOF spectrum is attained to with selection of the energy regions in both the start and the stop detector energy spectra. At first an energy spectrum was obtained from a carbon self-supporting foil which
is shown in figure 4.21. The spectrum shows peaks occurring from both the elastically and inelastically scattered alpha particles, carbon recoils (C), hydrogen (H) and finally carbon recoils that were ejected as a result of an inelastic collision (C'). The presence of high hydrogen peak in the spectrum is astonishing but might be explained by the method of preparation of the samples which occurred in water and the method of fabrication of the foils. The energy gate for the recoil detector is set from 3.7 up to 4.6 MeV, where the elastically ejected recoils are expected in the spectrum.

![Figure 4.21: The gates on the 71.6° start detector and the 45° stop detector.](image)

In order to do select the energy region in the start detector energy spectrum a sample consisting of a carbon layer positioned on an aluminum carrier is analyzed. The energy spectrum obtained is gated with the coincident criterion from the TAC and is shown in figure 4.21. It is clearly seen that the yield of the elastically, from aluminum scattered, alpha particles is suppressed due to the recoil gate and an alpha peak due to scattering from carbon occurs. The energy gate at the stop detector is set from 3.4 up to 4.9 MeV.

After the adjustment of both the energy gates, a CERDA-TOF spectrum with two extra gates for the TAC, the energy of both coincident alpha particle and ejected recoil atom, is obtained. Only when the energy of both alpha particle and recoil atom are within the selected regions, the TAC is triggered and the valid count is offered to the MCA. Figure 4.22 shows the measured spectrum.

This spectrum is free from unwanted background. The depth resolution at the surface, $d_x$, is $1.6 \, \mu g/cm^2$ (7 nm) when the correction for the glancing angle is made and the stopping power $S = 7.1 \, keV/\mu g/cm^2$ is used. This depth resolution is comparable with the analysis of a thin carbon film without the aluminum carrier foil and this is therefore a break-through in depth profiling of thin films on a thick carrier foil and might lead to applications in materials science.
Figure 4.22: The measured TOF spectrum measured from 20 µg/cm² carbon on a 20 µm aluminum carrier when multiple coincidence criteria are set.

4.7.2 Analysis of Self-Supporting Foils

A comparison of the spectra obtained from the carbon 20 µg/cm² self-supporting foil and the carbon foil positioned on an aluminum carrier are compared in figure 4.23.

Figure 4.23: A comparison of the obtained spectra from either a carbon 20 µg/cm² self-supporting foil and a carbon foil positioned on an aluminum carrier.
A shift of the fast timing edge is noted when the two obtained spectra are compared in figure 4.23. This shift is due to the different timing of the alpha particles at the start detector for the two cases. A simple calculation for the time interval between actual scattering and detection by the start detector can be made when the velocities of the alpha particles in both cases are known from the energy spectrum in figure 4.22, obtained from the start detector, and equals 4.6 MeV. The energy after elastic scattering from a carbon target nucleus would have been 7.8 MeV. From the energy loss in the aluminum layer follows, with use of the mean stopping power and equation 4.2, the thickness of the aluminum carrier foil, namely 22 μm. This implies a time shift of 940 ps when the distance of the start detector with respect to the target is 6 cm. However, the observed peak shift is 2.2 ns. From this measurement a detector distance of 11.6 cm would result from the calculation but the detector distance in the experiment was certainly smaller. It is therefore assumed that the timing properties of the start detector differ with energy of the detected particles.

From the obtained spectra shown in figure 4.23 can be concluded that the thickness of the self-supporting foil is smaller (18 μg/cm²) than the carbon layer on the aluminum carrier (25 μg/cm²). Furthermore the TOF spectrum of the self-supporting foil shows a longer tail due to a wrinkly sample causing thickness variations which results in extra contributions to the energy spread in the slow time edge. A closer view is given to a more thin self-supporting foil where the tail also occurs.

4.7.3 Thin Layer CERDA-TOF with a Glancing Exit Angle

The experiment concern the depth profiling of a 10 μg/cm² self-supporting carbon foil at the 45° geometry with a glancing exit angle of 15°. A closer view is given to the tail in the CERDA-TOF spectrum. The CERDA-TOF spectrum is shown in figure 4.24.

