Fine Structure in the Velocity Distributions of Slowed Fission Fragments*

R. B. LEACHMAN AND H. W. SCHMITT
Los Alamos Scientific Laboratory, Los Alamos, New Mexico
(Received August 13, 1954)

Fine structure is observed in the velocity distribution of U^{235} fission fragments that have been slowed by aluminum or nickel, but none is observed for unslowed fragments. Identification of this velocity fine structure with the fine structure in the fission mass yield confirms the influence of the 82-neutron shell in the fission act, as distinguished from its influence in post-fission neutron boil-off. The production of velocity fine structure by aluminum or nickel slowing is shown, by means of the measured range-velocity relation of fragments in these metals, to be the result of isobaric velocity bunching. The measured range-velocity relation of fragments in gold indicates no velocity bunching and therefore is consistent with the absence of velocity fine structure in measured velocity distributions of U^{235} fragments slowed in gold or platinum.

The absence of velocity fine structure in the velocity distribution of U^{233} and Pu^{239} fragments slowed by aluminum is discussed.

I. INTRODUCTION

FINE structure in the mass yield of the heavy-fragment group from U^{235} fission has been observed by Thode *et al.*[1–3] with mass spectograph measurements and by Pappas[4] by radiochemical measurements as an increase in the abundance of mass ~134 over the smooth mass yield curve. Mass spectograph measurement of the mass yield of the light-fragment group by Glendenin *et al.*[5] shows similar fine structure at the complementary mass ~100. The similarity of the fine structure peaks indicates the nuclear shell influence on the fission act itself, but the possibility of an additional influence on the post-fission neutron boil-off from the heavy-fragment group cannot be excluded from these data. The position of these fine structures at masses ~101 and ~135 in the primary-fragment (before neutron boil-off) yield indicates that the closed 82-neutron shell influences the fission act. It can be shown that, for the unstable fragments before decay, the 82-neutron shell is closed at a mass of approximately 135.

In the present work, the influence exerted by shell effects on the energy and velocity distributions of the fragments is of interest. The fission process is regarded initially as the separation of two fragments of equal and opposite momentum. The subsequent neutron emission from the moving fragments in various numbers and directions and with various energies disperses any fine structure initially existing in the distribution of fragment mass, velocity, or energy. The observed mass distribution is dispersed by the variation in the number of emitted neutrons. Further, it can be shown that the distribution in the fragment recoil momentum, resulting from neutron emission, causes a relatively greater spread in the velocity or energy distribution.

It is informative to neglect these dispersions and also any dispersion arising from the measurement and consider the quantities that must be measured to exhibit energy or velocity fine structure. To show fine structure, sufficient information is needed to obtain the mass distribution, for, according to existing information, it is primarily in the mass split of fission that the shell effects exert an influence. Because of the known spread of total kinetic energy[6–10] of the fragments for each fission mode, a coincidence measurement of the velocity or energy of the fragment pairs is needed to obtain the mass distribution from velocity or energy distributions. Thus, observations of energy or velocity of only single fragments give insufficient data to determine the mass distribution and, in view of the spread of masses associated with each fragment velocity, are not expected to follow the mass distribution closely enough to show fine structure. The lack of any fine structure in the good-resolution measurements[11] of the velocities of single fragments from U^{235} is in agreement with this expectation.

Several coincidence measurements[6–10] of fission fragment energies have been made by ionization chambers. Even though sufficient information is obtained to determine the mass distribution, no evidence of fine structure is observed in any of these measurements. A relatively large dispersion, which is presumably imposed by the statistics of the ionization process, was later shown[12] to exist in these energy measurements. This dispersion can be shown to be sufficient to eliminate any fine structure in these coincidence measurements of energy.

* This document is based on work performed under the auspices of the U. S. Atomic Energy Commission.

[1] H. G. Thode and R. L. Graham, Can. J. Research **A25**, 1 (1947).
[2] Macnamara, Collins, and Thode, Phys. Rev. **78**, 129 (1950).
[3] Wiles, Smith, Horsley, and Thode, Can. J. Phys. **31**, 419 (1953).
[4] A. C. Pappas, Massachusetts Institute of Technology, Laboratory for Nuclear Science Technical Report No. 63, 1953 (unpublished).
[5] Glendenin, Steinberg, Inghram, and Hess, Phys. Rev. **84**, 860 (1951).
[6] W. Jentschke and F. Prankl, Z. Physik **119**, 696 (1942).
[7] W. Jentschke, Z. Physik **120**, 165 (1943).
[8] Flammersfeld, Jensen, and Gentner, Z. Physik **120**, 450 (1943).
[9] M. Deutsch and M. Ramsey, U. S. Atomic Energy Commission Report MDDC-945, 1945 (unpublished).
[10] D. C. Brunton and G. C. Hanna, Can. J. Research **A28**, 190 (1950).
[11] R. B. Leachman, Phys. Rev. **87**, 444 (1952).
[12] R. B. Leachman, Phys. Rev. **83**, 17 (1951).

