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Yields and energy spectra of light charged particles emitted in neutron induced fission of ^{235}U

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Abstract. The yields and energy spectra of light charged particles emitted in the fission of ^{235}U have been measured in the neutron energy range of 100 keV to 1 MeV. The yield of long range alpha particles is found to increase around 200 keV neutron energy compared to thermal fission. A low energy component observed in the energy spectrum was assigned to the tritons emitted in fission. The yield of this triton component is seen to have a marked increase around 500 keV. These results indicate that LCP yield is influenced by the transition state level characteristics.

Keywords. Light charged particles; ^{236}U fission; yields and energy spectra.

1. Introduction

It is well known that once in about 500 fission events a light charged particle (LCP) is emitted along with the two heavy fragments. Long range alpha particles (LRA), having an average energy of about 15 MeV, constitute about 90% of these LCP and the remainder are mostly tritons having an average energy of about 8 MeV. From the sharp 90° peaked angular distribution of these particles with respect to the fragments, it is inferred that they are emitted close to the scission point in the region between the fragments. However, the mechanism governing their emission is still an open question. Very little is known about the factors which decisively influence the LCP emission probability (P_{LCP}). To understand the LCP emission mechanism it is desirable to study the dependence of P_{LCP} on the fissionability, excitation energy, angular momentum and spin of the fissioning nucleus. It has been observed (Nobles 1962) that P_{LCP} depends weakly on the fissionability parameter and is higher for spontaneous fission than for induced fission. Some investigations on P_{LCP} have been carried out for fissions induced by neutrons in the resonance energy region (Deruytter *et al* 1965, Melkonian and Mehta 1965, Schröder *et al* 1965, Wagemans and Deruytter 1972), by fast neutrons (Drapchinski *et al* 1964) and in charged particle induced fission at higher energies (Thomas and Whetstone 1966, Loveland *et al* 1967). Discrepancies exist in several investigations at much higher incident charged particle energies on the dependence of P_{LCP} on incident energy. It should be noted that at higher energies, a substantial fraction of fissions take place through second and third chance fissions and the observed P_{LCP} corresponds to an average over several nuclear species fissioning at different excitation energies, leading to uncertainties in the excitation energy dependence of P_{LCP}. However, if P_{LCP} is measured at those excitation energies where only first chance fissions contribute then a cleaner picture of the excitation energy dependence of P_{LCP} can emerge. In the case of neutron induced fission of ^{235}U this energy range is between thermal and about 5 MeV. Investigations on LRA emission probability (P_{LRA}) in neutron induced fission of ^{235}U up to 4 MeV (Nadkarni and Kapoor 1970) at intervals of 1 MeV indicated that P_{LRA} was equal to the thermal neutron value within the statistical error of 10%. Subsequent measurements (Nadkarni *et al* 1975) with neutrons of energies 0·75, 1·25, 1·5, and 1·75 MeV, showed that P_{LRA} did not vary with neutron energy. However, the spectra indicated the presence of a low energy component (8–12 MeV) whose yield varied with neutron energy.

In the present work we have carried out measurements on the yields and energy spectra of LCP in the fission of ^{235}U induced by neutrons of energies 120, 180, 500, 800 and 1020 keV. This energy range is of particular interest in the investigation of the influence of the transition (saddle point) states on the LCP emission probability. In this energy range some structures which are believed to be associated with the transition states have been observed in fragment anisotropy (Nesterov *et al* 1967, Nadkarni 1969), average fragment kinetic energy (Blyumkina *et al* 1968, Nadkarni 1969, Boldeman *et al* 1976, Meadows and Whalen 1967) and fragment mass distributions (Cuninghame *et al* 1961, 1964; Mehta *et al* 1967) in binary fission. In thermal neutron fission (s-wave) of ^{235}U ($7/2^-$) the states 3^- and 4^- are accessible to the fissioning nucleus. Based on fission characteristics, it has been possible to separate the resonances into two groups (Cowan *et al* 1966, 1970; Melkonian and Mehta 1965; Ryabov *et al* 1972) giving evidence that fission characteristics depend on the spin of the transition state. At neutron energies between 100 keV and 1 MeV the *p*-wave contribution to the fission cross section is significant and the states 2^+, 3^+, 4^+ and 5^+ are accessible. At still higher energies higher partial waves also contribute to the fission cross section and states of both parities are accessible.

