article by Douglass and Falicov[8] where the relevant references are given.

A comparison of the theory with experimental results is given in Fig. 1. [Note that the theoretical curve, $F(\alpha)$, is approximated to better than 1% by

$$F(\alpha)\simeq 1.1+(5/14)\alpha, \quad 1.4<\alpha<2.8$$

for the range of α, which is of interest here.] Here the experimental values of α are identified with $\Delta(0)/kT_c$.[3] The agreement with the experimental points is seen to be fair.

It follows from the approximation scheme adopted here [Eqs. (1) and (2)] that for a given $\Delta(0)/kT_c$, $C_s(T_c)$ is determined with *no* adjustable parameters. The agreement with the actual values of $C_s(T_c)$ is surprisingly good for T near T_c (which is the range of interest). Figure 2 shows this for the case of BCS.[9]

In closing this section we would like to remark once again that the scale of our Fig. 1 is large compared with the accuracy of the experimental results available. For example, the values of $2\alpha=2\Delta(0)/kT_c$ for aluminium obtained by photon absorption is 3.16,[8] while the value of 2α obtained from tunneling experiment is 3.37 ± 0.01.[8] This is illustrated in Fig. 1. The difficulties in determining $C_s(T_c)/\gamma T_c$ precisely are well known; here one has to determine the lattice contribution, which in some cases (e.g. indium) are appreciable, and to determine γ.[10]

IV. REMARKS AND CONCLUSIONS

No attempt was made to fit the experimental points to theory which would refine the results of Ref. 3. The theoretical curve is in good agreement with the experimental result. In particular, in view of the fact that the empirical rule proposed in this paper, namely that C_s near T_c is given by Eq. (2) with α identified with $\Delta(0)/kT_c$, does *not* contain any adjustable parameters. Should further investigations prove our formula to be correct, a more general theory than the BCS weak-coupling-limit theory[1] will be required to account for it. Furthermore, analysis of measurements on hard superconductors along the lines outlined in this paper could shed light on the magnetization of such materials.

To discriminate between the results offered here and Toxen's, better measurements are needed for elements wherein $\alpha>1.85$ and $\alpha<1.55$. A good candidate for this is zinc ($\alpha=1.5$), where no recent value for H_0 is available.

[8] D. H. Douglass, Jr., and J. M. Falicov, in *Progress in Low-Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Company, Amsterdam, 1964), Vol. IV.

[9] B. Mühlschlegel, Z. Physik **155**, 313 (1959).

[10] H. R. O'Neal and N. E. Phillips, Phys. Rev. **137**, A750 (1965).

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Energy Loss of Fission Fragments in Light Materials*

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A calibrated silicon detector provided range-energy data for Cf^{252} fission fragments in H_2, D_2, N_2 and Mylar. Bohr's classical stopping-power theory and Lindhard's more recent Thomas-Fermi model both underestimate the measured energy losses. For both theories, the discrepancy with experiment is a monotonic function of the atomic number of the stopping material, at least for the limited range of materials studied.

INTRODUCTION

At the beginning of its range, a fission fragment loses energy chiefly by ionization and excitation of the stopping material. Bohr's classical treatment of electronic energy loss by a charged particle[1] gives

$$-\frac{dE}{dx}=\frac{4\pi z^2e^4}{m_ev^2}NZ_2\ln\left\{\frac{1.123m_ev^3\hbar}{ze^2I}\right\}, \qquad (1)$$

where z and v are the ionic charge and velocity of the particle; m_e and e are the electronic rest mass and charge, N and Z_2 are the atomic density and number of the stopping material; and I is a mean excitation and ionization potential for the stopping material. The derivation of (1) requires that $2ze^2/\hbar v\gg1$ and that $Z_2e^2/\hbar v\ll1$.

Other early treatments resulted in different arguments for the logarithmic term in Eq. (1). Bethe[2] employed the Born approximation which requires that $2ze^2/\hbar v\ll1$, a condition that cannot apply to fission fragments because of their high ionic charges and relatively low velocities. Bloch[3] included the perturbation of the electronic wave function by the incident particle

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[1] N. Bohr, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **18**, No. 8 (1949).

[2] H. A. Bethe, Ann. Physik **5**, 325 (1930).

