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Detailed Study of Alpha Emission in ^{252}Cf Fission

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Long-range α-accompanied fission of ^{252}Cf is investigated in a five-parameter correlation experiment and the behavior of each component of total energy, including the prompt neutrons from the individual fragments, as well as from the fragment pairs and the prompt-γ emission, is studied and compared with the binary fission. The problem of determining the nucleon contribution by binary fragments to the α-particle formation is discussed. The average energy of the α particle is observed to increase towards the symmetric fission region, which is understood in terms of a possible shift of the scission point towards the heavy fragment. The dependence of the average number of total neutrons on the α-particle energy and the dependence of the α-particle energy on the fragment total kinetic energy is not linear, which is interpreted to yield some qualitative information on the initial energy of the α particle. Several differential correlations are also discussed.

I. INTRODUCTION

The α-particle accompanied fission [long-range-α (LRA) fission] in ^{252}Cf has been extensively studied in the past few years[1-4] and has recently been reviewed by Halpern.[5] Since the characteristics of the binary and LRA mode of fission, which are determined by the initial conditions at the moment of scission, are so similar, it is recognized that the study of the emission of the α particle could possibly yield some concrete information on the dynamical conditions at scission. A considerable effort in this direction has been made by comparing the LRA-fission experimental results with the asymptotic solutions of the trajectory calculations.[6-15]

Two main qualitative features of the dynamical conditions at the LRA scission point seem to emerge from these studies. First, the fissioning nucleus is on the average a little more elongated in the LRA mode of fission, and second, the fission fragments are already moving with an appreciable part of their final kinetic energy at the moment of scission. However, a quantitative agreement on these points is far from satisfactory, and moreover, the trajectory calculations made by some authors[13-15] yield appreciably different initial dynamical conditions.

The problem of arriving at a consistent set of initial dynamical variables is extremely complicated because of the multitude of free parameters and the lack of understanding of the mechanism of the α-particle emission which could possibly restrict the number of parameters or at least their range of variations. In the absence of the physical picture one has to rely more and more on experimental information. For example, the range of the variability of the initial dynamical variables and their interrelations reflect in the variances and the correlations of the experimental distributions. The experimental results on these, particularly the differential results, are very scanty. The present experiment is an attempt to obtain the detailed information, with sufficient statistical accuracy, on the energy and probability distributions of as many parameters as possible, and to study the variances and the various possible correlations. Moreover, it is believed that a reasonable picture of the dynamical conditions at the scission point could possibly emerge only after taking into account the deformation shapes of the fragments in the trajectory calculations. This experiment provides a large amount of information connected with the excitation energy of the fragments in order to augment the feasibility of such an attempt in future.

The experimental details are discussed in Sec. II, the data analysis in Sec. III, and the results and discussions are presented in Sec. IV. The global results, which have been obtained to a certain extent in previous experiments,[3, 4] along with the results on some of the variances, are discussed briefly, and more stress is given to the differential results and the new information obtained in the present experiment. Finally, a short summary of conclusions from our discussions is given in Sec. V.

II. EXPERIMENTAL DETAILS

The target consisted of a 1-cm-diam-thin deposit of ^{252}Cf, obtained by the self-transfer method, onto a thin VYNS backing. The source strength was 10^4 fissions/min. The target was mounted at 45° between two back-to-back surface-barrier detectors by means of which the energies of both the fragments from the fission events were measured. The fission-fragment detectors were 2 cm in diameter and their distance from the center of the target was 2 cm. The two semiconductor detectors for measuring α particles (2.5 cm diam) were mounted symmetrically at a distance of 1.1 cm from the center of the target and were at 90° to the axis of the fragment detectors.

The neutron detector was a 100-cm-diam gadolinium-loaded liquid scintillator tank. Fission neutrons entering the detector are thermalized by proton recoils and captured in about 10 μsec by the gadolinium nuclei. The 8.2-MeV γ-radiation cascades released from each capture event produce scintillations which are detected by the photomultipliers and give the neutron signal. It is possible to distinguish the prompt γ rays and prompt neutrons in this detector, by studying the pulses as a function of time with respect to the fission event. The γ rays give a prompt pulse, whereas the neutron pulses are produced after several microseconds. However, the prompt-γ pulse naturally contains the small effect of the proton recoils produced in the early part of neutron thermalization which has to be corrected to get the correct γ pulse.[16] The prompt-γ pulses were analyzed by means of an analog-to-digital converter, and the multiplicity of neutrons was recorded in a period of 35 μsec, starting 1 μsec after the arrival of the fission event signal. After a duration of 100 μsec from the fission event, a second 35-μsec-wide gate recorded the background counts. The details of this detector system have been published before.[17] This system has a high efficiency of ~80% for detecting fission neutrons. The background for the present experiment was about 0.04 counts per fission event. The detector tank consists of two hemispheres which are placed side by side for the 4π detection and can be used separately for 2π counting experiment. Each hemisphere has a 10-cm-diam diametrical hole to fix the fission-event detection system inside the neutron detector.

The present investigation consists of two separate sets of measurements which are as follows.

The fission chamber described in the first paragraph of this section was placed in the center of the diametrical hole in the neutron detector used in the 4π counting geometry. This was used to study the total number of neutrons ($\bar{\nu}_t$) emitted per fission event and the average prompt-γ energy for the binary and LRA fission of ^{252}Cf simultaneously. The α detectors were surrounded by $\approx$12-mg/cm^2 aluminium foil to shield these detectors from the fission fragments and the natural α particles from ^{252}Cf. For the LRA fission recording a coincidence was demanded between one of the α detectors and the two fission-fragment detectors and this coincidence signal opened the time gates for neutron detector systems. The recording of binary-fission data was exactly similar except for the absence of the signal from the α detector. A total of 70 000 LRA-fission coincidences were recorded in this experiment on an incremental magnetic tape recorder.

The second experiment was to measure the number of neutrons emitted by the individual fragments in the fission event of the binary and LRA type in ^{252}Cf. This was done in the usual fashion of utilizing the neutron detector in 2π counting geometry.

The angular correlation of the prompt neutrons with the fragment direction is used to associate the neutrons with the particular fragments that emit them. In order to have the better definition of the fragment direction the distance between one of the fragment detectors and the fissile source was increased to 4 cm. This naturally reduces the counting rate, and a total of ~9000 coincidence events were accumulated for LRA-fission events in the period of about 80 days.

The five parameters recorded for LRA fission were two fission-fragment pulse heights, α-particle energy, prompt-γ pulse height, and the number of neutrons emitted per triple coincidence event. The simultaneous recording of binary and LRA-fission data was achieved by recording 99 binary fission events for every LRA-fission event recorded, through a logic system.

III. DATA ANALYSIS

The energy calibration of fission-fragment detectors was done using the binary-fission data and the calibration method of Schmitt, Kiker, and Williams.[18] The α-particle detectors were calibrated using various natural α peaks and the α energy was corrected for absorption in the aluminium foil. The neutron data were corrected for background and electronic dead-time effects. The efficiency correction was determined from binary-fission data. In the case of 4π experiment, it is experimentally measured in an unambiguous fashion.[17] On the other hand, in the case of 2π experiment, it depends strongly on the neutron-fragment correlation.[19] For each pair of fragment energies (E_1, E_2), the efficiency was determined for the neutrons emitted by each fragment using a Monte Carlo code,[20] which assumed that all neutrons are evaporated isotropically (in the center-of-mass system) from fully accelerated fragments with a Maxwellian spectrum and a constant nuclear temperature of 1 MeV. A treatment of binary-fission neutron data with Bowman's nuclear temperatures[21] showed no significant differences in the $\bar{\nu}_1(M, E_K)$ values.

The masses and kinetic energies of fission fragments were not corrected for prompt-neutron emission, except in the case of the evaluation of the pre-neutron mass distribution. It is because the detailed information on prompt-neutron emission, $\bar{\nu}(M, E_K, E_\alpha)$, with sufficient statistical accuracy, which is necessary for realistic corrections, is still lacking in the case of LRA fission. It is believed that the comparison of binary and LRA-fission results is not hampered due to this because of the close similarity of the two processes. The fragment masses indicated are thus, in general, pseudomasses[18] unless otherwise mentioned. It was not possible to take into account in detail the recoil effects due to α emission in LRA fission, since we have not measured the angular direction of the α particle with respect to the fission fragments. However, we made the correction for the mass distribution curve (see Appendix).

The pre-neutron-emission mass distribution in LRA fission was obtained with the procedure given by Schmitt, Neiler, and Walter,[22] and by utilizing our results on prompt-neutron emission from individual fragments evaluated as described above.

