$$
\delta E_{\text{vac. pol.}} = \int \delta V(r) (F^2 + G^2) dr / \int (F^2 + G^2) dr. \quad (A12)
$$

Here  $F(r)$  and  $G(r)$  are the two radial components of as reported in the text.

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# $c$ alculated from the formula the  $K$ -electron wave function as obtained by numerical integration for  $Z = 170$ ,  $R = 11.3 \times 10^{-13}$  cm. In this way we obtained the value

 $\delta E_{\text{vac. pol.}} = -0.0185$  mc<sup>2</sup>, (A13)

# Capture-to-Fission Ratios for Fast Neutrons in  $U^{235}$ <sup>+</sup>

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The ratio  $\alpha = \sigma_c/\sigma_f$ , where  $\sigma_c$  is the neutron capture cross section and  $\sigma_f$  the neutron-induced fission cross section, has been measured for  $U^{255}$  as a function of neutron energy. A pulsed and collimated neutron beam is passed through a U<sup>235</sup> sample at the center of a large liquid scintillator. Captures and fissions are detected by means of their prompt gamma rays; elastic and inelastic scattering events are ignored because of smaller pulse heights. Fissions are distinguished from captures by means of delayed pulses from the capture of thermalized fission neutrons. It is found that in the neutron energy range  $E_n = 0.175$  to 1.0 Mev the value of  $\alpha$  is given approximately by  $\alpha = 0.190 - 0.116E_n$ . The accuracy of the determination of  $\alpha$  is 10 to 15% in terms of the standard deviation of individual points.

# INTRODUCTION

HE simplest and most widely used method of measuring neutron capture cross sections is activation, in which a radioactive end product is detected. For many nuclides, however, no radioactivity is produced. For capture reactions leading to stable or long-lived products, cross sections can be measured by the use of a mass spectrometer for determination of the product nuclides; this method usually requires the high fluxes present in reactors and does not seem to be suitable for fast monoenergetic neutrons. In addition to these methods involving detection of the end product of capture, two other basic methods are available. One is measurement of the change in neutron flux produced by capture, used, for example, in pile oscillator and reactivity measurements. The other basic method, used in the work reported here, is detection of the gamma rays emitted as a result of the capture process. For capture measurements with fast monoenergetic neutrons, this is in many cases the only practical method.

In the present experiment, neutron capture in  $U^{235}$ is detected by counting the capture gamma radiation in a large liquid scintillator<sup>1</sup> surrounding the sample; the cross section is determined by comparison with the fission rate of the same sample. Ideally the scintillator should be large enough to absorb all the energy of the gamma radiation emitted at its center. In this case,

capture gamma rays would produce a pulse corresponding to the sum of the energies of the gamma rays, which is equal to the binding energy of a neutron in  $U^{236}$ (6.29 Mev) plus the kinetic energy of the incident neutron. Fission is also accompanied by prompt gamma emission and the total energy of fission gamma rays is nearly the same as the total energy of capture gamma rays. $2-4$  Consequently, observation of the gamma pulses alone is not sufficient to distinguish between capture and fission. The prompt neutrons emitted in the fission process produce delayed pulses which enable us to identify a fission event. The scintillator is large enough to cause most of the fission neutrons to be thermalized and finally captured in the liquid. The addition of cadmium salt to the solution ensures that most neutrons will be captured in cadmium, and the resulting 9 Mev of gamma radiation provides an ample pulse for observation. The cadmium concentration is adjusted so that the mean life of neutrons in the solution is about 40  $\mu$ sec. The pulses due to fission neutrons are then spread out in time so that they may be counted individually with almost negligible losses due to resolving time. The neutron beam is pulsed so that the neutrons which cause capture or fission in the sample arrive in bursts of  $0.1$ - $\mu$ sec duration every 100  $\mu$ sec. Pulses in the scintillator which are caused by prompt gamma rays due to capture or fission are

t Work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>&</sup>lt;sup>1</sup> Liquid scintillator technique is described by Reines, Cowan, Harrison, and Carter, Rev. Sci. Instr. 25, 1061 (1954).

