contributes to $R_3(m)$, as can be seen from the energy denominators, and hence to $G_3$ and $F_3$.

After a tedious calculation, and neglecting terms contributing obviously less than 0.1 G for our experimental conditions, we get

$$R_1(m)=-\tfrac{1}{4}A^2(-35/4+m^2+17m)-8D_1^2+16D_2^2, \quad \text{(A9)}$$

$$G_1=(17/2)B_0^2+16D_1\Delta, \quad \text{(A10a)}$$

$$G_2=A^2(2m-1)[8D_0-(67/4)A-20b_0], \quad \text{(A10b)}$$

$$\begin{aligned}G_3=\tfrac{1}{4}A^4[-2995/4+399(m^2-m)]+578AD_1^2D_0\\+1088AD_1^2D_2-1508A^2D_1^2+192A^2D_2^2\\+16A^2D_0^2+16A^3D_0(25/4-3m^2+3m)\\+384AD_0D_2^2,\end{aligned} \quad \text{(A10c)}$$

where

$$D_1=D\sin\theta\cos\theta, \quad \text{(A11a)}$$

$$D_2=\tfrac{1}{4}D\sin^2\theta, \quad \text{(A11b)}$$

$$B_0=\tfrac{1}{2}(A\sin^2\theta+B+B\cos^2\theta), \quad \text{(A11c)}$$

$$\Delta=(A-B)\sin\theta\cos\theta. \quad \text{(A11d)}$$

We have taken $g_{\perp}=g_{\parallel}=g$, and neglected the small difference between $A$ and $B$ except in (A10a) where this difference contributes 0.2 G at $\theta=90°$. Substituting Eqs. (A9) and (A10) in (A6c) and neglecting terms in $A-B$ and $b_0$, we finally obtain the contribution third order in $1/H_0$ to $\Delta H$:

$$\begin{aligned}F_3=A^4[-2107/16+131/4(m^2-m)]\\-D_0A^3[-108+16(m^2-m)]-1508A^2D_1^2\\+578AD_0D_1^2+1088AD_1^2D_2+192A^2D_2^2\\+16A^2D_0^2+384AD_0D_2^2.\end{aligned} \quad \text{(A12)}$$

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# Ranges of $Ba^{126}$ and $Ba^{128}$ Recoil Fragments in Aluminum*

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Using thin-target, thin-catcher recoil techniques, we have measured the average ranges and range straggling of 97-min $Ba^{126}$ and 2.4-day $Ba^{128}$ in Al. The Ba ions were produced with initial energies from 3 to 14 MeV by bombardment of $Sn^{120}$, $Sb^{121}$, and $Sb^{123}$ targets with $B^{10}$, $B^{11}$, $C^{12}$, and $N^{14}$ ion beams. The assumption of a compound-nucleus mechanism in the nuclear reactions leads to a smooth relationship between average range and recoil energy. Theoretical predictions are in good agreement with the observed ranges, but relatively small (<10%) systematic deviations become apparent at the higher energies. The range distributions are consistent with a Gaussian representation, and yield straggling parameters which are substantially larger than theoretical estimates of straggling inherent in the stopping process. Possible sources of the discrepancy are discussed. In one experiment, the product Ba ion is believed to be formed in a reaction involving emission of an alpha particle, and an abnormally large straggling parameter is observed.

## I. INTRODUCTION

THE stopping of energetic heavy ions in matter has been of considerable interest in recent years.[1–5] The theoretical description of the energy-loss processes has attained a degree of sophistication which allows a reasonable attempt at predicting stopping properties over a wide region of initial velocities.[1] To determine the accuracy of the theory, it is important that comparisons be made with experimental data for a variety of ions moving in various stopping media. A number of such studies have been reported,[6] and general agreement between theory and experiment has been found for range-energy relationships, with somewhat less satisfactory agreement for range straggling. However, as the measurements have become more refined, systematic deviations have appeared which may require a more critical examination of the theoretical approximations.[4]

In this paper we report measurements of average ranges and range straggling in Al of 2.8 to 14.2-MeV $Ba^{126}$ and $Ba^{128}$ ions (22 to 113 keV/amu). These energies correspond to a velocity region where the contribution of electronic interactions to the stopping process becomes dominating, and the theoretical energy dependences of the stopping properties are quite sensitive to the details of the theory. The energetic Ba ions were

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† Alfred P. Sloan Foundation Fellow.