The aim of this work is to ascertain whether structures in the yield and average energy of LCP are present on account of the characteristics of the transition states accessible at the saddle point, in particular due to the change in parity of the available quantum states.

2. Experimental method and data analysis

A schematic diagram of the experimental set up and the electronics used is shown in figure 1. A ^{235}U source (93·9% enrichment) of thickness ~ 5 mg/cm² and area ~ 4 cm² formed the cathode of an ionization chamber. A 7 mg/cm² thick aluminium foil placed at a separation of 2 mm served as the collector. The ion chamber was filled with high purity argon gas to a pressure of 1·1 atmospheres. The natural alpha particles and fission fragments were stopped by the aluminium foil, allowing only the LCP to pass through and be detected by a semiconductor

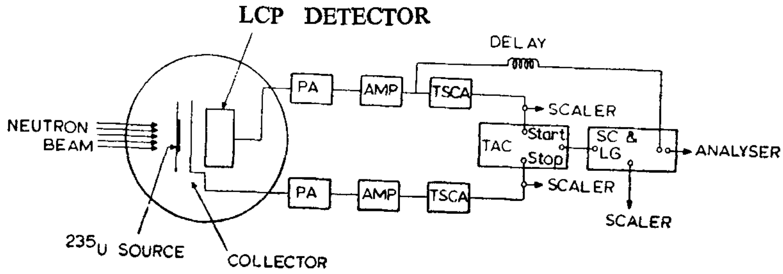

Figure 1. Block diagram of the electronic set-up.

detector (depletion depth of 400 μ at 170 V) mounted close to the collector foil. Very small activities of ^{241}Am and ^{237}Np sources deposited on the side of the Al foil facing the detector enabled an on-line energy calibration of the LCP detector. Neutrons were produced by ^{7}Li(p, n)^{7}Be and T(p, n)^{3}He reactions using the 2 MeV van de Graaff Accelerator at IIT, Kanpur. The lithium targets were prepared by vacuum evaporation of lithium on copper backing and the tritium targets were obtained from the Isotope Division of BARC. The neutron energy spread for 120 keV and 180 keV neutrons was estimated to be about 40 keV and for 500, 800 and 1020 keV neutrons it was about 160 keV. Thermal neutrons were produced by interposing a 5 cm thick paraffin block between the neutron source and the fission chamber. The average fission rate was $\sim 2\cdot 5 \times 10^{3}$ per sec and the neutron flux at the fissile target was estimated to be about 5×10^{7} n/cm^{2}/sec.

The pulses from the fragment and LCP detectors were amplified and fed to timing single channel analysers to get the timing signals. The timing signals were fed to a time to amplitude converter (TAC) set to achieve a timing resolution (2τ) of $0\cdot 5\,\mu$sec. The LCP detector pulses gated by the TAC output were recorded on a 400 channel analyser. The fission and alpha particle count rates were simultaneously monitored. After each fast coincidence run, an ungated spectrum of the alpha particles was recorded for a fixed time, which was used to determine the chance coincidence rates. Each fast run was preceded and followed by a thermal neutron run. The detector performance was monitored frequently and the detectors were replaced if any deterioration due to radiation damage was noticed.

The observed LCP energy spectra were corrected for chance coincidences by using the ungated LCP spectra recorded for a known duration of time. For the purpose of correcting the energy spectra for energy loss of alpha particles in the source, gas and aluminium foil, the average thickness encountered in the source, gas and the aluminium foil, was estimated by comparing the observed average LRA energy in the thermal neutron run with the known value of 15 MeV in thermal neutron fission (Nadkarni and Kapoor 1970). The energy loss corrections for all energies were estimated using energy loss tables.

3. Results and discussion

Figure 2 shows the corrected LCP energy spectra in thermal and fast neutron runs. The spectra show more low energy particles than that given by a Gaussian.

