Stopping of Energetic Copper Ions in Aluminum*

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By means of differential-range experiments, we have measured the average ranges and range straggling of 24-min Cu^{60} and 3.3-h Cu^{61} in Al. These species were produced with initial recoil energies from 3 to 18 MeV by bombardment of thin targets of Co^{59} and Mn^{55} with Li^6, B^{10}, and B^{11} heavy-ion beams. The average ranges increase smoothly with recoil energy and the distributions in range about the average values follow a Gaussian relationship. The experimental range-energy data are in good agreement with theoretical expectations, assuming total momentum transfer over the full energy region investigated. We conclude that the nuclear processes which produced the recoiling ions are pure compound-nucleus reactions. Although theory and experiment yield consistent average-range values, the stopping-theory predictions for range straggling are much smaller than experimental estimates of the straggling inherent in the stopping process. The source of the discrepancy is discussed in terms of the relative contributions of electronic and nuclear stopping.

I. INTRODUCTION

THE use of recoil techniques has been of great value in the study of nuclear reactions.[1] The recoil properties of residual nuclei are intimately related to the kinematics of specific nuclear processes, and may, in principle, yield quantitative, detailed information about these processes. To derive maximum benefit from such studies, however, one must have a thorough understanding of the relationships between the observed stopping phenomena and the energies and momenta of recoiling species. In recent years the available experimental data have increased greatly, and parallel theoretical developments have come a long way towards elucidating the stopping of heavy charged particles in matter.

Of particular note are the recent theoretical investigations of Lindhard *et al.*,[2,3] which have a direct application to the interpretation of recoil data. These authors consider electronic stopping (ionization) and nuclear stopping (ion-atom interaction) to be uncorrelated and continuous processes, and based upon particular models for relative contributions of each, have derived general range-energy relationships and distributions in ranges. It is important to compare the theoretical predictions with experiment over a wide energy region and for various moving and stopping species to determine the detailed validity of the theory and where modifications might be needed. Such comparisons have been given by Lindhard *et al.*,[3] and have recently been reported for 2 to 10 MeV Sm in Al,[4] 6 to 21 MeV Dy in gases and Al,[5] 0.07 to 1.0 MeV Ga in Cu and Zn,[6] and 0.5 to 4.5 MeV Co and Ni in Fe.[7] In general, the theoretical predictions of average ranges are in good agreement with experiment, but the range straggling, where measured,[4,5] indicates considerable discrepancy with the theory. Measurements of straggling in aluminum oxide at lower energies (10–150 keV) gives better agreement with predictions.[8]

In this paper we report measurements of average ranges and range straggling in Al of 3 to 18 MeV Cu^{60} and Cu^{61} ions (50 to 300 keV/amu). This corresponds to a velocity region where relatively little data are available, other than fission fragment ranges, and where the theoretical predictions are very sensitive to contributions from electronic stopping. The Cu atoms were produced in nuclear reactions induced by heavy-ion beams, and recoiled out of thin targets into stacks of Al catcher foils. The reactions were of the type (HI,pxn), (HI,xn) between Li^6, B^{10}, and B^{11} projectiles and Co^{59} and Mn^{55} targets. Assuming full linear-momentum transfer in the reactions, the product recoil-velocity is determined primarily by the incident-beam momentum. The emission of particles from the highly excited compound systems introduces small perturbations, but we have been able to estimate these effects experimentally.

II. EXPERIMENTAL PROCEDURE

Targets were prepared by vacuum evaporation of Co (99.9% purity) and Mn (99.5% purity) metals onto 0.00025-in. aluminum backings. The targets were always thin in comparison to the recoil ranges of the Cu products, and the actual thickness of each target was determined from the known area and weight of deposit. Uniformity of the evaporated layers was estimated as better than 5% by comparison of the thicknesses of different targets prepared in the same evaporation.

The catcher foils were cut from commercial aluminum leaf 120–200 $\mu g/cm^2$ thick using a special punch of accurately known area. Each punched foil was examined for pinholes and nonuniformities, and its thickness determined from its weight and area.

