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Introduction to Ion Beam Analysis: General Physics

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Introduction to Ion Beam Analysis:
General Physics

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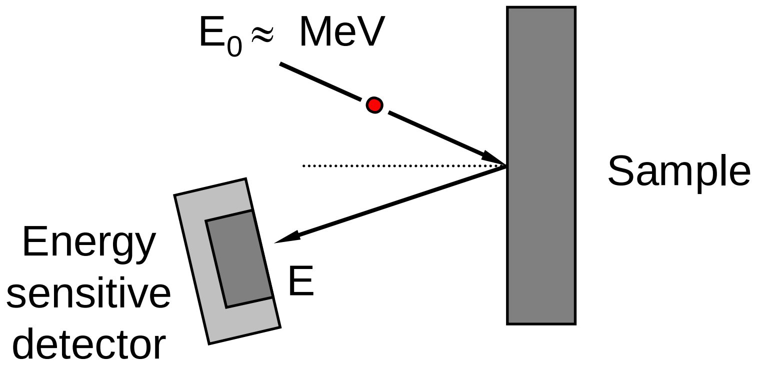
- **History of IBA**
- **Scattering kinematics**
- **Rutherford cross-section and beyond**
- **Stopping**
- **Energy spread:**
  - **Detector resolution**
  - **Energy straggling**
  - **Multiple scattering**
- **Depth resolution**

Introduction

IBA – Ion Beam Analysis

- Group of methods for the near-surface layer analysis of solids
- Elemental composition and depth profiling of individual elements
- Quantitative without reference samples
- No (or small) matrix effects
- Non-destructive
- Analysed depth: few 100 nm (heavy ions) to 40 µm (protons, high energy NRA)
- High sensitivity ≈ ppm



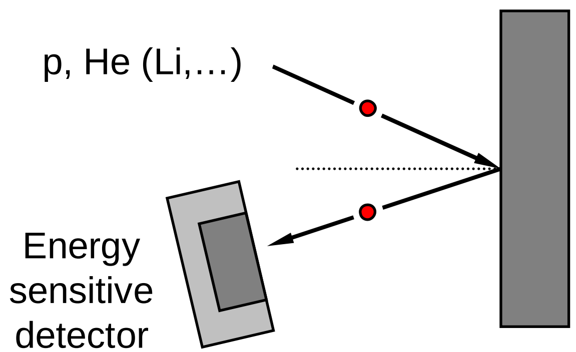
IBA methods: Detection of charged particles

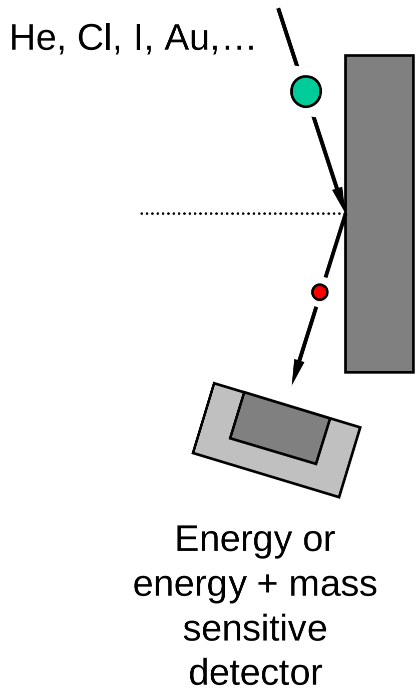
Common for all IBA methods: Incident particles with ~MeV energies

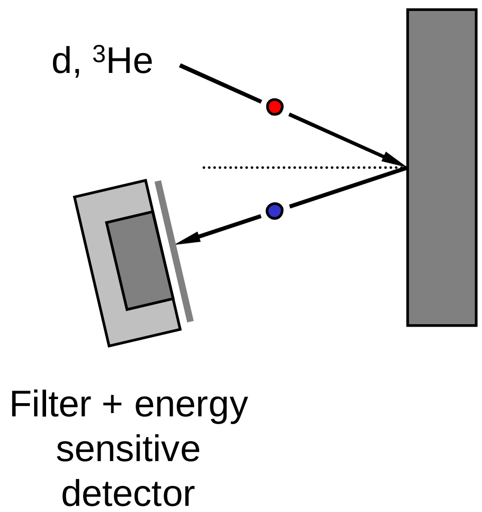
⇒ Methods can often be used in the same experimental setup

⇒ Methods can sometimes be used simultaneously

Rutherford backscattering (RBS)	**Elastic recoil detection analysis (ERDA)**	**Nuclear reaction analysis (NRA)**





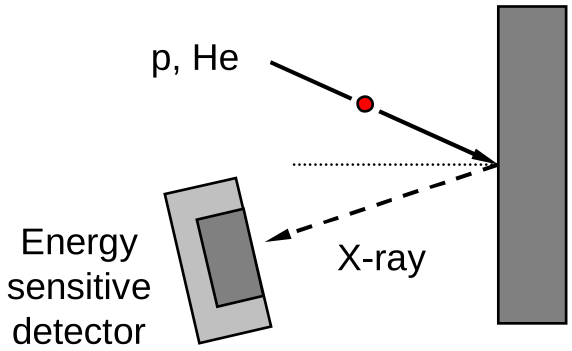


d(^{3}He,p)α

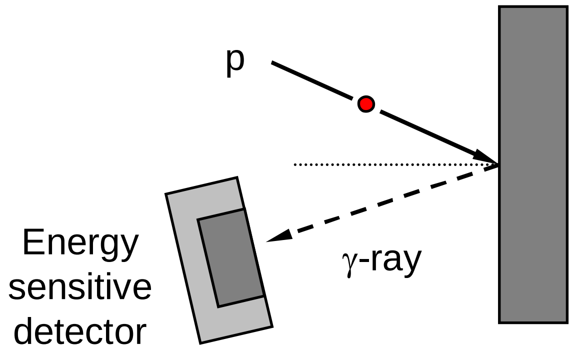
^{16}O(d,p)^{17}O

IBA methods: Detection of photons

Particle induced X-ray emission
(PIXE)



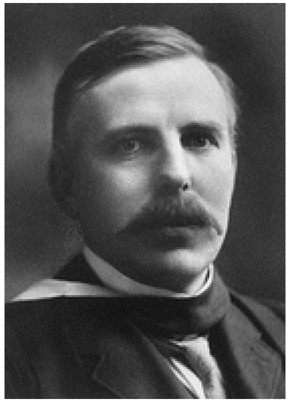
Particle induced γ-ray emission
(PIGE)



Ion beam Analysis (IBA) acronyms: G. Amsel, Nucl. Instr. Meth. B118 (1996) 52

History

Sir Ernest Rutherford (1871 - 1937)



- 1909: Rutherford’s scattering experiments: ^{4}He on Au

  ⇒ Atomic nucleus, nature of the atom

RBS as materials analysis method

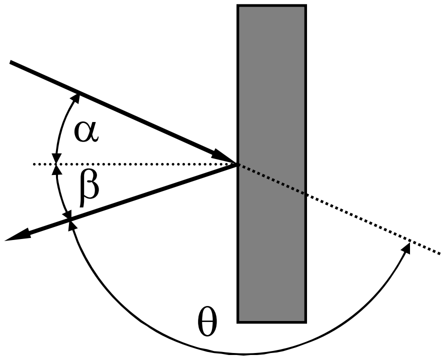
- 1957: S. Rubin, T.O. Passell, E. Bailey, “Chemical Analysis of Surfaces by Nuclear Methods”, *Analytical Chemistry* 29 (1957) 736

“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 µm, with sensitivities in the range of 10^{-8} to 10^{-6} g/cm^2.”

