TABLE II. Atoms whose $2Z_p$'s are given in the literature.

Atomic number	Element	Atomic number	Element
2	He	19	K^+, K^{++}
3	Li^+	20	Ca, Ca^{++}
4	Be, Be^{++}	26	Fe
9	F	29	Cu^+
10	Ne	37	Rb^+
11	Na	47	Ag^+
13	Al^+, Al^{+++}	55	Cs^+
14	Si^{++}	74	W
18	Ar	80	Hg, Hg^{++}

PHYSICAL REVIEW VOLUME 91, NUMBER 6 SEPTEMBER 15, 1953

Range-Energy Relations of 10- to 250-kev Protons and Helium Ions in Various Gases*

CHARLES J. COOK, EMERSON JONES, JR.,† AND THEODORE JORGENSEN, JR.
University of Nebraska, Lincoln, Nebraska

(Received May 5, 1953)

An analyzed beam of ions was brought through a differential pumping system into a low-pressure gas stopping cell, where the data necessary to plot a Bragg curve were obtained with the aid of a double grid ionization chamber of variable transverse radius. It was found that the resulting ionization extrapolated ranges may be uniquely determined only if the ionization chamber radius is greater than the transverse radius of the ionization envelope of the beam in the stopping gas. The ionization extrapolated range-energy relations for slow protons and alpha particles were determined in various gases. These data are also expressed in terms of an empirical relation. The observed range-energy relations are compared with previous data where they exist.

INTRODUCTION

THE range-energy relations of protons and alpha particles in various gases have been studied intensively in the past.[1] However, there has been a need for further measurements in this field at low energies.[2–5] The purpose of this investigation is to establish experimentally the range-energy relations for low-energy protons and helium ions in various gases. An ionization extrapolated range was determined by bringing a homogeneous beam of ions of known energy through a differential pumping system of negligible stopping power, into a low-pressure gas, where data for a Bragg curve were obtained by using a movable ionization chamber.

EXPERIMENTAL APPARATUS

The ion beam was produced by a Cockcroft-Walton accelerator, and after being magnetically analyzed, was brought to focus on the exit aperture of the differential pumping window leading into the stopping cell as shown in Fig. 1. The ion beam was accurately centered on this aperture by means of electrostatic deflection, which could move the beam both horizontally and vertically.

The differential pumping system consisted of three thin tungsten or tantalum diaphragms with a small aperture in each to pass the beam and to impede the flow of gas from the stopping cell; further details are shown in Fig. 2. Corrections to the ranges caused by the observed gas pressure in the intermediate regions of this window were calculated and were found to be less than 0.3 percent in all cases.

The energy of the ion beam was determined by measuring the potential of the anode of the high-frequency ion source, since the energy of the accelerated ions is known to be only about 100 volts less than this potential. The accelerating potential was measured by accurately determining the potential drop across a known fraction of a 1200-megohm voltmeter stack with a Leeds and Northrup K-2 potentiometer. This fraction was checked frequently and adjusted within 0.1 percent of the desired ratio so that temperature effects were compensated.

This arrangement was checked by means of the $Mg(p,\gamma)$ resonance at 222 kev.[6] A series of daily measurements of this resonance indicated that the voltmeter read 2 percent high with a 2 percent variation. These variations were random and not due to a possible film

* Research performed under contract with the U. S. Atomic Energy Commission.

† Now at the Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

[1] A. E. Taylor, Repts. Progr. Phys. **15**, 49 (1952).
[2] Cornog, Franzen, and Stephens, Phys. Rev. **74**, 1 (1948).
[3] H. A. Bethe, Revs. Modern Phys. **22**, 213 (1950).
[4] A. P. French and F. G. P. Seidl, Phil. Mag. **42**, 537 (1951).
[5] W. A. Wentzel and W. Whaling, Phys. Rev. **87**, 499 (1952).
[6] Hole, Holtsmark, and Tangen, Z. Physik **118**, 48 (1941).

