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HYDROGEN AND DEUTERIUM MEASUREMENTS BY ELASTIC RECOIL DETECTION USING ALPHA PARTICLES

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This paper presents recoil cross sections for both H and D for alpha particle bombardment in the energy range of 1.6 to 3.4 MeV. For hydrogen and deuterium, a non-Rutherford cross-section was found. For deuterium, a resonance at 2.15 MeV with fwhm of 75 keV was obtained.

Calculations were carried out to find the geometrical arrangement where the maximum information concerning the probing depth can be obtained in the energy range of 1–10 MeV.

In contrast to the generally accepted 30° scattering angle, another configuration is suggested.

1. Introduction

The elastic recoil detection (ERD) technique suggested by L'Ecuyer et al. [1] is a fast, nondestructive method for determining simultaneously the depth distribution of hydrogen isotopes in a heavy matrix. The energy distribution of light impurities recoiled from an inclined surface of a thick target is analysed by a surface barrier detector placed after a filter foil. The foil thickness is chosen in order just to absorb the scattered particles. In combination with the detection of backscattered ions (RBS), one can determine the depth distribution for most atomic components of the sample in a measurement of some minutes.

To identify the optimum values of parameters for different analytical problems, however, further theoretical considerations and detailed experimental studies are needed. The present paper wishes to contribute to this progress.

2. Experimental and theoretical considerations

The requirements for ERD are a low detection limit, good kinematics i.e. the best distinction between isotopes, and good depth resolution, with a simple experimental configuration where one can be able to carry out fast and routine measurements. It is clear that all these requirements cannot be satisfied simultaneously; therefore, it is necessary to find the best compromise.

The parameters that can be varied are: the analysing beam (usually atomic nuber $Z \geqslant 2$ and atomic mass $m \geqq 3$), the bombarding energy E_0, the recoil angle $0 < \theta < 90°$, the angle of incidence to the surface $0 < \alpha < \theta$, the size and shape of the beam spot, the distance between the detector and the target D, the detector aperture s, and the material and thickness of the absorber foil. Besides the kinematics, the recoil cross-section and the energy loss vs depth conversion factor $|S|$ need to be known.

Fig. 1 shows the kinematic factors as a function of scattering angle and the ratio of incident (m) and recoiled masses (M). It is clear that the lower the scattering angle θ and the lighter the particle of the probing beam the better the mass separation should be for ERD. If one analyses only hydrogen isotopes the $^4He^+$ beam is a satisfactory alternative to $^3He^+$ as the analysis in this case would be expensive without significant improvement.

The stopping factor $|S|$ can be calculated in the near surface region as:

$$|S| = \frac{kS_1}{\sin \alpha} + \frac{S_2}{\sin \beta}, \tag{1}$$

where $\beta = \theta - \alpha$ and S_1 and S_2 are the stopping powers of the incident and recoiled atoms respectively. The stopping powers are tabulated by Ziegler [2]. For larger depths $|S|$ must be evaluated by numerical methods, similar to RBS [3].

The energy calibration of stopper foil-detector and electronics system could be determined together in practice by measuring a sample with different θ angles. In our experimental setup it was linear with an accuracy of 1%. For energy resolution, however, the lateral inhomo-

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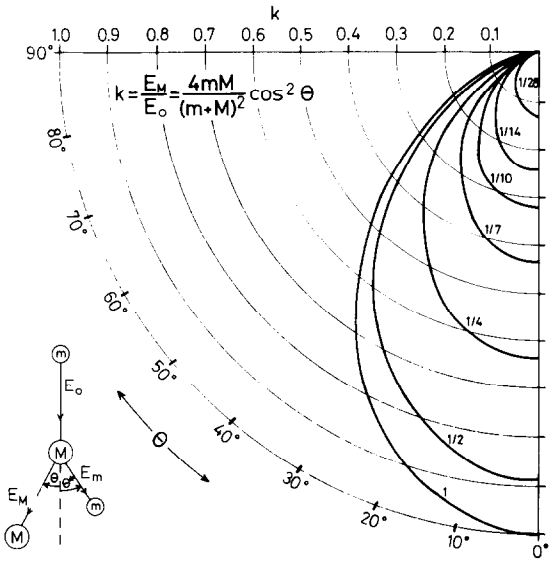

Fig. 1. Kinematic factor for the energy of recoiled particles. The plotting parameters are M/m ratios.

genity of the stopper foil is also crucial, which can be determined from taking spectra by different foils.

