An absolute measurement of ^{252}Cf prompt fission neutron spectrum at low energy range

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Abstract

Prompt neutron energy spectrum at low energies /25 keV < E_n < 1,2 MeV/ for ^{252}Cf spontaneous fission has been measured with a time-of-flight technique on a 30 cm flight -path. Ionization chamber and lithium-glass were used as fission fragment and neutron detectors, respectively. Lithium glasses of NE-912 /containing ^6Li/ and of NE-913 /containing ^7Li/ 45 mm in diameter and 9.5 mm in thickness have been employed alternatively, for the registration of fission neutrons and gammas. For the correct determination of the multiscattering effects - the main difficulty of the low energy neutron spectrum measurements - a special geometry for the neutron detector was used. A special attention was paid also to the determination of the absolute efficiency of the neutron detector. The real response function of the spectrometer was determined by a Monte-Carlo calculation. The scattering material content of the ionization chamber containing a ^{252}Cf source was minimized.

As a result of this measurement a prompt fission neutron spectrum of Maxwell type with a T = 1.42 MeV parameter was obtained at this low energy range. We did not find any neutron excess or irregularities over the Maxwellian.

1. Introduction

^{252}Cf fission prompt neutron energy spectrum is proposed as a reference standard. Californium sources are widely used for neutron detector calibration, in different neutron scattering and capture experiments, in fission, heavy-ion, reactor physics, defence physics, medical investigations and so a high accuracy of the spectrum in a wide energy range is required. A number of spectrum measurements have been done and being carried on recently [1-5]. One can conclude that ^{252}Cf fission prompt neutron spectrum at the 1 MeV < E_n < 6 MeV energy range can be described by a Maxwellian distribution with T = 1.42 MeV and with a not more then 3-5 % deviation. Outside of this energy range the complex experimental difficulties are arising. At low energies up to 1 MeV there are two different groups of results. In the first one [6-9] essential deviations /up to 30 %/ were found to the Maxwell spectrum, extrapolated from data measured at high energies. On the countrary, in the second group of experimental results, the energy spectra from 10 keV to 6 MeV can be fitted well by a Maxwellian distrubution of T = 1.42 MeV [1-3].

In an attempt to solve this discrepancy we have repeated the low energy californium spectrum measurements paying special attention to get absolute spectrum data and to minimize the uncertainities due to backgrounds of different type and the neutron detector efficiency.

2. Experimental method

The experimental arrangement is shown in Fig. 1. The fragment detector was a fast ionization chamber 38 mm in diameter and 120 mm in length made of 0.1 mm stainless steel filled with 3 atm. of gas mixture /Ar-90 %, CO_2-10 %/. Electrodes were of 0.1 mm stainless steel too. Diameters of electrodes and the distance between them were 25 mm and 1.5 mm respectively. The ^{252}Cf source of 10^4 fissions per second 6 mm in diameter was volatilized onto one of electrodes.

NE-912 lithium glass /45 mm in diameter and 9.5 mm in thickness/ and FEU-30 photomultiplier were used as neutron detector. For measuring the background of delayed gamma rays the NE-912 glass was replaced by NE-913 lithium glass of same dimensions because it is insensitive to neutrons in the studied energy range [10]. The glass was fixed in the centre of a thin-wall aluminium cell mounted at the photocatode of the photomultiplier. Details of the construction and characteristics of the detector can be find in Ref. [11,12]. This arrangement of neutrons and fission detectors essentially reduces the amount of scattering materials in the solid angles of neutron detection.

It is very important because the background due to neutron scattering on these materials cannot be measured directly in the experiment. Distortion of the spectrum due to these neutrons was taken into account by using the response function of the spectrometer.

The background component due to the neutron scattering on materials out of neutron detection solid angle Ω was determined in shadow cone experiments.

The effect of systematic random coincidencies was eliminated by a pile up rejector, which discriminates the double stop signals [13].

General characteristics of the spectrometer are follows: the neutron flight path was 30 cm, while the channel width of the analyser was 0.704 nsec. Amplitude spectra of neutron and fragment detectors, thresholds in the fast and slow channels are shown in Fig.2. The apparatus response functions of the fast channels were measured using an additional scintillation /stylbene/ detector for the fission gamma rays and for measuring γ-γ coincidences from ^{60}Co in the fission fragment and neutron channels, respectively, as shown in Fig. 3. Differential and integral nonlinearities of the spectrometer did not exceed 0.7 and 0.5 %, respectively.

Measurements have been performed in a cycle regime, each of them consists of four series of measurements of 24 hours: with NE-912 and NE-913 scintillators both with and without shadow cone, respectively. The total number of fission events recorded was $N_f = 1.523 \cdot 10^{10}$ for each of the four arrangements.

3. Data treatment

In general case the measured neutron spectrum can be described by a Fredholme's integral equation of the second kind:

$$P(t) = \int F(E,t)\varphi(E)dE$$

where $P(t)$ is the measured spectrum, $\varphi(E)$ is the prompt fission energy spectrum and $F(E,t)$ is the spectrometer response function.

