CLM-P 195

United Kingdom Atomic Energy Authority

RESEARCH GROUP

Preprint

BACK-SCATTERING OF keV HYDROGEN IONS IN SOLIDS

G. M. McCRACKEN
N. J. FREEMAN

Culham Laboratory
Abingdon Berkshire
1969

UNCLASSIFIED
(Approved for publication)

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G.M. McCRACKEN
N.J. FREEMAN*

(Submitted for publication in Proc. Phys. Soc. B)

A B S T R A C T

By assuming that incident ions are scattered in a single wide angle collision and that the energy loss in the solid is due only to interaction with electrons, an expression for the number of ions back-scattered from a surface is derived. Measurements have been made of the energy distribution of protons and deuterons back-scattered from titanium and niobium. Good agreement is obtained between the measured and calculated energy distributions both in general shape and in the absolute values of the number of particles back-scattered from titanium. In the case of niobium there is evidence that multiple scattering is taking place.

*U.K.A.E.A., A.W.R.E., Aldermaston, Berkshire, England.

U.K.A.E.A. Research Group,
Culham Laboratory,
Abingdon,
Berks.

February, 1969

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1. Introduction

It has been shown both experimentally (van Wijngaarden and Duckworth, 1962) and theoretically (Lindhard and Scharff, 1961) that the energy loss rate of fast ions in solids can be attributed to two separate processes. For ions with low velocities the energy loss to lattice atoms is dominant, while for ions with high velocities the energy loss is primarily to electrons.

The rate of loss of energy to electrons has been calculated by Lindhard and Scharff (1961) using a statistical model, and a simple analytical form has been obtained for the range of velocities $v \lesssim v_o$ (where v_o is the velocity of an electron in the first Bohr orbit). In the case of hydrogen ions moving in solids, the electron loss process is dominant above energies of 1 keV (while $v = v_o$ at 25 keV for hydrogen and 50 keV for deuterium). However, this does not preclude the possibility of a small number of scattering collisions, and where such collisions give rise to large angular deflections then reflection or back-scattering of incident ions can occur. These back-scattered ions have been observed by a number of investigators (cf. Carter and Colligon, 1968) and in recent years the cut off in the energy spectrum has been used as a method of determining the atomic masses of the elements in a target material (Davies et al. (1967), Patterson et al. (1965)). Patterson et al. (1965) have given the differential form of the energy distribution of back-scattered ions, but because only an approximate empirical expression was available for $\frac{dE}{dx}$ at the high energies then of interest only qualitative agreement between theory and experiment was obtained. Recently Behrisch (1968) has measured the energy and angular distributions of protons back-scattered from copper, though again at energies above those at which the Lindhard and Scharff theory is applicable. Morita et al. (1968) have measured angular and energy distributions of protons scattered in copper and gold in the energy range 6 - 40 keV and compared them with a single scattering theory using empirical data for the energy loss process. The present paper sets out to calculate the energy and angular distribution of ions back-scattered in a single collision in the energy range where the Lindhard and Scharff theory applies, and then goes on to compare the results with experimental measurements of the energy distribution of deuterium ions scattered from titanium and niobium targets. A preliminary report of the present paper has been published (McCracken and Freeman 1968), though an error of a factor 3 in the absolute value of these earlier results has since been discovered.

2. Theory

Let us consider an ion normally incident on a target, where the atomic number and mass of the incident and target atoms are z_1, m_1, z_2 and m_2 respectively. Let us assume that the incident ion slows down in the target material without undergoing any scattering, that it is then scattered by a single scattering event through a large angle and that it returns to the surface without further scattering.

From the theory of Lindhard and Scharff the rate of energy loss is given by

$$\frac{dE}{dx} = z_1^{\frac{1}{6}} \frac{z_1 \; z_2}{(z_1^{\frac{2}{3}}+z_2^{\frac{2}{3}})^{\frac{3}{2}}} \; 8\pi n e^2 a_o \sqrt{\frac{E}{E^1}} = KE^{\frac{1}{2}} \qquad \ldots (1)$$

where n is the density of ions in the solid, a_o the Bohr radius, and E^1 is the energy at which the ion velocity equals the velocity of an electron in the first Bohr orbit. From (1) we obtain the energy of the ion for any distance x travelled within the solid, $E(x) = (\sqrt{E_o} - \frac{1}{2} Kx)^2$, where E_o is the initial ion energy, and hence we obtain the probability of a scattering event in any element of length dx

$$dq = n\sigma \left[(\sqrt{E_o} - \tfrac{1}{2} Kx)^2, \theta \right] d\omega \, dx \qquad \ldots (2)$$

where $\sigma(E, \theta)$ is the differential scattering cross section derived from the appropriate interaction potential.

