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Studies of light-charged particle emission in fission at Trombay

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Abstract. Studies of prompt radiations emitted in fission were started at Trombay in the late 1950's by Dr R Ramanna and over the years extensive investigations on the emission of prompt neutrons, gamma ray and K x-rays in fission were carried out with neutron beams from APSARA and CIRUS reactors. In the early 1960's studies on the emission of light-charged particles in fission, which is a rare mode of fission, were also started. This paper reviews some of the recent studies on the emission of light-charged particles (LCP) in fission which were carried out with a view to investigate the mechanism of LCP emission, the scission configuration and the dynamics of the last stages of the fission process.

Keywords. Nuclear fission; light-charged particle emission; emission probability; energy-angular-distribution.

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1. Introduction

Nuclear fission process, which involves collective motion, large scale deformation and subsequent division of the fissioning nucleus into two fragments has provided over the years a convenient way to unfold macroscopic nuclear behaviour and to study nuclear dynamics. Only in recent years with the availability of energetic heavy ion beams the studies of nucleus-nucleus collision have provided a complementary and perhaps a more powerful way to investigate the domains of nuclear macrophysics which could earlier be probed only through the study of fission process. In the fisson process once the nucleus is given sufficient energy to reach the top of the fission barrier (saddle point) it continues to elongate until it splits into two fragments at what we call "scission point". The deformed fragment nuclei formed at scission convert almost instantly their deformation energies into excitation energy *via* damped vibrations around their stable shapes. Simultaneously, the resulting excited fragments undergo acceleration under the mutual Coulomb repulsion and acquire their full kinetic energies in a very short time ($\leqslant 10^{-20}$ sec) after scission. During the de-excitation of the fragments most ($\sim 90\,\%$) of the prompt neutrons are emitted after the fragments have reached their full kinetic energies. The excitation energy below the neutron binding energy is given off as prompt gamma rays *via* gamma transitions cascading down from the excited states to the ground state of the emitting fragment. A small fraction (about 6 %) of these transitions undergo internal conversion resulting in the emission of conversion electrons and the characteristic x-rays of the fragments. Occasionally, along with the two heavier fragment nuclei the fission process also results in the emission of a light-charged particle (LCP) and this process is called "LCP-accompanied fission" or simply as "ternary fission". As the bulk of these light particles are long range alpha particles (LRA) this process is also often referred to as "LRA-accompanied fission" particularly when measured without particle-identification of the LCP. As this process has a very low probability of occurrence of about 2×10^{-3} as compared to the normal binary fission, detailed investigations of this process have been possible only for thermally fissile nuclei with the use of high flux reactors or for the spontaneous fission of ^{252}Cf.

Investigations on the nuclear fission process were initiated at Trombay by Dr R Ramanna in the late 1950's and over the years considerable work has been carried out at Trombay with neutron beams from APSARA and CIRUS reactors on the various aspects of the fission process using physical and radiochemical techniques. The work using radiochemical techniques has been reviewed in another article by Dr Ramaniah in this volume. Among the earliest investigations in fission using physical techniques at Trombay were the measurements of the energy spectrum of the neutrons and neutron-fragment angular correlations in thermal neutron induced fission of ^{235}U (Ramanna *et al* 1961; Kapoor *et al* 1963). The neutron-fragment angular correlations showed that a small fraction of neutrons ($\sim 10\%$ of total) does not originate from the de-excitation of the moving fragments and is emitted isotropically in the laboratory system with an evaporation-like energy spectrum. In this work, emission spectrum of the neutrons emitted from fully accelerated fragments was shown to be a superposition of various evaporation spectra with a distribution of nuclear temperatures of the form $P(T) = 2T/T_{m^2}$ with a maximum temperature T_m. This type of distribution has formed the basis of several recent theoretical calculations of the fission neutron spectra (Madland and Nix 1982). In a subsequent study the prompt gamma-fragment angular correlations were measured which showed an anisotropic gamma ray emission (anisotropy $\sim$ 10–15 %) suggesting the occurrence of large angular momenta of fission fragments brought about possibly by the non-axial splitting of the neck-region of the fissioning nucleus at scission (Kapoor and Ramanna 1964). Several studies on the emission of K x-rays from fission fragments in spontaneous fission of ^{252}Cf and thermal neutron induced fission of ^{235}U were also later carried out. Among these were the measurements, for the first time, of the multiplicities of K x-rays emitted by the fission fragments of specified atomic numbers and the first and second moments of the x-ray emission distribution function (Kapoor *et al* 1971a,b). The average yields and emission times of K x-rays emitted from individual fission fragments were determined and from these the onset of a new region of nuclear deformation corresponding to neutron number $N > 88$ was established. It was also seen that there exists a significantly large probability of cascade emission of K x-rays in several cases.

