Energy Loss by 8.86-Mev Deuterons and 4.43-Mev Protons*

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Using deuterons of average energy 8.86 Mev, the stopping powers relative to air have been measured for the following gases: H_2, He, N_2, O_2, Ne, A, Kr, Xe, CH_4, and CO_2. Absolute stopping cross sections were measured, using 4.43-Mev protons (of equal velocity) for H_2, air, and Kr, and the deuteron results were normalized to give absolute stopping cross sections for all of the gases. Comparison is made with the theoretical results of Aron *et al.*, Walske, and Lindhard and Scharff.

It was found that the NaI(Tl) crystal exhibited a nonlinearity in its response curve of light output *versus* proton or deuteron energy.

INTRODUCTION

In recent years many measurements have been made of the energy loss by fast ions in both solids and gases with the result that dE/dx is well known as a function of proton energy in the low-energy region up to about two Mev[1] and at high energies—18 Mev [2] and 350 Mev.[3] There remains a significant gap in the measurements in the intermediate region where one is particularly concerned with the stopping powers of counter gases and absorbers for reaction products from nuclear disintegration experiments. Here calculations[4] of the stopping cross sections provide the only data on gases.

Aside from their use for practical purposes, further measurements in the intermediate energy region are of physical interest in testing the theories used in attempting to understand the energy-loss process. A recent interesting example of such a theory is that of Lindhard and Scharff[5] in which an attempt is made, on the basis of the statistical model of the atom, to arrive at a universal function which relates the stopping power and energy for all materials and particle energies.

In the present paper, we shall report stopping cross-section measurements at a single ion (deuteron or proton) velocity, corresponding to a proton energy of 4.43 Mev, for the gases H_2, He, N_2, air, O_2, Ne, A, Kr, Xe, CH_4, and CO_2.

* Work done under the auspices of the U. S. Atomic Energy Commission.

[1] Reviews of the low-energy data are given by S. K. Allison and S. D. Warshaw, Revs. Modern Phys. **25**, 779 (1953) and by Ronald Fuchs and W. Whaling of the California Institute of Technology in their unpublished report on stopping cross sections.

[2] D. G. Sachs and J. R. Richardson, Phys. Rev. **83**, 834 (1951); **89**, 1163 (1953).

[3] C. J. Bakker and E. Segrè, Phys. Rev. **81**, 489 (1951); R. Mather and E. Segrè, Phys. Rev. **84**, 191 (1951).

[4] Aron, Hoffman, and Williams, Atomic Energy Commission Report AECU-663 (UCRL-121), 1949 (unpublished). W. A. Aron, University of California Radiation Laboratory Report UCRL-1325, 1951 (unpublished). A more extensive compendium including the earlier work of Aron, Hoffman, and Williams is provided by M. Rich and R. Madey, University of California Radiation Laboratory Report UCRL-2301, 1954 (unpublished).

[5] J. Lindhard and M. Scharff, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **27**, No. 15 (1953).

APPARATUS AND EXPERIMENTAL ARRANGEMENT

Deuteron Measurements

In the first part of the experiment, the relative stopping powers of the gases were measured by observing the relative amounts of the gases required to give the same energy loss to deuterons of 10-Mev incident energy from the Los Alamos 42-inch cyclotron. The energy-sensitive detector used was a NaI(Tl) crystal whose light response is known to be approximately linear[6,7] in energy for intermediate-energy protons and deuterons. For the deuterons used in this experiment the resolution of the pulse-height groups was approximately 3.3 percent.