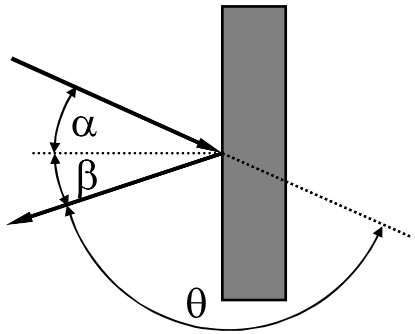
History (2)

- 1970's: RBS becomes a popular method due to invention of
  silicon solid state detectors (J.W. Mayer and B.R. Gossick 1956)
- 1976: J. L'Écuyer et al.
  Invention of ERDA
- 1977 - 1979: Compilations of stopping power and cross-section data
  H.H. Andersen and J.F. Ziegler, Stopping Powers of H, He in All Elements
  R.A. Jarjis, Nuclear Cross Section Data for Surface Analysis
- 1977: J.W. Mayer and E. Rimini
  Ion Beam Handbook for Materials Analysis
- 1985: M. Thompson
  Computer code RUMP for analysis of RBS spectra
- since 1997: M. Mayer, N.P. Barradas
  Advanced computer codes for analysis of RBS, ERDA, NRA spectra
- since 2003: Ion beam analysis nuclear data library (IBANDL)
  http://www-nds.iaea.org/ibandl/

Scattering Geometry



α: incident angle
β: exit angle
θ: scattering angle

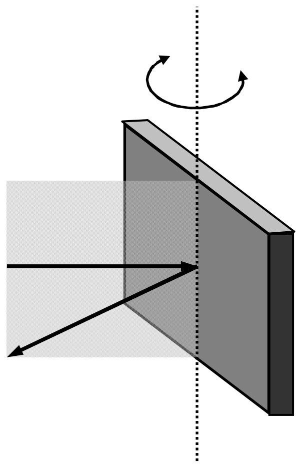


IBM geometry

Incident beam, exit beam, surface normal in one plane

$\Rightarrow \alpha + \beta + \theta = 180°$

Advantage: Simple



Cornell geometry

Incident beam, exit beam, rotation axis in one plane

$\Rightarrow \cos(\beta) = -\cos(\alpha)\cos(\theta)$

Often used in RBS

Advantage:

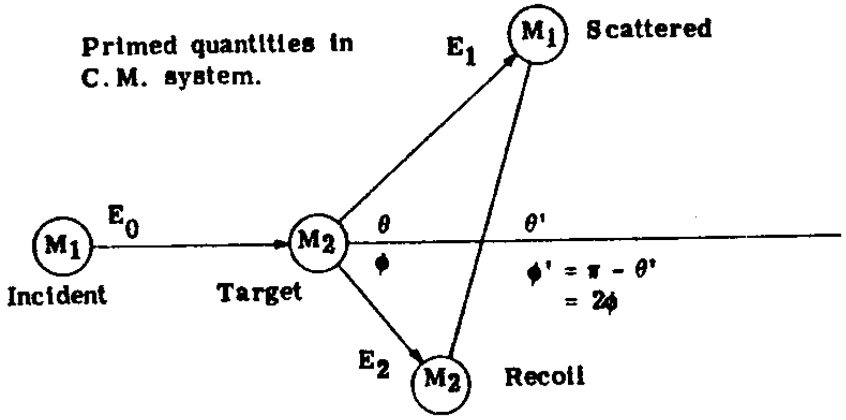
large scattering angle and grazing incident + exit angles

good mass and depth resolution simultaneously

General geometry

α, β, θ not related

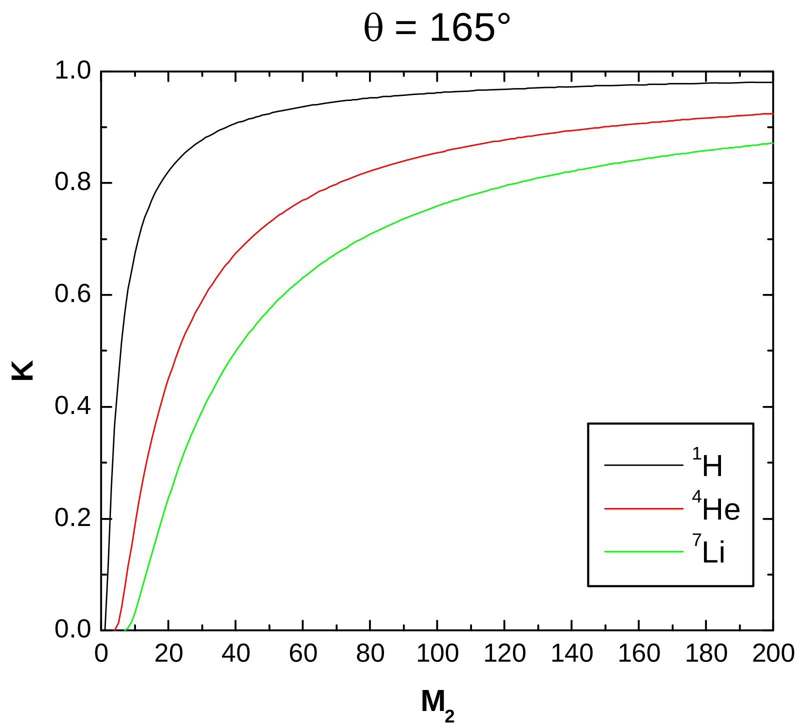
RBS: Scattering Kinematics



Elastic scattering: $E_0 = E_1 + E_2$

Kinematic factor: $E_1 = K\,E_0$

$$K = \left[\frac{\left(M_2^2 - M_1^2 \sin^2\theta\right)^{1/2} + M_1 \cos\theta}{M_1 + M_2} \right]^2$$



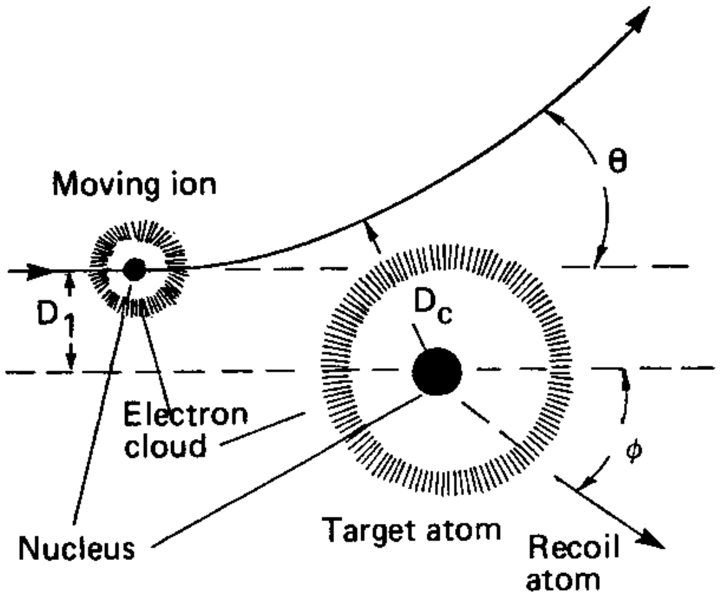
$$\Delta E_1 = E_0 \frac{dK}{dM_2} \Delta M_2$$

ΔE_1: Energy separation

ΔM_2: Mass difference

⇒ **Decreased mass resolution for heavier elements**

Rutherford Cross Section



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

  ⇒ Rutherford scattering cross section

$$\sigma_R(E,\theta) = \left(\frac{Z_1 Z_2 e^2}{4E}\right)^2 \times \frac{4\left[\left(M_2^2 - M_1^2 \sin^2\theta\right)^{1/2} + M_2 \cos\theta\right]^2}{M_2 \sin^4\theta \left(M_2^2 - M_1^2 \sin^2\theta\right)^{1/2}}$$

