Neutron correlations in spontaneous fission of ²⁵²Cf

A. Gavron and Z. Fraenkel

Nuclear Physics Department, Weizmann Institute of Science, Rehovoth, Israel (Received 26 September 1973)

The variance and covariance of the neutron distributions in fission fragments of ²⁵²Cf have been measured as a function of the fragment mass and total kinetic energy. The neutrons were detected by a time-of-flight technique. The covariance results indicate that there is no change in the degree of viscosity in the transition of the fissioning nucleus to the scission point as a function of the total excitation energy. There are some discrepancies between various features of the neutron evaporation cascade determined in our experiment and results of standard evaporation calculations. No simple explanation of these discrepancies is offered.

RADIOACTIVITY, FISSION ²⁵²Cf(sf); measured variance and covariance of $\gamma(A, E)$ of the two fragments.

INTRODUCTION

In the past, much effort has been invested in determining the average number of neutrons in low-energy fission as a function of various parameters of the fission fragments such as their mass and total kinetic energy.¹⁻⁵ These data have been summarized by Terrell⁶ and more recently by Nifenecker *et al.*⁷ The most important results are: (1) The average number of neutrons $\overline{\nu}$ exhibits a characteristic "saw-tooth-curve" dependence as a function of the fragment mass A; (2) the derivative of the average number of neutrons with respect to the total kinetic energy of the two fragments E_K , $(\partial \overline{\nu}/\partial E_K)(A)$, also exhibits a "sawtooth-curve" dependence on the fragment mass similar to that of $\overline{\nu}(A)$.^{4,8}

This information permits a description of the partition of the average excitation energy between the two fragments: The parallel behavior of the $\overline{\nu}(A)$ and $(\partial \overline{\nu}/\partial E_{\kappa})(A)$ curves indicates that the fragment which has more excitation energy is also more susceptible to receiving additional excitation energy.8 Additional details of the excitation energy partition and of the fragment deexcitation process can be obtained by measuring the second moments of the neutron number distribution. In particular, it is interesting to investigate whether two complementary fission fragments of given mass and fixed total excitation energy can exchange excitation energy between each other. The answer to this question is related to the degree of viscosity of the transition to the scission point-a highly viscous transition would keep the nascent fragments in thermodynamic equilibrium till the scission point, thus holding the partition of the excitation energy constant.

Measurement of the covariance of the neutron distributions in two complementary fragments can provide the necessary information regarding the viscosity of the transition to the scission point. The variance of the neutron distribution can provide details of the fragment deexcitation process through neutron emission.

In order to obtain these quantities we have performed the following experiments:

(a) " σ^2 experiment" to determine the variance of the distribution of the number of neutrons emitted from a single fragment. This variance, denoted $\sigma_v^2(A, E_r)$ is defined by

$$\sigma_{\nu}^{2}(A, E_{\kappa}) = \overline{\nu^{2}}(A, E_{\kappa}) - \overline{\nu}^{2}(A, E_{\kappa}). \qquad (1)$$

(b) " μ experiment" to determine the covariance of the two neutron distributions in two complementary fragments. The covariance $\mu(A, E_K)$ is defined by

$$\mu(A, E_{\kappa}) = \operatorname{av}[\nu(A, E_{\kappa})\nu(A', E_{\kappa})] - \overline{\nu}(A, E_{\kappa})\overline{\nu}(A', E_{\kappa}), \qquad (2)$$

where

$$A' = 252 - A$$
. (3)

Both av[] and the bar over a symbol imply average. Obviously $\mu(A, E_K)$ is symmetric around A = 126 and needs thus to be determined only for $A \leq 126$.

(c) " $\overline{\nu}$ experiment" in which we have determined $\overline{\nu}(A, E_{\kappa})$. Some systematic errors that exist in previous data^{2, 3, 9} were corrected in our own analysis of the experimental results.

The results obtained from our experiments contain several systematic errors which stem from insufficient knowledge of various details of the fis-

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sion and fragment deexcitation processes. These errors include the effect of scission neutrons, the effect of velocity correlations (in the center-ofmass system) between two neutrons in an evaporation cascade, and the effect of delayed γ radiation. These effects will be analyzed in detail and their contribution to the measured quantities will be estimated.

EXPERIMENTAL METHOD

A. Experimental arrangement and block diagram

A schematic representation of the experiment together with the block diagram of the electronics are shown in Fig. 1. A ²⁵²Cf source of $\sim 2 \times 10^5$ fission per minute deposited on a thin Ni backing was placed between two surface-barrier fissionfragment detectors denoted F1 and F2 in Fig. 1. These detectors together with the source were placed inside an aluminum vacuum chamber of 30-cm diameter and 0.5-cm wall thickness. The fission-fragment detectors F1 and F2 were placed at a distance of 5.5 cm from the source and subtended an angle of 19° with respect to it. The neutrons were detected by means of the time-of-flight method with the aid of two identical NE102 plastic scintillators (manufactured by Nuclear Enterprises Ltd.) denoted PM1 and PM2 in Fig. 1. The two scintillators, each of 5.1-cm length and 12.7-cm diameter were situated outside the chamber at a distance of 27.5 cm between the source and the front of the scintillator.

Two experiments were performed with this configuration: The first (the " σ^2 experiment") to determine the variance of the neutron distribution $\sigma_{\nu}^2(A, E_K)$. In this experiment both PM1 (at position I in Fig. 1) and PM2 were at an angle of $22^{\circ}30'$ relative to the source-fission detector F1 axis. The second (the " μ experiment") to determine the covariance $\mu(A, E_K)$ of the two neutron distributions in the two complementary fragments. In this experiment PM1 was shifted to an angle of



FIG. 1. Diagram of experiment setup and block diagram of electronic system.

 θ =157°30' relative to the same axis (position II in Fig. 1). An additional experiment, the " $\overline{\nu}$ experiment" to determine $\overline{\nu}(A, E_{K})$, was performed in which the scintillators were placed at a distance of 57.5 cm from the front of the detector to the source, and at angles of 0 and 180° relative to the F1-F2 axis.

The events were recorded on magnetic tape by a four-dimensional analyzer. They consisted of the following pulses: F1 and F2, the pulse heights from the two fission-fragment detectors; and TAC1 and TAC2, the pulses obtained from the time-ofamplitude converters. These last pulses are proportional to the time difference between the start signal furnished by the solid-state detector F1 and the stop signal obtained by the interaction of neutrons or photons with one of the scintillators. The pulses were coded into eight binary bits (256 channels) and an additional bit was set to unity when there was a coincidence between the pulses from TAC1 and TAC2 (routing). In addition to these pulses a gate pulse was fed into the analyzer (see Fig. 1). This pulse determined what type of experimental data was being recorded. The main experiments were fourfold coincidence experiments in which the events were of the $F1 \cap F2$ \cap TAC1 \cap TAC2 type. In addition we performed two calibration runs daily in which we obtained $F1 \cap F2$ events (for the calibration of the solidstate detectors) and $F1 \cap F2 \cap (TAC1 \cup TAC2)$ events (for calibration of the time-of-flight spectrum). This last type of event was also collected throughout the " $\bar{\nu}$ experiment."

The timing pulse from the photomultipliers was obtained from a constant-fraction-of-pulse-height trigger in order to obtain good timing resolution. The fast discriminator level was set as low as possible, and the final discrimination level was determined by a slow discriminator which gated the TAC output, to be at roughly 200-keV neutron energy. The timing pulses from the fission fragment detectors were obtained from commercial time-pickoff units. Their discrimination level was set just above the pulse height produced by the 6.1-MeV α particles from the decay of ²⁵²Cf.