[1] J. Lindhard, M. Scharff, and H. E. Schiott, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 33, 14 (1963).

[2] L. C. Northcliffe, Ann. Rev. Nucl. Sci. 13, 67 (1963).

[3] M. Kaplan and R. D. Fink, Phys. Rev. 134, B30 (1964).

[4] J. Gilat and J. M. Alexander, Phys. Rev. 136, B1298 (1964).

[5] V. Subrahmanyam and M. Kaplan, Phys. Rev. 142, 174 (1966).

[6] See, for example, Refs. 1–5, where referral to earlier work may also be found.

produced in nuclear reactions induced by heavy-ion (HI) beams, and recoiled out of thin targets into stacks of thin Al catcher foils. The reactions were mostly of the type (HI,*xn*) between $B^{10}$, $B^{11}$, $C^{12}$, and $N^{14}$ projectiles and $Sn^{120}$, $Sb^{121}$, and $Sb^{123}$ targets. All of our data are consistent with a compound-nucleus mechanism in these reactions, which permits the recoil energies of the Ba ions to be calculated directly.

## II. EXPERIMENTAL PROCEDURE

The techniques employed in the present studies were similar to those described previously.[3,5] Targets of $Sn^{120}$, $Sb^{121}$, and $Sb^{123}$ were prepared by vacuum evaporation of the isotopically enriched metals onto 0.00025-in. aluminum backings. The target layers were thin in comparison to the recoil ranges of the Ba products, and individual thicknesses were determined by weight and area measurements. The catcher foils were punched to known area from commercial aluminum leaf, and were individually selected and weighed. A target and a stack of catcher foils were mounted in a water-cooled holder, and irradiated with an appropriate collimated beam from the Yale heavy-ion linear accelerator. Bombarding energies less than 10.5 MeV/amu were obtained by inserting aluminum degrading foils of known thickness ahead of the target. Beam energies at the target were determined from the energy-loss measurements of Northcliffe.[2]

Following bombardment, the foils were separated and barium was isolated by standard radiochemical procedures. The purified samples were assayed for $\beta$ radioactivity on a series of end-window methane-flow proportional counters. The counters were intercalibrated and had counting efficiencies which were constant to within 1%. Blank corrections for activation of the catcher foils were determined from the activity observed in the last few foils of a stack. These corrections seldom exceeded a few tenths of 1%, and were easily applied. The decay curves obtained were cleanly resolved into 97-min and 2.4-day components corresponding to the half-lives of $Ba^{126}$ and $Ba^{128}$, respectively. (Most of the observed radioactivity was actually due to the short-lived Cs daughters in equilibrium with their Ba parents.) No significant contamination from other species was found.

## III. RESULTS

The relative activities of a series of catcher foils (corrected for chemical yields of the samples) gives directly the range distribution of recoil Ba ions. We have analyzed our data by means of probability plots, in the manner described previously.[3,5] The fraction of the total activity of the species which passes through a catcher thickness $t$ is plotted on a probability scale against $t$. In these coordinates, a Gaussian distribution of ranges will yield a straight line. In all of the experiments reported here, the activity distributions were in good agreement with a Gaussian relationship. We may, therefore, describe the distribution of $Ba^{126}$ and $Ba^{128}$ ranges in Al 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 $\rho$ is the straggling parameter. The quantity $R_0\rho$ is equivalent to the standard deviation of the Gaussian distribution. For each experiment, a probability plot analysis yielded an average range and a straggling parameter.

