In summary, the recent experimental data for pure $E2$ transitions reveals a consistent picture. The observed L_{II}/L_{III} ratios are in reasonable agreement with the calculated values, while the experimental L_I/L_{II} and L_I/L_{III} ratios are significantly larger than theory. The present results show that the mean deviation of the sum of the experimental $(L_I/L_{II}+L_I/L_{III})$ ratios from theory is 5.4%.

This deviation is consistent with that observed by Herrlander and Graham[24] for the spherical nuclei in the region $Z=76$–80. Present evidence thus suggests that this small deviation is independent of atomic number, and nuclear deformation. In view of the close agreement between the three available sets of theoretical values for this class of pure $E2$ transitions it seems likely that the observed discrepancy originates in the assumptions on which the theoretical computations are based rather than in the computations themselves.

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ACKNOWLEDGMENTS

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Slowing of Fission Fragments in Noble Gases*

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Measurements have been made which give information on the slowing of ^{235}U fission fragments by gaseous and metallic-foil absorbers. Emphasis was placed on slowing of fragments by noble gases: helium, neon, argon, krypton, and xenon. The data show significant discrepancies from the existing theory, and suggest that it is necessary to take into account the effects of electronic shell structure of the fragment in treating the slowing-down process.

I. INTRODUCTION

PERHAPS the most nearly complete and universally applicable treatment of the slowing of heavy charged particles in matter is that of Lindhard and co-workers.[1–3] The Lindhard treatment appears to give a particularly good description of range-energy relations and range straggling in the stopping of low-atomic-mass particles at lower energies in light materials,[4] but the limitations of the treatment in describing fission-fragment ranges have not yet been established.

In a previous paper,[5] some initial results of the present series of measurements were reported. In this earlier experiment, which relied on metallic foils as fission-fragment absorbers, it was assumed that the fragments which penetrated the absorbing foil were those fragments associated with the highest total-kinetic-energy release. The present study is an attempt to verify this assumption,[6] and to check the applicability of the theoretical treatment of Lindhard.

Lindhard, following the approach outlined by Bohr,[7,8] separates the stopping cross section for heavy ions in matter into two components: a nuclear stopping cross section which varies comparatively slowly with particle

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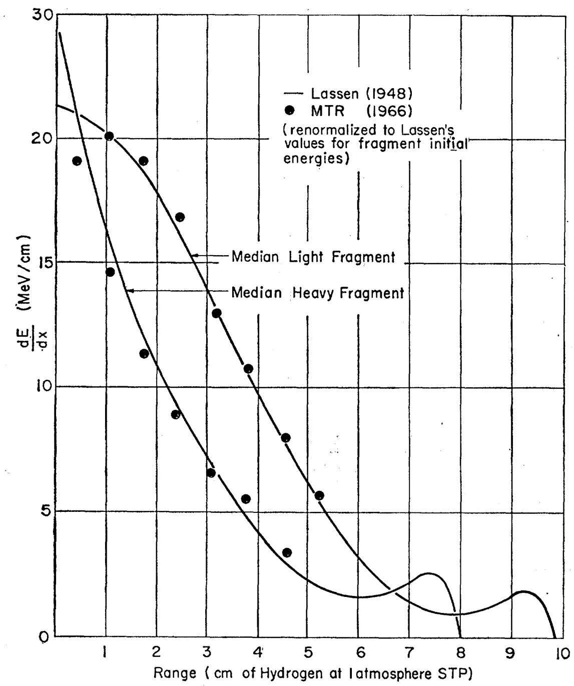

FIG. 1. Rate of energy loss of median light and heavy fragments from ^{235}U fission along their range in hydrogen. The solid curve is taken from early measurements made by Lassen (Ref. 13); the data points are the results of the present measurement, renormalized to the values given by Lassen for the initial fragment energies.

energy and for high energies and large deflections, has the form of Rutherford scattering, and an electronic stopping cross section which describes the energy loss by ionization and excitation of atoms in the stopping material due to the passage of the heavy ion. This separation into nuclear and electronic stopping cross sections is physically rather significant for fission fragments: it is the nuclear cross section which is responsible for the existence of the larger pulse-height defect, higher rate of radiation damage, and significantly poorer energy resolution of solid-state detectors for fission fragments than for lighter particles. This was demonstrated both by recent theoretical calculations of Haines and Whitehead[9,10] and in experimental studies of the channeling in solid-state detectors of mock fission fragments (accelerated heavy ions of iodine and bromine) by Moak *et al.*[11,12]

