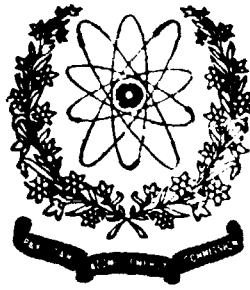


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**GAMMA RAY TRANSITIONS IN DE-EXCITATION OF
 ^{252}Cf SPONTANEOUS FISSION FRAGMENTS**

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**Pakistan Institute of Nuclear Science and Technology
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Abstract

Gamma rays in the range from 60 keV to 730 keV have been observed following the spontaneous fission of ^{252}Cf , with high resolution Ge(Li) detector, full width at half maximum (FWHM) of 700 eV at 122 keV, in coincidence with the two fission fragments observed with surface barrier detectors. A total number of 18,636,549 events were recorded over a run period of about 150 hours stretching over three weeks. The events were sorted to generate γ ray spectra belonging to 2 amu intervals of the fragment masses and 6 MeV intervals of the total kinetic energy released. Some of the prominent γ lines belonging to various masses of the fission fragments have been identified. For some γ lines, the intensities have been evaluated as a functions of the total kinetic energy of the fission fragments.

1. Introduction

The process of fission, spontaneous or induced, provides the means to produce neutron rich nuclei in highly excited states which decay by prompt and delayed γ rays, beta and neutron emissions. The bulk excitation energy of the excited fission fragments is released in the form of prompt γ rays which are emitted within 10^{-10} seconds after fission. The emission of prompt γ rays is believed to occur in the final de-excitation of the fission fragments following evaporation of the prompt neutrons. The overall spectrum of the γ rays is rather featureless, extending up to 10 MeV¹⁾. The combined application of high resolution Ge(Li) detectors for γ ray measurements and the semiconductor and time of flight techniques for fission fragment measurements have made possible the systematic study of the vast spectra of γ rays emitted during the fission process. Coincidence techniques enable the measurement of various products in correlation to each other. The literally numberless states available to the excited systems and the complex nature of the decay schemes of excited fission fragments provide valuable information on the fission process during scission and the decay scheme of neutron rich nuclei produced during fission, which are usually not accessible in any other way. Major studies on these lines have been made on ²⁵²Cf spontaneous decay mode. Many authors²⁻⁷⁾ have performed coincidence measurements between fission fragments, internal conversion electrons, x-rays, prompt γ rays, delayed γ rays etc. and decay schemes of many neutron rich nuclei have been derived.

In this report we present results of the γ rays observed in coincidence with the fission fragments. The γ rays were observed in the

range of 60 keV to 730 keV. A total of 18,636,549 events were recorded and analysed off-line on a computer to obtain the γ ray spectra. The dependence of intensities of γ ray on total kinetic energy release was determined for some known γ rays.

2. Experimental Procedure

The geometry of the experiment is shown in Fig. 1. A ^{252}Cf source on nickel backing was mounted on the fission fragment surface barrier detector F1. The second fission fragment surface barrier detector F2 was placed 8 mm away from F1 and the prompt γ ray detector was located 25 mm from F1. The γ ray detector was of Ge(Li) type cooled at liquid nitrogen temperature. It had a resolution of 700 eV at 122 keV. The complimentary fission fragments were detected in coincidence in detectors F1 and F2.

The criteria for the acceptance of detectors was that the resolution for fission fragments (peak to valley) be between 1.7 and 2.6. The γ rays emitted from the fission fragment were detected in coincidence with the coincidence of F1 and F2 detectors, i.e. every event satisfying the triple coincidence of detectors F1, F2 and γ ray was detected and recorded. The block diagram of the electronics is shown in Figure 2.

The three ADC's were coincident gated by the logic condition that there must have been a fast coincidence ($\tau = 10$ n sec) between the two surface barrier detectors (F1,F2) and this fast coincidence output must have been in fast coincidence with the γ rays detector. The pulse height from the three detectors for each triple coincident

event were recorded event by event on a magnetic tape. The fast discriminators were set just above the noise level. The γ ray spectrum was calibrated with ^{60}Co and ^{137}Cs sources. The total running time was about 150 hours with a coincidence rate of about 26 sec^{-1} . The gains of the amplifiers were checked several times daily. If any substantial gain drift occurred, the run was restarted after necessary adjustment. The surface barrier detectors were replaced when their resolution decreased due to radiation damage.

The data stored on magnetic tape was processed off-line on the IBM 360/44 computer of Quaid-e-Azam University. A total number of 18,636,549 counts in triple coincidence between F1, F2 and γ were recorded.

