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H4.SMR/1503 - 11

WORKSHOP ON NUCLEAR DATA FOR SCIENCE AND TECHNOLOGY: MATERIALS ANALYSIS

(19 - 30 May 2003)

NRA - Nuclear Reaction Analysis

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NRA - Nuclear Reaction Analysis

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- Measurement methods for NRA
- Reaction kinematics
- Resonant and non-resonant NRA, cross section data sources
- Proton induced reactions
- Deuteron induced reactions
- ^{3}He and ^{4}He induced reactions
- NRA for hydrogen analysis

Overview

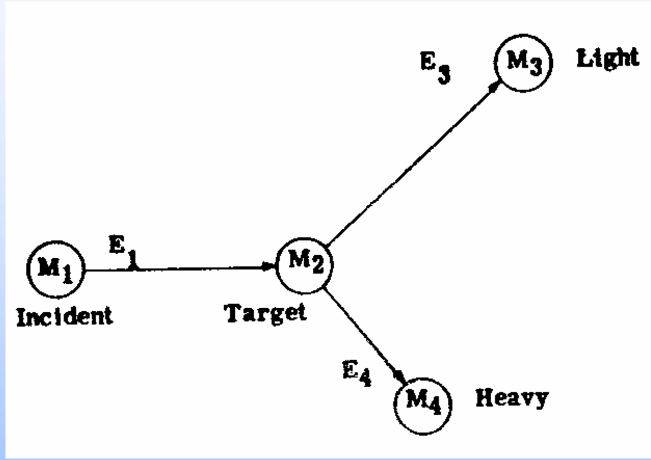
NRA - Nuclear Reaction Analysis

- Quantitative determination of selected light elements
- Quantitative depth profiling of selected light elements
- Isotopic tracing of specific light isotopes

- Particle - Particle reactions
- Particle - Gamma reactions
- Particle - Neutron reactions

Introduction: Particle - Particle reactions

$M_1 + M_2 \rightarrow M_3 + M_4 + \text{Energy}$ (endothermic reactions less useful)



- Sensitive for one (or few) light element(s)

  D(^{3}He,p)^{4}He at 800 keV

  D(^{3}He,p)^{4}He ^{9}Be(^{3}He,p)^{11}B ^{12}C(^{3}He,p)^{14}N at 2500 keV

⇒ Advantage: Background-free measurement

⇒ Disadvantage: Several measurements and/or combination with RBS necessary

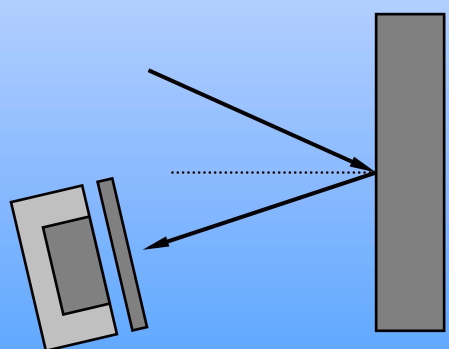
Measurement Methods

- Exothermic reactions result in high energetic reaction products

  ⇒ Above backscattered high Z components from target

  ⇒ Often allows absorber foil in front of detector

  - Background free
  - Allows detectors with large solid angle necessary due to small reaction cross sections
  - But: deteriorated depth resolution due to absorber foils



$^{2}D + {}^{3}He$ (0.8 MeV) → ^{1}H (13.2 MeV at 135°) + ^{4}He

^{11}B + p (2.5 MeV) → ^{4}He (6.1 MeV at 150°) + ^{8}Be

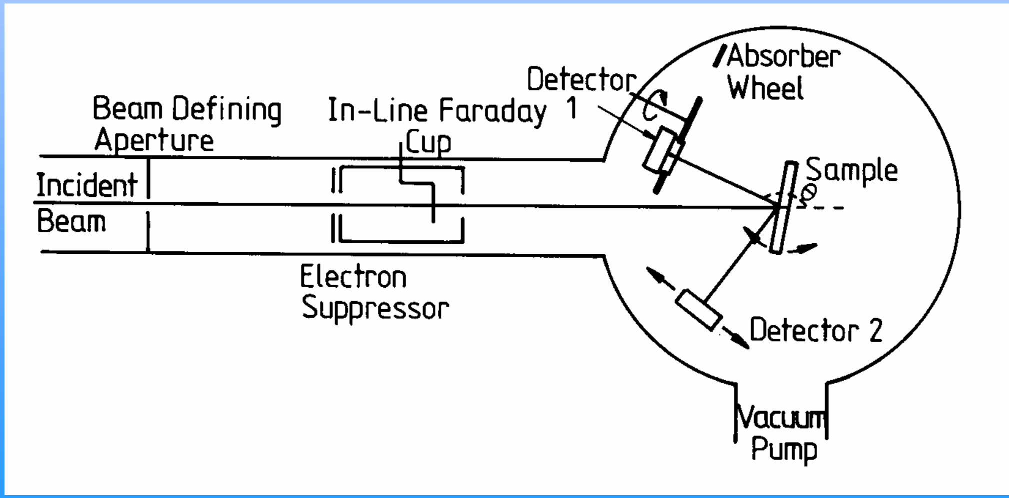
Measurement Methods (2)

Comparable equipment to RBS measurement

- Particle detection and energy measurement with solid state surface barrier detectors
- Accelerator, beam transport system identical to RBS

Additional needs for NRA:

- Absorber foil(s) in front of detector
- Optimum reaction angle may require additional detectors
- High energy protons may require thicker detector



Measurement Methods (3)

Filtering methods of unwanted particles

Unwanted particles may be

- Backscattered particles of incident beam
- Other reaction products

1. Absorber foil technique

   Advantage: Simple

   Disadvantage: Degraded depth resolution

2. Electrostatic or magnetic deflection

   Advantage: Excellent depth resolution

   Disadvantage: Large and complicated setup; rarely used

3. Time-of-flight (TOF) technique

   Discrimination of particles through TOF and energy

   Advantage: Excellent depth resolution

   Disadvantage: Complicated setup; small solid angle

   possibly large particle flux to detector $\Rightarrow$ detector lifetime

Measurement Methods (4)

Filtering methods of unwanted particles (continued)

4. Thin detector technique

Used when proton and α peaks overlap and α peak contains more information

5. Coincidence technique

Both reaction products are measured in coincidence at corresponding angles

Advantage: Low background; excellent depth resolution

Disadvantage: Only for transmission geometries (thin foil)

possibly large particle flux to detector $\Rightarrow$ detector lifetime

Warning!

Nuclear reactions (wanted or unwanted) can result in high levels of radiation

⇒ γ-radiation

⇒ neutron-radiation

⇒ activation of sample and beam system

Always contact your local radiation protection or health physics professional before undertaking measurements involving nuclear reactions!