A schematic diagram of the apparatus is shown in Fig. 1. The external, electrostatically deflected beam of the cyclotron was brought through a beam tube and scattered through an angle of 90 degrees from a 1.25-mg/cm² gold foil lying in a plane at 45 degrees to the unscattered beam direction. The scattered beam entered a brass absorption cell through a 3.01-mg/cm² Dural

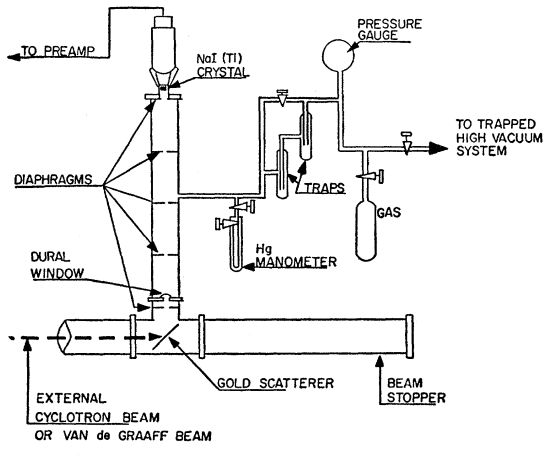

FIG. 1. Schematic diagram of apparatus for measuring stopping cross sections of gases.

[6] Taylor, Remley, Jentschke, and Kruger, Phys. Rev. **83**, 169 (1951); Taylor, Jentschke, Remley, Eby, and Kruger, Phys. Rev. **84**, 1034 (1951); F. S. Eby and W. K. Jentschke, Phys. Rev. **96**, 911 (1954); also private communication from Dr. Jentschke.

[7] J. G. Likely and W. Franzen, Phys. Rev. **87**, 666 (1952).

window and was collimated by three antiscattering diaphragms before entering the defining aperture in front of the crystal. The limiting aperture has a diameter of 1.016 cm. The total distance in which energy loss in the absorbing gas could take place between the entrance window and the surface of the scintillator was 90.43 cm. The scintillation crystal had a diameter of 1.25 cm and a thickness of 0.25 cm.

Attached to the absorption cell was a glass filling system followed by a high-vacuum pumping system with a liquid-nitrogen-trapped oil diffusion pump which produced a vacuum of 10^{-5} mm Hg in the absence of absorbing gas. Two glass traps were provided for the gas fillings: one of $Mg(ClO_4)_2$ for removing moisture, and the other of dry ice and acetone. Air could be drawn into the system through a P_2O_5 drying tube, followed by an "ascerite" trap for removing CO_2. All of the compressed gases with the exception of O_2 and CO_2 were of "refined" or "research" grade. Since the gases were stored at considerably more than an atmosphere of pressure, contamination due to air leakage was negligible. Two samples of Xe were used from tanks obtained on dates several years apart, one newly opened and the other previously used for counter fillings and purified by pumping the condensed Xe and subsequently circulating the gas through a hot-calcium purifier. No difference in measured stopping power of the two xenon samples was observed within experimental error.

All of the gas samples were checked for impurities by mass-spectrometric analysis. In this analysis measurements were made at each end of the mass scale corresponding to hydrogen and xenon and continuously between masses 14 and 88 for each gas. No gaseous impurity with molecular weight between one and 131 would have escaped detection. The impurities which occurred in more than "trace" (0.001 percent) amounts are listed for each gas sample in Table I. The last column gives the numerical factors which were applied to the stopping-cross-section results in order to correct for the impurities. The water-vapor impurities were disregarded since they were trapped out in the experiment.

Gas pressures in the absorption cell and filling system were read by means of Wallace and Tiernan absolute pressure gauges which were calibrated by means of an accurate mercury vacuum manometer to within 0.2 percent. The gas temperature was measured by means of a mercury thermometer strapped to the outside of the absorption cell.

The output pulses of the scintillator photomultiplier were amplified by a Los Alamos Model 250 preamplifier and amplifier and fed to the vertical plates of a Tektronix Model 513*D* oscilloscope through its vertical amplifier. Care was taken not to approach saturation of either amplifier. The amplified scintillator pulses were presented on the cathode-ray tube and photographed by means of a camera using 4 in.×5 in. cut film. The linearity of the electronics and optical system was carefully checked by feeding attenuated pulses from a Los Alamos Model 500 mercury-relay pulser through the amplifier system at various times during the runs and photographing the resulting pulses. The model 250 amplifier system was linear to within 0.1 percent, while the oscilloscope amplifier showed a nonlinearity of approximately three percent for which a calibration correction was made.

TABLE I. Measured amounts of impurities in the tank gases used in the stopping-cross-section measurements and correction factors applied to the experimental values to give results corresponding to pure-gas conditions.