3. Analysis

The fission fragment masses were calculated for each event from the measured pulse height from fission fragment signals, using the Schmitt calibration method. The pulse heights were converted to energies by the equations given by Schmitt⁸⁻⁹⁾, which correct for mass dependent pulse height defect. The analysis was carried out off-line event by event. The fragment energies were converted into total kinetic energy release and the masses of the fragments. A summary of the equations used is given in the appendix A. The total gamma-ray spectrum is quite complex. The events were sorted according to mass and total kinetic energy release. The spectra were tabulated in mass windows of 2 amu interval of fragment masses and 6 MeV intervals of total kinetic energy release. Because only small fast storage (64K) was available, external memory was created on disc storage data module. All the gamma-ray spectra generated have been written on a magnetic tape and are thus

directly accessible for any further evaluation. This sorting enables each gamma-ray to be analysed in terms of mass window of 2 amu and total kinetic energy window of 6 MeV. Some of the prominent lines were identified with respect to the masses of the excited fission fragments emitting these gamma-rays.

The gamma-ray spectra were analysed with the help of a computer code **GSFIT** developed by Haase¹⁰⁾ and adapted for the use on PDP 11/45 by Ikram, Saleem and Rashid¹¹⁾. This code fits and searches the peaks. The search is made by calculating the centroids and intensities of the peaks. The calculations are done by evaluating their standard deviations whose approximate channel position are given as input and whose probabilities are known through their dependence on energy. This programme assumes the background to be linear within the peaks. For energy pulse height conversion and for evaluation of peak parameters the programme uses the peak positions and line shapes of calibration lines which fall within the region of analysis. The speed of the energy calculation of the peak depends on how close the input trial values are to the final results. The closer the trial values are, the faster is the calculation. The evaluation of the gamma-ray is done by iterating peak position and the background until the best fit is obtained. For peaks having bad statistics it has been observed that the peak position is effected to some extent and peak area to a considerable extent. The peak positions were within a standard deviation but the peak area and thus the intensities differed by as much as four standard deviation for the weak lines.

The gamma-lines used for energy calibration upto 212.4 keV along with fitted values by the programme **GSFIT** are given in table I. The energies and intensities of gamma-rays from the fission fragments of

mass number 100 ± 1 and total kinetic energy from 0-300 MeV are obtained by this fit procedure in which known lines have been used as calibration lines are listed in table II.

4. Results.

The gamma-ray sorted spectrum for mass number 100 ± 1 and total kinetic energy window of 0-300 MeV is shown in Fig. 3. The gamma-lines of this spectrum are listed in table III alongwith available values from other references. There are a number of lines belonging to mass region 100 ± 1 which have not been listed by other authors and have yet to be assigned. Assignment of charge Z, to the γ -transition i.e. identification of the nucleus from which the gamma-rays originate, has been made by comparison with the results of references 1,2,4,5 and 6. The lines thus identified, are listed in table IV alongwith the charge and mass number and the internal conversion coefficient. These gamma-lines are among the most prominent which occur in the fission of ^{252}Cf . The relative yield of some of these lines, and thus of the nucleus from which they originate, were evaluated as a function of the total kinetic energy release in fission. The yield distributions of ^{100}Zr (212.7 keV), ^{102}Zr (151.9 keV), ^{104}Mo (192.3 keV), ^{106}Mo (171.7 keV), ^{108}Ru (242.8 keV), ^{110}Ru (240.8 keV) and ^{112}Ru (242.8 keV) are presented in figure 4. To every fragment we observe in fission, we can associate a mass division of the fissioning ^{252}Cf nucleus. This association is determined by the constraints.

$$(Z_1 + Z_2) = 98$$

$$(A_1 + A_2) = 252 \quad (\text{for pre neutron situation})$$

To every mass division is associated a certain amount of total kinetic energy, which can range from 150 to 250 MeV. The yield curves in figure 4 give us the probability of a particular mass division as a function of the total kinetic energy. The most probable value for the kinetic energy is at the maximum yield. These yield distributions are essentially gaussian. They show that the single fragments possess sizeable variation in kinetic energies from event to event. These variations are a consequence of the distribution of nuclear elongations at scission point. The relative variations of this value have been evaluated and are compared with the theoretical estimates. These experimental, $\Delta E(\text{exp})$ and theoretical, $\Delta E(\text{th})$ variations in kinetic energy release are also listed in Table V. These variations have been normalized to ^{100}Zr . The experimental variations have been evaluated from the yield curves in figure 4. The theoretical variation have been estimated in the touching sphere model. In the touching sphere model one assumes that the kinetic energy release is a result of the Coulomb repulsion of two spheres in contact.