A typical experiment consisted of assembling a target and 20 catcher foils in a water-cooled holder, and

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[1] B. G. Harvey, Ann. Rev. Nucl. Sci. **10**, 235 (1960).
[2] J. Lindhard and M. Scharff, Phys. Rev. **124**, 128 (1961).
[3] J. Lindhard, M. Scharff, and H. E. Schiott, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **33**, No. 14 (1963).
[4] M. Kaplan and R. D. Fink, Phys. Rev. **134**, B30 (1964).
[5] J. Gilat and J. M. Alexander, Phys. Rev. **136**, B1298 (1964).
[6] N. T. Porile, Phys. Rev. **135**, A1115 (1964).
[7] J. P. Hazan and M. Blann, Phys. Rev. **137**, B1202 (1965).
[8] B. Domeij, F. Brown, J. A. Davies, and M. McCargo, Can. J. Phys. **42**, 1624 (1964).

irradiating the stack with a collimated beam from the Yale heavy-ion linear accelerator. Beams with energies less than 10.5 MeV/amu were obtained by inserting aluminum foils of known thickness in the beam path, and magnetically analyzing the degraded beams upstream from the target assembly. Bombarding energies were determined from the range-energy data of Northcliffe,[9] in conjunction with the calibrated magnet settings.

Following bombardment, the catcher foils were separated and copper was isolated by radiochemical analysis. The foils were dissolved in HCl, Cu^{++} and Fe^{3+} carriers were added, and $Fe(OH)_3$ was precipitated by an excess of ammonium hydroxide. Copper remained in solution as an ammonia complex, and was separated from the scavenging precipitate by centrifugation. Samples were prepared for counting by reduction to metallic copper with ammonium hypophosphite, or by precipitating as cuprous cyanide following reduction with potassium sulfite. The duration of chemical separation ensured complete decay of any Zn precursors of Cu^{60} and Cu^{61}, but was sufficiently short to prevent substantial decay of 9-h Zn^{62} into 10-min Cu^{62}.

The purified samples were assayed for β radioactivity on a series of end-window methane-flow proportional counters. The counters were intercalibrated with a thick uranium source and their discriminator levels adjusted to yield equal counting efficiencies within one percent.

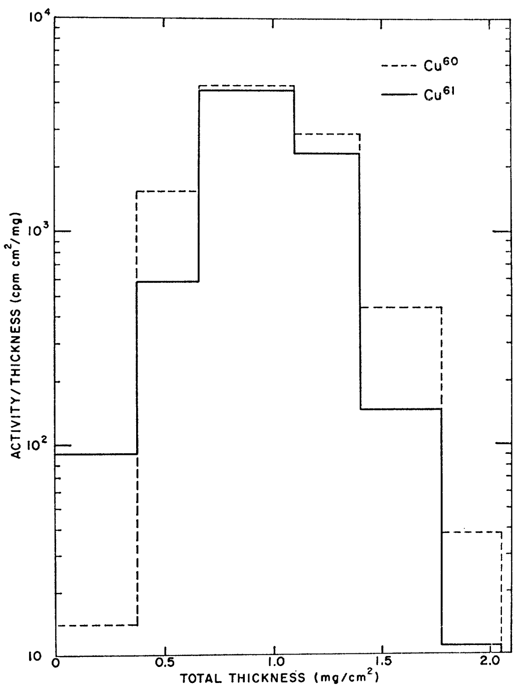

FIG. 1. Histogram of a typical differential-range experiment, showing the distribution of recoil Cu^{60} (dashed line) and Cu^{61} (solid line) activity in the catcher foils. The data are for the reaction of 62.7-MeV B^{11} with a Mn^{55} target (cpm = counts/min).

[9] L. C. Northcliffe, Phys. Rev. 120, 1744 (1960).

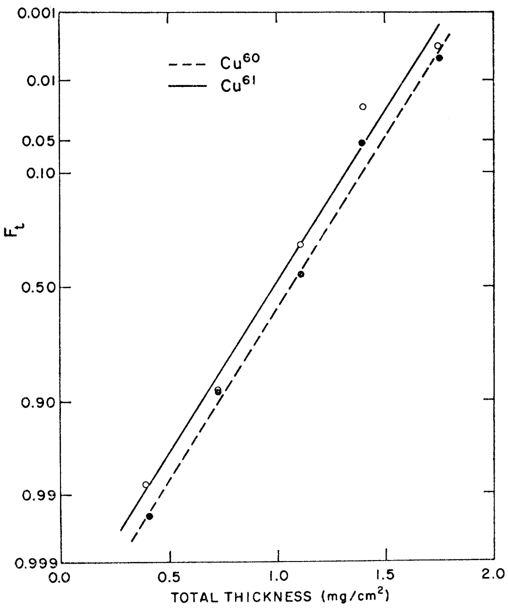

FIG. 2. Probability plot of the same data shown in Fig. 1. F_t, the fraction of the total recoil activity which passes through a catcher thickness t, is plotted on a probability scale against the total catcher thickness. In this representation, a straight line is indicative of a Gaussian distribution.