Gas	Molecular percent impurity	Correction factor
H_2	0.07 H_2O, 0.19 N_2	0.992
He	0.07 N_2, 0.03 O_2	0.995
N_2	0.06 H_2O	1.000
O_2	0.33 H_2O, 0.61 N_2, 0.28 A	1.001
Ne	0.05 H_2O, 0.11 N_2, 0.02 O_2	0.999
A	0.08 H_2O, 0.31 N_2	1.000
Kr	None	1.000
Xe	0.06 N_2	1.000
CH_4	0.28 N_2, 0.01 O_2, 0.14 C_2H_6	0.999
CO_2	0.11 H_2O, 0.77 N_2, 0.16 O_2	1.003

The experimental procedure consisted of photographing groups of deuteron pulses with gas in the absorption cell and with the cell evacuated to a pressure of 10^{-4} mm Hg. For each exposure about 1000 pulses were used. Preliminary runs were made to determine approximately how much gas was required to provide a 25 percent drop in pulse height, and then final data were taken in a continuous run using a 25 percent fractional drop in pulse height, during which the cyclotron's magnetic field, frequency, and deflection voltage were held as nearly constant as possible. During the measurements the vacuum pulse groups were frequently compared visually with a group from the pulser, and a relative comparison was made of the vacuum pulse heights on the films. The extreme relative variation of the cyclotron energy was thus found to be 0.6 percent throughout the final run.

The energy of the scattered external beam was measured by exposing a 200-micron Ilford C-2 nuclear plate in place of the scintillator with the absorption cell evacuated. We are indebted to Dr. Louis Rosen and his nuclear plate group for making the range analysis and providing us with the result that the deuteron energy was 10.05±0.13 Mev.

Proton Measurements

The deuteron measurements described above provide accurate determinations of the stopping powers of the various gases relative to air, since approximately the same fractional drop in pulse height was used in each case. If one assumes perfect linearity of the light response of the NaI(Tl) crystal to deuteron energy, one can determine the energy loss ΔE in terms of the fractional change $\Delta h/h_0$ in pulse height as $\Delta E = E_0$

$\times(\Delta h/h_0)$, where h_0 is the pulse height corresponding to incident deuteron energy E_0. One can then obtain the absolute rate of energy loss $\Delta E/\Delta x$. However, as will be discussed later, it appears that NaI(Tl) is slightly nonlinear in its light response to protons and deuterons, and this possibility made it desirable to determine the absolute scale for the stopping powers separately.

In order to make absolute measurements of $\Delta E/\Delta x$ a separate experiment was carried out with protons of the same incident velocity as that of the deuterons for the three gases H_2, air, and Kr. Protons of accurately known energy were obtained from the Los Alamos large van de Graaff accelerator, and the experiment was performed as in the deuteron case with the following exception: the lower-energy pulse-height group corresponding to partial absorption of the proton energy in the gas was "bracketed" by means of groups corresponding to protons of known energy from the van de Graaff with the absorption cell evacuated. The van de Graaff energies were very accurately determined by means of a 90-degree magnet whose field was measured with a radio-frequency proton-moment magnetometer. Interpolation in the small interval between the known bracketing energies yielded the energy of the proton group after passing through the gas absorption cell, independent of any slight nonlinearity of the NaI(Tl) crystal response.

In carrying out this part of the experiment, protons of incident energy 5.029 Mev were used. In order to obtain protons of this energy after energy loss in the gold foil, scattering through 90 degrees in the gold foil, and energy loss in the dural window, the van de Graaff voltage was set at 5.309 Mev. The gas pressures used were one-half those used in the deuteron experiment in order to give the same fractional energy loss and mean velocity in the gases.