Among other areas of fission physics research undertaken at Trombay were studies of fragment angular distributions (Nadkarni *et al* 1968b), correlation between fragment anisotropy and asymmetry in fast neutron-induced fission (Kapoor *et al* 1965) and correlation between fragment mass, charge and kinetic energy in thermal neutron fission (Govil *et al* 1983). We do not intend to discuss these investigations of binary fission process here but rather focus on some of the studies of the LCP-accompanied fission undertaken at Trombay.

The emission of LCP in fission has several characteristic features: (i) they are emitted with a very low probability (P_{LCP}) which is one in about 300 to 500 binary fissions, (ii) a majority of these LCP are isotopes of H and He, with almost 90 % of them being long range alpha particles (LRA), (iii) they are emitted with a broad energy spectrum with a near-Gaussian shape, which ranges from 5 to 30 MeV with a peak around 15 MeV and FWHM of about 10 MeV and (iv) they are emitted nearly perpendicular to the direction of fission fragment motion, more precisely at an average angle $\overline{\theta}_{\alpha L}$ of about 83° with respect to the direction of motion of the light fragment. It has been inferred from these characteristics that they are emitted close to the scission stage from the neck region between the two nascent fission fragments. In order to deduce information about the scission configuration as well as the dynamics of the fission process at the instant of their emission during the last stages of scission, more detailed information about the correlations among the various measured parameters is required. In the early 1960's Ramanna and coworkers started investigations on the emisson of LRA in fission and the first study in this series was measurements of the angular distribution of the LRA and ternary fission fragments in 14 MeV neutron induced fission of ^{238}U (Ramanna *et al* 1963). Numerous other studies of the LCP-accompanied fission followed in the subsequent years with the aim of investigating not only the mechanism of emission of these particles but also the scission configuration and the nuclear dynamics of the last stages of the fission process, using these LCP as probes. In what follows, some of the recent investigations on the LCP-accompanied fission carried out at Trombay are briefly reviewed.

2. Studies of LCP emission in ^{235}U (n_{th}, f)

With a view to study the effect of the expected sharp variation of the Coulomb field around the inter-fragment (neck) region on the energy of the LRA, the energy spectra of LRA emitted at several angles with respect to the fragment direction were measured (Nadkarni *et al* 1968, 1972). From the energy spectra observed at these angles the width represented by σ_θ of the Gaussian-shaped LRA-fragment angular correlation function was deduced for LRA of different energies. These values of σ_θ *vs* LRA energy E_α are shown in figure 1 where a shallow minimum of σ_θ at $E_\alpha \sim 17$ MeV is observed. It was noticed that the yield of LRA does not decrease to a very low value at such small angles as 27° or 11° as would be expected for a true Gaussian distribution. Most of the LRA yield at these