The beam size (d) in the scattering plane should be as narrow as possible to find an optimum depth resolution on a strongly tilted sample. During the present experiment $d = 0.2$ mm was chosen.

At the detector the acceptance angle should be a compromise between the depth resolution and the measuring time. Lowering it under a few msr is unpractical as the energy spread from the stopper foil will be the dominant contribution. The geometrical broadening at the surface [4]:

$$\Delta E_g = 2kE_0 \operatorname{tg}\theta \cdot \frac{1}{D}\left| s^2 + \frac{d^2 \sin^2\beta}{\sin^2\alpha}\right|^{1/2}, \qquad (2)$$

is determined by the ratio s/D provided the second term is negligible. It is reasonable that s/D is chosen in such a way that its contribution via ΔE_g to the energy spread would be in the same order as the others. As an example for hydrogen detection at 3 MeV and $\theta = 15°$, s/D should be 0.04 if the other contribution to the energy spread is about 40 keV. $D = 72$ mm and $s = 3$ mm satisfy this condition. These values were used in the present experiment.

To minimize the straggling of the absorber foil, low-Z materials are preferred. In our case it was an 11 μm thick aluminium foil for a 3 MeV $^4He^+$ beam.

With the above mentioned parameters fixed, E_0 and

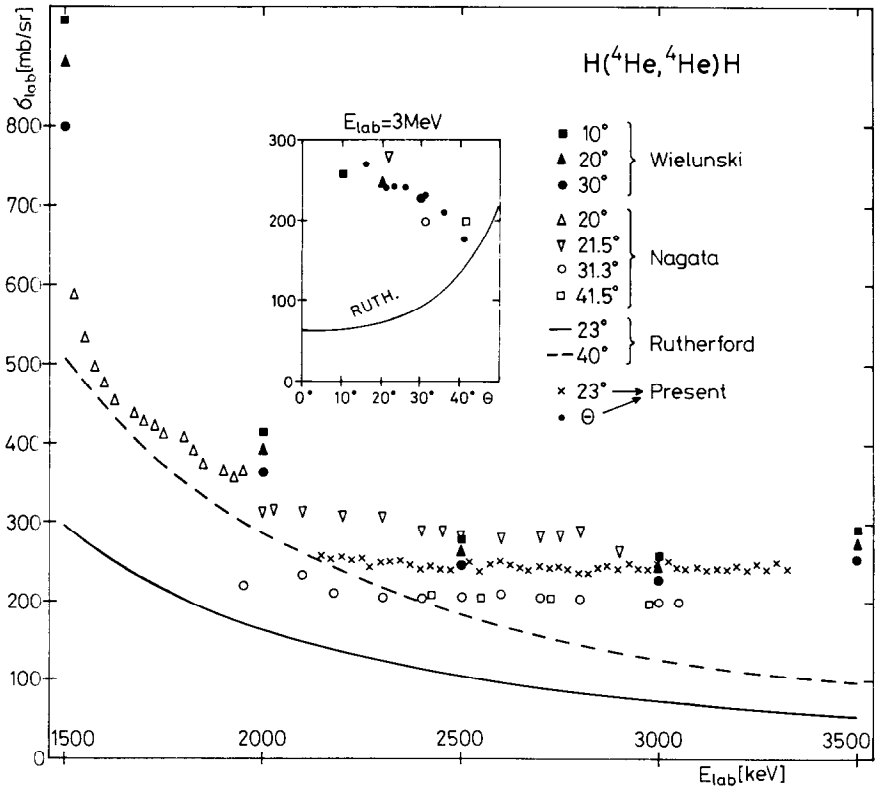

Fig. 2. Differential cross-section vs. energy (lab.) for the H(^{4}He, ^{4}He)H reaction.

θ and α can be varied to obtain the different optima of the technique.

The scattering cross-section is of non-Rutherford type and increases with decreasing scattering angle (fig. 2 and 3). Assuming an opposite behaviour, Turos and Meyer [4] concluded that $\theta = 30°$ is the optimum scattering angle. For hydrogen the sensitivity does not change much in the energy range of 1.5–3 MeV. For deuterium, however, the resonance at 2150 keV offers an improvement in sensitivity. Decreasing α and θ lower the minimum detectable concentration which is estimated ≈ 100 ppm or 10^{14} atoms/cm^2 for deuterium and a magnitude higher for hydrogen.