In his theoretical treatment, Lindhard introduces the dimensionless energy and range parameters ϵ and ρ, defined as

$$\epsilon=\frac{EaM_2}{Z_1Z_2e^2(M_1+M_2)}, \tag{1}$$

and

$$\rho=RNM_2(4\pi a^2)\frac{M_1}{(M_1+M_2)^2}, \tag{2}$$

where E is the particle energy and R is the range, N is the number of atoms of stopping material per unit volume, Z_1 and Z_2 are the nuclear charges of the incident particle and of the atoms of the stopping material, respectively, and M_1 and M_2 are the respective atomic masses. The parameter a is a Thomas-Fermi penetration constant, equal to $0.8853a_0(Z_1^{2/3}+Z_2^{2/3})^{-1/2}$, where $a_0=\hbar^2/me^2$, the Bohr radius in hydrogen. Over a considerable part of the range, the electronic stopping is expected to be proportional to the velocity of the incident particle, or

$$(d\epsilon/d\rho)=k\epsilon^{1/2}, \tag{3}$$

where the proportionality constant k can be estimated as

$$k\simeq\frac{0.0793Z_1^{1/6}(Z_1Z_2)^{1/2}(M_1+M_2)^{3/2}}{(Z_1^{2/3}+Z_2^{2/3})^{3/4}M_1^{3/2}M_2^{1/2}}. \tag{4}$$

The condition for the validity of Eq. 3 is that the velocity of the incoming particle be smaller than the velocity of its most energetic bound electrons, which permits electronic capture and loss to control the stopping. Typical fission-fragment velocities are of the order of 0.9×10^9 and 1.4×10^9 cm/sec for the average heavy and light fragment, respectively, so Eq. 3 should be valid for these fragments at their highest energies. However the earliest experiments on the rate of energy loss of fission fragments per unit range, carried out by Lassen,[13] showed that the rate of energy loss is not strictly proportional to velocity. As shown in Fig. 1, Lassen found that the rate of energy loss for fission fragments in hydrogen (and in other absorbers) shows different behavior for light and heavy fragments. Of particular interest is the "knee" in the light-fragment curve. Its existence has been confirmed a numbe1 of times, most recently by Nasyrov.[14,15]

The existing theoretical treatment does not describe this curve, but Bohr and Lindhard[8] suggest that it may be an electronic density effect. If the atomic charge on the fission fragment is in excess of $Z/2$, where Z is the nuclear charge, then one might expect departures from the energy-loss–velocity proportionality as given in Eq. 3. This occurs only for the light fragment near the beginning of the slowing-down process.

Lindhard, Scharff, and Schiøtt[1] compared theoretical estimates with a number of experimental measurements[16–18] of fission-fragment total ranges, and con-

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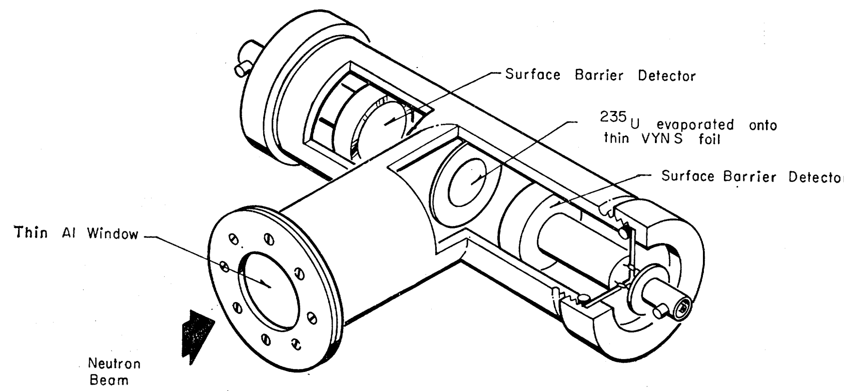

FIG. 2. Pictorial view of the fission chamber used. The back-to-back detectors were separated by a distance of 5.5 cm, with the ^{235}U target located approximately midway between them. The gaseous absorber was introduced through a pumping port which is not shown.

cluded that the agreement here between theory and experiment was satisfactory. The discrepancies of approximately 10–20% were not larger than the errors associated with many of the measurements. More recently, however, experiments have been done which indicate larger departures from the theory. Noshkin[19] carried out radiochemical measurements of ranges of selected fission fragments in a variety of stopping foils, and found that the experimental data were consistent with the Lindhard formalism, provided that the proportionality constant k (Eq. 4) is increased by about 50%. Aras, Menon, and Gordon[20] also concluded from absorber measurements that the Lindhard constant k should be increased. Mulas and Axtmann[21] studied residual pulse heights of ^{252}Cf fission fragments slowed in low-Z gases and absorbing foils (an experiment very similar to that reported here). They too found that the rate of energy loss was generally larger than that given by the Lindhard equations.