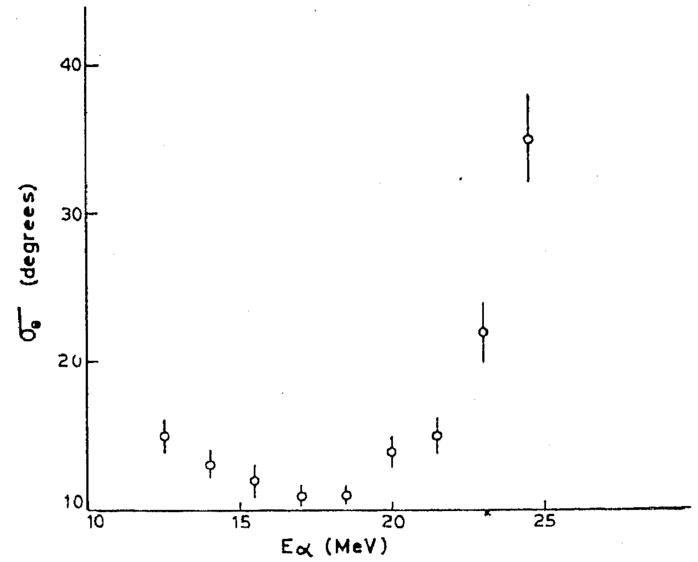

Figure 1. The variation of σ_θ of the Gaussian angular correlation function with alpha particle energy (Nadkarni *et al* 1972).

small angles, belonging to a non-Gaussian component, shows a significantly higher most probable energy (22–24 MeV) (Nadkarni *et al* 1972). LRA with energies beyond 26 MeV were found to be emitted preferentially along the direction of the fragments (Nadkarni and Rama Rao 1968a). This non-Gaussian LRA component, which has been called polar LRA (Piasecki *et al* 1970) was attributed to evaporation from moving fission fragments which then explains the observed most probable energies at these small angles. Estimates of Γ_α/Γ_n based on the statistical theory showed that at least some of the 'polar' LRA may be accounted for as evaporation from moving fission fragments (Nadkarni *et al* 1972).

Considerable insight into the mechanism of LCP emission in fission as well as of the fission process itself can be got from a knowledge of the kinetic energy E_{K_0} of the fragments at the moment of liberation of the light-charged particles. Considering that LCP are liberated near the scission configuration, the quantity E_{K_0} also corresponds to the prescission kinetic energy in the normal binary fission process. The magnitude of E_{K_0} in the fission process is expected to be dependent on the degree of coupling of the collective and single particle degrees of freedom, and is therefore related to the magnitude of nuclear viscosity operating during the descent of the nucleus from saddle to scission. A direct determination of the magnitude of E_{K_0} acquired by the fragments in a time of the order of few times 10^{-21} sec is not possible. But it has been realized for some time that the LCP can serve as probes to gauge this quantity through measurements of the various correlations in LCP accompanied fission (Frenkel 1967; Mehta *et al* 1973).

In the LRA-fission process several variables or parameters are involved such as the fission fragment mass (M_f), total fragment kinetic energy (E_K), LRA energy (E_α) and LRA-fragment angle $\theta_{\alpha f}$. Although in earlier studies (Fraenkel 1967; Mehta *et al* 1973) correlation of $\overline{E}_\alpha$ and E_K has been measured and analysed using trajectory calculations, a definite result on the magnitude of E_{K_0} has been difficult to deduce from a limited set of measured correlations. As with more detailed data on differential correlation among all possible LRA-fission parameters it beomes possible to obtain more accurately the scission parameters and hence E_{K_0}, detailed multiparameter correlation measurements between the parameters M_f, E_k, E_α and $\theta_{\alpha f}$ in thermal neutron induced fission of ^{235}U were carried out at Trombay with CIRUS reactor (Choudhury *et al* 1980). A back-to-back gridded ionization chamber (figure 2) was used to measure kinetic energies of the pair fragments and the angle $\theta_{\alpha f}$ was measured by an electronic method based on the measurement of coincident pulse heights of the grid and the collector pulses (Choudhury *et al* 1979). Pulse heights of the two collectors, the two grids and of the LRA detector placed along the electric field direction of the chamber were recorded event-by-event on a magnetic tape by means of a 6-parameter data acquisition system. The data were analysed to obtain M_f, E_K, E_α and $\theta_{\alpha L}$ for each event and a probability matrix P (M_f, E_K, E_α, E_T, $\theta_{\alpha L}$) was constructed, where total kinetic energy released is $E_T = E_K + E_\alpha$, and $\theta_{\alpha L}$ is the angle between LRA and the light fission fragment. From this matrix, the correlation between any two parameters for specific windows on the remaining parameters were obtained. Various correlations of interest were deduced and analysed in this work and we show here only a few selected results. Figure 3 shows the ternary fission fragment mass distribution, where the correction due to alpha particle recoil has been incorporated, together with the binary fission fragment mass distribution. The light fragment mass peak is observed to be shifted to the lower side whereas the heavy fragment peak is not shifted appreciably. This has been interpreted in terms of the