$$E_c = \frac{Z_1 Z_2 e^2}{(R_1 + R_2)}$$
$$= \frac{Z_1 Z_2 e^2}{r_0 (A_1^{1/3} + A_2^{1/3})}$$

where Z_1, A_1 and Z_2, A_2 are the charge and mass numbers of the two touching nuclei. The quantity r_0 is determined from the relation

$$E_{\text{kin}} = \frac{40.58 e^2}{r_0 (100^{1/3} + 152^{1/3})}$$

where E_{kin} here is the experimentally determined kinetic energy for the mass division associated with the production of ^{100}Zr .

This in effect is what is meant by normalization of variation with respect to ^{100}Zr nucleus. We observe that the experimental and theoretical values for the variation are consistent, indicating that the touching sphere model is a good enough approximation to explain the variation in kinetic energy from nucleus to nucleus. This consistency is to be expected since the value of the kinetic energy release at maximum yield is relatively insensitive to the internal excitations of the fission fragments.

Acknowledgment

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Appendix A

Pulse-height to energy and mass conversion

The conversion of pulse height to mass and kinetic energy was carried out by an iterative procedure which is based on the Schmitt's calibration formula. Let X_1 and X_2 be a given pair of pulse heights corresponding to a pair of fission fragments. The Schmitt's calibration formula relates pulse height X to energy E by a linear relation of the form

$$E \simeq M X + b \quad (1)$$

where

$$M = 24.40/P_L - P_H \text{ and } b = 3.3 + 0.00860 \times M$$

P_L is the pulse-height which corresponds to the midpoint between the 3/4 maximum points in the light fragment peak. P_H is the pulse height which corresponds to the midpoint between the 3/4 maximum points in the heavy fragment peak. P_L and P_H were determined from the double coincidence-stabilization fission fragment distributions and set equal to the values of 103.77 MeV and 79.37 MeV respectively. Since momentum is conserved, we have

$$E_{1p}/E_{2p} = M_{2p}/M_{1p} \quad (2)$$

The subscript p denotes pre-neutron emission quantities. Conservation of mass yields the relation

$$M_{1p} + M_{2p} = 252 \quad (3)$$

Using E_1 & E_2 , the post neutron emission kinetic energies as estimates for E_{1p} and E_{2p} , we have

$$M_{1p} = 252 / (1 - E_1/E_2) \quad (4a)$$

$$M_{2p} = 252 - M_{1p} \quad (4b)$$

The total kinetic energy is estimated from

$$E_T = E_{1p} + E_{2p} = E_1 + E_2 \quad (5)$$

To a first approximation

$$M_1 = M_{1p} - \nu_1 \quad (6a)$$

$$M_2 = M_{2p} - \nu_2 \quad (6b)$$

ν_1 and ν_2 are the average number of neutrons emitted. These values were obtained from the work of Bowman¹⁾.

At this point we use the more precise Schmitt's calibration formula,

$$E = (24.0203 + 0.03574M) \times (P_L - P_H) - (24.0202 + 0.03574M) \times P_L / (P_L - P_H) + 0.1370 \times M + 89.6083 \quad (7)$$

Since neutron emission does not significantly change the fragment velocities, therefore

$$E_{1p} = E_1 (M_{1p} / M_{1p} - \nu_1) \quad (8a)$$

$$E_{2p} = E_2 (M_{2p} / M_{2p} - \nu_2) \quad (8b)$$

Now we return to equation (5) using the values of equations (8a), (8b) and recalculating the post neutron emission masses. The process is repeated until the difference in mass values resulting from two consecutive iterations is less than 0.05%. On the average convergence was reached within four iteration.