II. EXPERIMENTAL RESULTS

The experimental arrangement is shown in Fig. 2. The fission foil consisted of a deposit, up to 10 $\mu g/cm^2$ in thickness, of $^{235}UCl_4$, on a thin (5–10-$\mu g/cm^2$) film of VYNS.[22] It was estimated that the heaviest fragments may have lost as much as 2 MeV of energy in the fission foil itself. The two fragments from reactor-neutron-induced binary fission of ^{235}U were detected in coincidence by two surface-barrier solid-state detectors. The pulse heights from the detectors were stored in a two-dimensional multichannel analyzer. The experiment consisted of the measurement of the residual pulse heights of the coincident fission fragments as a function of the absolute pressure of absorbing gas in the chamber. The geometrical solid angle intercepted by each detector was 3% of the unit sphere, but the requirement that the two fragments be detected in coincidence reduced the effective solid angle to about 1%. The coincidence requirement was also of great help in defining the path length over which loss of energy to the gas occurred. The average path length was about 2% larger than the measured distance from the fission foil to either detector.

It was assumed that these measurements represent only the electronic stopping cross section. Electronic stopping is dominant over the range of fragment energies considered, and, especially for the heavier gases, the nuclear stopping cross section should lead to fairly sizeable deflections of the fragment. Such events should not be detected in a low-geometry coincidence experiment. In the present experiment, it was found that the relative coincidence rate decreased markedly for the heavier gases when the fragments approached the end of the range, where nuclear stopping is expected to become relatively more important.

The raw data collected consisted of pulse-height contour plots, similar to those shown in Figs. 3 and 4 for

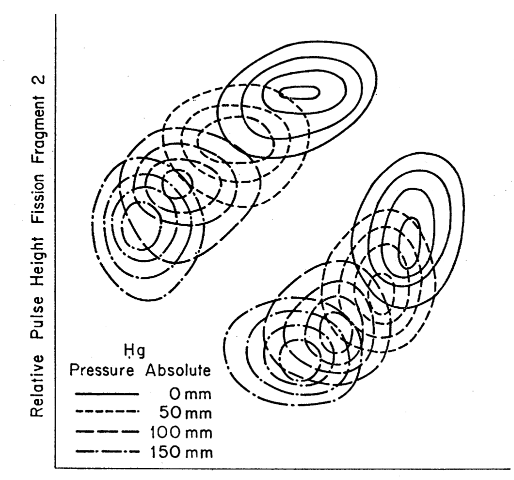

FIG. 3. A typical series of contour plots showing smoothed pulse-height distributions of coincident fission fragments from reactor neutron-induced fission of ^{235}U as a function of argon gas as an absorber, ranging between 0- and 150-mm pressure.

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[20] N. K. Aras, M. P. Menon, and G. E. Gordon, Nucl. Phys. **69**, 337 (1965).
[21] P. M. Mulás and R. C. Axtmann, Phys. Rev. **146**, 296 (1966).
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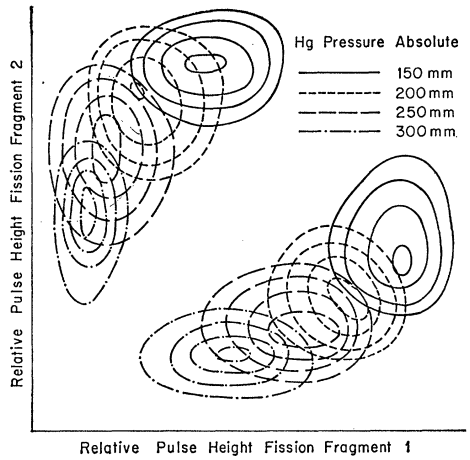

FIG. 4. A typical series of contour plots showing smoothed pulse-height distributions of coincident fission fragments from reactor neutron-induced fission of ^{235}U as a function of argon gas as an absorber, ranging between 150- and 300-mm pressure. A scale change of a factor of 2 is to be noted between these curves and those shown in Fig. 3.

the slowing of ^{235}U fission fragments by argon gas. The multichannel analyzer used for data collection was of modest size (giving an array of 32×32 channels), and the pulse-height resolution was insufficient to permit the observation of more than a hint of the fine structure which has been reported for high-fragment kinetic energies.[23,24] Nevertheless, the fine structure observed was of considerable help in giving a rough qualitative interpretation of the early data,[5] which the present study has shown to be essentially correct.