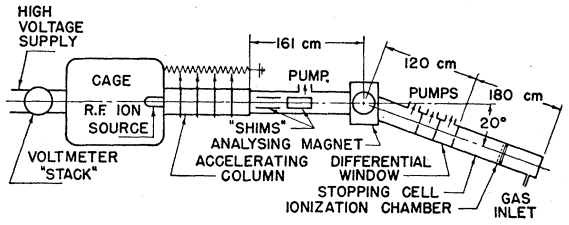

FIG. 1. Schematic diagram of the accelerator and experimental apparatus.

forming on the target. Therefore, the beam energy was considered to be known within 2 percent above 125 kev and 1 percent below. Variations in the accelerating potential detectable by the voltmeter were kept less than 0.015 percent of the desired potential from 20 to 250 kev by means of a galvanometer-phototube-amplifier system used to control a variable impedance transformer in series with the primary of the high voltage transformer.[7] The ripple of the accelerating potential was no greater than 0.15 percent for a well focused beam. The analyzing magnet current regulator maintained the magnet current within 0.2 percent of any selected value.

The gas stopping cell was a brass tube 6 inches in diameter and 200 cm long. The ionization chamber could be moved over most of the length of the stopping cell, and its position was measured to the nearest millimeter. Gas was let into the stopping cell through a manifold of five needle valves. The pressure of the stopping gas was measured with two McLeod gauges especially constructed for precise reading in the pressure range used. There were two traps, cooled with a dry ice-acetone mixture, in series between the gauges and the stopping cell. During the experiment the stopping gas flowed through the stopping cell at a nominal rate of one cc per minute, varying of course with the pressure and kind of gas. The secular equilibrium pressure could be maintained with only slight variation over long periods of time. The pressures were assumed known to ± 0.5 percent. Temperatures were determined to the nearest degree by a mercury thermometer in contact with the wall of the stopping chamber.

The ionization chamber consisted of a collecting plate and two grids, all of brass. The plate was divided into annular rings 1, 2, 3, 4, 5, and 6 cm outside radii.

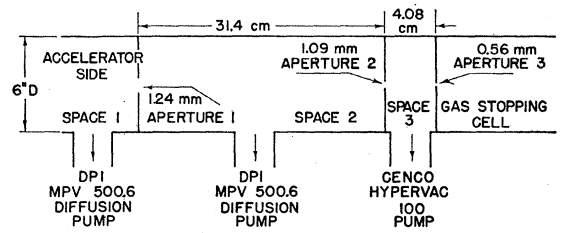

FIG. 2. Schematic diagram of the differential pumping system.

[7] L. N. Ridenour and C. W. Lampson, Rev. Sci. Instr. 8, 162 (1937).

The electrical connections to these rings were such that any one ring or any combination of them could be connected to a single galvanometer for measuring the ionization current, while the rings that were not used in this way served as guard rings. The largest ring always served as a guard ring. The collecting plate was kept at a positive potential, the middle grid was kept at a negative potential to repel electrons which would otherwise arrive at the plate from the region in front of the ion chamber, and the front grid was kept at ground potential. A careful study showed that the range measurements were independent of the ion chamber potentials over wide limits. The grid-grid and grid-plate spacings were 3.0 mm, and corrections for the depth of the chamber were made for all range determinations.

EXPERIMENTAL PROCEDURE

The ionization extrapolated range of an ion beam in a gas was determined by plotting a Bragg curve showing the relative ion chamber currents for various positions of the chamber along the stopping cell and extrapolating the straight portion of this curve to zero current. It is necessary, in order to obtain precise data for such a Bragg curve, that the beam current entering the stopping cell and the pressure of the stopping gas remain constant while the data is being taken. It was found that the major contributions to the instability of the beam were due to slight changes in the position of the focused spot caused by slight variations of the many parameters involved in producing a well-focused beam with this accelerator. These position changes need be very slight since the diameter of the focused beam was about the same as the diameter of the aperture of the window. Rather than attempt to stabilize the accelerator completely, the beam was swept vertically past the window aperture with 60-cycle modulation of large amplitude. The ion beam then passed the window at essentially a constant rate, and as a result, minor vertical changes in position of the beam that would ordinarily decrease the beam current into the stopping cell did not produce a detectable change of the average modulated beam current into the stopping cell. The horizontal changes in position of the focused spot were eliminated by slowly varying the magnet current in order to observe the maximum ionization current. Other factors influencing the average beam current into the stopping cell produced negligible variations. Careful investigations showed that ionization extrapolated ranges do not depend upon beam modulation even when the beam current into the stopping cell is reduced by a factor of 1000 by modulation.