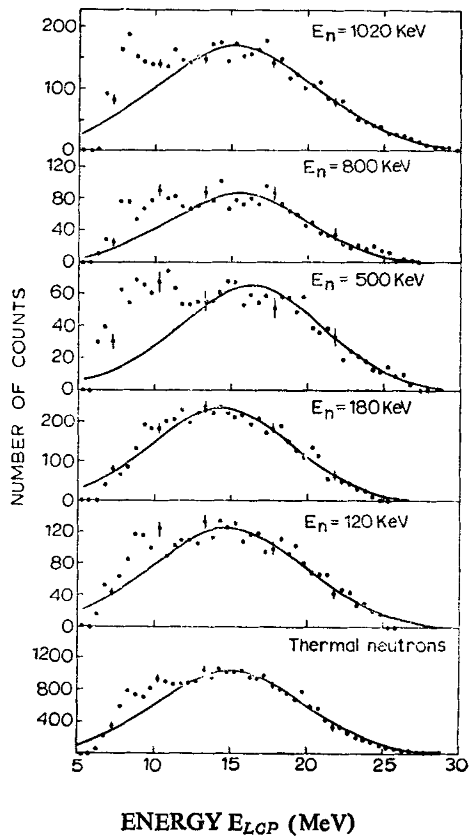

Figure 2. The LCP energy spectra for various neutron energies.

The lower limit of the Gaussian fitting function was fixed at 12 MeV since the yield in the region beyond 12 MeV is predominantly due to alpha particles. The least squares Gaussian fits to the spectra are also shown in the figures as continuous curves.

From the Gaussian fits to the spectra, the most probable value and standard deviation of LRA energy spectra, $\bar{E}_\alpha$ and σ_{E_α} respectively, were obtained as a function of incident neutron energy and these are shown in figures 3 *a* and 3 *b*. It can be seen that there is no significant variation in the average LRA energy ($\bar{E}_\alpha$) in the range of incident neutron energies studied. However, some structure of the order of 0·6 MeV is seen to be present in $\bar{E}_\alpha$ in the incident neutron energy range of 200 to 700 keV. Since there is no indication of a systematic trend with neutron energy, no conclusions are drawn. The width σ_{E_α} does not appear to be sensitively dependent on incident neutron energy.

The results on the LCP yield above 6·5 MeV are shown in figure 4 *a*. A marked increase (20%) in LCP yield compared to the yield at thermal neutron energies is evident in the neutron energy range of 100–500 keV. At higher incident neutron energies the yield tends to become the same as that for thermal neutron fission. Since this yield has contributions from both LRA particles and tritons, we have attempted to determine their individual yields assuming that above 12 MeV the contribution to LCP yield is mostly due to LRA.

The observed LRA yield for $E_{LCP} > 12$ MeV in fast neutron fission relative to that in thermal neutron fission is shown in figure 4 *b*. There is a noticeable increase

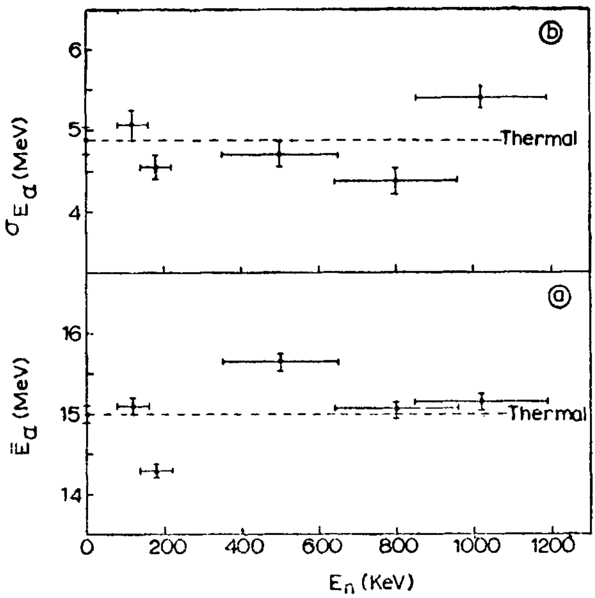

Figure 3. Variation of (*a*) the average alpha energy ($\bar{E}_\alpha$) and (*b*) standard deviation (σ_{E_α}) with neutron energy.