In this work it will be shown that the greater resolution of measurement afforded by observations of velocities by a time-of-flight method allows observation of the fine structure in the velocity distribution of U^{235} fragments, provided that the velocities of the fragments are decreased in such a way as to decrease the velocity spread associated with each fragment isobar. This results in an approach to the condition in which each fragment mass has associated with it a unique velocity. When such a condition is approached, the resulting velocity distribution follows the structure of the mass yield curve.

Regardless of this isobaric velocity (and energy) bunching, however, ionization-chamber measurements of slowed-fragment energies do not give any indication of fine structure.[13] Again, it can be shown that the large dispersion which apparently is inherent in ionization measurements prevents the observation of fine structure.

II. EQUIPMENT

In the present measurements fragments were slowed through thin metal foils on one side of which the fissile material had been deposited. The residual velocity of these fragments was measured, as shown schematically in Fig. 1, by their time of flight through an evacuated drift tube. The drift length for each absorber was chosen so that the drift times were roughly the same for all absorbers. The fragment drift times were recorded and the data analyzed by a method essentially the same as that previously described.[11] The method of determining the resolution of the velocity measurements was similarly described. Fission was induced by thermal neutrons from the Los Alamos homogeneous reactor. The background in the data was caused by pulses from the detector nearer the source foil followed by chance pulses from the remote detector. This background was measured with Po^{210} alpha sources substituted for the fissile foil in the beam from the reactor.

In the present efforts to observe fine structure in the velocity distribution of fragments it was important to maintain good velocity resolution both in measurement and in the slowing process. For the latter, particular attention was given to the uniformity of the slowing

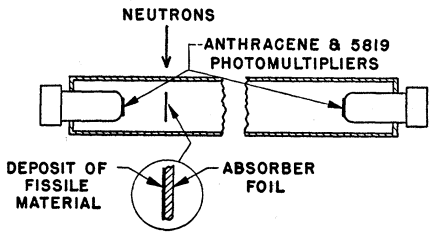

FIG. 1. Schematic diagram of the apparatus used to detect the time of flight of slowed fission fragments. Fragments were slowed by the metal backing of the fissile material. The longest of the drift lengths used was 199 cm and the shortest was 125 cm.

[13] H. W. Schmitt, dissertation, University of Texas, 1954 (unpublished).

TABLE I. Characteristics of the foils used to slow fission fragments. The numbers following the thickness nonuniformities represent (1) measurement of the foil used for slowing fragments and (2) measurement of an equal area of the foil neighboring the area used for velocity measurements.

Foil material	Thickness (mg/cm²)	Foil type	Method of U or Pu deposition	Foil diameter (in.)	Standard deviation of thickness (%)	Standard deviation of velocity (10^8 cm/sec)
Al	1.1	rolled	painted	1.5	0.93 (2)	0.023
Al	1.8	rolled	painted	1.5	2.0 (2)	0.060
Ni	1.1	plated	painted	1.5	2.7 (2)	0.050
Ni	2.6	plated	painted	1.5	3.9[a]	0.123
Au	5.15	evaporated	painted	1.0	0.90 (1)	0.042
Pt	5.1	rolled	evaporated	1.5	2.5 (2)	0.117

[a] One 1.24-mg/cm² foil and one 1.32-mg/cm² foil; uniformities measured by methods (1) and (2), respectively.

foils. Both thickness and uniformity were measured by means of a specially designed beta-ray thickness gauge. This gauge consisted of a 1.8-mm collimated beam of beta particles from a C^{14} source and a Geiger tube. Standard absorbers of the same element to be measured were used to calibrate the absorption of the beam and this calibration was combined with the beam attenuation at various positions on the unknown foil to determine uniformity. Tabulated in Table I are the characteristics of the foils used and the uniformity of these foils. The method for obtaining the velocity dispersion resulting from foil nonuniformity is explained in Sec. IV.