Note that: $\sigma_R \propto \frac{Z_1^2 Z_2^2}{E^2}$

Sensitivity increases with
- increasing Z_1
- increasing Z_2
- decreasing E

Shielded Rutherford Cross Section

Shielding by electron clouds gets important at

- low energies
- low scattering angles
- high Z_2

Shielding is taken into account by a shielding factor $F(E, \theta)$

$$\sigma(E,\theta) = F(E,\theta)\,\sigma_R(E,\theta)$$

$F(E, \theta)$ close to unity

$F(E, \theta)$ is obtained by solving the scattering equations for a shielded interatomic potential:

$$V(r) = \frac{Z_1 Z_2 e^2}{r}\varphi(r/a)$$

φ: Screening function

Use Thomas-Fermi or Lenz-Jenssen screening function

a: Screening radius $a = 0.885 a_0 \left(Z_1^{2/3} + Z_2^{2/3}\right)^{-1/2}$

a_0: Bohr radius

Shielded Rutherford Cross Section (2)

For 90° ≤ θ ≤ 180°:

L'Ecuyer et al. (1979)

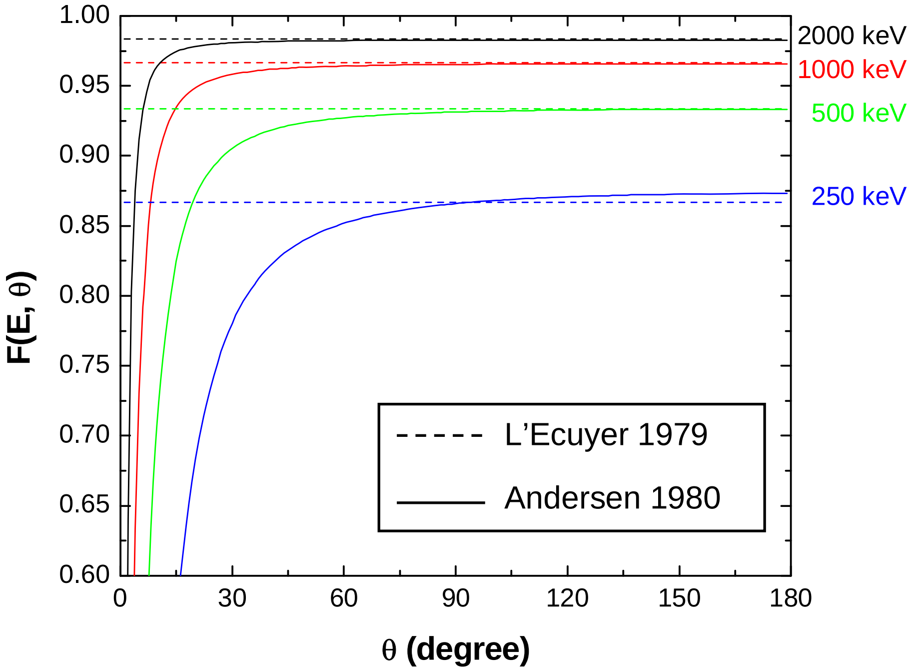
$$\frac{\sigma}{\sigma_R} = 1 - \frac{0.049\, Z_1 Z_2^{4/3}}{E_{CM}}$$

Wenzel and Whaling (1952)

$$\frac{\sigma}{\sigma_R} = 1 - \frac{0.0326\, Z_1 Z_2^{7/2}}{E_{CM}}$$

For all θ: Andersen et al. (1980)

^{4}He backscattered from Au



Non-Rutherford Cross Section

Cross section becomes non-Rutherford if nuclear forces get important

- high energies
- high scattering angles
- low Z_2

Energy at which the cross section deviates by > 4% from Rutherford at $160° \leq \theta \leq 180°$

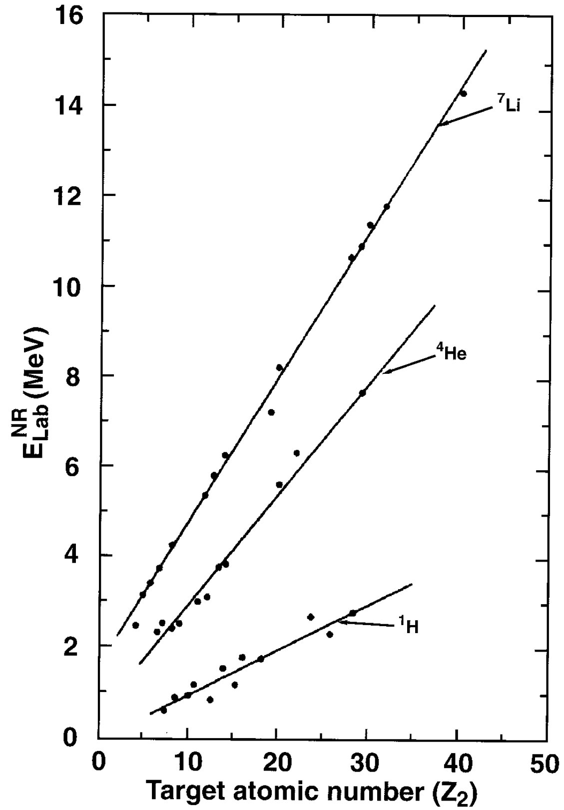
Bozoian (1991)

^{1}H: $E^{NR}_{Lab} = 0.12\, Z_2 - 0.5$ [MeV]

^{4}He: $E^{NR}_{Lab} = 0.25\, Z_2 + 0.4$ [MeV]

Linear Fit to experimental values (^{1}H, ^{4}He) or optical model calculations (^{7}Li)

Accurate within ± 0.5 MeV



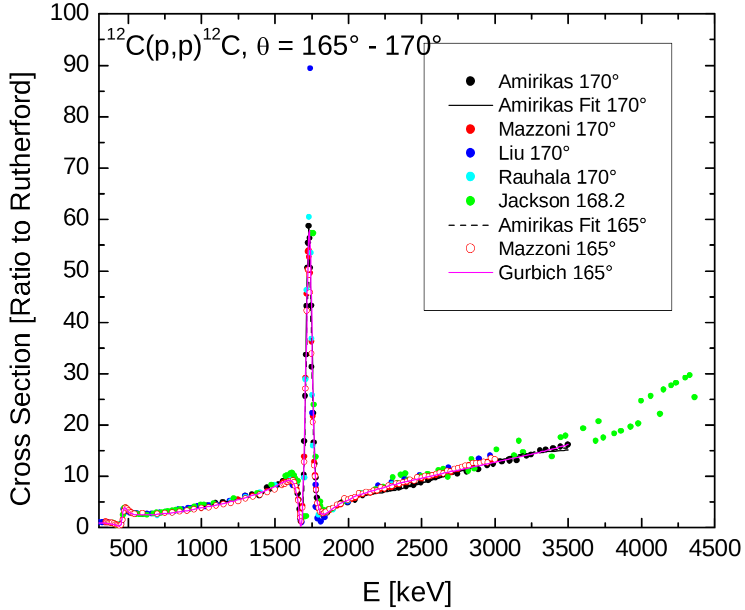
J.R. Tesmer and M. Nastasi,
Handbook of Modern Ion Beam Materials Analysis,
MRS, 1995