DATA AND RESULTS

The data were obtained in the form of photographs of groups of pulses which were delay-line clipped to provide almost flat tops approximately one microsecond in duration. A base line was either exposed or drawn on each photograph. In order to make a quantitative analysis of the pulse heights, the photographic negatives were scanned with a Leeds and Northrup microdensitometer, the Brown recorder of which provided a graph of photographic density as a function of displacement from the base line of the negative. A sample microdensitometer trace corresponding to scanning in one direction is shown in Fig. 2. In each case the negative was scanned in both directions, giving mirror-image traces, in order that small instrumental errors should cancel. The background seen in this graph was caused by neutrons and γ rays produced in various parts of the cyclotron external-beam apparatus. Very low-energy background pulses were excluded by the setting of the sweep threshold of the oscilloscope. In scanning, the light spot (of negligible dimensions) moved in the direction perpendicular to the base line and to the flat tops of the scintillator pulse traces.

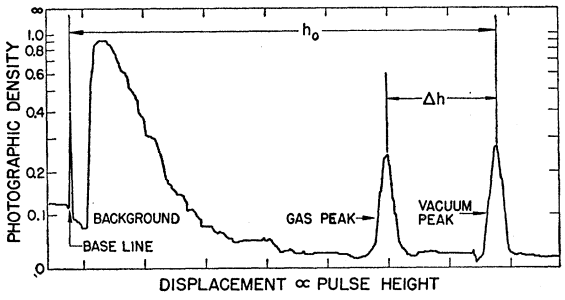

FIG. 2. Microdensitometer trace obtained by scanning the photographic negative of scintillator pulses appearing on the oscilloscope.

The centers of the pulse-height groups corresponding to the most probable pulse-height values were obtained in each case as the locations of the intersections of lines drawn along the sides of the peaks of the densitometer traces. For each gas there were at least two traces corresponding to runs at the same pressure, and it was observed that one could determine the fractional drop in pulse height with an average consistency of about 1.4 percent.

In the case of the deuteron experiment, the fractional decreases in pulse height $\Delta h/h_0$ were not precisely the same but differed by small amounts for different gases. The relative stopping powers were then proportional to $(1/p)\Delta h/h_0$, where p is the absolute pressure of gas in the absorption cell. Values of p, $\Delta h/h_0$, and the stopping power relative to air for the various gases are given in the third, fourth, and fifth columns of Table II.

The results of the proton measurements are given in the sixth, seventh, and eighth columns of Table II. Here the absolute energy loss ΔE was measured, rather than $\Delta h/h_0$. The stopping powers of H_2 and Kr relative to air are seen to agree with those obtained in the deuteron measurements; the value for H_2 is more accurate however, because multiple scattering decreased the intensity of the proton pulse-height group in the case of krypton to a greater extent and made its location somewhat more uncertain.

The ninth column gives the proton energy at which the measurement applies. In the cases of H_2 and air, this is the energy directly measured in the proton experiment, while in the other cases the energies are taken in the ratios of the mean pulse heights $h_0(1-\Delta h/2h_0)$ to that of air. An exception is the case of Kr where the energies found by the two methods were averaged to give the value listed. It can be shown[8] that the error in taking $E_0(1-\Delta E/2E_0)$ as the energy value at which $\Delta E/\Delta x$ is equal to dE/dx is negligible. It is seen that these mean energies are nearly the same for all of the gases, the small variations arising from the fact that

[8] Reference 1, p. 791.

TABLE II. Data and results of stopping cross-section measurements. No correction has been made for impurities in the tank gases.

Gas or element	Atomic number	Pressure, p (mm Hg, 25°C)	$\Delta h/h_0$	Stopping power rel. to air	Pressure, p (mm Hg, 25°C)	ΔE (Mev)	Stopping power rel. to air	Equiv. proton energy (Mev)	Absolute stopping cross section dE/dx (ev-cm²×10⁻¹⁵)
		Deuteron measurements			Proton measurements				
1	2	3	4	5	6	7	8	9	10
½ H_2	1	1175	0.251	0.185±0.003	585.4	1.163	0.186±0.003	4.44	0.338±0.007
He	2	1336	0.248	0.321±0.005				4.45	0.585±0.013
½ N_2	7	233.7	0.261	0.966±0.014				4.42	1.76 ±0.04
½ air		225.8	0.261	1.000	112.9	1.206	1.000	4.42	1.82 ±0.04
½ O_2	8	214.0	0.269	1.088±0.016				4.40	1.98 ±0.04
Ne	10	355.0	0.258	1.258±0.019				4.43	2.29 ±0.05
A	18	219.2	0.258	2.042±0.031				4.43	3.72 ±0.08
Kr	36	138.4	0.271	3.393±0.051	70.0	1.235	3.302±0.082	4.41	6.10 ±0.15
Xe	54	97.6	0.249	4.425±0.075				4.44	8.06 ±0.18
CH_4		285.4	0.263	1.598±0.024				4.43	2.91 ±0.06
CO_2		143.9	0.252	3.037±0.046				4.43	5.53 ±0.12