B. Data collection

A total of 10^6 fourfold coincidence events were obtained in each of the two main experimental configurations (" σ^2 " and " μ "). Of these, about 2×10^5 were neutron-neutron coincidence events. In addition, a total of 4×10^5 binary fission events and a similar number of $F1 \cap F2 \cap (TAC1 \cup TAC2)$ -type events were obtained. Each of the two main experiments lasted for about six months during which calibration runs of the solid-state detector and TAC system were performed daily. The " ν experiment" lasted for two weeks during which 4 $\times 10^5$ triple (neutron-fission) coincidences were detected.

DATA ANALYSIS

The object of the experiments we performed was to obtain the following functions: (1) the variance $\sigma_{\nu}^{2}(A, E_{K})$ of the neutron distribution emitted from fragments of mass A and total kinetic energy E_{K} , (2) the covariance $\mu(A, E_{K})$ between the neutron distributions of two complementary fragments, and (3) the variance $\sigma_{T}^{2}(A, E_{K})$ of the total number of neutrons $\nu_{T}(A, E_{K})$ emitted from the two fragments. $\sigma_{T}^{2}(A, E_{K})$ can be determined from the equation

 $\sigma_{T}^{2}(A, E_{K}) = \sigma_{v}^{2}(A, E_{K}) + \sigma_{v}^{2}(A', E_{K}) + 2\mu(A, E_{K}), \quad (4)$

where

A' = 252 - A.

In addition to these experiments we performed the " $\overline{\nu}$ experiment" to obtain new, more reliable data for $\overline{\nu}(A, E_{\kappa})$ which is necessary in order to obtain the final results of the other experiments.

In the following we describe our method of data analysis in detail, in order to allow a full analysis of the possible errors inherent in it.

A. Determination of the mass and kinetic energy of the fission fragments

The kinetic energies of the fission fragments were obtained from the pulse heights they produced in the solid-state detectors, using the calibration procedure of Schmitt, Kiker, and Williams.¹⁰ The pre-neutron emission masses and total kinetic energy were obtained using an iterative procedure similar to that of Watson et al.,¹¹ with the following modifications: (1) We adopted our own values of $\overline{\nu}(A, E_{\kappa})$ rather than those of Bowman *et al.*³ (2) The fragment masses and kinetic energies were corrected for the recoil of detected neutrons.⁹ (3) The values of $\overline{\nu}(A, E_{\kappa})$ used to determine masses and energies in events in which neutrons were detected were conditional averages; i.e., the average number of neutrons (at mass A and kinetic energy E_{κ}) subject to the condition that at least one neutron (or 2 in the " σ^2 experiment") was emitted.⁹ (4) The resulting distributions were corrected for mass and kinetic energy dispersion using a new correction method.⁹

B. Determination of the neutron detection efficiency

The detection efficiency of neutrons of velocity v, $\epsilon(v)$, was determined by normalizing our neu-

tron spectrum to that of Bowman *et al.*,² with some modifications: Since $\epsilon(v)$ was utilized in the " $\overline{\nu}$ experiment" it was desirable to determine it in this specific configuration. We therefore extrapolated the data of Bowman *et al.* to the angle θ = 0 by fitting 15 Legendre polynomials of $\cos\theta$ to their $\rho(v, \theta)$ function for every velocity in their tables. The coefficients of these polynomials were corrected for the angular dispersion of the fission detectors and the scintillators by the method of Rose.¹² We then obtained corrected values of $\rho(v, \theta = 0^{\circ})$ between v = 1 and 4.5 cm/nsec by interpolating and smoothing between the original tabulated velocities.

Our results of $\epsilon(v)$ are presented in Fig. 2, together with those of Milton and Fraser⁴ (for exactly the same scintillators). At small and large velocities, our value of $\epsilon(v)$ is high. At the low velocity this is probably due to our low discrimination level relative to that of Bowman *et al.*, whereas at the high velocity end the high value of $\epsilon(v)$ may be the results of effects of scattered prompt γ radiation, and (n, γ) and $(n, n'\gamma)$ reactions of fission neutrons with surrounding materials.⁴

C. Determination of $\overline{\nu}(A, E_K)$

We introduce the following notation: $N_T(A, E_K)$: the total number of binary coincidence events between two fission fragments, one of which (we assume the one hitting F1) has mass A, and the total kinetic energy of the two fragments is E_K .

 $N_1(A, E_K, v)$: the number of the above events that also include a neutron of velocity v detected by one of the scintillators (summed over the two scintillators).

 $N_2(A, E_k, v_1, v_2)$: the number of the above events in which a neutron of velocity v_1 is detected by PM1 and another of velocity v_2 is detected by PM2 (σ^2 configuration).

 $N_c(A, E_K, v_1, v_2)$: same definition as $N_2(A, E_K, v_1, v_2)$ but in the " μ experiment" configuration.

A' denotes 252 - A, the mass of the complementary fission fragment; ω is the solid angle of the neutron detector; v' is the neutron velocity in the c.m. frame of the fragment that emits it; v_F is the velocity of the fission fragment emitting the neutron; θ is the angle defined by the fission-detectorsource-neutron-detector axis (see Fig. 1); and $\overline{\eta}$ is the average neutron kinetic energy in the center-of-mass system.

Our determination of $\overline{\nu}(A, E_R)$ is based on the assumption that neutrons are emitted from fission fragments isotropically in the center-of-mass frame. Therefore most of the neutrons detected in the direction of one of the fission fragments generally originate from this fragment rather than from the complementary one. Bowman *et al.*^{2,3} have found that at least 90% of the emitted neutrons are consistent with the hypothesis of isotropic emission in the center-of-mass frame. Thus it is easy to integrate the neutron spectrum over the angle of emission in the center-of-mass frame, giving³

$$\overline{\nu}_{0}(A, E_{K}) = \frac{4\pi}{2N_{T}(A, E_{K})\omega} \sum_{v} \frac{N_{1}(A, E_{K}, v)}{\epsilon(v)} \times \frac{v'(v - v_{F}\cos\theta)}{v^{2}} .$$
 (5)

The subscript 0 denotes lack of higher order corrections (see following); the factor of 2 in the denominator is due to the employment of two scintillators in this experiment. The results obtained by this equation were corrected for the fact that a small portion of the detected neutrons originates from the fragment moving in the opposite direction utilizing the standard shape function $\phi(\eta/\bar{\eta})$ of Bowman *et al.*³ A correction was also introduced to account for the possibility that two neutrons can hit the same detector simultaneously (causing only a single time-of-flight event).⁹

The results obtained in this manner are essentially independent of the $\overline{\nu}(A, E_K)$ results of Bowman *et al.* despite the fact that $\epsilon(v)$ was obtained by normalizing our velocity spectrum to their spectrum. The reason for this is that the *trend* of $\overline{\nu}(A)$ and $\overline{\nu}(E_K)$ is practically independent of the exact values of $\epsilon(v)$ within reasonable limits. The *normalization* of $\overline{\nu}(A, E_K)$ is determined by normalizing the average total number of neutrons $\overline{\nu}_T$ to a currently accepted value.¹³ Thus neither the trend nor the normalization of $\overline{\nu}(A, E_K)$ is sensitive to errors in $\epsilon(v)$.

As shown elsewhere⁹ various corrections have a significant effect on the trend of the $\overline{\nu}(A, E_K)$ results. It was therefore necessary to obtain new results for $\overline{\nu}$ in our experiments and make the various necessary corrections.