Some notorious reactions:

$D(d,n)^3He$	No threshold! Observed for all energies Always observed with d-beams due to D-implantation in apertures of beam system
$^9Be(^4He,n)^{12}C$	High cross section at E > 2 MeV

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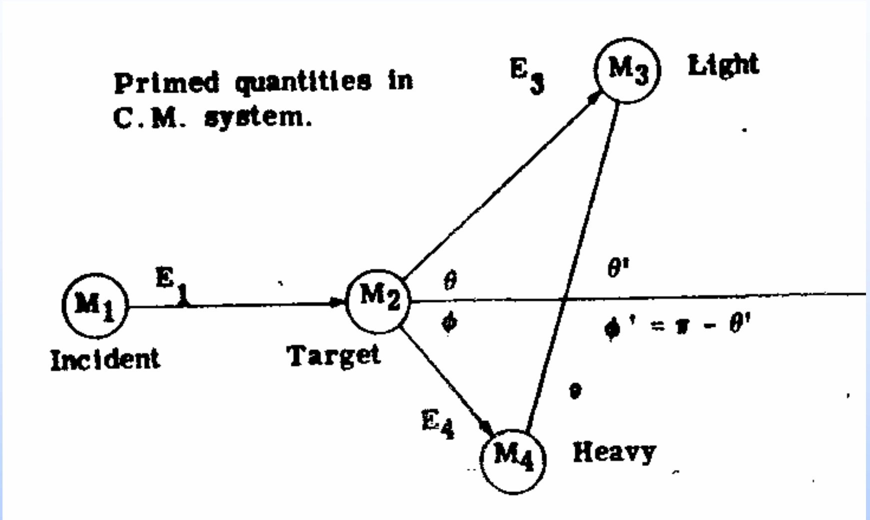
Reaction Kinematics



$$E_T = E_1 + Q = E_3 + E_4$$

$$A = \frac{M_1 M_4}{(M_1 + M_2)(M_3 + M_4)} \frac{E_1}{E_T}$$

$$B = \frac{M_1 M_3}{(M_1 + M_2)(M_3 + M_4)} \frac{E_1}{E_T}$$

$$C = \frac{M_2 M_3}{(M_1 + M_2)(M_3 + M_4)} \left(1 + \frac{M_1 Q}{M_2 E_T}\right)$$

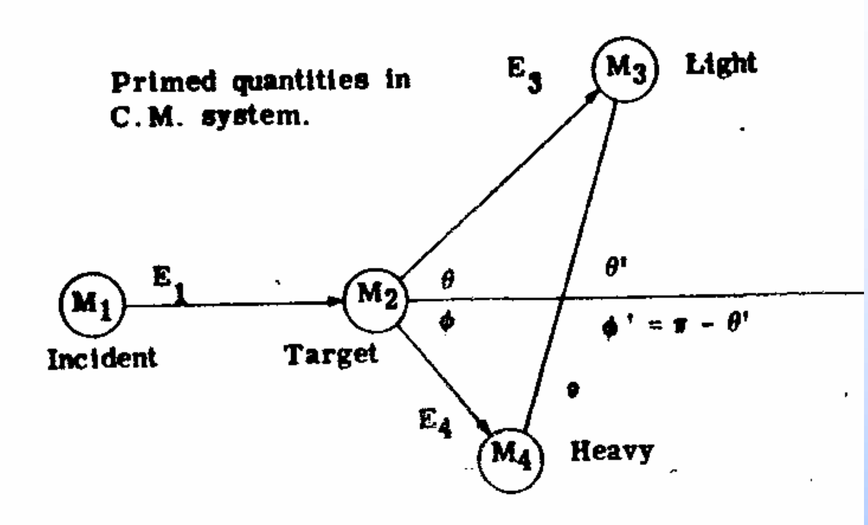
$$D = \frac{M_2 M_4}{(M_1 + M_2)(M_3 + M_4)} \left(1 + \frac{M_1 Q}{M_2 E_T}\right)$$

Note that:

$$A + B + C + D = 1$$

$$AC = BD$$

Reaction Kinematics (2)



Energy of light product: $$\frac{E_3}{E_T} = B\left[\cos\theta \pm \left(\frac{D}{B} - \sin^2\theta\right)^{1/2}\right]^2$$ Use only + unless $B > D$

Energy of heavy product: $$\frac{E_4}{E_T} = A\left[\cos\phi \pm \left(\frac{C}{A} - \sin^2\phi\right)^{1/2}\right]^2$$ Use only + unless $A > C$

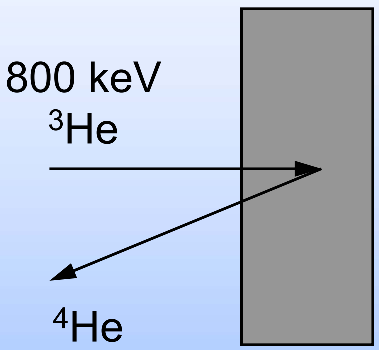
Reaction Kinematics (3)

Some reactions result in "reverse kinematics" at backward angles:

D(d,p)T D(^{3}He,p)^{4}He D(^{3}He,^{4}He)p

D(^{3}He,^{4}He)p at 165°
Energies in keV

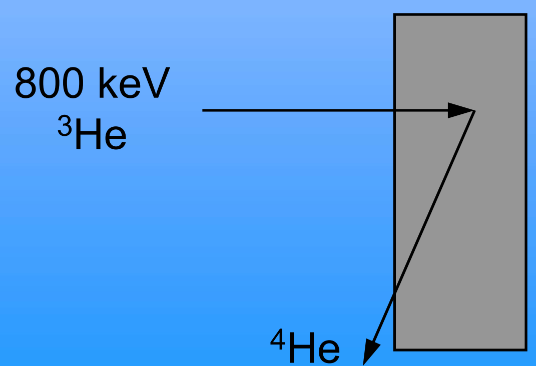
^{3}He	^{4}He
800	1780
700	1880
600	1990



Particles in deeper layers start with higher energy

⇒ not suitable for depth profiling

Solution: θ = 105°
normal incidence



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Non-Resonant and Resonant NRA

- **Non-Resonant NRA:**

  Slowly varying cross section

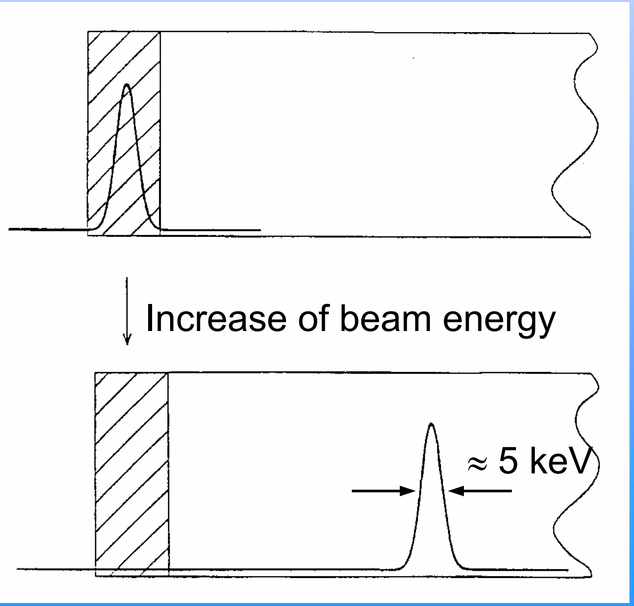
  ⇒ Analysis identical to RBS, taking NRA kinematics into account