To make sure that a given Bragg curve depended only on the ionizing power of the beam, the data were taken, once for successive positions of the chamber for increasing chamber-window distances, and then again for successive positions for decreasing chamber-window distances. If both curves were essentailly the same,

there could have been no pressure change, beam intensity changes, etc., and the curves were accepted. All other Bragg curves were rejected. The ionization extrapolated ranges can be determined within ± 0.5 percent from the original data.

While the apparatus was being developed so that Bragg curves could be obtained with suitable precision, preliminary experiments showed that the extrapolated ranges of protons of a known energy in a gas determined with an ionization chamber of fixed radius, when normalized to standard conditions of pressure and temperature, were not independent of the pressure. This effect is caused by the scattering of the beam by the stopping gas. At lower pressures, the ionization envelope of the beam in the gas is so large that the ionization current collected by the ionization chamber decreased rapidly with the increasing distance along the stopping cell, primarily because the beam is scattered outside the volume swept by the chamber and only partially because of a decrease in the ionizing power of the beam. A set of Bragg curves taken with various sized chambers showing this effect is given in Fig. 3.

If it is assumed that the size of the ionization envelope is inversely proportional to the pressure, it would appear that an increase in pressure should produce the same effect in the normalized range as a corresponding proportional increase in the radius of the ionization chamber. In other words, the normalized range for a given product of pressure and chamber radius Pr should be unique for a given stopping gas and particle energy. Normalized ranges for 60-kev protons in hydrogen are plotted in Fig. 4 against the values of Pr. This curve shows the range to be a function of Pr within experimental error. These curves were obtained for various gases at various energies, and all had similar shapes. They all showed the characteristic "saturation" effect at the higher Pr values. This saturation occurs when the pressure of the gas and the radius of the chamber are such that essentially all of the ionization envelope is contained in the volume swept by the ionization chamber.

To assure that saturation occurred in every range determination, and therefore that a unique range was obtained, it was not necessary to obtain a Pr curve for every energy. It was enough to show that the observed range is the same for ionization chambers of 4- and 5-cm radius. The procedure adopted was to observe the Bragg curves at as low a pressure as possible and yet have a saturated range. This procedure reduced the relative error inherent in observing the position of the ionization chamber as well as the correction due to the finite depth of the ionization chamber.

After the criterion for unique normalized ionization extrapolated ranges was established, it was possible to measure these ranges in gases with good precision. The errors in the high potential determination were reflected into errors in the normalized range amounting

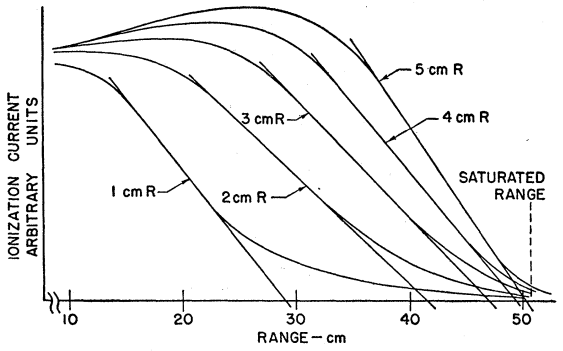

FIG. 3. Bragg curves for 50-kev protons in CO gas at 1.06-mm Hg for 1-, 2-, 3-, 4-, and 5-cm radius ionization chamber.

to 3 percent above 125 kev and 1.5 percent below. These errors, together with errors of ± 0.5 percent in the pressure and ± 0.3 percent in the temperature of the stopping gas, ± 0.5 percent in the measurement of the position of the ion chamber, and ± 0.5 percent in the extrapolation of Bragg curves are consistent with actual variations in the range determinations. The root-mean-square deviations for proton ranges were investigated for all stopping gases at various energies from 50 to 200 kev and were always found to be less than 1.4 percent.