FIG. 2. The neutron detection efficiency $\epsilon(v)$. The squares are the results of Milton and Fraser (Ref. 4).

D. Determination of the variance $\sigma_v^2(A, E_K)$

The variance of the neutron distribution $\sigma_{\nu}^{2}(A, E_{\kappa})$ is determined by the data obtained in single neutron and double neutron coincidence experiments as follows: We define $\psi(A, E_{K}, \overline{\eta}; v, \theta)$ to be the neutron distribution function in the laboratory system; i.e., $\psi(A, E_{\kappa}, \overline{\eta}; v, \theta) v^2 dv d\Omega$ is the probability that a neutron emitted from a fragment [of mass A, total kinetic energy E_{κ} , average neutron c.m. energy $\overline{\eta}$ $= \overline{\eta}(A, E_{K})$] will have a velocity between v and v + dvand direction within the solid angle $d\Omega$ around θ . ψ is analogous in meaning to the function $\rho(v, \theta)$ of Bowman $et al.^2$ except for being related to a given fragment of specific A and E_{κ} , and normalized to unity. ψ can be obtained from the standard shape function $\phi(\eta/\overline{\eta})$ by transforming the variable η to the laboratory system. It will be seen later however, that ψ obtained in this manner is only approximately correct. We will therefore use ψ for second-order corrections only when the small errors in ψ do not affect the results.

The probability $P(A, E_{\mathbb{R}}, \overline{\eta}, \theta)$ that a neutron emitted with velocity between v_a and v_b will produce a detectable event in PM1 or in PM2 is given by

$$P(A, E_{K}, \overline{\eta}, \theta) = \int_{v_{a}}^{v_{b}} \omega \psi(A, E_{K}, \overline{\eta}; v, \theta) \epsilon(v) v^{2} dv$$
(6)

for an infinitesimal solid angle ω . We define the following matrices of experimental results:

•1 -

$$S_{1}(A, E_{K}) = \sum_{v=v_{a}}^{v} N_{1}(A, E_{K}, v) / [2N_{T}(A, E_{K})],$$

$$S_{2}(A, E_{K}) = \sum_{v_{1}, v_{2}=v_{a}}^{v_{b}} N_{2}(A, E_{K}, v_{1}, v_{2}) / N_{T}(A, E_{K}).$$
(7)

Using the probability given in Eq. (6) we obtain to first order in P

$$S_1(A, E_{\kappa}) = \overline{\nu}(A, E_{\kappa}) P(A, E_{\kappa}, \overline{\eta}, \theta), \qquad (8)$$

$$S_2(A, E_{\mathcal{K}}) = \left[\overline{\nu^2}(A, E_{\mathcal{K}}) - \overline{\nu}(A, E_{\mathcal{K}})\right] P^2(A, E_{\mathcal{K}}, \overline{\eta}, \theta) .$$
(9)

frame we obtain

In these equations we have made a number of implicit assumptions. Among them, we have assumed that the detected neutrons are emitted from the fragment moving towards the detectors, and that the detection probabilities of two detected neutrons in coincidence are independent.

These equations have to be corrected for the following higher-order effects: (1) the possibility that a detected neutron was emitted from a fragment moving in the opposite direction to that of the scintillator³; (2) the possibility that two (or more) neutrons emitted by a single fragment hit the scintillator producing only a single time-of-flight event⁹; (3) the possibility that a neutron and a photon both produce detectable pulses in the same scintillator, thus giving rise to a photon time-of-flight event.

Corrections due to (1) were performed using the method described in Ref. (3). Corrections due to (2) and (3) were performed as described in Ref. (9).

The problem of probability correlations is not easily dealt with. Ignoring this possibility for the present, we obtain from Eqs. (8) and (9)

$$\frac{\nu^{2}(A, \underline{E}_{K}) - \overline{\nu}(A, \underline{E}_{K})}{\nu^{2}(A, \underline{E}_{K})} = \frac{S_{2}(A, \underline{E}_{K})}{S_{1}^{2}(A, \underline{E}_{K})} \left[1 + \delta_{2}(A, \underline{E}_{K})\right].$$
(10)

 $\delta_2(A, E_K)$ contains the contribution of the higherorder terms which affect the ratio by roughly 5%.¹⁴ These terms depend on the calculated probabilities P and are therefore affected by errors in $\epsilon(v)$ and in the standard shape function ϕ by which P is determined.

We recall that Eq. (10) involves no special assumptions regarding the angular or velocity distribution of neutrons, other than assuming that the distribution of each neutron is independent of all others. If this assumption is incorrect, the effect of correlations has to be considered. This can be done by assuming that the neutrons are emitted isotropically in the fragment c.m. frame and that there are no *angular* correlations. Then by integrating over the angles of emission in the c.m.

$$\overline{\nu^{2}}(A, E_{K}) - \overline{\nu}(A, E_{K}) = \frac{(4\pi)^{2}}{\omega^{2} N_{T}(A, E_{K})} \sum_{v_{1}, v_{2}} \frac{N_{2}(A, E_{K}, v_{1}, v_{2})}{\epsilon(v_{1})\epsilon(v_{2})} \frac{v_{1}'(v_{1} - v_{F}\cos\theta)}{v_{1}^{2}} \frac{v_{1}'(v_{2} - v_{F}\cos\theta)}{v_{2}^{2}} .$$
(11)

The assumptions on which Eq. (11) is based cannot be considered more accurate than the assumptions on which Eq. (9) is based. The basic assumption of lack of velocity correlations in the c.m. frame, which can be only approximately correct, was replaced by the assumption of isotropic emission which is correct only for 90% of the emitted neutrons.

E. Determination of the covariance $\mu(A, E_K)$

The covariance of the two neutron distributions in two complementary fragments is defined in Eq. (2). It can be determined in the " μ experiment" configuration [PM1 at position II in Fig. (1)] by measuring the number of coincidence events which include two neutrons, one emitted by each fragment. Defining the S_c matrix of experimental data as

$$S_c(A, E_K) = \sum_{v_1, v_2} N_c(A, E_K, v_1, v_2) / N_T(A, E_K),$$

we obtain to first order in P:

$$S_{c}(A, E_{K}) = \operatorname{av}[\nu(A, E_{K})\nu(A', E_{K})]$$
$$\times P(A, E_{K}, \overline{\eta}, \theta)P(A', E_{K}, \overline{\eta}, \theta) \qquad (12)$$

where

A' = 252 - A.

Combining with Eq. (8) (written once for A and once for A') we obtain

$$\frac{\operatorname{av}[\nu(A, E_{K})\nu(A', E_{K})]}{\overline{\nu}(A, E_{K})\overline{\nu}(A', E_{K})} = \frac{S_{c}(A, E_{K})}{S_{1}(A, E_{K})S_{1}(A', E_{K})} \times [1 + \delta_{c}(A, E_{K})].$$
(13)

 $\delta_c(A, E_K)$ contains the contribution of the higher-order terms which affect the ratio by roughly 5%.¹⁴

F. Scission neutron corrections

The existence of scission neutrons in fission of ²⁵²Cf modifies the various equations we have presented because of their different spectrum and angular distributions. Very'little is known about the properites of these neutrons. We therefore make some plausible assumptions about them in order to perform the necessary corrections. The corrections themselves are discussed in Appendix A. Their accuracy is discussed in Appendix B in the framework of the general analysis of errors.

We assume the following:

(1) 0.4 scission neutrons are emitted per fission in the velocity range of 1.0 to 4.0 cm/nsec. These neutrons are assumed to have a Maxwellian energy distribution with a mean energy of 1.5 MeV.