<sup>&</sup>lt;sup>2</sup> I. Francis and R. Gamble and also F. Maienschein et al., Oak Ridge National Laboratory Report ORNL-1879, October, 19SS (unpublished). '

Smith, Fields, and Friedman, Phys. Rev. 104, 699 (1956).

<sup>&#</sup>x27;Kinsey, Hanna, and Van Patter, Can. J. Research 26, 79 (1948).

essentially coincident with the beam pulse. Fission events are identified by the observation of one or more pulses in the  $100$ - $\mu$ sec interval following the prompt pulse, with corrections both for background pulses and for the small fraction of fissions known to be followed by no pulse.

The only interaction other than capture and fission produced in  $U^{235}$  by neutrons of energy less than 1 Mev is scattering. Elastic scattering of neutrons by the sample leads to a delayed capture pulse in the solution but provides only a small prompt pulse produced by recoil protons during the slowing down of the neutron in the solution. Inelastic scattering can produce scintillator pulses due to gamma radiation and the kinetic energy of the scattered neutron, but the sum remains less than the prompt pulse bias of 1 Mev. If we require that the prompt pulse correspond to more than 1 Mev of gamma radiation, all scattering events will be ignored. The prompt pulses observed are thus the result of fissions and captures, plus a small number due to random coincidences and other minor effects. The ratio of the capture to fission cross sections of  $U^{235}$ , a quantity which will hereafter be designated by  $\alpha$ , may thus be determined experimental in a straightforward way.

The characteristics of a large liquid scintillator which make this experiment feasible are the high efficiencies for detection of gamma rays and of neutrons. The efficiencies for detection of capture and fission by prompt gamma rays are assumed to be equal under the conditions of this experiment, and are about  $90\%$ ; this assumption has been checked by taking data at various biases for prompt pulses. The efficiency for detection of fission neutrons is found to be  $57\%$ . This relatively low figure (for a liquid scintillator) is the result of losses caused by a bias for delayed pulses high enough to eliminate most background pulses, a gate length somewhat shortened for the same reason, and a scintillator size smaller than was used in previous work.<sup>5</sup> This efficiency for neutrons results in a probability of  $18\%$  that no neutron pulse will be observed following a U<sup>235</sup> fission. In order to correct the data for this effect, measurements are made with a fission counter in place of the capturing sample. The probability that no detectable neutron pulses follow fission is observed under the same conditions as with the capturing sample, except that coincidences between fission counter pulses and beam pulses are required to, open the  $100$ - $\mu$ sec gate.

It is particularly useful to measure  $\alpha$  as a function of neutron energy for  $U^{235}$  because the fission cross section of this isotope is well known<sup>6,7</sup> and serves as a

convenient laboratory standard. Thus we may use  $\sigma_t + \sigma_c = \sigma_f(1+\alpha)$  as a standard for the measurement of capture cross sections of other nuclei by the method described here (such measurements are in progress; some preliminary results are given in the compilation by Hughes and Schwartz<sup>6</sup>). Furthermore, the ratio  $\alpha$ for  $U^{235}$  is of considerable interest to reactor designers because it specifies the rate at which fuel is wasted by capture.

## EXPERIMENTAL PROCEDURE

#### General

The equipment and techniques used in the  $\alpha$  measurement were suggested by, and do not differ greatly from, those used in the measurement of the multiplicities of fission neutrons.<sup>5</sup> Electronic counting has been substituted for photographic recording of single oscilloscope sweeps. The. scintillator assembly has been replaced by a smaller one because of shielding requirements, and the collimator geometry has been improved. Pulsing the proton beam from a 2.5-Mev electrostatic accelerator has made possible a large increase in the peak intensity of the  $T(p,n)$  neutron source; the maximum target current is about 40  $\mu$ a, but the average current of  $0.04$   $\mu$ a gives reduced background in the scintillator and long life for the aluminum window of the gas target. At the lowest neutron energies used (below 285 kev) a slower second group of neutrons is always present with a  $T(p,n)$ source, but the pulsed beam prevents it from affecting the data, because of its considerably longer time of flight. Similarly, the effects of any neutrons of degraded energy, originating, for example, in the shielding, are essentially eliminated by the pulsing technique. Fission chamber data show that slow neutrons arriving at almost random times are responsible for considerably less than  $1\%$  of the fissions occurring at the time of the beam pulse. Primarily, however, pulsing the beam makes it possible to discriminate against the delayed pulses from scattered neutrons, without which the experiment could not be done. To maintain adequate neutron flux, fairly thick targets, and hence broad bands of neutron energy, are used. Proton beam pulses 0.1  $\mu$ sec wide are produced at a repetition rate of 10<sup>4</sup> per second by the use of an electrostatic beam deflector. The beam is deflected from the gas target by a fixed potential on the deflector except for the  $0.1$ - $\mu$ sec intervals when the voltage is removed and the beam is allowed to enter the target.

