Coulomb barrier, and no $Al^{28}$ activity was observed until a neutron energy of 4.5 Mev was reached. Below this energy, the cross section is $\lesssim 2$ mb, the lower limit which could be detected by the method used.

The neutron total cross section of silicon has been measured from 3 to 12 Mev with a resolution of about 10% by Nereson and Darden,[10] who find two broad resonances at 4.8 and 6.0 Mev. Above these energies the value of the cross section is approximately constant at 1.7 barns. The $Si^{28}(n,p)Al^{28}$ cross section does not indicate resonances at either 4.8 or 6.0 Mev, but the low value of the $(n,p)$ cross section near 4.8 Mev and the general rise due to penetrability would tend to obscure the effects of broad resonances in these regions. It is not difficult to understand why the total cross-section data failed to show the 0.1-Mev wide resonances, since the resolution was approximately 0.6 Mev in this energy region. Even the peak at 7.45 Mev would have been averaged over, despite its 0.3-Mev width.

[10] N. Nereson and S. Darden, Phys. Rev. **89**, 775 (1952).

TABLE I. Resonances in the $Si^{28}(n,p)Al^{28}$ reaction.

| Neutron energy (Mev) | Peak cross section (barns) (±50%) | Γ (Mev) |
|---|---|---|
| 5.14 (?) | (0.03) | |
| 5.62±0.05 | ≥0.09 | ≲0.1 |
| 6.51±0.05 | ≥0.28 | ≲0.1 |
| 6.81±0.05 | ≥0.35 | ≲0.1 |
| 7.45±0.10 | 0.37 | ≈0.3 |

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# Range and Range Dispersion of Specific Fission Fragments

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The range and dispersion in range of specific $U^{233}$ and $U^{235}$ fission fragments; namely, those associated with delayed-neutron emitters, have been determined in gases of different atomic number. These measurements give the $Z$-dependence of the range and range dispersion for fragments associated with a specific mode of fission. After corrections for foil thickness and geometry the $Z$-dependent part of the range dispersion which arises from the nuclear stopping process was determined and values were obtained for the residual component corresponding to the energy dispersion associated with the fission process itself. The values thus obtained for this energy dispersion are found to be in fair accord with recent theoretical results of Fong.

## INTRODUCTION

MEASUREMENTS have been made of the range and dispersion in range of specific fission fragments in various stopping gases. From the range dispersion it has been possible to obtain information on the dispersion in the energy release accompanying fission and these results can now be compared with some recent theoretical work of Fong.[1] Experimentally, the dispersion in energy accompanying a given mode of fission has been given in the literature only indirectly. The fission-product energy spectra measurements of Brunton and Hannah,[2] Demmers,[3] and Leachman,[4,5] and others involve the gross fission process. The dispersion in energy observed in these experiments is therefore partly associated with the variation in energy release accompanying different modes of fission. Another class of experiments employs the measurements of range. Katcoff[6] has measured the dispersion in range corresponding to what might well be ranges of fragments of a unique mode of fission. These measurements, however, were made in air only and complete reliance must be placed upon the theory of the stopping of fission fragments to derive from the dispersion in range the dispersion in the energy of the given fragment. Boggild[7] has measured the fission fragment ranges and the dispersion in the range in gases of different atomic weights but the measurements were made upon gross fission fragments. It seems thus not untimely to present now some measurements made in 1948 which have a somewhat more direct bearing than the experiments just mentioned upon the question of the dispersion in the energy of a given fragment associated with a single mode of fission. For various reasons these results have not previously been presented in detail; some results have, however, appeared in abstract.[8]

The experiment here described consisted of measuring

[1] P. Fong, (private communication).
[2] D. C. Brunton and G. C. Hannah, Can. J. Research **A28**, 190 (1950).
[3] P. Demers, Can. J. Phys. **31**, 78 (1953).
[4] R. B. Leachman, Phys. Rev. **83**, 17 (1951).
[5] R. B. Leachman and H. W. Schmitt, Phys. Rev. **96**, 1366 (1954).
[6] Katcoff, Miskel, and Stanley, Phys. Rev. **74**, 631 (1948).
[7] Boggild, Arroe, and Sigurgeirsson, Phys. Rev. **71**, 281 (1947).
[8] Good, Wollan, and Strauser, Phys. Rev. **74**, 1225 (1948); Good, Campbell, and Strauser, Phys. Rev. **75**, 1292 (1949).

