Rutherford Backscattering Spectrometry

Part I: Why Ions? Ion-solid interactions
- Stopping and Range of Ions in Matter (SRIM)
- Rutherford backscattering spectrometry (RBS)
  - Introduction
  - Scattering geometry and kinematics
  - Rutherford cross section and limitations (non-Rutherford)
  - RBS spectra from thin and thick films
  - Stopping power and energy loss
  - Detector resolution
  - Energy straggling
  - Non-Rutherford cross sections

References:
3) The Stopping and Range of Ions in Matter (SRIM), http://www.srim.org/
4) SIMNRA. User’s Guide. Max Plank-Instut für Plasmaphysik

Part II: Treatment of experimental data

Elastic Collisions

(Eq.1) \[ \frac{1}{2} M_1 v_1^2 + \frac{1}{2} M_2 v_2^2 = \frac{1}{2} M_1 v'_1^2 + \frac{1}{2} M_2 v'_2^2 \]  
(Eq.2) \[ M_1 v_1 \cos \theta = M_1 v_1' \cos \phi \]  
(Eq.3) \[ 0 = M_1 v_1 \sin \theta - M_2 v_1' \sin \phi \]
From Eq. 2 and 3, eliminating \( \phi \) first, then \( v_2 \), one finds the ratio of particle velocities, and we can show that the energy of projectile \( (M_1) \) after collision can be found by the following relationship:

\[
E_2 = E_1 \left( \frac{M_1 - M_2 \sin \phi}{M_1 + M_2} \right)^2
\]

Ratio of \( E_1 \) and \( E_0 \) is called the **kinematic factor**:

\[
E_2 \sim E_1 \left( \frac{M_1 - M_2 \sin \phi}{M_1 + M_2} \right)^2
\]

Plot of the kinematic factor, \( k \), vs scattering angle for H\(^+\) scattering from various targets.

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**Advantages of Ion Beams**

- Can be used for material modification and analysis
- Mass Specific
  - Kinematic factor
  - Cross sections are very well known
- Good depth resolution
- Penetrating (can access buried interfaces)
- What about substrate?
  - can use channeling and blocking effects

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**Ion channeling and blocking**

Si (diamond structure)

- Si(111)
  - (011)
  - (110)
- Si(111) – side view
  - \( \overline{11} \)
Electrons vs Ions

When an ion collides with electron clouds in the solid, it does not lose much energy and its direction of motion is hardly change, in a contrast with electrons colliding with electrons.

18keV e⁻ and 18 keV He⁺ striking a Si₃N₄ layer with a SiO₂ substrate

SRIM

http://www.srim.org/ → Download SRIM-2013

SRIM Setup Window
Calculated Ion Trajectories

- 2MeV He⁺ in Si
- 50keV He⁺ in Si
- 50keV Au⁺ in Si

Tandetron Accelerator Laboratory (WSC G49)

- 1.7 MV Tandetron Accelerator
- PIXE system
- stripper gas
- Rutherford Backscattering System (RBS)
- Medium Energy (MEIS) Ion Scattering System
- Group III,V Molecular Beam Epitaxy System
- Group IV Molecular Beam Epitaxy System
- High Energy Magnet
Tandetron Operating Principle

1. Begin with negative ions via sputtering for most species, or He (\(H_2\)) gas
2. Accelerate to kinetic energy = \(qV\), where \(V\) = terminal voltage (MV) and \(q = -1\) so that \(E_t = V\) [MeV]
3. Ions traverse a stripper gas at the high voltage terminal to produce a charge state distribution of positive ions
4. Accel/decel mode is available when the stripper gas is OFF: used for \(E_{ion} \leq 100\) keV and the incident ions then have \(q = -1\)

Rutherford Backscattering Spectroscopy

- Typical parameters: 500keV-4MeV \(H^+, He^+\)
- Ion range: 1.5-14 \(\mu\)m
- Ion beam is close to surface normal
- Small fraction of incident ions will scatter back
  - Introduction
  - Scattering geometry and kinematics
  - Rutherford cross section and limitations (non-Rutherford)
  - RBS spectra from thin and thick films
  - Stopping power and energy loss - aside
  - Detector resolution
  - Energy straggling
  - Non-Rutherford cross sections

RBS- Rutherford Backscattering Spectrometry

- Widely used for near-surface layer analysis of solids
- Elemental composition and depth profiling of individual elements
- Quantitative without reference sample (unlike SIMS, XPS with depth profiling)
- Non-destructive (unlike SIMS, XPS with depth profiling)
- Analyzed depth: \(~2\mu m\) for He ions
- Very sensitive for heavy elements: \(~ppm\)
- Less sensitive for light elements ⇒ ERD
History

Sir Ernest Rutherford (1871 - 1937)

- 1911: Rutherford’s scattering experiments: \(^4\text{He}\) on \(\text{Au}\)
  - Atomic nucleus, nature of the atom

RBS as materials analysis method


“Nuclear scattering and nuclear reactions induced by high energy protons and deuterons have been applied to the analysis of solid surfaces. The theory of the scattering method, and determination of O, Al, Si, S, Ca, Fe, Cu, Ag, Ba, and Pb by scattering method are described. C, N, O, F, and Na were also determined by nuclear reactions other than scattering. The methods are applicable to the detection of all elements to a depth of several \(\mu\text{m}\), with sensitivities in the range of \(10^{-8}\) to \(10^{-6}\ \text{g/cm}^2\)."

