

We left off after developing the capability to construct and interpret RBS spectra for doing **non-destructive** and **quantitative** analysis of surface and near-surface regions of “thin” films. The term ‘thin’ refers to the thickness with respect to the probing depth of the particular charged particle that is being used. One needs tables of ranges and stopping powers to see what depths we are talking about for MeV light ions (H,He).

Let’s now talk about how to increase the sensitivity for light element detection (C,N,O for example) on **SINGLE CRYSTAL** heavier substrates. In this case, we need to exploit a technique that is called **CHANNELING**, whereby the incident *light* ions are steered gently between the crystal planes or along a crystal axis, and lose less energy than for incidence in a so-called random direction. Now we have the capability to do lattice location studies, to look for defects in the crystal, *etc.* In these cases, the RBS signal lies ON TOP OF THAT OF THE SUBSTRATE (remember the superposition principle!) so that it will be hard to see due to the substrate scattering yield, together with the Z_2^2 dependence on target species.

Below is shown a stereographic projection for a fcc (100) crystal lattice.

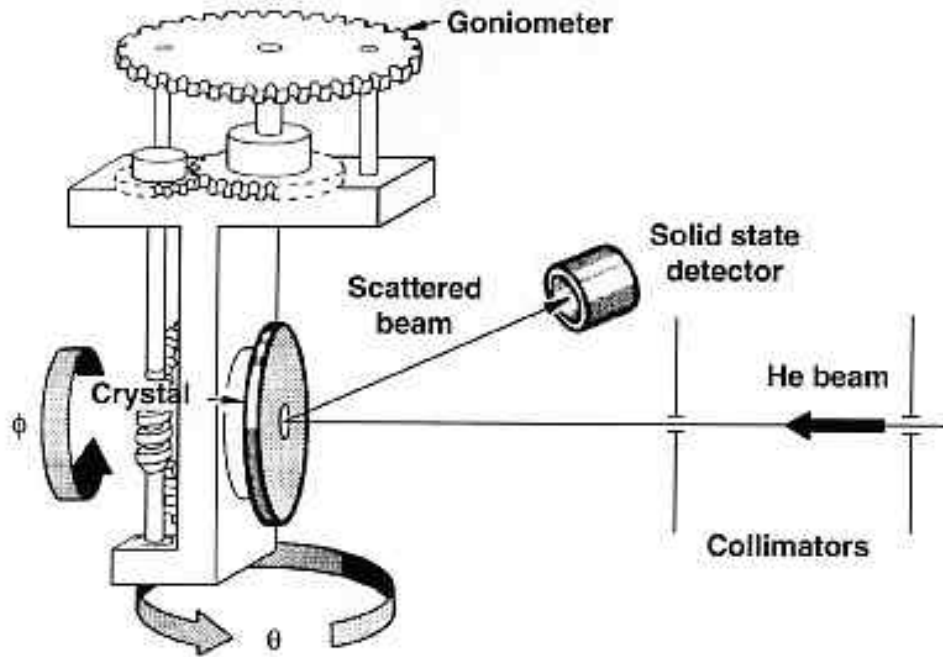
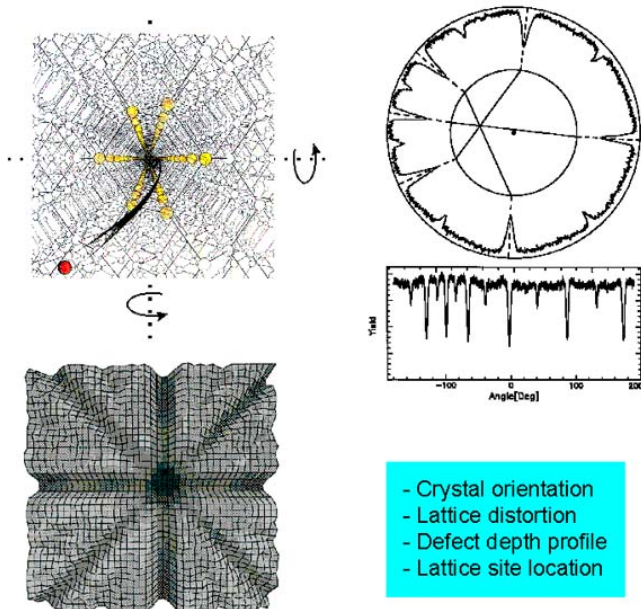


FIG 10.10. Schematic view of the setup for channeling experiments. The ion beam impinges on a crystal mounted on a two axis goniometer, with tilt angle θ (rotation about the vertical axis), and azimuthal angle ϕ (rotation about the crystal normal). The nuclear particle detector measures the energy of the backscattered ions (from Feldman et al., 1982).

Below is shown the yield as one rotates azimuthally about a $\langle 111 \rangle$ channel for a cubic structure.

Ion Beam Channeling

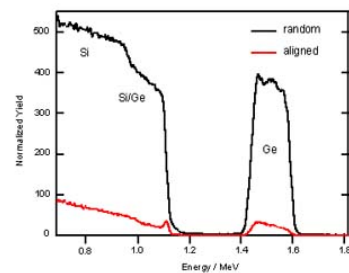
Principle:



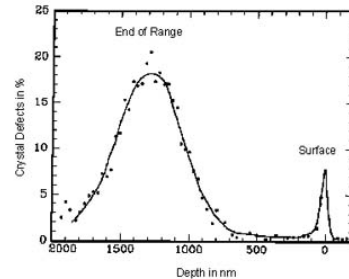
- Crystal orientation
- Lattice distortion
- Defect depth profile
- Lattice site location

Applications:

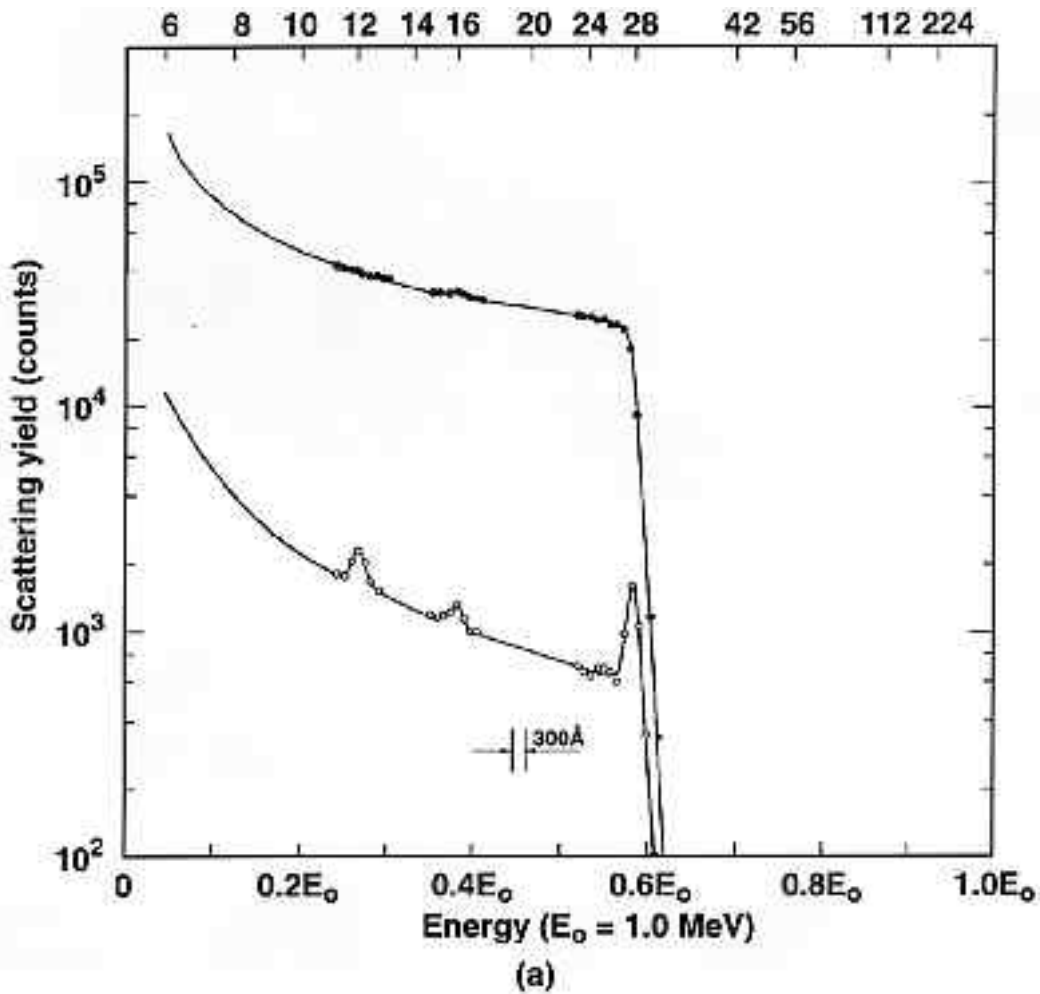
$\text{Ge}_x\text{Si}_{1-x}$ Epitaxy on Si



Crystal damage in InP after 2 MeV ^{16}O Implantation



Below is shown a “random” spectrum and an aligned spectrum for a Si sample which has some carbon and oxygen on the surface. One can immediately see the impracticality of trying to determine areas of the C and O signals, which sit on top of the Si signal (remember superposition!), for the random spectrum. However, for the aligned spectrum, one can extract these areas. The surface peak corresponds to scattering from those Si atoms that the ion beam “sees” even under channeling conditions. The mass (if located on the surface) is shown at the top.