The initial energies of the recoiling Ba ions were computed assuming compound nucleus formation in the nuclear reactions. This implies a complete transfer of linear momentum from the incident beam followed by particle evaporation symmetric about 90° in the center-of-mass system. Under these conditions, the recoil energy is given by

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

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. The internal consistency of all our range-energy data, coupled with Gaussian range distributions and general agreement with stopping theory predictions (see Sec. IV), provides strong evidence for the validity of our assumption and the applicability of Eq. (2).

Table I summarizes our results. The first three columns list the characteristics of the experiment: target and beam, bombarding energy, and target thickness, respectively. Columns 4 and 5 give the recoil energies of $Ba^{126}$ and $Ba^{128}$, obtained from Eq. (2). Columns 6–9 present the average ranges and straggling parameters measured for $Ba^{126}$ and $Ba^{128}$ in aluminum. Whenever it was practical, data for $Ba^{126}$ and $Ba^{128}$ were determined in the same experiment. However, when conditions of bombardment or counting time led to grossly unequal contributions from $Ba^{126}$ and $Ba^{128}$ in the decay curves, only the dominant activity was used for the range determination.

## IV. DISCUSSION

It is most convenient to discuss our results in terms of the theoretical relationships developed by Lindhard, Scharff, and Schiott (LSS).[1] To do this effectively we shall first outline the elements of the LSS theory. The stopping of energetic heavy ions in matter is described as arising from electronic collisions (ionization) and nuclear collisions (ion-atom interaction), with the two processes being considered as uncorrelated and continuous. A Thomas-Fermi (statistical) model is used as a basis for the ion-atom interaction, and electronic stopping is assumed 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

TABLE I. Results of differential-range experiments for $\mathrm{Ba}^{126}$ and $\mathrm{Ba}^{128}$ stopping in aluminum.

| Target and beam | Bombarding energy $E_b$ (MeV) | Target thickness $W$ (mg/cm$^2$) | Recoil energy $E_R$ (MeV) | | Average range $R_0$(mg/cm$^2$) | | Straggling parameter $\rho$ | |
|---|---|---|---|---|---|---|---|---|
| | | | $\mathrm{Ba}^{126}$ | $\mathrm{Ba}^{128}$ | $\mathrm{Ba}^{126}$ | $\mathrm{Ba}^{128}$ | $\mathrm{Ba}^{126}$ | $\mathrm{Ba}^{128}$ |
| $\mathrm{Sn}^{120}+\mathrm{C}^{12}$ | 115.8 | 0.128 | 10.07 | ... | 0.808 | ... | 0.242 | ... |
| | 108.2 | 0.121 | 9.40 | ... | 0.748 | ... | 0.224 | ... |
| | 98.3 | 0.121 | 8.55 | ... | 0.699 | ... | 0.227 | ... |
| | 95.6 | 0.121 | 8.32 | 8.44 | 0.650 | 0.646 | 0.252 | 0.241 |
| | 92.0 | 0.121 | 8.00 | ... | 0.714 | ... | 0.227 | ... |
| $\mathrm{Sb}^{121}+\mathrm{B}^{11}$ | 87.5 | 0.034 | 6.96 | ... | 0.607 | ... | 0.232 | ... |
| | 72.7 | 0.030 | 5.79 | 5.88 | 0.510 | 0.510 | 0.265 | 0.247 |
| | 66.6 | 0.028 | 5.30 | 5.38 | 0.478 | 0.502 | 0.274 | 0.272 |
| $\mathrm{Sb}^{123}+\mathrm{B}^{11}$ | 114.1 | 0.044 | 8.84 | ... | 0.715 | ... | 0.217 | ... |
| $\mathrm{Sb}^{121}+\mathrm{B}^{10}$ | 72.2 | 0.028 | 5.31 | ... | 0.468 | ... | 0.257 | ... |
| | 56.6 | 0.037 | 4.16 | ... | 0.368 | ... | 0.267 | ... |
| | 49.5 | 0.031 | ... | 3.69 | ... | 0.323 | ... | 0.304 |
| | 37.0 | 0.030 | ... | 2.76 | ... | 0.263 | ... | 0.360 |
| $\mathrm{Sb}^{123}+\mathrm{B}^{10}$ | 103.6 | 0.037 | 7.38 | ... | 0.620 | ... | 0.232 | ... |
| | 98.0 | 0.041 | 6.99 | ... | 0.621 | ... | 0.257 | ... |
| | 92.1 | 0.042 | 6.57 | 6.68 | 0.580 | 0.543 | 0.245 | 0.278 |
| | 86.2 | 0.039 | 6.15 | 6.25 | 0.533 | 0.512 | 0.268 | 0.293 |
| | 76.5 | 0.041 | ... | 5.55 | ... | 0.498 | ... | 0.264 |
| | 74.3 | 0.041 | ... | 5.38 | ... | 0.480 | ... | 0.278 |
| $\mathrm{Sn}^{120}+\mathrm{N}^{14}$ | 138.2 | 0.128 | 13.58 | ... | 0.985 | ... | 0.199 | ... |
| $\mathrm{Sb}^{121}+\mathrm{C}^{12}$ | 124.1 | 0.031 | 10.62 | ... | 0.800 | ... | 0.217 | ... |
| $\mathrm{Sb}^{121}+\mathrm{N}^{14}$ | 144.2 | 0.043 | 13.96 | 14.18 | 0.965 | 0.985 | 0.232 | 0.200 |
| | 80.9 | 0.028 | ... | 7.95 | ... | 0.580 | ... | 0.432 |