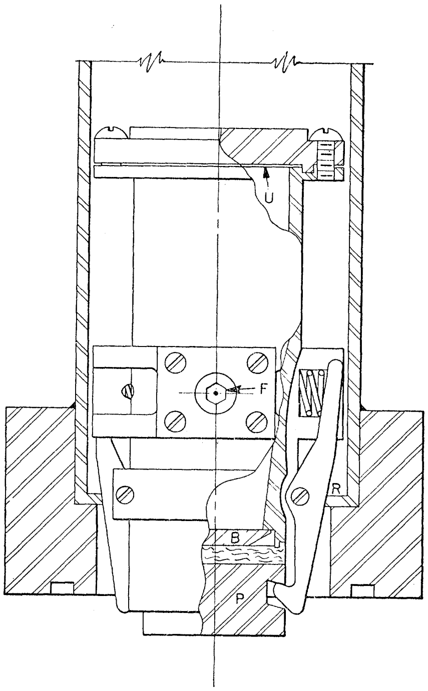


FIG. 1. "Rabbit" near outer terminal of pneumatic tube. *U*—uranium foil, *B*—Bakelite fission fragment catcher button, *P*—sealing plug, *R*—release ring, and *F*—filling valve.

the range and the dispersion in range of those fission fragments associated with the 55-sec, the 22-sec and the 4.5-sec delayed neutron activities. These ranges were measured in gases of different atomic weights for both $U^{233}$ and $U^{235}$.

## APPARATUS AND PROCEDURE

The apparatus for measuring the ranges of the delayed-neutron-emitting fission fragments is represented in part in Fig. 1. This shows a gas-tight chamber, the "rabbit," situated near the outside terminal of a pneumatic tube which extends into the central high-flux region of the ORNL graphite reactor. At the top of the "rabbit," which is made of steel, is a cover plate under which is clamped a 25-mil nickel foil *U* on the bottom side of which is electroplated a thin (0.3 mg/cm$^2$) layer of enriched uranium. This is the source of the fission fragments which are emitted when the "rabbit" is driven pneumatically into the center of the pile.

At the lower end of the "rabbit" is a bakelite fission-fragment catcher button *B* held in position by a steel plug with a bonded rubber disk which gives a gas tight seal when the plug is held in position by the arms shown on the side of the rabbit.

The basic elements of the experiment can now be indicated as follows. The "rabbit" is filled with a gas to a given pressure, it is then sent into the pile where the emitted fission fragments from the fission foil traverse the gas (if the pressure is at a sufficiently low value) and collect on the catcher button. After a proper irradiation time the "rabbit" is sent out, and when the arms strike the release ring *R* the bottom plug *P* is released and the fission-fragment catcher button is discharged into a neutron counter system where the delayed-neutron activity of the button is measured as a function of time.

The saturation activity for a given neutron period is then determined from the measured delayed-neutron decay curve, proper account being taken of irradiation time and the time between the end of irradiation and start of neutron counting. This measured saturation activity for a given gas pressure and delayed-neutron period then corresponds to one point on a curve of the type shown in Fig. 2. A repetition of the process for various gas pressures in the "rabbit" gives the range curve for this particular gas.

An opening on the side of the pneumatic tube assembly allowed access to the gas filling valve shown on the front of the "rabbit." It consisted of a $\frac{1}{4}$ in. thick, pure rubber washer with a hole in the center. This rubber was put under compression by means of an Allen set screw and an Allen wrench down the centers of which were clearance holes for a No. 19 hypodermic needle. This needle was connected by means of a flexible rubber hose to a gas-filling manifold with mercury monometer. A given pressure of gas in the "rabbit" was then attained by inserting the needle into the "rabbit" through the rubber valve and setting the desired pressure on the filling manifold. The needle and Allen wrench were attached to long handles in order to minimize the radiation hazard in the filling process. A small thermocouple concentric with and terminating near the end of the hypodermic needle was used as a check on the equilibrium temperature of the gas. The final temperature was determined by a thermometer embedded in mercury in the pneumatic tube assembly. After equilibrium was reached, the needle was withdrawn and the rubber compressed by means of the Allen screw and Allen wrench. The leakage of the sealed "rabbit" was negligible over several hours.