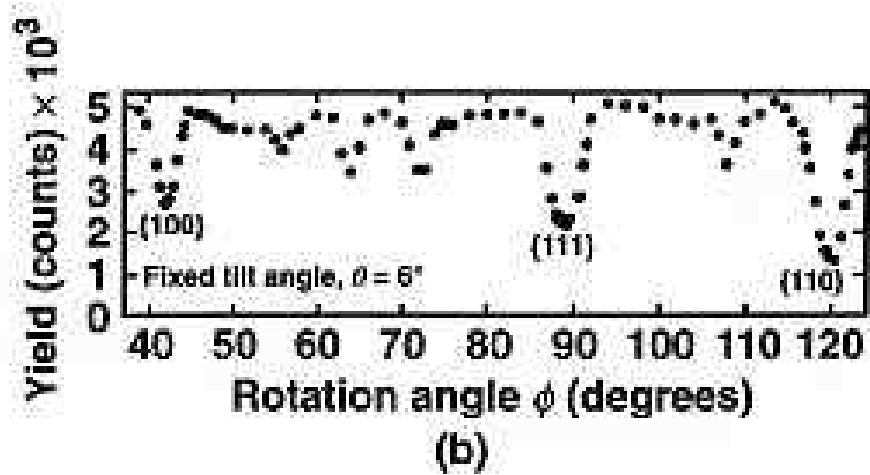
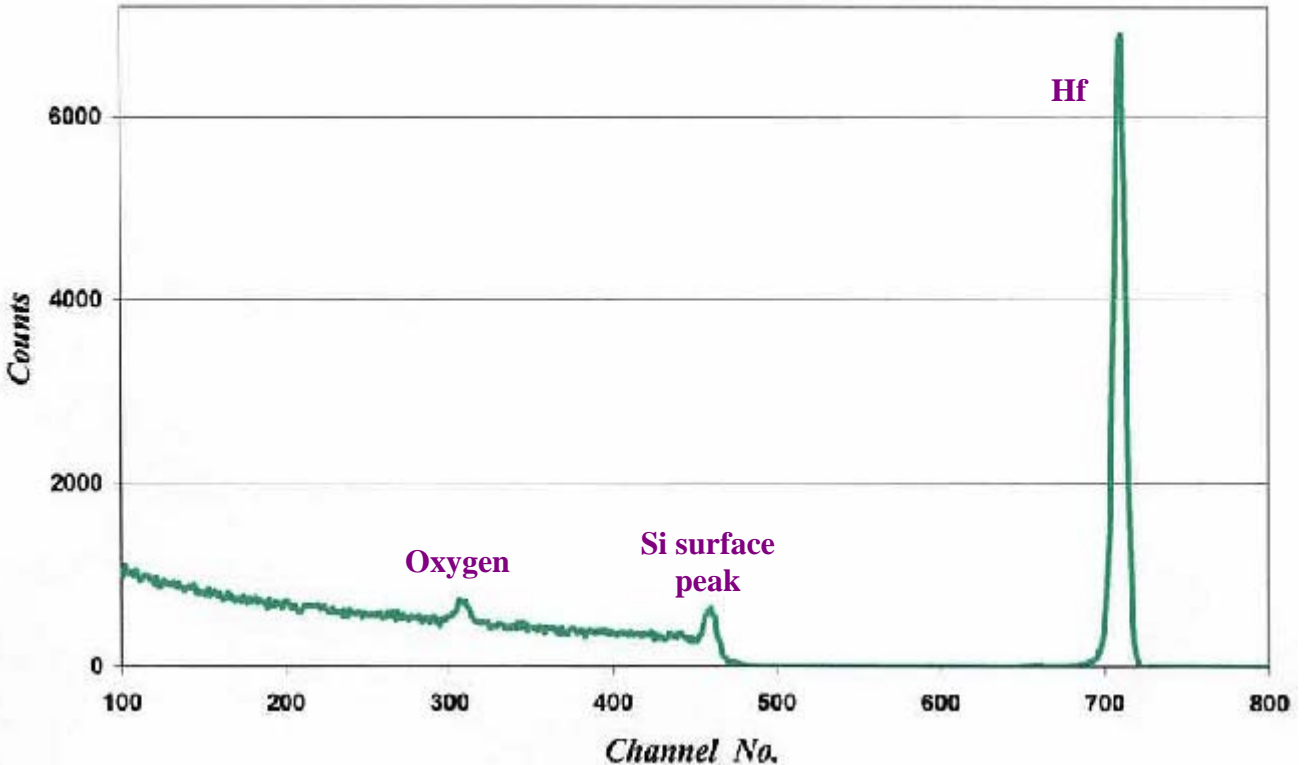


FIG. 10.19. How to align a Si (110) crystal for channeling. (a) RBS spectra for a randomly-oriented crystal (top) and an aligned crystal (bottom). A narrow energy window is shown below the surface peak. (b) RBS yield as a function of the azimuth angle ϕ for a tilt angle of 6° .

Now let's look at the RBS channeled spectrum for a thin (~ 4 nm) Hf ($Z=72$, $A=178.49$) silicate sample on a Si(100) substrate using incident 1.5 MeV $^4\text{He}^+$ ions.

Hafnium Silicate on Si(100)



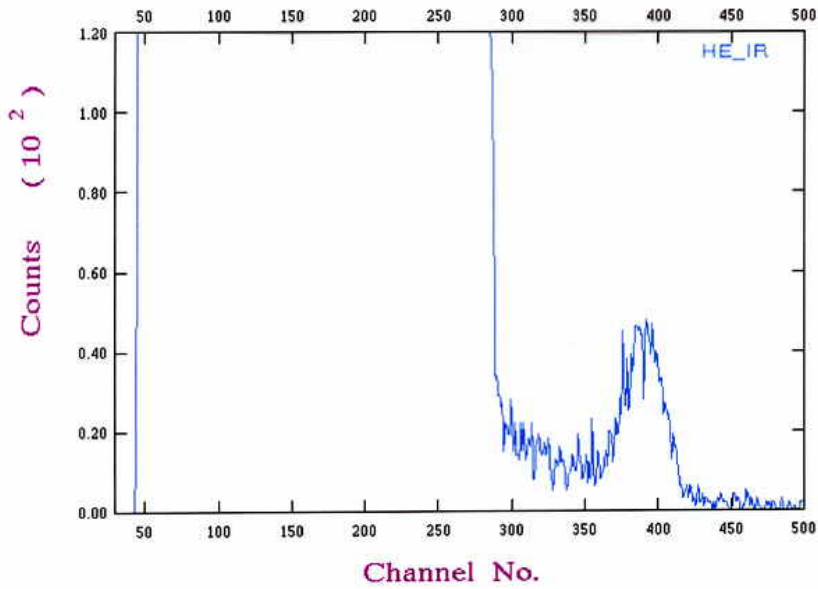
We can easily measure the Hf/O ratio since we know the RBS cross sections very accurately. The absolute [Hf] can be easily measured *without channeling*.

HEAVY ION RBS

Instead of using ^4He ions, use $^{12}\text{C}^{q+}$ (6 MeV) or $^{18}\text{O}^{q+}$ (8 MeV) ions to measure heavy impurities, **assuming one has a particle accelerator capable of producing such beams**; specifically, let's look at ^{193}Ir ($Z=77$) implanted into Si at 1.0 MeV to a dose of 2×10^{14} at./cm²:

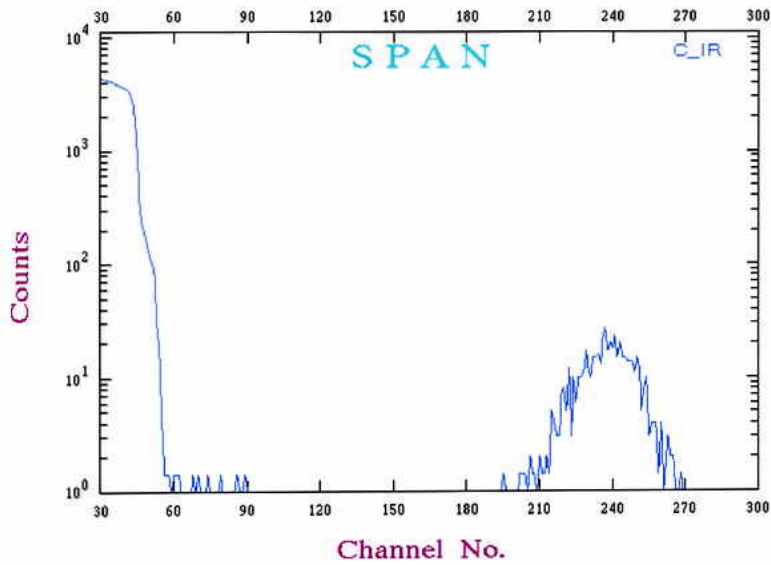
Let's examine a conventional RBS spectrum using 1.8 MeV ^4He ions and compare that to HIRBS using incident carbon ions (the latter ion beam can be generated on the Tandatron accelerator, not the Van de Graaff accelerator):