Intercalibration with Cu^{60}–Cu^{61} sources showed no difference in relative counter efficiencies. The samples were counted for at least 7 h and the decay curves resolved into 24-min Cu^{60} and 3.3-h Cu^{61} components. No evidence was found for contamination by 10-min Cu^{62}. The counting data were corrected for counter background, chemical yield of the sample (obtained by iodimetric determination of copper after counting), and activation of the catcher foils. The foil activation was determined from the activity observed in foils at the end of the stack (beyond the maximum range of the recoils) or by measurements in blank experiments. This correction amounted to about 10% of the peak activity.

III. RESULTS

The corrected relative activities of a series of catcher foils gives directly the range distribution of recoil Cu ions. Figure 1 shows a pair of histograms (for Cu^{60} and Cu^{61}) obtained in a typical differential-range experiment. This particular case is for the reaction of 62.7-MeV B^{11} with a Mn^{55} target. We have plotted the activity of each species in a catcher foil, divided by the foil thickness, as a function of the total thickness of (or penetration depth into) the foil stack. Small corrections for finite target thickness have been applied to all of our differential-range data by adding one-half the target thickness (converted to aluminum equivalent) to the total catcher thickness. We can determine that the distribution of activity for each species follows a Gaussian relationship by plotting on a probability scale the quantity F_t, defined as the fraction of the total

TABLE I. Results of differential-range experiments for Cu^{60} and Cu^{61} stopping in aluminum.

Target and beam	Bombarding energy, E_b (MeV)	Recoil energy, E_R [a] (MeV) Cu^{60}	Cu^{61}	Target thickness, W (μg/cm²)	Average range, R_0 (mg/cm²) Cu^{60}	Cu^{61}	Straggling parameter, ρ Cu^{60}	Cu^{61}
$Co^{59}+Li^6$	39.3	3.35	3.40	17	0.470	0.465	0.322	0.319
	62.5	5.33	5.41	17	0.585	0.613	0.362	0.336
$Mn^{55}+B^{10}$	74.4	10.55	10.75	95	0.985	0.980	0.312	0.318
$Mn^{55}+B^{11}$	39.4	5.96	6.07	95	0.660	0.630	0.343	0.470
	49.1	7.44	7.56	95	0.840	0.820	0.290	0.505
	54.3	8.22	8.36	95	0.930	0.905	0.304	0.374
	59.3	8.98	9.14	95	1.110	1.140	0.268	0.279
	62.7	9.50	9.66	97	1.070	1.030	0.280	0.257
	71.2	10.79	10.98	97	1.105	1.105	0.240	0.214
	85.1	12.90	13.12	97	1.025	1.245	0.314	0.290
	100.1	15.17	15.43	97	1.280	1.260	0.276	0.234
	114.1	17.30	17.60	47	1.330	1.290	0.272	0.316

[a] Calculated from Eq. (2).

activity of the species which passes through a total catcher thickness t, against the total thickness.[10] In these coordinates, a Gaussian distribution will yield a straight line. Figure 2 shows such a probability plot for the same data as in Fig. 1. Thus we may represent the distributions of Cu^{60} and Cu^{61} in the aluminum catcher foils by an equation of the form

$$P(R)dR=\frac{1}{R_0\rho(2\pi)^{1/2}}\exp\left[-\left(\frac{R-R_0}{(2)^{1/2}R_0\rho}\right)^2\right]dR, \quad (1)$$

where R_0 is the average range and ρ is the straggling parameter. The quantity ρ is a measure of the distribution in ranges about the average value. On a probability plot, as in Fig. 2, the catcher thickness at which $F_t=0.5$ is the average range, and the slope of the line gives the straggling parameter.