IV. RESULTS AND DISCUSSIONS

A. Mean Values

A summary of results is presented in Table I. Our value of the mean kinetic energy of fission fragments in LRA fission is lower than the values obtained in other measurements.[3, 4] In all the previous measurements the energy calibration of the fission-fragment detectors was done by assuming a simple linear dependence of the energy on the pulse height, whereas we have used the more accurate method of Schmitt, Kiker, and Williams.[18] To understand this difference, we also tried a simple linear-dependence calibration procedure, and in this case we obtained the value of $\bar{E}_K^{\mathrm{LRAF}}$ (post-neutron emission) to be 171.2 MeV in agreement with above-mentioned authors. The difference between the mean kinetic energies of the fragments of binary and LRA fission is found to be 14.16 ± 0.04 MeV, after the correction for neutron emission. This value is significantly higher than the values obtained so far, which is understandable from the reason given above.

The average number of neutrons $(\bar{\nu}_t)$ emitted per fission event in LRA fission has been measured in an almost 4π geometry. Our value of the difference $(\bar{\nu}_t^B - \bar{\nu}_t^{\mathrm{LRAF}})$ is in good agreement with all previous measurements except that of Adamov *et al.*[23] who give the value as 0.95. A new quantity measured in the present work is the average energy associated with the prompt-γ emission which is listed in the table. The difference in the energy associated with γ emission between the binary and LRA fission is measured to be 1.01 ± 0.02 MeV, which had been assumed to be zero so far.[3] Our $\bar{E}_\gamma$ values are corrected for small effects of proton recoils caused by fission neutrons. The mean α energy given in the table is after the correction for absorption in the aluminium foil and thus has the usual uncertainty connected with the foil thickness, the correction procedure, and the artificial bias introduced by the foil.

Since we have access to all components of the total energy in binary and LRA fission process we attempt to compare the total energy available in the two fission processes. The difference in energy available in the two processes can be written as

$$\Delta Q=\Delta\bar{E}_K+\Delta\bar{E}_\gamma+\Delta\bar{E}_\nu-\bar{E}_\alpha\,, \qquad (1)$$

where $\Delta\bar{E}_K=\bar{E}_K^{\rm BF}-\bar{E}_K^{\rm LRAF}$ etc.; $\Delta\bar{E}_\nu$ is calculated using the same binding energy of 5.2 MeV for binary and LRA fission, and the kinetic energies from the formula[24] $E_{\rm cm}=0.65(\bar{\nu}_t+1)^{1/2}$. By using the values listed in the table and correcting $\Delta\bar{E}_K$ for neutron emission we get

$$\Delta Q=4.00\pm 0.06\ \text{MeV}\,.$$

B. Comparisons Between Binary and LRA Fission

The mean values of the quantities $\bar{E}_K$, $\bar{E}_\gamma$, and $\bar{\nu}_t$ as a function of light fragment mass in LRA and binary fission are shown in Fig. 1. The remarkable similarity (except for a small over-all shift) between these characteristics for the two fission processes has already been demonstrated by previous experiments. The present experiment provides much higher statistical accuracy and gives additional information on the similar behavior of the variances as a function of fragment mass, shown in Fig. 2.

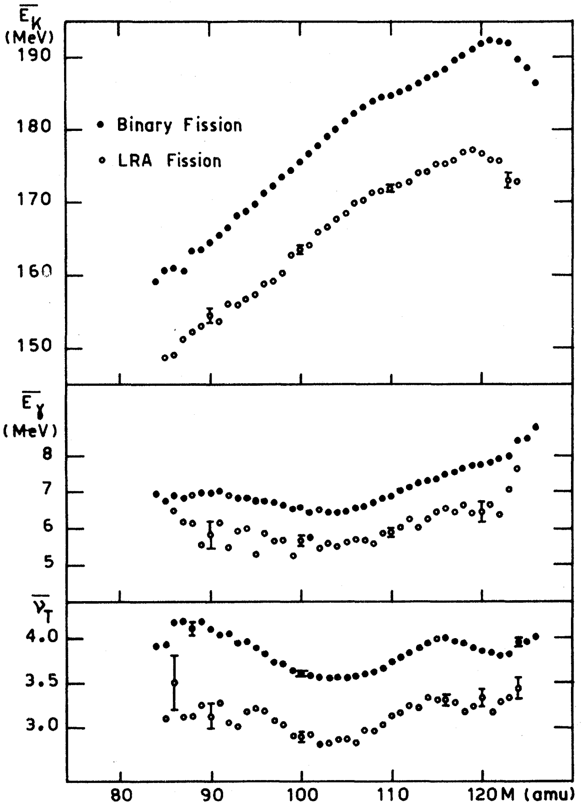

FIG. 1. The mean total kinetic energy of both fragments ($\bar{E}_K$), the average prompt-γ energy ($\bar{E}_\gamma$) and the average total neutron emission from fragment pairs ($\bar{\nu}_t$) as a function of light fragment mass, for binary and LRA fission.

The results on prompt-neutron emission from individual fragments ($\bar{\nu}_1$) in binary and LRA fission are shown in Fig. 3. The $\bar{\nu}_1^{\rm LRAF}$ results from our 2π geometry experiment suffer from insufficient statistical accuracy and the difficulties discussed in Sec. III. On the other hand, our results on the total neutron emission from both fragments, $\bar{\nu}_t$, are free from these problems. Thus the good agreement between the $\bar{\nu}_t$ values in the two measurements (2π and 4π geometry), shown in the upper part of the figure, gives us confidence in the reliability of our $\bar{\nu}_1^{\rm LRAF}$ results. The ratio of neutrons emitted from light and heavy fragment ($\bar{\nu}_L/\bar{\nu}_H$) is observed to be 1.13 in the case of LRA fission and 1.16 for binary fission. Our results on $\bar{\nu}_1$, both for binary and LRA fission, are markedly different than those Nardi and Fraenkel,[3] particularly for neutron emission from heavy fragments. The flat response of our large liquid scintillator to neutrons of varying energies gives neutron data with considerably improved accuracy, whereas the efficiency corrections in the experiments utilizing small scintillators introduce several uncertainties as also emphasized by Nardi and Fraenkel.[3] This may explain the observed differences, but it should be pointed out that the

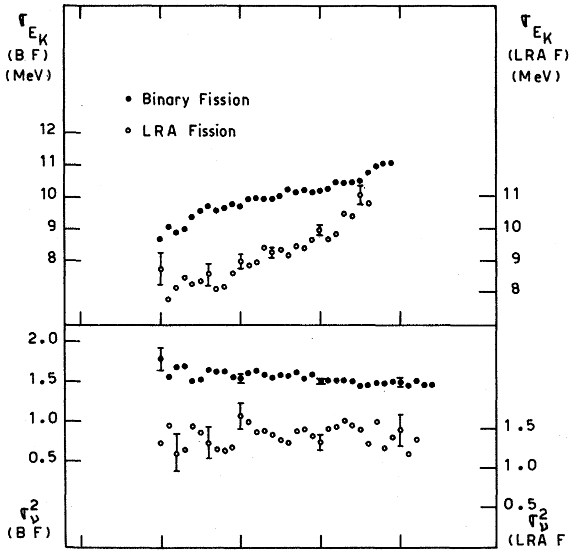

FIG. 2. The variances of the total kinetic energy and the total neutron emission distributions as a function of the light fragment mass for binary and LRA fission.

comparison of the two results is difficult because Nardi and Fraenkel indicate only four points in the region of reliable statistical accuracy.

The pre-neutron-emission mass-distribution curves shown in Fig. 4 differ in details with those obtained by Nardi and Fraenkel,[3] which arise due to the calibration procedure as discussed before, and the differences in the results on the neutron emission from individual fragment. The binary- and LRA-fission curves in our figure are normalized to the total yield and not at the peak values. The average shifts in the light and heavy fragment peaks are indicated in Table I.

In Fig. 5 we show the results on $\bar{\nu}_t$ and $\bar{E}_\gamma$ (averaged over the mass distribution) as a function of kinetic energy. The LRA fission results are plotted as a function of both E_K and total kinetic energy, $E_K + E_\alpha$. In the light of previous results the similarity between the binary and LRA fission, except again possibly a small shift, is not surprising. The differential slopes, $d\bar{\nu}_t/dE_K$ for individual masses, are plotted in Fig. 6. The values of the slopes $d\bar{\nu}_t/dE_K$ and $d\bar{\nu}_t/d(E_K + E_\alpha)$ are more or less identical mass by mass.

This comparison of all the characteristics of experimentally measured parameters leads to a definite conclusion that the α-emission process does not change in a significant way any of the characteristics of the scission process. If we turn the argument around we can say that the α-emission process as a whole is very nearly independent of the scission configuration, except for possible difference in elongation.

