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Studies of mass and energy correlations in thermal neutron fission of U-235 accompanied by long range alpha particles

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Abstract. Several characteristics of fission accompanied by long range alpha particles (LRA) have been studied in the thermal neutron induced fission of ^{235}U. The kinetic energies of fission fragments and the LRA were measured with a back-to-back ionization chamber and semiconductor detectors respectively. The kinetic energies of the two fragments and the LRA in LRA fission, along with the energies of pair fragments in the normal binary fissions, were recorded event by event on a magnetic tape by means of a four-parameter data acquisition system. The data were analysed to study the dependence of different quantities in LRA fission on the fragment mass ratio, LRA energy and the total kinetic energy of the fission fragments. It is seen that the most probable energy of LRA increases significantly for near symmetric mass divisions. The total kinetic energy for all mass ratios in LRA fission is found to be $(2 \cdot 6 \pm 0 \cdot 7)$ MeV larger than that in binary fission. The difference in the total kinetic energies in LRA and binary fissions is seen to be dependent on mass ratio. This result may suggest that the scission configuration in LRA fission is different for different mass ratios. Correlations between the fission fragment and LRA energies have been studied for several mass ratios. It is seen that the most probable fragment kinetic energy $\bar{E}_k$ varies nearly linearly with the LRA energy E_α for various mass divisions but the variation of the most probable LRA energy $\bar{E}_\alpha$ with fragment kinetic energy E_k is found to deviate from linearity for several mass ratios. From a least square fit to the variation of $\bar{E}_k$ with E_α it is found that the slope $(d\bar{E}_k/dE_\alpha)$ increases with the increase in mass ratio. The present results are discussed to arrive at a better understanding of the scission configuration in the fission accompanied by LRA emission.

Keywords. LRA emission; thermal neutron fission of ^{235}U; mass, energy correlations.

1. Introduction

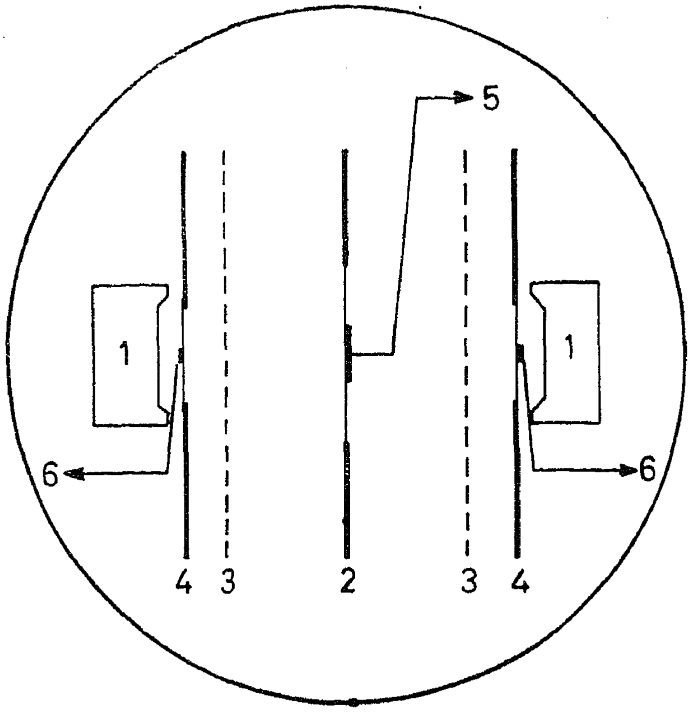
It is now well known that once in a few hundred fissions, a light charged particle, which is predominantly an alpha particle, accompanies the two heavy fragments. The energy and angular distributions of these long range alpha particles (LRA) suggest that they are emitted at a time very close to the scission of the fissioning nucleus. A number of experimental studies of this mode of fission has been undertaken in the past with a view to investigate the scission stage. Attempts have been made in the past to determine a set of initial parameters characterising the scission configuration by comparing the experimental observations with the results of trajectory calculations, based on Coulomb repulsion of three point charges. In order to determine the set of initial parameters in an unambiguous manner, it is necessary to have detailed experimental information on the distribution of energies and angles of LRA and the energies of the fission fragments of specified masses and the correlation between these quantities.

The first detailed work on the energy and angle measurements in LRA fission was reported by Fraenkel (1964) for the spontaneous fission of ^{252}Cf, where the fragment energies and LRA energies were measured at different angles. Various correlations on the mass, energy and angle of the three particles were obtained in these studies. Another detailed study for spontaneous fission of ^{252}Cf, ininvolving measurements of the energies of the prompt neutrons, LRA and fission fragments for specified fragment mass ratios was carried out by Mehta *et al* (1973). In this work also, various correlations between the fission fragment and the LRA energy were obtained as a function of fragment mass ratio. For the case of thermal neutron induced fission of ^{235}U, a three parameter measurement of the energies of the LRA and the pair fragments was carried out by Schmitt *et al* (1962) in which they obtained results on the mass distribution and the variation of average fragment kinetic energy with fragment mass ratio for different alpha particle energy groups. Recently, measurements on the alpha particle angle and energy and the fission fragment energy were carried out by Gazit *et al* (1971), but detailed correlations between fragment and LRA energies as a function of mass ratio were not reported. Experimental information on energy correlations between the energies of fragments and the LRA for specified mass divisions are, however, important for the study of the scission stage. Also from the point of view of comparing the Coulomb configuration at the scission point in binary and LRA fission, it is of interest to measure the distributions in the total kinetic energy (fragments + LRA energy) in LRA fission and that in binary fission for different mass divisions, which are not available from any of the earlier measurements. Moreover, all the previous measurements on the energy and mass correlations in LRA fission have been carried out with semiconductor detectors where the LRA were detected mainly around 90° with respect to the fission fragment direction. It is, therefore, of interest to carry out such multiparameter studies in which either the fragments or the LRA are detected in 4π geometry so as to eliminate any possible biasing of the data which might result due to the restricted angles of acceptance of the LRA. Such a geometry has also the obvious advantage of increasing the count rate in these experiments.