In the collision the ion will have a discrete energy loss and will then continue to lose energy continuously at a rate given by Equation (1). We will denote the energy remaining after the collision by R^2E_c, where E_c is the energy before the collision and R is given by the usual expression for elastic collisions calculated from the conservation of energy and momentum.

$$R = \frac{m_1 \cos\theta + \sqrt{m_2^2 - m_1^2 \sin^2\theta}}{m_1 + m_2} \qquad \ldots (3)$$

If the energy of the ion when it returns to the surface is E_s then it can readily be shown that the depth at which the scattering event took place is given by

$$X_c = \frac{2(R\sqrt{E_o} - \sqrt{E_s})}{K(R - \sec\theta)} \qquad \ldots (4)$$

and hence using (2) we obtain the probability of an ion being scattered and emerging from the solid with an energy E_s

$$N(E_s) d\omega \, dE_s = \sigma \left[\left(\frac{\sqrt{E_s} - \sqrt{E_o} \sec\theta}{R - \sec\theta} \right)^2, \theta \right] \frac{n \, d\omega \, dE_s}{K E_s^{\frac{1}{2}} (R - \sec\theta)}$$

$$\ldots (5)$$

When the ion beam is incident at an angle φ with respect to the normal the corresponding distribution in the plane of the ion beam is

$$N(E_s)d\omega\ dE_s = \sigma\left[\left(\frac{\sqrt{E_s}-\sqrt{E_o}\cos\varphi\sec(\theta+\varphi)}{R-\cos\varphi\sec(\theta+\varphi)}\right)^2,\theta\right]\frac{n\ d\omega\ dE_s}{KE_s^{\frac{1}{2}}(R-\cos\varphi\sec(\theta+\varphi))} \quad \ldots (6)$$

If one assumes that simple Rutherford scattering applies to the collisions of interest then

$$\sigma(E,\ \theta)\ d\omega = \frac{1}{16}\left(\frac{z_1^2 z_2^2 e^4}{E^2}\right)\text{cosec}^4\left(\frac{\theta}{2}\right)d\omega$$

and one obtains for normal incidence

$$N(E_s)d\omega\ dE_s = \frac{f(z)e^2\sqrt{E^1}\text{cosec}^4\left(\frac{\theta}{2}\right)(R-\sec\theta)^3 d\omega\ dE_s}{128\pi a_o\sqrt{E_s}\ (\sqrt{E_s}-\sqrt{E_o}\sec\theta)^4} \quad \ldots (7)$$

where $f(z) = z_1^{\frac{5}{6}}\ z_2(z_1^{\frac{2}{3}}+z_2^{\frac{2}{3}})^{\frac{3}{2}}$. It is noted that for hydrogen isotope bombardment of most metals $z_1^{\frac{2}{3}} << z_2^{\frac{2}{3}}$ and so $f(z) \simeq z_2^2$.

In fact only in a comparatively few situations, when using low z targets, will both the ion velocity and the screening be low enough that the Lindhard and Scharff and the Rutherford equations be good approximations simultaneously. In the comparison with experiment described later, we have used the screened coulomb differential scattering cross section as calculated by Everhart et al. (1955). Nevertheless the expression for N(E) given in Equation (7) is useful in giving a simple analytical description of the back-scattering behaviour of hydrogen and deuterium ions at low energies.