TABLE I. Comparison of various moments of the yield $Y(A, E_K)$ to those of Whetstone (Ref. 17) and of Schmitt, Neiler, and Walter (Ref. 18). The statistical error in the various moments is approximately 0.1%. Notation: $\overline{A}_L(\overline{A}_H)$, average light (heavy) fragment mass; σ_{AL} , standard deviation of light mass yield; \overline{E}_K , standard deviation of total kinetic energy; σ_{E_K} , standard deviation of total kinetic energy yield.

| | Our results | Whetstone | Schmitt et al. | Unit |
|--------------------|----------------|--------------|----------------|------------|
| \overline{A}_L | 109.0 | 108.4 | 108.6 | amu |
| | 6.77 | 6.77 | 6.72 | amu |
| $E_K \sigma_{E_K}$ | 187.3 11.06 | 186.6 11.3 | 186.5 12.0 | MeV MeV |

(2) Scission neutrons are distributed isotropically in the laboratory system. (Fragment neutrons are assumed to be likewise distributed in the fragment c.m. system.)

(3) The total number of fragment neutrons is correlated with the total number of scission neutrons. Specifically, we assume that if a scission neutron is emitted in a fission event, one of the fragments emits one neutron less. We also assume that not more than one scission neutron is emitted in a fission event.

The average value of 0.4 scission neutrons is compatible with the results of Bowman *et al.*² and of Mayek.¹⁵ The assumed energy spectrum is entirely *ad hoc*; however (see Appendix B), the results are not sensitive to it.

The third assumption is based on the properties of long-range α (LRA) emission.^{8, 16} These results indicate that when a LRA particle is emitted, the average number of emitted neutrons is decreased by 0.7. Since the $\overline{\nu}_L/\overline{\nu}_H$ ratio is practically unaffected by this, we assume that the decrease is shared equally between the two fragments. If we assume that long-range α particles and scission neutrons are emitted by the same mechanism our third assumption seems plausible.



FIG. 3. The pre-neutron emission mass yield Y, the average total kinetic energy \overline{E}_K , and the total kinetic energy standard deviation σ_{E_K} as a function of the mass A. The standard errors are too small to be shown.

These assumptions are sufficient to perform the corrections described in Appendix A. The variance we will henceforth present will be of *the fragment* neutron distribution unless otherwise stated.

RESULTS

A. Mass yield and kinetic energy distribution

We present the results we obtained for the various quantities associated with the masses and kinetic energies of the fission fragments (all the quantities are pre-neutron emission ones). Table I summarizes average masses of the light (L) and heavy (H) fragments, their variance, and the average and variance of the total kinetic energy distribution. These results are compared to those of Whetstone¹⁷ and of Schmitt, Neiler, and Walter.¹⁸ The statistical errors in our values are roughly 0.1%.

Figure 3 presents the mass yield Y(A), the average and the standard deviation of the total kinetic energy E_K as a function of the mass A. Compared to other data in the literature,^{17, 18} the agreement is good except in the region of symmetric mass distribution; in this region the results are affected by mass dispersion^{11, 19} and vary according to the type of correction used.

B. Average neutron distribution

In Fig. 4 we present the average number of neutrons $\overline{\nu}$ emitted as a function of the mass A and as a function of E_K (these results are not corrected for scission neutrons). We also present the average derivative of $\overline{\nu}$ with respect to E_K , $\langle \partial \overline{\nu} / \partial E_K \rangle$, as a function of A. The results of $\overline{\nu}(A)$ are compared to those of Signarbieux *et al.*²⁰ Our results of $\overline{\nu}$ were renormalized to the latest value of the average total number of neutrons¹³ of 3.725. We obtain a ratio of $\overline{\nu}_L/\overline{\nu}_H = 1.05$ which is between that obtained by Bowman *et al.*³ and by Nardi and Fraenkel.⁸ The average value of $(\partial \overline{\nu}_T / \partial E_K)$ (A, E_K) is -0.125 MeV⁻¹.

The average kinetic energy of the neutrons in the c.m. system $\overline{\eta}(A)$ is also presented in Fig. 4. The average value we obtain for $\overline{\eta}$ is 1.30 MeV.

The accuracy of the detection probability $P(A, E_K, \overline{\eta}; \theta)$ was determined for $\theta = 0^\circ$ by determining $\overline{\nu}(A, E_K)$, once using Eq. (5) and once using Eq. (8). In order to calculate P, we used the standard shape function of Bowman *et al.*³ together with our own results of $\overline{\nu}(A, E_K)$. We conclude in general that for kinetic energies above 170 MeV the two results agree precisely. For lower kinetic energies Eq. (8) gives a result which is somewhat low. At $E_K = 160$ MeV the error in $\overline{\nu}_0$ (and thus in P) is of the order of 10% when using Eq. (8); there-

fore the values of *P*are sufficiently accurate to use use in higher-order corrections, due to their small effect.

C. Results of the variance and covariance

As we have already explained, the values of σ_{ν}^{2} are obtained from those of $av[\nu(\nu-1)]/\overline{\nu}^{2}$ and the values of the covariance μ from $av[\nu_{L}\nu_{H}]/\overline{\nu_{L}}\overline{\nu}_{H}$.

In the following we shall discuss several types of variances and covariances: (1) the variance $\sigma_{\nu}^{2}(A, E_{K})$ and covariance $\mu(A, E_{K})$ at given mass and kinetic energy defined in Eqs. (1) and (2), and (2) averages of these values over the mass or over the total kinetic energy or over both. We denote

$$\langle \sigma_{\nu}^{2} \rangle \langle A \rangle = \sum_{E_{K}} Y(A, E_{K}) \sigma_{\nu}^{2} \langle A, E_{K} \rangle / \sum_{E_{K}} Y(A, E_{K}) ,$$

$$\langle \mu \rangle \langle A \rangle = \sum_{E_{K}} Y(A, E_{K}) \mu \langle A, E_{K} \rangle / \sum_{E_{K}} Y(A, E_{K}) ,$$

$$(14)$$

and similarly $\langle \sigma_v^2 \rangle (E_K)$ and $\langle \mu \rangle (E_K)$ (average over A) and $\langle \sigma_v^2 \rangle$ and $\langle \mu \rangle$ (average over A and E_K). These averages are to be distinguished from the



FIG. 4. The average number of neutrons $\overline{\nu}$, the average derivative of $\overline{\nu}$ with respect to E_K , and the average c.m. energy of the neutrons $\overline{\eta}$ as function of A. Our results are presented by filled circles. The $\overline{\nu}$ results of Signarbieux *et al.* are shown for comparison (open circles). The statistical errors are too small to be shown.

neutron variance and covariance at a given mass A where $E_{\mathbf{F}}$ is free to vary (and vice versa). These last quantities we denote as $\Sigma_{\nu}^{2}(A)$ and $\mathfrak{M}(A)$. They are defined as

$$\Sigma_{\nu}^{2}(A) = \overline{\nu}^{2}(A) - \overline{\nu}^{2}(A),$$

$$\mathfrak{M}(A) = \operatorname{av}[\nu(A)\nu(A')] - \overline{\nu}(A)\overline{\nu}(A').$$
 (15)

The bar and the av[] function denote averages over all the variables we do not measure; in this case E_{κ} is one of them. Likewise we can define $\Sigma_{\nu}^{2}(E_{\kappa})$ and $\mathfrak{M}(E_{\kappa})$ as the variance and covariance regardless of A.