III. ISOBARIC VELOCITY BUNCHING

Velocity bunching occurs for a given fragment mass in slowing materials of which the absorption characteristics are such that the faster fragments of that mass are slowed more than the slower fragments. In this manner the spread of velocities associated with each fragment isobar will be decreased as the fragments are slowed. In view of the complexities in the velocity loss process of fission fragments in matter, we shall consider the loss expressed simply by the somewhat arbitrary expression,

$$dv/dx = -Av^n, \tag{1}$$

where, for each absorber, A and n are considered to be functions of only the fragment mass, the velocity v is in (10^8) cm/sec, and the thickness x is in mg/cm². It can be seen that isobaric velocity bunching occurs only when $n>0$.

Determinations[14–18] of the range-velocity relation needed for the evaluation of A and n in Eq. (1) have been made for several slowing elements. Those of Demers[18] are for slowing in photographic emulsion

[14] Bøggild, Brostrøm, and Lauritsen, Kgl. Danske Videnskab. Selskab, Mat. fys. Medd **18**, No. 4 (1940).
[15] R. Sherr and R. Peterson, Rev. Sci. Instr. **18**, 567 (1947).
[16] Bøggild, Arrøe, and Sigurgeirsson, Phys. Rev. **71**, 281 (1947).
[17] N. O. Lassen, Kgl. Danske Videnskab. Selskab, Mat. fys. Medd. **25**, No. 11 (1949).
[18] P. Demers, Can. J. Phys. **31**, 78 (1953).

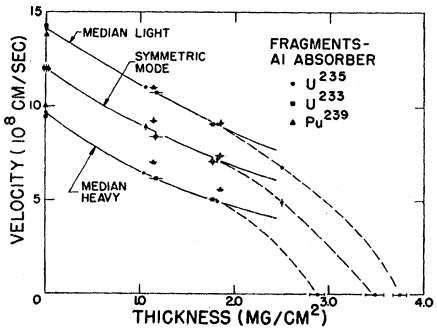

FIG. 2. Range-velocity data of fission fragments in aluminum. The velocities of unslowed fragments from U^{233} and Pu^{239} fission are from reference 11 and the end-point ranges are obtained from references 19 and 20. Solid curves are fits of the equation $dv/dx = -Av^n$ to the unslowed, 1.1-mg/cm², and 1.8-mg/cm² datum points of U^{235} fragments. The dashed curve represents experimental results. When no uncertainties are indicated they are smaller than the datum point.

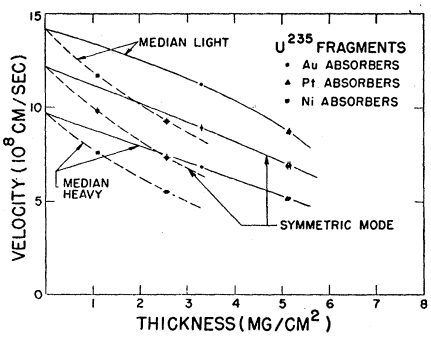

FIG. 3. Range-velocity data of fission fragments in nickel, gold, and platinum. A 3.3-mg/cm² gold foil was used in addition to the foils described in the text. Curves are fits of the equation $dv/dx = -Av^n$ to the data of nickel and gold. When no uncertainties are indicated they are smaller than the datum point.

and those of Sherr and Peterson[15] are known to be inaccurate near the beginning of the fragment range. Except for the end of the fragment range, which is not of importance in the present considerations, the data of Lassen are best fitted with a negative n for light fragments in all the slowing gases used; for heavy fragments in hydrogen and deuterium n is positive and for heavy fragments in argon n is zero. The data of Bøggild *et al.*[14,16] are consistent with the data of Lassen. From this limited information it appears that heavy fragments slowed in materials of low nuclear charge will undergo velocity bunching.

IV. VELOCITY LOSSES OF FRAGMENTS

In Figs. 2 and 3 are plotted the range-velocity data obtained from the present velocity measurements. These measurements are described in greater detail in the next section. Because of the uncertainties associated with any correlation of fragment velocity with fragment mass, the velocity distributions were used in this section only to obtain the median velocities of the heavy- and light-fragment groups and, with less accuracy, the velocity of the least probable yield, which corresponds to the symmetrical fission mode. For each of the mass groups and for each absorber, three range-velocity datum points are used in Eq. (1) to solve for A and n. The values of n are given in Table II.