In the reducing of the data, it was assumed that the centroids of the light- and heavy-fragment distributions are representative of the slowing of fragments with nuclear charges of 37.4 and 54.6, and nuclear masses of 94.9 and 138.6 amu, respectively. In converting from pulse height to energy, the procedure described by Schmitt[25] was found to be inadequate at lower energies unless the pulse-height defect of the detectors was small. Much better consistency was found if it was assumed that the response of the detectors varies linearly with the fragment energy. (For the best detectors, this procedure gave answers which were virtually identical with those from the Schmitt formula[25] over the range of validity of that formula.)

One other correction was necessary for several of the runs. It was observed that radiation damage of the detectors by fission fragments could not always be neglected. This caused a gradual, mass-dependent decrease of the pulse height for a given set of gas absorber conditions. A corresponding deterioration of the resolution could also be noticed as the radiation damage progressed, although data taken under conditions of poor resolution were discarded in the final analysis. The effects of radiation damage were minimized by keeping the number of fissions detected per run to a minimum (30 000 coincidences, or 10^5 singles per detector per distribution). Frequent rechecks of the most easily reproduced situation (no gaseous absorber material) permitted corrections for progressive radiation damage. For these corrections, it was assumed that the radiation damage which occurred depended linearly on the number of fission fragments detected, but was independent of their energy. For most of the runs, the pulse-height deterioration was fairly small ($<1\%$); in only one series of runs (using krypton as the absorbing gas) was the cumulative radiation-damage effect larger than 5%. In all measurements, gas pressures were

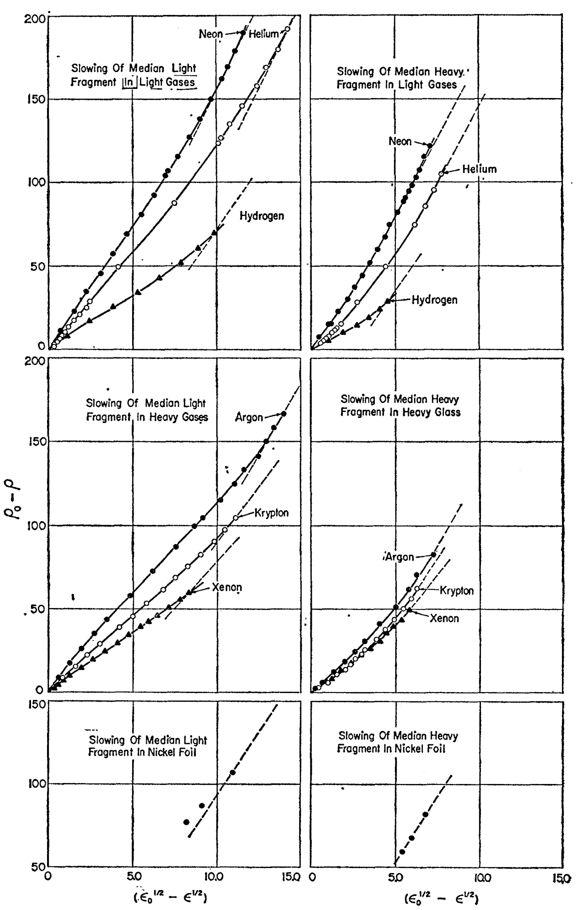

FIG. 5. The slowing of median light and heavy fragments from ^{235}U fission in various gases and in nickel foil. The ordinate is proportional to the fragment range expended in the absorber, and the abscissa is proportional to the velocity loss of the fragment in the absorber. The suppressed origin should be noted for the slowing in nickel foil. In all cases, the linear relationship between the range and velocity parameters, expected from the Lindhard treatment (Refs. 1–3) is given by the dashed line.