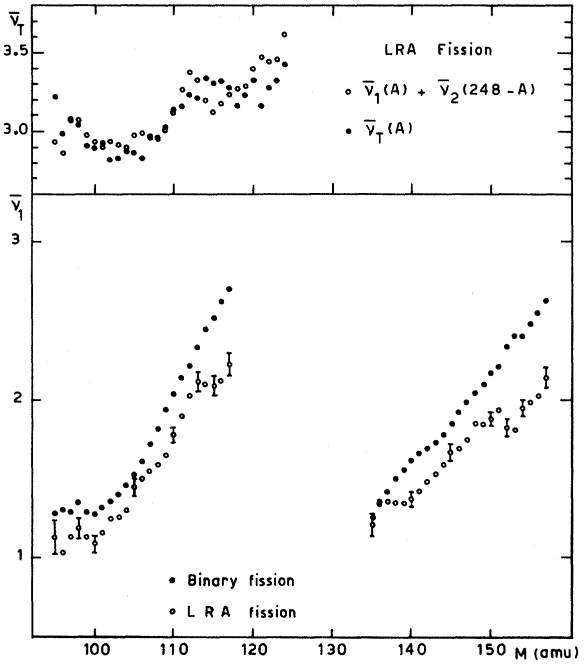

FIG. 3. The average neutron emission from individual fragments ($\bar{\nu}_1$) as a function of fragment mass in binary and LRA fission. On top is the comparison of the results on the average neutron emission from fragment pairs in LRA fission, obtained from the $\bar{\nu}_t$ and the $\bar{\nu}_1$ experiments.

C. Correspondence Between Binary and LRA Fission

There are two questions to be answered in connection with the α-emission process. First, how does the nucleon contribution (X) to form an α-particle depend on the binary fragment mass, and second, how does the probability of emission of the α-particle vary as a function of mass ratio. The shape of the various distributions, such as the mass distribution, in LRA fission depend on both the probability of α emission and the origin of nucleons, and thus it is impossible to extract results on both the quantities just from the comparison of mass distributions. If one makes a reasonable assumption about P_α it is possible to evaluate X, or vice versa. It has been clearly demonstrated recently by Halpern[5] that the two are completely equivalent ways of characterizing the transformation from binary- to LRA-fission mass distribution. In order to have a complete correspondence between all binary- and LRA-fission distributions one would like to have consis-

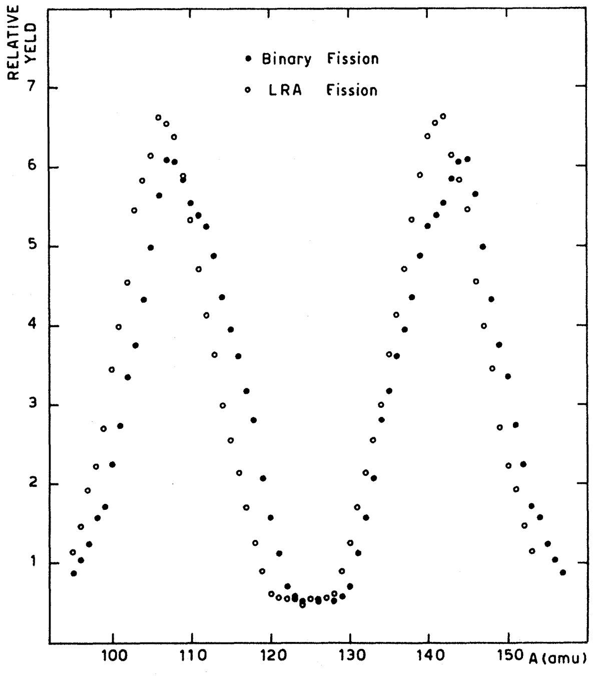

FIG. 4. Pre-neutron-emission mass distribution curves for the two fission processes. The curves are normalized to the same total yield.

tent information on both the quantities P_α and X.

Since there are many ways in which the α particle can be formed, in general, binary yields of several masses contribute to the yield of a particular mass in LRA fission, and one gets a general relation

$$Y^{\mathrm{LRAF}}(A_L)=\sum_{n=0}^{4} Y^{\mathrm{BF}}(A_L+n)P_\alpha^n(A_L+n), \qquad (2)$$

where $P_\alpha^n(A_L+n)$ is the probability that the binary fragment of mass (A_L+n) contributes n nucleons to α formation. The average number of nucleons contributed by binary fragments is given by

$$\overline{X}(A_L)=\sum_{n=0}^{4} nP_\alpha^n(A_L), \qquad (3)$$

and the probability of LRA fission for a given mass ratio by

$$P_\alpha(A_L/A_H)=\sum_{n=0}^{4} P_\alpha^n(A_L). \qquad (4)$$

It should perhaps be emphasized that P_α is the probability that a binary fission with fragments of mass A_L and A_H actually evolves out into LRA fission and that P_α is related to the mass pair.

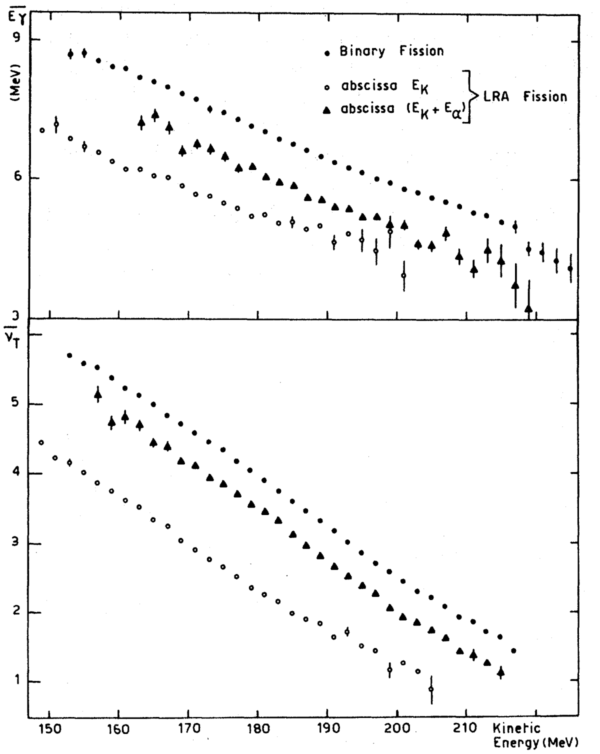

FIG. 5. The dependence of the average prompt-γ energy ($\overline{E}_\gamma$) and the average total number of neutrons ($\overline{\nu}_t$) on the total kinetic energy of both fragments in the two fission processes. For LRA fisssion, the dependence on the total kinetic energy of the three particles (E_K+E_α) is also shown (triangles).

Schmitt and Feather[25, 26] have obtained the probability of LRA fission with the assumption that all the nucleons are contributed by one fragment. They express it as

$$Y^{\mathrm{LRAF}}(A-4)=Y^{\mathrm{BF}}(A)P_\alpha(A), \qquad (5)$$

which gives rise to two curves, one corresponding to the α emission from the light fragment only, and the other corresponding to the α emission from the heavy fragment only. If both light and heavy fragments could emit α particles, then Eq. (2) would lead to the expression

$$Y^{\mathrm{LRAF}}(A_L)=Y^{\mathrm{BF}}(A_L)P_\alpha^0(A_L)+Y^{\mathrm{BF}}(A_L+4)P_\alpha^4(A_L+4). \qquad (6)$$

Nardi and Fraenkel[3] have evaluated $P_\alpha(A_L/A_H)$ by making a plausible assumption regarding the nucleon contribution $X(A_L)$. However, to obtain $P_\alpha(A_L/A_H)$ they use the expression

$$Y^{\mathrm{LRAF}}(A_L-X)=Y^{\mathrm{BF}}(A_L)P_\alpha(A_L/A_H). \qquad (7)$$

This expression seems to be valid, with the restrictions that X is unique for each A (physically it implies that X is integral), and that only one binary fragment can contribute to the yield of a particular fragment in LRA fission.

It is possible to evaluate $P_\alpha(A_L/A_H)$ with some assumption about $\overline{X}(A_L)$, or vice versa, according to Eqs. (2), (3), and (4) without any restrictions if one uses the cumulative yields derived from the

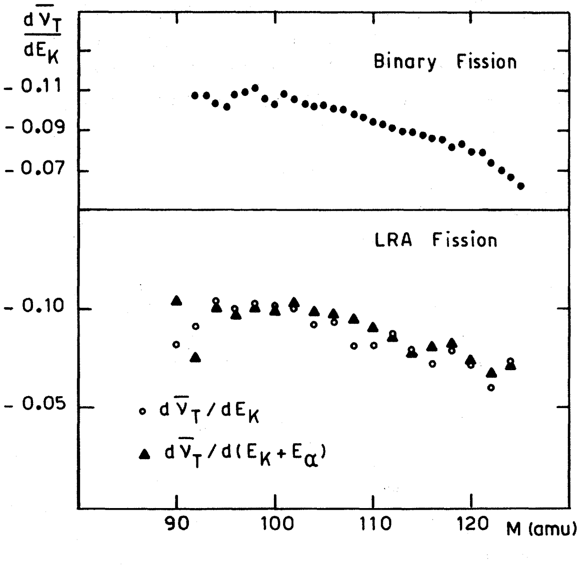

FIG. 6. The slope $d\overline{\nu}_t/dE_K$ for fragment pairs in binary fission as a function of the light fragment mass (top curve). The lower curves show the slopes $d\overline{\nu}_t/dE_K$ (circles) and $d\overline{\nu}_t/d(E_K+E_\alpha)$ (triangles) in LRA fission.

measured differential mass yields of binary and LRA fission, as shown by Terrell[24] for the case of neutron emission.