- **Resonant NRA:**

  Resonant cross section, width ≈ several keV

  ⇒ Change of incident ion energy for depth profiling

  ⇒ Special analysis techniques required



Cross Section Data Sources

- Most nuclear reaction cross sections were **measured in the years 1950 - 1970** for nuclear physics research

  ⇒ goal was nuclear physics, not materials analysis

  ⇒ many data for non-optimal angles

  ⇒ most data published only in graphical form

- **Data compilation by R.A. Jarjis**, *Nuclear Cross Section Data for Surface Analysis*, University of Manchester, UK 1979

  2 volumes, 600 pages → unpublished

  still the most comprehensive compilation of cross section data (RBS + NRA)

- **Data compilation by G. Vizkelethy** *et al.* for J.R. Tesmer and M. Nastasi, *Handbook of Modern Ion Beam Analysis*, MRS 1995

  Digitised data from original publications in R33 file format

- These data are **available at SigmaBase**:

  ibaserver.physics.isu.edu/sigmabase/ or www.mfa.kfki.hu/sigmabase/

  Also included in the computer code **SIMNRA**

R33 File Format

```
Comment: These cross sections have been digitized from the publication
cited below. No error of either the energy and or the sigma is given.

Version: R33
Source: P.F.Alkemade et al. Nucl. Instr. Meth. B35 (1988) 135
Name: Gyorgy Vizkelethy
Serial Number: 0
Reaction: 18O(p,a)15N
Distribution: Energy
Composition:
Masses:    1.000,   18.000,    4.000,   15.000
Zeds: 1, 8, 2, 7
Qvalue:   3980.40,      0.00,      0.00,      0.00,      0.00
Theta:    155.00
Sigfactors: 1.00, 0.00
Enfactors: 1.00, 0.00, 0.00, 0.00
Units: mb
Data:
 1694.000,  0.000,   8.570, 0.000
 1695.000,  0.000,   8.590, 0.000
.
.
.
EndData:
```

R33 file format proposed by I. Vickridge, see R33Help.htm

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Proton Induced Reactions

Most useful reactions with protons:

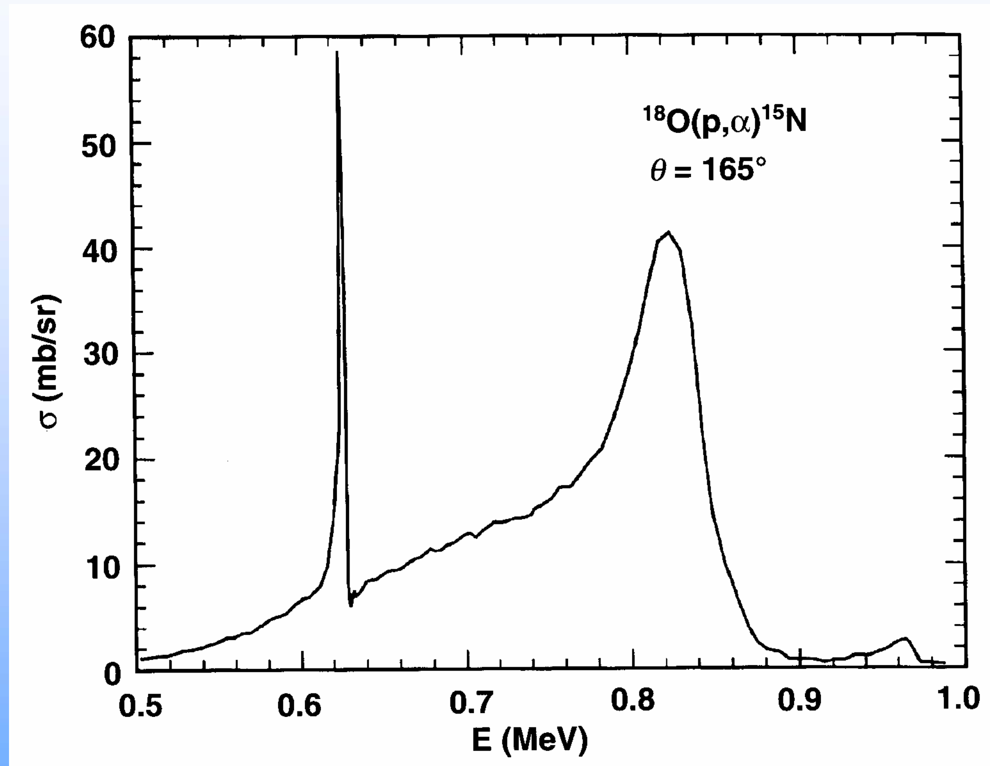
- $^{7}Li(p,\alpha)^{4}He$ Q = 17.3 MeV
- $^{11}B(p,\alpha)^{8}Be$ Q = 8.5 MeV
- $^{18}O(p,\alpha)^{15}N$ Q = 4.0 MeV

Can be used for depth profiling

Other reactions exist, but suffer from low Q-values

$\Rightarrow$ only limited use

Proton Induced Reactions (2)



Q = 3980 keV

E_α = 3310 keV for

E_p = 629 keV

- Natural abundance of ^{18}O: 0.2%
- Slow variation of cross section around 750 keV
- Resonance at 629 keV, width 2.1 keV

  ⇒ Resonant depth profiling

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Deuteron Induced Reactions

- Almost all light elements have deuteron induced reactions with positive Q-value
- Mostly (d,p) is used, but (d,α) and (d,^{3}He) are available
- Compound nuclei usually has many excited states
  $\Rightarrow$ many groups of emitted particles: ^{14}N(d,$p_{1\text{-}7}$)^{15}N, ^{19}F(d,$p_{1\text{-}16}$)^{20}F
  $\Rightarrow$ may result in interference of different peaks

Warning when using deuterium beams:

$$D + d \rightarrow \begin{cases} n + {}^3He & 50\% \\ p + T & 50\% \end{cases}$$

- May result in high radiation level due to D-implantation in beam transport system
- May result in additional p-peak due to D-implantation in target

Deuteron Induced Reactions (2)

Most useful reactions with deuterons:

- $^{12}C(d,p)^{13}C$ Q = 2.72 MeV
- $^{14}N(d,p_{0-6})^{15}N$ Q = 8.62 MeV (p_0)
- $^{16}O(d,p_{0,1})^{17}O$ Q = 1.92 MeV (p_0)