Attached to the lower end of the pneumatic tube assembly was a box fastened through a funnel shaped bottom to a tube leading into the center of the neutron counter system. This box was attached to a line which sucked the contaminated air from the pneumatic tube when the "rabbit" was being discharged from the pile. A method of handling the seal-off plug in the bottom of the "rabbit" on discharge from the pile was contained within this box. This consisted of a hinged arm with an electromagnet which was located at the end of the pneumatic tube when the "rabbit" was being irradiated in the reactor. Upon discharge of the "rabbit," the bottom plug was released and caught by the electromagnet which then swung out of position to allow the catcher button to fall into the counter system. After counting, the button was retrieved by opening a trap door in the tube in the center of the counter system, it was then replaced by a pair of tongs on top of the "rabbit" sealing plug through an opening in the box, the "rabbit" was clamped in position and by means of a hinged arm the plug and catcher button were brought into position to seal the "rabbit" for the start of a new run.

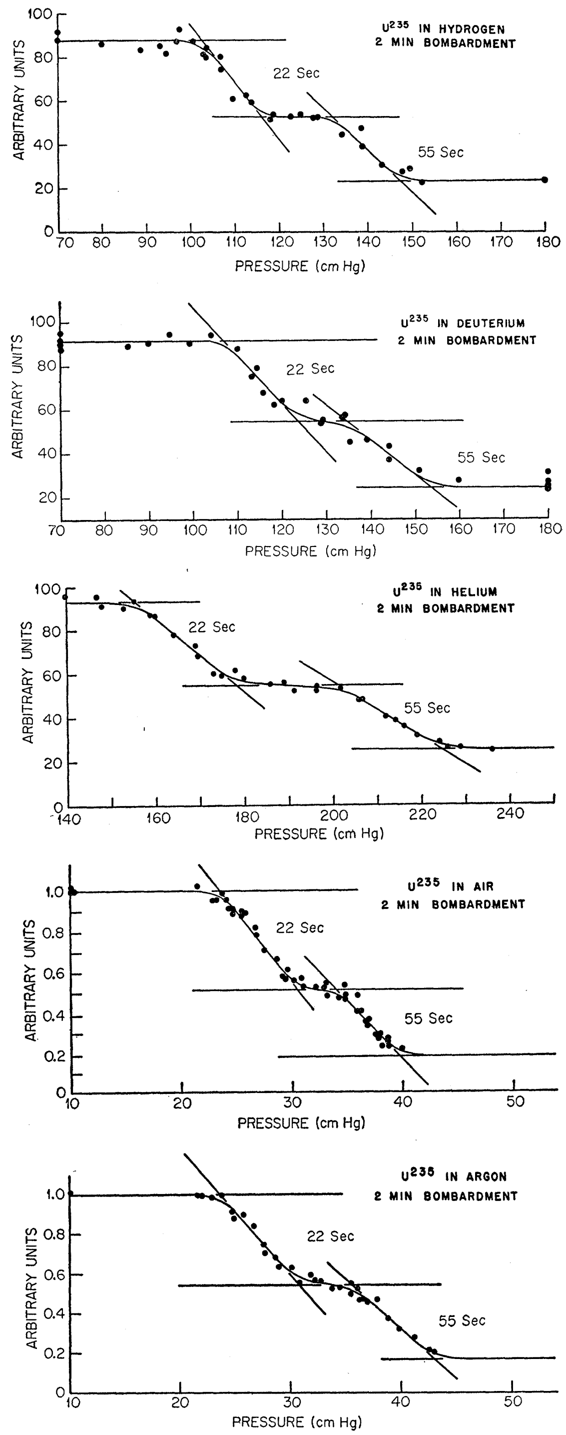


FIG. 2. Counting rates for delayed neutrons from $U^{235}$ fission *versus* gas pressure in "rabbit" for various stopping gases.

TABLE I. Range and range dispersion for light fission fragments.

| Gas | $R_0$[a] | $\rho$(obs)% | $\rho'$%[b] | $\lambda$%[c] |
|---|---|---|---|---|
| $H_2$ | 143.3 | 5.4 | 2.8 | 4.9 |
| $D_2$ | 147.3 | 6.9 | 4.3 | 7.8 |
| He | 218.2 | 5.4 | 2.8 | 4.9 |
| Air | 37.9 | 8.4 | 5.8 | 10.5 |
| Argon | 40.3 | 9.9 | 7.3 | 13.4 |

[a] Observed range ×foil correction factor (1.026), cm Hg NTP.
[b] $\rho' = \rho - 2.6$.
[c] $\lambda = [(1.84\rho')^2 - 2.0]^{\frac{1}{2}}$.