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high enough that no corrections were necessary for saturation of the density effect.[26,27]

The data obtained are plotted in Fig. 5. This figure shows the slowing of the median light and heavy fragments for ^{235}U fission in the lighter gaseous absorbers H, He, and Ne; in heavier noble gas absorbers Ar, Kr, and Xe; and finally for slowing in metallic foil. The nickel foil data are extremely scanty, because of the limited range of thicknesses of foil absorber available at the time. The data are presented with the aid of Lindhard's dimensionless parameters ϵ and ρ as defined in Eqs. (1) and (2). If ϵ_0 and ρ_0 are proportional to the energy and range, respectively, of an unslowed fragment, and ϵ and ρ are the residual energy and range of the fragment which has been slowed by passing through a certain amount of absorber, then the quantities of interest are $\epsilon_0-\epsilon$, which is proportional to the energy expended by the fragment in the absorber, and $\rho_0-\rho$, which is proportional to the thickness of absorber present. If it is assumed that the conditions of the experiment (the low geometry, and the requirement that a coincidence exist between the two fragments) are such that only electronic stopping is being observed, then Eq. (3) can be integrated to give the relation

$$(\rho_0-\rho)=\frac{2}{k}(\epsilon_0^{1/2}-\epsilon^{1/2}). \quad (5)$$

We have chosen to present the data in a way which uses Eq. (5), by plotting a Lindhard dimensionless range parameter $(\rho_0-\rho)$ as a function of the dimensionless velocity difference $(\epsilon_0^{1/2}-\epsilon^{1/2})$. Shown also as dotted lines in Fig. 5 are the theoretical slopes $2/k$ as calculated from Eq. (4). It can readily be seen that in general the slopes are not in agreement with the theory. The discrepancy is roughly the same as has been reported by Noshkin,[19] Aras *et al.*,[20] and Mulás and Axtmann.[21]

III. DISCUSSION

One may differentiate the curves shown in Fig. 5, solving for an experimental value of the proportionality factor k in Eq. (5). If one plots the ratio of $k_{\rm exp}=2(\epsilon_0^{1/2}-\epsilon^{1/2})/(\rho_0-\rho)$ to $k_{\rm th}$ as given by Eq. (4), the ratio shows a significant dependence on the dimensionless range parameter $(\rho_0-\rho)$. For all absorbing gases, the behavior pattern is the same. For the light fragment, the ratio of $k_{\rm exp}/k_{\rm th}$ rises to a broad maximum and then monotonically decreases as the fragment approaches the end of its range. For the heavy fragment, only a monotonic decrease in the ratio is seen. This behavior is very suggestive: Since it is the same for all absorbing gases, it must be attributed to some property of the fragment rather than to the absorbing medium.

Lindhard has pointed out[1–3] that the constancy of the proportionality factor k in Eq. (4) cannot be expected

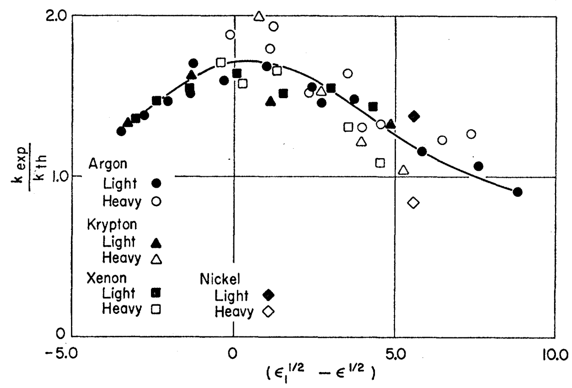

FIG. 6. The ratio of the measured proportionality factor $k_{\rm exp}$ to the constant $k_{\rm th}$ as given by the Lindhard treatment (Refs. 1–3), as a function of the difference from the critical velocity as described in the text. This ratio appears to be independent of both the fragment mass and the atomic mass of the absorbing gas, for the heavier gases.

over the whole range of energy. When the velocity of the particle exceeds a certain critical velocity $v_1=Z_1^{2/3}e^2/\hbar$, where Z_1 is the nuclear charge of the particle, then the electronic slowing is no longer proportional to the velocity of the particle. The slowing instead goes through a maximum and thereafter decreases as approximately v^{-2} (following the stopping formula of Bethe[28–30]). This velocity v_1 is the velocity of the most strongly bound electron of the incoming particle, treated statistically as a Thomas-Fermi atom. Whenever the velocity of the particle exceeds the critical velocity v_1 to any significant extent, then the usual electronic capture and loss equations no longer apply; the particle can instead be treated as a nucleus which is completely stripped of electrons.