- Low stopping power for incident deuterons and exit protons

  ⇒ not suitable for depth profiling

  ⇒ use (d,α) for depth profiles

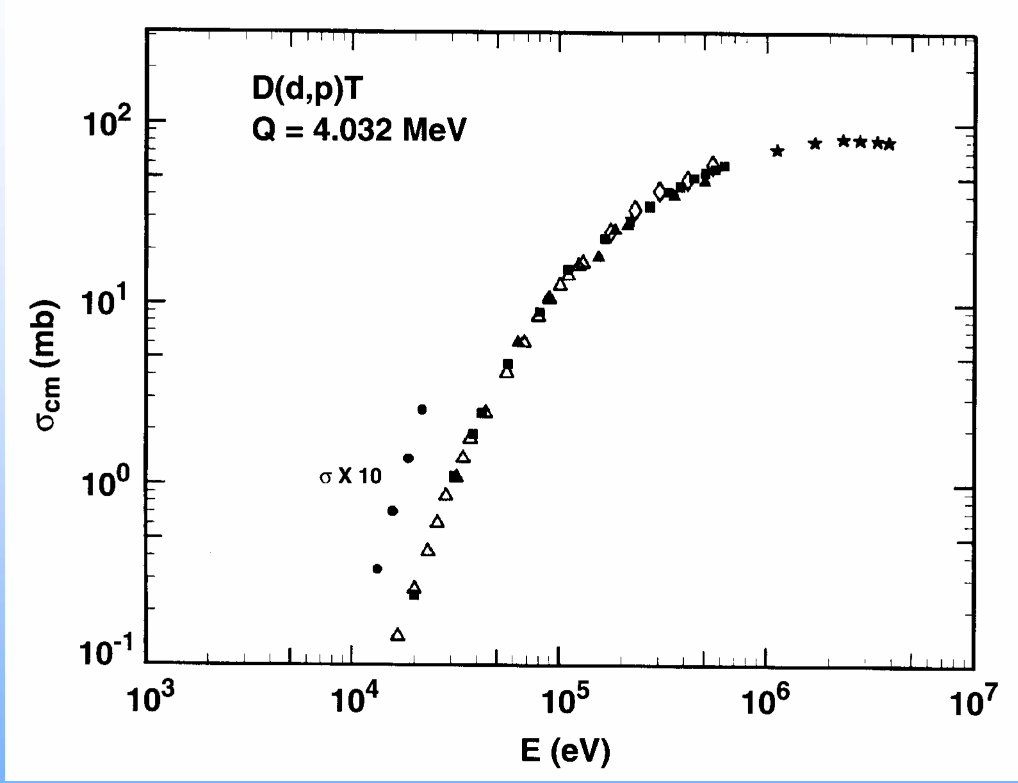
- Easy determination of total near surface content

- Many more d-induced reactions for other light elements

  See J.R. Tesmer and M. Nastasi,

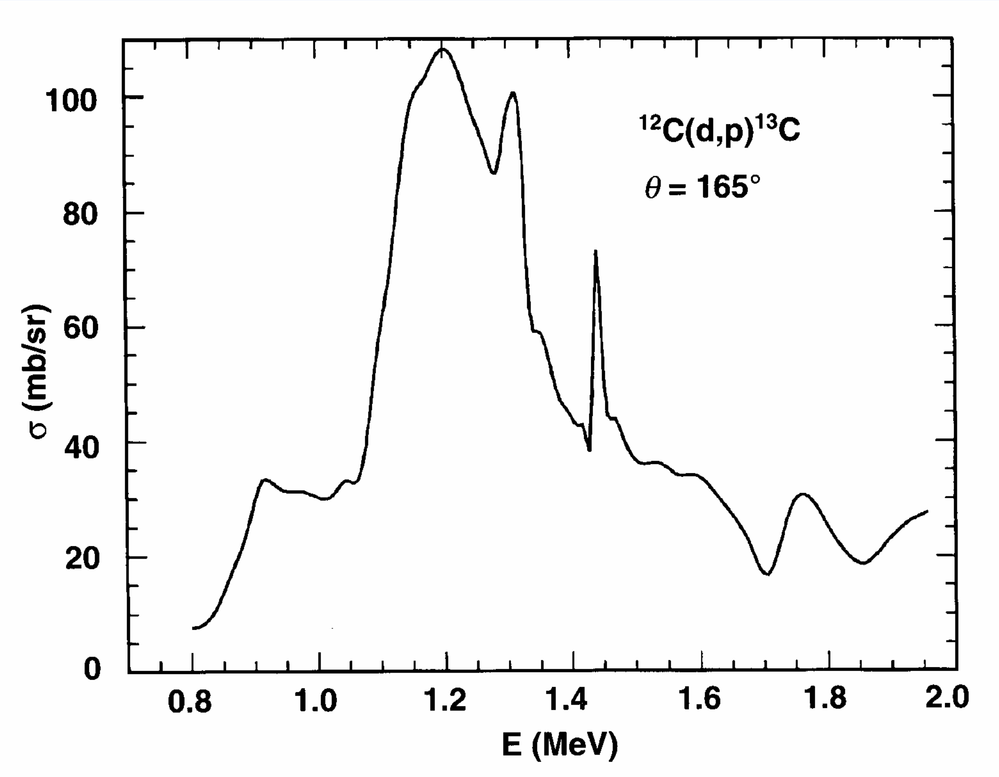
  Handbook of Modern Ion Beam Analysis, MRS 1995

Deuteron Induced Reactions (3)