Figure 1 shows the arrangement of neutron source, collimator, scintillator, and shielding. The collimator provides a narrow beam of neutrons which pass through the axial tube of the liquid scintillator. The neutron beam diameter is 0.75 inch at the sample position. The scintillator tank is a cylinder 19 inches in diameter and 19 inches long, with a porcelain-enameled interior. The detectors are six 5-inch-diameter photomultiplierg

<sup>~</sup> Diven, Martin, Taschek, and Terrell, Phys. Rev. 101, 1012

<sup>&</sup>lt;sup>6</sup> D. J. Hughes and R. B. Schwartz, Neutron Cross Sections, Brookhaven National Laboratory Report BNL-325, (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1957), Suppl. 1.<br><sup>7</sup> B. C. Diven, Phys. Rev. 105, 1350 (1957).



FIG. 1. Schematic diagram of equipment used in the  $\alpha$  measurement, showing neutron source, collimator, liquid scintillator, and shielding.

(DuMont 6364), all mounted on the end of the tank opposite the collimator, with outputs in parallel. The gains of the photomultiplier tubes are equalized by adjustment of the individual voltage dividers. The stabilized high voltage for the tubes is monitored continuously by a differential voltmeter, allowing the voltage to be maintained at  $1600 \pm 1$  volts. The liquid scintillator solution is triethylbenzene and terphenyl, with POPOP<sup>8</sup> as a wavelength shifter and cadmium octoate to increase the neutron sensitivity. The amount of cadmium is such as to give a cadmium-to-hydrogen atomic ratio of 0.0005.

The primary capture (plus fission) sample is composed of 40 disks of  $93\%$  U<sup>235</sup>, each 0.75 inch in diameter, 0.005-inch thick  $(9.5 \text{ g/cm}^2 \text{ total})$ , and spaced 0.12 inch apart in a light aluminum frame. Each disk is suspended by two 0.005-inch steel wires and a 0.008-inch steel helical spring. A similar set of 0.010-inch polyethylene disks, used to produce scattering only, is mounted in the same way, and a third identical frame, with no sample or wires, is used as a blank. The dimensions and arrangement of the  $U^{235}$  disks were chosen to provide a sample which would be as transparent as possible to capture and fission gamma rays and to fission neutrons. The mass of the  $U^{235}$  sample is limited both by the mean free path of primary neutrons and by the need to minimize multiplication, i.e. , the production of secondary fissions by fission neutrons, either promptly or after some energy loss in the solution. The latter effect is minimized by surrounding the U<sup>235</sup>, as well as the polyethylene and blank samples, with 0.030 inch of cadmium. The fraction of fissions producing prompt secondary fission is estimated to be  $1\%$  in the present arrangement.

The fission counter, which may be inserted in the 2.5-inch-diameter central hole of the scintillator in place of the capture sample, has 36 surfaces, each coated with about 1.2 mg/cm<sup>2</sup> of  $U^{235}$ .

### Electronics

Pulses from the scintillator are amplified by two systems,  $A$  and  $B$ , in parallel, as shown in the block

diagram, Fig. 2. Amplifier  $A$  delivers pulses to a Los Alamos Model 12 time-to-pulse-height converter,<sup>9</sup> which also gets a synchronizing pulse from the beam pulser to establish a zero of time. The converter produces an output pulse whose amplitude is essentially proportional to the time interval between the synchronizing pulse and the scintillator pulse. A Los Alamos Model 2A 100-channel pulse-height analyzer<sup>10</sup> at the output of the time converter records the time distribution of scintillator pulses during a  $2$ - $\mu$ sec interval, centered on the beam pulse time. Figure 3 shows a typical set of such data, with the prompt peak due to capture and fission gamma rays from  $U^{235}$ superimposed on a background of random pulses in the scintillator which are produced mostly by cosmic rays and radioactivity. This display provides a method of determining what fraction of the total number of pulses in the peak are random and not related to prompt events produced in the scintillator by the neutron beam.