D. Evaluation of $\bar{X}(A)$

In terms of the only available picture[6] of the α-emission process, the α particle is probably emitted by a sudden change in potential. One could envisage the situation that an α particle exists in the neck prior to LRA scission, and the probability of α emission depends on the change of having an α particle at the time of scission. If one assumes that the snap position does not depend on whether an α particle exists at the time of scission or not, then one could say that P_α should be independent of mass ratio. The other quantity $[\bar{X}(A)]$ (which provides the information how, if binary scission took place instead of LRA scission, these nucleons would have been distributed between the two binary fragments) might depend on the details of the energetics involved as calculated by Feather.[26] If this way of looking at α-emission process is reasonable, then one can learn about the variation of $\bar{X}$ with binary fragment mass. However, the picture is clearly intuitive, and its reasonability can only be checked by its consistency.

The average nucleon contribution by binary fragments, $\bar{X}(A)$, evaluated by using the cummulative mass yields (corrected for prompt-neutron emission) under the assumption that P_α is independent of mass ratio is shown in Fig. 7. The statistical accuracy of our results on $\bar{\nu}_1^{\mathrm{LRA}}(A)$ does not permit to evaluate $\bar{X}(A)$ near the symmetric fission region. The first conclusion from this is that the contribution to the α-particle formation is at the expense of both binary fragments (the situation will of course change if P_α was not really independent of mass ratio). The average contribution from the light fragments is slightly greater ($\simeq 2, 2$). This is reflected as a shift of 2.3 amu in the mean light fragment mass in the two mass distributions. Furthermore, it is observed that the variation of $\bar{X}$ is reasonably linear with fragment mass, in each peak, with an over-all saw-tooth behavior as suggested by Nardi and Fraenkel.[3]

A rough linear dependence of $\Delta\bar{\nu} = \bar{\nu}^B(A) - \bar{\nu}^T(A - \bar{X})$ on $\bar{X}$ is observed considering light and heavy fragments separately, which can be expressed as

$$\frac{\bar{X}_L - \langle \bar{X}_L \rangle}{\Delta\bar{\nu}_L - \langle \Delta\bar{\nu}_L \rangle} = \frac{\bar{X}_H - \langle \bar{X}_H \rangle}{\Delta\bar{\nu}_H - \langle \Delta\bar{\nu}_H \rangle}, \tag{8}$$

where $\langle \bar{X}_L \rangle$ is the average taken over all the light fragments,

$$\Delta\bar{\nu}_L = \bar{\nu}_L^{\mathrm{BF}}(A) - \bar{\nu}_L^{\mathrm{LRA}}(A - \bar{X}),$$

$$\Delta\bar{\nu}_H = \bar{\nu}_H^{\mathrm{BF}}(A) - \bar{\nu}_H^{\mathrm{LRA}}(A - 4 + \bar{X}), \text{ etc.}, \ldots .$$

The consequence of this is that

$$\Delta\bar{\nu}_t = \bar{\nu}_t^{\mathrm{BF}} - \bar{\nu}_t^{\mathrm{LRA}}(A - \bar{X}) = \text{constant}.$$

It should be noted that $\Delta\bar{\nu}_t = \text{constant}$ had been taken as one of the assumptions by Nardi and Fraenkel[3] to determine $\Delta\bar{\nu}(A)$ and $P_\alpha(A_L/A_H)$. However, they had made an additional assumption that $X_L/X_H = \nu_L^B/\nu_L^{\mathrm{LRA}}$ which is not in agreement with our results. The logical consequence of the deduction that $\Delta\bar{\nu}_t$ is constant is that the decrease of the total excitation energy due to the α emission is constant over all mass ratios. This seems to be an indication of the consistency between our starting assumption and its consequences.

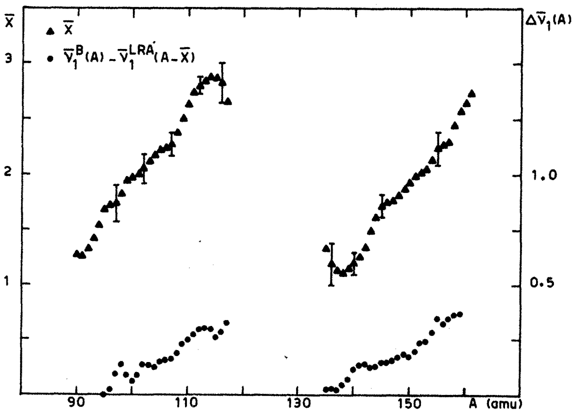

FIG. 7. The average nucleon contribution to the α-particle formation, with roughly estimated errors, as a function of binary fragment mass and the function $\Delta\bar{\nu}_t(A) = \bar{\nu}_t^{\mathrm{BF}}(A) - \bar{\nu}_t^{\mathrm{LRA}}(A - \bar{X})$.

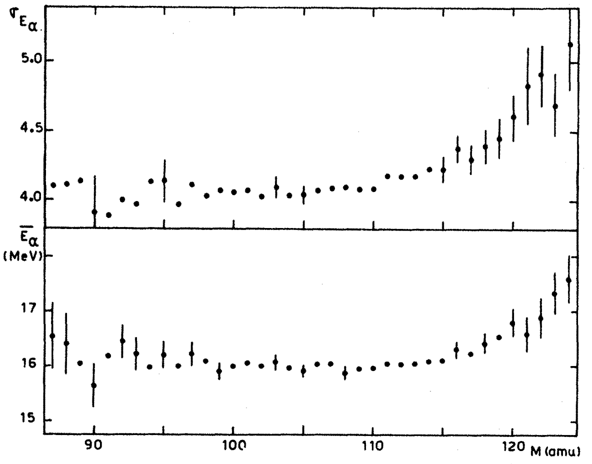

FIG. 8. The dependence of the average energy of α particle ($\bar{E}_\alpha$) and the variance (σ_{E_α}) on the fragment mass ratio, plotted as a function of the light fragment mass, in LRA fission.

E. Average Energy of the α Particle and Its Variance

Our measurement of the average energy of the α particle as a function of the mass of the fission fragment, $\overline{E}_\alpha(M)$, shows an increase of about 1.5 MeV as the symmetric fission region is approached, which is shown in Fig. 8. We might emphasize that the mass-resolution effects and the spurious events caused by the energy loss of fission fragments due to scattering in detector mounts can only reduce the observed effect. Similar behavior is observed for the variance of E_α, also shown in Fig. 8.

These effects observed near the symmetric fission region are interesting for trajectory calculations. The results of the various trajectory calculations on this point are not conclusive. Geilikman and Khlebnikov[7] show that the most probable energy E_α varies very little (by ~500 keV) as the mass ratio increases from 1 to 2. The recent trajectory calculations of Fong[13] show an effect similar to our experimental results, but he attributes it to not taking into account the effects of uncertainty in the initial position of the α particle. It is pointed out by Fong that if an uncertainty in position of about 3 fm is introduced, the distribution tends to smooth out, removing the effect of increase in $\overline{E}_\alpha$ towards the symmetric fission region.

One can understand qualitatively the observed behavior of $\overline{E}_\alpha$ as a function of the fragment mass, if we assume the configuration near the scission point as two asymmetric fragments joined by a neck.[27, 28] As the symmetric fission region is approached, the neck snaps closer to the heavy fragment and the shift in the α-particle emission point gives it relatively more kinetic energy than in other regions. Thus the increase in $\overline{E}_\alpha$ can be attributed to the fact that the scission point shifts towards the heavy fragment as the symmetric fission region is approached. This is also borne out from the angular-distribution results of Fraenkel.[2] The trajectory calculations of Boneh, Fraenkel, and Nebenzahl[8] indicate that the most probable initial distance of the α particle from the heavy frag-

TABLE I. Summary of results on mean values of various quantities in binary and LRA fission. The widths of the distributions given in our results are the root-mean-square widths. The errors quoted are statistical only. The results of other authors are listed for comparison.