Unfortunately, the function $F(E,t)$ for thick lithium glass cannot be determinated with sifficient accuracy, and some difficulties can take place at solving this equation.

In the present work the data were processed as follows: at the first step the background of random coincidencies was subtracted from the measured spectra. Then using of expression

$$P(t) = P_1(t) - P_2(t) - P_3(t) + P_4(t)$$

where $P(t)$ is the flight time spectrum of fission neutrons, as shown in Fig. 4. $P_1(t)$, $P_2(t)$, $P_3(t)$, and $P_4(t)$ are the flight time spectra from measurements with NE-912, NE-913 scintillators, without and with shadow cone, respectively. At the same time the $P_3(t)$ spectrum has been corrected for some possible shadow cone transmission.

The $P(t)$ distribution has been corrected for the deviation of the real response function of spectrometer from the δ-function:

$$P'(t) = \alpha(t)P(t)$$

The correction factor $\alpha(t)$ was calculated by the following expression

$$\alpha(t)=\left\{\int \delta(t-t(E))\mathcal{E}'(E)\varphi(E)dE\right\}/\left\{2\int F(E,t)\varphi(E)dE\right\} ,$$

where $\varphi(E)$ is a Maxwellian distribution with $T = 1.42$ MeV and $\mathcal{E}'(E)$ is the theoretical efficiency of neutron detection. $\mathcal{E}'(E)$ was calculated by the following expression:

$$\mathcal{E}'(E)=\int F'(E,t)dt ,$$

where $F'(E,t)$ is the probability of detection of a neutron, at the time t, emitted from the source with energy E, at t=0 into solid angle Ω , when the source and the detector are in vacuum /see Table 1/. $F(E,t)$ is the real response function of the spectrometer determined as

$$F(E,t)=\int F''(E,t)F'(t,t')dt'$$

where $F''(E,t)$ the same as $F'(E,t)$ but when the source and detectors are in the real experimental conditions /see Table 1/. $F'(t,t')$ is the response function of the apparatus. The distribution $F'(t,t')$ was obtained by the following expression:

$$F'(t,t')=\int F_f(t,t'')F_n(t',t'')dt'' ,$$

where $F_f(t,t'')$ and $F_n(t',t'')$ are apparatus response functions of fast channels for fragments and neutrons, respectively /Fig. 3/.

Functions $F'(E,t)$ and $F''(E,t')$ were calculated by Monte -Carlo method, basic input data of which are presented in Table 1. The concentration of ^{6}Li in the lithium glass NE-912 was determined by the data of Nuclear Enterprises Ltd. Catalogue [10]. The ^{7}Li, O, Si concentrations were determined by using data from Ref. [14]. The neutron cross-section data for ^{6}Li, for ^{7}Li, Si, O, N and for Fe were taken from the file ENDF/B-V from file ENDF/B-IV and from Ref. [15] respectively.

It was assumed in the calculations that the neutrons are detected only by $^6Li(n,\alpha)^3T$ reaction. The Monte-Carlo programme BRAND [16] for 200 energies of monoenergetic source of neutrons in the energy range $0 < E_n < 2$ MeV was used. Results of calculations for $F''(E,t')$ functions at neutron energies 0.025, 0.245, 0.445 and 1.005 MeV are shown in Fig. 3, while Fig. 5 shows the correction factor $\alpha(t)$. It can be seen that the difference of the real spectrometer response function from the δ-function is essential. When this correction is ignored, a softening of the measured spectrum and essential spectrum oscillations near the strong resonances of lithium and oxygen of energies 0.242 and 0.442 MeV, respectively, can be observed. For example, the response function broadening on the high time side due to the before-detection-scattering of neutrons at the 0.442 MeV oxygen resonance can lead to a dip of 25 % in the spectrum at this energy /Fig. 5/.

For speeding of the Monte-Carlo calculation of $F''(E,t)$ function /see Table 1/ some simplifications have been used. In particular instead of the gas mixture of the fission chamber air, instead of stainless steel iron were assumed, respectively. The window thickness /80 micron thick aluminium foil/ of the neutron detector has been ignored. Estimated errors caused by these simplifications are within the statistical accuracy of the Monte-Carlo calculation.

4. Neutron detection efficiency

The efficiency of the thick lithium glass detector was measured [11] by time-of-flight method with IPPE pulsed Van de Graaff neutron generator, using the efficiency of 0.835 mm thin NE-908 ^{6}Li glass detector as a reference. For efficiency determination cross section data for $^6Li(n,\alpha)^3T$ were taken from the ENDF/B-V file. To get more precise neutron detection efficiency data a Monte-Carlo calculation were performed for the thin NE-908 glass scintillator, too [16]. The calculation model was the same as for calculation of $F'(E,t)$ with an exception in the zone 10 /see Table 1/, where the zone thickness and the ^6Li, ^7Li, O and Si concentrations were 0.0835 cm, 172.4, 8.0, 480.7 and 182.1 nuclei/cm^3 x 10^{20}[17], respectively. Table 2 shows the real detection efficiency data for the NE-912 glass obtained from the evaluation of the measured data for thick NE-912 scintillator [11] with the help of the calculated detector efficiency values of the thin NE-908.