The neutron counting system consisted of a paraffin cylinder about two feet long and one foot in diameter with a two-inch hole along its axis. Around the central tube embedded in the paraffin were located six 1-in. diameter $BF_3$ counters connected in parallel. The catcher button was counted in the center of this assembly, the response of which is known to be relatively independent of neutron energy.

For the success of this method it was necessary to show that the fragments being studied would not stick to the collector button unless they impinged with greater than thermal energy and hence that diffusion played no observable role. That low-energy fragments do not stick to the collector was shown in two ways. It was found (a) that the background counting rate at pressures just exceeding the fragment range pressure was unaffected by the presence of a few micrograms of foil covering the collector button and (b) that the background counting rate at pressures exceeding the fragment range pressure did not increase with arbitrarily long exposures. It was hence concluded that diffusion is not observed because a certain minimum energy is required to make the fragments stick to the bakelite.

## RESULTS AND DISCUSSION

Figure 2 shows the experimental results corresponding to the 55-sec and 22-sec delayed-neutron fragments from the fission of $U^{235}$. Interpretation is made as follows: Horizontal lines are drawn to represent the low-pressure and background counting rates. Judgment as to where to draw the "horizontal line" in the region between the 55-sec and 22-sec groups is assisted for different gases by the knowledge that the ratio of the number of fragments in the 55-sec and 22-sec groups must be the same, obviously, for all stopping gases. Tangents are drawn through the points of maximum slope. Then, if the points of intersection between the line of maximum slope and the adjacent horizontal lines are called $R_1$ and $R_2$, the mean range is given by $R_0 = (R_2 + R_1)/2$ and the straggling parameter is defined as $\rho = (R_2 - R_1)/2R_0 = (R_2 - R_1)/(R_2 + R_1)$. In the following discussion the range distribution is assumed Gaussian. Then $\lambda$, the full percentage width at half-maximum, is related to the straggling parameter $\rho$ by $\lambda = 1.84\rho$. Before discussing the data of Fig. 2

TABLE II. Range and range dispersion for heavy fission fragments.

| Gas | $R_0$[a] | $\rho$(obs)% | $\rho'$%[b] | $\lambda$%[c] |
|---|---|---|---|---|
| $H_2$ | 113.2 | 5.8 | 2.5 | 4.3 |
| $D_2$ | 118.7 | 7.7 | 4.4 | 8.0 |
| He | 172.8 | 6.8 | 3.5 | 6.3 |
| Air | 27.8 | 11.9 | 8.6 | 15.7 |
| Argon | 28.2 | 13.2 | 9.9 | 18.2 |

[a] Observed ranges ×foil correction factor (1.033), cm Hg NTP.
[b] $\rho'=\rho-3.3$.
[c] $\lambda=[(1.84\rho')^2-2.0]^{\frac{1}{2}}$.

it will be well to dispose of those data not explicitly represented therein. Curves of the same type as those of Fig. 2 were obtained not only for 55-sec and 22-sec activities but also for the 4.5-sec activity and for $U^{233}$ as well as for $U^{235}$. The general statement can be made that the ranges for 235 fission are of the order of 2 to 3% greater than the corresponding ranges for 233 fission. In the cases of both $U^{235}$ and $U^{233}$ the 4.5-sec ranges were the same within the experimental error as the 55-sec range. The essential content of the present report is contained in an analysis of the data of Fig. 2 given in Tables I and II.

The observed ranges of the delayed-neutron-emitting fission fragments can be directly compared for the case of stopping in air with the results of Katcoff[6] and a less direct comparison can be made with the data of Sugarman[9] who measured the extrapolated ranges of these fragments in aluminum. For this comparison, the ranges of Sugarman have been reduced to mean ranges in air by the relation

$$R_{\text{mean}}(\text{air}) =R_{\text{extrap}}(\text{Al})\left[1-\frac{\rho_{\text{air}}+\rho_{\text{argon}}}{2}\right]\left[\frac{2R_{\text{air}}}{R_{\text{air}}+R_{\text{argon}}}\right],$$

where the ranges and straggling in air and argon are taken from our data. This relation is justified by the fact that the atomic number of aluminum is approximately the average of that of argon and air. A comparison of these mean ranges in air is given in Table III, and the results are seen to be in good accord. The observed percentage dispersion $\rho_{\text{obs}}$ is assumed to be composed of the range dispersion associated with nuclear stopping, electronic stopping, geometry, and foil thickness, and in addition an initial energy dispersion, including recoil from the prompt neutrons. The dispersion due to foil thickness was determined from an experiment in which the heavy fragment range was studied in helium for different uranium foil thicknesses. The assumption was then made for the heavy fragment that the fission foil represented approximately the same fraction of the observed dispersion for all the gases studied, *viz.*: $H_2$, $D_2$, He, air, A. For the light fragment the fission foil was taken to contribute a smaller amount to the range dispersion in first approximation inversely as the light and heavy particle ranges.