	Binary fission				Long-range α fission			
	Our results	Schmitt *et al.* (Ref. 6)	Whetstone (Ref. 7)	Nardi and Fraenkel (Ref. 4)	Our results	Nardi and Fraenkel (Ref. 4)	Piekarz *et al.* (Ref. 3)	Fraenkel (Ref. 5)
$\overline{E}_K$ (MeV)	186.26 ± 0.01 (183.34 ± 0.01) [a]	186.5 ± 1.2 (183.2 ± 0.7) [a]	185.7 ± 1.8	187.3 ± 0.1	169.79 ± 0.04 [a]	174.5 ± 0.1	171.0 ± 1.1 [a]	169.1 ± 0.1 [a]
σ_{E_K} (MeV)	11.4	12.0	11.3	10.8 ± 0.1	11.0 [a]	9.8 ± 0.06		12.6 ± 0.1 [a]
$\overline{M}_L$ (amu)	108.5 (108.9) [b]	108.55	108.39	108.7 ± 0.1	106.2 (107.7) [b]	105.9		
$\overline{M}_H$ (amu)	143.4 (143.1) [b]	143.45	143.61	143.3 ± 0.1	141.8 (140.3) [b]	142.1		
σ_{M_L} (amu)	6.8 (7.5) [b]	6.72	6.77	6.91	6 5	6.08		
$\overline{\nu}_T$	3.766 ± 0.002				3.072 ± 0.006	3.11 ± 0.05	3.10 ± 0.08	
σ_{ν_T}	1.25 ± 0.07				1.20 ± 0.10			
$\overline{E}_\gamma$	7.00				5.99 ± 0.02			
$\overline{E}_\alpha$ (MeV)					16.08 ± 0.02			
σ_{E_α} (MeV)					4.2 ± 0.3			
$\overline{E}_K^{\mathrm{BF}} - \overline{E}_K^{\mathrm{LRAF}}$ (MeV)					13.55 ± 0.04 [a]	12.8 ± 0.1		12.1 ± 0.1 [a]
$\overline{\nu}_T^{\mathrm{BF}} - \overline{\nu}_T^{\mathrm{LRAF}}$					0.694 ± 0.006	0.60 ± 0.05	0.69 ± 0.06	

[a] Post-neutron-emission quantities.

[b] Pseudomass.

ment shifts appreciably towards the heavy fragment as the mass ratio R tends towards 1, which also corresponds to an effective increase in $\overline{E}_\alpha$ considering their calculations on the dependence of $\overline{E}_\alpha$ on the α particles, initial position. Thus the qualitative arguments used to understand our results on $\overline{E}_\alpha$ variation seem to be in agreement with the trajectory calculations of Boneh, Fraenkel, and Nebenzahl.[8]

The behavior of σ_{E_α} can be understood from the argument that the slope of the potential energy curve becomes steeper as the heavy fragment is approached, and thus a small variation in the initial position of the α particle can produce the observed larger variation in $\overline{E}_\alpha$ near symmetric fission.

So far our discussion was based on the observed behavior of $\overline{E}_\alpha$ (averaged over E_K) as a function of mass split. Continuing in the realm of the Vladimirski-Whetstone picture, this corresponds to the situation of fixed snap position (fixed mass ratio) and variable snap time. It should be interesting now to examine the behavior of $\overline{E}_\alpha$ for a fixed snap time (state of elongation) as a function of mass ratio, R. Some representative curves of $\overline{E}_\alpha(M)$ for several values of $\overline{E}_K$ are exhibited in Fig. 9, with a chosen grid of 2 amu and 4 MeV. The increase in $\overline{E}_\alpha$ as the mass ratio decreases is evident in all the curves. Moreover, the increase in $\overline{E}_\alpha$ near the symmetric region becomes more pronounced as $\overline{E}_K$ increases (i.e., for smaller elongation), which again is in line with the qualitative explanation used so far. As far as the variance of E_α for fixed $\overline{E}_K$ values is concerned, the statistical accuracy is the limiting factor. It is not possible to draw any definite conclusion, but the tendency seems to be that σ_{E_α} does not change with mass for fixed values of E_K.

In our discussion on the variation of $\overline{E}_\alpha$ with R, we have concentrated on the increase in $\overline{E}_\alpha$ as R tends to 1. Now from the qualitative picture of the shift in the scission point as R changes, one would expect that as R becomes very large the scission point should shift closer to the light fragment, thus producing some characteristic influence on on $\overline{E}_\alpha$ and σ_{E_α}. Our results do not show any statistically significant effect for $R \gg 1$. Boneh, Fraenkel, and Nebenzahl[8] found in their trajectory calculations that $\overline{E}_\alpha$ increases as the initial position of the α particle is closer to the lighter fragment, but the increase is considerably less than in the case when the α particle is initially closer to the heavy fragment.

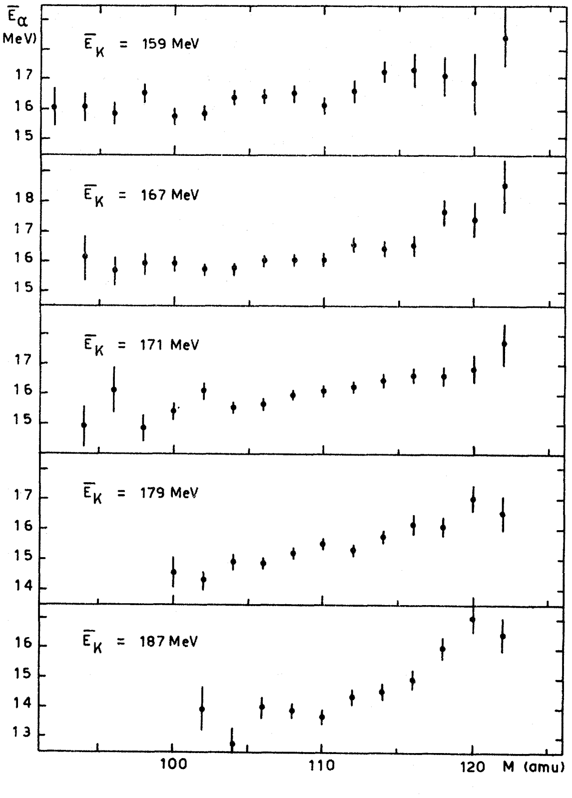

FIG. 9. The variation of the α-particle energy (averaged over 2 amu and 4-MeV grid) with the fragment mass, for the fixed values of the total kinetic energy of both fragments, in LRA fission.

F. Correlation of $\overline{\nu}_t$ and $\overline{E}_\gamma$ with E_α

The average number of neutrons emitted by both fragments decreases as E_α increases, which is shown in Fig. 10. However, the decrease in $\overline{\nu}_t$ does not appear to be linear with E_α in contradiction with existing results.[3,4,23] On examining this

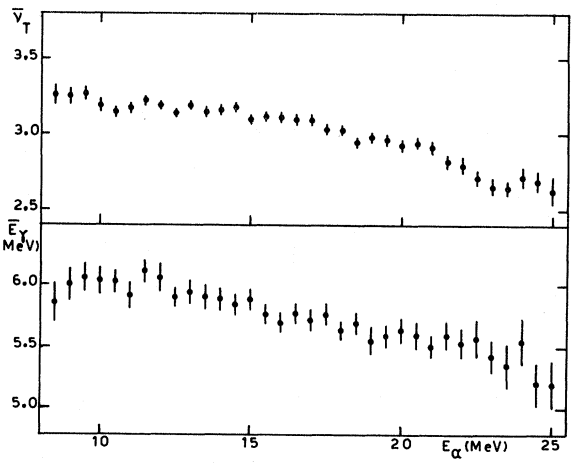

FIG. 10. The dependence of the average total neutron emission and the average γ energy on the α-particle energy in LRA fission.

dependence of $\bar{\nu}_t$ on E_α for fixed fragment mass (Fig. 11) it is apparent, in spite of the poor statistical accuracy, that it is not linear even when the fragment mass is fixed. The variation is possibly such that the slope $d\bar{\nu}_t/dE_\alpha$ increases with E_α.