TABLE II. The exponent n from range-velocity data of fission fragments. Data of Figs. 2 and 3 are fitted by the relation $dv/dx = -Av^n$. Uncertainties in the symmetrical fission data make the symmetrical fission results of n relatively uncertain. The n value for light fragments of Pu^{239} is similarly uncertain.

Fission source	Absorber	n for median heavy fragment	n for fragments from symmetrical fission	n for median light fragment
U^{235}	Al	1.3	1.2	0.6
U^{233}	Al	1.4	1.4	0.3
Pu^{239}	Al	0.9	−0.4	−0.4
U^{235}	Ni	0.9	1.0	1.4
U^{235}	Au	−0.1	−0.3	−1.7

The velocity dispersion resulting from the absorber nonuniformities is determined from these range-velocity relations and the magnitude of the foil nonuniformities. Equation (1) with the appropriate A and n values is used with the absorber nonuniformities of Table I to obtain the velocity dispersions listed in Table I.

It is to be emphasized that Eq. (1) is not necessarily expected to be a true representation of the actual range-velocity relation. The inadequacy of Eq. (1) in describing the range-velocity relation near the end of the fragment range is seen in Fig. 2, where points corresponding to a 2.5-mg/cm² aluminum absorber and to the end points of the fragment ranges[19,20] have been included. However, over the absorber thicknesses used in the velocity measurements to be discussed, Eq. (1) is an adequate approximation for the study of velocity bunching and velocity fine structure.

From the positive values of n in Table II velocity bunching of U^{233} and U^{235} fragments in aluminum, and of U^{235} fragments in nickel, is anticipated. Similarly, velocity bunching of the heavy Pu^{239} fragments in aluminum is anticipated. In contrast, the negative value of n for U^{235} fragments in gold implies a velocity expansion for fragment isobars rather than velocity bunching. On this basis, no fine structure is expected to be observed in the velocity distribution of U^{235} fragments slowed in gold. Because of the similarity of gold and platinum in respect both to the measured range-velocity data of Fig. 3 and to expected behavior in fragment slowing, it is expected that n is negative also for platinum. The similarity of the n values for the heavy fragments from U^{233}, U^{235}, and Pu^{239} is not unexpected in view of the close similarity of the mass distributions[21] of these heavy-fragment groups.

[19] F. Suzor, Ann. phys. 4, 269 (1949).
[20] Finkle, Hoagland, Katcoff, and Sugarman, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 46, National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV.
[21] See reference 19; appendix B.

It is noted in Table II that the decrease in the heavy-fragment value of n with the nuclear charge of the slowing material is consistent with the velocity data of Lassen.[17]

To illustrate the effect of isobaric velocity bunching we now calculate the velocity loss of each U^{235} fragment slowed by two different aluminum thicknesses. These calculations require the use of Eq. (1), where A and n values as functions of fragment mass are needed. For simplicity, the A and n values are assumed to be linear functions of the primary mass M of each fragment group. This yields $n=2.29(10^{-2})M-1.55$ and $A=-1.71(10^{-2})M+2.23$ for the light fragment group and $n=4.26(10^{-3})M+0.647$ and $A=1.07(10^{-3})M+0.080$ for the heavy fragment group.

These approximations of A and n as functions of mass are used in Fig. 4 with Eq. (1) and the probability $P(v,M)$ of occurrence as a function of mass and unslowed velocity. These latter data are obtained from a transformation we have made of the U^{235} data of Brunton and Hanna,[10] which give probability as a function of light- and heavy-fragment energies. However, before this transformation was made, 6.7 Mev was added to the heavy-fragment energies and 5.7 Mev to the light-fragment energies to bring the resulting velocities into agreement with measured unslowed velocities. It is to be noted that the transformed data of Fig. 4 do not indicate a preference for the primary masses ~101 and ~135, just as the energy data from which these contours were obtained did not show this preference. On the basis of the discussion of Sec. I, it must be assumed that the resolution of these original energy data was inadequate to resolve the fine structure which is known to exist. Similarly, the dispersion in the energy data is believed to have led to an increase in the velocity spread beyond the actual spread for each fragment isobar.