Table I summarizes our differential range measurements. The first two columns give the irradiation conditions: target and beam, and bombarding energy, respectively. Columns 3 and 4 list the recoil energies of Cu^{60} and Cu^{61}, calculated on the assumption that the velocity of the recoiling atom is equal to the center-of-mass velocity. This implies that the nuclear reaction proceeds by complete transfer of linear momentum from the incident beam and particle evaporation is symmetric about 90 deg in the center-of-mass system. Under these conditions, the recoil energy is given by[11]

$$E_R=A_RA_bE_b/(A_b+A_T)^2, \quad (2)$$

where kinetic energy and mass are denoted by E and A with subscripts R for the recoil, b for the projectile, and T for the target. Column 5 gives the target thickness, and columns 6–9 list the average ranges and straggling parameters for Cu^{60} and Cu^{61} in aluminum. In every case, the experimental range distribution could be characterized by a Gaussian function, and the average ranges and straggling parameters were obtained from probability plots.

Figure 3 represents the measured average ranges of Cu^{60} and Cu^{61} as a function of recoil energy calculated from Eq. (2). The solid line is the theoretical prediction based on the work of Lindhard, Scharff, and Schiott.[3] For purposes of comparison, we have converted the calculated relationship (which represents a total path-length) to ranges projected along the beam direction (the experimentally measured quantity) in the manner suggested in Ref. 3. (The correction varied from 6% at the lowest energies to 3% at the highest energies.) In Fig. 3 we have assumed that the average range is a function only of the center-of-mass velocity. If range is proportional to momentum, then the effects of particle evaporation will not alter the average range.[10] However, for higher power dependences on velocity, the additional velocity imparted to the recoils by particle emission will tend to increase the average range. This point has been investigated as follows. The data in Fig. 3 are consistent with a power dependence of 1.4 upon velocity. Combining this value with independent data on the energetics of nuclear evaporation, obtained from angular distribution experiments for the same reactions,[12] we may estimate[10,13] that the increase in average range is of the order of 1%, and may, therefore, be neglected.

With regard to the assumed applicability of Eq. (2), we believe that the internal consistency of our range-energy data over a wide energy-region, coupled with Gaussian range distributions and the agreement with stopping-theory calculations, provide strong evidence for a compound-nucleus reaction mechanism. We conclude that the observed Cu^{60} and Cu^{61} were formed in such processes.

IV. DISCUSSION

The stopping theory of Lindhard, Scharff, and Schiott[3] (LSS) is based upon a Thomas-Fermi (statistical) model for the ion-atom interaction (nuclear

[10] L. Winsberg and J. M. Alexander, Phys. Rev. 121, 518 (1961).
[11] J. M. Alexander and D. H. Sisson, Phys. Rev. 128, 2288 (1962).
[12] M. Kaplan and V. Subrahmanyam (unpublished results).
[13] G. N. Simonoff and J. M. Alexander, Phys. Rev. 133, B104 (1964).

stopping), and assumes electronic stopping (ionization) to be proportional to velocity. For an ion of mass M_R and nuclear charge Z_R, moving in a stopping medium of atomic mass M_s and nuclear charge Z_s, the kinetic energy E_R and corresponding true range (total path-length) R are expressed as dimensionless variables ε and ρ_L given by

$$\varepsilon = E_R a M_s / Z_R Z_s e^2 (M_R + M_s), \tag{3}$$

$$\rho_L = RNM_s 4\pi a^2 M_R / (M_R + M_s)^2, \tag{4}$$

where

$$a = 0.8853 (\hbar^2/me^2)(Z_R^{2/3} + Z_s^{2/3})^{-1/2} \tag{5}$$

is a Thomas-Fermi screening length, m and e are the mass and charge of an electron, respectively, N is the atomic density of the stopping medium, and $\hbar$ is Planck's constant. Taking the energy transfer to the nuclei (atoms) and electrons of the stopping medium as being continuous and uncorrelated processes, the total stopping power may be written as the sum of nuclear and electronic contributions:

$$\left(\frac{d\varepsilon}{d\rho_L}\right) = \left(\frac{d\varepsilon}{d\rho_L}\right)_n + \left(\frac{d\varepsilon}{d\rho_L}\right)_e. \tag{6}$$