Table I

γ -lines used for calibration from sorted spectra
of Mass No 100 \pm 1 with energy window 0-300 MeV

E_{γ} up to 212.4 (KeV)	
Assumed Energy (KeV)	Fitted Energy (KeV)
74.57	74.61
98.70	98.56
119.27	119.68
126.98	126.38
152.10	151.23
172.02	171.73
192.00	192.00
212.37	212.31

Table II

Computer output of γ -ray energies and their Intensities from sorted spectra of Mass No 100 \pm 1 with energy window 0-300 MeV

ENERGY (KEV)	INTENSITY	FWHM
57.27(0.05)	391.(94.)	0.72
62.34(0.12)	509.(240.)	0.74
65.58(0.07)	961.(384.)	0.76
67.77(0.05)	546.(94.)	0.76
72.29(0.05)	1605.(351.)	0.79
74.61(0.04)	2064.(242.)	0.79
79.74(0.09)	877.(339.)	0.81
85.14(0.05)	1234.(169.)	0.83
87.77(0.10)	514.(143.)	0.84
91.42(0.22)	243.(112.)	0.86
95.06(0.13)	552.(191.)	0.87
98.58(0.03)	2725.(313.)	0.88
102.83(0.07)	904.(216.)	0.89
104.50(0.07)	1033.(196.)	0.90
109.86(0.09)	505.(111.)	0.92
114.90(0.01)	533.(9.)	0.93
119.60(0.05)	1156.(156.)	0.95
122.04(0.08)	744.(219.)	0.96
126.56(0.12)	723.(162.)	0.97
131.60(0.09)	541.(242.)	0.99
133.90(0.09)	579.(138.)	0.99
136.27(0.08)	608.(208.)	1.00
137.96(0.38)	333.(383.)	1.01
142.79(0.20)	622.(401.)	1.02
151.53(0.16)	732.(264.)	1.05
158.19(0.11)	572.(240.)	1.06
162.41(0.05)	1093.(216.)	1.08
171.13(0.28)	298.(139.)	1.10
191.65(0.12)	397.(82.)	1.16
203.04(0.21)	283.(88.)	1.19
212.31(0.07)	991.(117.)	1.21

Table III

γ -rays from Sorted Spectra

Mass No. = 100 ± 1

Energy Window = 0-300 MeV

Present E_{γ} (KeV)	Others E_{γ} (KeV)	Mass
55.8	55.0 ^{a)}	Zr
61.7	54.8 ^{a)}	Y
65.0	65.5 ^{a)}	Y
67.2		
72.0		
74.2		
80.2		
85.2		
87.0		
92.2		
95.5	95.1 ^{a)}	Y
98.7	98.2 ^{a)}	$^{101}_{Zr}$
	98.2 ^{a)}	$^{101}_{Y}$
	99.0 ^{c)}	$^{101}_{Z}$
102.8	102.7 ^{a)}	Y
105.7		
110.1	109.2 ^{a)}	Y
115.5		
120.0	119.3 ^{a)}	Y
122.5	122.0 ^{e)}	$99 + 1$ - 0
127.0		

Table III (Continued)

Present E_{γ} (KeV)	Others E_{γ} (KeV)	Mass
127.0		
132.1		
133.9		
136.0		
138.3		
143.1	143.1 ^{a)}	Zr
152.1	151.9 ^{d)}	Zr
158.9	158.7 ^{d)}	Zr
162.9		
171.9	171.0 ^{a)}	Zr
	171.9 ^{d)}	Zr
192.0		
202.8		
212.4	212.7 ^{b)}	^{100}Zr
	212.9 ^{a)}	^{100}Zr
	213.3 ^{c)}	100 ± 0

a. From reference 5

b. From reference 2

c. From reference 1

d. From reference 6

e. From reference 4

Table IV Some gamma lines with Charge No and Mass No

NUCLEUS	CHARGE NO	MASS NO	ENERGY (keV)	TOTAL ICC
Zr	40	100.0	212.7	7.290E-2
	40	102.0	151.9	2.499E-1
Mo	42.0	102.0	296.0	2.572E-2
	42.0	104.0	192.3	1.166E-1
	42.0	106.0	171.7	1.793E-1
Ru	44.0	106.0	269.0	3.912E-2
	44.0	108.0	242.3	5.691E-2
	44.0	110.0	240.8	5.804E-2
	44.0	112.0	236.8	6.071E-2
Pd	46.0	112.0	348.8	1.866E-2
	46.0	114.0	332.9	1.993E-2
	46.0	116.0	340.6	2.119E-2
Cd	48.0	118.0	488	7.277E-3
Xe	54.0	138.0	589.5	5.645E-3
	54.0	140.0	376.8	2.498E-2
Ba	56.00	140.0	602.2	6.071E-3
	56.0	142.0	359.7	2.579E-2
	56.0	144.0	199.4	1.1812E-1
	56.0	146.0	181.0	2.512E-1
Ce	58.0	144.0	397.5	2.086E-2
	58.0	146.0	258.6	7.929E-2
	58.0	148.0	158.7	4.197E-1
	58.0	150.0	97.1	2.311E0

Nucleus	Charge No	Mass No	Energy (keV)	Total ICC*
Nd	60.00	152.0	75.9	6.091E0
	60.00	154.0	72.8	7.189E0
Sm	62.0	156.0	76.0	6.656E0
	62.0	158.0	72.8	7.887E0

*Internal Conversion Coefficient.