[3] F. Bloch, Ann. Physik **16**, 285 (1933).

and obtained an expression that reduces to Eq. (1) for $2ze^2/\hbar v \gg 1$. Several authors have concluded, therefore, that of the early theories, Bohr's should best describe energy loss by fission fragments.[1,4,5] Because of the restriction $Z_2e^2/\hbar v \ll 1$, Eq. (1) should hold best for the very lightest stopping materials.

In recent years Lindhard and co-workers have constructed a "unified"-range theory for heavy ions. The analogous expression to Eq. (1) is

$$-\frac{dE}{dx}=N\xi_e 8\pi e^2 a_0 \frac{Z_1 Z_2}{(Z_1^{2/3}+Z_2^{2/3})^{3/2}}\frac{v}{v_0}, \qquad (2)$$

where a_0 and v_0 are the radius and velocity of the first Bohr orbit of hydrogen, Z_1 is the nuclear charge of the moving ion, and ξ_e is a constant "of the order of $Z_1^{1/6}$."[6] Northcliffe has emphasized that since Eq. (2) is based on a Thomas-Fermi description of both projectile and stopping material, its applicability is limited to $Z_1>10$ and $Z_2>10$.[6] We conclude, then, that Eq. (1) should still provide the best description for the energy loss of fission fragments in light materials.

As pointed out in a preliminary communication,[7] there have been no previous direct attempts to verify the applicability of Eq. (1) to fission fragments. Such a verification, of course, requires knowledge of the ratio z/v in addition to range-energy (E versus x) or stopping power (dE/dx versus x) data.

Lassen's early measurements of dE/dx for U^{235} fragments in gases[8] did not take into account the substantial "ionization defect,"[9] then unknown, nor can they be corrected for the defect in a straightforward manner. In any case, Lassen assumed the correctness of Eq. (1) in order to derive values for z from the data. Fulmer later reported range-energy curves for U^{235} fragments in several gases[10] and although he made simultaneous measurements of z/v,[11] he made no attempt to compare his measurements with theory. Neither Fulmer's range-energy data on solids,[10] nor those of Schmitt and Leachman,[12] nor the results from various radiochemical investigations[13] provide a good test for Eq. (1) because of the restriction, $Z_2e^2/\hbar v \ll 1$, mentioned above.

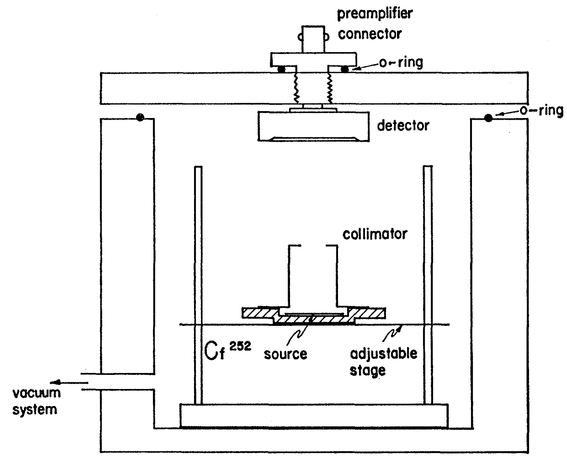

FIG. 1. Experimental arrangement.

EXPERIMENTAL

1. Apparatus

Figure 1 is a scale drawing of the range-energy apparatus. The silicon surface-barrier detector (Ortec Model SBQN-450-60) had an active area of 450 mm^2, a 75-Å-thick gold layer, and a maximum resolution of 60 keV, full width at half-maximum (FWHM), for alpha particles. A bias of 100 V provided a depletion region 95 μ deep. The detector was mounted inside the removable cover of a hollow aluminum cylinder of inside diameter 3 in. and height 2-17/64 in. A Cf^{252} source, Mylar films, and various cylindrical collimators were mounted on an adjustable stage below the detector. The collimators restricted the variation in fragment path lengths and prevented edge effects in the detector's response.

After amplification by a charge-sensitive preamplifier and linear amplifier, fission-fragment or alpha spectra were recorded with a 400-channel pulse-height analyzer. The stability of the electronic system was checked with a standard signal from a mercury pulser before and after each experiment.

The Cf^{252} source,[14] plated on a platinum foil over a circular area 4 mm in diameter, had a fission activity of approximately 10^5 disintegrations/min.