In the present work, a detailed study of the energy correlations between the fission fragments and the LRA has been carried out in the thermal neutron induced fission of ^{235}U by using a back-to-back gridded ionisation chamber for the measurement of the kinetic energies of the pair fragments in a 4π geometry and semiconductor detectors for the measurement of the energies of the LRA.

2. Experimental arrangement

Figure 1 shows a schematic diagram of the experimental assembly. A back-to-back gridded ionisation chamber was used for the measurement of the energies of the pair fragments in which the cathode-grid and the grid-collector distances were 2·1 cm and 0·7 cm respectively. A thin gold coated VYNS film, on which, a thin source of ^{235}U (94% enriched) of thickness $\sim 25\ \mu$ gm/cm^2 was deposited



1. LRA detectors
2. Cathode and source plate
3. Grids
4. Collectors
5. VYNS film with U^{235}
6. Al. windows with calibration sources.

Figure 1. Schematic diagram of experimental assembly.

at the centre by electro-spraying technique, formed the common cathode of the ionisation chamber. The chamber was filled with a mixture of argon (97%) and methane (3%) to a pressure of 1·5 atmospheres to ensure that the fragments are stopped in the cathode-grid region. The grid and cathode voltages were optimised for the best energy resolution of the ionisation chamber. The central region of the collectors had thin aluminium windows of 2 mg/cm^2 thickness which enabled the LRA to pass through and get detected in the semiconductor detectors mounted close to them (see figure 1). These detectors were of 2 cm^2 area and had depletion depths sufficient to stop alpha particles of energy up to 30 MeV. The detectors were mounted such that the line joining the centres of the detectors was parallel to the electric field direction in the ionisation chamber. The purpose of using two LRA detectors was to increase the LRA fission count rate. The LRA detectors were energy calibrated using natural alpha sources. These calibrations were checked at periodic intervals by means of two calibrated precision pulsers and also by means of low intensity alpha sources of ^{241}Am and ^{237}Np coated on the outer side of the thin aluminium windows of the collector plates facing the LRA detectors. The experiment was carried out at CIRUS reactor with the neutron beam of flux of about 3×10^7 cm^{-2} sec^{-1}.

A block diagram of the electronic arrangement used is shown in figure 2. Whenever a coincidence occurred between the fission pulses and any of the two LRA detector pulses within a resolution time (2τ) of 2 μ secs, a coincidence output pulse was generated. The pulses from the collectors of the ion-chamber and the LRA detectors, after suitable amplification, were gated by this coincidence output pulse and were recorded event by event on a magnetic tape by means of a four-parameter data acquisition system. For the coincidence requirements the fission

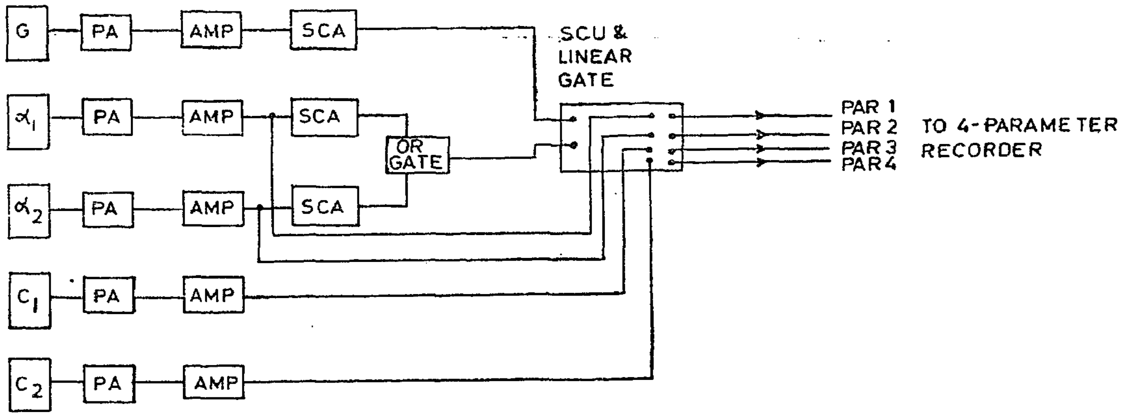


Figure 2. Block diagram of the electronic set up. PA—Preamplifier, AMP—Amplifier, SCA—Single Channel analyser, SCU—Slow Coincidence Unit.

pulse was derived from one of the grids of the ionisation chamber instead of the collectors because the latter pulses have intrinsic time jitter resulting from the different orientations of the fragment tracks with the electric field direction. The binary fission events, after suitable scaling down were also recorded along with the LRA fission events. A total of about 5×10^5 binary fission events and 3×10^4 LRA fission events were recorded in several runs spread over a period of two months.

3. Data analysis and experimental results

The data recorded on the magnetic tapes were analysed with a CDC–3600 computer to determine the various distributions in binary and LRA fission. The pulse heights of the LRA detectors were converted into energies on the basis of on-line energy calibrations as described earlier. The alpha particle energies E_α were then corrected for the energy loss in the gas and the thin aluminium window on the collector by using the energy loss tables (Northcliffe and Schilling 1970). Figure 3 shows the corrected LRA energy spectrum as measured by both the detectors, where the most probable value of the LRA energy, $\bar{E}_\alpha$, is seen to be $(16 \cdot 8 \pm 0 \cdot 5)$ MeV.