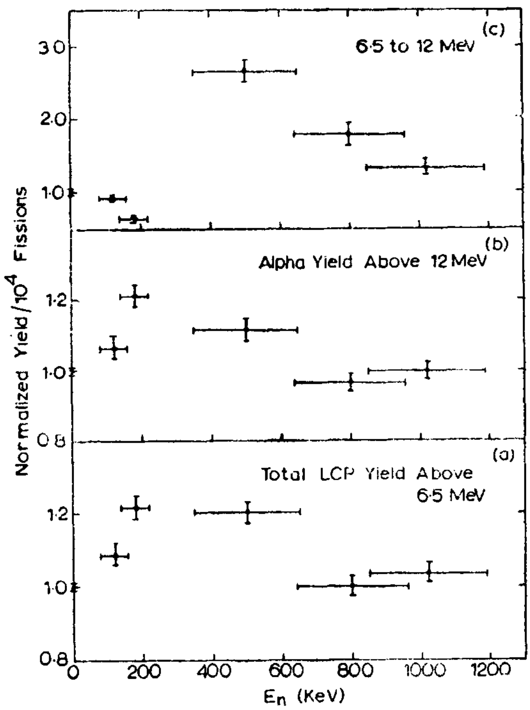

Figure 4. Variation of (*a*) total LCP yield above 6·5 MeV (*b*) alpha yield above 12 MeV and (*c*) LCP yield between 6·5 and 12 MeV.

of about 20% in the neutron energy range of 200–500 keV, over the thermal neutron value. At higher neutron energies, the LRA yield is the same as that for thermal neutron fission as can be seen from the results of the present work and those by Nadkarni *et al* (1975) where measurements were made up to $E_n = 1 \cdot 75$ MeV.

The yield in the energy region $6 \cdot 5 < E_{LCP} < 12$ MeV has contributions from both LRA and tritons. To estimate the yield of tritons alone, the low energy tails of the Gaussian fits to the LRA spectra have been subtracted from the observed yields to give the 'triton' yield. Yields of the triton component determined in this way for different neutron energies (figure 4 *c*) show a significant increase (2–2·5 times) at $E_n \sim 500$ keV. Fluss *et al* (1972) have measured the yields of charged particles in fast neutron fission of ^{235}U by radio chemical and particle identifier methods. They have also observed a significant (2 to 3 times) yield of tritions in fast fission as compared to thermal fission.

The increase in LRA yield in the neutron energy region of 200 to 500 keV as compared to thermal neutron fission, cannot be attributed to any excitation energy dependence of LRA yield, since the change in excitation energy is very small and also the yield does not vary monotonically with neutron energy. Thus the increase in LRA yield around $E_n \sim 200$ keV may be associated with the increase in the relative number of fissions proceeding *via* the even parity states populated by the *p*-wave interaction at these energies. Since the positive parity levels lie lower ($\sim 0 \cdot 6$ MeV) than the negative parity levels, the extra energy available at the saddle point for *p*-wave fission can go into the degrees of freedom such as deformation, kinetic energy of relative motion, fragment excitation or into LCP emission. In binary fission it has been observed (Blyumkina *et al* 1964, Nadkarni 1969, Boldeman *et al* 1976, Meadows and Whalen 1967) that there is a drop in the average kinetic energy of fission fragments and an increase in prompt neutron yield at these neutron energies as compared to thermal neutron fission. The higher LRA yield in this neutron energy region observed in the present work indicates that the extra energy may also be available for LRA emission. At higher energies higher *l* waves also contribute to the fission cross section and this may result in the averaging out of the channel effects resulting in an LRA yield equal to the thermal neutron value.

From figure 4 *c* it is seen that the yield of 'tritons' in the neutron energy range of 200–500 keV is about twice that in thermal neutron fission. It is seen that for neutron energies of 120 keV and 180 keV, the extracted triton yield is the same as the thermal neutron value but is significantly higher around $E_n = 500$ keV. The yield decreases for higher energies but is always higher than the thermal neutron value. The variation of this yield with neutron energy is different from that of LRA yield and may throw further light on the mechanism of LCP emission at scission. However, to draw quantitative conclusions further studies, using a $\triangle E$–E set up for particle identification are necessary.

In conclusion, the results on the LRA yield and energy spectra in fast neutron fission for neutron energies up to about 1 MeV indicate that the LRA yield appears to be influenced by transition state level characteristics. The yield of the extra low energy component, ascribed to tritons, also shows significant variation with incident neutron energy in the energy region investigated.

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