the pressures were not set for identical energy absorption in all cases.

The last column of Table II gives the absolute stopping cross sections. In the cases of H_2 and air, this is the value directly measured in the proton experiment. The values for the other cases are obtained by multiplying the relative stopping powers of column 5 by the absolute stopping cross section for air. The value for Kr is the average of those obtained by the two methods. None of the results of Table II have been corrected for impurities.

DISCUSSION OF RESULTS

In the absence of other experimental results on gases in the energy range investigated here, a direct comparison of our dE/dx values can be made only with calculated values. In Table III, we present a comparison of our results, corrected for impurities according to Table I, with the calculations of Aron, Hoffman, and Williams[4] and Walske.[9] The stopping cross section of carbon was obtained from those of CH_4, CO_2, H_2, and O_2 by assuming the applicability of Bragg's rule of additivity of stopping powers:

$$CH_4-2H_2=2.91-1.34=1.57,$$
$$CO_2-O_2=5.55-3.96=1.59.$$

The average value for C is 1.58.

In the calculations of UCRL-121,[4] the ionization potential I in the Bethe-Bloch formula was obtained from the Bloch relation, $I=kZ$, using $k=11.5$ ev. For elements in the atomic-number range from carbon to aluminum the K-shell correction, C_K, given by Livingston and Bethe[10] was used. For heavier elements K-, L-, and M-shell corrections were taken somewhat crudely into account by neglecting the stopping power of a shell when the ion energy fell below four times the "critical energy" $(M/m)A_{K,L,M}$. Here M denotes the ion mass, m the electron mass, and $A_{K,L,M}$ the excitation potential of the shell in question. For H and He the theoretical values of Williams[11] for I were used. In the calculations of UCRL-1325,[4] Aron computed values of I to fit the experimental data[3] of Bakker and Segrè and Mather and Segrè, where applicable, preferring the latter for those elements measured by both.

Walske has calculated the L-shell correction C_L, using hydrogen-like wave functions, analogous to his previously calculated K-shell correction C_K. He obtains the average ionization potential I from the measurements of Bakker and Segrè, using their values of the I/Z, normalized at aluminum to the value corresponding to $I=160$ ev. For hydrogen he uses the value $I=16.2$ ev based on Mano's experimental determination.[12]

In Table III, it is seen that the agreement with both sets of calculated values is generally good. An exception is Walske's value for Xe. He estimates however, that the M-shell correction in this case would decrease his stopping cross section by about four percent. This would bring it into agreement with the measured value to within experimental error.

Another theory with which to compare the present results is that of Lindhard and Scharff.[5] By plotting

TABLE III. Comparison of experimentally determined stopping cross sections with the calculated values of Aron *et al.*[a] and Walske.[b] The experimental values have been corrected for impurities in the tank gases.

Element	Experimental dE/dx (ev-cm²×10⁻¹⁵)	UCRL-121[a]	UCRL-1325[a]	Walske[b]
H	0.335±0.007	0.331	0.331	0.341
He	0.582±0.013	0.601	0.601	
C	1.58 ±0.03	1.62	1.59	1.53
N	1.76 ±0.04	1.95		
Air	1.82 ±0.04			
O	1.98 ±0.04	2.09		
Ne	2.29 ±0.05			2.36
A	3.72 ±0.08	3.89	3.94	3.81
Kr	6.10 ±0.15			6.03
Xe	8.06 ±0.18			8.57

[a] See reference 4.
[b] See reference 9.