RBS: Scattering geometry and kinematics

\[ E_f = k E_o \]

\[ k = \frac{E_f}{E_o} = \left[ \frac{(M_1 - M_1 \sin^2 \theta)^{1/2} \times M_2 \cos \theta}{M_1 + M_2} \right] \]

- \(\alpha\): incident angle
- \(\beta\): exit angle
- \(\theta\): scattering angle

Optimized mass resolution for:

\(160^\circ < \theta < 170^\circ\)

Scattering Geometry

IBM geometry

Cornell geometry → Choose this one for RBS Tandetron experiments
Scattering kinematics: example 1

2 MeV \(^{4}\)He\(^{+}\) \(\theta=165^\circ\)
Backscattered from C, O, Fe, Mo, Au
3 \times 10\(^{16}\) atoms/cm\(^{2}\) each on Si substrate

Key features of RBS

Ability to quantify depth profile of buried species with a precision of ~ 3%

Qualitative information: kinematic factor, \(k\)
\[
k = \frac{E_s}{E_o} \left[ \left( \frac{M_2 - M_1 \sin^2 \theta}{M_1 + M_1} \right)^2 + \frac{M_1 \cos \theta}{M_1 + M_1} \right]\]

Quantitative: scattering cross section, \(\sigma\)
\[
\frac{d\sigma}{d\Omega} = \left( \frac{Z_o Z_s \mu}{4E \sin \left( \frac{\theta}{2} \right)} \right)^2
\]

Scattering kinematics: example 2

\(\Rightarrow\) Decreased mass resolution for heavier elements
Rutherford Cross Section

- Neglecting shielding by electron clouds
- Distance of closest approach large enough that nuclear force is negligible

Rutherford scattering cross section

\[ \frac{d\sigma}{d\Omega} = \sigma(\theta) = \frac{Z_1 Z_2 e^4}{4\varepsilon_0 m_e c^2 \sin^2 \left( \frac{\theta}{2} \right)} \]

Note that sensitivity increases with:
- Increasing \( Z_1 \)
- Increasing \( Z_2 \)
- Decreasing \( E \)

Scattering kinematics: example 1

2MeV 4He\(^+\) \( \theta = 165^\circ \)
Backscattered from C, O, Fe, Mo, Au
3\( \times 10^{14} \) atoms/cm\(^2\) each on Si substrate

- Increased sensitivity for heavier elements
- Good for heavier elements on lighter substrates
- Bad for light elements on heavier substrates

RBS spectra from thin and thick films

The integrated peak count \( A_i \) for each element on the surface can be calculated using this equation:

\[ A_i = \left( Nt \right) \times Q \times T \times \frac{\sigma(E, \theta)}{\cos\theta} \]

where
- \( N \) is areal density, atoms per unit area;
- \( Q \) - ion beam fluency;
- \( T \) - solid angle of the detector;
- \( \sigma(E, \theta) / \cos\theta \) - cross section of an element
Ion dose (fluence), solid angle, cross section

- Ion dose (fluence), the number of incident particles (collected charge)
  - measured by Faraday cup
  - \( Q = I \times t \)
- Solid angle, in steradians, \( \text{sr} \)
  - stays constant for a particular detector/detector slit
  - need to be verified by the calibration standard measurements
- Cross section (or differential cross section), in \( \text{cm}^2/\text{sr} \) of the element
  - well known (tabulated) in Rutherford cross section regime

Areal density: note about units

Areal density = \( \rho \, t \, [\text{g/cm}^2] \),
where \( \rho = \text{g/cm}^3 \), \( t = \text{cm} \)

\[ \frac{N_0 \, \rho \, t}{M} \, [\text{at./cm}^2] \]
where \( M = \text{atomic mass \, [amu]} \), \( N_0 = \text{Avogadro's number} \)
In absolute numbers – close to thickness in Å

RBS Spectrum of a thick film

- Target is divided into thin sublayers (“slabs”)
- Calculate backscattering from front and back side of each sublayer taking energy loss into account
- For each isotope of each element in sublayer

\[ E_i \]
\[ E_f \]
\[ E_{	ext{coul}} \]
\[ E_{	ext{inel}} \]
RBS spectrum: bad choice of the substrate

RBS Spectrum: Stoichiometry

2MeV 4He+, backscattered from ceramic films on Si substrate

Ion channeling and blocking

Si (diamond structure)
Use crystal structure of the substrate

- Substrate can be aligned to a major crystallographic direction to minimize background signal in some cases