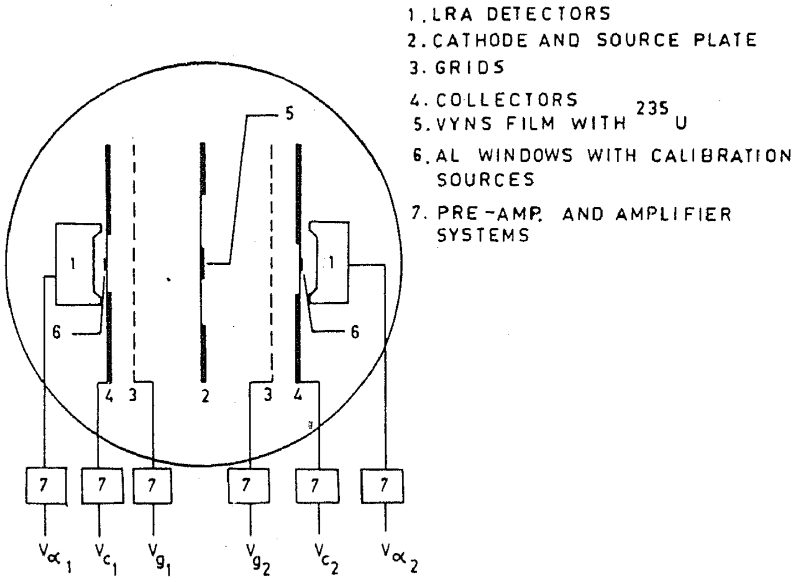

Figure 2. Schematic diagram of the experimental set up used in the LRA correlation experiment (Choudhury *et al* 1980).

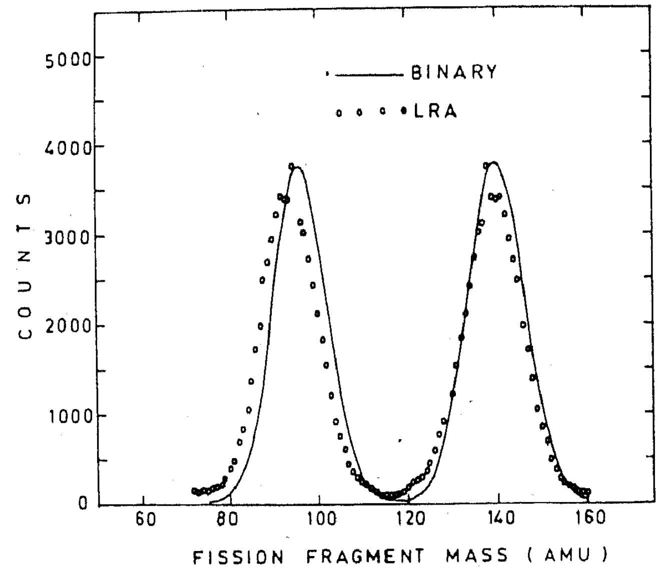

Figure 3. Mass distribution of the fission fragments in binary and LRA accompanied fission (Choudhury *et al* 1980).

dependence of the alpha particle emission probability P_α on fragment mass (Choudhury *et al* 1980). Figure 4 shows the variation of the most probable value of the total kinetic energy E_T and σ_{E_T} with heavy fragment mass M_H in LRA and binary fission, where a significantly larger σ_{E_T} is observed in LRA accompanied fission for $M_H \sim 130$ (near closed shell region) whereas nearly equal σ_{E_T} is observed for $M_H \gtrsim 140$ (away from closed shell). These results were interpreted in terms of differences in the dependence of P_α on the interfragment separation (d) for these two mass regions. An important correlation is the dependence of the width $\sigma_{\theta_{\alpha L}}$ of the LRA-light fragment