For fission fragments, the above considerations are not strictly applicable, since the critical velocity v_1 corresponds to energies of 0.5–1 GeV. These ideas do, however, suggest a mechanism for the discrepancy between experiment and theory. All the theoretical approaches to the stopping of fission fragments in matter are based on treating both the fragment and the atoms of the stopping medium as Thomas-Fermi atoms. The velocity distribution of electrons in a Thomas-Fermi atom is smoothly varying and monotonic. In actual nuclei, this is not the case; the velocity distribution of electrons has rather pronounced maxima and minima, corresponding to the binding energies of the electronic shell structure.

For fission fragments, the charge on the fragment is such that the electronic stopping of the lighter fragment corresponds to capture and loss of M-shell electrons in the fragment. For the heavier fragments, over

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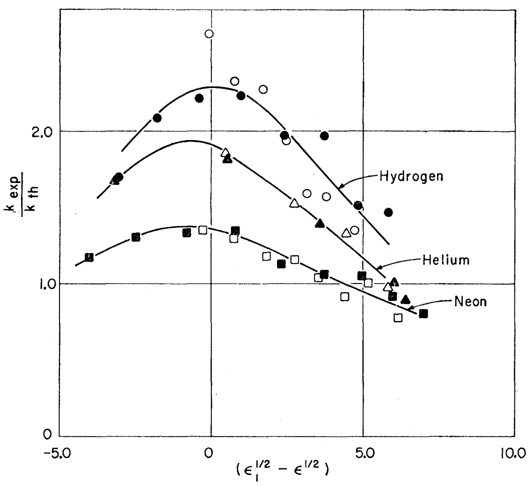

FIG. 7. The ratio of the measured proportionality factor k_{exp} to the constant k_{th} as given by the Lindhard treatment (Refs. 1–3), as a function of the difference from the critical velocity as described in the text. This ratio appears to be independent of the fragment mass but not of the atomic mass of the light absorbing gases.

most of the range the appropriate charge on the fragment corresponds to N-shell electrons. We therefore define a critical velocity v_1' as the velocity of the most energetic electron, not in the entire atom, but in the electronic shell in question. The energy of the median fragment having this velocity is in the region of 57 MeV for both the heavy and the light fragment. (In estimating the critical velocity, we have assumed constant charge density in fission, to give nuclear charges of 37.4 and 54.6 for the average light and heavy fragment, respectively, from ^{235}U fission, and have then used M_1 and N_1 shell binding energies for these charges to calculate appropriate electron velocities.)

In Fig. 6 is plotted the ratio k_{exp}/k_{th}, for energy loss of fission fragments in heavy noble gases, as a function of the dimensionless velocity difference $(\epsilon^{1/2}-\epsilon_1^{1/2})$, where ϵ_1 is calculated from Eq. (1) with $E=57$ MeV, It is of interest to note that for the rate of energy loss in heavier gases—argon, krypton, and xenon—as well as for nickel foil, the data appear to follow a single curve which describes both the heavy and the light fragment. For the lighter gases—hydrogen, helium, and neon—there is a departure from the behavior common to the absorption in heavier gases. Data for the lighter gases are shown in Fig. 7. It may be noted that Bell[31] treated absorption of fission fragments in hydrogen and helium as special cases. We find that neon appears to need special consideration as well. [Following Bell, we have assumed $Z_{eff}=1.2$ for hydrogen atoms in diatomic hydrogen, in converting to the dimensionless range and energy parameters as described in Eqs. (1) and (2).]

The above approach, in particular the derivation of a "universal" curve in Fig. 6 for the stopping of fission fragments in heavy materials, has been shown to be valid for only the average light and heavy fragments of ^{235}U fission. One further test has been made: We have calculated the changes in shape of the typical contours as a function of gas pressure, and find good qualitative agreement with the experimentally observed data in Figs. 3 and 4. In making the calculations, it was assumed that the critical velocity parameter $\epsilon_1^{1/2}$ varies with nuclear charge as the square-root of the binding energy which gives a dependence of approximately $Z^{3/2}$ for the M and N shells over the region of interest. It was also assumed that "light" fragments were by definition any fragment with $Z<46$, where Z was calculated under the assumption of constant charge density in fission. The agreement of the calculated contours with the experimentally observed ones must be to some extent fortuitous. For certain mass splits (involving the heavier of the light fragments and the lighter of the heavy fragments) it must be necessary to take into account both the M- and N-shell electrons. Our data do not give any information on such cases, since we cannot tell whether the heavy-fragment stopping begins to decrease as the fragment velocity exceeds the N-shell electron velocity. Perhaps further range-energy experiments, with californium fission or with accelerated heavy ions, could be used to extend the present data to significantly higher energies.

ACKNOWLEDGMENTS

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