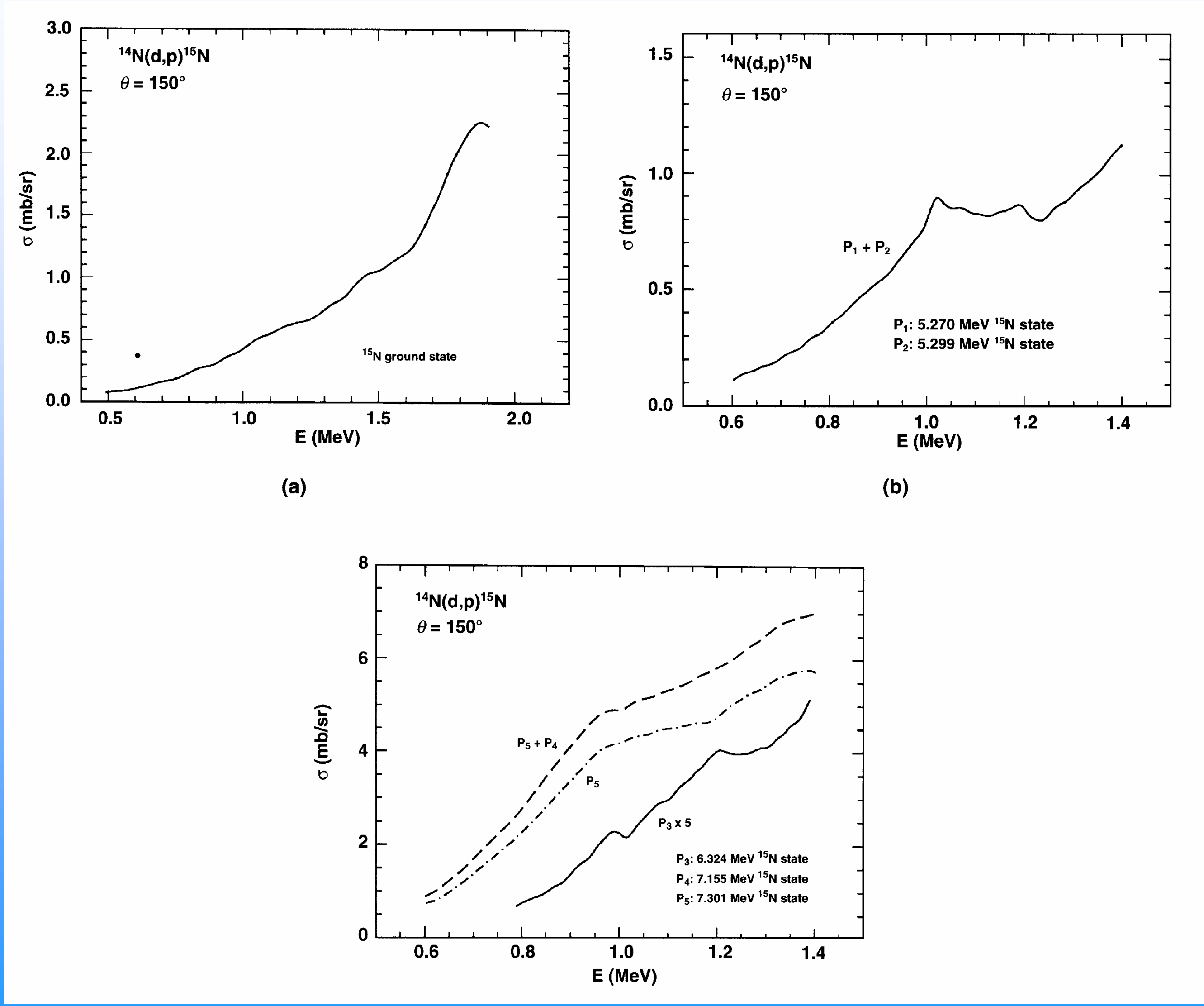
- Attention: Associated D(d,n)^{3}He reaction
- D-implantation in target

Deuteron Induced Reactions (4)



- Very high cross section
- Plateau around 1 MeV

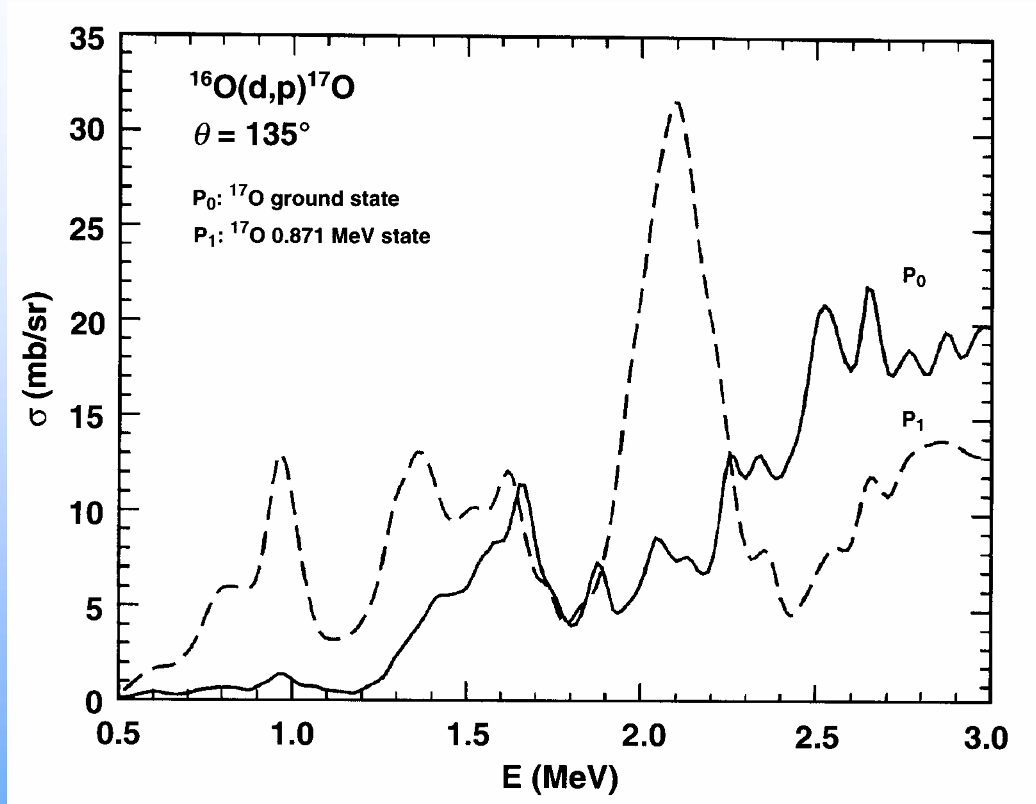
Deuteron Induced Reactions (5)



- Many excited states

  ⇒ complicated spectra

Deuteron Induced Reactions (6)