In some experiments the total number of back-scattered ions have been measured rather than the energy or angular distribution. It is therefore of interest to integrate Equation (7) to obtain the total number of back-scattered ions N_{TE} with energy greater than E.

$$N_{TE} = \int_{\frac{\pi}{2}}^{\pi}\int_{E}^{E_o} N(E_s)\,d\theta\,dE_s = \frac{f(z)e^2\sqrt{E^1}}{64a_o}\int_{\frac{\pi}{2}}^{\pi}\int_{E}^{E_o}\frac{(R-\sec\theta)^3\sin\theta\,d\theta\,dE_s}{\sin^4\left(\frac{\theta}{2}\right)\sqrt{E_s}(\sqrt{E_s}-\sqrt{E_o}\sec\theta)^4}$$

$$= -\frac{f(z)e^2\sqrt{E^1}}{96\,a_oE_o^{\frac{3}{2}}}\int_{\frac{\pi}{2}}^{\pi}\frac{\sin\theta}{\sin^4\left(\frac{\theta}{2}\right)}\left(\frac{R-\sec\theta}{1-\sec(\theta)}\right)^3\left\{1-\frac{(1-\sec\theta)^3}{\left(\sqrt{\frac{E}{E_o}}-\sec\theta\right)^3}\right\}d\theta \cdots \quad (8)$$

$= -1.42 \times 10^{-3}\ f(z)\ I(\theta)\ E_o^{-\frac{3}{2}}$ where E is in kV, and the integral is written as $I(\theta)$.

The integral $I(\theta)$ has been evaluated numerically as a function of $\frac{E}{E_o}$ for values of R and is shown in Fig. 1. Of course for low values of $\frac{E}{E_o}$ where E is less than 1 keV and the energy loss to lattice atom is therefore appreciable, Equation (8) is a very poor approximation.

Calculated values for N_{TE} for hydrogen (or deuterium) ions striking a number of metals at normal incidence are shown in Fig. 2. The earlier provisos concerning the application of Rutherford scattering of course apply, so that the results at high z_1 or z_2 and low energy should be treated as a qualitative indication only. The curves have been dotted in the region where screening makes a reduction of more than 10% in the differential scattering cross section, as calculated by Everhart et al. (1955). The full curves have been calculated using the values of I(θ) when R = 1.0. This introduces a slight error in N_{TE} for light elements, though even for aluminium (where R = 0.93 to 0.96) N_{TE} is only about 10% high. The values of N_{TE} for aluminium and beryllium have been separately computed, integrating I(θ) with the correct values of R included as a variable, and these are shown in Fig. 2 also.

The calculated values of N_{TE} may be compared with the experimental values obtained by a number of investigators shown in the following Table.

<u>Measurements of total number of back-scattered ions N_T</u>

	Target	Incident Particle	Energy keV	N_T% Theory	N_T% Exp.	Method	
Barnett (1966)	Si	H^o	20	0.7	3	Energy loss:	All charge states measured
Panin (1962)	Mo	H^+	7	20	2.5	Current Measurement	Only +ve ions measured
	Mo	H^+	35	3	2.3		
Fogel et al. (1960)	Mo	H^+	22	6	1.56		
	Mo	H^-	22	6	2.03		

In the first result quoted in the Table the percentage of incident ions back-scattered has been estimated indirectly, but all charge states are taken into account. Results taken by current measuring techniques have to be interpreted very carefully to allow for secondary and tertiary reactions with the collector as discussed by Kaminsky (1965). Moreover, by current measurement one can at best measure those ions which are back-scattered in a charged state, while neutrals go undetected. Hence some correction for charge exchange has to be made as discussed later. The very small variation in reflection coefficient with energy in the results of Panin is probably due to the product of decreasing scattering cross section and increasing charge exchange cross section ratio σ_{01}/σ_{10}.

3. Experiment

The apparatus used to measure the energy distribution of back-scattered ions is shown in Fig. 3. The primary beam is produced, accelerated and mass analysed in an e.m. separator (Freeman et al., 1960). The target is mounted in a chamber differentially pumped with respect to the separator using sputter ion pumps and maintained at a pressure $\sim 10^{-8}$ torr. The incident beam strikes the target at 45° with respect to the normal and the back-scattered ions which are emitted in a direction normal to the target surface are analysed with a conventional 30 cm radius, 90° sector magnetic field, mass spectrometer. Ions are detected by single particle counting techniques using a scintillation detector similar in principle to that described by Daly (1960). The high voltage electrode has been modified so that the ions are accelerated by 25 kV directly onto it so that 100% detection efficiency should be maintained over the whole energy range.