[9] Nathan Sugarman, J. Chem. Phys. **15**, 544 (1947).

The third columns in Tables I and II give the observed "straggling parameter" and the fourth column gives the "straggling parameter" corrected for the fission foil thickness.

For the final values conversion has been made from straggling parameters to full widths at half-maximum, $\lambda$. In this terminology,

$$\lambda^2=(\lambda_{\text{obs}}-\lambda_{\text{foil}})^2-\lambda_{\text{geom}}^2.$$

From the third column of Tables II, III and $\lambda_{\text{geom}}=1.4\%$, values are obtained for $\lambda$, which are listed in columns four of Tables I and II.

These listed values of $\lambda$ contain contributions from nuclear stopping, electronic stopping and from variations in energy in the fission process. From the theory,[10] it is found that the electronic stopping contributes only about 0.2% to the straggling and hence compared to the observed values is entirely negligible. There remains then only the nuclear stopping contribution and the contribution associated with the energy spread in the fission process. The nuclear contribution will depend on the mass of the stopping gas whereas that arising from the fission process will of course be independent of the stopping medium. It is on this basis that an attempt has been made to separate these two effects. This has been done with the aid of the general principles involved in the theory of range and range-straggling. The expression for the range of a fission fragment as given by Bohr[11] can be considered in an approximate sense as consisting of two parts, one in which electronic stopping predominates and one in which nuclear stopping predominates. Bohr's approximate expression for the nuclear range straggling $\lambda_N$ when represented as a percentage of the total range $R$ can be written as

$$\lambda_N^2=\text{Const}\left(\frac{R_N}{R}\right)^2\times\frac{M_1M_2}{(M_1+M_2)^2}, \qquad (1)$$

where $R_N$ is the nuclear part of the range and $M_1$ and $M_2$ are the masses of the fission fragment and the stopping nucleus.

Now the observed total range straggling listed in the last column of Tables I and II is given by

$$\lambda^2=\lambda_N^2+\lambda_F^2, \qquad (2)$$

in which $\lambda_F^2$ represents the contribution from energy fluctuations in the fission process; and when Eq. (1) is

TABLE III. Mean ranges of specific $U^{235}$ fission fragments in air.

| Fragments | Ranges (mgs/cm²) measured by G and W | Katcoff | Sugarman[a] |
|---|---|---|---|
| 22 sec | 2.40 | 2.56 | 2.62 |
| 55 sec | 3.27 | 3.36 | 3.25 |

[a] Converted data—see text for method used.

[10] N. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **18**, No. 8 (1948).
[11] N. Bohr, Phys. Rev. **59**, 270 (1941).

substituted for $\lambda_N^2$ one obtains

$$\lambda^2\left(\frac{R}{R_N}\right)^2=\text{Const}\frac{M_1M_2}{(M_1+M_2)^2}+\lambda_F^2\left(\frac{R}{R_N}\right)^2. \quad (3)$$

The ratio $R/R_N$ of the total range to the nuclear part of the range which depends to only a minor degree on the mass of the stopping nucleus has been calculated with the aid of Bohr's $(-dv/dx)$ formula and $\alpha$-particle data. The values thus calculated are approximately the same for the light and heavy fragments and the average is listed in the second column of Table IV. Values of $\lambda^2(R/R_N)^2$, where the $\lambda$'s are the corrected values given in Tables I and II, are listed in the third and fifth columns of Table IV and values of the reduced-mass factor are listed in the fourth and sixth columns. These quantities which according to Eq. (3) are linearly related are plotted in Fig. 3. The linear dependence is seen to be in satisfactory accord with the data except for the case of helium. No reason can at present be given for this anomalous behavior.