Errors introduced into the range determinations by contaminants in the stopping cell were difficult to estimate. The stopping cell was pumped down to a low pressure, as shown on an ionization gauge, and was tested for rate of pressure rise each day before the stopping gas was let into the stopping cell. This was done to insure that no appreciable vapors from within the apparatus nor any air leaking into the stopping cell could contaminate the stopping gas. Gases were admitted to the stopping cell through two dry ice-acetone traps in series to remove water vapor. Hydrogen gas was further purified by removing a trace of oxygen catalytically. All gases were from commercial tanks and were stated to be 99.5 percent pure or better except for nitrous oxide which was stated to be 98 per-

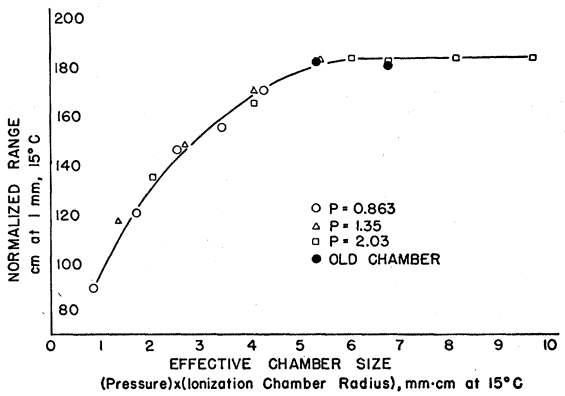

FIG. 4. Variation of the normalized range with effective chamber size for 60-kev protons in hydrogen gas.

TABLE I. Observed ionization extrapolated ranges for protons.

	Ranges—cm at 1-mm Hg, 15°C								
E kev	H_2	O_2	A	Air	N_2	CO	CH_4	N_2O	CO_2
4.00	29.4								
4.90	32.0								
6.66			15.6	12.5	10.3				
8.00						14.4			
10.0			20.9			17.5	18.3		
11.0									14.3
11.5				19.4					
12.0	57.9				19.7	19.4	20.4	14.5	
13.0		23.9							
15.0	68.4				22.8				18.0
16.0						24.4	24.4		
20.0		32.1	31.5	28.9	28.0	28.6	27.8	20.5	22.1
21.5	87.1								
25.0	97.0				32.2				
30.0	109	43.9	42.8	39.2	36.6	37.3	36.3	28.4	30.1
35.0					41.2				
37.5	129								
40.0		54.3	52.0	48.1	45.0	45.8	43.9	34.5	36.3
45.0	146								
46.0	147								
50.0		63.4	58.6	57.2	53.2	53.7	51.1	40.6	42.6
57.0	174								
60.0		71.5	67.2	64.3	62.0	62.3	58.2	46.0	48.2
70.0	203	80.3	73.5	71.9	69.3	71.3	65.0	50.8	53.9
80.0		88.1	82.8	80.0	76.9	78.5	72.3	56.8	59.9
85.0	237								
90.0		97.0	92.7	89.0	85.4	86.1	78.8	62.7	64.6
95.0	266								
100			102	96.3	94.2	95.1	84.8	68.4	70.0
110		114							
120	331								
125		125	124	115	114	115	104	81.0	85.0
140	388								
150		149	147	137	136	137	124	96.7	99.8
170	485								
175		169	172	159	159	158	146	111	114
200		192	200	182	182	182	170	126	131
202	594								
205							175		
225		217		209		207	195	144	
240					219				
241	739								
250		243		232		236	222	164	166
252			259						

cent pure with unknown impurities. Air was drawn from outside the laboratory and carbon dioxide was obtained from the solid form. Since the ranges of protons in all gases except hydrogen are about the same, small amounts of likely impurities would cause insignificant errors in the ranges. Since the ranges in hydrogen from four different tanks from different suppliers were experimentally identical, it was assumed that the nitrogen contaminant was negligible in each. Care was taken that no mercury vapors from the McLeod gauges entered the stopping cell except that which came past the two traps. No evidence could be found of possible mercury contamination.