$Z_s$, the kinetic energy $E_R$ and corresponding true range $R$ (total path length) are expressed as reduced (dimensionless) variables $\epsilon$ and $\rho_L$ given by

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

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

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, $N$ is the atomic density of the stopping medium, and $\hbar$ is Planck's constant divided by $2\pi$. In the approximation that the energy loss in nuclear and electronic stopping are independent, the total stopping power may be expressed as a linear combination of the two contributions:

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

LSS take the electronic stopping to be proportional to velocity,

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

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

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

where $\Delta(k,\epsilon)$ represents the effect of nuclear stopping and is treated as a correction to the electronic range. The parameter $k$ characterizes the static properties of the moving ion and the stopping medium. An approximate expression for this parameter is given as

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

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

Figure 1 is a range-energy plot for $\mathrm{Ba}^{126}$ and $\mathrm{Ba}^{128}$ in terms of the reduced variables $\rho$ and $\epsilon$ of the LSS theory. The points represent the experimental data from Table I, translated by means of Eqs. (3) and (4). The solid lines are theoretical predictions for $k=0.11$ and $k=0.13$. The former value of $k$ is the theoretical estimate from Eq. (9). The calculated curves have been converted[7] from true ranges to projected ranges (the experimentally measured quantity) as prescribed by LSS.[1] The general agreement between experiment and theory is good; however, above about $\epsilon=15$, a significant departure becomes noticable. At these higher energies, the experimental points no longer follow the $k=0.11$ line but tend to approach somewhat higher $k$ curves. This may be interpreted as an underestimation of the theoretical stopping at higher energies where electronic contributions have become of major importance. The apparent dependence of experimental $k$ value on energy may indicate that Eq. (7), which assumes electronic stopping to be proportional to velocity, is an oversimplification. Similar observations over the same energy region have been reported by Gilat and Alexander[4] for mass 149–151 fragments stopping in Al and various gases.

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[7] The projected range correction varied from 6% at $\epsilon=2$ to 3% at $\epsilon=25$.