The variation of $\bar{E}_\gamma$ with E_α, which is also shown in Fig. 10, is rather similar to the variation of $\bar{\nu}_t$. Since the average photon energy is expected to be practically constant, our results on $\bar{E}_\gamma$ mostly reflect the change in average number of γ quanta yields. Ajitanand[29] found that the γ quanta yield remains fairly constant with E_α up to about 18 MeV and then drops off, which was interpreted as a rather sudden change for $E_\alpha > 17$ MeV. Our results on $\bar{E}_\gamma$, as well as $\bar{\nu}_t$, do not show any trend of sudden change. It is also interesting to note that Fraenkel[2] had found that up to an energy of $E_\alpha = 19$ MeV the angular distributions are more or less independent of E_α, but for $E_\alpha > 19$ MeV they broaden considerably. This again must be a gradual change and may possibly be related to the effect observed here in $\bar{\nu}_t$ vs E_α curves. From the observation that the slopes $d\bar{\nu}_t/dE_\alpha$ and $d\bar{E}_\gamma/dE_\alpha$ increase with α-particle energy, one can argue that as E_α increases more and more energy comes at the expense of the excitation energy, which may be connected with the increase in the initial kinetic energy of α particle (E_α^0). The broadening of angular distribution with increasing E_α is expected if E_α^0 increases. However, an alternate explanation of the broadening of the angular distributions could well be associated with late scission, which corresponds to larger separation between the two fragments and thus less focusing effects by the fragment Coulomb fields. Thus it is not possible to make a reasonable conclusion from these global results (averaged over the mass and kinetic energy distribution of fission fragments).

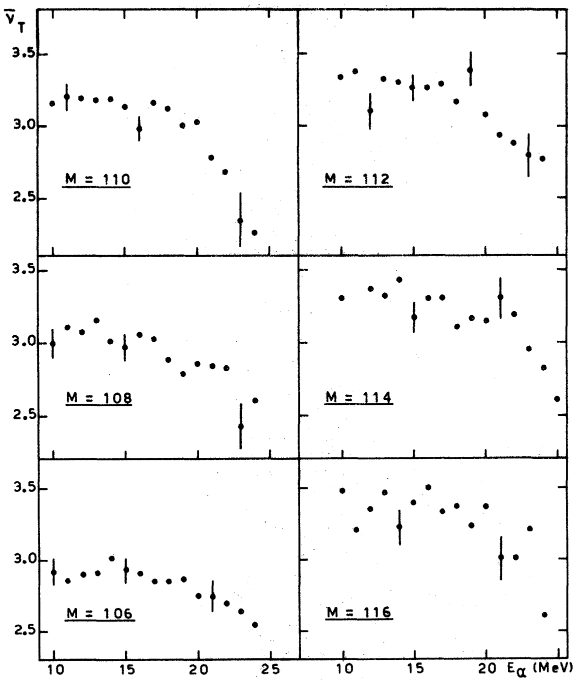

FIG. 11. The dependence of the total neutron emission by the fragment pairs (averaged over the kinetic energy distribution) on the α-particle energy, for the fixed fragment masses (the mass grid is 2 amu).

In Fig. 12 is shown the dependence of $\bar{\nu}_t$ on E_α for several groups of total kinetic energies of the fragments. It is noticed that in this case $\bar{\nu}_t$ seems to decrease a little more linearly with E_α. It can possibly be argued that by fixing E_K we have put a restriction on the variations of the separation between the two fragments and thus on the electrostatic energy. This would in turn imply that now the variations in E_α should be more or less entirely due to E_α^0 (for fixed mass) which probably comes at the expense of excitation energy and thus $\bar{\nu}_t$ decreases linearly. Thus it appears that the nonlinear variation of $\bar{\nu}_t$ with $\bar{E}_\alpha$ is brought about by the averaging over E_K. More useful information can perhaps be derived by examining variation of $\bar{\nu}_t$ with E_α and $\bar{\nu}_L/\bar{\nu}_H$ with E_α for fixed values of M and E_K, but the statistical limitation does not allow us to pursue it further.

The existence of a competition between prompt-neutron and prompt-γ emission in binary fission has been recently studied by Nifenecker *et al.*,[16] and it was found that for binary fission the average prompt-γ energy $\bar{E}_\gamma^B$ and the average number of total neutrons $\bar{\nu}_t^B$ show a linear relationship. In

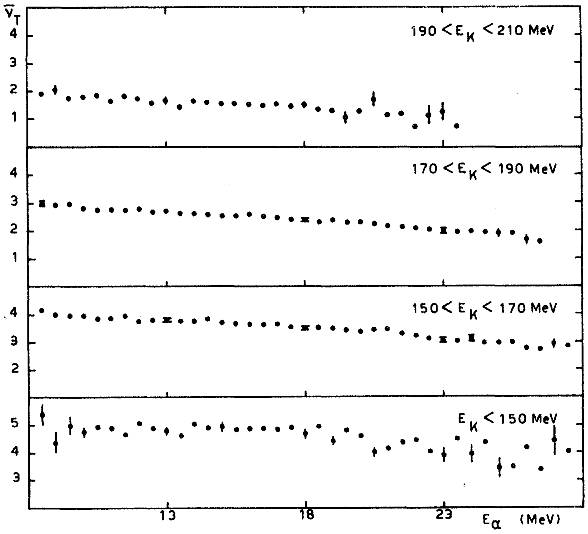

FIG. 12. The average total neutron emission for the fixed groups of total kinetic energy of both fragments in LRA fission versus α-particle energy.

Fig. 13 we show a plot of $\bar{E}_\gamma$ as a function of $\bar{\nu}_t$ for both binary and LRA fission. These curves are obtained from the results of $\bar{E}_\gamma$ and $\bar{\nu}_t$ as a function of E_K, by determining values of $\bar{E}_\gamma$ and $\bar{\nu}_t$ corresponding to the same values of E_K. The slopes of the two curves are almost equal, which indicates that the neutron-γ competition in LRA fission is identical to that in binary fission. The intercept of these curves at $\bar{\nu}=0$, which is related to the average binding energy of neutron, indicates that the average binding energy of neutrons in the case of LRA fission is slightly lower than in the binary fission. It is reasonable, since LRA fragments are supposed to be more neutron rich than binary fragments. A rough estimate shows that the difference in the average binding energy is about 0.4 MeV. It should be mentioned that the average difference in the energy available in the two fission processes obtained in Sec. A would increase by roughly 0.3 MeV if this difference in binding energy of neutron is taken into account.

G. Correlations Between E_K and E_α

The variation of $\bar{E}_K$ and σ_{E_K} with E_α is shown in Fig. 14. The slope $d\bar{E}_K/dE_\alpha = -0.44$ gives the correlation between the average fragment kinetic energy and the α-particle energy which is in excellent agreement with the value obtained by Fraenkel.[2] Our differential results demonstrate that the anticorrelation between $\bar{E}_K$ and E_α increases as the mass ratio decreases (Fig. 15). The negative correlation is about 0.2 in far asymmetric region and increases to about 0.6 near symmetric region. Although the errors in evaluating the slopes are in general large, the trend of increase is clear, if it is not produced due to some systematic errors. However, an independent result obtained on $d\bar{\nu}_t/dE_\alpha$ (upper part of the figure) indicates a decrease as mass ratio decreases, which is expected if $d\bar{E}_K/dE_\alpha$ increases. Since these two quantities measured independently point towards the same effect, it can be asserted that the effect of the change in the anticorrelation between $\bar{E}_K$ and E_α with mass ratio is not due to any systematic errors. Since these variations in $d\bar{E}_K/dE_\alpha$ and those of σ_{E_K} (Fig. 2) with fragment mass ratio show a similar trend it might appear that the correlation between $\bar{E}_K$ and E_α depends on the distribution of the initial distance between the fragments (D), particularly in connection with the remark made by Blocki and Krogulski[9] that the negative correlation between $\bar{E}_K$ and E_α can be reproduced in trajectory calculations by including some distribution of D. But the trajectory calculations of Boney, Fraenkel, and Nebenzahl[8] indicate that just the initial distribution of E_α^0 can account for the observed anticorrelation between $\bar{E}_K$ and E_α. However, it has been pointed out by Boneh, Fraenkel, and Nebenzahl,[8] and emphasized recently, by Halpern,[5] that the actual negative correlation observed in the experiment is perhaps a complicated mixture of the correlations of varying signs produced by the several initial parameters. The trajectory calculations have so far attempted to derive the correlation between the fragment kinetic

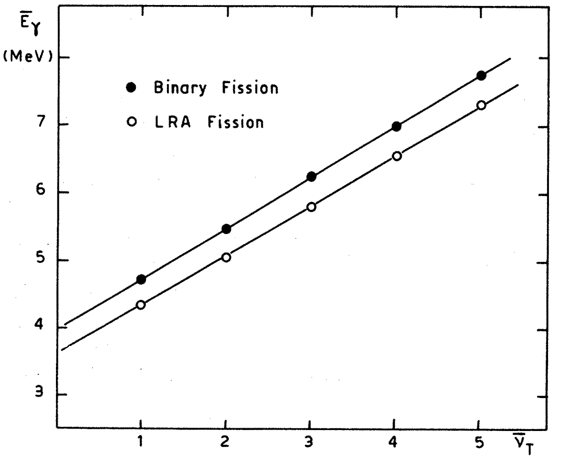

FIG. 13. The average prompt-γ energy as a function of the average total neutron emission from fragment pairs in binary and LRA fission.