If, however, the general linear trend of the data is accepted then the intercepts on the ordinate will give the last term in Eq. (3) from which $\lambda_F$ can be determined with the use of the average value of $R/R_N$. Also, since it is known that the range of fission fragments varies approximately as the square root of the energy, one has that

$$\lambda_F=\Delta E_F/E_F=2(\Delta R_F/R_F),$$

and hence the intercepts give a measure of the energy spread in the fission process.

TABLE IV. Data as plotted in Fig. 3.

| Light or heavy fragments Gas | $R_0/R_N$ | Light fragment $\frac{M_1M_2}{(M_1+M_2)^2}$ | $\left(\lambda\frac{R_0}{R_N}\right)^2$ | Heavy fragment $\frac{M_1M_2}{(M_1+M_2)^2}$ | $\left(\lambda\frac{R_0}{R_N}\right)^2$ |
|---|---|---|---|---|---|
| $H_2$ | 2.39 | 0.0112 | 0.0137 | 0.0072 | 0.0106 |
| $D_2$ | 2.00 | 0.0220 | 0.0243 | 0.0142 | 0.0256 |
| He | 2.35 | 0.0420 | 0.0137 | 0.0276 | 0.0218 |
| Air | 2.25 | 0.123 | 0.0558 | 0.0857 | 0.1250 |
| Argon | 2.76 | 0.207 | 0.1405 | 0.165 | 0.2530 |
| | Av = 2.35 | | | | |

For the heavy fragments for which the data are most reliable, the observed intercept in Fig. 3 gives about 5% for the energy spread. For the light fragments, the corresponding value is about 8%. If the energy of fission of $U^{235}$ is taken to be 165 Mev, the energy associated with the 22-sec delayed-neutron emitter $({}_{53}I^{137})$ will be about 65 Mev, and with the 55-sec emitter $({}_{35}Br^{87})$ it will be about 100 Mev. The energy straggling as given by the lines drawn through the data in Fig. 3 for the light and heavy fragments will thus correspond to about 8 Mev and 3.5 Mev, respectively. The errors to be associated with these figures are somewhat difficult to determine. Nevertheless, it is of interest to compare these values with those obtained from the theory of the fission process as developed by Fong.[1] Fong has shown that in the neighborhood of the most probable fission mode, the $1/e$ half-width associated with the total intrinsic energy spread is about 5.8 Mev. The intrinsic energy spread does not include the energy spread resulting from fragment recoil from prompt neutrons. This latter spread is given[4] approximately by $4.8(M_H/M_L)^{\frac{1}{2}}$ and $4.8(M_L/M_H)^{\frac{1}{2}}$ for the light and heavy fragments respectively. By appropriately combining these quantities, one obtains for the energy full widths at half-maximum 8.4 Mev and 5.6 Mev for the light and heavy fragments. These values are in good accord with those obtained from the lines drawn through the data of Fig. 3. It is evident, however, from the figure that there is a considerable spread in the measured values, but it would seem reasonable to conclude that the energy dispersion is probably not appreciably greater than the values derived from the line intercepts.

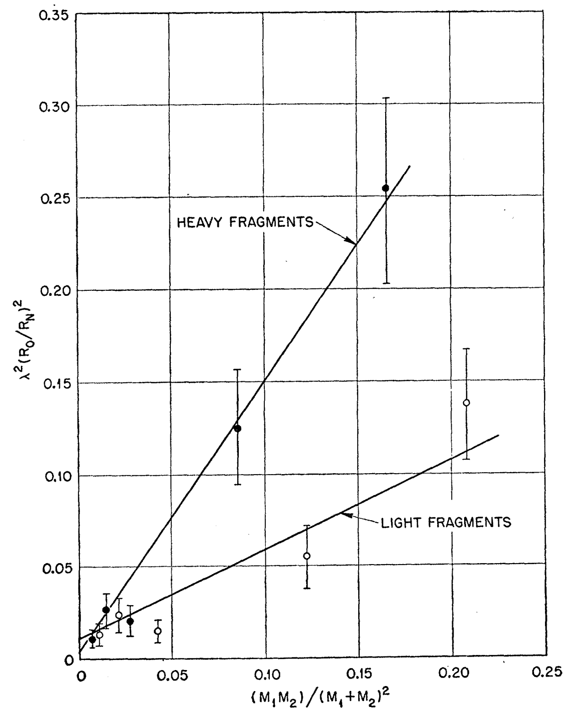


FIG. 3. Square of dispersion width *versus* $M_1M_2/(M_1+M_2)^2$ for fission fragment and stopping nucleus.

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