The fission fragment energy calibration was achieved by using the results of Schmitt *et al* (1966) for the peak energies of the light and heavy fragment groups. Due to the fragment–LRA angular correlations, in the present geometry, the fragments suffer more average energy loss in the source foil in LRA events as compared to those in the binary events. Therefore the data on the fragment energies in LRA events, obtained by using the same calibration as in binary events, were corrected for this additional energy loss in the foil as described in the appendix. It was estimated that the most probable light and heavy fragments in LRA fission suffer an extra energy loss of $(2 \cdot 1 \pm 0 \cdot 1)$ MeV and $(2 \cdot 3 \pm 0 \cdot 1)$ MeV respectively as compared to the binary case. Therefore, a correction of $4 \cdot 4$ MeV was applied to the average total fission fragment kinetic energies in the LRA fission to make comparisons with the results of binary fission. This correction was also calculated for different LRA energies taking into account the appropriate fragment-LRA angular correlations as described in the appendix. Figure 4 shows the results on the fragment kinetic energy distributions in binary and LRA fission,

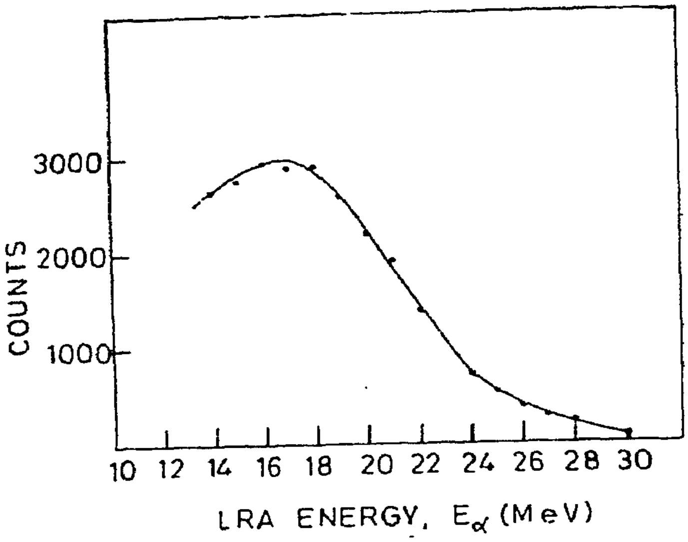


Figure 3. Summed up LRA spectrum of both the detectors.

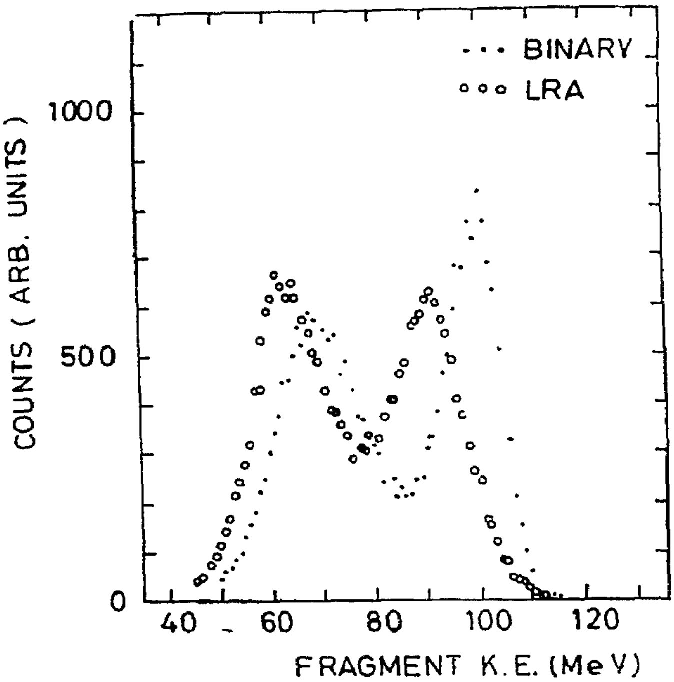


Figure 4. Kinetic energy distributions in binary and LRA fission.

It is found that in LRA fission, the energies of the heavy and light peaks are lower by $(5\cdot 2 \pm 0\cdot 5)$ MeV and $(8\cdot 9 \pm 0\cdot 5)$ MeV respectively as compared to the binary fission case. Figure 5 (*a*) shows the distribution of total fragment kinetic energy E_k in the case of LRA fission. The most probable value of the total fragment kinetic energy E_k in LRA fission is found to be $(155\cdot 4 \pm 0\cdot 5)$ MeV which is in agreement with the earlier measurements of Schmitt *et al* (1962) and Asghar *et al* (1970) but not with that of Gazit *et al* (1971) who have reported somewhat higher values.

The present data were also analysed to obtain for each event, the total kinetic energy E_T, which is the sum of the kinetic energy of the fragments and the LRA. The distribution of total kinetic energy in LRA fission is shown in figure 5 (*b*) along with that in the binary fission. It is found that the most probable values of the total kinetic energy in LRA and binary fission are $(171\cdot 3 \pm 0\cdot 5)$ MeV and $(168\cdot 7 \pm 0\cdot 5)$ MeV respectively. Table 1 summarizes the present results on the observed energy distributions in LRA and binary fission along with the results of Gazit *et al* (1971) for the sake of comparison.