Time-converter pulses also drive two single-channel analyzers in parallel (Fig. 2). Each of these analyzers selects a pulse-height (and therefore a time) interval of interest. One analyzer is adjusted to respond only to "prompt" scintillator pulses, i.e., those coincident with the beam pulse. The output of this analyzer is used to open the  $100$ - $\mu$ sec gate for delayed pulses. Most of the trigger pulses thus obtained are due to capture and fission in the  $U^{235}$  sample, though some are due to the random pulses in the scintillator. In order to obtain the distribution of numbers of pulses following such random pulses, the second single-channel analyzer is used to select "early" trigger pulses which occur in a  $0.5$ - $\mu$ sec interval closely preceding the beam pulse.

The delayed pulses from system  $B$  pass through a discriminator and pulse shaping circuit to the gate



FIG. 2. Block diagram of electronics. For the purpose of analyzing fissions only, the fission chamber is placed within the liquid scintillator and is then connected to amplifier  $A$  in place of the scintillator output.

<sup>9</sup> Weber, Johnstone, and Cranberg, Rev. Sci. Instr. 27, 166  $(1956).$ 

<sup>10</sup> Developed at Los Alamos largely by C. W. Johnstone and P. W. Byington.

<sup>8</sup> Hays, Rogers, and Ott, J. Am. Chem. Soc. 77, 1950 (1955).



FIG. 3. Time distributions of scintillator pulses with various samples in the scintillator, as determined with a time-converter and 100-channel pulse-height analyzer. Certain groups of channels are designated as "early, " "prompt, " and "late" for analysis of at designator as carry, prompt, and rate for analysis of channels, and are discussed in the text. All sets of data groups of channels, and are discussed in the text. All sets of data are for equal integrated beam current and similar times except for the beam-off data.

which is opened by pulses from the single-channel analyzers of system A. Time delays in the two systems are so arranged that prompt pulses (corresponding to the time converter pulses which triggered the gates) arrive at the gate about 1  $\mu$ sec too early to be passed. A Burroughs tube (MO-10R) is used in a fast sealer to count the number of pulses,  $n$ , which arrive during the 100  $\mu$ sec that the gate is open. Each count of  $n$  is recorded in one of a bank of 11 registers designated 0, 1, 2,  $\cdots$ , 9,  $\geq$ 10 by the advance by one unit of the *n*th register. A three-position switch (actually an array of mercury relays) selects "prompt," "early," or "background" trigger pulses which open the gate ("background" trigger pulses are arbitrarily selected beam pulses not necessarily associated with scintillator pulses). The switch moves automatically to the next position in rotating sequence whenever the circuit is triggered. The 6nal result is that three banks of eleven registers record the distribution of numbers of pulses following "prompt," "early," and "background" trigger pulses.

By placing a fission chamber in the center of the scintillator and gating on the fission pulse, it is possible to measure with the same equipment the probabilities of observing  $0, 1, 2, \dots, 9, \ge 10$  pulses following a fission, and an appropriate background distribution. From such observations are deduced the probabilities  $C_n$  that a fission event is followed by *n*-detectable neutron pulses during the  $100$ - $\mu$ sec gate. These data

have been taken primarily at one neutron energy (500 kev), for the distribution  $C_n$  changes slowly with neutron energy and several days of running are required to obtain sufficiently accurate data at one energy. During the course of this experiment 28000 fissions induced by 500-kev neutrons were observed and analyzed. Slight changes in  $C_n$  with neutron energy were computed on the assumptions (1) that the average number of neutrons per fission changes with energy at the rate of 0.14 per Mev and (2) that the width of the neutron number distribution from fission is substantially constant with energy. These assumptions are in accord with all available experimental evidence<sup>11</sup> for U<sup>235</sup>, including that taken during this experiment. The result of this computation is a maximum correction to  $\alpha$  of 0.014 at 1000 kev, with correspondingly smaller correction for energies nearer 500 kev. For these fission chamber runs the bias for delayed pulses must be the same as during all other types of runs. Ca1ibration runs are made each day using a  $Co<sup>60</sup>$  source in the scintillator, during which the (normally gated) discriminator bias is set for a fixed counting rate in ungated operation. The over-all gain has been found in this way to be stable to within a few percent over a few months. The bias setting of the time converter discriminator is checked at the same time, although this is not a critical setting.