Cylinder gases ($H_2>99.8\%$, $D_2>99.5\%$, $N_2>99.9\%$) were admitted to the apparatus without further purification. The cell was sequentially evacuated to 10 μ Hg and flushed with gas several times before a run was made. Pressures were measured with a McLeod guage or a mercury manometer.

2. Measurement of Film Thickness

A series of Mylar films[15] of nominal thicknesses between 15–50 mils were used in the range-energy

[4] H. A. Bethe and J. Ashkin, in *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley & Sons, Inc., New York, 1953), Vol. I, Chap. 2.
[5] R. D. Evans, *The Atomic Nucleus* (McGraw-Hill Book Company, Inc., New York, 1955), p. 584.
[6] L. C. Northcliffe, Ann. Rev. Nucl. Sci. **13**, 67 (1963).
[7] R. C. Axtmann and P. Mulás, Bull. Am. Phys. Soc. **10**, 611 (1965).
[8] N. O. Lassen, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **25**, No. 11 (1949).
[9] E. K. Hyde, *The Nuclear Properties of the Heavy Elements: Fisson Phenomena* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1964), Vol. 3, pp. 167–9.
[10] C. B. Fulmer, Phys. Rev. **108**, 1113 (1957); Oak Ridge National Laboratory Report No. ORNL-2320, 1957 (unpublished).
[11] C. B. Fulmer and B. L. Cohen, Phys. Rev. **109**, 94 (1958).
[12] H. W. Schmitt and R. B. Leachman, Phys. Rev. **102**, 183 (1956).
[13] B. G. Harvey, Ann. Rev. Nucl. Sci. **10**, 235 (1960).
[14] Very kindly loaned by Professor T. D. Thomas.
[15] Graciously supplied by J. P. Harrington of E. I. du Pont de Nemours and Company.

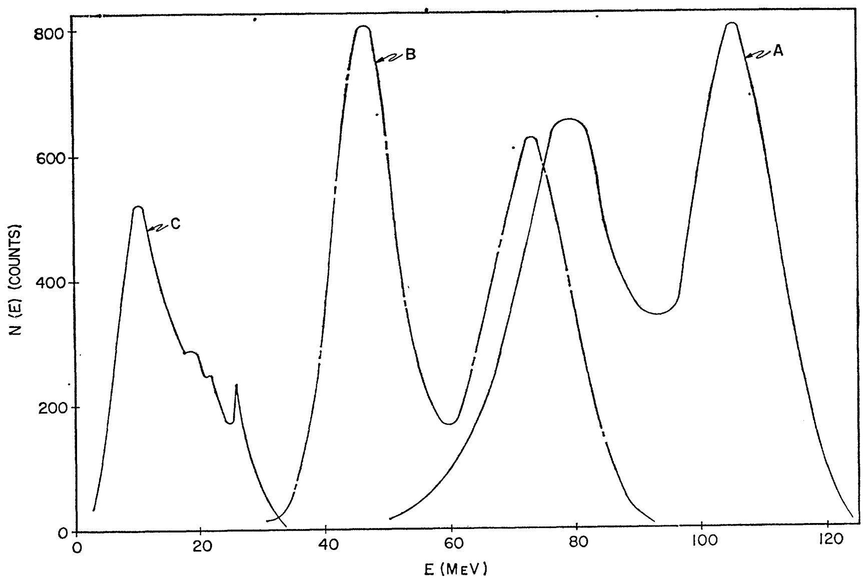

FIG. 2. Experimental spectra of fission fragments from Cf^{252} obtained with a silicon surface-barrier detector. Curve A represents data obtained with nondegraded fragments. Curve B was obtained with a pressure of hydrogen corresponding to 15.29 mg/cm². Curve C corresponds to 51.26 mg/cm² of hydrogen.

studies of Mylar itself and, in some cases, to predegrade the fission fragments so that the pressure required for the gas studies could be kept below atmospheric pressure. A combination gravimetric and alpha-particle thickness-gauge procedure[16] provided a relationship between $E(\alpha)$, the residual energy of a Cf^{252} alpha particle (6.11 MeV), and S_M, the thickness of Mylar in mg/cm², through which the alpha particle had passed:

$$S_M = 6.247 - 0.7541E(\alpha) - 0.0444E(\alpha)^2. \quad (3)$$

3. Calibration of the Detector

Although semiconductor particle detectors respond linearly to light particles, including alphas, they exhibit a pronounced "pulse-height defect" for fission fragments. Some recent studies[17–19] indicate that most of the defect arises near the end of the fragment track. Slowly moving atoms, produced by screened Coulomb scattering with degraded fission fragments, are inefficient at creating hole-electron pairs.