In addition we define the variance and covariance of the *average* number of neutrons $S^2(A)$ and M(A);

$$S_{\nu}^{2}(A) = \langle \overline{\nu}^{2}(A, E_{K}) \rangle_{B_{K}} - \langle \overline{\nu}(A, E_{K}) \rangle_{B_{K}}^{2},$$

$$M(A) = \langle \overline{\nu}(A, E_{K}) \overline{\nu}(A', E_{K}) \rangle_{B_{K}}$$

$$- \langle \overline{\nu}(A, E_{K}) \rangle_{E_{K}} \langle \overline{\nu}(A', E_{K}) \rangle_{B_{K}}.$$
(16)

The angular brackets denote here average over E_K . Likewise we can define $S^2(E_K)$ and $M(E_K)$ (averages over A) and S^2 and M (averages over A and E_K). One can easily show that

$$\Sigma_{\nu}^{2} = S_{\nu}^{2} + \langle \sigma_{\nu}^{2} \rangle,$$

$$\mathfrak{M} = M + \langle \mu \rangle$$
(17)

(the brackets can imply average over A, E_{R} or both these variables.)

TABLE II. Results of various averages using different assumptions regarding fragment neutron correlations and scission neutron-fragment neutron correlations. Assumptions: (1) No angular or velocity correlations in the c.m. system, no scission neutrons [Eq. (10)]. (2) Isotropic evaporation in c.m. system, no scission neutrons [Eq. (11)]. (3) Isotropic evaporation in c.m. system of fragment neutrons, 0.4 scission neutrons emitted. No correlation between number of fragment neutrons and number of scission neutrons. (4) Same as (3), but assuming negative correlation between number of fragment neutrons and number of scission neutrons.

| | Assumption No. | | | | |
|----------------------------------|----------------|-------|-------|-------|--|
| | (1) | (2) | (3) | (4) | |
| $\overline{\nu}_{f}$ | 3.72 | 3.72 | 3.32 | 3.32 | |
| $\overline{\nu}_s$ | 0 | 0 | 0.40 | 0.40 | |
| $\langle \sigma_L^2 \rangle$ | 0.95 | 0.71 | 0.61 | 0.66 | |
| $\langle \sigma_{\!H}^2 \rangle$ | 1.04 | 0.82 | 0.72 | 0.79 | |
| $\langle \mu \rangle$ | -0.23 | -0.23 | -0.25 | -0.18 | |
| $\langle \sigma_T^2 \rangle$ | 1.53 | 1.07 | 0.83 | 1.09 | |
| S_{T}^{2} | 1.38 | 1.38 | 1.18 | 1.18 | |
| $\langle \sigma_s^2 \rangle$ | 0 | 0 | 0.24 | 0.24 | |
| $\langle \mu_{s} \rangle$ | 0 | 0 | 0 | -0.24 | |
| Σ_T^2 | 2.91 | 2.45 | 2.25 | 2.03 | |

Similar equations are obtained for the total number of neutrons $\overline{\nu}_{T}$: Defining

$$S_T^2 = \langle \overline{\nu}_T^2(A, E_K) \rangle_{A, E_K} - \langle \overline{\nu}_T(A, E_K) \rangle_{A, E_K}^2$$
(18)

(the angular brackets denote averages over A and E_{K}),

$$\sigma_T^{2}(A, E_K) = \overline{\nu_T^{2}}(A, E_K) - \overline{\nu}_T^{2}(A, E_K), \qquad (19)$$

$$\langle \sigma_T^2 \rangle = \langle \sigma_T^2(A, E_K) \rangle_{A, B_K}.$$
 (20)

Using Eqs. (18)-(20) we calculate Σ_r^2 , the variance of the total number of neutrons regardless of the fission made. This result can be compared to that obtained from multiplicity measurements of neutrons using large gadolinium-loaded liquid-scintillation tanks.²¹⁻²³

Results of these various averages are presented in Table II, using different assumptions regarding fragment neutron velocity correlations and scission neutron-fragment neutron correlations. In this table, σ_s^2 denotes the variance of the scission neutrons, and the covariance of the total fragment neutron ($\nu_{\rm tf}$) and the scission neutron distributions, μ_s , is

$$\mu_{s} = \langle \nu_{\rm tf} \nu_{s} \rangle - \langle \nu_{\rm tf} \rangle \langle \nu_{s} \rangle, \qquad (21)$$



FIG. 5. $\langle \sigma_v^2 \rangle$, Σ_v^2 , $\langle \sigma_T^2 \rangle$, and Σ_T^2 variances and $\langle \mu \rangle$ and \mathfrak{M} covariances as a function of mass A. (a) $\Sigma_v^2(A)$ (the open circles are the results of Signarbieux *et al.*); (b) $\langle \mu \rangle \langle A \rangle$, open circles; $\mathfrak{M}(A)$, full triangles; (c) $\Sigma_T^2 \langle A \rangle$; (d) $\langle \sigma_v^2 \rangle \langle A \rangle$, filled circles; $\langle \sigma_T^2 \rangle \langle A \rangle$, filled triangles. The statistical errors can be inferred from fluctuations between neighboring points.

 Σ_{τ}^{2} is given by

$$\Sigma_{\mathbf{T}}^{2} = \langle \sigma_{\mathbf{T}}^{2} \rangle + S_{\mathbf{T}}^{2} + \langle \sigma_{\mathbf{s}}^{2} \rangle + 2 \langle \mu_{\mathbf{s}} \rangle.$$

Considering Table II it is evident that assumption (4) provides a value of Σ_r^2 which is closest to that of 1.6 obtained in liquid-scintillation-tank experiments.²¹⁻²³ We will therefore present all our results using assumption (4). The difference between our Σ_r^2 results and those of Refs. 21-23 should be considered as indicative of the order of magnitude of the systematic errors involved in our results. (For complete error analysis see Appendix B).

Results of $\langle \sigma_v^2 \rangle(A)$, $\Sigma_v^2(A)$, $\langle \mu \rangle(A)$, $\langle \sigma_T^2 \rangle(A)$, $\mathfrak{M}(A)$, and $\Sigma_T^2(A)$ are presented in Fig. 5. The $\Sigma_v^2(A)$ results of Signarbieux *et al.*²⁰ are shown for comparison. In Fig. 6 we show the $\langle \sigma_v^2 \rangle(E)$, $\langle \mu \rangle(E)$, and $\Sigma_v^2(E)$ values. The statistical errors are not shown since they are generally much smaller than the systematic ones.

As expected from our comparison of Σ_r^2 values, our $\Sigma_v^2(A)$ results are generally higher than those of Signarbieux. Our $\mathfrak{M}(A)$ values are slightly positive (approximately +0.1) whereas those of Signarbieux *et al.*²⁰ are slightly negative. However, the results of Ref. 20 were not corrected for scission neutron emission and part of the difference could be due to them.

DISCUSSION

The variance of the neutron distribution at a given mass and kinetic energy is a result of three



FIG. 6. $\langle \sigma_{\nu}^2 \rangle$, $\langle \mu \rangle$ (circles), and Σ_T^2 (triangles) as function of total kinetic energy E_K . The statistical errors can be inferred from fluctuations between neighboring points.

different factors: (1) the statistical nature of the neutron evaporation process—various evaporation cascades produce different numbers of neutrons; (2) the effect of the correlation between the excitation energies of the two fragments; and (3) the effect of variations of the fragment charge.