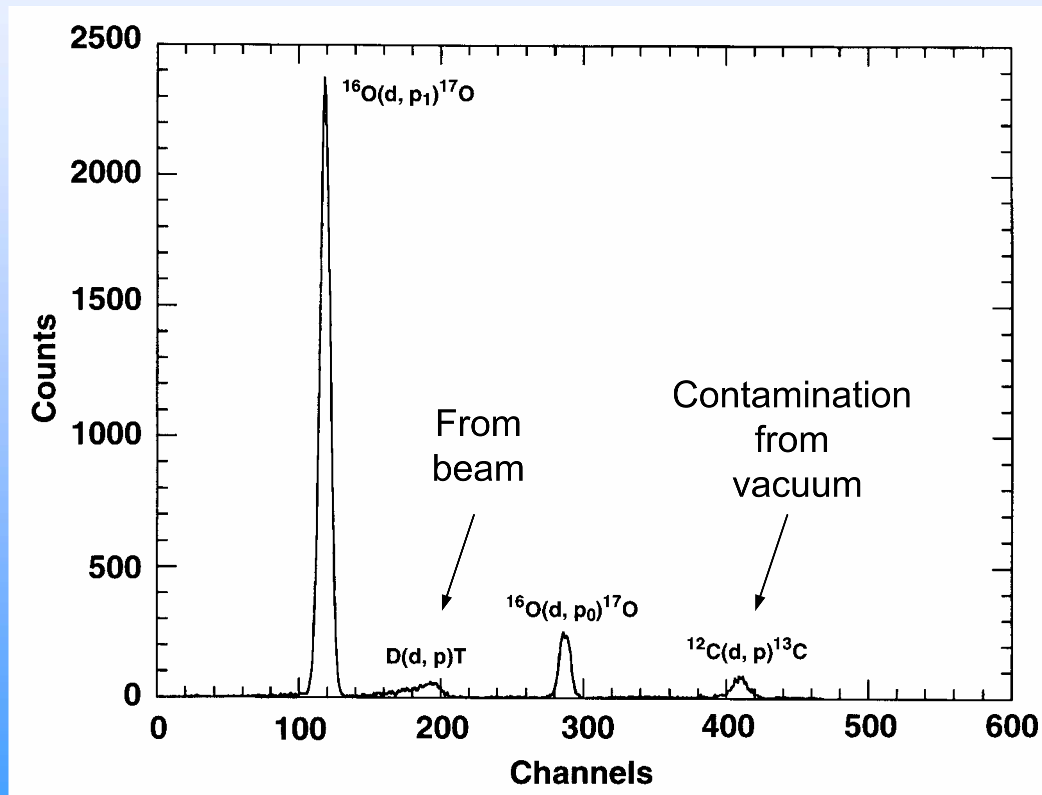


- Plateau in (d,p_0) at E < 0.9 MeV

Deuteron Induced Reactions (7)

Example: 834 keV deuterons on SiO_2/Si

θ = 135°, 12 µm Mylar absorber



G. Vizkelethy, Nucl. Instr. Meth. **B45** (1990) 1

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^{3}He Induced Reactions

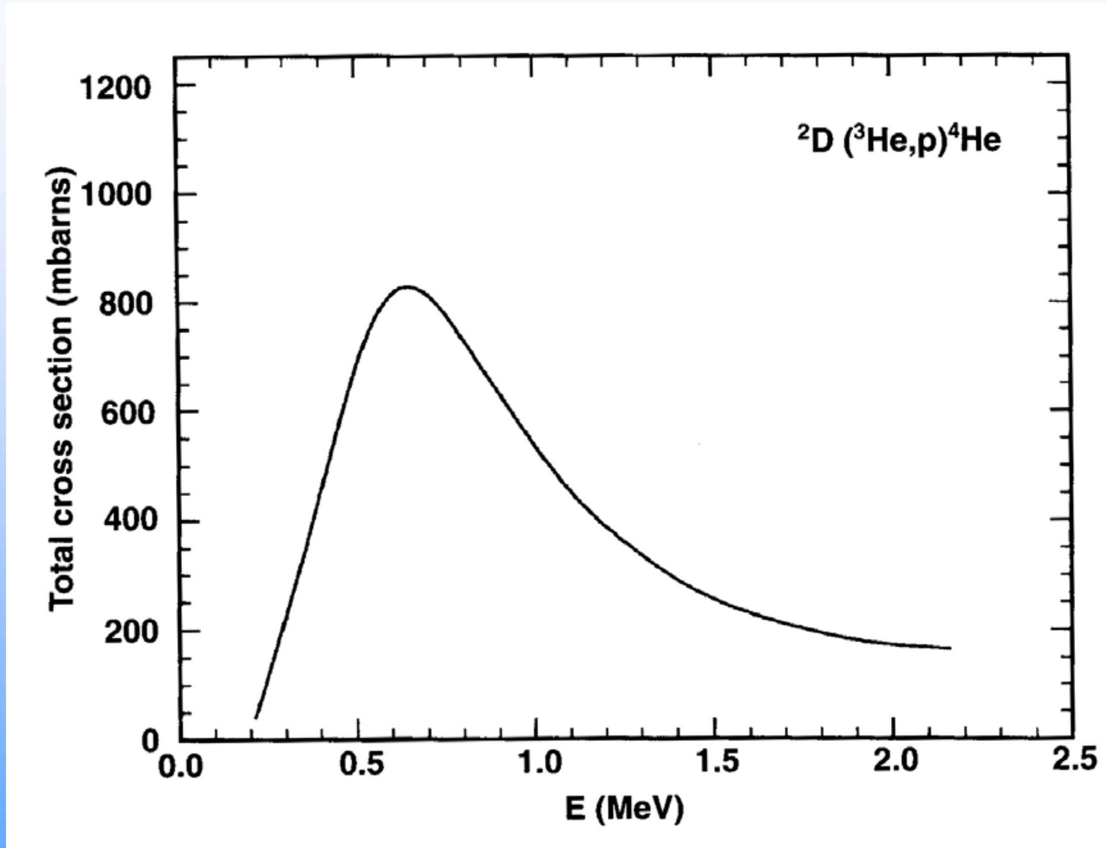
Most useful reactions with 3He:

- $D(^{3}He,p)^{4}He$ Q = 18.35 MeV
- $D(^{3}He,\alpha)^{1}H$ Q = 18.35 MeV
- $^{9}Be(^{3}He,p_{0,1})^{11}B$ Q = 10.32 MeV (p_0)
- $^{12}C(^{3}He,p_{0-11})^{14}N$ Q = 4.78 MeV (p_0)

- Many other light elements have ^{3}He induced reactions with positive Q-value
  However, only seldom used, except D + ^{3}He

- Usually many excited states
  ⇒ many groups of emitted particles
  ⇒ may result in interference of different peaks