Approximating the electronic stopping by

$$(d\varepsilon/d\rho_L)_e = k\varepsilon^{1/2}, \tag{7}$$

LSS obtain from Eqs. (6) and (7) the range-energy relationship

$$\rho_L(\varepsilon) = \tfrac{1}{2}k\varepsilon^{1/2} - \Delta(k,\varepsilon), \tag{8}$$

where $\Delta(k,\varepsilon)$ is the correction to the electronic range due to the effects of nuclear stopping. The single parameter k, which appears in Eqs. (7) and (8), characterizes the particular projectile-stopper combination, and thus the derived relationships are expected to be general. LSS give as an approximate expression for this parameter:

$$k = \xi\left[\frac{0.0793 Z_R^{1/2} Z_s^{1/2} (M_R + M_s)^{3/2}}{(Z_R^{2/3} + Z_s^{2/3})^{3/4} M_R^{3/2} M_s^{1/2}}\right]; \quad \xi \approx Z_R^{1/6}. \tag{9}$$

The theoretical estimates of $\Delta(k,\varepsilon)$ as a function of ε are given by LSS for various values of k.

Based upon Eq. (8), we have derived the predicted range-energy curve shown as the solid line in Fig. 3. [For our experiments Eq. (9) gives $k=0.12$.] The agreement with the measured ranges is good. (In LSS terminology, our data cover the energy region $\varepsilon = 23$ to 120.) A better fit to the data would be obtained by using a somewhat larger value of k at the higher energies, but the experimental uncertainties in the data do not justify seeking relatively small deviations. Similar extents of agreement (at somewhat lower energy) have been reported for Tb and Dy ions stopped in Al and gases[5] and for Sm stopped in Al.[4] These latter works were able to attain greater experimental accuracy,

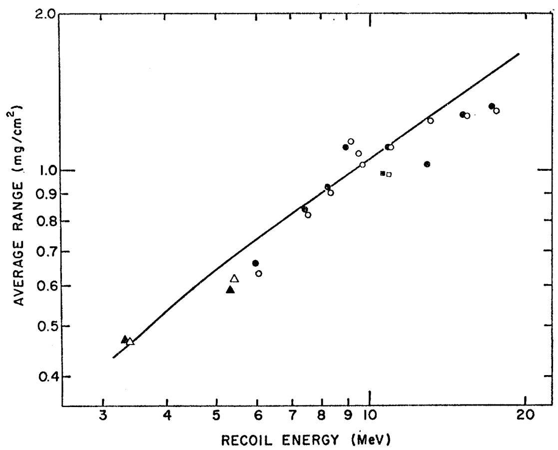

FIG. 3. Range-energy data for Cu^{60} and Cu^{61} stopping in aluminum. The filled points are for Cu^{60} and open points for Cu^{61} produced in the following reactions: circles, $B^{11}+Mn^{55}$; squares, $B^{10}+Mn^{55}$; triangles, Li^6+Co^{59}. The solid line is a theoretical prediction based on the work in Ref. 3.

and, as pointed out by Gilat and Alexander,[5] a small but real discrepancy exists which manifests itself as an apparent dependence of the parameter k on energy.

The range straggling is a more critical test of the details of the theory, because of its sensitive dependence on the ratio of nuclear to electronic stopping. The straggling parameters presented in Table I contain contributions from ρ_s, the straggling inherent in the stopping process; ρ_n, the distribution of velocities due to nuclear evaporation, ρ_w, the effect of finite target thickness, and ρ_f, inhomogeneities in the catcher foils. These components combine approximately as the squares:

$$\rho^2 = \rho_s^2 + \rho_n^2 + \rho_w^2 + \rho_f^2. \tag{10}$$

In order to extract ρ_s^2 for comparison with stopping theory, we estimate the effects of the other contributions as follows. The target thicknesses used were very thin compared to the average range values, and hence the effect of ρ_w is small and can be subtracted accurately.[10] The straggling due to nuclear evaporation processes, ρ_n, arises from the velocity and angular distributions imparted to the recoil ions by particle emission. Simonoff and Alexander[13] have derived expressions for ρ_n^2 in terms of range and angular-distribution variables. We have been able to estimate ρ_n^2 from angular distribution experiments[12] on Cu^{61} recoils from the $Mn^{55}+B^{11}$ reaction, assuming an isotropic distribution of center-of-mass recoil velocity vectors. We have no direct measure of microscopic inhomogeneities in the catcher foils, but comparison experiments using thick and thin Al foils provide some evidence that ρ_f^2 is relatively small.[11] Also, the discrepancies between theory and measurement for straggling of Dy ions in Al are comparable to the discrepancies observed using gas stoppers,[5] further indicating that ρ_f^2 is not large. On the other hand,