EXPERIMENTAL RESULTS

Ionization extrapolated ranges of protons and helium ions in the various gases studied, normalized to 1-mm Hg and 15°C, are shown in Tables I and II.

The relation between the normalized range R and the incident energy E can be very closely approximated by empirical relations of the form

$$R=kE^n, \quad E<E_0; \quad R=C(E+E_1)^{1.5}, \quad E>E_0,$$

where k, C, E_1, and n are parameters chosen so that these relations fit the data, and E_0 is the energy for which the rate of energy loss is a maximum. The first equation is suggested by the linear portion of the curves when the data are plotted on log-log paper. The second equation is suggested by Geiger's relation[8] for high energies. The requirement that these two empirical relations must be continuous with their first derivatives at E_0 reduces the number of independent parameters, including E_0, to three. The empirical relations fit the data to within 2 percent except near E_0. Values of the parameters are shown in Tables III and IV.

DISCUSSION OF RESULTS

Since, at the present time, there are no existing theoretical range-energy relations for ions below the energy for which Bethe's equation[9] is valid, here called slow ions, it is only possible to compare the range-energy relations found in this experiment with similar range-energy relations found in previous experiments.

Bethe[3] and Jesse and Sadauskis[10] have constructed the most precise range-energy relation for alpha particles in air previously available in this energy range. It was based on the experiment of Holloway and

TABLE II. Observed ionization extrapolated ranges for alpha-particles in various gases.

	Ranges—cm at 1-mm Hg, 15°C		
E kev	A	Air	H_2
20.0	35.2		125
30.0	45.2	31.5	168
40.0	54.3	40.4	203
50.0	60.7	46.9	234
60.0	68.7		251
70.0	74.3		276
70.5		60.0	
80.0	81.4		302
90.0	86.8	71.0	324
100	91.4	76.1	342
125	105	89.0	391
150	118	101	433
175	129	114	467
200	141	124	506
225	151	131	534
250	161	143	585

[8] H. Geiger, Proc. Roy. Soc. (London) **83**, 492 (1910).
[9] M. S. Livingston and H. A. Bethe, Revs. Modern Phys. **9**, 261 (1937).
[10] W. P. Jesse and J. Sadauskis, Phys. Rev. **78**, 1 (1950).

Livingston[11] in which the specific ionization of a single alpha particle in the air was determined and the mean range of a beam of alpha particles in air was then calculated using W(air), the energy lost by the incident alpha particle per ion pair produced in air. Jesse and Sadauskis found that above about 300 kev, W(air) was not a constant as was assumed for all energies by Holloway and Livingston. They then recalculated the range-energy relation from the specific ionization as observed by Holloway and Livingston for energies greater than 271 kev. However, the work of Jesse and Sadauskis may be extrapolated to lower energies by extending the linear relation that exists from 300 kev to 2 Mev for the ratio of W(argon) to W(air).[10] The range-energy relation that is calculated from the specific ionization curve as found by Holloway and Livingston, using this low energy extrapolation, is shown in Fig. 5 and is labeled the Jesse and Sadauskis (1950) extrapolated range-energy relation.

The range-energy relation for alpha particles in air found in this investigation [the University of Nebraska (U. of N.) 1953 relation)] is also shown in Fig. 5. The Bethe (1950) relation for alpha particles in air was also determined by extrapolating the work of Jesse and Sadauskis and thereby correcting the ranges calculated by Holloway and Livingston. This curve is essentially the same as the U. of N. relation. It is apparent that the agreement between the relations presented so far depend upon fortuitous extrapolations of the work of Jesse and Sadauskis.