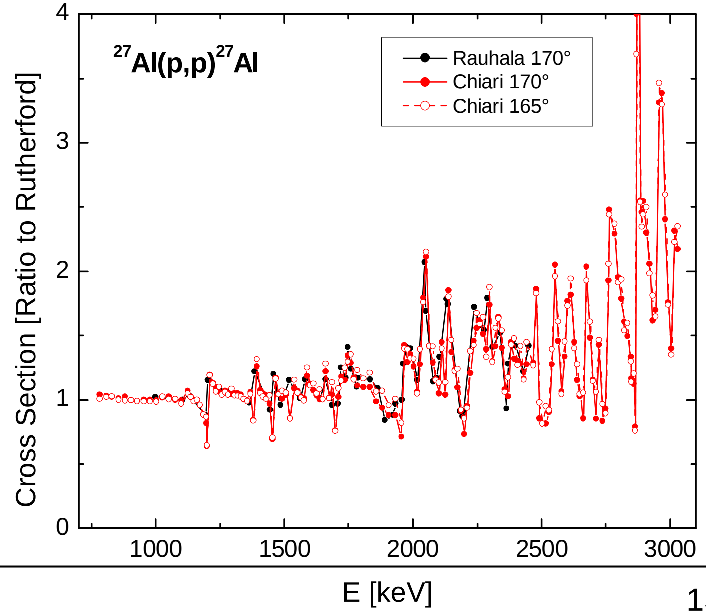
Non-Rutherford Cross Section (2)

Deviations from Rutherford may be huge

⇒ Carefully check for

possible deviations from Rutherford

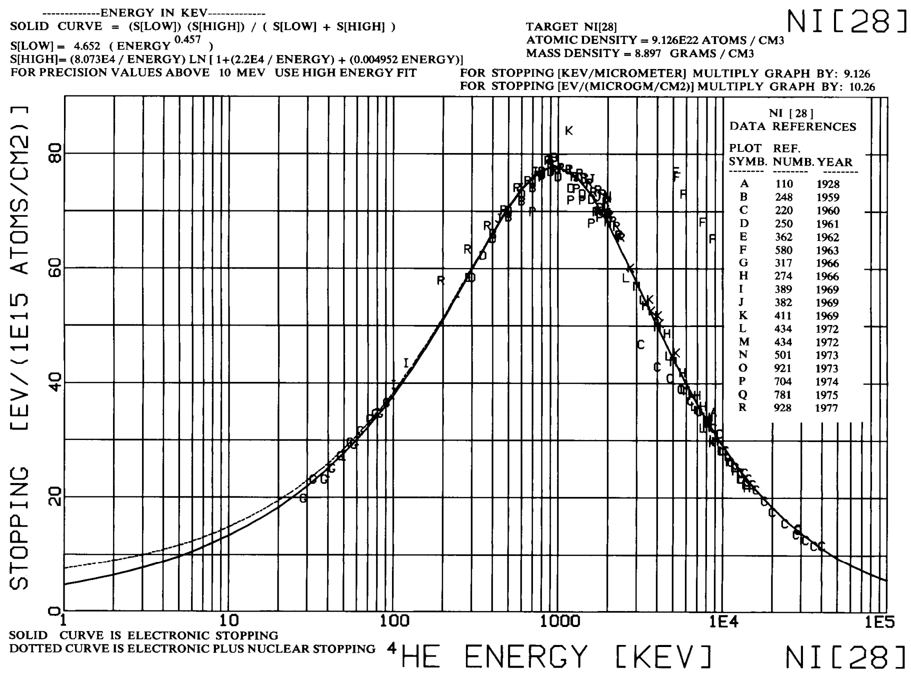




Stopping Power

^{4}He in Ni

- **Electronic stopping power:**
  - Andersen, Ziegler (1977):
    H, He in all elements
  - Ziegler, Biersack, Littmarck (1985):
    All ions in all elements
  - Several SRIM-versions since then
  - Additional work by Kalbitzer, Paul, ...
  - Accuracy: 5% for H, He
    10% for heavy ions
- **Nuclear stopping power:**
  - Only important for heavy ions
    or low energies
  - Ziegler, Biersack, Littmarck (1985):
    All ions in all elements using ZBL potential



J.F. Ziegler, Helium - Stopping Powers and Ranges in All Elements, Vol. 4, Pergamon Press, 1977

Stopping Power (2)

Stopping in compounds:

consider compound A_mB_n, with m + n = 1

S_A is stopping power in element A

S_B is stopping power in element B

What is stopping power S_{AB} in compound?

Bragg's rule (Bragg and Kleeman, 1905):

$$S_{AB} = m\, S_A + n\, S_B$$

Bragg's rule is accurate in:

- Metal alloys

Bragg's rule is inaccurate (up to 20%) in:

- Hydrocarbons
- Oxides
- Nitrides
- ...

Other models for compounds

- Hydrocarbons:
  Cores-and-Bonds (CAB) model
  Ziegler, Manoyan (1988)
  Contributions of atomic cores
  and chemical bonds between atoms
- Large number of experimental data,
  especially for hydrocarbons, plastics, ...

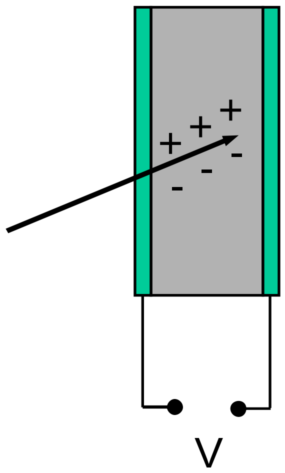
Silicon Detector Resolution

Principle of operation:

- Creation of electron-hole pairs by charged particles
- Separation of electron-hole pairs by high voltage V

$\Rightarrow$ Number of electron-hole pairs $\propto$ Particle energy

$\Rightarrow$ Charge pulse $\propto$ Particle energy



Limited energy resolution due to:

- Statistical fluctuations in energy transfer to electrons and phonons
- Statistical fluctuations in annihilation of electron-hole pairs

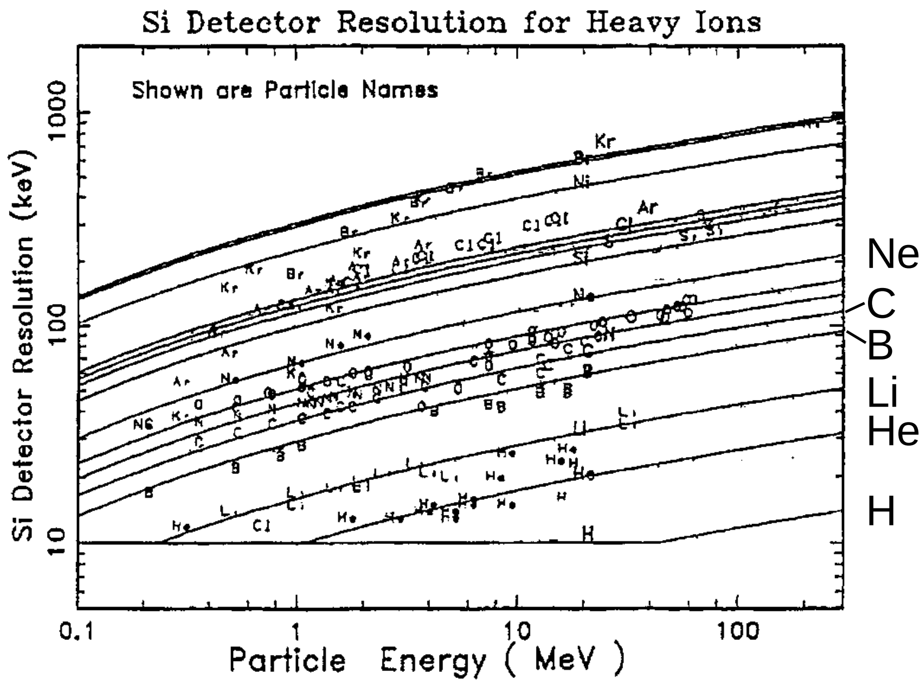
Additional energy broadening due to:

- Preamplifier noise
- Other electronic noise

Silicon Detector Resolution (2)

Typical values (FWHM):