The depth resolution is the ratio of the total energy spread ΔE_t to $|S|$. Near to the surface

$$\Delta E_t^2 = \Delta E^2 + \Delta E_g^2, \tag{3}$$

where ΔE is the contribution of detector, foil and electronics and is independent of the angles, so

$$\mathrm{d}x = \left[\Delta E^2 + 4k^2E_0^2\ \mathrm{tg}^2\theta\left(1/D^2\right)\right.$$
$$\times \left|s^2 + d^2\sin^2\beta/\sin^2\alpha\right|\Big]^{1/2}$$
$$\times \left(kS_1/\sin\alpha + S_2/\sin\beta\right)^{-1}. \tag{4}$$

In principle with simultaneous decrement of θ and α, the depth resolution can be improved, the only limitation being is the accomplishment of the measurements. Turos and Meyer [4], Nagata et al. [5] made calculations to find an optimum for α. They fixed the scattering angle (30° or 20°) and assumed a larger bean size ($d \cong 1$ mm). It was found that the near-surface depth resolution was very good with $(\theta - \alpha) \cong 1$–$2°$ but in deeper regions became poor. On the other hand if α is small (2–5°), good resolution can be achieved at larger depth. Presumably the limited depth resolution was due to the big values of d and θ both used in experiments and calculations in previous papers.

Therefore we chose $d = 0.2$ mm, $\alpha = 4.5°$ and $\theta = 15°$, and better depth resolution was reached by minimizing the geometrical broadening (table 1).

For a depth larger than ~ 100 nm, the depth resolution cannot be improved further as multiple scattering and straggling in the sample are the main contributions to the energy spread.

To achieve the maximum probing depth, one has to find the proper θ and α at a given energy. As an example fig. 4 shows the variation of the energy loss factor for hydrogen in silicon at 3 MeV as a function of α and θ. For all energies and isotopes, by this type of

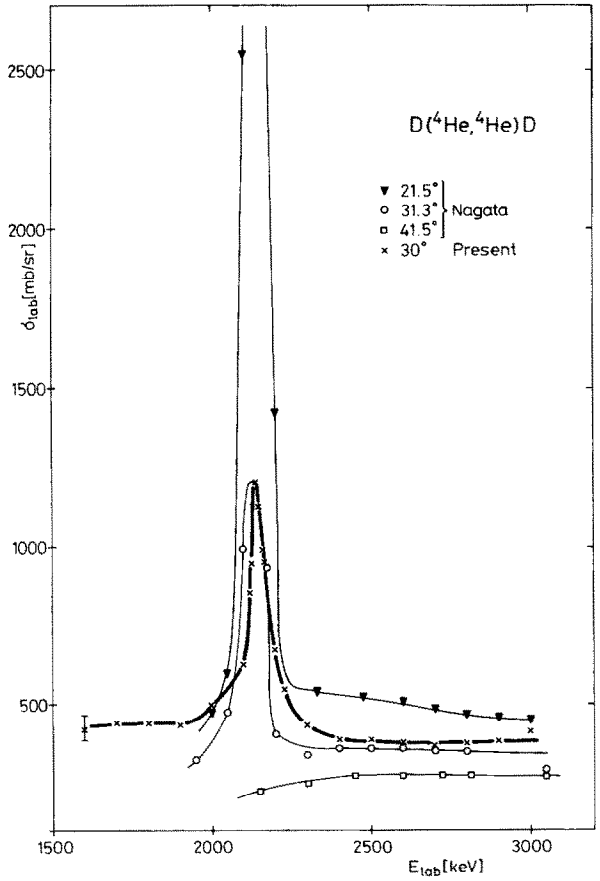

Fig. 3. Differential cross-section vs. energy (lab.) for the D(^{4}He, ^{4}He)D reaction.