From the width of the spectrum (FWHM) follows that the film thickness D = 12 μg/cm². The depth resolution at the fast time leading edge (34 keV) is 1.3 μg/cm² (5.5 nm), which is extremely good. Note that the improved depth resolution is of course due to the glancing exit angle.

The contribution to the slow time edge are multiple scattering, straggling and target thickness variations. However the slow time leading edge shows also a long tail due to an extra contribution. This extra contribution is due to the wrinkly structure of the investigated target. This structure will result in an apparent thickness variation seen by the recoil detector. The effect is aggravated when the sample is located at the glancing angle geometry.
The wrinkly structure is seen with the naked eye and is therefore on mm-scale. The beam spot on the target is also approximately 1 mm. One demand for appropriate depth profiling is therefore that a flat target is used so that the tails will not occur. The demanded flatness must be on a mm-scale and therefore these used self-supporting films should not be used when the contributions to the slow time edge is investigated in the near future.

4.7.4 Discussion of the 45° CERDA-TOF Setup

The 45° TOF setup, discussed in the previous sections, has some advantages over the 30° setup. It was used in the first place for depth profiling of the surface of a thick sample carried out with the same projectile energy used in the 30° geometry.

The disadvantage is the enormous counting rate in both start and stop detector when both detectors are located at a forward angle. The enormous flow of scattered alpha particles in the start detector (20000 sec⁻¹) is still a problem and can cause damage to the detector. This problem of forward scattered projectiles will increase if the choice for the position of the start detector at a more forward angle is made because the scattering cross section in creases with decreasing scattering angle.

Moreover the limiting feature to the depth resolution was due to beam divergence which contributes to energy spread in the ejected recoils which is higher in case of the 45° geometry because it is proportional with dK²/dφ which has its maximum at 45°.
However an advantage of the chosen setup is that the contribution in the timing spread due to the energy variance of the incident projectiles is smaller because it is transferred to the recoils with the $K_2$-factor for elastic scattering. The $K_2$ factor decreases with increasing recoil angle $\phi$. Moreover the advantage of the possibility for the analysis of a thick sample using the same 12.6 MeV incident beam energy combined with a high stopping power for the recoils and a high scattering cross section was profitable.

The different contributions that limit depth resolution in both the 30° and the 45° geometry are denoted in the next table. The incident beam conditions are $d = 2.5$ mm, $E_0 = 12.6$ MeV and the diameter of the recoil detector is 24 mm. The best obtained system time resolution was 150 ps and is used in the table. This system time resolution was used in the previously described experiments. The improved time resolution was accomplished with a better detector combination.

<table>
<thead>
<tr>
<th>Contribution</th>
<th>30° (2.00 m)</th>
<th>45° (2.00 m)</th>
<th>30° (3.50 m)</th>
<th>45° (3.50 m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time res. (150 ps)</td>
<td>11.3 keV</td>
<td>6.1 keV</td>
<td>6.4 keV</td>
<td>3.5 keV</td>
</tr>
<tr>
<td>Energy res. ($dE_0=21$ keV)</td>
<td>11.8 keV</td>
<td>7.9 keV</td>
<td>11.8 keV</td>
<td>7.9 keV</td>
</tr>
<tr>
<td>Beam div. (d=2.5 mm)</td>
<td>12.0 keV</td>
<td>14.0 keV</td>
<td>12.0 keV</td>
<td>14.0 keV</td>
</tr>
<tr>
<td>Total:</td>
<td>20.3 keV</td>
<td>17.2 keV</td>
<td>18.0 keV</td>
<td>16.5 keV</td>
</tr>
</tbody>
</table>

From the tabulated values can be concluded that the 45° has better energy resolution. However if the alpha detector should be saved from suffering under the enormous amount of forward scattered particles, the 3.50 m, 30° geometry is recommended. Note that the beam divergence is the limiting contribution and that the new Canberra fast timing detectors slightly contribute.