#### ANALYSIS

A cycle of measurements at one neutron energy consists of runs with  $U^{235}$ , polyethylene, and blank samples. For each of these runs there is a set of data from the time-converter, which shows the time spectrum of scintillator pulses at prompt time  $\pm 1$  µsec. For the  $U^{235}$  there is also a set of data from the cyclic selected counter, which gives the pulse number distribution during gates triggered by prompt pulses, by early pulses, and at random. From the time-converter data for the  $U^{235}$  sample it is possible to determine what fractions of the gates are tripped by events other than capture or fission in the  $U^{235}$ , such as random pulses and other minor effects. After corrections have been made for the effects of such trigger pulses, the cyclic counter data allow the ratio of capture to fission events to be determined.

The gate trigger pulses with the  $U^{235}$  sample in place may be divided into five categories, given by the trigger probabilities:

 $T_c$ =the fraction of gates triggered by capture gamma rays.

 $T_f$  = the fraction triggered by fission gamma rays.

 $T<sub>r</sub>$ =the fraction triggered by random coincidences between scintillator pulses and beam pulses; this includes effects of cosmic rays, radioactivity, and neutrons from earlier beam pulses. This rate is measured

<sup>&</sup>lt;sup>11</sup> J. Terrell, Phys. Rev. 108, 783<sup>3</sup>(1957).

at "early" time, but is assumed to be the same at "prompt" time.

 $T<sub>o</sub>$ =the fraction triggered by scintillator pulses associated with the beam, but not with the  $U^{235}$  sample, such as target gamma rays and collimator leakage; it is measured with the blank sample holder.

 $T<sub>s</sub>$  = the fraction of trigger pulses due to neutrons scattered by the  $U^{235}$  sample into the scintillator and captured early enough to appear prompt, plus any other pulses which are beam- and sample-associated but are not caused by capture or fission; it is measured by means of the polyethylene sample.

Neglecting the extremely small probability that a trigger pulse will be due to the combination of pulses from two separate events, there is the necessary relation

$$
T_c + T_f + T_r + T_o + T_s = 1.
$$
 (1)

The quantities  $T_r$ ,  $T_o$ , and  $T_s$  are determined from time-converter data. Figure 3 shows typical timeconverter data for  $U^{23\bar{5}}$ , polyethylene, and blank samples, in addition to a long run with no beam. Certain groups of channels of the 100-channel pulseheight analyzer are chosen to represent "early," "prompt," and "late" time intervals, as is shown in Fig. 3. All channels would have equal weight if the time-converter and associated amplifiers were perfectly linear in operation. Since this is not quite the case, periodic beam-off runs are taken to determine with very good statistics the ratios of counts in "early, ""prompt, " and "late" channels with beam off. The ratios of interest are  $P/E$  and  $L/E$ , in which E, P, and L are the total counts in the three groups of channels mentioned. The corresponding totals with U235, polyethylene, and blank samples in place, with beam on, are designated by subscripts  $u$ ,  $\phi$ , and  $b$ , respectively. It is assumed here that all runs with beam on are for the same integrated beam current and under as nearly identical conditions as possible. Since  $(P/E)E_u$  is the number of counts out of the prompt total  $P_u$ which are due to random background, it is easy to show that

$$
T_r = (P/E)(E_u/P_u). \tag{2}
$$

Similarly,

$$
T_o = [P_b - (P/E)E_b]/P_u.
$$
 (3)