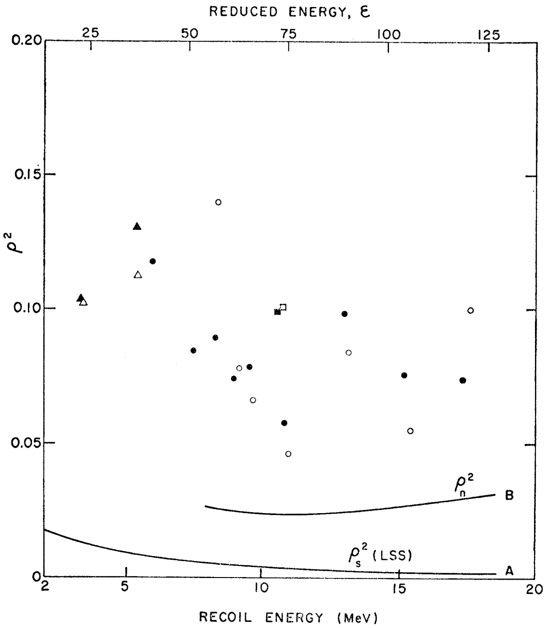

FIG. 4. Squares of experimental straggling parameters plotted against energy. The symbols are as in Fig. 3. Curve A is a theoretical prediction based on Ref. 3 of straggling inherent in the stopping process. Curve B represents the effect of an initial velocity distribution resulting from particle evaporation in the nuclear reaction. The sum of these two curves is to be compared with the experimental data.

work with Al_2O_3 films suggests that ρ_f^2 in Al may not be negligible.[14] In view of the inconclusive evidence, we assume that the straggling due to foil inhomogeneities is not significant, but are aware that this may require modification.

In Fig. 4 we have plotted the squares of the observed straggling parameters, corrected for target thickness, as a function of energy. The appreciable scatter of the data points is greatly amplified in taking the squares, but for the simple argument to be presented, the outlines of a general region and trend are adequate. Curve A, labeled ρ_s^2 (LSS), is the predicted relative-square-straggling (equivalent to our straggling parameter squared) inherent in the stopping process, as calculated from the LSS theory using $k=0.12$. Curve B, labeled ρ_n^2, is the contribution to straggling from nuclear evaporation, obtained as described above. We would expect the sum of these two curves to approximate the experimental data, but as can be seen, the theoretical estimate of ρ_s^2 is very small and leads to a rather large discrepancy with the observations. The theory considers the range straggling to arise primarily from nuclear stopping, with any contribution from electronic stopping being neglected. The computed values of relative square straggling begin to decrease very rapidly with energy for ε about 5–10, and in the energy region ε greater than about 50, the various relationships for different k values have all converged to quite small stragglings. It is this predicted sharp drop with energy (as compared to the straggling for pure Thomas-Fermi, or nuclear stopping) that produces the divergence from experiment, and small adjustments in the parameter k have little effect at the high energies. Disagreement between theoretical and experimental ρ_s^2 has been observed previously by Kaplan and Fink[4] and by Gilat and Alexander.[5] In those cases, however, the discrepancies were less (being about a factor of 2), which may be related to the lower energy regions covered where the predicted values have not yet fallen as far towards their very low values.

If the entire stoppin process (the full range) were due to nuclear stopping, then the predicted range straggling would be given by the Thomas-Fermi value. On the other hand, if all the stopping arose from electronic interactions, the predicted straggling would be zero (within the assumed framework). For contributions from both of these processes, the straggling must be somewhere between the two limits, and depends strongly on the admixture. It would seem that the disagreement between the observed and calculated effect implies (barring severe experimental difficulties) either an underestimation of the relative contribution of nuclear stopping or a substantial straggling contribution from electronic stopping, or both. Another indication of possible difficulty stemming from the same source is in the theoretical conversion from true ranges (total path-length) to projected ranges. This correction is also a function of the mixture of electronic and nuclear stopping (although much less sensitively than the straggling), and seems to fall off somewhat more rapidly with energy than the data would indicate. The experimental range and straggling data of Gilat and Alexander[5] suggest departures from the approximation of proportionality of electronic stopping to velocity, Eq. (7), which would, of course, have a significant effect on the relative contributions of nuclear and electronic stopping.

[14] P. D. Croft, Lawrence Radiation Laboratory Report UCRL-11563, Berkeley, 1964 (unpublished).