28 Sep 1998



1.8 MeV ^4He RBS

28 Sep 1998



6 MeV ^{12}C RBS

170°

$\sim 2 \times 10^{14}$ Ir/cm^2 into Si @ 1 MeV

Notice that this RBS spectrum has *zero background*. This is because the mass resolution gets much better. Remember that at 180°, the k-value is:

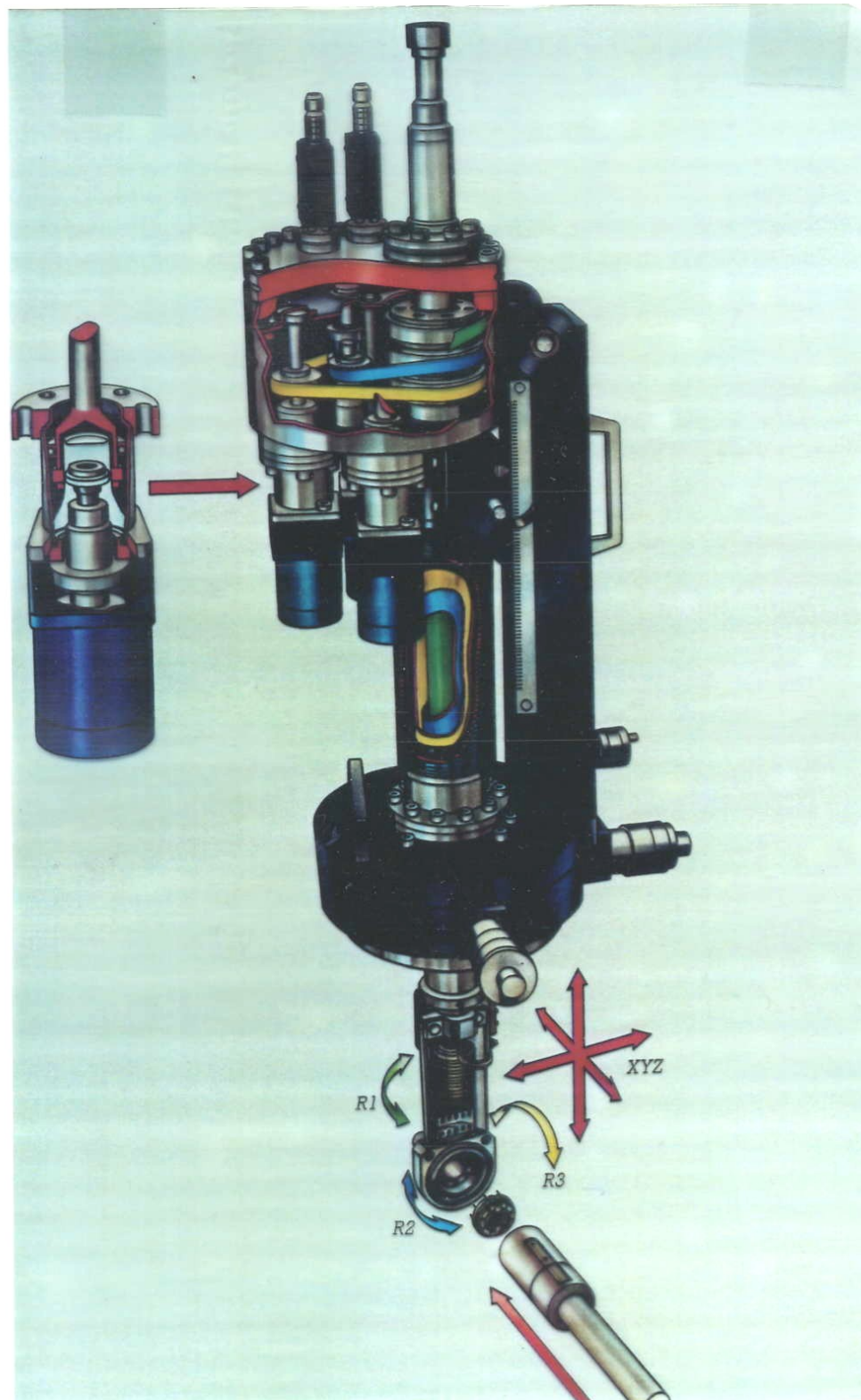
$$k = \left(\frac{M_2 - M_1}{M_2 + M_1} \right)^2$$

so that the yield from the substrate Si is pushed to much lower energies. The negative feature is that the energy resolution of the Si charged particle detector degrades for heavier ions.

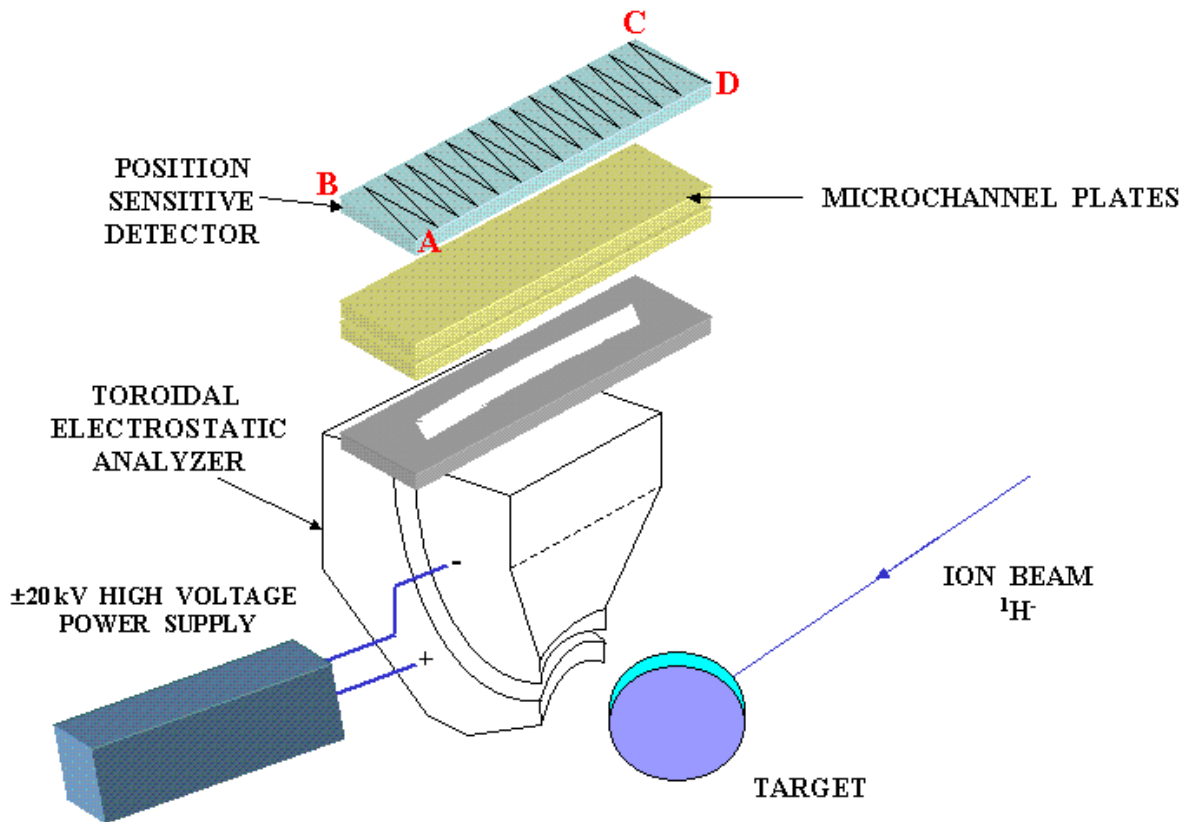
The scattering cross sections are not so different here: remember that $d\sigma/d\Omega$ varies as $(Z_1/E)^2$: thus, $(2/1.8)^2 \approx (6/6)^2$ (^4He vs. ^{12}C incident ions).

MEDIUM ENERGY ION SCATTERING (MEIS)