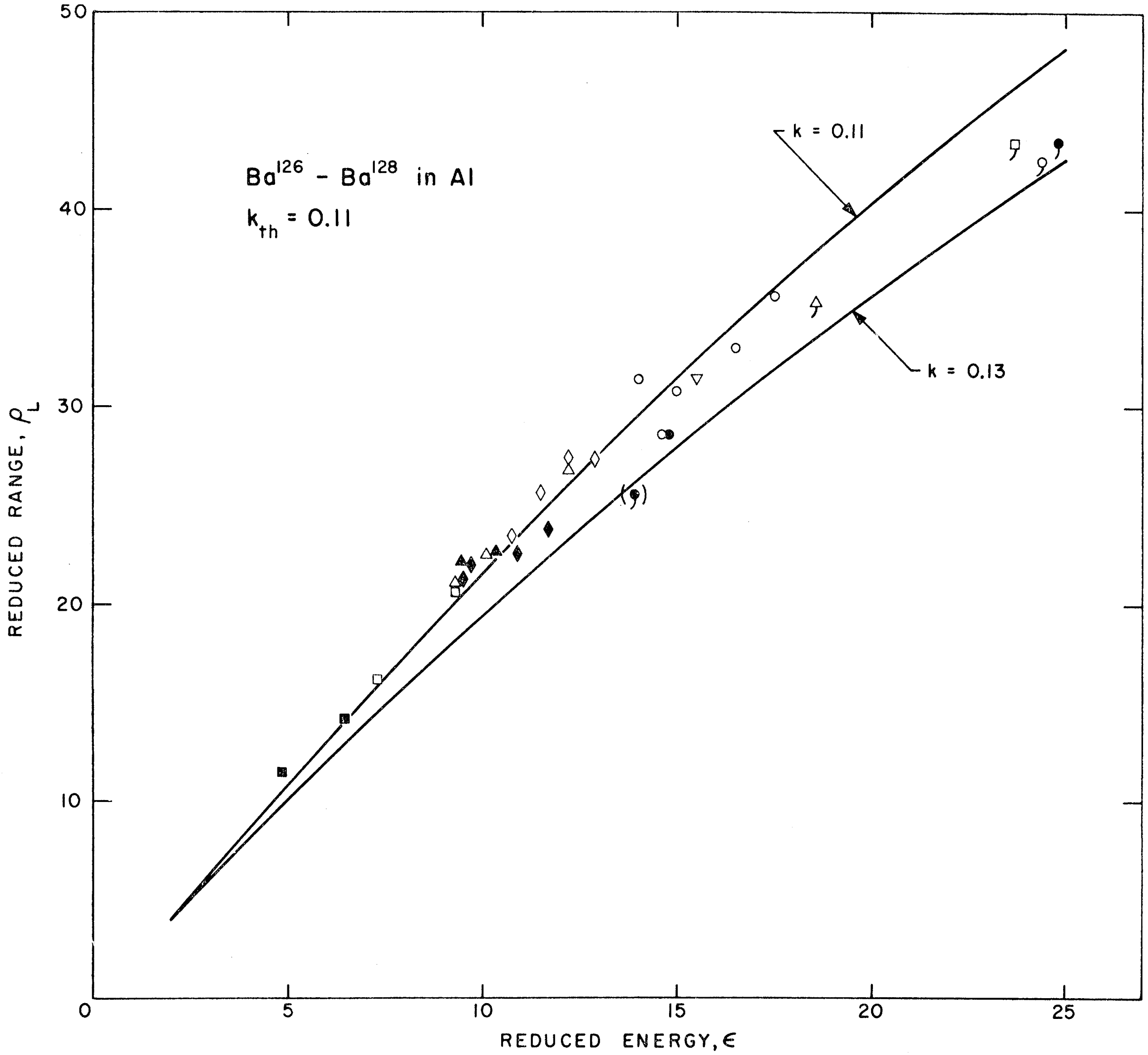

Fig. 1. Range-energy data for $Ba^{126}$ (open points) and $Ba^{128}$ (filled points) stopping in Al. The coordinates are expressed as dimensionless variables (see text). The various symbols refer to the nuclear reaction which produced the recoiling Ba ions, and are as follows: circles, $Sn^{120}+C^{12}$; squares, $Sb^{121}+B^{10}$; triangles, $Sb^{121}+B^{11}$; diamonds, $Sb^{123}+B^{10}$; inverted triangles, $Sb^{123}+B^{11}$; circles with tails, $Sb^{121}+N^{14}$; squares with tails, $Sn^{120}+N^{14}$; triangles with tails, $Sb^{121}+C^{12}$. The single data point in parentheses is for 81-MeV $N^{14}+Sb^{121}$ where the product $Ba^{128}$ is believed to have been formed via emission of an alpha particle. The solid lines are theoretical range-energy curves for two values of the electronic stopping parameter $k$.

Our range-straggling results are shown in Fig. 2. The ordinate is the relative square straggling, $\langle\Delta R^2\rangle/\langle R\rangle^2 \equiv\langle\Delta\rho_L{}^2\rangle/\langle\rho_L\rangle^2$, which is equivalent to the straggling parameter squared $\rho^2$. The experimental straggling parameters, taken from Table I, have been corrected for the very small effect of finite target thickness as described previously.[3] The data points do, however, contain contributions from straggling inherent in the stopping process, the initial energy spread of the recoiling ions resulting from particle evaporation, and the effects of inhomogeneities in the catcher foils. The solid lines are predictions from the LSS theory and refer only to the stopping process. Calculated curves are shown for two values of the electronic stopping parameter $k$, and for the case of pure nuclear stopping (labeled T.F. for Thomas-Fermi). As can be seen, the data lie considerably above the predicted $k=0.11$ position. The contribution from nuclear evaporation to the observed straggling may be estimated from other studies of very similar reactions[8] to be about 0.01–0.02 in $\rho^2$ units. This leads to the conclusion that the theoretically predicted straggling in the stopping process is about one-half of the comparable experimental quantity. Little is known concerning any straggling introduced by inhomogeneous catcher foils (of which no direct account has been taken here). Arguments have been given

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[8] J. M. Alexander, J. Gilat, and D. H. Sisson, Phys. Rev. **136**, B1289 (1964).