The contribution of the first two effects was evaluated using the Monte-Carlo evaporation code of Nardi, Moretto, and Thompson.²⁴ The correlation of the excitation energies of the two fragments causes fragment excitation energy fluctuations with a standard deviation of approximately 3 MeV. The average excitation energy is determined by the average number of neutrons emitted by the fragment. Using the average excitation energy and its standard deviation as input data to the computer program²⁴ we calculated the variance of the number of neutrons emitted from each fragment. In order to check the results, we performed a similar calculation using the evaporation code of Dostrovsky, Fraenkel, and Friedlander.²⁵ For both cases we obtained $\langle \sigma_v^2 \rangle = 0.2 - 0.3$ for fixed mass A. This is smaller than both our $\langle \sigma_{\nu}^{2} \rangle$ values and those of Babinet et al.²⁶

The effect of charge variation was estimated to contribute an average variance of 0.03 units, which is much too small to explain the discrepancy between the evaporation calculations and the experimental results.

There is an additional discrepancy between the calculations and experimental results concerning the average center-of-mass neutron kinetic energy. In our experiments we have obtained: $\langle \eta \rangle^{(1)}$, the average c.m. energy in events in which one neutron was detected; $\langle \eta \rangle^{(2)}$, the average in events in which two neutrons were detected; $\langle \eta_1 \eta_2 \rangle^{(2)}$, the average



FIG. 7. Center-of-mass kinetic energy of neutrons in " σ^2 experiment" configuration. $\langle \eta_1 \eta_2 \rangle$ as function of A denoted by circles; $\langle \eta \rangle^2$ as function of A denoted by triangles. The statistical errors are too small to be shown.

of the product of the c.m. energies of two neutrons emitted from the same fragment. All these quantities were measured in the same (" σ^2 ") experimental configuration in order to avoid biasing of the results due to different scattering effects. We obtain essentially that

$$\langle \eta \rangle^{(1)} = \langle \eta \rangle^{(2)}$$

and

$$\left[\langle \eta \rangle^{(2)}\right]^2 = \langle \eta_1 \eta_2 \rangle^{(2)}$$

The differences are of the order of 1%, and do not show any significant systematic behavior as a function of the fragment mass (see Fig. 7) or of the total kinetic energy. The physical implications of these results are: (1) There is no correlation between the average center-of-mass kinetic energy of the neutrons and the total excitation energy; and (2) there is no correlation of evaporation energies of two neutrons in an evaporation cascade.

These results have no simple explanation. If the total excitation energy E_x was constant in a given nucleus we would expect a negative correlation between η_1 and η_2 , the c.m. kinetic energies of neutrons "1" and "2" in the cascade. This is because an increase in η_1 leaves less excitation energy for the emission of the second neutron. However, if E_x varies we would expect η_1 to increase as E_x increases. Thus an increase in η_1 implies an increase in the total initial excitation energy which in turn causes an increase in the kinetic energy of all the neutrons. Thus the experimental lack of correlation between two neutrons could be explained by the existence of two correlation mechanisms which cancel each other. It is, however, most surprising that this cancellation exists in the entire range of fragment masses and excitation

energies.

We have no explanation for the lack of correlation between the average kinetic energy and the total excitation energy. Fragments emitting one neutron on average should exhibit relatively high neutron c.m. kinetic energies when two neutrons are detected. Evaporation calculations show that an 0.1-0.2-MeV difference should be observed. In practice, the observed difference is an order of magnitude smaller and could be due to recoil effects, dispersion effects, etc.

Regarding conclusions pertaining to the degree of viscosity of the fragments when they approach the scission point, we note that although μ approaches zero when the total excitation energy decreases (see Fig. 6), the covariance per neutron $\mu/\bar{\nu}_L \bar{\nu}_H$ remains constant. Thus the *fraction* of the total excitation energy that can be exchanged remains constant. We find no indication for increased viscosity either at low or at high excitation energies, which is not in agreement with the conclusion of Babinet *et al.*²⁶

APPENDIX A: SCISSION NEUTRON CORRECTIONS

We present the analysis of the effect of scission neutrons on $\sigma_v^2(A, E_k)$ and on $\mu(A, E_k)$. We will assume throughout that the analysis is performed at mass A and kinetic energy E_k and therefore omit these variables. We denote: ν_f , the number of fragment neutrons; ν_s , the number of scission neutrons; P_f , the fragment neutron detection probability; P_s , the scission neutron detection probability; P_1 , the probability of detecting a single neutron; P_2 , the probability of detecting two neutrons in the " σ^2 configuration," P_c , the probability of detecting two neutrons in the " μ configuration."

Obviously

$$P_1 = \nu_f P_f + \nu_s P_s , \tag{A1}$$

$$P_2 = \operatorname{av}[\nu_f(\nu_f - 1)]P_f^2 + 2\operatorname{av}[\nu_f \nu_s]P_f P_s , \qquad (A2)$$

$$P_{c} = \operatorname{av}\left[\nu_{fL}\nu_{fH}\right]P_{fL}P_{fH} + \operatorname{av}\left[\nu_{fL}\nu_{s}\right]P_{fL}P_{s} + \operatorname{av}\left[\nu_{fH}\nu_{s}\right]P_{fH}P_{s} .$$
(A3)

The subscript L and H denote light and heavy fragments, respectively. In Eq. (A2) we make use of the assumption that not more than one scission neutron is emitted per fission event. Assuming that $v_s P_s \ll v_f P_f$ we obtain

$$\frac{P_2}{P_1^2} = \frac{\operatorname{av}[\nu_f(\nu_f - 1)]}{\overline{\nu}_f^2} \left(1 + \frac{2\operatorname{av}[\nu_f \nu_s]}{\operatorname{av}[\nu_f(\nu_f - 1)]} \frac{P_s}{P_f} - \frac{2\overline{\nu}_s P_s}{\overline{\nu}_f P_f} \right) \quad , \tag{A4}$$

$$\frac{P_{c}}{P_{1L}P_{1H}} = \frac{\operatorname{av}[\nu_{fL}\nu_{fH}]}{\overline{\nu}_{fL}\overline{\nu}_{fH}} \left(1 + \frac{\operatorname{av}[\nu_{fL}\nu_{s}]}{\operatorname{av}[\nu_{fL}\nu_{fH}]} \frac{P_{s}}{P_{fH}} + \frac{\operatorname{av}[\nu_{fH}\nu_{s}]}{\operatorname{av}[\nu_{fL}\nu_{fH}]} \frac{P_{s}}{P_{fL}} - \frac{\overline{\nu}_{s}P_{s}}{\overline{\nu}_{fL}P_{fL}} - \frac{\overline{\nu}_{s}P_{s}}{\overline{\nu}_{fH}P_{fH}}\right).$$
(A5)

The correction factors f_2 and f_c for the probability ratios (A4) and (A5) due to scission neutrons are

1 + + 1

therefore

$$f_2 = 1 + 2 \frac{P_s}{P_f} \left(\frac{\operatorname{av}[\nu_f \nu_s]}{\operatorname{av}[\nu_f(\nu_f - 1)]} - \frac{\overline{\nu}_s}{\overline{\nu}_f} \right) \quad , \tag{A6}$$

$$f_{c} = 1 + \frac{P_{s}}{P_{fL}} \left(\frac{\operatorname{av}[\nu_{fH}\nu_{s}]}{\operatorname{av}[\nu_{fL}\nu_{fH}]} - \frac{\overline{\nu}_{s}}{\overline{\nu}_{fL}} \right) + \frac{P_{s}}{P_{fH}} \left(\frac{\operatorname{av}[\nu_{fL}\nu_{s}]}{\operatorname{av}[\nu_{fL}\nu_{fH}]} - \frac{\overline{\nu}_{s}}{\overline{\nu}_{fH}} \right). \tag{A7}$$

In order to calculate f_2 and f_c we need to know the following: (1) the detection probabilities P_s and P_f (this demands knowledge of the angular and velocity distribution of the fragment and scission neutrons), and (2) the number of fragment and scission neutrons, their variance and covariance.