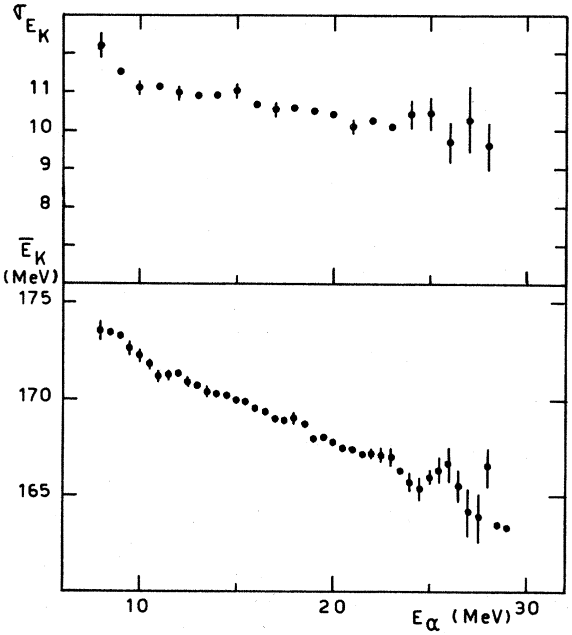

FIG. 14. The mean total kinetic energy of both fragments and its variance as a function of the α-particle energy in LRA fission.

energy averaged over the mass distribution (or for the most probable mass) and the α-particle energy. The differential results and the variances presented here when compared with the corresponding quantities obtained in trajectory calculations might be able to provide better information on the relative importance of the various possible initial parameters.

The correlation between the average α-particle energy ($\bar{E}_\alpha$) and E_K can also provide interesting information, which is shown in Fig. 16 along with the results on the variance σ_{E_α}. The dependence of $\bar{E}_\alpha$ on E_K is not linear, which is in contradiction with the observation of Asghar *et al.*[30] in the case of ^{235}U neutron-induced LRA fission. It should perhaps be pointed out that the fact that $\bar{E}_K$ vs E_α curve is a straight line, whereas $\bar{E}_\alpha$ vs E_K curve shows a complicated dependence, is not surprising. From the regression analysis one knows that these two slopes are related by the expression

$$\frac{d\bar{E}_K}{dE_\alpha}=\frac{d\bar{E}_\alpha}{dE_K}\frac{\sigma^2_{E_K}}{\sigma^2_{E_\alpha}},$$

and thus the variation of these slopes as a function of fragment mass can in principle show a completely different behavior. Our results show that even when we consider the variation of $\bar{E}_\alpha$ with E_K for fixed fragment masses the over-all dependence of $\bar{E}_\alpha$ on E_K remains unchanged, and in fact it is more or less insensitive to the fragment mass ratio (Fig. 17) except for the effect (Sec. E) that $\bar{E}_\alpha$ increases as the symmetric fission region is approached. The obvious qualitative conclusion from this is that independent of the snap position the correlation between $\bar{E}_\alpha$ and E_K is small for low values of E_K and it increases with E_K.

Although these correlations are presumably produced by a complicated mixture of several initial variables one could perhaps still try to look for a simple interpretation. The experimental result that the correlation between $\bar{E}_\alpha$ and E_K increases with E_K can possibly be an effect of decrease in the initial kinetic energy of fragments (E_K^0) as deformation decreases (corresponding to increase in E_K). If the initial energies E_α^0 and E_K^0 are not correlated, then higher initial energy leads to less

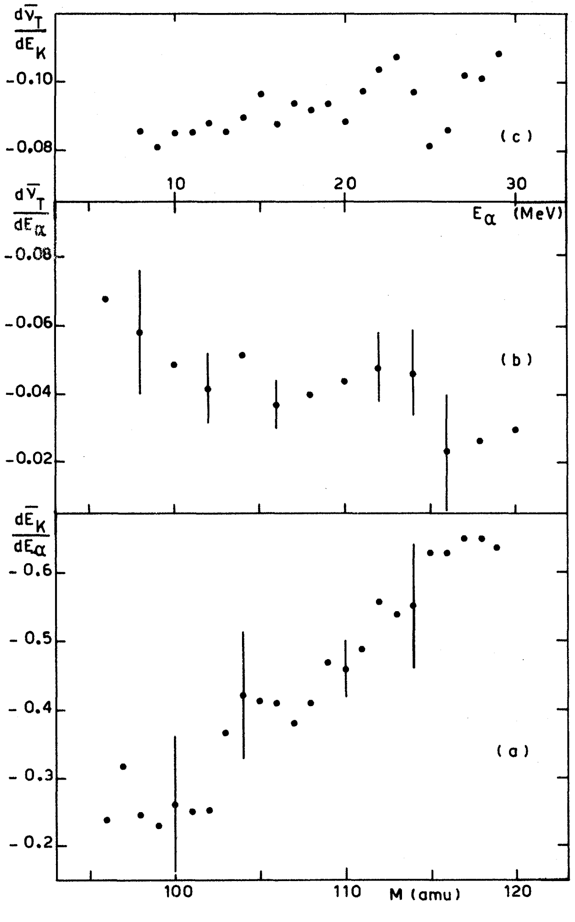

FIG. 15. (a) The derivative of the mean total kinetic energy of the fragments with respect to the α-particle energy, (b) the derivative $d\bar{\nu}_t/dE_\alpha$, as a function of the light fragment mass in LRA fission, and (c) the derivative $d\bar{\nu}_t/dE_K$ (averaged over $107 < M < 111$) as a function of E_α.

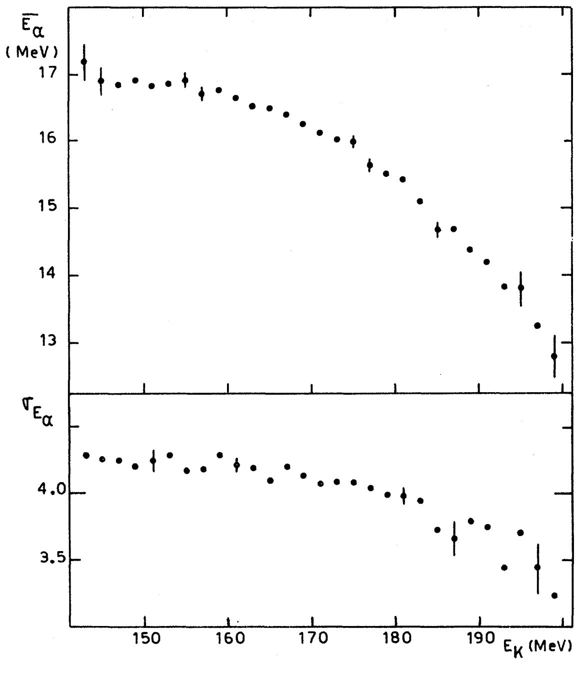

FIG. 16. The mean α-particle energy and its variance as a function of the total kinetic energy of the fragments in LRA fission.

correlation. Similarly if we assume that E_α^0 is the most relevant initial parameter in determining the correlation between $\bar{E}_K$ and E_α, as indicated by Boneh, Fraenkel, and Nebenzahl,[8] then the results on $d\bar{E}_K/dE_\alpha$ as a function of mass ratio suggest that E_α^0 decreases as one approaches the symmetric fission region. This should be considered in conjunction with the discussion in Sec. E that the increase in $\bar{E}_\alpha$ towards the symmetric fission region is due to the shift in the scission point towards the heavy fragment. This would imply that as the scission point shifts towards the heavy fragment the initial energy of the α particle may decrease but the final energy acquired by the α particle increases.

The results show that for a fixed fragment mass, both $\bar{\nu}_t$ and $\bar{E}_K$ decrease with increase in E_α, but the decrease of $\bar{\nu}_t$ is not linear and the variations in $\bar{E}_\gamma$ are small. The energy available must be constant (within the uncertainty of charge) when the mass is fixed and thus the above information indicates that the energy associated with neutrons must vary with E_α to compensate the nonlinear behavior of $\bar{\nu}_t$ with E_α. It is known that the kinetic energy of the neutron depends on $\bar{\nu}_t$ $[E_{\text{c.m.}}=0.65\times(\bar{\nu}_t+1)^{1/2}]$. However, the statistical accuracy of our slopes of these various energy components with respect to E_α is not sufficient to say whether the variation of E_n expected from this relation is enough to explain the effect.