Velocity bunching is seen in Fig. 4, especially in the heavy-fragment group, as a contraction of the probability contours with increasing absorber thickness. Even the illustrated spread of isobaric velocities of the preferred mass ~135 decreases sufficiently to resolve the velocities of this mass, for example, from the velocities of the most probable mass ~140. In addition to the effect of velocity bunching, the increasing inclination of the light-fragment contours with absorber thickness aids in resolving the velocities of the complementary light-fragment masses. The observation, however, of the preferred mass ~101 as a velocity fine structure is difficult for the following reasons: the smaller n value for light fragments results in less velocity bunching in the light-fragment group and, further, the resolution of the velocity measurements is seen in the next section to be poorer for light fragments than for heavy fragments.

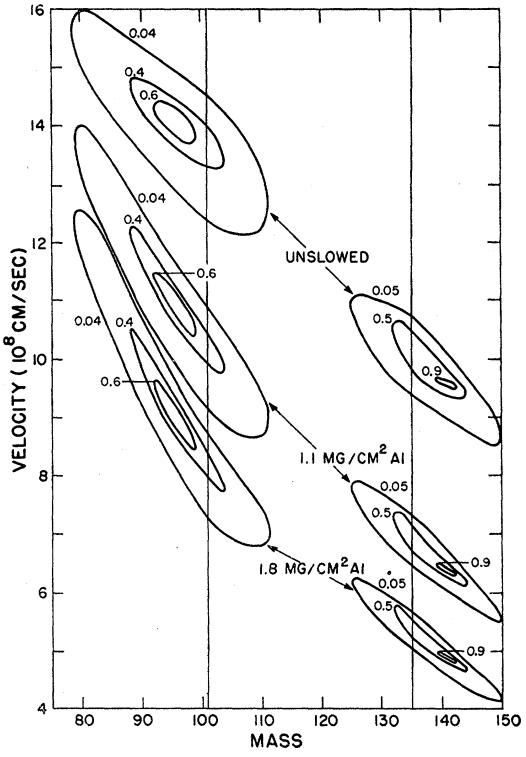

FIG. 4. Illustration of the contraction of mass-velocity probability contours for the slowing of U^{235} fragments by aluminum. The position of the known high yields at primary masses 101 and 135 are indicated by vertical lines. The probability contours of the unslowed fragments are obtained from a transformation of the data of reference 10 with the two groups normalized so that the integrated probability $\int\int P(v,M)dvdM$ for each group is the same. Velocity losses were calculated from the equation $dv/dx=-Av^n$, where A and n are given in Sec. IV of the text. The maximum $P(v,M)$ for each of the three light-fragment distributions are equal. Similarly, the maximum $P(v,M)$ for each of the three heavy-fragment distributions are equal.

V. VELOCITY DISTRIBUTIONS OF SLOWED FRAGMENTS

(a) U^{233}, U^{235}, and Pu^{239} Fragments; Al Absorbers

The velocity distributions of U^{235} fragments slowed by the aluminum thicknesses used in the calculations of the previous section were measured. They are shown in Fig. 5 in comparison with the velocity distribution of these fragments before being slowed. The latter measurement is a repeat of a previous measurement[11] and agrees with it within statistics. The expected fine structure in the heavy-fragment (low-velocity) group appears as a step on the high side of the slowed heavy-fragment distributions. Although smooth curves are drawn for the light-fragment (high-velocity) group, small but inconclusive indications of fine structure exist in the datum points on the low side of the slowed light-fragment distributions. As seen in Fig. 5, the decrease in velocity resolution with increasing velocity makes the observation of light-fragment fine structure more difficult than that of heavy-fragment fine structure.

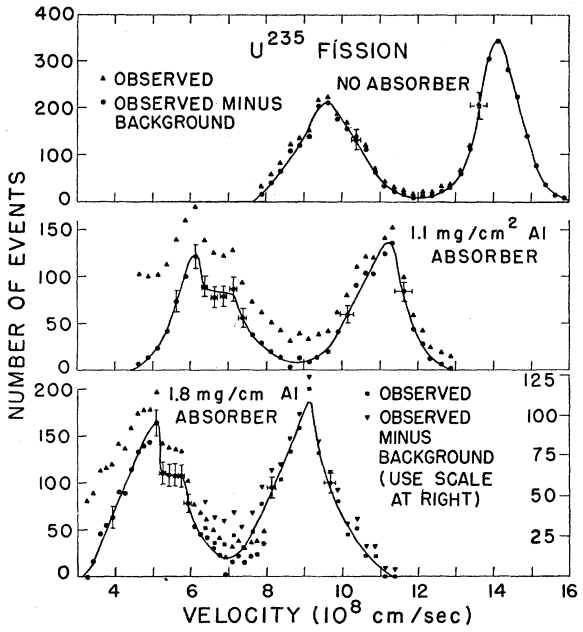

FIG. 5. Velocity distributions of unslowed and aluminum-slowed fragments from U^{235} fission. Horizontal wings through datum points represent the standard deviations associated with measurement of velocity. Different drift lengths were used in separate measurements of the heavy- and light-fragment groups for 1.8-mg/cm² aluminum absorber to improve the velocity resolution of the light fragment group.