TABLE V

Total kinetic energy release at maximum yield. ΔE is the relative charge in TKE with the normalization ΔE (Exp) for $^{100}\text{Zr} = 0$.

ΔE (Th) is the change in TKE as obtained from the expression $E_c = \frac{Z_1 Z_2 e^2}{r_0(A_1^{1/3} + A_2^{1/3})}$ for the mass divisions belonging to the nuclei above, with the normalization ΔE (Th) for $^{100}\text{Zr} = 0$.

Z		E_T (keV)	TKE(MeV)	ΔE (Exp)	ΔE (Th)
40	^{100}Zr	212.7	181	0	0
40	^{102}Zr	151.9	182	1	-0.1
42	^{104}Mo	192.3	185	4	3.4
42	^{106}Mo	171.7	185	4	3.2
44	^{108}Ru	242.8	187	6	6
44	^{110}Ru	240.8	187	6	6
44	^{112}Ru	236.8	187	6	6

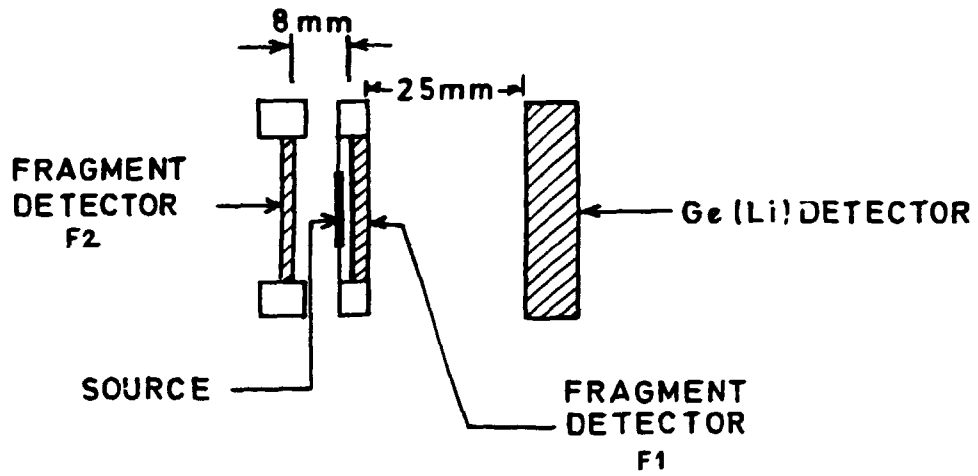
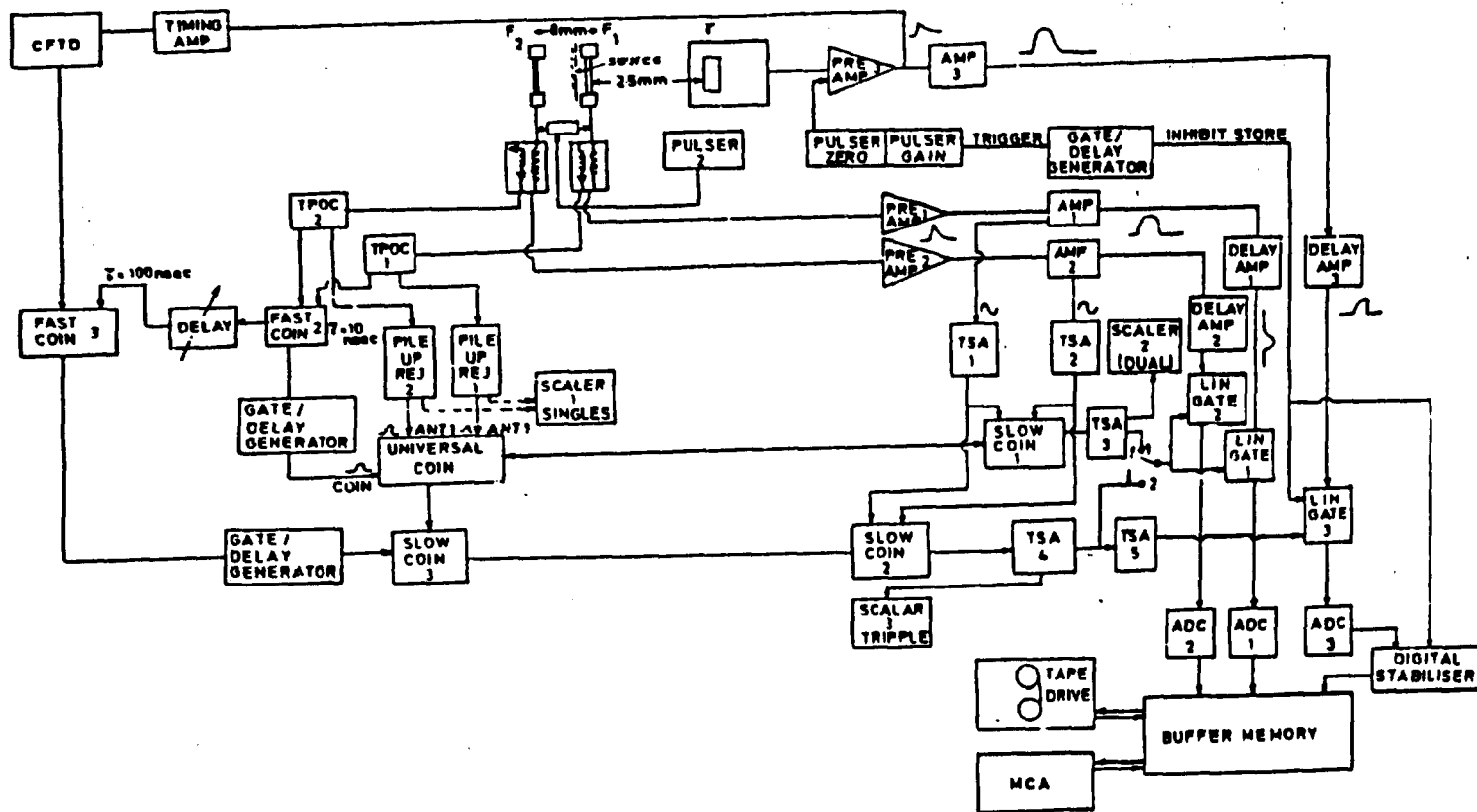