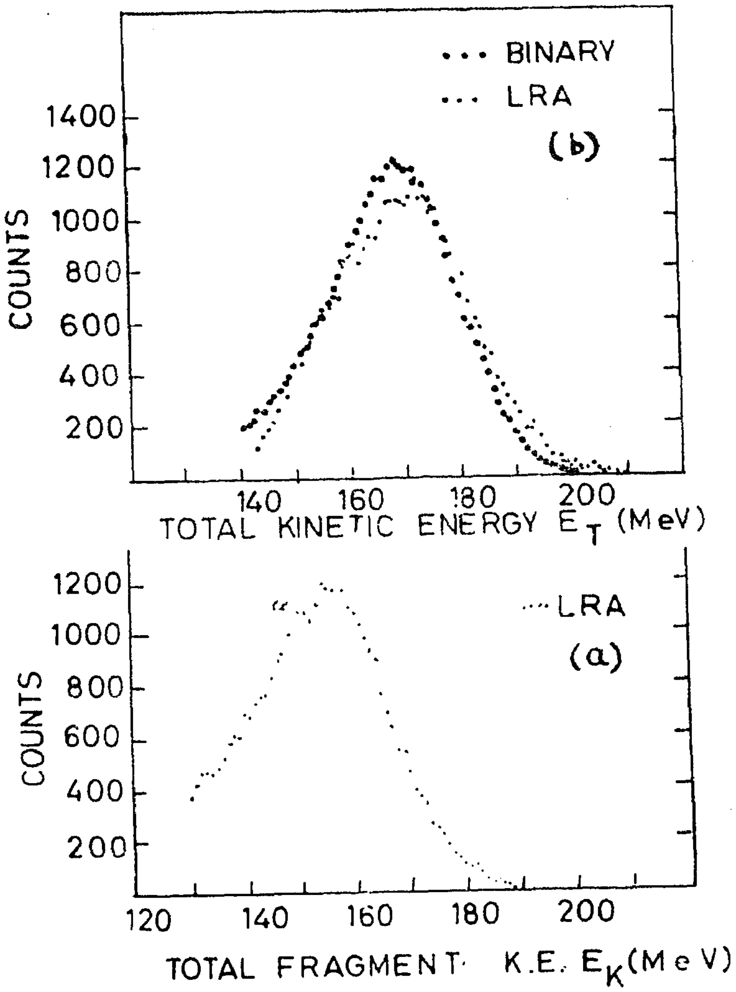


Figure 5. (*a*) Fragment kinetic energy spectrum in LRA fission. (*b*) Total kinetic energy distributions in binary and LRA fission.

Table 1. Observed energy distributions in LRA and binary fission along with results of Gazit *et al*

		Present work		Gazit *et al* (1971)	
		LRA (MeV)	Binary (MeV)	LRA at 90° (MeV)	Binary (MeV)
Heavy fragment energy	$\bar{E}_H$	63·9±0·3	69·1±0·2	64·77±0·15	69·91±0·13
	σ_{E_H}	7·2±0·2	8·3±0·1	6·4 ±0·02	8·0 ±0·02
Light fragment energy	$\bar{E}_L$	90·2±0·3	99·1±0·2	92·82±0·10	100·42±0·10
	σ_{E_L}	6·9±0·1	5·1±0·1	4·47±0·02	4·66±0·02
Total fragment energy	$\bar{E}_k$	155·4±0·5	168·7±0·5	157·46±0·4	168·55±0·04
	σ_{E_k}	10·8±0·2	10·8±0·2	8·74±0·02	9·88±0·02
Total kinetic energy	$\bar{E}_T$	177·3±0·5	168·7±0·5	..	..
	σ_{E_T}	11·4±0·2	10·8±0·2		
Alpha particle energy	$\bar{E}_\alpha$	16·8±0·5	..	15·8±0·2	..
	σ_{E_α}	4·1±0·4	..	4·5±0·2	..

The most probable total kinetic energy, $\tilde{E}_T$, was also obtained as a function of fragment mass division. The masses of the two pair fragments M_1 and M_2 were obtained by the simple momentum and mass conservation relation, $E_1M_1 = E_2M_2$ and $M_1 + M_2 = M$, where $M = 236$ for binary and $M = 232$ for LRA fission. For E_1 and E_2 the measured kinetic energies were used neglecting the neutron emission effects, as the neutron emission data for different fragment mass and kinetic energy are not available for LRA fission. The mass distributions obtained for binary and LRA fission are shown in figure 6. From a comparison of the observed mass distribution in binary fission with the radiochemical data (Wahl 1965), the mass dispersion σ in the present measurement was estimated to be ~ 2 amu for binary fission, and a comparison of the present LRA mass distribution with that obtained by Schmitt *et al* (1962) shows that for LRA events the mass dispersion is increased to $\sim 4 \cdot 5$ amu. This increase in the mass dispersion for LRA events is due to the neglecting of the alpha particle recoil effect in the determination of fragment masses and also due to a somewhat larger energy loss of fragments in the source foil for LRA fission events in the present geometry.

The most probable values of the total kinetic energy released in binary and LRA fission as a function of fragment mass ratio are plotted in figure 7.

The most probable LRA energy $\bar{E}_\alpha$, obtained by fitting Gaussian distribution to the LRA spectra, are shown in figure 8 (a) for different mass ratios.

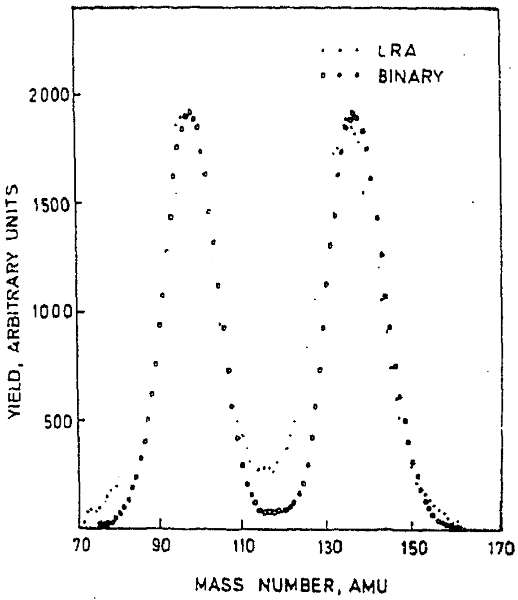


Figure 6. Mass distributions in binary and LRA fission.