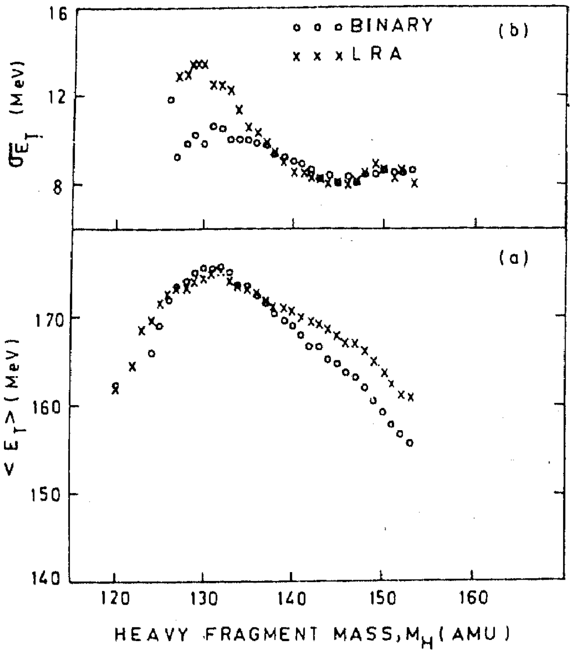

Figure 4. Variation of $\bar{E}_T$ and σ_{E_T} of the total kinetic energy (E_T) distribution with heavy fragment mass M_H in binary and LRA-accompanied fission (Choudhury *et al* 1980).

angular correlation on the total fragment kinetic energy E_K (figure 5). If the variation of $\sigma_{\theta_{\alpha L}}$ with E_K is examined under the two assumptions that the variation of the final E_K can be due either to (i) the variation of the prescission kinetic energy E_{K_0} or (ii) the variation in the Coulomb energy, the case (i) leads to a larger $\sigma_{\theta_{\alpha L}}$ with increasing E_K whereas the case (ii) leads to a smaller $\sigma_{\theta_{\alpha L}}$ with increasing E_K. The observed $\sigma_{\theta_{\alpha L}}$ *vs* E_K favours the case (ii) implying that E_{K_0} does not increase with E_K which is expected only if E_{K_0} has a small magnitude. Trajectory calculations based on three-point charge approximation have been done to fit these data to arrive at an initial set of scission parameters and their widths. From the above studies it has been concluded that the fission fragments have a very small prescission kinetic energy and that the alpha particle is liberated close to either of the two fragments and slightly off the axis joining the two fragment centres. This result has a direct relevance to the nature of fission dynamics and points to a strong coupling between the collective and internal degrees of freedom suggesting applicability of the statistical models during the saddle to scission.

In thermal neutron induced fission of ^{235}U a still more rare mode of fission in which two light charged particles are emitted simultaneously together with the two heavier fragments was first observed at Trombay (Kapoor *et al* 1972). The probability of such events was estimated to be about one per million binary events and the gross features of LCP energy spectrum observed in this case were found to be similar to those in ternary fission. Later in spontaneous fission of ^{252}Cf these LCP were identified as ^{1}H, ^{3}H and ^{4}He and their relative yields were found to be similar to that in ternary fission. Although their energies were not strongly correlated their angles of emission were found to be well correlated (Kataria *et al* 1973). Trajectory calculations were done using different initial conditions and the one which assumed that the two LCP are emitted in the neck region statistically independent of each other at different times near scission gave a better fit to the experimental data.