The Bethe[9] (1937) range-energy relation for alpha particles in air is also given in Fig. 5. This relation was

TABLE III. Empirical range-energy relation constants for protons in various gases.

Gas	K cm/(kev)^n	C cm/(kev)$^{\frac{1}{2}}$	n	E_0 kev	E_1 kev
Hydrogen	11.2	0.147	0.67	42.5	53
Oxygen	3.90	0.0400	0.71	79.0	88
Argon	4.39	0.0440	0.67	58.4	73
Air	3.26	0.0393	0.73	76,5	80
Nitrogen	3.34	0.0414	0.71	62.9	70
Carbon monoxide	3.35	0.0376	0.71	77.3	86
Methane	3.80	0.0385	0.67	56	70
Nitrous oxide	2.50	0.0251	0.71	73	92
Carbon dioxide	2.64	0.0259	0.71	85	95

TABLE IV. Empirical range-energy relation constants for alpha particles in various gases.

Gas	n	K cm/(kev)^n
Hydrogen	0.558	25.2
Argon	0.596	5.95
Air	0.687	3.23

[11] M. G. Holloway and M. S. Livingston, Phys. Rev. **54**, 18 18 (1938).

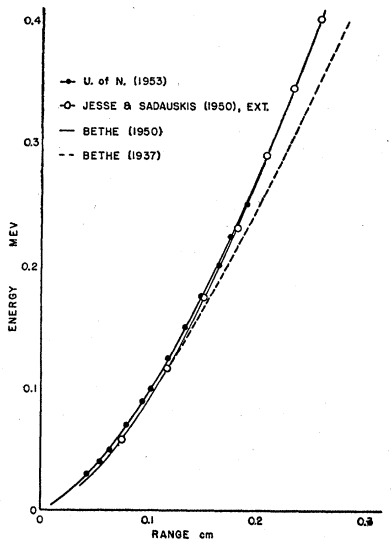

FIG. 5. Range of alpha particles in air at 760-mm Hg and 15°C.

derived from cloud chamber work by Blackett and Lees[12] and is admittedly quite inaccurate.[3,10]

The range-energy relation for protons in air has been previously derived from the range-energy relation for alpha particles in air, at 760-mm Hg, 15°C, by the semi-empirical expression[9]

$$R_H(E)=1.0072R_\alpha(3.971E)-0.20,$$

where $R_H(E)$ is the range in cm of a proton of incident energy E, $R_\alpha(3.971E)$ is the range in cm of an alpha particle of incident energy $3.971E$, and 0.20 = Blackett's empirical constant. This relation is valid in the energy region where Bethe's expression for the rate of energy loss of the ion is valid, that is, for proton energies above 300 kev and alpha-particle energies above 1200 kev. Below this region Blackett's constant becomes a decreasing function of energy.

Jesse and Sadauskis[10] used the above expression to calculate the range-energy relation of protons in air down to an energy of 302 kev (calculations from their table of alpha-particle ranges give 0.40-cm range at 306 kev for protons in air). Bethe[3] used this expression down to a proton energy of only 500 kev. From 0 to 170 kev Bethe calculated the ranges from the rate of energy loss of protons in air as determined by Crenshaw.[13] The region from 170 to 500 kev was fitted by interpolation. Bethe's 1950 relation, as well as Jesse and Sadauskis' range at 306 kev, are shown in Fig. 6. The Bethe 1937 alpha-particle ranges, which are based on the work of Blackett and Lees, appear to be too long, and it would seem that the proton ranges should thus also be too long. In fact, the Bethe ranges at 306 kev are nearly 20 percent longer than that calculated at this energy by Jesse and Sadauskis.

[12] P. M. S. Blackett and D. S. Lees, Proc. Roy. Soc. (London) **134**, 658 (1932).

[13] C. M. Crenshaw, Phys. Rev. **62**, 54 (1942).