In order to compute  $T_s$ , it is necessary to assume that the part of the polyethylene prompt peak which is not included in the blank prompt peak, after both have been corrected for random counts, is due almost entirely to neutrons scattered from the sample which are captured promptly in the scintillator tank.  $T_s$  may then be calculated on the further assumption that the number of such pulses has the same ratio to the corrected counts in late channels, due also to scattered neutrons being captured in the solution, for  $U^{235}$  and polyethylene. The polyethylene sample is thus intended to simulate the scattering of  $U^{235}$  without the capture and fission. It should be pointed out that fission neutrons from the  $U^{235}$  sample will not in general produce counts in the late channels, because the prompt 6ssion gamma rays will almost always have previously set the time-converter into operation for those particular beam pulses. The fact that the timeconverter can produce only one output pulse per beam pulse causes no appreciable counting loss, however, since only about one beam pulse out of a thousand is associated with a scintillator pulse falling in the time range of the time-converter. Typical values of  $T_r$ ,  $T_o$ , and  $T_s$  are 0.07, 0.03, and 0.01, respectively. The remainder of prompt triggers are due to capture and fission in the  $\hat{U}^{235}$  sample.

In order to separate captures from fissions, it is necessary to use the cyclic counter data for  $U^{235}$ , for which the following probabilities may be defined:

 $A_n$ =probability of *n* pulses occurring in any gate tripped by a prompt pulse.

 $B_n$ =probability of *n* (background) pulses occurring in any gate triggered at prompt time by a randomly selected beam pulse.

 $C_n$ =probability of *n* correlated pulses (i.e., after correction for background) occurring in a gate triggered by a fission in the sample.

 $E_n$ =probability of *n* pulses occurring in a gate triggered by an early pulse, or by a prompt pulse in the category  $T<sub>r</sub>$  (the distribution  $E<sub>n</sub>$  is not the same as  $B_n$ , because of the occurrence of groups of pulses in the scintillator; such groups may arise from cosmic rays, for example).

Obviously  $A_n$ ,  $B_n$ , and  $E_n$  are given directly by the distributions of pulse numbers from the cyclic selected counter with the  $U^{235}$  sample in place. The fission-pulsenumber distribution  $C_n$  is determined by the use of the fission chamber with the cyclic counter; slight corrections for the expected multiplication and absorption of fission neutrons in the  $U^{255}$  sample must be made. The background-corrected distribution  $C_n$  is used here because the background is not the same with the fission chamber in place as with the capture sample in place. The removal of background from the fission distribution is done by means of the set of equations

$$
F_n^* = C_0^* B_n^* + C_1^* B_{n-1}^* + \dots + C_n^* B_0^*, \qquad (4)
$$

in which  $F_n^*$ ,  $B_n^*$  and  $C_n^*$  are the uncorrected fission distribution, the background distribution, and fission distribution corrected for background, all as measured with the fission chamber in place.

The simplest way of determining the fractions  $T_c$ and  $T_f$  of capture and fission triggers is to use the probabilities that no counts follow the various types of trigger pulses. Since trigger pulses of the types  $T_c$ ,  $T_o$ , and  $T_s$  are not followed by correlated pulses,

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 $\mu$  it can be shown that  $\alpha$ .

$$
\frac{T_c}{T_f} = \frac{(1 - T_r - T_s - T_o)(1 - C_0)}{1 - T_r(1 - E_0/B_0) - A_0/B_0} - 1.
$$
 (5)

As was mentioned earlier, the total prompt gammaray energy is roughly the same for fission and capture. Since, in both cases, the energy is almost always divided among a number of gamma rays,<sup>3</sup> the chance of total escape of the gamma-ray energy from the scintillator is essentially negligible. Hence, for a sufficiently low bias on prompt pulses,

$$
\alpha = \sigma_c / \sigma_f \cong T_c / T_f. \tag{6}
$$

For capture and fission gamma-ray spectra sufficiently similar in total energy and multiplicity this should also be true at higher biases. The procedure used has been to measure  $T_c/T_f$  for several biases at one neutron energy, in order to establish the limiting value  $\alpha$ . The result is that  $T_c/T_f$  is relatively insensitive to bias changes in the vicinity of the 1.25-Mev bias used. This will be discussed in more detail in the next section.