The advantages of a glancing angle were already mentioned but one contribution should further be illustrated. The glancing geometry is favourable, especially for thick samples, because it detects recoils that originate from the front of the investigated target. The incident projectiles are not due to extra energy spread in the sample arising from multiple scattering and energy straggling, which is the case in when the transmission geometry is used. Remind that the energy spread of the incident projectiles was already mentioned in chapter 3 where a transmission geometry was used. The extra energy spread of the projectiles also result in a higher energy spread in the ejected recoils. The extremely glancing geometry should only be used in case of the analysis of samples with flat surfaces in order to deter the occurrence of a tail at the slow time edge.
4.8 The CERDA-TOF Multi Mass Spectrometer

The CERDA-TOF technique is very powerful for depth profiling of light elements in a heavy matrix. The next step in the development of the method was the request to profile other light elements. For example nitrogen and oxygen, important elements in semiconductor manufacturing. In order to determine the nitrogen in a sample the first demand, a high differential scattering cross section for the elastic collision of an incident alpha particle from nitrogen should be fulfilled.

Because the design of the scattering chamber only allowed the 30° and 45° geometry, a resonance was requested for the centre of mass angles $\theta_{cm} = 120°$ or $\theta_{cm} = 90°$. The differential scattering cross section as a function of incident alpha energy was investigated by Tollefsrud [Tol 70]. Figure 4.25 shows the scattering cross section as a function of the incident alpha particle energy.

![Figure 4.25: The differential scattering cross section as a function of incident energy for elastic scattering of alpha particles from nitrogen. The centre of mass angle for the collision is $\theta_{cm} = 121°$.](image)

The second demand for depth profiling with CERDA-TOF is a high stopping power for the ejected recoil atoms. Both demands can be fulfilled when 11 MeV incident alpha particles are used for analysis. Note the broad energy region where the scattering cross section is large which is favourable in the case of depth profiling of thick samples.

4.8.1 Depth Profiling of Si$_3$N$_4$

In order to depth profile nitrogen the experiment was carried out on a 2000 Å Si$_3$N$_4$ test foil obtained from Philips Eindhoven. The incident alpha energy was 10.9 MeV and the recoil detection angle was 30° with respect to the incident beam direction. The coincident angle $\theta$ for the elastic scattered alpha is 104°. The recoil detector distance was 2.00 m and the start detector distance was 6 cm. The target tilt angle was 40° which implies that $\alpha_i = 40°$ and $P_o = 10°$. The obtained TOF spectrum is shown in figure 4.26.
The TOF spectrum shows a substantial peak between 226 ns and 236 ns occurring from the nitrogen recoil. However there are more peaks noted in the measured spectrum. The small peaks at the left side of the nitrogen are due to the detected carbon recoils that are ejected as a consequence of an elastic collision and arise from unwanted deposition on the target. Maybe due to this carbon and oxygen growth one sample was destroyed which was a pity because they were very hard to manufacture.

The coincident alpha particle, which collided with a carbon target atom is elastically scattered at 101° and was still detected by the start detector due to the start detector acceptance angle of 7°. The carbon recoils obtain a higher energy in the collision because they have a lower mass and higher energy, compared to nitrogen target atoms, resulting in a decreased time of flight. The small fast carbon peak arises from a collision at the backside, the right peak is due to the coincident detection of carbon recoils that were ejected from the target front and lost kinetic energy due to electronic stopping in the Si₃N₄ medium. The small peaks at the right side of the nitrogen peak are due to the elastic collision between alpha particles and oxygen recoils on the sample front and back side, respectively. The elastically, from oxygen scattered, alpha particles are detected at 106°. However this technique is very powerful for the depth profiling of multiple elements in one spectrum. The advantage of one measurement for three elements is especially favourable because a measurement of a single CERDA-TOF spectrum takes almost one hour when good counting statistics are requested.
The energy resolution can be determined from the leading edge of the nitrogen peak which is shown in figure 4.27.

![Close-up N peak, Si₃N₄](image)

Figure 4.27: A close up of the nitrogen peak from figure 4.26.

From figure 4.27 follows an energy resolution of 30 keV and a depth resolution for the nitrogen depth profiling of 11 nm (4 μg/cm²). The thickness of the Si₃N₄ substrate is determined from the FWHM of the nitrogen peak and equals 160 nm which is less than claimed by the manufacturer.