Schmitt, Kiker, and Williams have reported a simple calibration procedure that involves a measurement of the detector's response to Cf^{252} fission fragments.[20] The procedure is valid down to about 25 MeV—roughly the upper limit for screened Coulomb scattering of the fragments by silicon nuclei. The detailed response of silicon detectors to fission fragments of less than 25 MeV has not yet been studied so we have rather arbitrarily assumed a linear response from 0–25 MeV. The combination of the Schmitt procedure with the above assumption gives, for our detector, $\bar{E}$, the average fission-fragment energy as a function of $\langle PH \rangle_{av}$, the average pulse height

$$\bar{E} = 17.166\langle PH \rangle_{av} + 9.020 \quad 25 \leq \bar{E} \leq 100 \text{ MeV}, \quad (4a)$$

$$\bar{E} = 26.850\langle PH \rangle_{av} \quad 0 \leq \bar{E} < 25 \text{ MeV}. \quad (4b)$$

In order to limit the uncertainties in the data due to the arbitrariness of Eq. (4b), we only present data corresponding to $\bar{E} \geq 31.5$ MeV for which the contribution to the spectrum from fragments of less than 25 MeV is insufficient to affect $\bar{E}$ by more than 1 MeV, irrespective of the true calibration below 25 MeV.

The detector was also calibrated by an earlier technique[21] that involves an extrapolation to higher energies of its response to four alpha emitters. The two methods agreed at 100 MeV to within 0.6 MeV, but it is the Schmitt and Kiker procedure that was actually used to interpret the data. We estimate that the probable errors of the reported values for $\bar{E}$ are, in every case, less than plus or minus 1 MeV.

Figure 2 displays a virgin fission-fragment spectrum obtained with the apparatus and compared with two other spectra for fragments degraded to lower energies. As the pressure of the gas is increased the excellent resolution demonstrated in the nondegraded case becomes meaningless for distinguishing between heavy and light fragments but still permits an accurate calculation of the average energy.

As a check on the correctness of the procedures, measurements of geometry and gas pressures, and on the absence of systematic error, the apparatus was used to determine I, the mean excitation and ionization

[16] P. Mulás, Ph.D. thesis, Princeton University, 1965 (unpublished). Available on microfilm.
[17] A. R. Sattler, Phys. Rev. **138**, A1815 (1965).
[18] R. C. Axtmann and D. Kedem, Nucl. Instr. Methods **32**, 70 (1965).
[19] E. L. Haines and A. B. Whitehead, Rev. Sci. Instr. (to be published).
[20] H. W. Schmitt, W. E. Kiker, and C. W. Williams, Phys. Rev. **137**, B837 (1965).
[21] See, for example, H. C. Britt and H. E. Wegner, Rev. Sci. Instr. **34**, 274 (1963).