- H   2 MeV: 10 keV
- He 2 MeV: 12 keV
- Li   5 MeV: 20 keV



J.F. Ziegler, Nucl. Instr. Meth. B136-138 (1998) 141

Ad-hoc fit to experimental data:

$$\text{FWHM resolution (keV)} = C_1(Z_1)^{C2}(\ln E_{\text{keV}})^{C3} - C_4(Z_1)^{C5}/(\ln E_{\text{keV}})^{C6}$$

$C_n = 0.0999288,\ 1.1871,\ 1.94699,\ 0.18,\ 2.70004,\ 9.29965$

Better energy resolution for heavy ions can be obtained by:

- Electrostatic analyser (Disadvantage: large)
- Magnetic analyser (Disadvantage: large)
- Time-of-flight (Disadvantage: length, small solid angle)

Energy Straggling

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

$\Rightarrow$ **energy straggling**

- Electronic energy loss straggling due to statistical fluctuations in the transfer of energy to electrons
- Nuclear energy loss straggling due to statistical fluctuations in the nuclear energy loss
- Geometrical straggling due to finite detector solid angle and finite beam spot size
- Multiple small angle scattering

Electronic Energy Loss Straggling

Due to statistical fluctuations in the transfer of energy to electrons

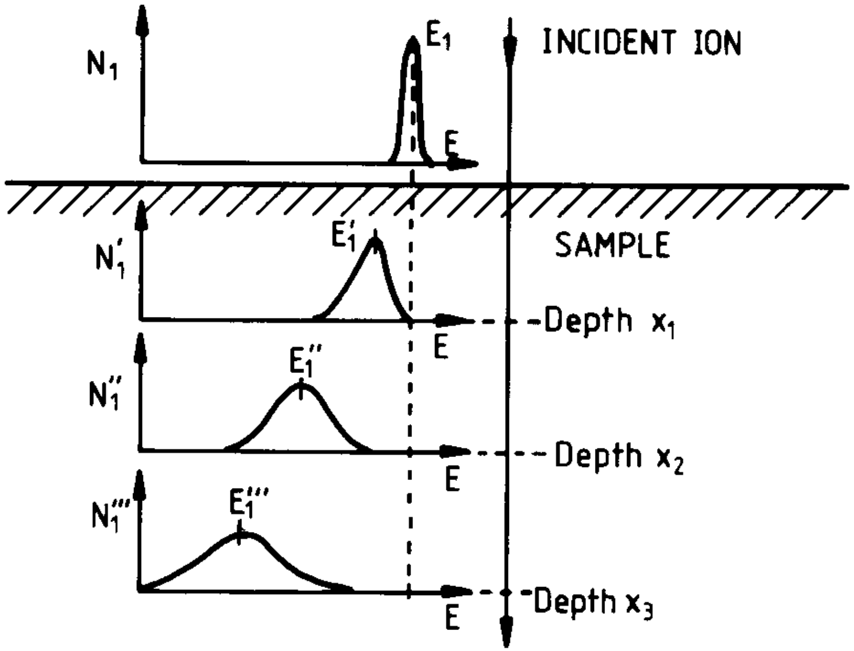
$\Rightarrow$ statistical fluctuations in energy loss

Energy after penetrating a layer Δx: $\langle E \rangle = E_0 - S\,\Delta x$

$\langle E \rangle$ mean energy

S stopping power

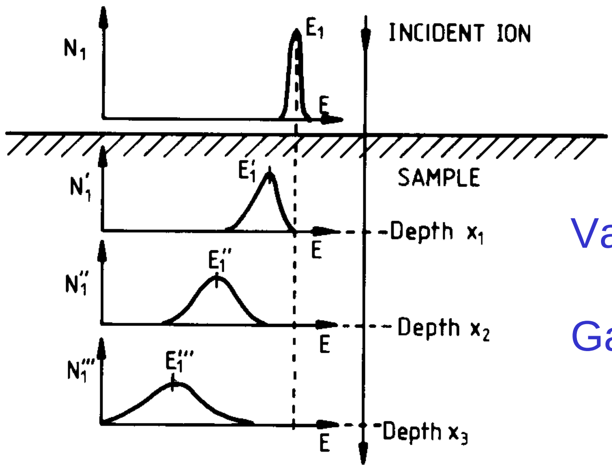
$\Rightarrow$ only applicable for mean energy of many particles



Electronic Energy Loss Straggling (2)

Shape of the energy distribution?

$\Delta E/E_0$		
< 10%	Vavilov theory	Low number of ion-electron collisions Non-Gaussian
10% – 20%	Bohr theory	Large number of ion-electron collisions Gaussian
20% – 50%	Symon, Tschalär theory	Non-stochastic broadening due to stopping Almost Gaussian
50% – 90%	Payne, Tschalär theory	Energy below stopping power maximum Non stochastic squeezing due to stopping Non-Gaussian



Vavilov

Gaussian

Vavilov Theory

Vavilov 1957

P.V. Vavilov, Soviet Physics J.E.T.P. **5** (1957) 749

Valid for small energy losses

⇒ Low number of ion-electron collisions

⇒ Non-Gaussian energy distribution with tail towards low energies

Mostly replaced by Bohr's theory and approximated by Gaussian distribution

⇒ Total energy resolution near the surface usually dominated by detector resolution due to small energy loss straggling

⇒ Only necessary in high resolution experiments

Bohr Theory

Bohr 1948

N. Bohr, Mat. Fys. Medd. Dan. Vid. Selsk. **18** (1948)

Valid for intermediate energy losses

⇒ Large number of ion-electron collisions

⇒ Gaussian energy distribution

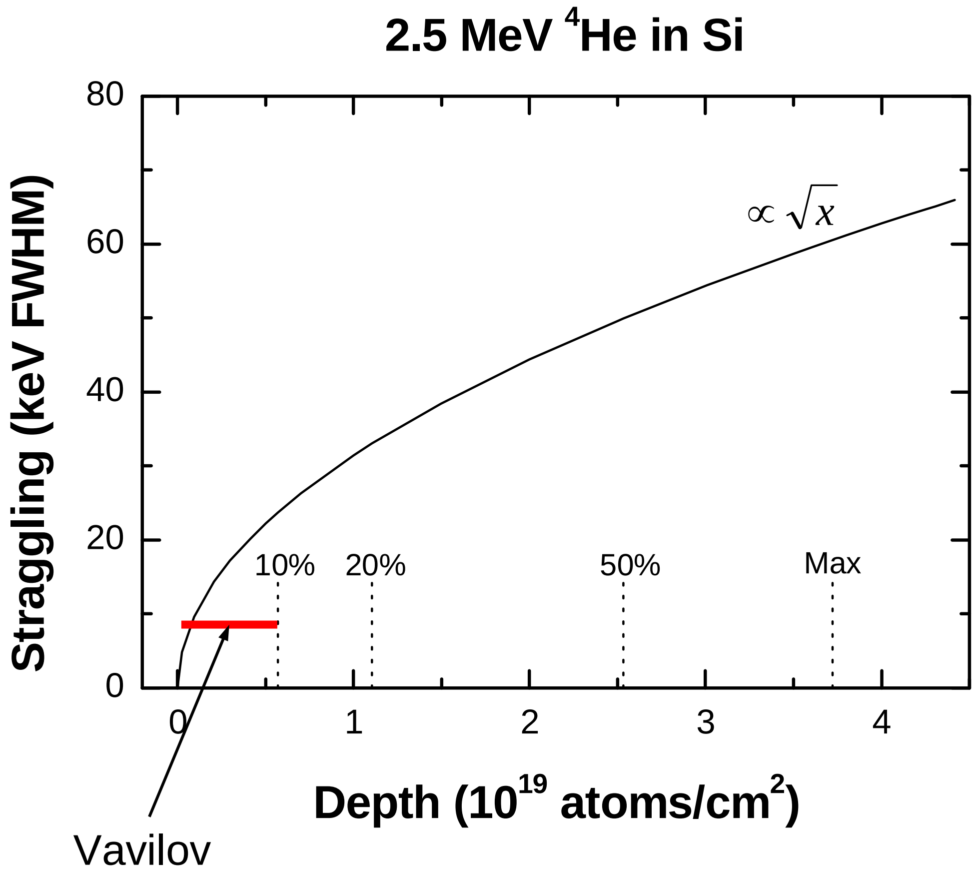
Approximations in Bohr's theory:

- Ions penetrating a gas of free electrons
- Ions are fully ionised
- Ion velocity >> electron velocity ⇒ stationary electrons
- Stopping power effects are neglected

$$\sigma_{Bohr}^2 = 0.26\, Z_1^2\, Z_2\, \Delta x$$

σ^2_{Bohr}	variance of the Gaussian distribution [keV^2]
Δx	depth [10^{18} atoms/cm^2]
Z_1	atomic number of incident ions
Z_2	atomic number of target atoms

Bohr Theory: Example



Improvements to Bohr Theory

Chu 1977

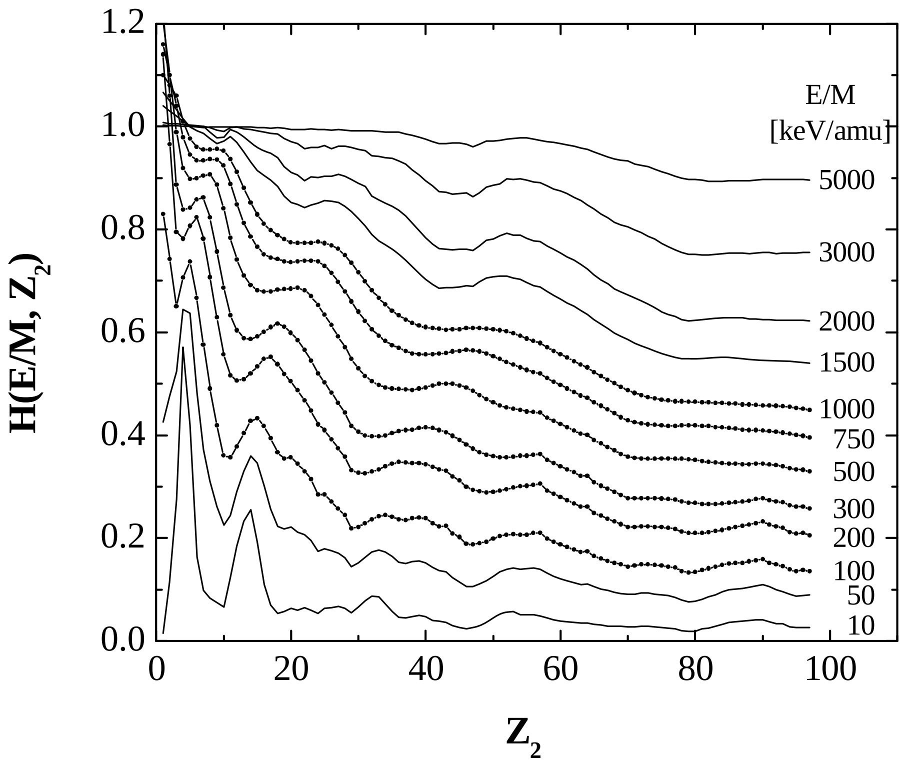
J.W. Mayer, E. Rimini, Ion Beam Handbook for Material Analysis, 1977

Approximations in Chu's theory:

- Binding of electrons is taken into account
- Hartree-Fock electron distribution
- Stopping power effects are neglected

$$\sigma_{Chu}^2 = H\left(\frac{E}{M_1}, Z_2\right)\sigma_{Bohr}^2$$

⇒ Smaller straggling than Bohr



Straggling in Compounds

Additive rule proposed by Chu 1977

J.W. Mayer, E. Rimini, Ion Beam Handbook for Material Analysis, 1977

consider compound A_mB_n, with m + n = 1

σ_A^2 is variance of straggling in element A

σ_B^2 is variance of straggling in element B

$$\sigma_{AB}^2 = \sigma_A^2 + \sigma_B^2$$

σ_A^2 Straggling in element A in a layer $m\,\Delta x$

σ_B^2 Straggling in element B in a layer $n\,\Delta x$

Propagation of Straggling in Thick Layers

Consider particles with energy distribution

- **Energy above stopping power maximum**

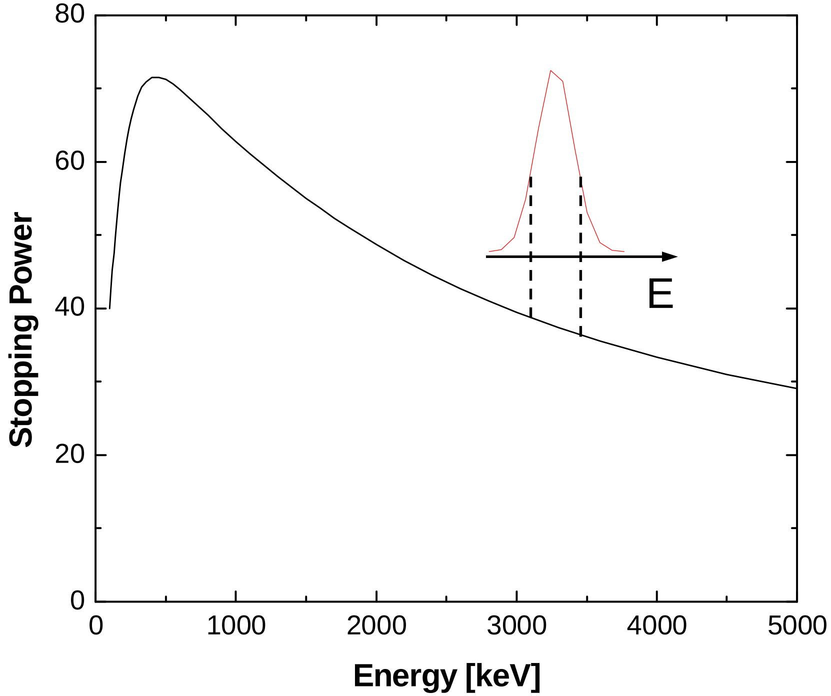
  higher energetic particles ⇒ smaller stopping