The peak shape of the nitrogen TOF peak shows an extra decrease for the yield at higher time of flight which is not seen in the simulation. The decreased yield can not be explained by a change in composition of the sample but the explanation for the decreased yield is the change of the scattering cross section with target depth. In figure 4.27 is seen that the used energy of 10.9 MeV is located on an edge in the cross section. The cross section for elastic scattering increases with decreasing energy. The scattering cross section at the back side of the investigated target is therefore larger and will result in more detected coincidences for this depth. Therefore experiments in the near future should be carried out with a slightly lower energy of the incident alpha particles so that the cross section is continuously over a certain depth so that yield variations, as a function of target thickness, can be referred to composition changes. A limited depth that can be profiled when this energy is used is 2.7 μm which follows from the width of the resonance in figure 4.25. In case of depth profiling of more thick samples an other energy with both a broad and high resonant scattering cross section should be found.
4.9 Conclusion and Recommendations

It is concluded that CERDA-TOF is a useful technique for the depth profiling with a depth resolution of 5 nm, for light elements in a heavy matrix when incident alpha particles are used and the advantage of resonant scattering is taken into account. Contributions that limit good depth resolution were the beam divergence and energy spread of the incident particles in case of the depth resolution at the surface, and energy straggling and energy spread arising from multiple scattering for the depth resolution of buried layers and multi layered samples. The best depth resolution was 5 nm obtained from the depth profiling of a 15 µg/cm² (66 nm) carbon self-supporting thin film.

The CERDA-TOF technique was further developed for the depth profiling of thin films positioned on a thick carrier foil and showed the possibility of depth profiling without aggravation of the depth resolution. Furthermore a possibility of simultaneously depth profiling of multi elements has been shown and resulted in a depth resolution of 11 nm for the depth profiling of nitrogen in a 160 nm Si₃N₄ test sample. A carbon and oxygen growth was noted on both the aluminum layers and the Si₃N₄ test sample. The growth rate was approximately 2 monolayers in every 1000 seconds.

Moreover, research in order to depth profile more light elements in a heavy matrix should be continued. Furthermore it could be profitable when a higher beam current and smaller beam divergence could be obtained. An improvement of the vacuum is not necessary yet but might be requested for the depth profiling of carbon or in case that samples are investigated that may not be exposed to deposition of hydro-carbon molecules. A general aim should be the development of a powerful simulation program which is able to simulate CERDA-TOF spectra which might result in a better understanding of the different contributions and limitations to depth resolution below the surface.
REFERENCES

Stopping Power of C and Al ions in Solids.

Surface Modification Using a Cascade Arc Plasma Source.

Phys. Rev. 133 (1964) B1421-B1433
Elastic Scattering of Alpha Particles by C\textsuperscript{12} in the Bombarding Energy Range 10 to 19 MeV.

Academic Press (1978),
Backscattering Spectrometry.

Scattering of 18-MeV Alpha Particles by C\textsuperscript{12}, O\textsuperscript{16} and S\textsuperscript{32}.

A Simple Representation for the Angular Dependence of Scattered and Recoil Particle Energies.

Fundamentals of Surface and Thin Film Analysis.

Separate Determination of Concentration Profiles for Atoms with Different Masses by Simultaneous Measurement of Scattered Projectile and Recoil Atom Energies.

John Wiley and Sons, Inc. (1988)
Introductory Nuclear Physics.
Non-Rutherford $^4$He Cross Section for Ion Beam Analysis.

Reference Stopping Cross Section for Hydrogen and Helium ions in Selected Elements.

[Rau 89] E. Rauhala.  
Proton Backscattering and Computer Data Analysis in the Non-Rutherford Energy Region.

to be published, IBA 10 proceedings, Nucl. Instr. and Meth.  
Improved Depth Resolution in CERDA by Recoil Time Of Flight Measurement.

to be published, IBA 10 proceedings, Nucl. Instr. and Meth.  
Subnanosecond Timing with Ion Implanted Detectors.

[Sat 90] G.R. Satchler.  
Macmillian Education LTD. (1990)  
Introduction to Nuclear Reactions.

[Sig 74] P. Sigmund and K.B. Winterborn.  
Small-Angle Multiple Scattering of Ions in the Screened Coulomb Region.

Nuclear Reactions Resulting from the Proton Bombardment of Aluminum.

M. Sc. Thesis, Eindhoven University of Technology, the Netherlands  
Onderzoek naar tijdbepalende eigenschappen van verschillende PIPS-
detectoren.