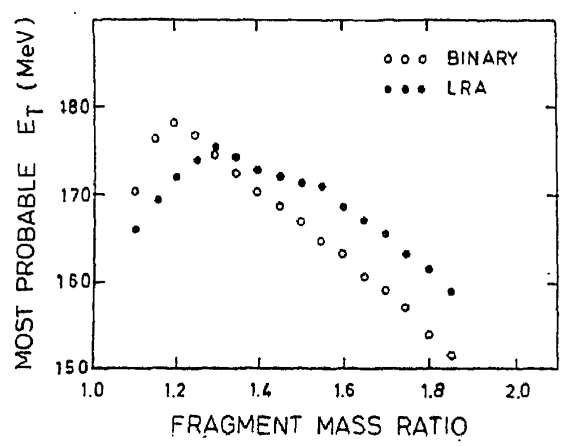


Figure 7. Most probable values of total kinetic entrgy in binary and LRA fission as a function of fragment mass ratio.

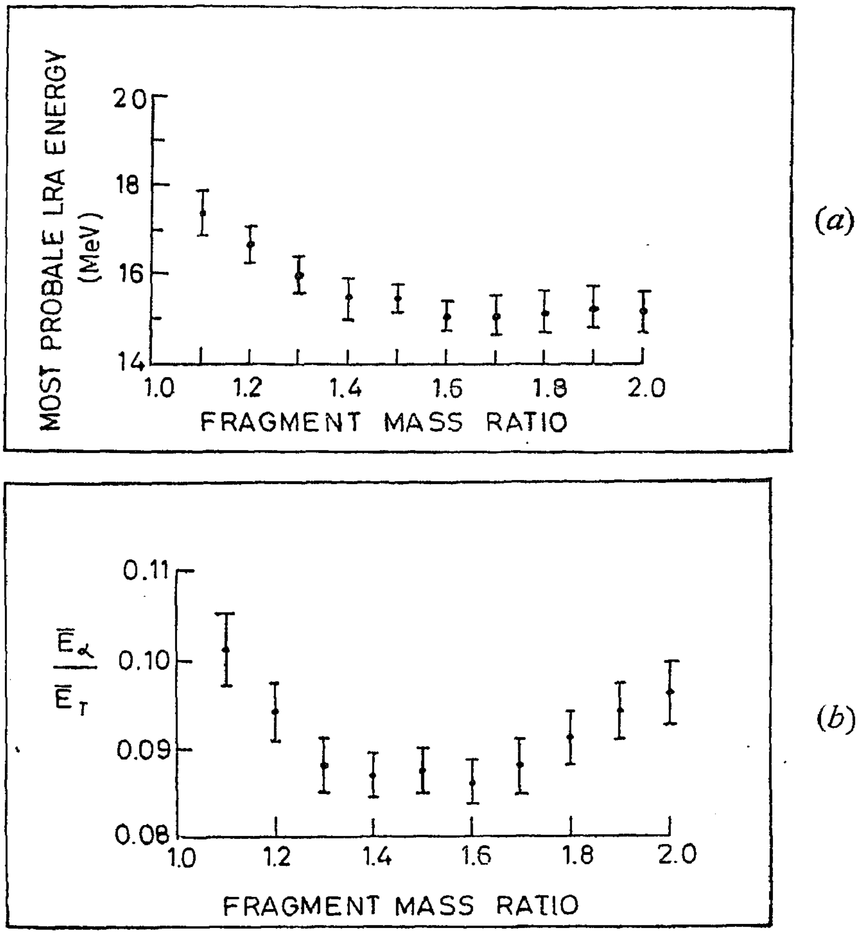


Figure 8. (*a*) Most probable valaes of LRA energy as a function of fragment mass ratio.
(*b*) A plot of the quantity $\bar{E}_a/\bar{E}_T$ as a function of fragment mass ratio. $\bar{E}_a$ and $\bar{E}_T$ are the most probablt values of the LRA and total kinetic energy.

The data were also analysed to obtain detailed correlations between the fragment kinetic energy and the LRA energy for different mass divisions. Figure 9 shows the dependence of the most probable value of fission fragment kinetic energy $\bar{E}_k$ on the LRA energy E_a for various mass ratios. This correlation averaged over all mass ratios is also shown in figure 9. The slope $d\bar{E}_k/dE_a$ of the correlation averaged over all mass ratios is found to be $(-0{\cdot}2 \pm 0{\cdot}05)$. Similarly the correlations between $\bar{E}_a$ and E_k have been obtained for different mass ratios and are plotted in figure 10, where $\bar{E}_a$ are the most probable energies obtained by fitting Gaussian distributions to the observed LRA energy spectra for different fragment kinetic energies.