The U. of N. ranges of protons in air are also shown in Fig. 6. The U. of N. range at 306 kev found by means of the empirical relation is 0.393 compared to 0.40 cm calculated by Jesse and Sadauskis. These two ranges differ by less than the experimental error and indicate that Blackett's constant is probably not energy-dependent for energies greater than 300 kev.

The U. of N. and the Brookhaven[14] range-energy relations for protons in hydrogen are shown in Fig. 7. The range-energy relation published by the Brookhaven National Laboratory was obtained from the theoretical stopping power of protons in hydrogen as calculated by Hirschfelder and Magee.[15] The range-energy relations determined by Crenshaw[13] and Phillips[16] agree with the U. of N. range-energy relation within experimental error. The agreement found between these various range-energy relations for protons in hydrogen is most gratifying.

In the comparison of present results for the other gases with previous work it is necessary either to integrate the rates of energy loss data to obtain ranges or to differentiate the present data to obtain rates of energy loss. Satisfactory comparisons have not been obtained by either procedure.

Probably the best way of obtaining rates of energy loss from the present data is to differentiate empirical

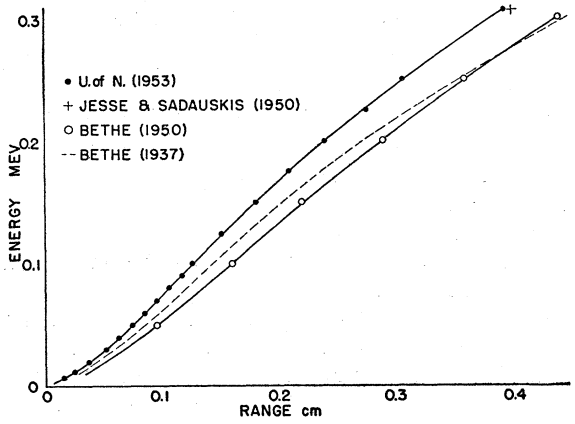

FIG. 6. Range of protons in air at 760 mm Hg and 15°C.

[14] H. Bethe, Brookhaven National Laboratory Report BNL-T-7, 1949 (unpublished).
[15] J. O. Hirschfelder and J. L. Magee, Phys. Rev. **73**, 207 (1948).
[16] James A. Phillips, Phys. Rev. **90**, 532 (1953).

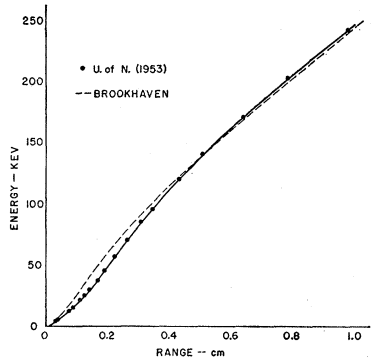

FIG. 7. Range of protons in hydrogen gas at 760 mm Hg and 15°C.

relations suitable for this purpose. The empirical relations previously given have discontinuous second derivatives at E_0. This, of course, creates errors in the rate of energy loss curves obtained from the present data. Mechanical differentiation likewise is subject to errors. Nevertheless, all attempts to find rates of energy loss from the present data give values that are greater than the results given by the differential type experiment. For protons in argon, the present experiment gives rates of energy loss of the order of 20 percent greater than the Los Alamos results.[16] The difference appears to be too large to be accounted for by experimental errors or errors in differentiation.

Although this difference in the rates of energy loss could be caused by a large systematic error, it should probably be attributed to the fundamental difference of the two experiments. The differential type experiment excludes from consideration any particle which has suffered an appreciable angular scattering, while the present integral type of experiment gives a unique result only when essentially all the ions are collected.

ACKNOWLEDGMENTS

We wish to express our appreciation to Mr. J. S. Heiser for his expert assistance in the construction of the accelerator and other apparatus and to Mr. Arthur J. Meyerott and Mr. William A. Barrett, Jr. for their technical assistance. We wish to express our gratitude to the Board of Regents of the University of Nebraska for a special grant which enabled us to start this work and to the U. S. Atomic Energy Commission for its continued support.