  ⇒ non-stochastic broadening

- **Energy below stopping power maximum**

  higher energetic particles ⇒ larger stopping

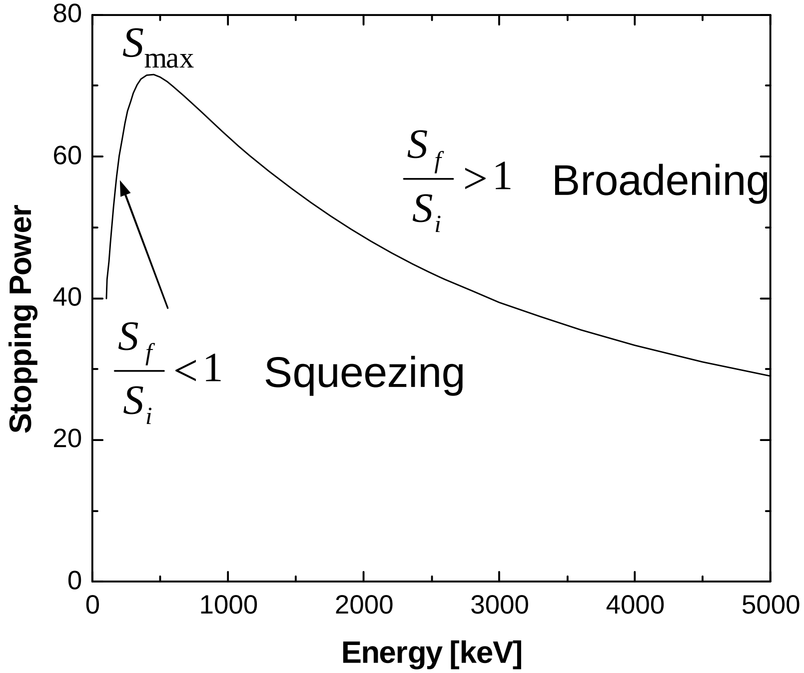
  ⇒ non-stochastic squeezing



C. Tschalär, Nucl. Instr. Meth. **61** (1968) 141

M.G. Payne, Phys. Rev. **185** (1969) 611

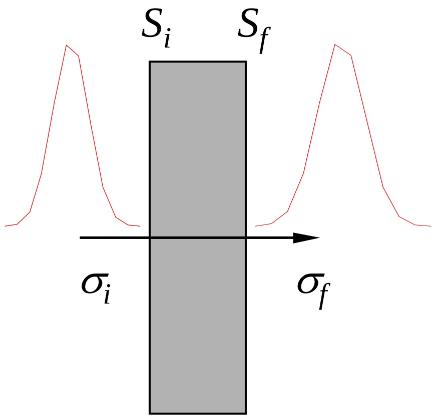
Propagation of Straggling in Thick Layers (2)



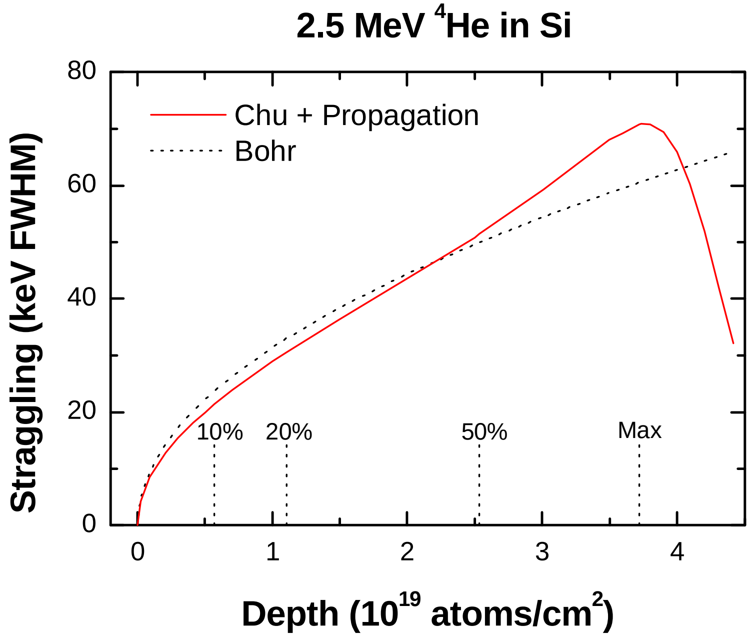
Adding stochastic and non-stochastic effects:

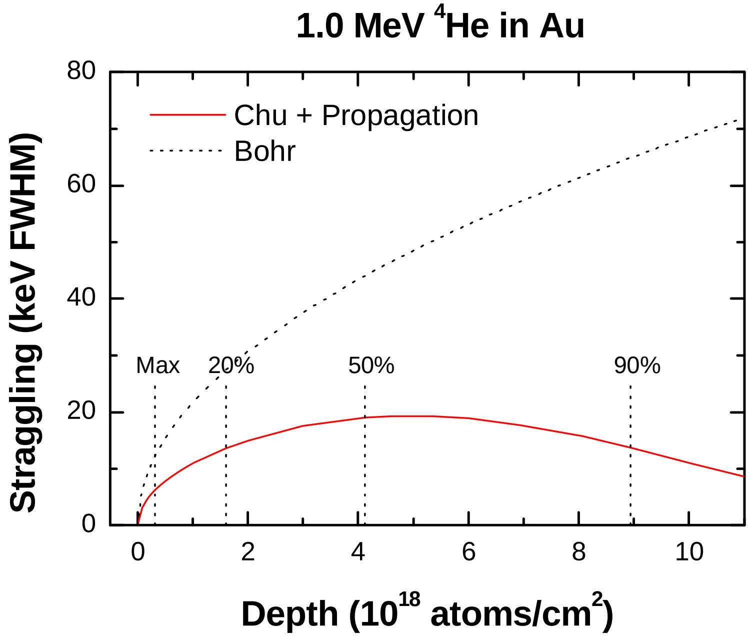
- Statistically independent

$$\sigma_f^2 = \left(\frac{S_f}{S_i}\right)^2 \sigma_i^2 + \sigma_{Chu}^2$$



Propagation of Straggling: Examples



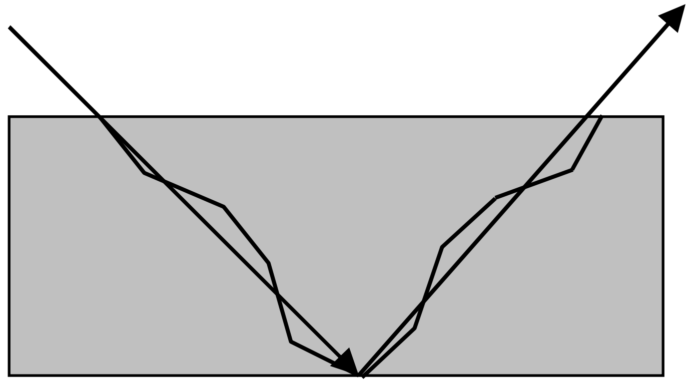


$$\sigma_f^2 = \left(\frac{S_f}{S_i}\right)^2 \sigma_i^2 + \sigma_{Chu}^2$$

Multiple and Plural Scattering

Multiple small angle deflections

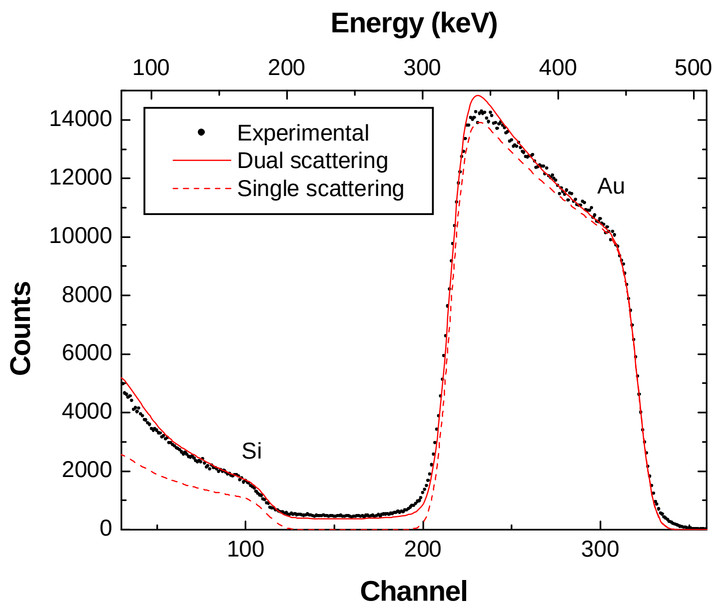
- Path length differences on ingoing and outgoing paths
  ⇒ energy spread
- Spread in scattering angle
  ⇒ energy spread of starting particles