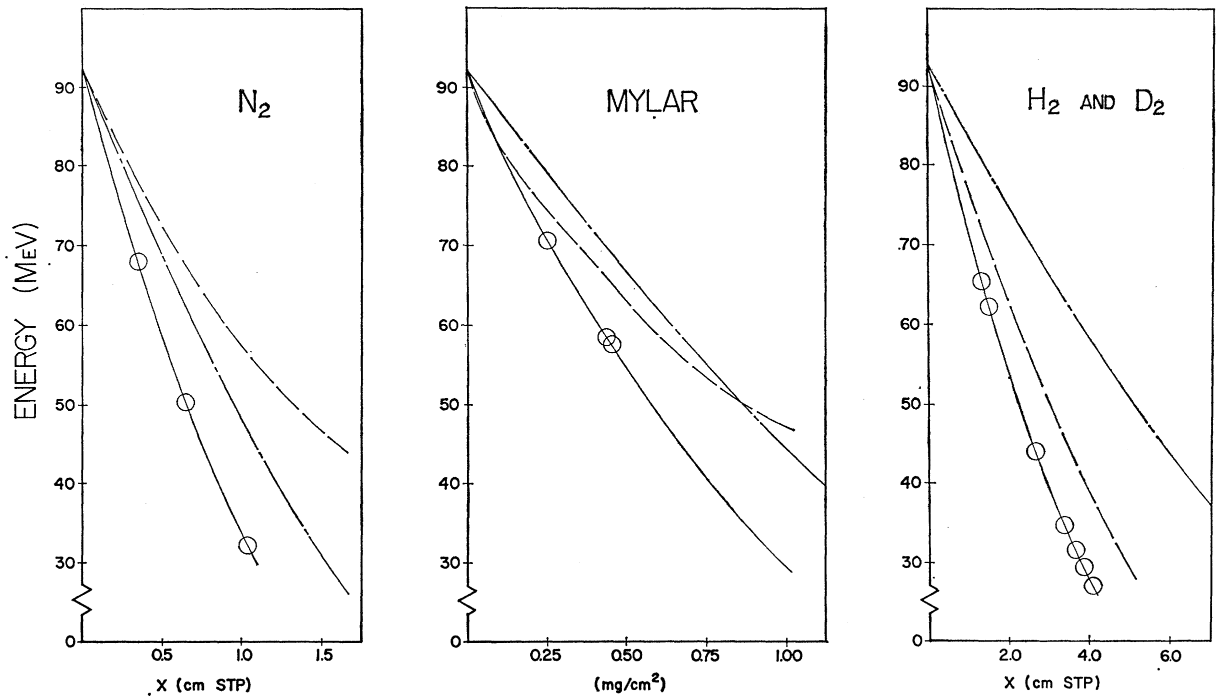

FIG. 3. Range-energy data for Cf^{252} fission fragments compared with theory. The solid curves are fits of the experimental data. The regularly dashed curves represent the integral of Eq. (1)—the Bohr theory, while the alternately dashed curves are plots of Eq. (6)—the Lindhard theory. In the case of H_2 and D_2, approximately one-half of the data was obtained with each gas. The diameter of the circles that represent the data reflect the estimated probable error in the average energies (±1 MeV).

potential, of H_2 and N_2 from measured values of dE/dx at the beginning of the range of the 6.11-MeV alpha particles from Cf^{252}. The stopping power for light charged particles in a light stopping medium is given by[22]

$$-\frac{dE}{dx}=\frac{4\pi z^2e^4}{m_ev^2}NZ_2\left\{\ln\frac{2m_ev^2}{I}-\frac{C_K}{Z_2}-\ln(1-\beta^2)-\beta^2\right\}, \quad (5)$$

where C_K is a screening constant[22] and $\beta\equiv v/c$. From the constant slope of the range-energy data in the interval of alpha energy from 5.7–6.11 MeV for H_2 and 5.2–6.11 MeV for N_2, the results were $I_{H_2}=14.8$ eV and $I_{N_2}=81.0$ eV, in quite reasonable agreement with values in the literature.[2,5]

A final check on the ability of the apparatus to measure fission-fragment energies was offered by comparison of the final results on nitrogen (see below) with those obtained by a completely independent method based on the ability of nitrogen to act as a gaseous scintillation counter. This comparison, published elsewhere,[23] gave excellent agreement down to fission-fragment energies of about 40 MeV, the lower limit of validity of the scintillation data.

RESULTS AND DISCUSSION

Figure 3 presents the range-energy data for N_2, Mylar and H_2 and D_2. In each case the experimental data are compared with the Bohr and the Lindhard theories in integral form. Equation (1) was integrated with a digital computer; Eq. (2) was integrated analytically to give

$$x(E)=2/k[E_0^{1/2}-E^{1/2}], \quad (6)$$

where E is the energy of a heavy particle of original energy E_0 which has penetrated to distance x in a stopping material; and k is a collection of constants that includes M_1 and M_2, the mass numbers of the projectiles and stopping materials, respectively. All the theoretical curves displayed represent averages of two curves: one computed for the mean heavy fragments and one for the mean light fragments. This procedure was necessary since the experiment did not distinguish between the two groups of fragments but rather yielded an average energy for all the fragments. Table I gives the values for the various parameters used in the computations.