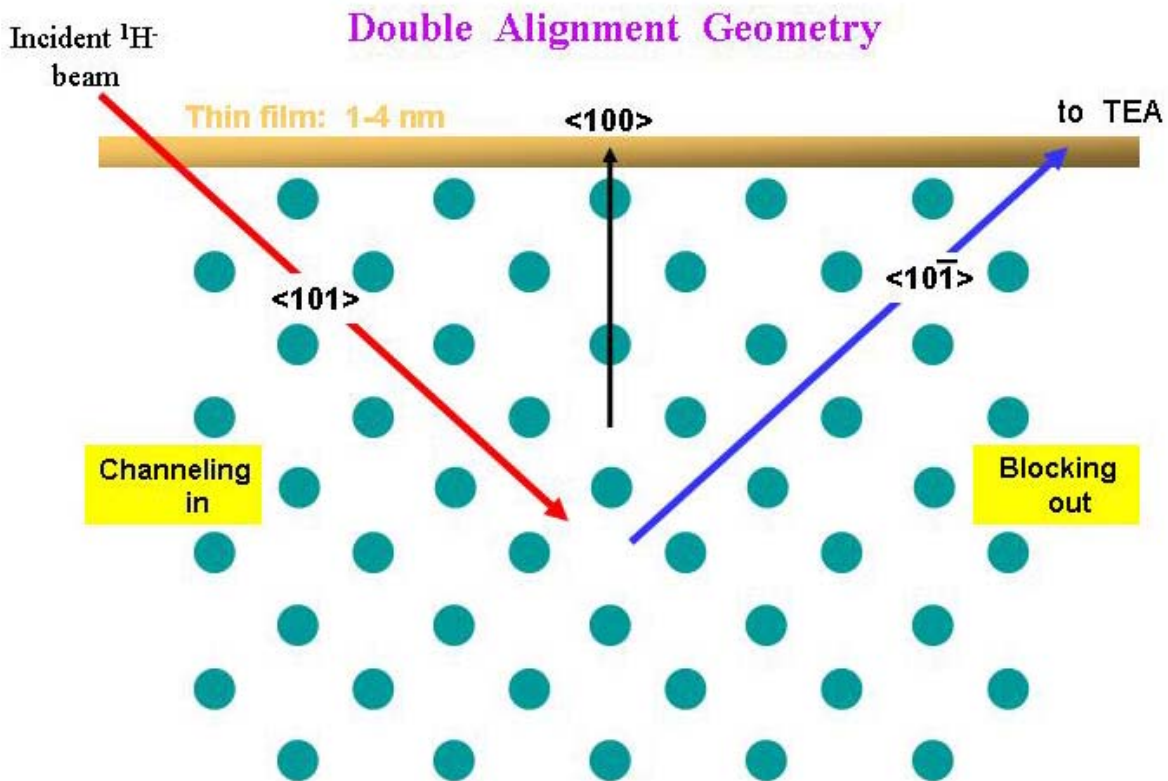
For very high depth resolution, we cannot use the Si charged particle detectors due to their limited (and fixed) energy resolution. Therefore, use an (toroidal) electrostatic analyzer, or TEA, to energy-analyze the scattered particles. Here, we normally use elastic scattering of 100 keV protons to keep the voltages on the TEA reasonable, although this procedure limits the mass resolution (remember the kinematic factor, k !).



We need a 6-axes goniometer in order to perform **channeling in / blocking out** measurements, also done in UHV. The 3 orthogonal rotations are computer controlled, the 3 orthogonal translations are manually actuated.

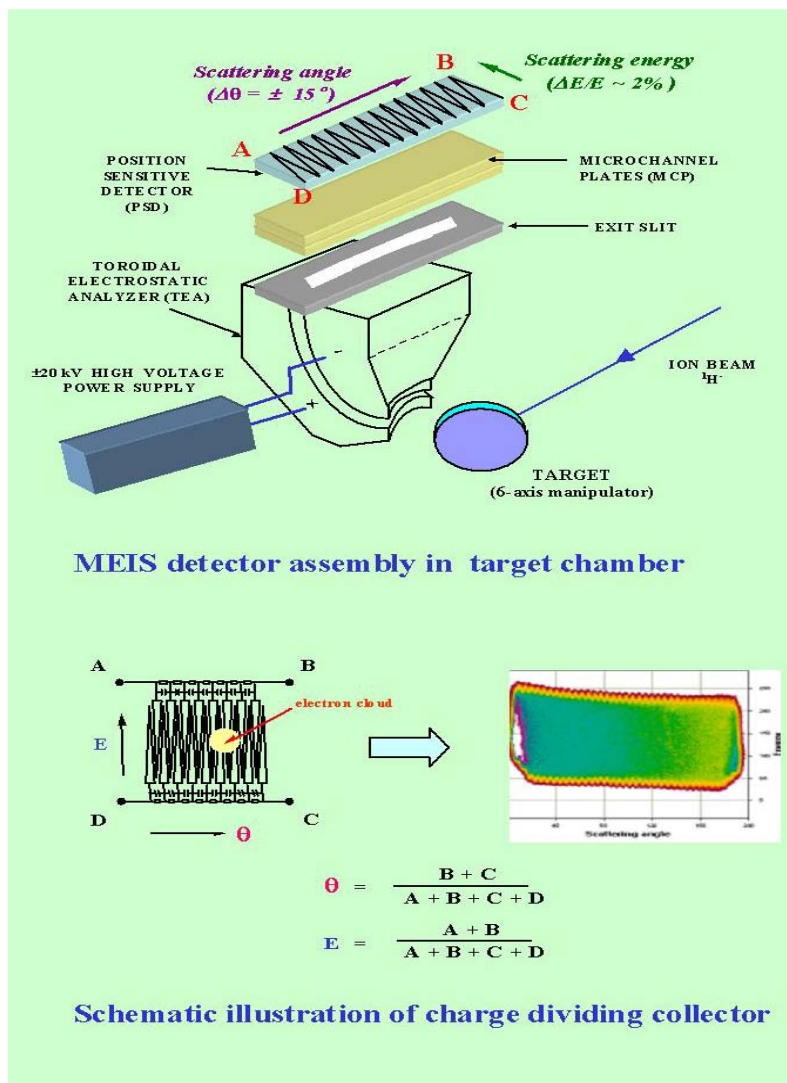


Here, the fractional resolution of the TEA is $\Delta E/E \cong 1.5 \times 10^{-3}$ or ~ 150 eV for $E_0=100$ keV. The PSD (position sensitive detector) yields a 2D image (energy and angle) for each setting of the TEA voltage, V_{TEA} , and we step V_{TEA} along to cover all the relevant masses. Also, in order to measure elements lighter than the substrate (which is most often Si, so we want to measure C, N, O), we not only use a channeling *IN* configuration, we employ a blocking direction on the *OUTWARD* path so that the substrate yield is further reduced.

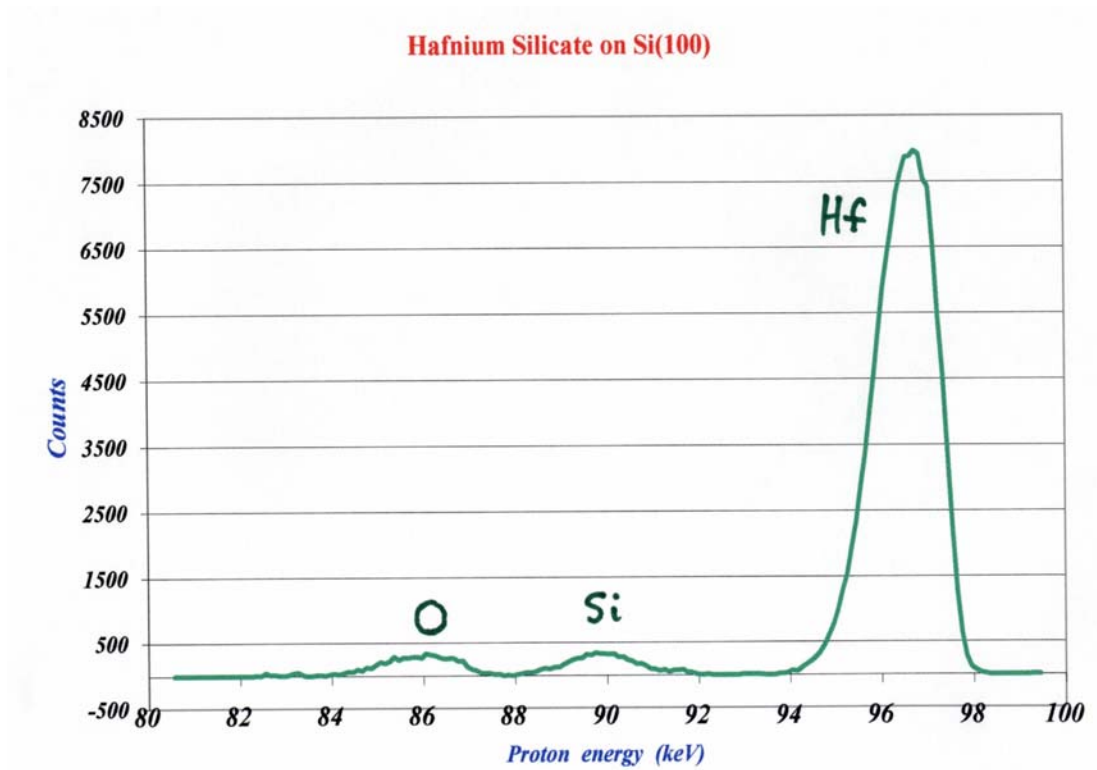


fcc (100) surface with a thin film on top of Si(100) substrate

The use of negative incident ions is irrelevant: the electron is quickly detached as soon as the ion strikes the target since its binding energy is so small. The depth resolution of this instrument is <1 nm using 100 keV protons. The target manipulator has 6 degrees of freedom: 3 rotations that are computer controlled, and 3 translations that are manually controlled.