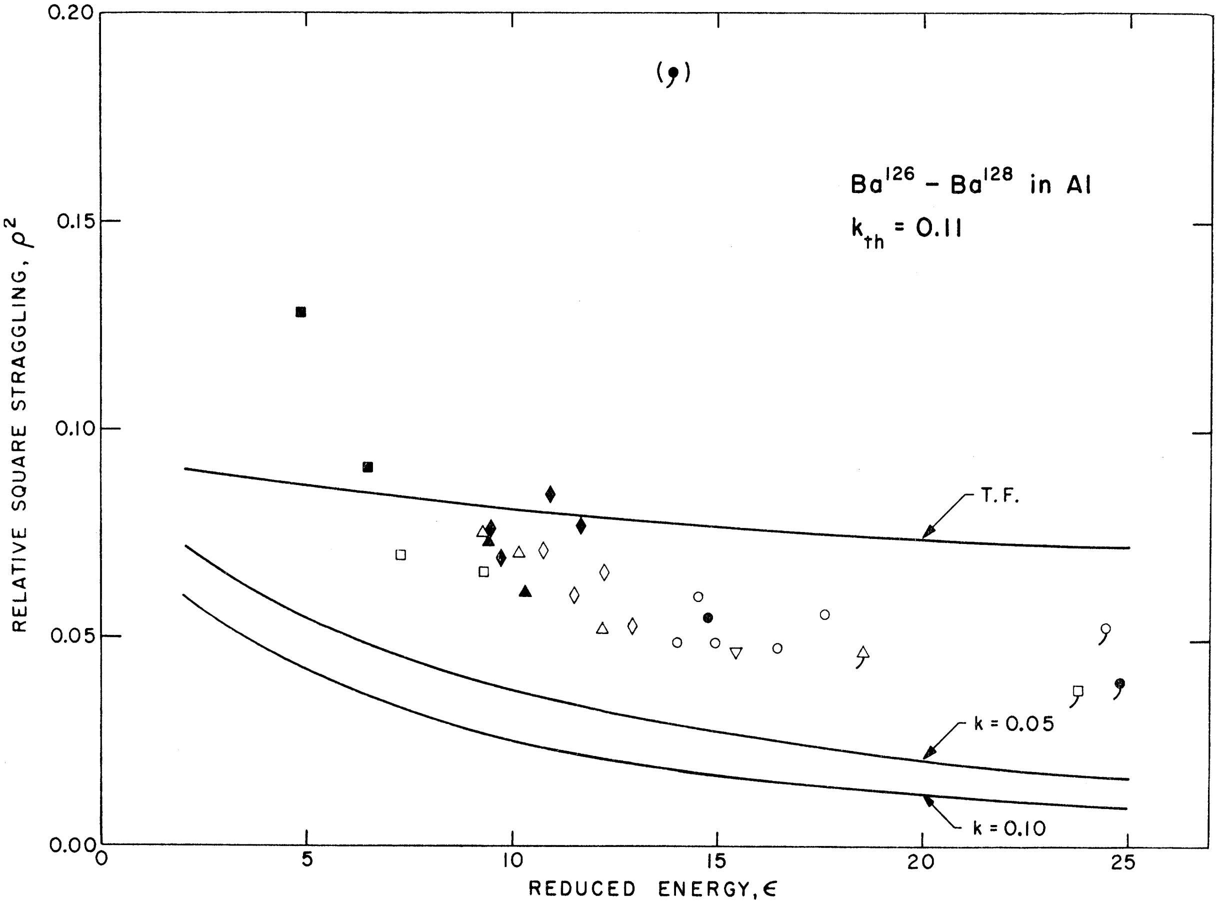

FIG. 2. Experimental relative square straggling in range plotted against reduced energy for $Ba^{126}$ (open points) and $Ba^{128}$ (filled points) stopping in Al. The data have been corrected for target thickness effects. The various symbols refer to the nuclear reaction which produced the recoiling Ba ions, and are as in Fig. 1. The solid lines are theoretical predictions for two values of the electronic stopping parameter $k$ ($k=0.10$ and $k=0.05$) and for pure nuclear stopping (T.F.). These curves refer only to straggling inherent in the stopping process. The single data point in parentheses is for 81-MeV $N^{14}+Sb^{121}$ where the product $Ba^{128}$ is believed to have been formed via emission of an alpha particle.

previously,[5] and we shall only point out that if significant effects are present, the apparent disagreement will be diminished.

The single point shown in parentheses in Figs. 1 and 2 is from the bombardment of $Sb^{121}$ with 81-MeV $N^{14}$ ions. Simple calculations of energetics indicate that at this bombarding energy the formation of $Ba^{128}$ is most likely to occur by de-excitation involving emission of an alpha particle. The observed effects are a slightly reduced average range (Fig. 1), and an enormously increased straggling parameter (Fig. 2). A relationship between alpha-particle emission and range straggling has been developed by Kaplan,[9] and we only wish to note here that the present $Ba^{128}$ measurement is consistent with the earlier $Sm^{142}$ data.[9]

Previous studies of range straggling at comparable energies ($\epsilon$ values) have also indicated that LSS predictions are too low.[3–5] The theory assumes that all of the straggling arises in nuclear stopping, and any straggling effects in electronic stopping are neglected. In the energy region under consideration, the relative contribution of electronic stopping is increasing rapidly and the straggling parameter predictions are very sensitive to the stopping cross sections. Thus the predicted straggling does not change much in going from $k=0.10$ to $k=0.05$ (as shown in Fig. 2) and both lie considerably below the T.F., or nuclear-stopping, value. It would seem that either there is a significant straggling contribution from electronic stopping, or perhaps the assumption of no correlation between nuclear and electronic stopping is too approximate when applied to calculations of straggling.

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[9] M. Kaplan, Phys. Rev. 134, B37 (1964).