Silicon detector resolution

Principle of operation:
- Creation of electron-hole pairs by charged particles
- Separation of electron-hole pairs by high voltage \( V \)
  \[ \Rightarrow \text{Number of electron-hole pairs} \propto \text{Particle energy} \]
  \[ \Rightarrow \text{Charge pulse} \propto \text{Particle energy} \]

Limited energy resolution (~1keV/100Å) due to:
- Statistical fluctuations in energy transfer to electrons and phonons
- Statistical fluctuations in annihilation of electron-hole pairs

Additional energy broadening due to:
- Preamplifier noise
- Other electronic noise

Depth resolution and concentration profiling

Depth resolution for 100 keV protons (resolution of the detector=1keV)
- Stopping power \( \text{SiO}_2 \approx 12 \text{ eV/Å}; \text{Si}_3\text{N}_4 \approx 20 \text{ eV/Å}; \text{Ta}_2\text{O}_5 \approx 18 \text{ eV/Å} \)
- "Near surface" depth resolution \( \approx 50 \text{ Å}; \) worse for deeper layers due to energy straggling

- Areas under each peak corresponds to the concentration of the element in a 50Å slab
- Peak shapes and positions come from energy loss, energy straggling and instrumental resolution
- The sum of the contributions of the different layers describes the depth profile.
Energy Straggling

Slowing down of ions in matter is accompanied by a spread of beam energy ⇒ energy straggling

Electronic or nuclear energy loss straggling due to statistical fluctuations in the transfer of energy to electrons or nuclear energy loss

Energy after penetrating a layer \( \Delta x \): \(<E> = E_0 - s \Delta x\)

\(<E>\) - mean energy; \(s\) - stopping power

⇒ only applicable for mean energy of many particles

Non-Rutherford cross sections

Typical problem for light elements: overlap with thick layers of heavier elements ⇒ High cross sections wishful!!!

Elastic Recoil Detection (ERD)

\(^{4}\text{He}, E_r = 1800\text{ keV}\)

\(^{2}\text{He}, E_r < 870\text{ keV}\)

\(^{3}\text{He}, E_{	ext{det}} < 600\text{ keV}\)
Part II: Treatment of experimental data

To do list:
- Download and install SIMNRA (use 26100 to register)
- File: "follow.xls"
- Data file for SiSb standard
- Data file for unknown sample
- Data files for your known samples

1. Download SIMNRA 6.0 software from http://home.rzg.mpg.de/~mam/Download.html
2. Install it (as a Demo version) on your laptop.
3. After installation, run the program, go to Help -> Register, and enter "26100" as a registration code.

SIMNRA main menu

SiSb standard
Some trivial examples: common pitfalls and their origin
Simulation codes and user knowledge
Over-interpretation and under-interpretation
What to do? – the best and the worse practices

Trivial pitfalls 1: assumptions about experimental parameters

- Scattering angle
- Angle of incidence
- Solid angle, beam fluence/flux
- Energy calibration

How to solve/to remedy the problem:
- Acquire data at 2-3 different detector scattering angles, and find consistent positions
- Treat experimental parameters as fitting parameters
- If possible – find a channeling/major crystallographic direction and relate it to the incident angle
- Collect more data with different ion doses
- Measure independent calibration standards

Layer mixing: GaInN/GaN on Al₂O₃

- Film interacts with the substrate leading to extensive intermixing
Roughness: GaInN/GaN on Al$_2$O$_3$

- Very few samples have atomically flat surface
- Very often “diffusion” or “mixing” is actually roughness
- Lateral homogeneity has to be proven with other methods than IBA

Trivial pitfalls 2: Assumptions about the sample

- Roughness
- Missing element - ignored or falsely postulated
- Postulate your favorite depth profile

How to solve/to remedy these problems:

- Use complementary techniques
- Measure samples are different energies
- Keep an open mind?

RBS: SiO$_2$ on SiC

Make sure to use right cross sections in the non-Rutherford regime
Trivial pitfalls 3: assumptions about the basic data used

- Scatter cross sections in the non-Rutherford regime
- Stopping powers (including use of Bragg rules)

How to solve/to remedy these problems:
- Try simulations with different cross sections
- Accept lower accuracy than achievable (often the only practical alternative)

Less trivial data analysis pitfalls

Over-interpretation
- Analyst unjustifiably imposes a given model on the data, when other models would also lead to an equivalent or better solution

Under-interpretation
- Analyst does not extract all information that could be extracted, often due to lack of knowledge or lack of proper code/cross section

In general
- A good fit does not mean analysis is reliable
- Good use of a suitable code requires extensive knowledge

Over-interpretation
- Nominal (Ti_{0.4}Al_{0.6}N 5 nm/Mo 5nm) ×50
- Simulation consistent with extended layer intermixing
Under-interpretation: same sample

• AFM and TEM for few samples
• Reasonable report quantifies roughness for all the samples

The best and the worse practices

➢ Trust a computer
  • all codes have limitations, sometimes severe
  • the more advanced features you need, the more knowledge you need to justify using them

➢ Take the stopping powers and cross sections for granted
  • you must be aware of the values used in your analysis

➢ Codes are a tool
  • you are the analyst: know your system, know your parameters