The energy spectrum of the secondary ions is measured by scanning the magnetic field with all electrodes in the target chamber at ground potential. A momentum spectrum is thus obtained which can readily be corrected to an energy spectrum if the mass of the scattered ion is assumed and the resolution of the system known. The measured resolving power at half peak height was 380. The effective solid angle submitted by the detector to the target was estimated to be 3.2×10^{-6} steradians, and is probably accurate within a factor of 2.

Two targets, one of niobium and the other titanium, were used. Both were of minimum purity 99.9% and were prepared from standard sheet by degreasing and heating in vacuum to 700°C. Surface erosion due to sputtering is expected to clean the target and to be the principal factor controlling surface conditions during the experiments. A primary beam current of about 5 μA was used on a target area of 0.025 cm^2. The target temperature was maintained in the range 100 - 200°C.

4. Discussion

Results are shown in Fig. 4b of the energy distribution of D^+ ions scattered (as D^+ ions) from the titanium and niobium targets. The relative error on the basis of the counting statistics is better than ± 8% for titanium and ± 5% for niobium. The absolute error depends primarily on the estimate of the effective solid angle subtended by the detector. The results can be compared with the energy distribution calculated from Equation (6) and the differential scattering cross section for screened coulomb collisions as calculated by Everhart et al. (1955), Fig. 4a. There is seen to be agreement between the experimental values of the cut off, and the values calculated on the basis of a single scattering model to better than 0.5% in all cases.

In comparing experimental and theoretical distributions allowance must however be made for charge exchange in the target. Secondary negative ions have been detected and it is likely that secondary neutral atoms are also produced. Measurements have been made by Phillips (1955) of the fraction of an incident proton beam which was transmitted through thin targets in various charged states, as a function of energy. Unfortunately no measurements have been made of the fraction transmitted through titanium or niobium as positive ions. However, the behaviour of the five metals investigated by Phillips was very similar, the fraction of positive ions increasing linearly with energy in the range 4 - 50 keV. A linear relationship was therefore assumed for titanium and the total number of particles back-scattered in all charge states was then calculated from the experimental results using the linear relationship which gave the best fit with the E_o = 60 keV theoretical curve for titanium, i.e.

$$\% D^+ = 67 + 3.8E \quad \text{where E is in keV} \qquad \ldots (9)$$

Since the percentage of protons measured by Phillips at 50 keV was in the range 58 - 74% there appears to be a discrepancy in the absolute values of the experimental and theoretical results of about a factor 4. Part of this may be accounted for by the uncertainty in the estimate of the solid angle subtended by the detector.

Corrections calculated from Equation (9) have been applied to both titanium and niobium results for comparison with theory as shown in Fig. 4a. It is seen that there is good agreement with titanium, less good with niobium. No single correction curve could produce agreement between the three experimental and theoretical niobium results. It must therefore be concluded that a single collision model is not adequate in this case, due to the higher z than in titanium and therefore the greater probability of multiple collisions. The fall off in the measured distribution at very low energies is also probably due to multiple scattering and thus a significant energy loss to lattice atoms. However, there is also the possibility that the linear correction for charge exchange is not a good approximation for niobium. It was found that with niobium the shape of the energy spectrum depended more critically on surface conditions than for titanium and that in an unbaked vacuum system the slope of the energy distribution was steeper than results after thorough degassing.

Measurements of the energy spectrum of scattered hydrogen ions show very similar behaviour to those for deuterium even at energies up to 50 keV, where the Lindhard and Scharff energy loss formula, Equation (1), is not expected to hold.

The absolute value of the scattered hydrogen ion spectra is slightly higher than the deuterium spectra, but is within the reproducibility of the absolute values of the counting rates, i.e. $\pm$ 5–8%. On the basis of Equation (6) one would expect the proton spectra to be ~ 3% higher for niobium and ~ 6% for titanium.

A more direct comparison with theory should be obtained by measuring ions scattered in all charge states. Some recent measurements of this type have been made by Behrisch (1968) on the back-scattering of protons from copper using a silicon surface barrier detector. The primary energy in this case was 120 keV so that simple Rutherford scattering will be applicable. However, at this energy the ion velocity will be much greater than v_o and hence the Lindhard and Scharff expression for $\frac{dE}{dX}$ will not strictly apply. Nevertheless, good agreement is obtained between experimental results and the distributions calculated from Equation (7) (modified for non-normal incidence). Results for the angular distribution are shown in Fig. 5. A normalization factor of 3.1 has been used to obtain the best fit.