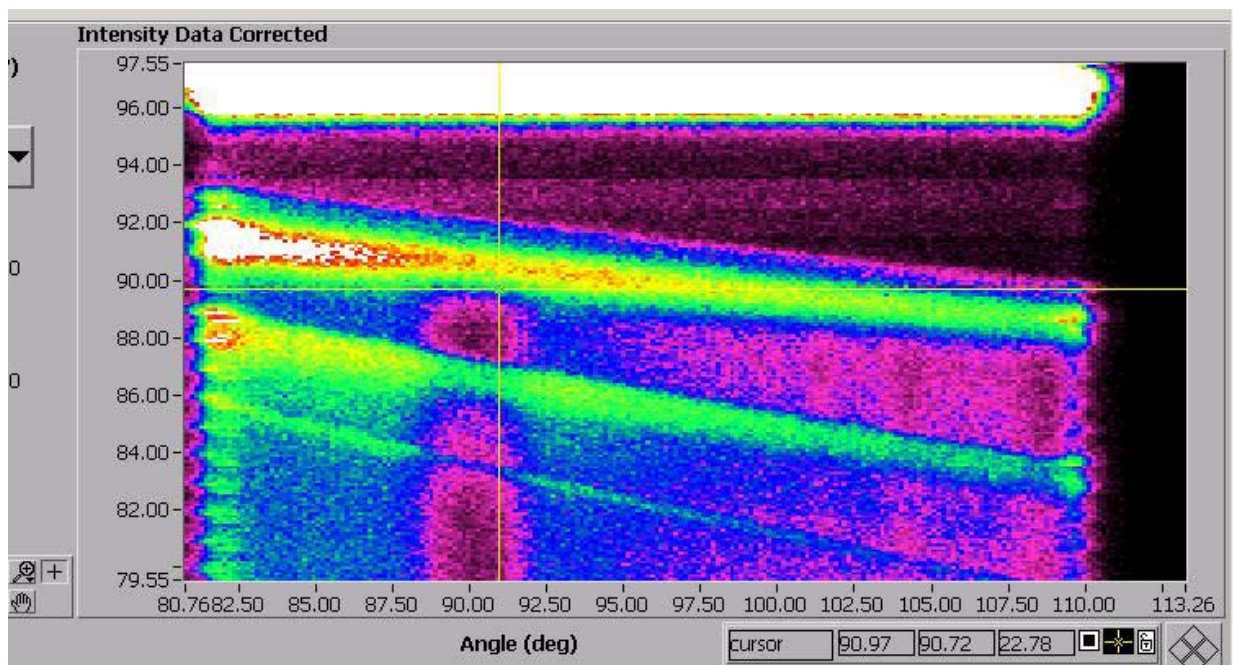
What does the corresponding RBS spectrum look like? Data were acquired by stepping the TEA from 81-99 keV in 1 keV steps. We first remove the (almost) flat Si background arising from the double alignment technique.



Below, TEA intensity-corrected 2D spectrum for the same thin film

(ordinate $\Rightarrow E_p$, abscissa \Rightarrow lab. angle)

Note the blocking direction !



ELASTIC RECOIL DETECTION (ERD)

There is a variant of RBS that was alluded to in the first lecture, when we mentioned the energy given to the recoiling target atom: remember the formulae for E_1/E_0 and E_2/E_0 , representing the fraction of kinetic energy that is retained by the projectile (“1”) and transferred to the target (“2”), respectively:

$$\frac{E_1}{E_0} = \frac{M_1^2}{(M_1 + M_2)^2} \left\{ \cos \psi \pm \left[(M_2 / M_1)^2 - \sin^2 \psi \right]^{1/2} \right\}^2$$

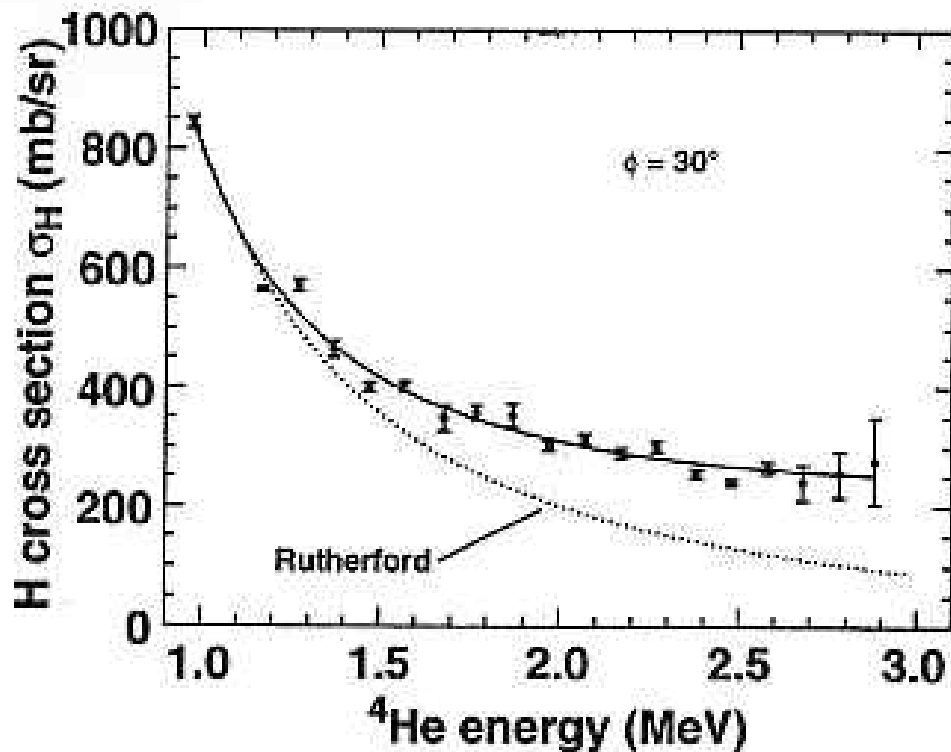
$$\frac{E_2}{E_0} = 1 - \frac{E_1}{E_0} = \frac{4M_1M_2}{(M_1 + M_2)^2} \cos^2 \zeta$$

In the second equation, $\zeta \leq 90^\circ$, obviously, since the recoil can't come back in the direction from which the projectile is approaching due to momentum considerations. Let us consider ${}^4\text{He}$ incident on hydrogen, ${}^1\text{H}$: so $M_1=4$ and $M_2=1$ (accurate enough for the present calculations). For $M_1 > M_2$ as we have here, then the maximum lab. scattering angle, ψ , is given by $\sin \psi = M_2/M_1 = 1/4$, or $\psi = 14.5^\circ$.

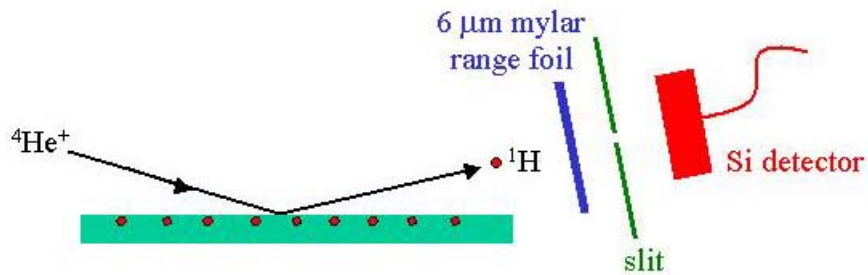
Suppose we place our detector in the **FORWARD DIRECTION**, e.g. $\psi = 30^\circ$. We then find $E_2/E_0 = 0.48$ (use $M_1=4$ and $M_2=1$, which is accurate enough instead of $M_1=4.0026031$ amu and $M_2=1.0078252$ amu; if we use the correct masses, we find $E_2/E_0 = 0.482$). However, suppose the substrate is Au ($Z_2=79$, $M_2=197$ amu)? Suppose again that the hydrogen target atoms are not sitting by themselves but are located on top of or at shallow depths into the Au substrate. The ${}^4\text{He}$ projectiles are then scattered with a large cross section from the (heavy) substrate atoms to the *same* detector angle of 30° , and will have an energy

calculated using the above equation (for E_1/E_0) with $M_1=4$, $M_2=197$: $E_{He}/E_0 = 0.9946 \cong 1$.

If we use $E_0 = 1.6$ MeV, which is a reasonable value for these experiments, then the recoil protons have $E_p=0.768$ MeV and the scattered projectiles (SCATTERED FROM THE HEAVY Au SUBSTRATE) have $E_{He} \leq 1.59$ MeV. The relevant cross sections for these two events are: Recoils – $d\sigma/d\Omega=0.43$ b/sr. The scattering cross section for such light particles may not be equal to the Rutherford value due to some probabilities for inducing nuclear reactions, as shown below -