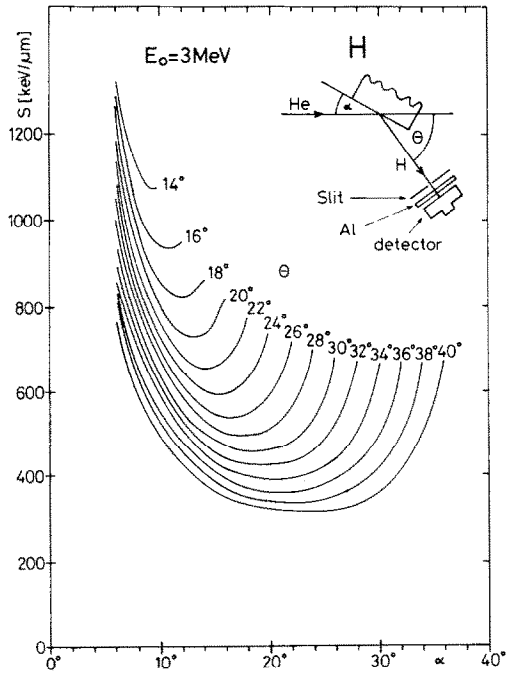

Fig. 4. Energy loss factor in Si substrate for He recoiled protons as a function of α and θ.

Table 1
ERD depth resolutions

Target	E (MeV)	θ (deg)	α (deg)	Depth (nm)	dx (nm)	Ref.
a-Si:H	2.1	30	15	0	50 exp	[6]
Si	2.5	30	2	100	23 th	[4]
a-Si:H	2.5	30	15	0	80 exp	[4]
Si	2.5	30	7	100	50 exp	[4]
Al	3	20	3	0	35 th	[5]
Al	3	20	7	310	62 th	[5]
Al	3	20	6	0	50 exp	[5]
a-Si:H	3	23	12.5	0	75 exp	a)
a-Si:H	3	15	4.5	0	22 exp	a)
a-Si:H	3	15	4.5	100	30 exp	a)
a-Si:H	3	15	4.5	200	35 exp	a)
a-Si:H	3	15	4.5	350	60 exp	a)

th: theoretical estimation.
exp: experimental data.
a) This work.

calculation one can obtain an optimum α for each θ. Fig. 5 shows the achievable maximum probing depth h as a function of E_0 and θ using the optimum value of α. Table 2 summarizes the results in the energy range of 1–10 MeV together with the optimum angles. The maximum probing depth increases to about two orders of magnitude while θ and α were around 29° and 18°, respectively.

The conclusions are the following. The sensitivity and isotope separation and depth resolution are good simultaneously if one chooses small θ and α. To obtain the maximum probing depth, the θ and α should be around 29° and 18° respectively, depending on the isotope and incident energy.

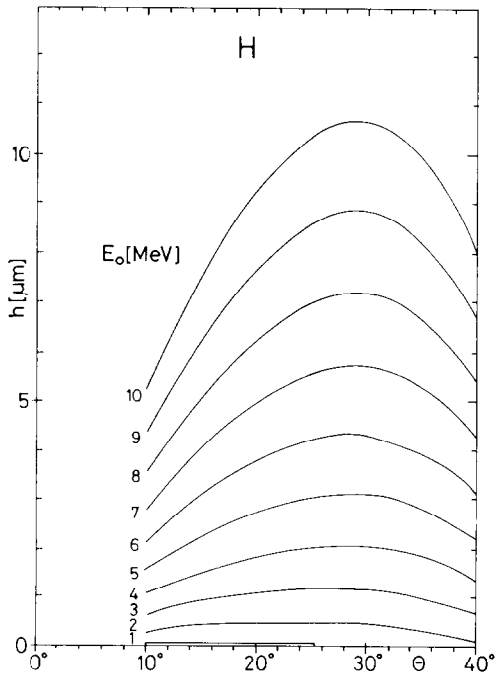

Fig. 5. The maximum analysis depth for H in Si vs. θ at different incident energies.

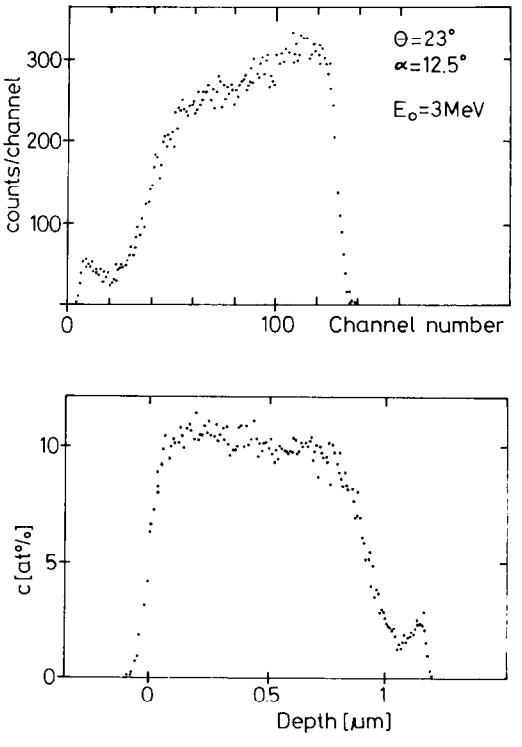

Fig. 6. The ERD spectrum and the conversion to H depth-profile in a-Si:H layer deposited onto Si. The layer thickness and deposition temperature was 0.9 μm and 350°C respectively. At low energies, the peak comes from noise.