[9] M. C. Walske, Phys. Rev. **88**, 1283 (1952); also private communication.
[10] M. S. Livingston and H. A. Bethe, Revs. Modern Phys. **9**, 265 (1937).
[11] E. J. Williams, Proc. Cambridge Phil. Soc. **33**, 179 (1937).
[12] Georges Mano, J. phys. radium **5**, 628 (1934).

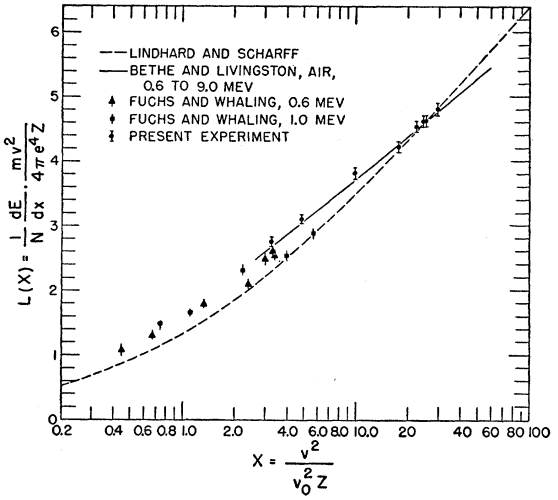

FIG. 3. Plot of stopping cross-section data for protons in gases using the functions $L(dE/dx)$ and $X(E)$. The dashed curve shows the function found by Lindhard and Scharff for protons in solids.

the quantity

$$L(X)=(4\pi e^4 Z/mv^2)^{-1}dE/dx$$

against the quantity

$$X=v^2/Zv_0^2$$

they found that a universal curve could be obtained which approximately brought all of the data for various atomic numbers and proton energies together in a single continuous function. Here dE/dx is the atomic stopping cross section, v the ion velocity, and $v_0=e^2/\hbar$.

In order to compare the curve of Lindhard and Scharff with the present data and other data on the stopping of gases, we have determined the values of X and $L(X)$ both for our results and for the results at proton energies 0.6 and 1.0 Mev for various gases as given by the curves in the compilation of Fuchs and Whaling.[1] These results are plotted in Fig. 3. The points corresponding to the present results run in order of increasing X from Xe to C and those corresponding to the compilation of Fuchs and Whaling from Xe to N. We have also replotted the calculated stopping cross-section curve for air of Livingston and Bethe[10] for proton energies between 0.6 and 9.0 Mev.

The experimental points for gases lie approximately on a continuous curve, with a few exceptions. However a curve fitting the gas data would clearly not coincide with that obtained by Lindhard and Scharff for the data on solids. It is interesting to note that the points corresponding to the present results lie approximately on the replotted air curve of Livingston and Bethe.

RESPONSE OF SODIUM IODIDE TO PROTONS AND DEUTERONS

The measurements of the Illinois group[6] of the response of NaI(Tl) to protons, H_2^+ ions, and deuterons showed that the response curves of light output *versus* ion energy are straight lines in the energy region from one to 11 Mev. In addition, it was shown that for alpha particles saturation and other effects result in a response curve having a knee at about 8 Mev convex downward but tending asymptotically to a straight line, so that for high energies one may consider the curve as a straight line having a negative intercept on the axis of ordinates. In the cases of the singly-charged ions the accuracy of the data (primarily the energy determinations) left some doubt whether or not the straight lines pass through the origin. The accuracy of the data of Likely and Franzen[7] on the response curve for protons from five to 18 Mev likewise leaves some doubt whether or not the high-energy response curve effectively has a small nonzero intercept, due perhaps to a saturation knee at low proton energies.

It was with the possibility of such nonlinearity in mind that the stopping-power measurements for deuterons were performed with approximately the same energy change ΔE in order that the relative stopping-power determinations would not be affected by crystal response. If one formally calculates the energy change in this part of the experiment as $E_0(\Delta h/h_0)$, one should arrive at absolute stopping cross sections which are too great by a factor $(1-\delta h/h_0)^{-1}$, where δh is the magnitude of a negative intercept of the response curve (see Fig. 4). Such calculations were made for the cases of H_2, air, and Kr and the results compared with the stopping cross sections measured absolutely for protons in the manner described earlier, where ΔE was measured directly, independent of crystal response. The resulting values of $\delta h/h_0$ [for deuterons in NaI(Tl)] were 8.2, 8.5, and 11.6 percent respectively, with a weighted average value of 8.5 ± 1.3 percent.