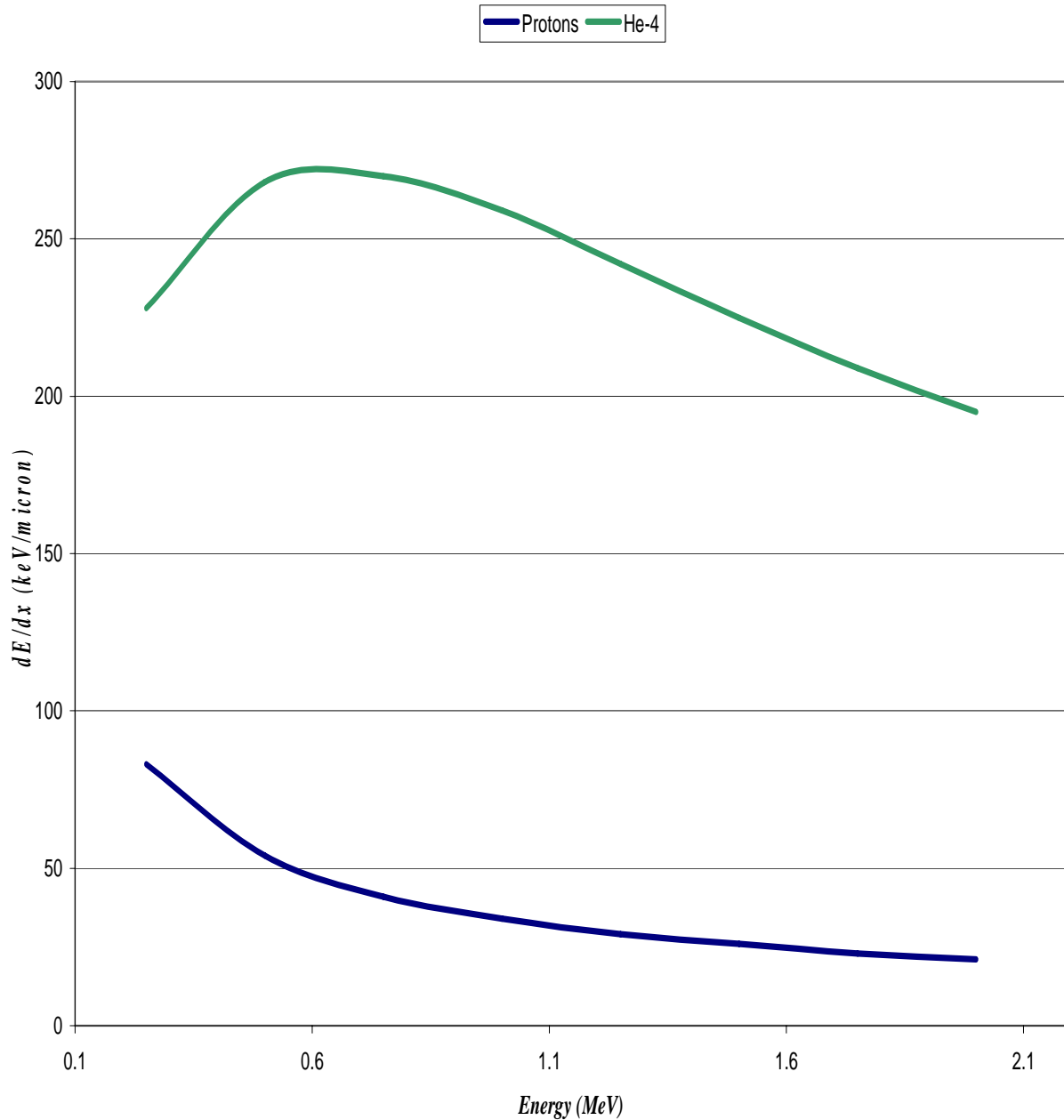


Scattered projectiles – $d\sigma/d\Omega= 2750$ b/sr. So the detector will be ‘flooded’ with the scattered projectiles. So we must put in what is called a **RANGE FOIL**, which should totally stop ${}^4\text{He}$ ions with energies up to 1.6 MeV, and ‘pass’ the protons, although the protons will suffer an energy loss in traversing this foil. The setup is shown below:



Note the large tilt that we must give to the target since this corresponds to FORWARD scattering rather than BACKSCATTERING. The detector is of the same type as we use for RBS: a Si charged particle detector. We use a fairly narrow slit to accept not too large an angular spread for the recoil protons, since this recoil energy is changing rather rapidly with angle (proportional to $\cos^2\psi$: $dE_2/d\psi=15\ [\text{keV}/^\circ]$ at $E_0=1.6\ \text{MeV}$ and $\psi=30^\circ$). Here we show a $6\ \mu\text{m}$ mylar range foil. If we calculate the energy loss of protons in this foil (remember that the stopping powers for high energy equivelocity ions scale as Z_1^2 , so that ${}^4\text{He}$ ions will lose more energy than protons in traversing the same thickness of a solid), then the proton energy is degraded to $0.495\ \text{MeV}$, whereas the ${}^4\text{He}$ ions are reduced to essentially zero energy.

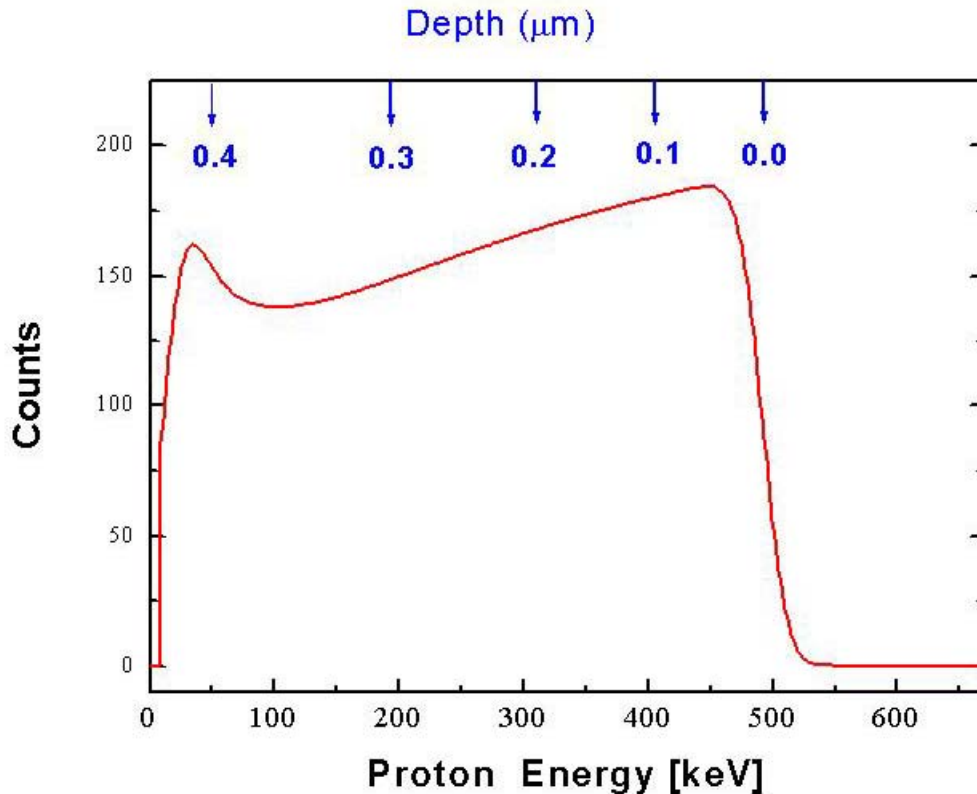
Stopping powers in Mylar



Our depth resolution is now not great since the protons suffer this energy loss straggling that we mentioned in **Lecture 1**, which broadens their distribution.

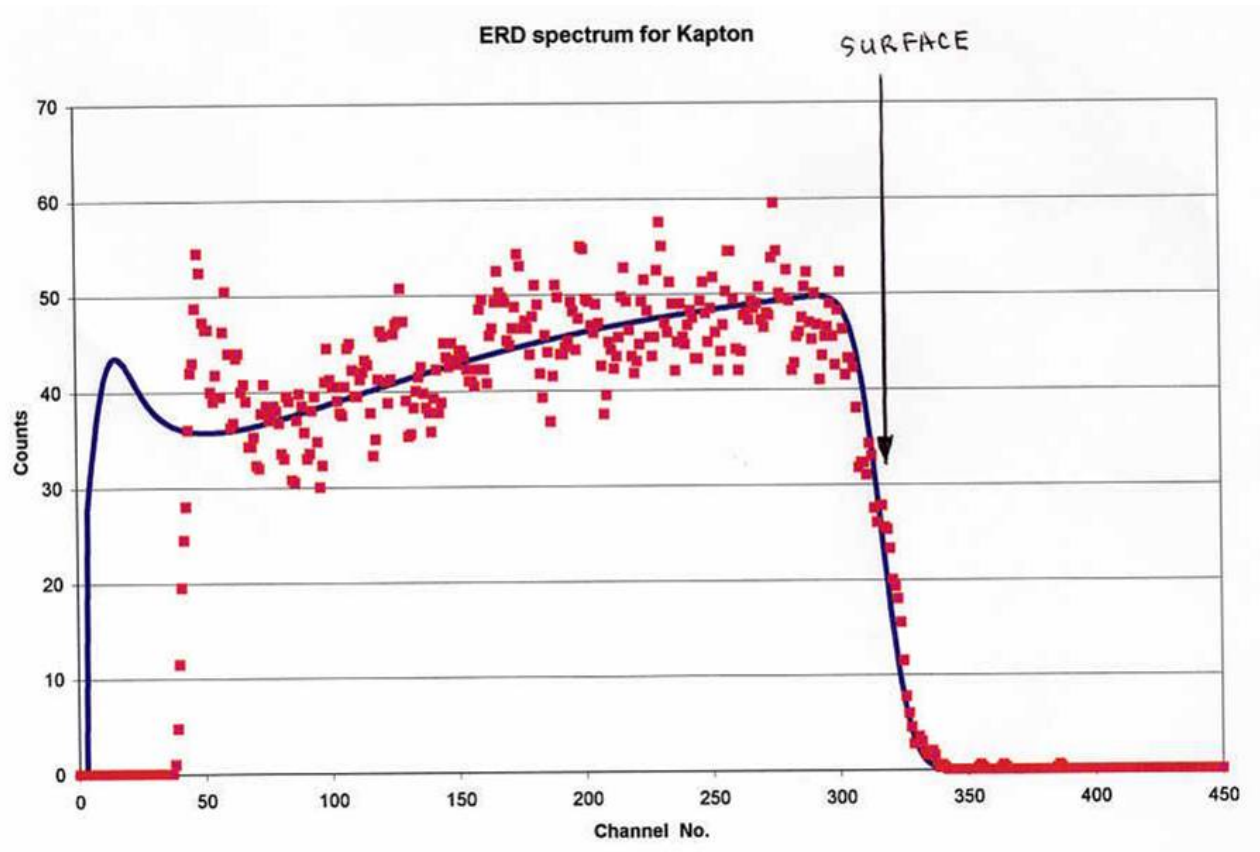
How do we quantify our measurements? We could use a solid target that has a precisely known amount of hydrogen implanted to shallow depths, or we could use a solid target that contains a significant (and precisely known) amount of

hydrogen (examples: mylar – $C_{10}H_8O_4$; kapton – $C_{22}H_{10}N_2O_5$). There are other choices of polymers, but we must be careful to check that there is no beam-induced change/damage during the measurement. We generally choose **kapton**, which is more radiation resistant than mylar. So how does the recoil proton energy spectrum corresponding to a polymer target spectrum appear?