$^{18}$F States with Large Isospin Impurities.

M. Sc. Thesis, Eindhoven University of Technology, the Netherlands 
Ionenverstrooiing met hoog energetische $\alpha$-deeltjes.

The Stopping and Ranges of Ions in Matter.

Emperical Formulae for Energy Loss Straggling of Ions in Matter.

The Stopping and Ranges of Ions in Solids.
Appendix

Appendix A: Kinematic Factors

The incoming projectile with mass $M_1$ has a incident energy $E_0$. Mass $M_2$ is at rest. After the collision projectile and atom have energy $E_1$ and $E_2$ respectively. The inelastic contribution $Q$ has a positive quantity. The projectile leaves at an angle $\theta$, the recoil atom is ejected at $\phi$ with respect to the direction of the incident projectile.

Conservation of energy:

$$E_0 = E_1 + E_2 + Q.$$  \hfill (A.1)

When $E$ is replaced by $p^2/2M$ conservation of momentum is given by:

$$p_0/2M_1 = p_1/2M_1 + p_2/2M_2 + Q.$$  \hfill (A.2)

Conservation of momentum:

$$p_0 = p_1 \cos \theta + p_2 \cos \phi$$ \hfill (A.3)

$$0 = p_1 \sin \theta - p_2 \sin \phi.$$  \hfill (A.4)

For further calculations A.3 and A.4 are used, $p_1$ and $p_2$ should be squared and added. If A.2 is substracted it follows for $p_1$ and $p_2$ respectively:

$$p_1^2 = p_0^2 - 2p_0 p_2 \cos(\phi) + p_2^2.$$  \hfill (A.5)

$$p_2^2 = p_0^2 - 2p_0 p_1 \cos(\theta) + p_1^2.$$  \hfill (A.6)

From energy conservation, equation A.2, follows for $p_1$ and $p_2$:

$$p_1^2 = p_0^2 - \frac{M_1}{M_2} p_2^2 - 2M_1 Q.$$  \hfill (A.7)

$$p_2^2 = \frac{M_2}{M_1} p_0^2 - \frac{M_1}{M_2} p_1^2 - 2M_2 Q.$$  \hfill (A.8)

Combination of previous equations and introducing the mass ratio $\mu$, equalling $M_2/M_1$, gives quadratic equations for $p_1$ and $p_2$:

$$p_2^2 (1 + \frac{1}{\mu}) - 2p_0 p_2 \cos \phi + 2M_2 Q = 0.$$  \hfill (A.9)

$$p_1^2 (1 + \mu) - 2p_0 p_1 \cos \theta + p_0^2 (1 - \mu) + 2M_2 Q = 0.$$  \hfill (A.10)
Solutions for these equations are found with use of the abc-formula. Note that \( E_1/E_0 \) equals \( p_1^2/p_0^2 \), but \( E_2/E_0 = (1/\mu)(p_2^2/p_0^2) \). First the \( K_1 \) is considered.

\[
K_1 = \frac{E_1}{E_0} = \left\{ \frac{\cos \theta \pm \sqrt{\cos^2 \theta -(1+\mu)(1-\mu+\mu Q/E_0)}}{(1+\mu)} \right\}^2
\]

This equation demands that the numerator has a positive quantity as it represents the momentum of the projectile. The argument of the square root should also have a positive quantity. These two requirements state that there is only one solution when:

\[
(1+\mu)(1-\mu+\mu Q/E_0) \leq 0
\]

This demand is fullfilled in the case that \( Q \leq (\mu-1)E_0/\mu \). For elastic collisions the \( Q \) value is limited to \( Q=\mu E_0/(1+\mu) \). As a consequence there are two possible solutions for \( (\mu-1)E_0/\mu \leq Q \leq \mu E_0/(1+\mu) \). There are always two solutions for the case that \( \mu<1 \).

The scattering angle \( \theta \) is limited because of the requirement of:

\[
(1+\mu)(1-\mu+\mu Q/E_0) \leq \cos^2 \theta
\]

From these calculations the next example can be derived. In case of a collision between an alpha projectile and carbon atom a possibility for an existing second solution is possible when the \( Q \) value is beyond \( 2/3 E_0 \). The maximum \( Q \) value is \( 3/4 E_0 \). In the case of 6 MeV alphas the 4.43 MeV excited state of the carbon atom gives two solutions for the energy after scattering inelastically.