Fig.1. EXPERIMENTAL GEOMETRY OF THE 3-PARAMETER FISSION EXPERIMENT.

Fig.2. ELECTRONICS DIAGRAM FOR (Y_1, F_1, F_2) COINCIDENCE MEASUREMENT



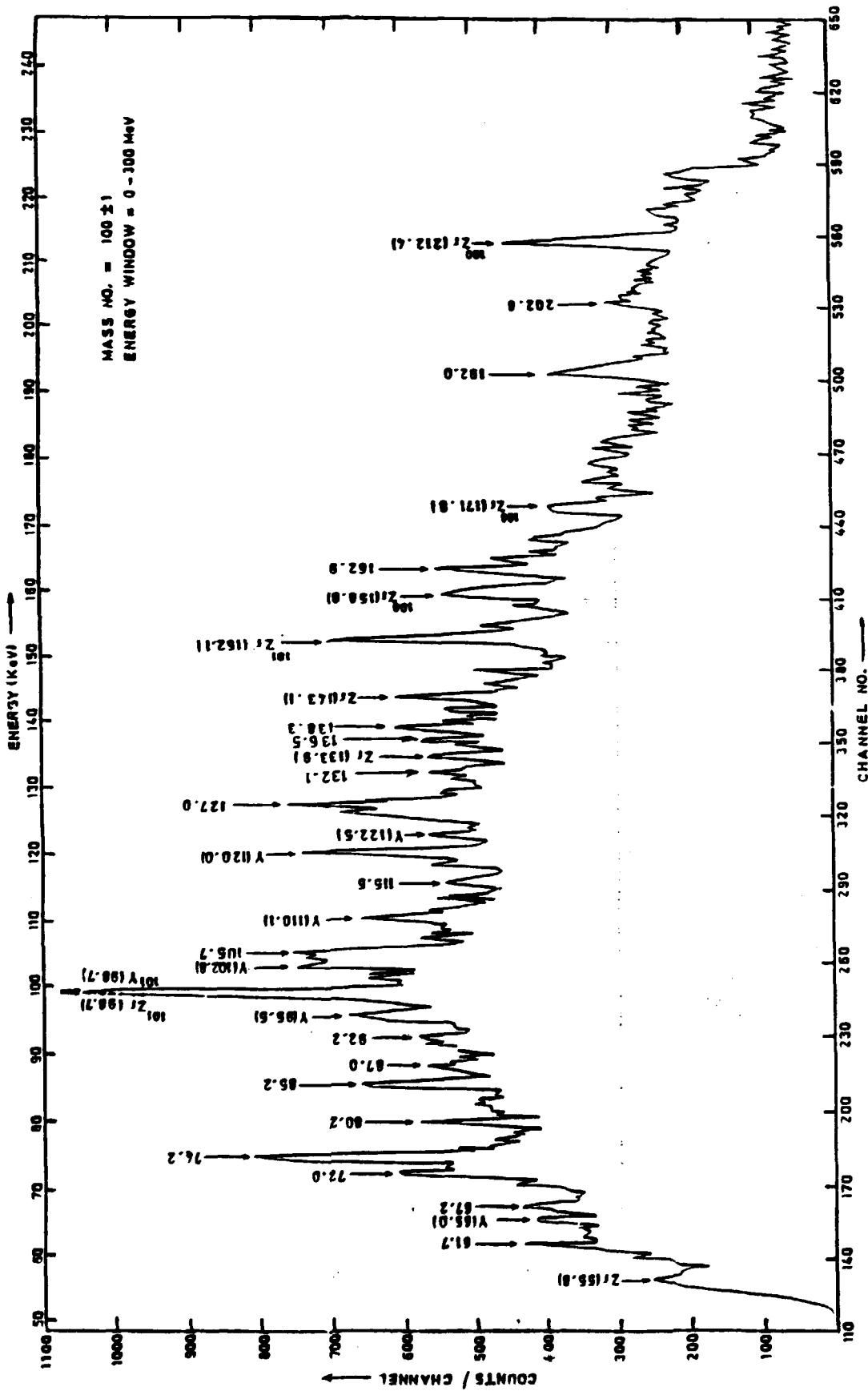