It is interesting that, despite the target being a single crystal, the theory gives a good overall fit to the distribution apart from the channelling directions. The poorer fit at low scattering angles is probably due to surface roughness, since these particles are being emitted at grazing angles.

5. Conclusion

At low energies a significant number of protons and deuterons incident on solid surfaces are back-scattered. Both the angular and energy distributions seem to be quite well described by a model taking separately into account a continuous energy loss process in the solid and a single elastic scattering event. In the case of niobium the atomic number z is sufficiently high apparently for multiple scattering to make an appreciable contribution. The absolute number of scattered ions measured experimentally is greater than that calculated and the discrepancy after allowing for charge exchange is slightly greater than the estimated error, although with the geometry in the present experiment this estimate is difficult to make precisely. In the experiments by Behrisch on copper where the geometry is simple and no charge exchange corrections have to be made the experimental results are also a factor of 3 higher than the calculated ones. Thus it is possible that there is a contribution from multiple scattering events in the case of titanium and copper, though the effect on the shape of the spectra is small.

There is still a difficulty in calculating the proportion of ions back-scattered in the charged state. The direct comparison allowed between theory and experiment when all charged states are measured makes it desirable to extend the techniques used by Behrisch to lower energies where the Lindhard and Scharff theory applies. It would also be interesting to extend the theory to two or more collisions to see whether there should be a significant contribution from multiple scattering.

Acknowledgements

One of us (G.M. McC) is grateful to Dr. R. Behrisch for some valuable discussions and for unpublished details about his experimental measurements.

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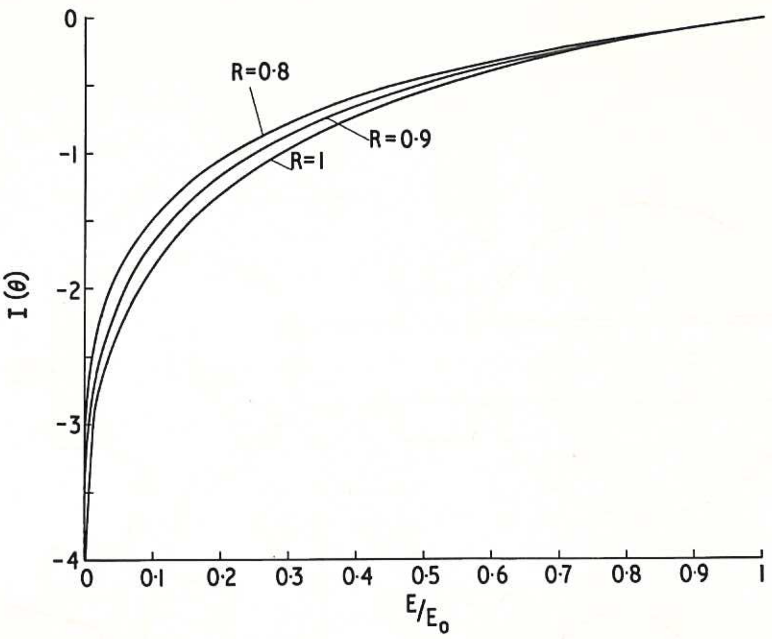

Fig. 1 (CLM-P 195)

Integral I(θ) as a function of energy for different energy losses in the scattering collision