Calculations, the results of which are shown in Fig. 4, predict that the fine structure in the velocity distribution should become more pronounced with increasing absorber thickness. However, no growth in the observed fine structure is noted in Fig. 5. The expected growth is presumably canceled by the greater velocity dispersion associated with the thicker absorber relative to the thinner absorber; the velocity dispersion for the 1.8-mg/cm² aluminum listed in Table I is appreciable compared with the velocity width of fine structure while that for the 1.1-mg/cm² aluminum absorber is small.

Mass fine structure, even after dispersion by the variation in the number of emitted neutrons, is about two mass numbers wide and so appears as a sharp spike on the mass distribution. Because of the larger dispersions associated with the velocity measurement of fine structure, on the other hand, the width of the velocity fine structure shown in Fig. 5 is appreciable compared with the width of the distribution. These dispersions in the velocity fine structure arise from measurement, from slowing, and from recoil momentum in neutron emission. It is doubtful if such a low-resolution detection of velocity fine structure would be seen if it occurs very near the peak of a velocity distribution. Accordingly, the velocity fine structure of U^{235} fragments is observed only because it occurs on the side of the velocity distribution.

Mass yield measurements indicate that the primary mass 135 occurs slightly nearer the center of the mass peak for U^{233} fission than for U^{235} fission[21,22] and still nearer the center for Pu^{239} fission.[21,23] As a consequence, the apparent lack of fine structure in the heavy-fragment velocity groups of U^{233} and Pu^{239} fragments shown in Fig. 6 may be the result of a relatively low-resolution detection of fine structure near the peak of the general velocity distribution. Since the position of the mass 135 is off the peak of the U^{233} mass distribution almost to the same extent as for U^{235} fission, we believe any fine structure in U^{233} fission that is comparable in magnitude to that of U^{235} fission would be observed in our velocity data. On this basis, the present results are in accord with those of Fleming *et al.*,[22] which indicate a smooth heavy-fragment mass yield from U^{233} fission. They are not, however, in accord with the results of Steinberg *et al.*,[24] which indicate mass fine structure in the light-fragment group. The failure of the velocity distribution to show the heavy-fragment fine structure[23] in the mass yield of Pu^{239} fission is attributed to the smaller magnitude of the fine structure compared to that of U^{235} and to its position near the peak of the general distribution.

These U^{233} and Pu^{239} data were taken under the same

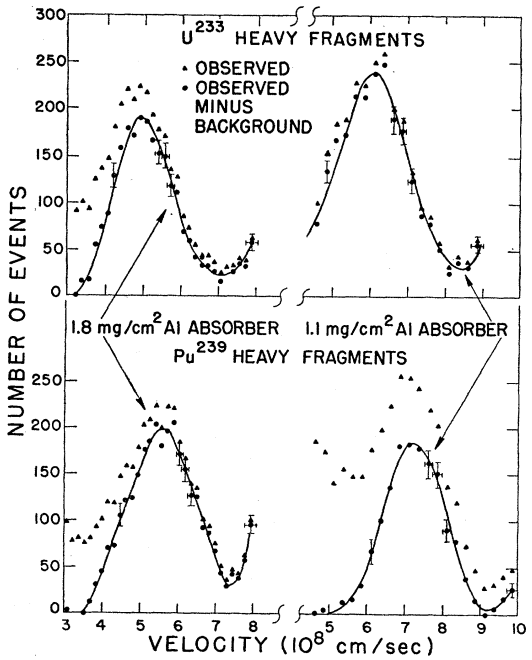

FIG. 6. Velocity distributions of U^{233} and Pu^{239} fragments slowed by aluminum. Horizontal wings through datum points represent the standard deviations associated with measurement of velocity.

[22] Fleming, Tomlinson, and Thode, Can. J. Phys. **32**, 522 (1954). The authors are indebted to Professor R. H. Tomlinson for information on these results and those of reference 23 in advance of publication and for suggesting the present measurements with U^{233} fragments.