Fig. 3 Gamma ray spectra of Mass No 100 ± 1 with energy window 0-300 MeV

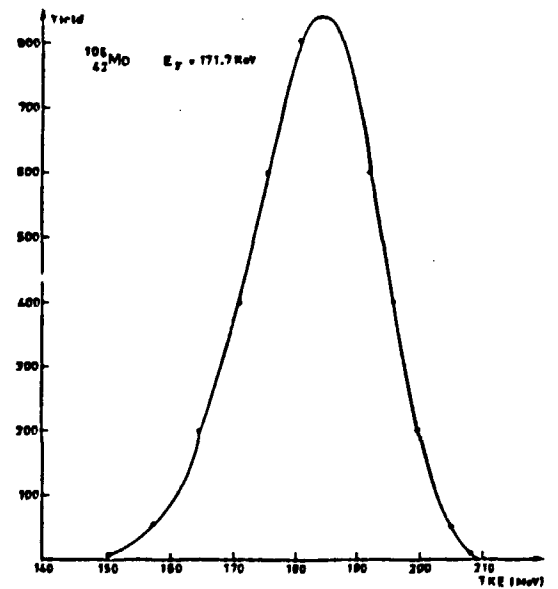
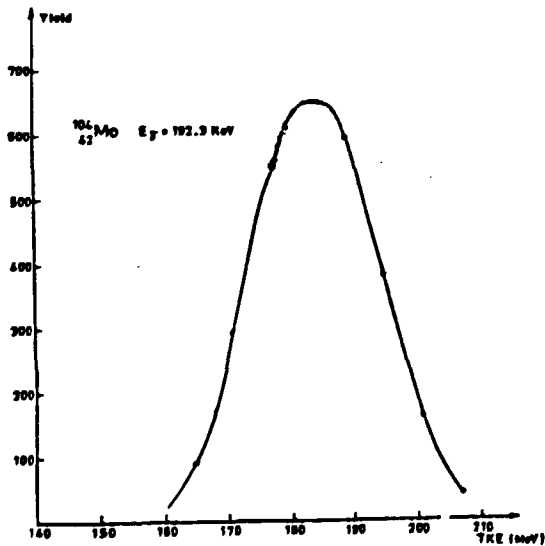
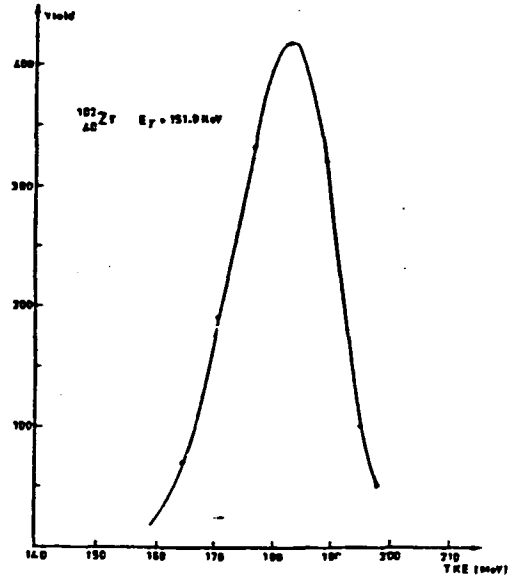
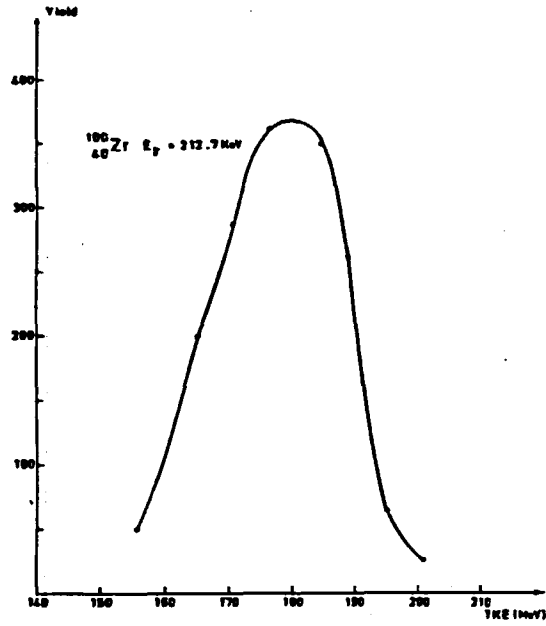