Values for z/v for use in the Bohr equation were derived from the magnetic spectrographic measurements of Fulmer and Cohen[11] and of Lassen[24] on U^{235} fragments. Where values of the ratio were available from both sources, the greater value was adopted in order to minimize the discrepancy between our measurements and Bohr's theory. All of the present experimental measurements were made at sufficiently high

[22] Reference 5, p. 638.

[23] R. C. Axtmann and J. T. Sears, Nucl. Sci. Eng. 23, 299 (1965).

[24] N. O. Lassen, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 26, No. 12 (1951).

TABLE I. Parameters of fission fragments from Cf^{252} for comparison with stopping-power theories.

	Light fragments	Heavy fragments
Mass–M_1 [a]	108 amu	144 amu
Nuclear charge–Z_1 [b]	42.5	55.5
Initial velocity–V_0 [a]	1.36×10^9 cm/sec	1.03×10^9 cm/sec
Initial atomic charge–z [c]		
in H_2 and D_2	19.6	17.1
in Mylar	22.7	26
in N_2	18.8	16.7

[a] S. L. Whetstone, Phys. Rev. **131**, 1232 (1963).
[b] Computed from Glendenin's rule. See Ref. 9, p. 143.
[c] See text.

pressures that z/v had attained a constant value.[11,24] The z/v measurement for U^{235} fragments of a given mass group were scaled to the case of the corresponding Cf^{252} fragments with the assumption that $z/E^{1/2}$ is a constant. This latter relationship has some theoretical validity[1] but is justified here on the empirical basis that $z/E^{1/2}$ is remarkably constant, even for fragments from different mass groups, in most materials for which data are available.[11,24]

Values for Z_2 and A_2 for Mylar, a polyester formed by condensation of ethylene glycol and terephthalic acid, were computed from formulas given by Evans.[25] The initial charges z of U^{235} fragments for a solid with the effective atomic number so computed ($Z_2=4.53$), were obtained from the work of Lassen.[24]

The experimental data agree qualitatively with the work of Fulmer on U^{235} fragments[10] and that of Moak and Brown[26] who produced the synthetic fission fragments Br^{79}, Br^{81}, and I^{127} with an accelerator. Quantitative comparison of our data with these other experiments is unprofitable because of differences not only in the projectiles used but in the stopping materials and, in the case of Fulmer's data, the experimental conditions.[27]

As may be seen from Fig. 3, the data for H_2 and D_2 fall on the same smooth curve, a circumstance that confirms the usual assumption that energy loss by fission fragments in the energy range down to about 30 MeV is primarily via electronic interactions. The figure demonstrates, however, that both the Bohr and the Lindhard theories underestimate the actual energy loss for the limited number of stopping materials tested. In the case of the Bohr description, the discrepancy increases with Z_2, the atomic number of the stopping material, while in the case of the Lindhard formalism, the opposite tendency obtains.

The same trends are observed when Fulmer's data on U^{235} fragments, corrected for pressure effects,[28] are compared with theory. Moak and Brown have compared their data,[26] which covers a greater range of Z_2, with the Lindhard theory and have found the same general trend with Z_2 that was observed in the present work.

The fact that the Lindhard treatment tends to agree with experiments more closely at higher values of Z_2 is not, of course, surprising since that theory is based on Thomas-Fermi considerations. Likewise the Bohr description's poorer agreement as Z_2 increases is to be expected since the condition $Z_2e^2/\hbar v<1$ holds over the entire range of electronic energy loss only for H_2 and D_2 and is violated even at the beginning of the fragment range in the case of N_2.

In summary, the present work indicates that, in spite of injunctions to the contrary,[1,4,25] use of available detailed theories for estimates of fission-fragment energy loss appears unwise; and points up the existence of a significant void in the nearly complete fabric of the stopping-power theory for charged particles.

ACKNOWLEDGMENTS

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[25] Reference 5, Chap. 22.
[26] C. D. Moak and M. D. Brown, Bull. Am. Phys. Soc. **10**, 611 (1965); Dr. Moak (private communication).
[27] See Ref. 18, p. 73, Note added in proof.
[28] C. B. Fulmer, Phys. Rev. **139**, B54 (1965).