[23] Fleming, Wanless, and Tomlinson, 1954 (private communication).

[24] Steinberg, Glendenin, Inghram, and Hayden, Phys. Rev. **95**, 867 (1954).

conditions of velocity bunching as the U^{235} data. With our method of measurement, fine structure is difficult to observe in the light-fragment distribution and so in these and the following data only the heavy-fragment data are included.

The possibility that velocity fine structure is the result of a discontinuity in the stopping power as a function of mass can be excluded by comparison of the velocity distributions of slowed fragments of U^{233} and U^{235} fission. The close similarity of these heavy fragments in mass, velocity, and nuclear charge results in essentially the same velocity loss characteristics for both. Thus, since the velocity distributions of slowed fragments from U^{233} fission are smooth and those of slowed fragments from U^{235} fission contain fine structure, the appearance of the velocity fine structure cannot be explained on the basis of a discontinuity in stopping power.

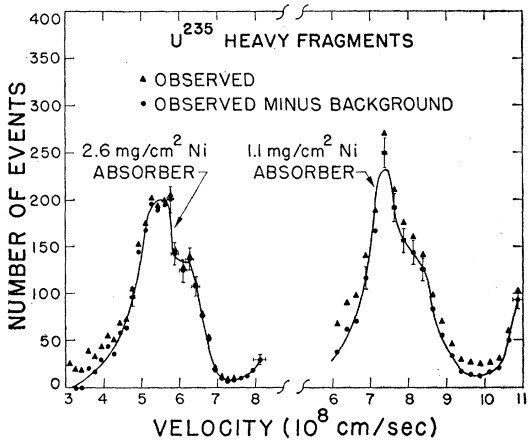

FIG. 7. Velocity distributions of U^{235} fragments slowed by nickel. Horizontal wings through datum points represent the standard deviations associated with measurement of velocity.

(b) U^{235} Fragments; Ni, Au, and Pt Absorbers

Slowed-fragment velocity distributions obtained when U^{235} fragments were slowed by nickel absorbers are similar to those obtained when U^{235} fragments were slowed by aluminum absorbers. These data of the heavy-fragment group for two nickel thicknesses are shown in Fig. 7. Although smaller than in the case of aluminum absorption, the velocity fine structures in these nickel data, when considered together, are well established. Indeed, fine structure is expected on the basis of the positive n values in Table II. The smaller

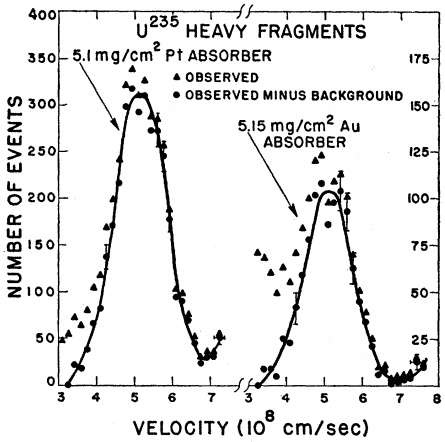

FIG. 8. Velocity distributions of U^{235} fragments slowed by platinum and gold. Horizontal wings through datum points represent the standard deviations associated with measurement of velocity.

extent of the velocity fine structure in these nickel data compared to aluminum data is attributed to three factors: For both nickel thicknesses, the smaller n values of Table II lead to less velocity bunching than for equivalent slowing by aluminum. For both nickel thicknesses, but especially in the case of the thicker absorber, the velocity dispersion from foil nonuniformity as listed in Table I is appreciable compared with the width of the velocity fine structure. Finally, the thinner nickel absorber is not thick enough to produce adequate velocity bunching.

In order to facilitate comparison of velocity data, foils of gold and platinum were selected to have similar stopping powers and somewhat smaller velocity dispersions compared to the thicker aluminum and nickel foils. As seen in Table I, the gold and aluminum foils are approximately equivalent in these respects, as are the platinum and nickel absorbers. The measured velocity distributions of heavy fragments from U^{235} fission slowed in these gold and platinum foils are shown in Fig. 8. The lack of fine structure in these velocity distributions is consistent with the negative n values for gold and platinum.

VI. ACKNOWLEDGMENTS

The authors are indebted to Miss E. Pierce for reading much of the velocity data and to Mr. W. D. Schafer for construction of the thickness gauge and for measurements with it.