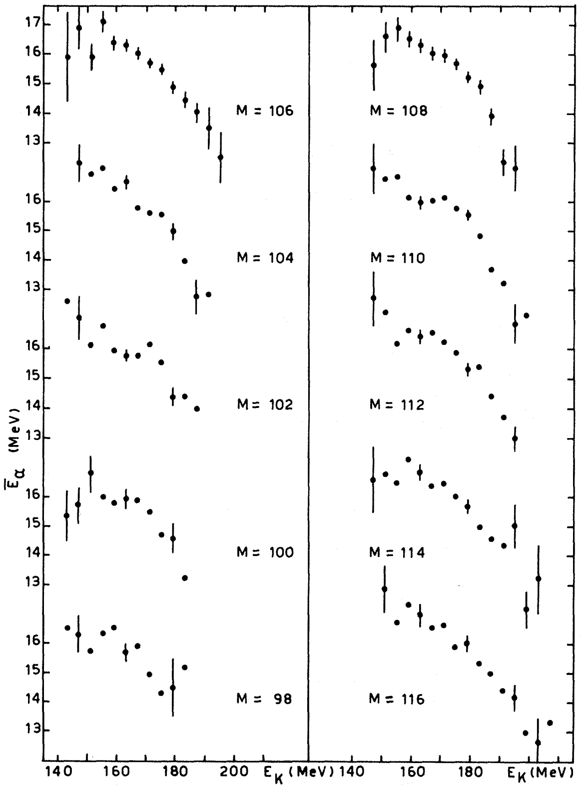

FIG. 17. The variation of the average energy of α particle with E_K for fixed values of fragment masses (mass grid = 2 amu).

An alternate source of this information is the variation of the slope $d\bar{\nu}_t/dE_K$ with E_α. This is shown in Fig. 15(c), for an average over a group of masses ($107 < M < 111$) to get a reasonable statistical accuracy. Within this limitation of having to average over certain masses we find that the observed effect of increase in $d\bar{\nu}_t/dE_K$ with E_α is consistent with the energy balance. That is, the relation

$$\frac{\Delta\bar{Q}}{\Delta E_\alpha}=\frac{\Delta\bar{E}_K}{\Delta E_\alpha}+\frac{\Delta\bar{E}_\gamma}{\Delta E_\alpha}+\frac{\Delta[\bar{\nu}_t(d\bar{\nu}_t/dE_K)^{-1}]}{\Delta E_\alpha}+1=0\,,$$

is satisfied by our experimental values of slopes.

V. SUMMARY

The present experiment has yielded an extensive information on the LRA fission. The results on the mean values of the quantities, such as $\bar{E}_K$ and $\bar{\nu}_t$, as a function of fragment mass ratio are in general agreement with those reported previously. However, the trend of variation of average neutron emission per fragment ($\bar{\nu}_1$) with fragment mass is observed to be quite different in comparison to the existing results. Moreover, markedly different results are obtained for the average α-particle energy ($\bar{E}_\alpha$) as a function of different fission parameters and for the experimentally observable variables as a function of E_α. A summary of the results follow:

(1) The detailed experimental results obtained here lead to a strong confirmation of the close similarity between the LRA and the binary scission configuration. But, it has not been possible to get any *direct* indication from the experiment to comment either way on the question whether or not the scission configuration in LRA fission is on the average a little more elongated than in binary fission.

(2) The differences in the average values of the

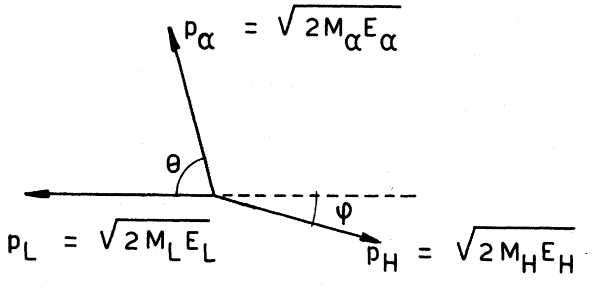

FIG. 18. Schematic diagram of the kinematics.

parameters characteristic of the fission process are indicated below:

$\overline{E}_K^B - \overline{E}_K^{\mathrm{LRA}} = 14.16 \pm 0.04$ MeV (pre-neutron emission),

$\overline{\nu}_t^B - \overline{\nu}_t^{\mathrm{LRA}} = 0.694 \pm 0.006$ MeV,

$\overline{E}_\gamma^B - \overline{E}_\gamma^{\mathrm{LRA}} = 1.01 \pm 0.02$ MeV,

$\overline{Q}^B - \overline{Q}^{\mathrm{LRA}} = 4.00 \pm 0.06$ MeV,

$\overline{\nu}_L^{\mathrm{LRA}} / \overline{\nu}_H^{\mathrm{LRA}} = 1.13$ (= 1.16 for binary fission).

(3) In as far as the assumption that the probability of α emission is independent of fragment mass ratio is reasonable, it is found that both light and heavy fragments contribute to the formation of the α particle. However, it should be emphasized that only the average nucleon contribution to the α formation is determined, and thus one cannot rule out the possibility that the α particle is emitted by the light or the heavy fragment, with some probability attached to each possibility, since it can yield the same result on the average nucleon contribution.

(4) The average energy of α particle increases as the fragment mass ratio approaches unity which is understood in terms of the shift of the scission point towards the heavy fragment as the symmetric fission region is approached. The variation of σ_{E_α} is consistent with this picture.

(5) The average number of neutrons emitted by both fragments ($\overline{\nu}_t$) has a complicated dependence on E_α indicating that the slope $d\overline{\nu}_t/dE_\alpha$ increases with E_α. Similar behavior is observed even when the fragment mass is fixed. The nonlinear dependence seems to be due to the averaging over the kinetic energy distribution of the fragments.

(6) The anticorrelation between $\overline{E}_K$ and E_α, for fixed fragment mass ratio, shows a strong dependence on fragment mass.

(7) The average binding energy of neutron in LRA fission is roughly 0.5 MeV lower in comparison with the binary fission fragments.

One of the aims of this experiment was to obtain some information on the initial energy of the α particle (E_α^0) at the scission point, which seems to be the critical parameter in determining the applicability of different models used in fission. It has really not been possible to get any concrete information on this and also on the initial kinetic energy of fission fragments (E_K^0) at the scission point. However, some qualitative arguments based on the results of various trajectory calculations indicate that:

(a) E_α^0 decreases towards the symmetric fission region;

(b) E_K^0 decreases as the deformation decreases.

These conclusions would only be reasonable if E_α^0 is really the most predominant factor in determining the correlations between E_K and E_α. If the distribution of fragment shapes at scission play a major role in producing these correlations, then these conclusions would perhaps be questionable. It is, in fact, due to this uncertainty that we are not able to comment on the magnitude of E_α^0. It is clear that the answer to this problem can only emerge from the detailed trajectory calculations and not directly from the experiment. We hope that the extensive results reported here will enhance the capabilities of the trajectory calculations for obtaining more physical information on the fission process.

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APPENDIX. CORRECTION OF THE MASS DISTRIBUTION CURVE FOR α RECOIL EFFECTS

The conservation of momentum is written as

$$p_\alpha \sin\theta = p_H \sin\phi , \tag{A1}$$

$$p_\alpha \cos\theta + p_L = p_H \cos\phi . \tag{A2}$$

Notations are those of Fig. 18. From (A1) we get

$$\sin\phi = p_\alpha / p_H \sin\theta$$

and

$$\cos\phi = (1 - p_\alpha^2 / p_H^2 \sin^2\theta)^{1/2} .$$

From (A2)

$$p_\alpha \cos\theta + p_L = (p_H^2 + p_\alpha^2 \sin^2\theta)^{1/2} ,$$

or

$$p_L^2 (1 + 2p_\alpha / p_L \cos\theta + p_\alpha^2 / p_L^2) = p_H^2 ,$$

which can be written as

$$a M_L / M_H = E_H / E_L ,$$

where

$$a = 1 + 2p_\alpha / p_L \cos\theta + p_\alpha^2 / p_L^2 ,$$

or

$$a = 1 + 2\cos\theta \left(\frac{M_\alpha E_\alpha M_0}{M_L M_H E_K} \right)^{1/2} + \frac{M_\alpha E_\alpha M_0}{M_L M_H E_K} .$$

Let us denote M_L^* and M_H^* the "masses" calculated without taking into account the α recoil effect:

$$M_L^*/M_H^* = E_H/E_L = aM_L/M_H ,$$

which can be written as

$$M_L^* = aM_L/[1+(a-1)M_L/M_0] .$$

The coefficient a varies very slowly with M_L so that we may assume in our calculations $da/dM_L = 0$. Then we get for the yields

$$y(M_L) = y^*(M_L^*)a/[1+(a-1)M_L/M_0]^2 ,$$

where y^* is the uncorrected yield and y the corrected one.

For our correction we need the variation of θ with mass ratio R. Detailed data on this point exist only for ^{235}U.[31,32] However, these results show an opposite trend as a function of R. In the case of ^{252}Cf we have only the results of Fraenkel, which show that the average θ as a function of R varies very little around the value of 81°. This led us to use this unique value for all the masses in our calculation of the corrections.

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