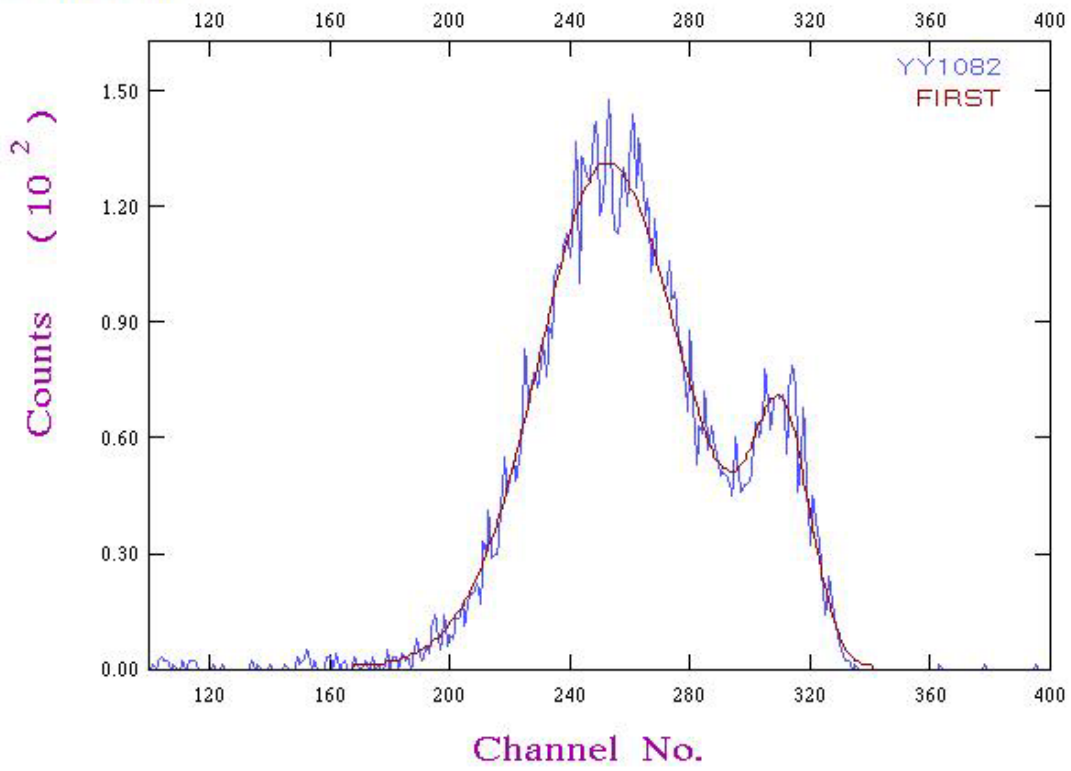
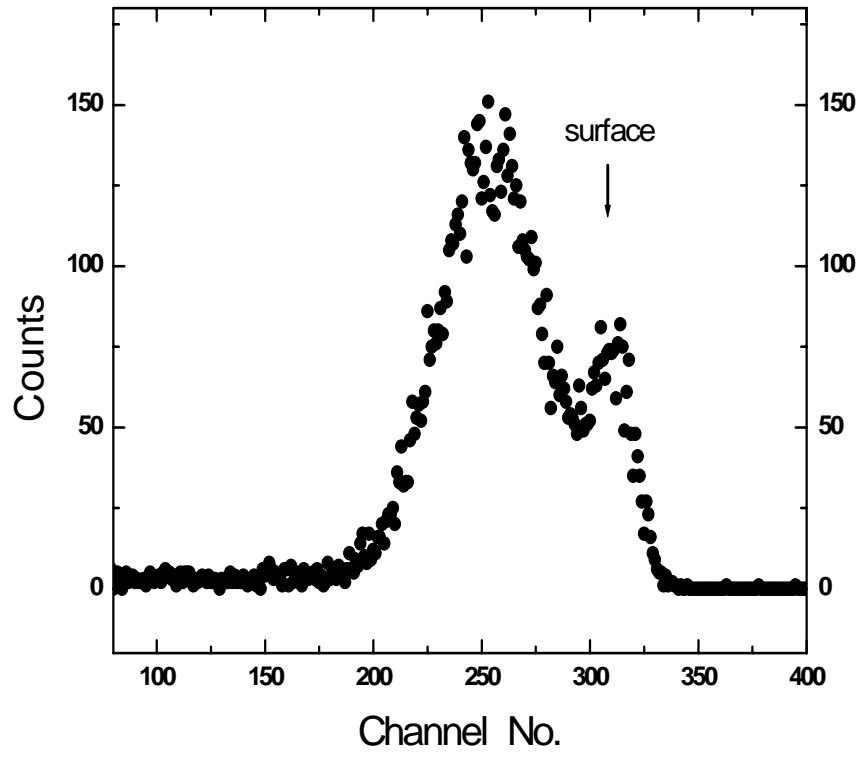


The detailed shape of this ‘thick target’ spectrum arises from a convolution of the stopping power with the scattering cross section, since the H is distributed uniformly with depth. The depth into the target as a function of detected energy is shown at the top of this **calculated** spectrum. (We can calculate this spectrum exactly, and the software to do this is available from my website: QUARK-ERD package). We use this spectrum to determine the detector solid angle, as we used the Bi-implanted a-Si target for the same purpose when using RBS. A typical solid angle in these experiments is 1 msr – smaller than for RBS.

Spectrum for a **kapton** target, $E_0=1.6$ MeV, $\psi = 22^\circ$, target tilt = 79° , together with a simulation.



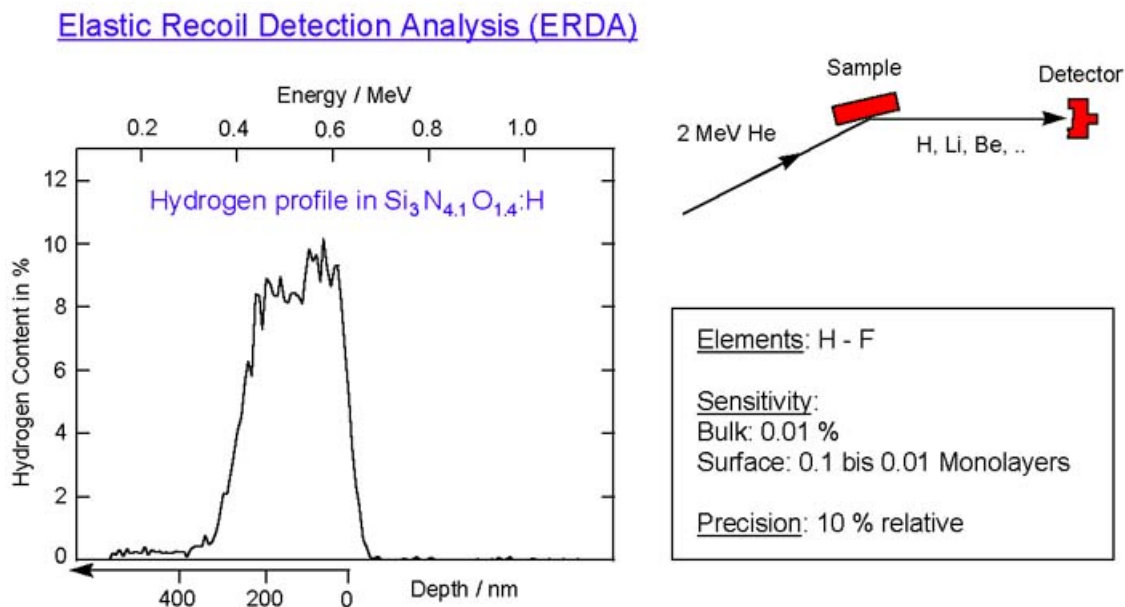
Below is a recoil proton spectrum: 1.6 MeV $^4\text{He}^+$ ions incident with the H-implanted (6 keV into a-Si) target tilted to 75° and the detector positioned at 30° and a $6 \mu\text{m}$ mylar filter in front of the detector:



Above is a calculated fit to the measured spectrum. Note the H peak at the surface, which is always seen even for UHV conditions (residual water vapour in the vacuum system corresponding to an H coverage $\sim 5 \times 10^{15}$ at./cm² for this sample). The implanted H dose ($\sim 5 \times 10^{16}$ at./cm²) is located at a depth of ~ 95 nm. Proton energy is increasing with channel number, and depth into the Si is increasing to the **LEFT**.