Equation (5) does not exhaust the information Equation (3) does not exhaust the information<br>contained in the distributions  $A_n$ ,  $B_n$ ,  $C_n$ , and  $E_n$ . It is only one of a set of equations,

in the distributions 
$$
A_n
$$
,  $B_n$ ,  $C_n$ , and  $E_n$ . It  
to of a set of equations,  

$$
\frac{T_c}{T_f} = \frac{(1 - T_r - T_s - T_o)(F_n - B_n)}{A_n - B_n - T_r(E_n - B_n)} - 1,
$$
(7)

in which  $F_n$  is the distribution of pulses following a fission in the capture sample, which may be obtained from fission-chamber data with some small corrections. Solution of Eq. (7) yields a set of values for  $T_c/T_f$ , with increasing uncertainty and decreasing weight as  $n$  increases. It is found that, under the conditions of this experiment, about  $70\%$  of the weight is contained in the first of these equations, equivalent to Eq. (5). For this reason the rest of the distribution data has been used only as a check.

The result of Eq. (5) has been checked primarily by calculating  $T_c/T_f$  from the first and second moments of the various number distributions. The method is straightforward and the equations will not be enumerated here. The result is similar to that of Eq. (5) but is consistently higher by about 0.015, an effect which would be expected from neglecting to correct for a very small amount of multiplication. Since  $T_c/T_f$ from Eq. (5) is at least ten times less sensitive to this effect, somewhat more confidence can be placed in that result because of the agreement with the "moments" calculation.

Among other processes which could require correction of the data are absorption of capture and fission gamma rays in the sample and pile up of prompt pulses from capture and fission with small slowing-down pulses from scattered neutrons. The first could cause trouble only by a differential effect between capture and fission;



FIG. 4. Experimental results for  $\alpha$  of U<sup>235</sup> as a function of neutron energy. Standard deviations are indicated, as well as energy spreads. The straight line is a least-squares fit to the data.

the second could cause increased counts in many triggered gates. These effects, however, have been shown to be negligible in this experiment. The almost negligible effect of the  $0.2$ - $\mu$ sec over-all resolving time on the probabilities  $A_n$ ,  $B_n$ ,  $C_n$ , and  $E_n$  has been corrected for;  $A_0$ ,  $B_0$ ,  $C_0$ , and  $E_0$  do not require such correction.

### RESULTS

The experimental values of  $\alpha$  are shown in Fig. 4. The standard deviations are based on a combination of the statistical uncertainties, related to the number of events observed, and estimates of uncertainties in the relative efficiencies for detection of capture and fission gamma rays. A total of twelve runs of 10000 cycles each on the cyclic selected counter were taken at a neutron energy of 400 kev. Biases of 1.25, 2.5, and 5.0 Mev were used on the time-converter discriminator, which determines the minimum acceptable height of prompt fission and capture pulses. The value of  $T_c/T_f$ at 1.25-Mev bias was  $1.08\pm0.06$  and  $1.8\pm0.4$  times as large as at 2.5 and 5.0 Mev, respectively. These values are consistent with the assumption that the total energy of fission gamma rays is 1 or 2 Mev higher than the 6.7 Mev from capture and that the pulseheight distributions are similar. It is observed that the scintillator pulses from fissions range in height from less than 1 Mev to a maximum of perhaps 9 Mev. Since the exact spectra of gamma rays from capture and fission are unknown, as are the detailed shapes of the pulse-height distributions at low pulse heights, the extrapolation of  $T_c/T_f$  to zero bias and the assumption that the extrapolated value is  $\sigma_c/\sigma_f$  are subject to some uncertainty.

On the basis of the results of these runs, all values of  $T_c/T_f$  were extrapolated to zero bias by multiplying values obtained at 1.25 Mev and 2.50 Mev biases by  $1.08 \pm 0.08$  and  $1.17 \pm 0.08$ , respectively. The standard deviation of the 400-kev point arises mostly from the uncertainty of this extrapolation and partly from small variations of the efficiency for detection of neutrons. All other points of Fig. 4 consisted of either one or two runs of 10 000 cycles each. The standard deviations are relatively larger than at 400 kev because of the smaller amount of data taken. All data have been corrected for capture in the  $7\%$  of U<sup>238</sup> which was present in the sample; the cross sections used in this correction were taken from a published compilation.<sup>6</sup> Neutron energy spreads are also indicated for each point in Fig. 4.