P. Sigmund and K. Winterbon, Nucl. Instr. Meth. **119** (1974) 541

E. Szilagy et al., Nucl. Instr. Meth. **B100** (1995) 103

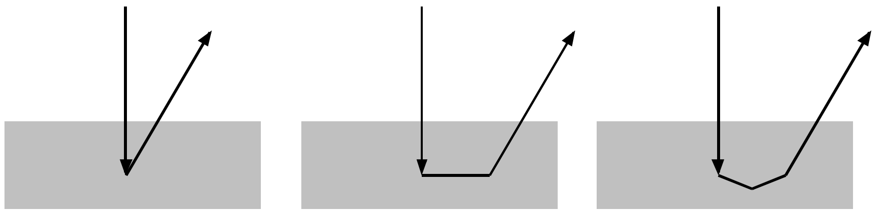
500 keV ^{4}He, 100 nm Au on Si, θ = 165°



Large angle deflections (Plural scattering)

- For example: Background below high-Z layers

W. Eckstein and M. Mayer, Nucl. Instr. Meth. **B153** (1999) 337

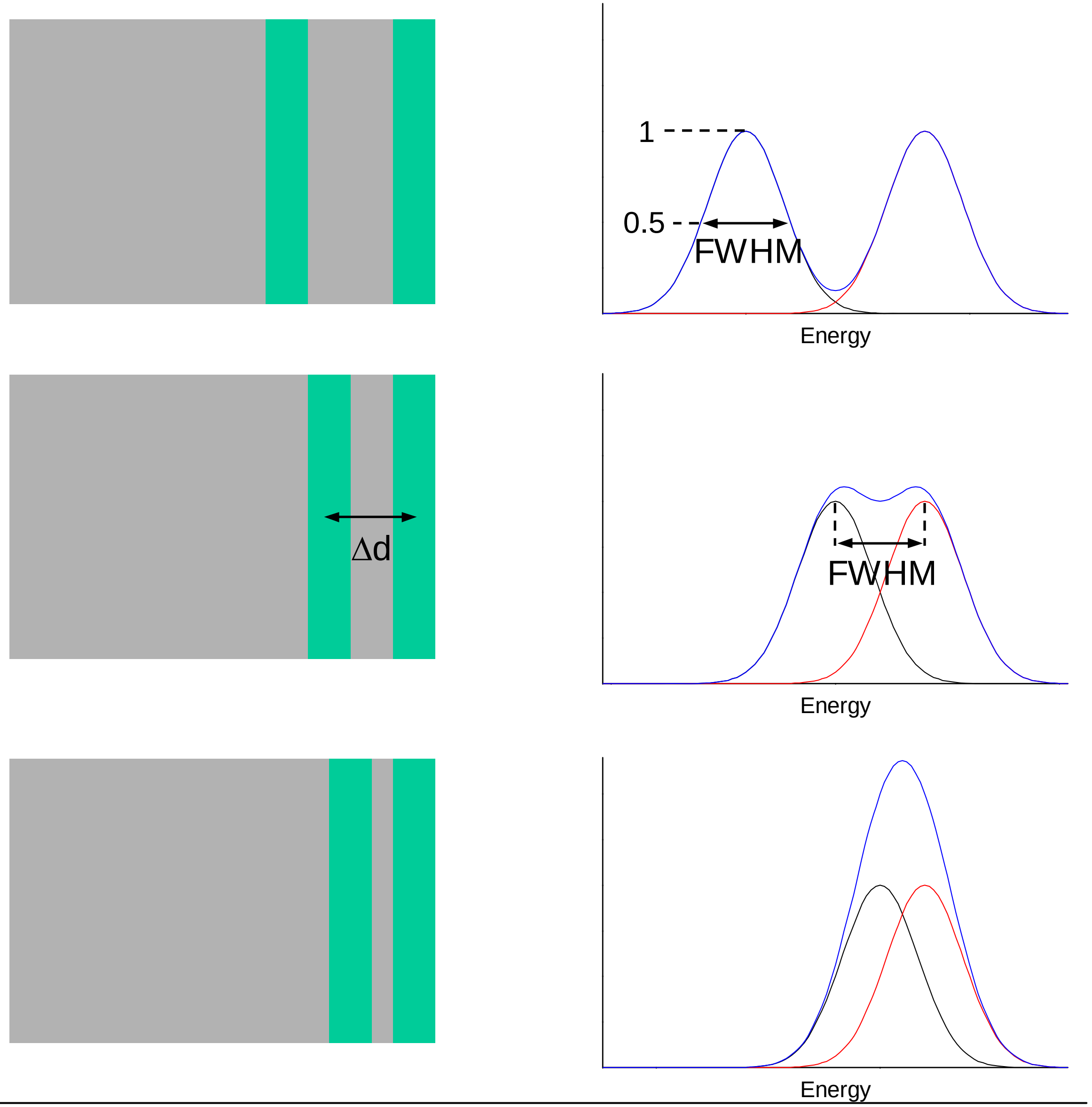


etc.

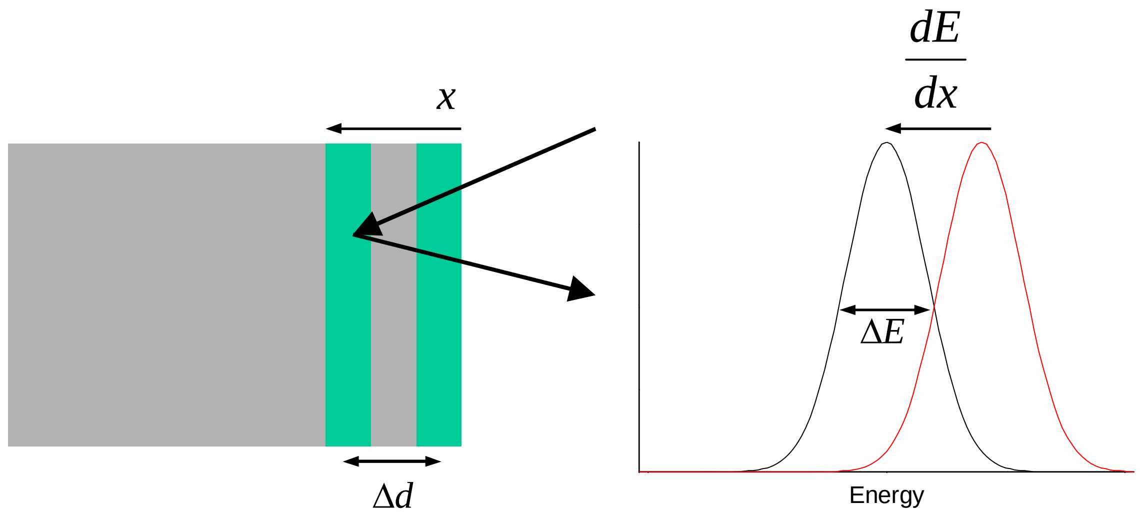
Depth resolution

- Depth resolution Δd:
  Distance between 2 layers,
  so that their energy separation is
  identical to the energy spread
- Energy spread is measured
  in FWHM
  $\Rightarrow$ Depth resolution in FWHM

- **It is not possible to obtain information about the depth profile better than the depth resolution**



Depth resolution (2)



$$S_{eff} = \left| \frac{dE}{dx}(x) \right|$$

S_{eff}: Effective stopping power
E: Mean energy in detector
x: Depth

$$\Delta d(x) = \frac{\Delta E(x)}{S_{eff}(x)}$$

ΔE: Energy straggling

Summary

- **IBA: Group of methods** for the near-surface layer analysis of solids
  Charged particles or photons can be detected
- **Scattering kinematics**: Mass resolution
- **Stopping power**: Depth resolution
- **Rutherford cross-section**: Shielding by electrons; non-Rutherford at high energies
- **Silicon detector**: Limited energy resolution
- **Energy spread** due to
  Energy-loss straggling + propagation in thick layers
  Multiple small-angle scattering; Plural scattering
- **Limited depth resolution** due to effective stopping power + energy spread