4. Discussion

From table 1 it is seen that, within the experimental errors, the most probable total kinetic energy $\bar{E}_T$, *i.e.*, $\overline{(E_k + E_a)}$ in LRA fission is found to be equal to the sum of the most probable kinetic energies $\bar{E}_k$ and $\bar{E}_a$ of the fission fragments and the LRA respectively. It is also found that the total kinetic energy E_T averaged over all mass ratios in LRA fission is $(2{\cdot}6 \pm 0{\cdot}7)$ MeV higher than that in binary

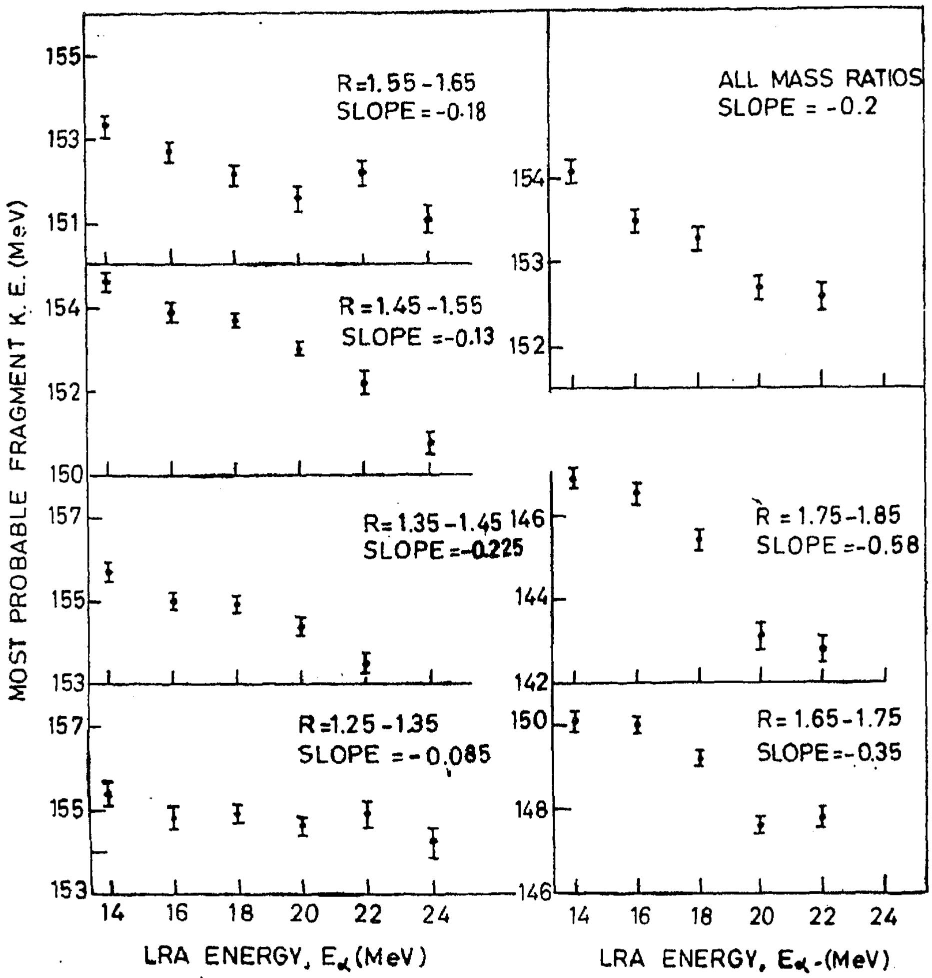


Figure 9. Most probable values of fragment kinetic energy as a function of LRA energy for various mass divisions.

fission. This result is in agreement with the results of earlier investigations (Gazit *et al* 1971). Combined with the trajectory calculations this result suggests that, on the average, the scission configuration of LRA fission is more stretched than that of binary fission. From figure 7 it is seen that the difference in $\bar{E}_T$ in LRA and binary fissions is dependent on the mass division. This may suggest that the scission configurations in LRA fission are different for different mass divisions.

Although the most probable LRA energy $\bar{E}_\alpha$ is seen to be constant over a large range of mass divisions ($R > 1 \cdot 4$), it increases appreciably in the symmetric region. A similar result has also been observed in the case of spontaneous fission of ^{252}Cf (Mehta *et al* 1973). This result indicates that for near symmetric mass divisions, either the alpha particle is emitted with a higher initial kinetic energy or it is born in a region of higher Coulomb energy. The alpha particles may be liberated in a region of higher potential energy in symmetric mode if in this mode of splitting the scission point is nearer to the heavy fragments as has often been assumed to explain the neutron emission characteristics of the fragments. An increase in the alpha particle energy for symmetric mode was also suggested by the trajectory calculations of Fong (1970) in which the fragments and the alpha particle were assumed to have very small kinetic energy at scission.