Having thus indirectly measured the amount of nonlinearity of response for deuterons for the crystal used in this experiment, it was interesting to investigate the response curve for protons also. This was done

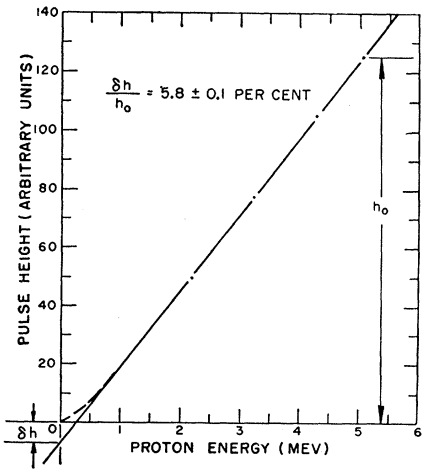

FIG. 4. Response curve of the NaI(Tl) scintillator pulse height (proportional to light output) *versus* proton energy.

directly by setting the van de Graaff energy at known values and measuring scintillator pulse height as a function of proton energy, correcting for 90-degree recoil, energy loss in the gold scatterer, and energy loss in the Dural entrance window. The results are shown in the graph of Fig. 4. It is seen that the proton response curve has an effective negative intercept, presumably due to a nonlinearity in the form of a knee as indicated by the dashed portion of the curve. The value of $\delta h/h_0$ for protons is 5.8 ± 0.1 percent, where h_0 is the pulse height corresponding to a proton energy of 5.03 Mev.

It may be noted that a nonscintillating layer in which energy is lost by the protons or deuterons in entering the crystal would cause the measured response curves to have effective negative intercepts. It should be mentioned that the surface of the crystal on which the ions impinged was cleaved in a dry box and consisted of a single cleavage plane. Transfer to the crystal housing was made in the dry box, and the crystal thereafter was kept sealed under vacuum or in contact with the dry gases used in the experiment. No observable clouding of the glass-like crystal surface occurred during the experiment. The large single crystal from which the disk used in this experiment was cleaved was obtained from the Harshaw Chemical Company and had a thallium activation of 0.2 ± 0.05 percent in content.

The following argument also indicates that the negative intercepts are not due to a nonscintillating surface layer: Since the protons and deuterons at the values of h_0 in question had equal velocities, their energy losses in the layer were the same. The deuterons had twice the energy, and hence approximately twice the pulse height, of the protons. Therefore, if the negative intercepts were due solely to surface energy loss, the quantity $\delta h/h_0$ should have had approximately one-half the value for deuterons that it had for protons. This is not the case since the value eight percent for deuteron is larger than the value six percent for protons.

In this connection the results of der Mateosion and Yuan[13] are of interest. They showed that for Po^{210} alpha particles the surface effect for chemically cleaned NaI crystals was at most five percent.

The present measurements are not inconsistent with those of Allison and Casson[14] who found that the response of NaI(Tl) to protons with energies between 50 and 400 kev is a straight line passing through the origin. A portion of the response curve like the one shown dashed in Fig. 4 would probably be indistinguishable from the straight line determined by their measurements.

We are pleased to acknowledge assistance received in this experiment from Dr. R. L. Henkel who operated the large van de Graaff and assisted in setting up the equipment at that installation; Miss Patricia Fain who performed the mass-spectrometric analysis of our gas samples; Mr. Berlyn Brixner who kindly furnished the plate camera; Mr. Grenfell Boicourt who constructed much of the equipment; and Captain Ralph Speece who instructed us in the use of the microdensitometer.

[13] E. der Mateosian and Luke C. L. Yuan, Phys. Rev. **90**, 868 (1953).
[14] S. K. Allison and H. Casson, Phys. Rev. **90**, 880 (1953).