Using the assumptions regarding the scission neutrons which were previously presented, f_2 and f_c were calculated. These factors enable us to obtain the final values of the variance and covariance of the fragment neutron distribution.

APPENDIX B: DISCUSSION OF ERRORS

A. General remarks

As already noted, our value of σ_r^2 is larger than values obtained in 4π geometry measurements using gadolinium-loaded liquid-scintillation tanks. We assume that this discrepancy is indicative of the magnitude of systematic errors in all our σ_v^2 results. We discuss below in detail the various errors inherent in our experiment and their effect on our results.

The errors can be divided into two different classes: (1) errors due to processes which interfere with neutron detection, and (2) errors due to incomplete knowledge of the details of neutron emission in fission. Causes for errors which belong to the first class are: (a) neutron seattering, especially from one detector to the other, and (b) emission of delayed γ radiation.

The effects about which we do not have the sufficient information to perform accurate corrections are: (a) the spectrum and angular distribution of scission neutrons, (b) the correlation of the number of scission neutrons with the number of the fragment neutrons, (c) deviation of the angular distribution of fragment neutrons from the assumed isotropic distribution in the c.m. frame, (d) angular correlations between fragment neutrons, (e) velocity correlations (in the c.m. system) between fragment neutrons. We next analyze the effect of the various errors on our results.

B. Effect of neutron scattering

The effect of neutron scattering has to be taken into account only as far as the scattering effects increase the number of particles (neutrons and photons). This is because the detection efficiency $\epsilon(v)$ is actually an *effective* efficiency which normalizes the experimental neutron spectrum to that of Bowman *et al.*² However, if the effective number of particles is increased [such as in $(n, n'\gamma)$ reactions or by scattering from one detector to the other], the number of double neutron events is increased and this affects the measured values of σ_{ν}^{2} and μ .

These effects can be taken into account by reducing the size of the neutron velocity interval in which the number of neutron events is counted. When considering neutrons with velocity between 2.0 and 2.5 cm/nsec all events due to scattering of neutrons from one detector to the other are eliminated. A neutron hitting one detector with a velocity of 2.5 cm/nsec would appear to have a velocity below 2.0 cm/nsec if scattered and redetected by the second detector. These velocity limits also discriminate against $(n, n'\gamma)$ events, in which the detected photon appears as a fast neutron.

Limiting the neutron velocity range to 2.0–2.5 cm/nsec reduces the av $[\nu(\nu-1)]/\overline{\nu}^2$ ratio by 3% and does not affect the av $[\nu_L \nu_H]/\overline{\nu}_L \overline{\nu}_H$ ratio. The reduction is independent of the mass and kinetic energy of the fragments. Since we have found that the neutron velocity correlations are too small to be responsible for this decrease (see below), we assume that it was entirely due to the elimination of the various scattering processes. All results of the av $[\nu(\nu-1)]/\overline{\nu}^2$ were therefore subsequently reduced by 3% in order to correct the results for the effects of neutron scattering.

C. Emission of delayed γ radiation

Delayed γ radiation would be identified as neutrons if emitted in the time range corresponding to the velocity limits used in the various results. Measurement of the total delayed γ radiation has been performed by Skarsvag.²⁷ From his results we find that approximately 5% of the total number of photons emitted would be detected as neutrons in the " σ^2 " and " μ " experiments, and approximately 3% in the " ν experiment." John *et al.*²⁸ found that the chief delayed γ emitters in this time interval have masses of 91, 95, 134, and 146 amu.

No significant fluctuations are discernable in our $\sigma_{\nu}^{2}(A)$ or $\mu(A)$ results at these masses (or at those of the complementary fragments). If indeed 5% of the total number of photons are identified as neutrons, this should cause an increase of approximately 0.1 units in σ_{ν}^{2} and 0.02 units in μ . This increase is sufficient to explain a significant part of the difference between our Σ_{ν}^{2} result (see Table II) and previous values.²¹⁻²³ However no accurate knowledge of the dependence of this increase on the fragment mass and total kinetic energy is available and therefore no correction for delayed γ radiation was performed.

D. Scission neutrons

The large effect of scission neutrons on the variance and covariance of fragment neutrons has already been noted (see Table II); therefore accurate corrections for their effects are important. These corrections demand accurate knowledge of the number of scission neutrons, their correlation with fragment neutrons, their angular distribution, and velocity spectrum.

Bowman *et al*.² have found that about 10% of the emitted neutrons in their experimental velocity range can be ascribed to an isotropic angular distribution in the laboratory system. Mayek¹⁵ found that the number of scission neutrons is independent of the fragment mass and kinetic energy and is equal to 0.4 ± 0.1 neutrons. However, since the neutron detection efficiency he used was obtained by normalizing his neutron spectrum to that of Bowman *et al*.,² the average number he obtained is not an independent result.

The energy spectrum of the scission neutrons determines the average detection probability P_s (see Appendix A). Since $\epsilon(v)$ does not change rapidly as a function of the neutron velocity v (see Fig. 2), the exact shape and average energy of the spectrum do not have a large effect on the magnitude of the correction. Errors of the order of 0.05 units in $\langle \sigma_v^2 \rangle$ and 0.01 units in $\langle \mu \rangle$ are possible if we assume that scission neutrons have a Maxwellian energy distribution with an average energy of 1.5 MeV and that the error in the average energy is 0.5 MeV.

The possibility of correlation between the number of fragment neutrons and the number of scission neutrons introduces an additional error. Considering Table II, columns (3) and (4), we see that this effect can cause an error of approximately 0.05 units in $\langle \sigma_v^2 \rangle$ and 0.03 units in $\langle \mu \rangle$.

The uncertainty in the average number of scission neutrons causes a similar uncertainty in the normalization of the total number of fragment neutrons. This gives rise to an error of about 0.05 in S_T^2 in columns (3) and (4) in Table II.

Summarizing the errors due to scission neutrons, we find: (1) an error of approximately 0.08 in $\langle \sigma_{\nu}^{2} \rangle$, (2) an error of approximately 0.03 in $\langle \mu \rangle$, and (3) an error of 0.05 in S_{T}^{2} .

As a result, the total error in Σ_T^2 due to scission neutrons is about 0.2.

E. Angular distribution of fragment neutrons

In Eqs. (5) and (11) we have assumed that the detected neutrons are emitted isotropically in the fragment c.m. system. Apart from scission neutrons (which are assumed to be emitted isotropically in the laboratory system) deviation from this assumption is possible due to the angular momentum of the fragments. Wilhelmy et al.29 find that fission fragments of ²⁵²Cf have an average angular momentum of 7 units which is aligned perpendicular to the fission axis. Neutron evaporation causes an average decrease of 1 unit per neutron.³⁰ We denote by J the fragment spin, Mits projection on the fission axis, l and m the orbital angular momentum and projection of the emitted neutron. The probability ratio for emitting a neutron with $(l=1, m=\pm 1)$ and (l=1, m=0) is

$$\left|\frac{\langle J,M | J_f, M_f, l=1, m=\pm 1 \rangle}{\langle J,M | J_f, M_f, l=1, m=0 \rangle}\right|^2.$$

Assuming $J_f = J - 1$, $M \ll J$ (see Ref. 29), we find that for $J \sim 7$, the angular distribution in the c.m. system is approximately given by $1 + 0.2 \cos^2 \theta'$. The effect of this anisotropy on $\langle \sigma_{\nu}^2 \rangle$ and $\langle \mu \rangle$ is estimated to be small relative to other errors. The reason for this is that the forward focusing of neutrons due to the angular momentum is small relative to that caused by the fragment motion.