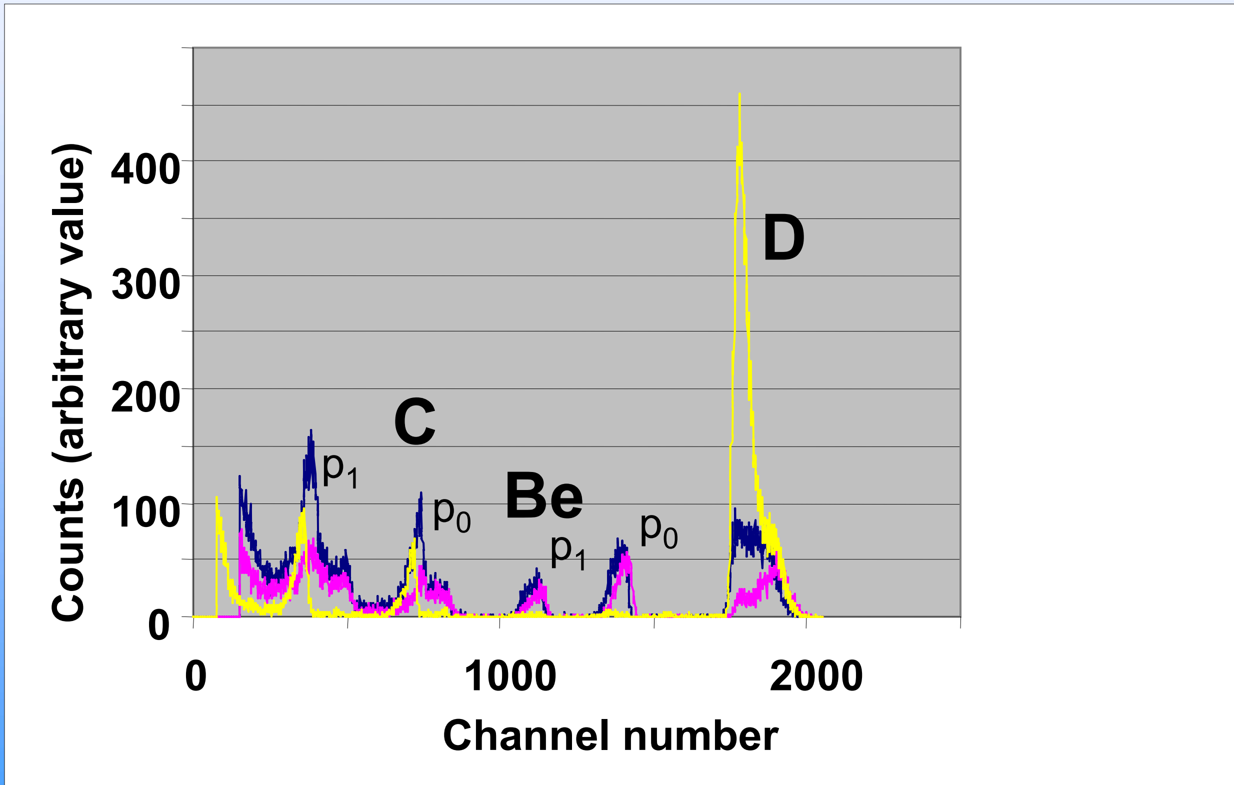
^{3}He Induced Reactions (2)



- Maximum at 640 keV

^{3}He Induced Reactions (3)

2.5 MeV ^{3}He, θ = 135°, Mylar absorber
Sample containing D, Be, C



D(^{3}He,p)^{4}He, Q = 18.35 MeV

E_p = 12.4 MeV

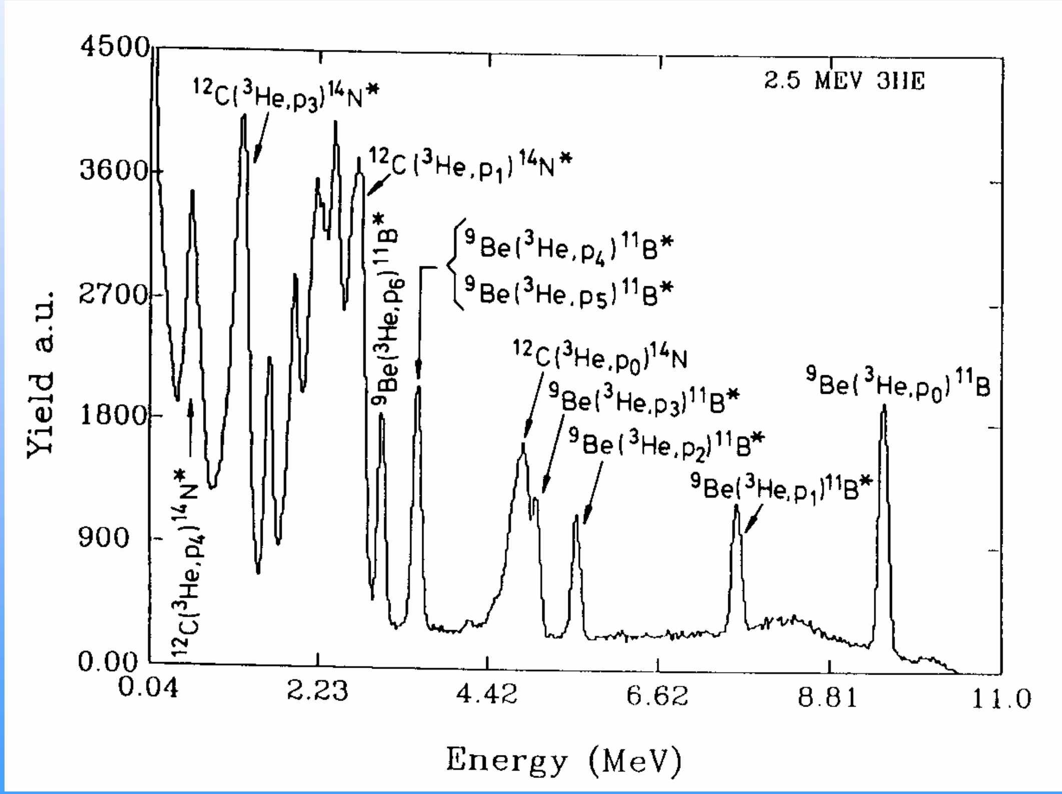
⇒ Range in Si: 1 mm

⇒ Only partly stopped in Si detector, typical thickness 100 - 500 µm

⇒ Thorough selection of detector thickness and absorber foil

M. Rubel, unpublished, 2002

2.5 MeV ^{3}He, θ = 165°, no Mylar absorber
Be on C



R. Reichle et al., Nucl. Instr. Meth. B50 (1990) 68

Very complicated spectra without absorber

^{4}He Induced Reactions

- Only few light elements with positive Q-values
- Complicated cross sections with many resonances

⇒ **Usually not useful**

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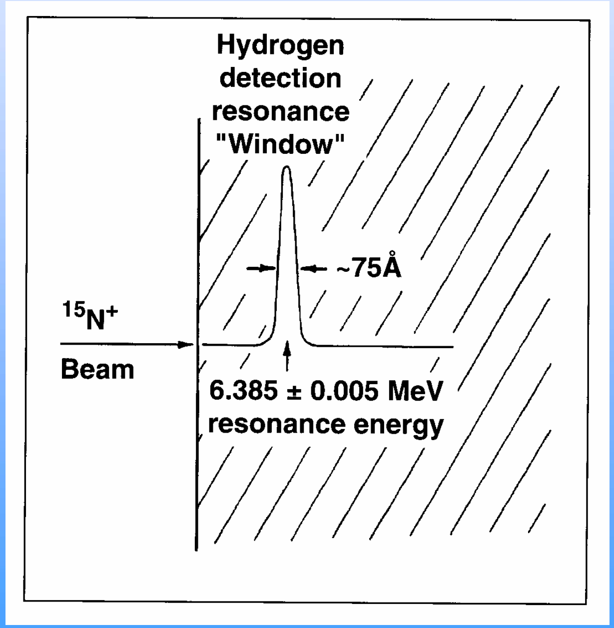
NRA for Hydrogen Analysis