In providing a calculation to match the experimental spectrum, we must assume that we know the rest of the sample composition, as the depth scale is determined by the stopping powers of both the ⁴He (ingoing) and recoil ¹H (outgoing).

Now let's look at a ERD (or FRS, Forward Recoil Spectroscopy) spectrum for a 'real' target -



HEAVY ION ELASTIC RECOIL DETECTION USING TIME-OF-FLIGHT SPECTROMETRY

Suppose we use very heavy incident ions to recoil atoms/ions from within or on a target with glancing angle geometry as shown above: *e.g.* 200 MeV ^{197}Au ions. In this case, all target masses can be recoiled with appreciable energies,

remembering that (for 180°) -

$$k(180^\circ) = \left(\frac{197 - M_2}{197 + M_2} \right)^2$$

A detector ‘telescope’ is necessary for these measurements. The flight path, s , necessary for such a detection apparatus is $s \sim 1$ m. If carbon foil time detectors are used, then the flight time, Δt , between the two thin carbon foils determines the particle velocity and the total energy is measured in a downstream Si charged particle detector, as shown below -

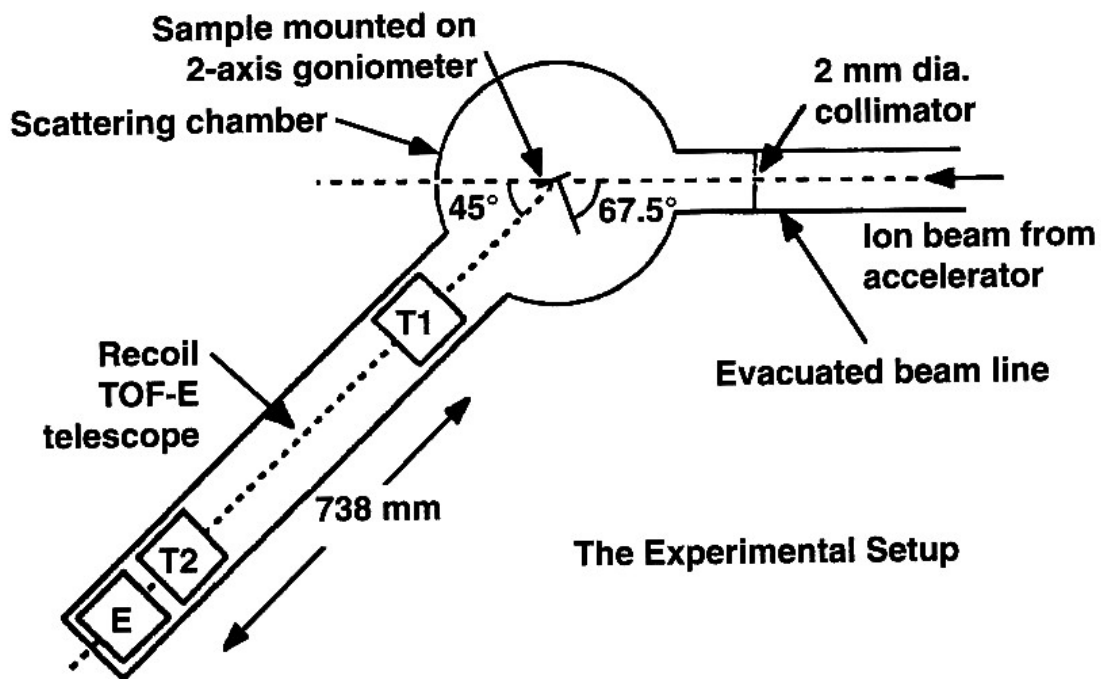
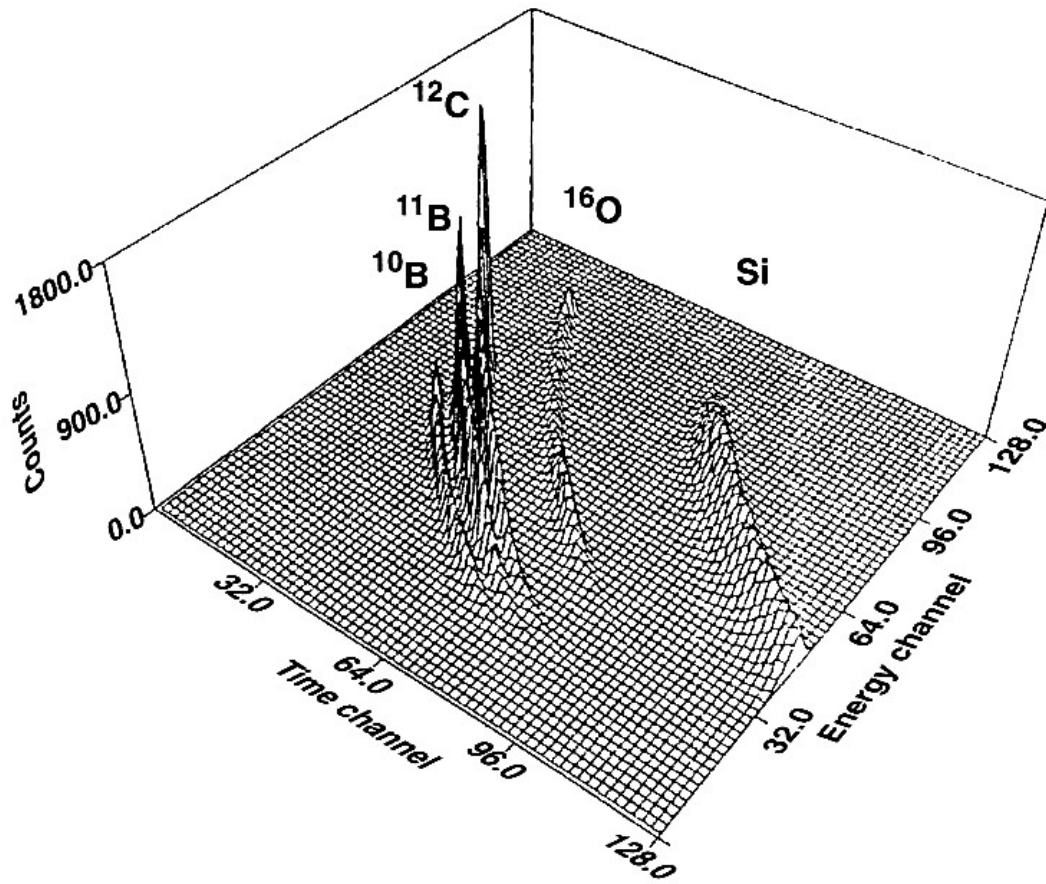


FIG. 5.32. A typical experimental configuration for mass and energy dispersive time-of-flight (TOF) ERD. The scattering angle is 45° , T1 and T2 are carbon foil time detectors which are separated by a flight path of 0.738 m, and E is a silicon surface barrier detector. [From Whitlow, 1990a].

One can also use a gas detector in conjunction with a solid state detector in a so-called ΔE -E configuration, where the gas detector provides both a timing signal and an energy loss measurement. The 'start' and 'stop' time signals are produced by detecting the electrons 'splashed' from the back surface of the (thin) carbon foils as the recoiling ions exit. This procedure allow the simultaneous measurement of *ALL* target species, in principle. The absolute yields are slightly more difficult to calibrate, but this can be accomplished. Below is shown the 2D output (flight time vs. energy) that results from the recoil particles recorded for a recoil angle of 45° –



The flight time is determined using a TAC (time-to-amplitude convertor) and the energy is determined from the Si charged particle detector. Each curved

‘island’ shows the mass-resolved yield for the mass indicated, so that even individual isotopes are resolved for the system shown here. For this geometry, the incident ^{197}Au ions cannot be scattered to 45° except for film/substrate species with atomic numbers exceeding $Z_2 = 57$, since for $M_1 > M_2$ the maximum laboratory

scattering angle is given by $\sin \Psi_{\max} = \frac{M_2}{M_1}$

Let's put in some values -

remembering that \Rightarrow
$$\Delta E_{\text{recoil}} = \frac{4M_1M_2E_0}{(M_1 + M_2)^2} \cos^2 \zeta$$

using ^{12}C as a target constituent, the recoil energy at 45° is 21.67 MeV for incident 200 MeV ^{197}Au ions. If the flight pathlength is 1 m, then the flight time will be 54 ns ($\Delta t = s/v$ and noting that $\beta = v/c = 0.046337 [E_{\text{MeV}}/M_{\text{amu}}]^{1/2}$). The time resolution for typical carbon foil detectors is ~ 0.4 ns (or better), which is more than adequate to obtain good mass resolution. Incidentally, the elastic scattering cross sections will still be Rutherford in this regime, and relatively large: *e.g.* ~ 25 b/sr in this case. One deleterious effect can be the damage inflicted by such heavy ions (*i.e.* **non-destructive??**), but we do not have time to discuss this issue in these lectures. As well, such T-o-F analyses are expensive given the extensive accelerator infrastructure needed to generate appropriate ion beams.