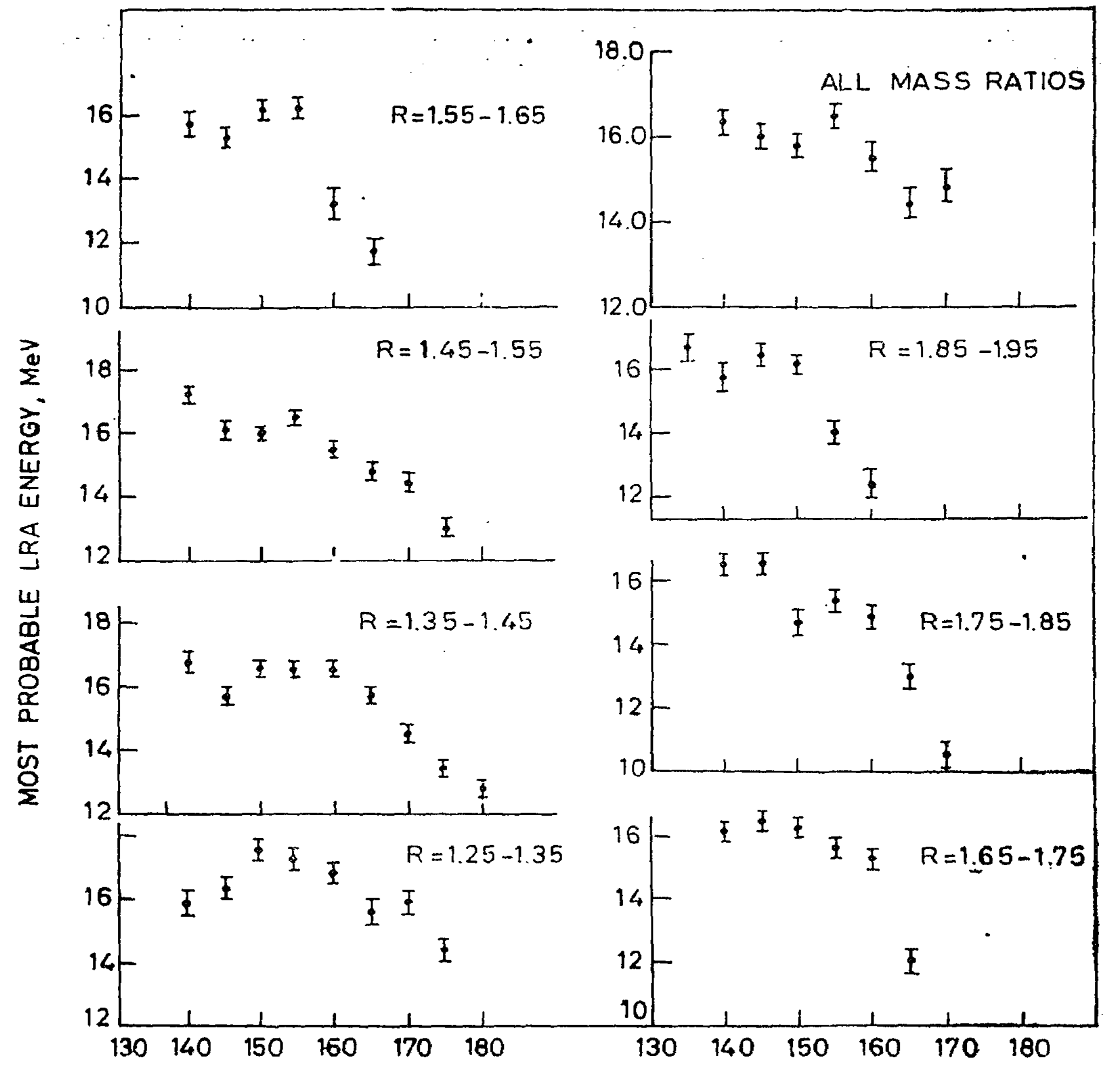


Figure 10. Most probable values of LRA energy as a function of fragment kinetic energy for various mass divisions.

Another quantity of interest is the fraction of total Coulomb energy at scission which appears as the LRA kinetic energy. The quantity $\bar{E}_\alpha/\bar{E}_T$ is a good measure of this fraction, if it is assumed that the initial energies of the alpha particle and the fission fragments at the time of scission are very small as compared to the observed final energies. A plot of $\bar{E}_\alpha/\bar{E}_T$, as a function of fragment mass ratio, is shown in figure 8 (*b*), where it is seen that this fraction increases both for very asymmetric and near symmetric mass divisions. This result suggests that for very asymmetric and near symmetric modes of fragment mass divisions, the alpha particle spends relatively more time in the vicinity of the two heavy fragments and hence it acquires a larger fraction of the total energy available. This is possible if for symmetric and very asymmetric modes the alpha particle appears nearer to one of the two fragments in which case it gains a larger fraction of energy due to reflections in the vicinity of the heavy fragments before moving out (Fong 1970).

The observed anticorrelation between the fission fragment and LRA energies contains information about how the total energy available at scission is shared between the LRA and the two heavy fragments. If the final energies were entirely derived from the Coulomb field and if no averaging factors were involved, this anticorrelation should be unity. The observation of a significantly reduced anti-correlation even for specified mass ratios, as seen in figures 9 and 10, reflects the non-uniqueness of the Coulomb confiugration at the scission point and possibly the presence of appreciable initial kinetic energies of the three particles at scission as has also been pointed out earlier (Halpern 1971). It is seen from figures 9 and 10, that while $\bar{E}_k$ varies nearly linearly with E_a for all the mass ratios, the dependence of $\bar{E}_a$ on E_k significantly deviates from linearity for several mass ratios. The slopes of the anticorrelation $d\bar{E}_k/dE_a$, obtained by a linear least square fit to the data, are also given in figure 9. It can be seen from figure 11 that the anticorrelation increases with the increase in mass asymmetry. This result does not agree with the results obtained by Mehta *et al* (1973) for spontaneous fission of ^{252}Cf. The correlation of $\bar{E}_k$ and E_a averaged over all mass ratios is also shown in figure 9 and this anticorrelation is found to have a slope of $(-0{\cdot}2 \pm 0{\cdot}05)$ which is lower than the slope $(-0{\cdot}44 \pm 0{\cdot}06)$ reported by Gazit *et al* (1971). This difference appears to be due to the different geometries used in these two measurements, since the slope of the anticorrelation reported by Gazit *et al* (1971) corresponds to the case where LRA are detected mostly around 90° with respect to fission fragment, while in the present geometry the observed correlation is that averaged over all angles between LRA and the fission fragments. This difference then implies that the slope $d\bar{E}_k/dE_a$ depends on the angle between LRA and fission fragments which nevertheless needs to be investigated by further experiments, since it is also possible that this difference in the results of these two measurements may partly arise due to uncertainties in the data on energy-angle correlations of LRA for specified mass divisions which were used in the estimation of the energy loss corrections in the present work.