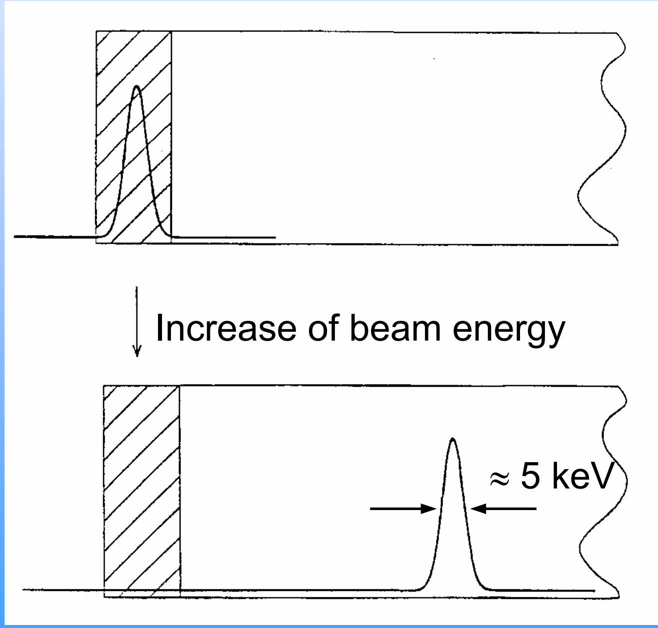
NRA for hydrogen analysis uses the resonant nuclear reaction

$^{15}N + ^{1}H \rightarrow ^{12}C + ^{4}He + \gamma$ (4.43 MeV) at 6.385 MeV ^{15}N energy

The γ is observed, not the charged particles

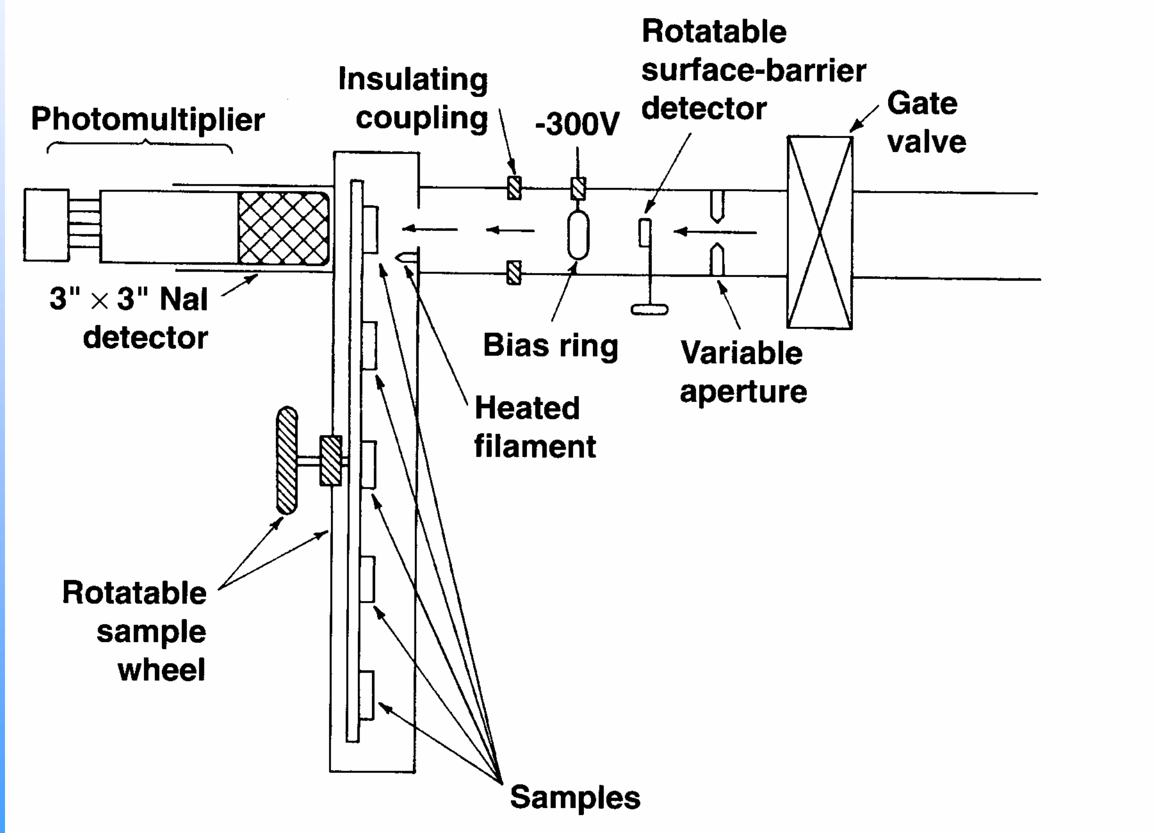
As in all resonance methods, the beam energy is varied





NRA for Hydrogen Analysis (2)

Typical experimental setup (University at Albany)

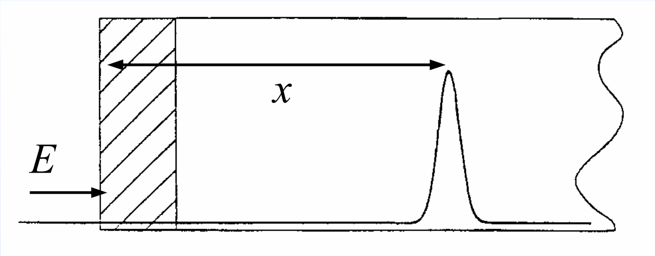


NRA for Hydrogen Analysis (3)

Depth x as function of incident energy E

$$x = \frac{E - E_{res}}{dE/dx}$$

E_{res} energy of resonance
dE/dx stopping power ≈ constant



Yield Y as function of hydrogen concentration $n(x)$

$$Y = N_0 \int n(x)\,\sigma(x)\,dx$$

Use $dx = \frac{dE}{dE/dx}$

$$\Rightarrow \quad Y = N_0 \int \frac{n(E)\,\sigma(E)}{dE/dx}\,dE$$

Use $n(x)$ = const
and Breit-Wigner formula $\sigma(E) = \frac{\sigma_0 \frac{\Gamma^2}{4}}{(E - E_{res})^2 + \frac{\Gamma^2}{4}}$

$$\Rightarrow \quad \boxed{Y = \frac{N_0 \pi}{2\sigma_0 \Gamma} \frac{n}{dE/dx}}$$

NRA for Hydrogen Analysis (4)

Advantages:

- Very good sensitivity and depth resolution
- Can be used at normal incidence ⇒ less sensitive to surface roughness

Disadvantages:

- Low probing depth
- Not applicable for delicate samples (hydrocarbons): Ion beam induced loss of H
  - Larger sample damage by ^{15}N than by light ions
  - Change of beam energy requires large fluence

Alternative methods:

- $^{7}Li + {}^{1}H$ at 3.07 MeV: Larger analysed depth, smaller cross section
- $^{19}F + {}^{1}H$ at 16.44 MeV: Easier beam handling, smaller cross section
- ERD with ^{4}He or heavy ions