Table 2
The maximum probing depth parameters in silicon

E_0 (MeV):	1	2	3	4	5	6	7	8	9	10
$m=1$										
θ	18	24	27	28	28	28	28	29	29	29
α	11.6	15.4	17.2	17.8	17.9	17.9	18	18.5	18.5	18.5
$h[\mu m]$	0.07	0.5	1.18	2.06	3.12	4.31	5.72	7.2	8.85	10.7
$m=2$										
θ	23	27	29	29	29	30	30	30	30	30
α	14.9	17.4	18.6	18.7	18.7	19.2	19.3	19.3	19.3	19.3
$h[\mu m]$	0.14	0.68	1.46	2.43	3.59	4.89	6.41	8.01	9.78	11.76
$m=3$										
θ	23	28	29	29	29	29	29	29	29	29
α	14.7	17.8	18.3	18.3	18.4	18.4	18.4	18.4	18.3	18.3
$h[\mu m]$	0.15	0.67	1.41	2.32	3.38	4.56	5.93	7.35	8.93	10.71

If one wants to apply both optima, a movable detector is necessary.

As a compromise we attached the detector to the sample holder in such a way that $(\theta - \alpha)$ was 10.5°. For routine measurements $\theta = 23°$ and $\alpha = 12.5°$ angles were used to obtain a probing depth of 9% lower than the maximum and 30% better depth resolution than at $\theta = 27°$ and $\alpha = 17°$ geometry. To achieve the best resolution for this arrangement the θ and α were diminished to 15° and 4.5°, respectively.

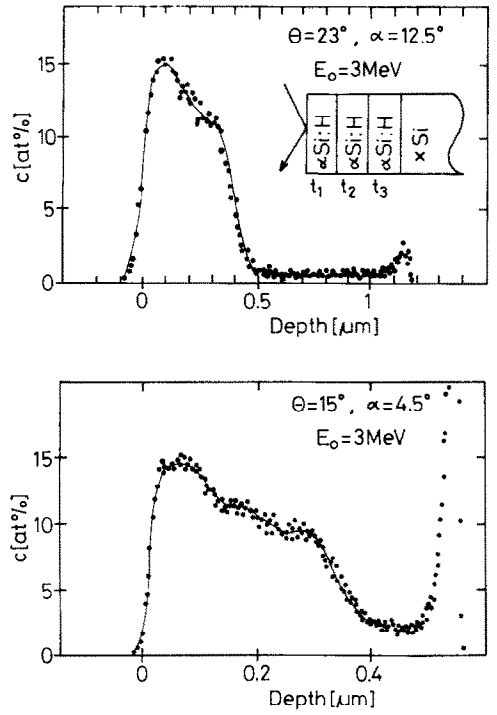

Fig. 7. H depth-profile in a-Si:H multilayer on Si at different analysing conditions. The t_1, t_2, t_3 deposition temperatures are 150, 250 and 350°C respectively. Note the improved depth resolution at $\alpha = 4.5°$. At high depth, the peak arises from noise.

3. Cross-section measurement

To determine the absolute cross-section for hydrogen, a thick, plasma-deposited silicon nitride, containing 20 ± 1 at.% hydrogen and calibrated by infrared spectroscopy, was used. For deuterium a 400 eV, 4.5×10^{16} D/cm^2 implant in silicon was applied. The results are shown in fig. 2 and 3 together with a compilation of earlier experimental and theoretical works.

4. Applications

The measured spectra can be converted into depth profiles by cross-sections and $|S|$. The latter was determined by numerical methods for larger depths. Fig. 6 illustrates this conversion for 0.9 μm thick plasma-deposited amorphous silicon on a single crystal silicon target. To determine the depth resolution a three-layer sandwich was chosen, each layer of nominal thickness 0.1 μm. The deposition temperatures were 350, 250 and 150°C respectively. Fig. 7 demonstrates the effect of θ and α for the depth resolution.

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