Attempts have been made in the past to obtain information about the scission configuration by comparing the results of a three-point-charge trajectory calculation with the experimental results. These studies (Boneh *et al* 1967, Fong 1970, Gazit *et al* 1971) have however led to different conclusions regarding the initial parameters at scission, due to varying assumptions about the number of degrees of freedom at scission and also due to fitting of different types of observations. Another factor which is not taken into account in the trajectory calculations is the possible correlation between these initial parameters themselves. In

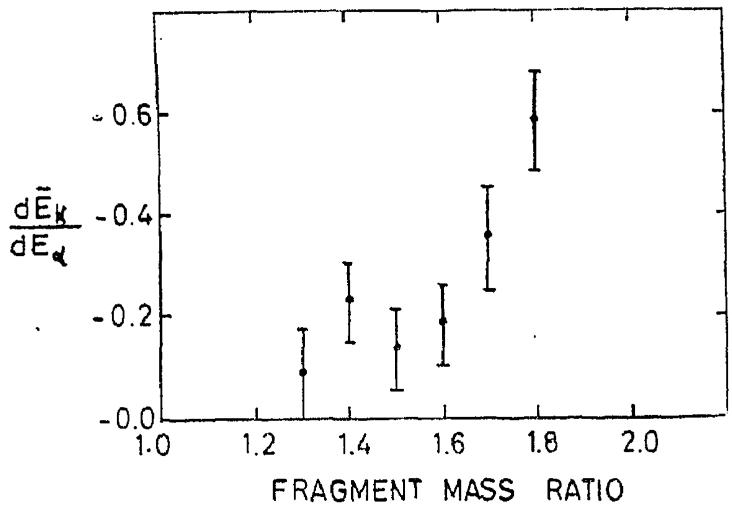


Figure 11. Variation of $d\bar{E}_k/d\bar{E}_a$ with fragment mass ratio.

some earlier works (Boneh *et al* 1967, Gazit *et al* 1971, Krishnarajulu and and Mehta 1975) in which the anticorrelation observed between $\bar{E}_k$ and E_a for the case of ^{252}Cf were fitted to the results of trajectory calculations, rather large initial (prescission) fragment kinetic energies ($E_k^\circ = 10$–40 MeV) had to be assumed. On the other hand, the anticorrelation between $\bar{E}_k$ and E_a and the non-linear correlation between $\bar{E}_a$ and E_k as observed for ^{252}Cf fission have been obtained by Choudhury and Ramamurthy (1974) in trajectory calculations with only three parameters in which the initial energies of the alpha particle and the fission fragments were assumed to be negligible and the distributions of the other parameters were assumed to be of Gaussian shape. Thus it appears that widely different assumptions about the initial kinetic energies could explain the experimental results on the anti-correlations. It is, however, hoped that a number of other experimental results obtained in the present work will be able to decide the validity of the different models of the trajectory calculations, and to determine the initial parameters unambiguously. We propose to undertake further experimental studies of the LRA fission including the measurement of the angle between the LRA and the fission fragments and then attempt to utilize all the results for ^{236}U fission to arrive at a unique set of parameters characterizing the scission configuration.

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Appendix

In the present geometry, the average thickness seen by the fission fragments is dependent on the angular distribution of the fission fragments with respect to the direction of LRA detection. In binary fission, fission fragments are emitted isotropically and so the average target thickness for these events is given by

$$t_{\text{binary}} = \frac{\int_0^{\theta_{\max}} \frac{t}{\cos\theta}\, 2\pi \sin\theta \, d\theta}{\int_0^{\theta_{\max}} 2\pi \sin\theta \, d\theta} \quad (1)$$

where t is the half-thickness of the source foil and θ is the angle between the fission fragment direction and the electric field, which is same as the direction of LRA detection. $\theta_{\max}$ is the maximum value of the acceptance angle which was restricted to about 86° in the present geometry due to the mounting arrangement of the source on the cathode plate. For LRA fission events, due to fission fragment-alpha particle angular correlation, the average thickness was obtained by the relation,

$$t_{LRA} = \frac{\int_0^{\theta_{max}} \frac{t}{\cos\theta} \exp\left(-(\theta-\bar{\theta})^2/2\sigma^2\right) 2\pi \sin\theta \, d\theta}{\int_0^{\theta_{max}} \exp\left(-(\theta-\bar{\theta})^2/2\sigma^2\right) 2\pi \sin\theta \, d\theta} \quad (2)$$

where the exponential expression represents the fission fragment-LRA angular correlation, with $\bar{\theta}$ and σ as the average angle and the standard deviation of the angular distribution.

For the fragment-LRA angular correlation, we have used the results of earlier workers (Gazit *et al* 1971, Nadkarni *et al* 1972) and estimated the average thickness seen by the binary and LRA fission fragments for various fragment mass ratios and for different alpha particle energies. As a typical value for all the events, the average thickness seen by the binary and LRA fission fragments was estimated to be $2 \cdot 83\, t$ and $4 \cdot 1\, t$ respectively. Thus an extra thickness of $1 \cdot 27\, t$ is seen by the LRA fission fragments as compared to the binary case. The energy correction to be applied to the observed fragment energies in LRA fission was calculated by knowing the average energy loss of the fission fragments in the normal thickness of the source foil. Since the energy loss data for fission fragments in various materials is not very well known, and also since the thickness of VYNS foil and the gold coating cannot be measured very accurately, we adopted a more direct method for obtaining this value. The energy shifts of the light and heavy peaks of a spontaneous fission source of ^{252}Cf were measured by means of a solid state detector when the source foil is introduced between them. In this way it was found that the extra average energy loss by the light and heavy fragments in LRA fission are $(2 \cdot 1 \pm 0 \cdot 1)$ and $(2 \cdot 3 \pm 0 \cdot 1)$ MeV respectively, giving rise to a total average energy loss of $4 \cdot 4 \pm 0 \cdot 2$ to the total fission fragment kinetic energy. Similarly, this extra energy loss in LRA fission was determined for different values of LRA energy, by using the appropriate LRA-fragment angular distributions for the calculation of the extra thickness.

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