Fig. 4 Variation of yield as a function of total kinetic energy of fission fragments in ^{252}Cf spontaneous fission.

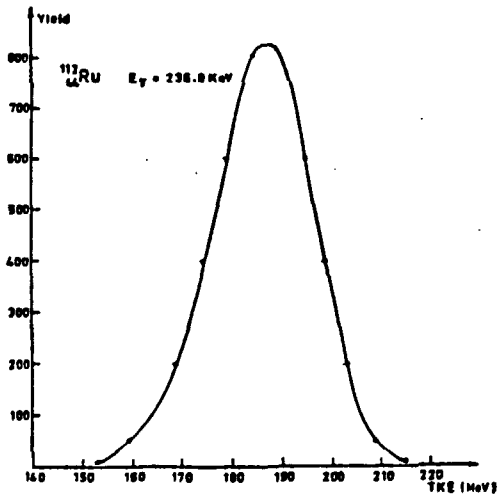
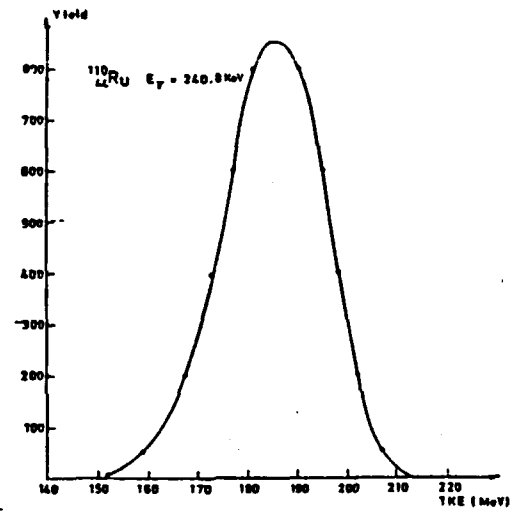
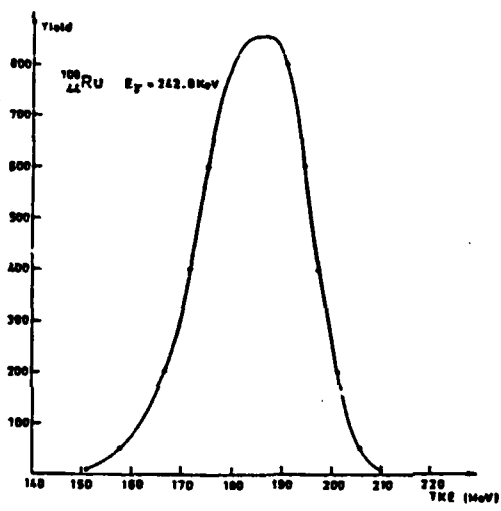


Fig.4 Variation of yield as a function of total kinetic energy of fission fragments in ^{252}Cf spontaneous fission.

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