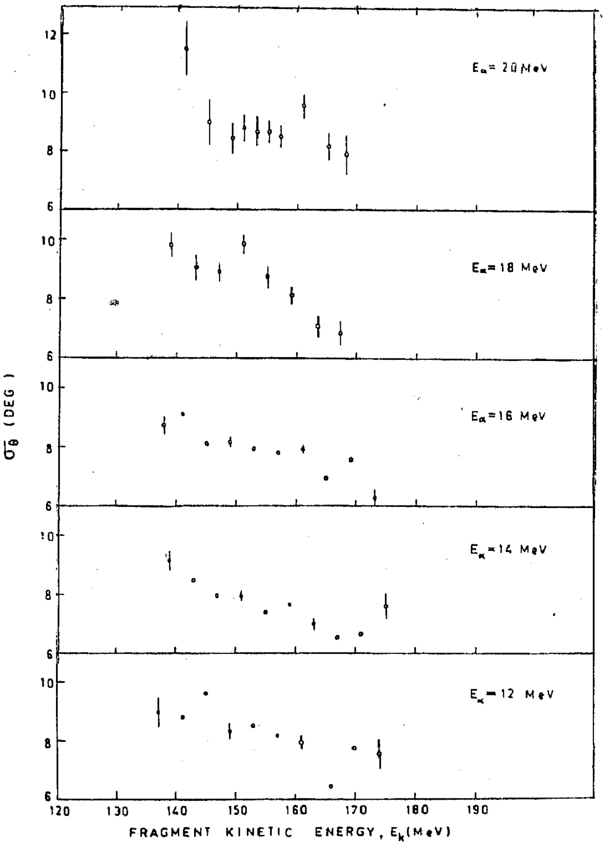

Figure 5. Dependence of $\sigma_{\theta_{\alpha L}}$ of the angular correlation function on the total fragment kinetic energy for different windows of LRA energy (Choudhury *et al* 1980).

3. Studies of LRA emission in fast neutron induced fission

3.1 *Emission probability of* LRA *and* LCP *in fission*

As the mechanism of LCP emission in fission is not well understood and very little was known about the factors which decisively influence the emission probability P_{LCP} one of the aspects of LCP emission investigated in detail was the dependence of P_{LCP} on the excitation energy of the fissioning nucleus, in $^{235}\mathrm{U}\,(n,f)$. In this case the region of incident neutron energy of particular interest is thermal to about 5 MeV, because at much higher energies contributions from second and higher chance fissions complicate the interpretation of the results. The results of measurements at thermal, 2,3 and 4 MeV neutrons showed no appreciable change in the P_α with neutron energy E_n implying that if P_α is dependent on the scission configuration or excitation energy at scission, these latter quantities do not change significantly with the initial excitation energy of the fissioning nucleus (Nadkarni and Kapoor 1970). In a later work the yield of different LCP, their energy spectra were measured in $^{235}\mathrm{U}(n,f)$ for different values of E_n using a ΔE–E detector telescope to identify the LCP (Sharma *et al* 1981) using the 2 MV Van de Graaff machine at IIT Kanpur. In this study increase $\sim$ 20 % in P_α for E_n $\sim$ 200 keV was observed compared to that in thermal neutron fission in good agreement with an earlier measurement (Krishnarajulu *et al* 1977). However, the yield of tritons increased with E_n such that at E_n $\sim$ 550 keV, it was about 3 times higher than the thermal neutron value. A still higher increase in the yield of protons was observed. These observations of dependence of P_{LCP} with E_n have not yet found satisfactory explanation and need further investigations. Measurements were also carried out to study the dependence of the yield of polar and equatorial protons and alpha particles on incident neutron energy ($E_n \sim 600$ keV) where a similarity in the dependence of emission probability on the excitation energy of the fissioning nucleus was observed both for the polar and equatorial LCP (Sinha *et al* 1980, 1982, Sharma *et al* 1981).

3.2 *Angular distribution of LRA and ternary fission fragments with respect to the incident neutron direction*

The angular distribution of LRA and ternary fission fragments with respect to the incident neutrons was first measured by Ramanna *et al* in 14 MeV neutron fission of ^{238}U. They observed a forward-backward peaked angular distribution of LRA and a 90°-peaked distribution of ternary fission fragments. These results suggested that the LRA emission is analogous to the evaporation of particles from a deformed and excited fissioning nucleus prior to scission. The observed results also implied that LRA emission is dependent on the *K*-quantum number, LRA emission being less probable for $K = 0$ than for the case where K is large. In further investigations the anisotropy of the angular distribution of LRA with respect to the incident beam was measured in 3MeV neutron-induced fission of ^{235}U (Hattangadi *et al* 1965) where the anisotropy was found to be in agreement with the value calculated on the basis of the statistical theory of evaporated particles. The observed LRA anisotropy was compared with the anisotropy of the ternary fission fragments measured in 3 MeV neutron fission of ^{235}U and it was found that the magnitude of the observed ternary fragment anisotropy is equal to that expected on the basis of the fact that LRA are emitted prior to scission at right angles to the fission fragments (Nadkarni 1968). The question of *K*-dependence of the LCP emission probability is also of current interest in the studies of LCP emission in heavy ion-induced reactions.