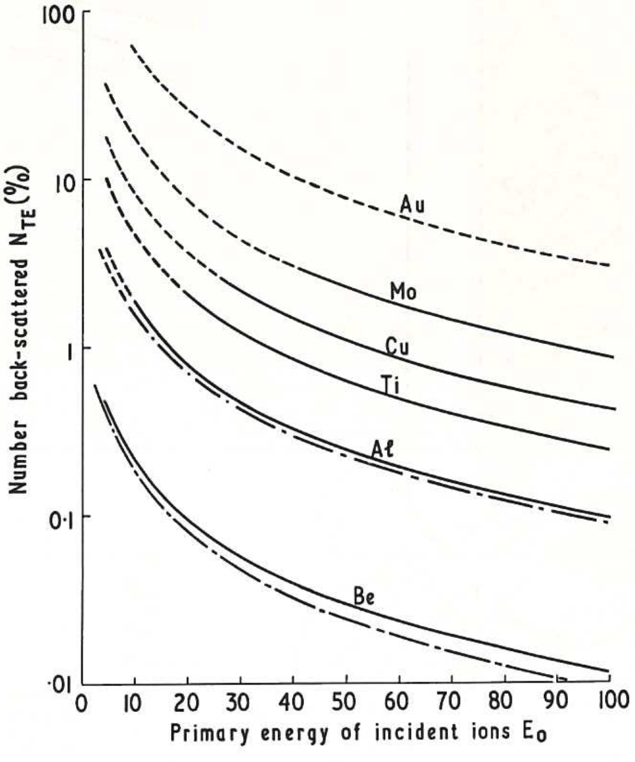

Fig. 2 (CLM-P 195)

Number of ions backscattered with an energy greater than 1 keV assuming Rutherford scattering