Angular correlations between fragment neutrons could be induced by the high angular momentum of the fragments. However, we have already noted that since the fragment spins are aligned initially in a perpendicular direction to the fission axis, the anisotropy in the c.m. system is determined by the ratio (J+1)/J. Since the anisotropy is small, detection of a neutron in any direction (in the c.m. system) does not determine J or M to any reasonable precision. All other neutrons in this cascade will be emitted with approximately the same degree of anisotropy and this rules out the existence of any significant correlations.

F. Velocity correlations between fragment neutrons

Velocity correlations between two neutrons in the fragment center of mass have an important effect in the determination of σ_{ν}^{2} using Eq. (10). As explained in the Discussion, correlations which are expected from evaporation cascade considerations are not detected experimentally. When comparing results of $av[\nu(\nu-1)]/\overline{\nu}^2$ obtained from Eqs. (10) and (11), we find that Eq. (11) gives a result which is smaller by about 5%. However, this difference decreases rapidly as the size of the neutron velocity range is decreased. Therefore the observed difference could be due to scattering effects we have previously mentioned. The 3% decrease in the av $[\nu(\nu-1)]/\overline{\nu}^2$ ratio performed (which we found to be equivalent to decreasing the velocity range) leaves us with the necessity to decrease these results by a further 2%, if indeed the difference is due to physical correlation effects. Since a 1-2% neutron c.m. kinetic energy shift can be caused by various recoil and dispersion effects, we did not perform this correction. This gives rise to an error of 0.05-0.1 units in $\langle \sigma_{v}^{2} \rangle$.

G. Summary

In this Appendix we have enumerated the various sources of errors in this experiment and evaluated their contribution to the total error. Accurate corrections can be made for some of these errors: Effects of neutron scattering and of velocity correlations can be determined with considerable precision, and their contribution to the single and

- ¹W. E. Stein and S. J. Whetstone, Jr., Phys. Rev. <u>110</u>, 476 (1958).
- ²H. R. Bowman, S. G. Thompson, J. C. D. Milton, and W. J. Swiatecki, Phys. Rev. <u>126</u>, 2120 (1962).
- ³H. R. Bowman, J. C. D. Milton, S. G. Thompson, and W. J. Swiatecki, Phys. Rev. <u>129</u>, 2133 (1963); UCRL Report No. UCRL-10139, 1962 (unpublished).
- ⁴J. C. D. Milton and J. S. Fraser, in *Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, 1965* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 39.
- ⁵H. Nifenecker, private communication.
- ⁶J. Terrell, in *Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, 1965* (see Ref. 4), Vol. II, p. 3.
- ⁷H. Nifenecker, C. Signarbieux, R. Babinet, and J. Poitou, in Proceedings of the Third Symposium on the Physics and Chemistry of Fission, Rochester, 1973 (to be published).
- ⁸E. Nardi and Z. Fraenkel, Phys. Rev. Lett. <u>20</u>, 1248 (1968); Phys. Rev. C <u>2</u>, 1156 (1970).
- ⁹A. Gavron, Nucl. Instrum. Methods (to be published).

¹⁰H. W. Schmitt, W. E. Kiker, and C. W. Williams, Phys. Rev. 137, B837 (1965).

¹¹R. L. Watson, J. B. Wilhelmy, R. C. Jared, C. Rugge, H. R. Bowman, S. G. Thompson, and J. O. Rasmussen, double neutron coincidence rate can be subtracted. Other effects can only be corrected in part. These effects are:

(1) Delayed γ radiation. The over-all magnitude of this effect in known but its possible mass dependence does not allow its accurate subtraction from the detected neutrons. The error in $\langle \sigma_v^2 \rangle$ due to this effect is 0.1, the error in $\langle \mu \rangle$ is 0.02, and thus the error in $\langle \sigma_T^2 \rangle$ is 0.2.

(2) Scission neutrons. The effect of scission neutrons was corrected using available data. However, since these data are insufficient to perform a complete analysis of the effect of scission neutrons, an error of 0.2 units in Σ_T^2 seems likely.

We see that these effects may be responsible for most of the discrepancy between our final value of $\Sigma_T^2 = 2.03$ and the accepted value of 1.6. In order to eliminate the various errors, future experiments designed to determine $\langle \sigma_v^2 \rangle$ and $\langle \mu \rangle$ should be performed by complete measurement of the angular distribution and angular and velocity correlations of the emitted neutrons. The effect of delayed γ radiation should be accurately evaluated and subtracted from the experimental neutron spectra. This should permit measurements with errors of less than 10%.

In the present experiment, the error in $\langle \sigma_v^2 \rangle$ and in $\langle \mu \rangle$ is roughly 25%. Thus $\langle \mu \rangle$ is determined here with a small absolute error due to the small effect of the various errors on the av $[\nu_L \nu_H]/\bar{\nu}_L \bar{\nu}_H$ ratio.

- Nucl. Phys. A141, 449 (1970).
- ¹²M. E. Rose, Phys. Rev. <u>91</u>, 610 (1953).
- ¹³A. Volpi and K. G. Porgess, Phys. Rev. C <u>1</u>, 683 (1970).
- ¹⁴A. Gavron, Ph.D. thesis, Weizmann Institute of Science, 1972 (unpublished).
- ¹⁵I. Mayek, M. Sc. thesis, Weizmann Institute of Science, 1973 (unpublished).
- ¹⁶G. K. Mehta, J. Poitou, M. Ribrag, and C. Signarbieux, Phys. Rev. C 7, 373 (1973).
- ¹⁷S. L. Whetstone, Jr., Phys. Rev. <u>131</u>, 1232 (1963).
- ¹⁸H. W. Schmitt, J. H. Neiler, and F. J. Walter, Phys.
- Rev. <u>141</u>, 1146 (1966).
- ¹⁹J. Terrell, Phys. Rev. <u>127</u>, 880 (1962).
- ²⁰C. Signarbieux, J. Poitou, M. Ribrag, and J. Matuszek, Phys. Lett. <u>39B</u>, 503 (1972).
- ²¹B. C. Diven, H. C. Martin, R. F. Taschek, and J. Terrell, Phys. Rev. <u>101</u>, 1012 (1956).
- ²²D. A. Hicks, J. Ise, Jr., and R. V. Pyle, Phys. Rev. <u>101</u>, 1016 (1956).
- ²³C. Signarbieux, M. Ribrag, J. Poitou, and J. Matuszek, Nucl. Instrum. Methods <u>95</u>, 585 (1971).
- ²⁴E. Nardi, L. G. Moretto, and S. G. Thompson, Phys. Lett. 43B, 259 (1973).
- ²⁵I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. <u>116</u>, 683 (1960).

- ²⁶R. Babinet, H. Nifenecker, J. Poitou, and C. Signarbieux, in Proceedings of the Third Symposium on Physics and Chemistry of Fission, Rochester, 1973 (to be ²⁷K. Skarsvag, Nucl. Phys. <u>A153</u>, 82 (1970).
 ²⁸W. John, F. W. Guy, and J. J. Wesolowski, Phys. Rev.

- C 2, 1451 (1970). ²⁹J. B. Wilhelmy, E. Cheifetz, R. C. Jared, S. G. Thompson, H. R. Bowman, and J. O. Rasmussen, Phys. Rev. C 5, 2041 (1972). ^{30}E . Cheifetz, private communication.