The next consideration takes care of the description of the inelastical kinematic factor of the recoil atom.

It can easily be derived that:

\[
\frac{E_2}{E_0} = \frac{\mu}{(1+\mu)^2} \cos^2 \phi \left\{ 1 \pm \sqrt{1-(\mu+1)Q/\mu E_0 \cos^2 \phi} \right\}^2
\]

The argument of the square root should have a positive quantity and therefore \( Q \leq \mu E_0 \cos^2 \phi/(1+\mu) \) should hold. There are two possible solutions for the kinematic energy gained by the recoil atom in the collision.
Appendix B: Detector Tilt Angles

B.1 The Recoil Detector Tilt Angle

Equation 4.15 gives an equation for the recoil detector tilt angle:

\[
\tan \tau = \frac{1}{2} \frac{dK_2}{d\phi} \frac{E_0}{E_2}
\]

(B.1)

An expression for \( K_2 = E_2/E_0 \) is given by equation 2.2:

\[
K_2 = \frac{4\mu}{(1+\mu)^2} \cos^2\phi
\]

(B.2)

Furthermore the expression for the derivative of the recoil energy with respect to the scattering angle is:

\[
\frac{dK_2}{d\phi} = -\frac{8\mu \sin \phi \cos \phi}{(1+\mu)^2}
\]

(B.3)

Substitution of both B.2 and B.3 in B.1 gives:

\[
\tan \tau = \frac{1}{2} \frac{8\mu \sin \phi \cos \phi}{(1+\mu)^2} \frac{(1+\mu)^2}{4\mu \cos^2 \phi} = \tan \phi
\]

This will result in \( \tau = \phi \).

B.2 The Start Detector Tilt Angle

Calculations for the start detector tilt angle have been carried out. It has been shown in Chapter 4 that the following equations hold:

\[
\tan \tau_s = \frac{1}{2} \frac{dK_1}{d\theta} \frac{E_0}{E_1}
\]

(B.4)

\[
K_1 = \frac{E_1}{E_0} = \left( \frac{\cos \theta + \sqrt{\mu^2 - \sin^2 \theta}}{1+\mu} \right)^2
\]

(B.5)

\[
\frac{dK_1}{d\theta} = -\frac{2\sin \theta}{(1+\mu)^2} \left( \frac{2\cos \theta + \mu^2 + \cos(2\theta)}{\sqrt{\mu^2 - \sin^2 \theta}} \right)
\]

(B.6)
Finally substituting both B.5 and B.6 in B.4 results in:

$$\tan \tau_\alpha = \frac{\sin \theta}{\sqrt{\mu^2 - \sin^2 \theta}} \quad \text{(B.7)}$$

### B.3 Derivation of $\theta + \tau_\alpha = \theta_c$

Starting with the following equations:

$$\tan \theta = \frac{\mu \sin \theta_c}{1 + \mu \cos \theta_c} \quad \text{(B.8)}$$

$$\tan \tau_\alpha = \frac{\sin \theta}{\sqrt{\mu^2 - \sin^2 \theta}} \quad \text{(B.7)}$$

$$\cos \theta_c = -\frac{\sin^2 \theta}{\mu} + \cos \theta \sqrt{1 - \sin^2 \theta} \quad \text{(B.9)}$$

Substituting B.9 in B.7 results in:

$$\sin \theta_c = \frac{\sin \theta}{\mu} \left[ \cos \theta + \sqrt{\mu^2 - \sin^2 \theta} \right] \quad \text{(B.10)}$$

It is useful to start with:

$$\tan (\theta + \tau_\alpha) = \frac{\tan \theta + \tan \tau_\alpha}{1 - \tan \theta \cdot \tan \tau_\alpha} \quad \text{(B.13)}$$

The next step is the substitution of B.7 en B.8:
Finally substituting both B.9 and B.10 results in:

\[
\tan(\theta + \tau_a) = \frac{\mu \sin\theta_c}{\mu \cos\theta_c} = \tan\theta_c
\]  

(B.15)

which implies that \( \theta + \tau_a = \theta_c \).