#### DISCUSSION

The experimental values of  $\alpha = \frac{\sigma_c}{\sigma_f}$ , when plotted against neutron energy as in Fig. 4, lie approximately on a straight line in the energy range 175 to 1000 kev. The equation of the line is given by a least-squares fit as  $\alpha=0.190-0.116E_n$ , in which  $E_n$  is the neutron energy in Mev. There is no theoretical reason to expect such a linear decrease of  $\alpha$  with energy, and it is purely coincidental that the extrapolation of this line passes near the thermal value<sup>6</sup> of 0.192. The thermal value is strongly influenced by the properties of one or two resonances, and  $\alpha$  is known<sup>6</sup> to fluctuate widely from level to level in the resonance region. vel to level in the resonance region.<br>An integral experiment,<sup>12,13</sup> involving irradiation of

suitably shielded  $U^{235}$  samples in a Hanford reactor to give average values of  $\alpha$  above a cutoff energy, has given values ranging from 0.41 to 0.52 for median energies of 15 kev and lower. Values of  $\alpha$  obtained at higher energies were 0.10 at 215 kev and 0.17 at 300 kev, which are in reasonable agreement with those reported here. Similar values of  $\alpha$  for U<sup>235</sup> have been obtained by Kafalas, Levenson, and Stevens<sup>14</sup> from integral measurements in the reactor EBR-1. Measurements of  $\eta = \bar{\nu}/(1+\alpha)$ , the number of neutrons emitted per neutron absorbed in  $U^{235}$  ( $\bar{\nu}$  is the average number of neutrons emitted per fission), have been made by of neutrons emitted per fission), have been made by Spivak  $el$   $al.^{15}$  using photoneutron sources. These values of  $\eta$ , when combined with the known<sup>6,11</sup>  $\bar{\nu}$ , yield values of  $\alpha$  at 140, 250 and 900 kev of 0.16 $\pm$ 0.06,  $0.13\pm0.08$  and  $0.13\pm0.05$ , respectively. These results also agree satisfactorily with those of the present paper. region is a matter of some difficulty, involving the effects of various angular momenta, competition with inelastic scattering, and varying fission and radiation widths. If the distributions of fission and radiation widths were exactly the same in this energy region as in the resonance region where these parameters have been measured, and if the other effects mentioned could be neglected, a decrease in  $\alpha$  would be expected for increasing energy. As Wigner<sup>16</sup> has pointed out, the relative weights of energy levels are different in the two regions; when the neutron emission width, which varies approximately as  $E^{\frac{1}{2}}$ , is larger than the fission width, as is true in the 100- to 1000-kev region, levels with large fission widths are given relatively more weight than in the resonance and kev regions (this conclusion depends on the fact that the radiation widths are smaller than the fission widths). The result is a decrease of  $\alpha$  with energy, the amount depending on the extent of the fluctuations of the various level widths. However, this effect alone is not enough, as is shown by calculations of Wigner<sup>16</sup> and Oleksa,<sup>13</sup> to account for the considerable decrease in  $\alpha$  for high energies from the values of 0.4 to 0.5 found in the kev region. Weisskopf<sup>17</sup> and Bethe<sup>18</sup> have found evidence that fission widths increase more rapidly with energy than radiation widths, thereby accounting for the experimental decrease in  $\alpha$ .

It is interesting to note that the capture cross section for  $U^{235}$ , which is essentially in the range 100 to 250 mb in the present data, is very similar in magnitude to capture cross sections for Th $^{232}$  and U<sup>238</sup> in the same energy range.<sup>6</sup>

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A theoretical prediction of  $\alpha$  in the 100- to 1000-kev

<sup>&</sup>lt;sup>12</sup> Kanne, Stewart, and White, *Proceedings of the International* Conference on the Peaceful Uses of Atomic Energy, Geneva, August,  $1955$  (United Nations, New York, 1956), Vol. 4, p. 315.<br><sup>18</sup> S. Oleksa, J. Nuclear Ener

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