———— Uncorrected for energy loss in scattering collision

—·—·— Corrected for energy loss in scattering collision

---------- Effect of screening greater than 10%

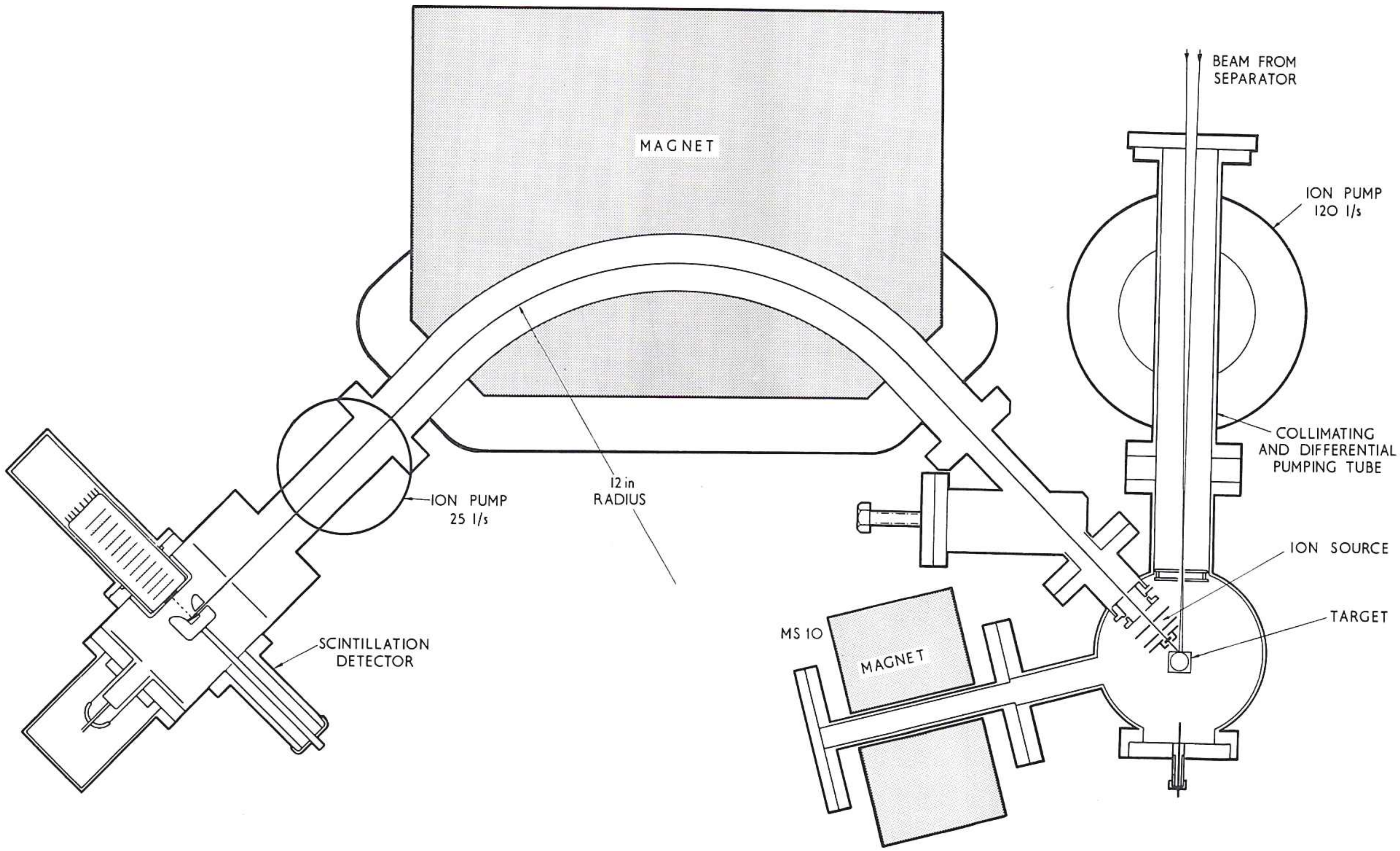

Fig. 3 Schematic of experimental apparatus (CLM-P195)

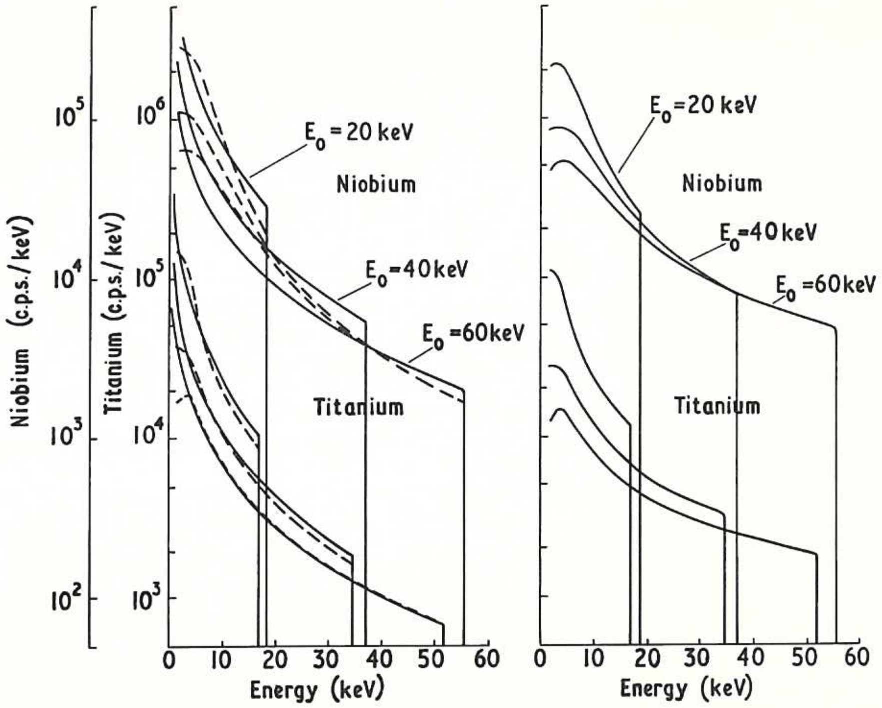

Fig. 4 (CLM-P 195)

Energy distribution of D^+ backscattered from Nb and T_i

(a) ——— Theoretical distribution

- - - - Experimental distribution normalized and corrected for charge exchange

(b) ——— Experimental distribution

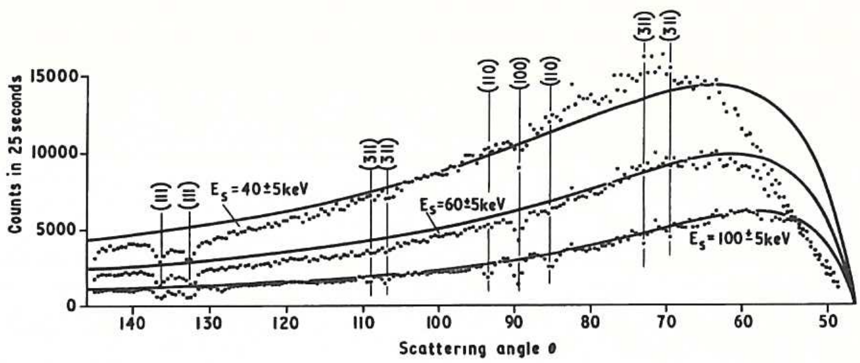

Fig. 5 (CLM-P 195)

Angular distribution of H^+ backscattered from single crystal copper

——— Theory

.... Results from R. Behrisch (Can. J. Phys., 46, 527, 1968)