4. Study of prompt radiation emitted in LCP accompanied fission

Emission of K x-rays in LCP accompanied fission was investigated for the first time at Trombay where the yields of K x-rays emitted by fission fragments in LCP accompanied fission were compared with that in binary fission in the case of spontaneous fission of ^{252}Cf. The observed K x-ray yields in these two cases did not seem to favour the assumption of exclusive emission of LRA from either of the two fragment groups (Kapoor *et al* 1968). If the emission of LRA takes place after scission at the expense of nucleons only from some fragments the isotopic yield distribution of the fragment of a given charge would vary in a definite manner as compared to that in binary fission. With this in view the isotopic yield distribution for fragments of specified nuclear charge in LRA fission relative to that in binary fission was measured in spontaneous fission of ^{252}Cf using the method of discrete γ-ray lines emitted from fragments. A number of γ-ray lines were assigned to specific fission fragments and the ratios of intensities of these γ-ray lines in LCP fission and binary fission were determined. Taking $2^+ \rightarrow 0^+$ transition intensities of the even-even nuclei to be a measure of their yields, the yields of several individual fission fragments of known mass and charge in LCP accompanied fission relative to that in binary fission were obtained (Ajitanand *et al* 1975). Figure 6

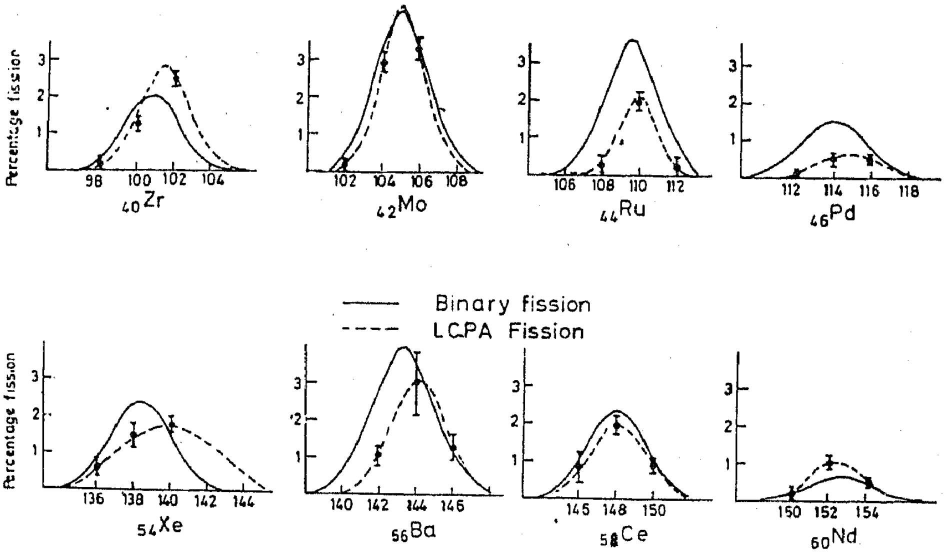

Figure 6. Isotopic mass distributions for different fission fragment charges in LRA accompanied fission compared to those in binary fission (Ajitanand *et al* 1975).

shows the isotopic yield distributions for different fragment charges ($Z = 40$–60) in LCP accompanied fission derived from this experiment and these are compared with those in binary fission. The isotopic mass distributions were observed to be altered in the light and heavy fragment groups almost to the same extent. If LCP were emitted exclusively from either of the fragment groups (light or heavy), substantially different shifts in the isotopic yield distribution would be expected. Thus these results also supported the view that LRA are emitted at the expense of nucleons from both the light and heavy fragment groups or from the fissioning nucleus as a whole.

To sum up, the investigations of LCP emission in fission briefly reviewed above, have provided information on the various facets pertaining to the mechanism of emission of these particles and have also proved to be useful in exploring the dynamics of the last stages of the fission process.

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