Errors include the statistical errors of measurements [11], the statistical accuracy of the calculation of the thin glass efficiency and the accuracy of the $^6Li(n,\alpha)^3T$ cross sections. The last one was taken to be equal ± 2 % for the energy range E_n < 100 keV and ± 5 % for higher energies.

5. Results and discussion

^{252}Cf fission prompt neutron spectrum N(E) /shown in Fig. 6. and Table 2/ was obtained using the following expression

$$N(E) = \frac{P(E)}{N_f\ \Omega \cdot \mathcal{E}(E)}$$

P(E) is the converted to energy scale P'(t) distrubution N_f is number of fission events, Ω and $\mathcal{E}(E)$ are the solid angle and efficiency of the neutron detection. The spectrum data errors include the statistical errors of the measurements of $P_1(t)$, $P_2(t)$, $P_3(t)$, $P_4(t)$ distributions, the errors of the $\alpha(t)$correction, the error of the neutron detection efficiency, the errors of N_f and Ω . The error of $\alpha(t)$ which less then 2 % was obtained from the deviation of two $\alpha(t)$ calculated for different Maxwellians with parameters T of 1.2 and 1.6 MeV, respectively, taking into account the statistical accuracy of calculations. The error in N_f was determined using the fission fragments spectrum /see Fig. 2/ and was equal ± 3 %. Error in Ω , defined by the accuracy of the flight path determination and the diameter of the lithium glass, was equal to ± 2 %.

Fig. 6 shows a Maxwellian distribution with parameter $\bar{\nu}_p$ = 3.757 /ENDF/B-V/ and T = 1.42 MeV, which describes the experimental data quite well. With some exceptions the deviation of experimental data from the values of a Maxwellian distribution do not exceed ± 5 %. The ratio of the three-point averaged experimental data to the same Maxwellian is shown in Fig. 7. This result confirms the conclusion made by M.V. Blinov and his coworkers [1-3] and contradicts to experimental data of Ref. [6-9], where essential deviations were found from a Maxwellian distribution extrapolated from higher energies.

It is difficult to find the exact reasons for these disagreements, including our earlier measurements, too [7,8]. We can point out only some factors which were not taken into account in our earlier measurements [7,8]. For example the real response function of the spectrometer was not taken into account though it plays a very important role specially at the geometry used in Ref.[7]. Inclusion of delayed neutrons might cause a softening of the measured spectrum [8].

6. Acknowledgements

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Table 1: Models for Monte-Carlo calculations of functions F'(E,t) and F''(E,t)

No of zone	r [cm]	h [cm]	Concentration [nucl. x cm^{-3} x 10^{20}] ^{6}Li		^{7}Li		N		O		Fe		Si	
			F'(E,t)	F''(E,t)	F'(E,t)	F''(E,t)	F'(E,t)	F''(E,t)	F'(E,t)	F''(E,t)	F'(E,t)	F''(E,t)	F'(E,t)	F''(E,t)
1	2.25	100	-	-	-	-	-	0.42	-	0.12	-	-	-	-
2	2.25	0.01	-	-	-	-	-	-	-	-	-	8.5	-	-
3	2.25	1.73	-	-	-	-	-	0.42	-	0.12	-	-	-	-
4	2.25	0.01	-	-	-	-	-	-	-	-	-	8.5	-	-
5*	2.25	0.15	-	-	-	-	-	0.42	-	0.12	-	-	-	-
6	2.25	0.01	-	-	-	-	-	-	-	-	-	8.5	-	-
7	2.25	1.88	-	-	-	-	-	0.42	-	0.12	-	-	-	-
8	2.25	0.01	-	-	-	-	-	-	-	-	-	8.5	-	-
9	2.25	27.62	-	-	-	-	-	0.42	-	0.12	-	-	-	-
10	2.25	0.95	175.4	175.4	8.6	8.6	-	-	477.8	477.8	-	-	191.2	191.2
11	2.25	100	-	-	-	-	-	0.42	-	0.12	-	-	-	-

*Source is located on the boundary between 5 and 6 zone.

Table 2: Efficiency of neutron detection $\mathcal{E}(E)$ [%] and prompt neutron spectrum N(E) for spontaneous fission of ^{252}Cf [neutr. x fission^{-1} x MeV^{-1} x sterad.$^{-1}$ x 10^{-2}]

E_n, keV	$\mathcal{E}(E)$	N(E)	E_n, keV	$\mathcal{E}(E)$	N(E)
25	1.89±0.04	3.27±0.46	35	1.70±0.04	2.72±0.45
45	1.53±0.03	4.34±0.48	55	1.39±0.03	4.37±0.49
65	1.35±0.03	4.55±0.48	75	1.29±0.03	5.05±0.49
85	1.29±0.03	4.89±0.46	95	1.32±0.03	5.67±0.45
105	1.34±0.03	5.67±0.50	115	1.38±0.04	6.44±0.53
125	1.40±0.05	7.35±0.56	135	1.48±0.07	7.12±0.52
145	1.58±0.08	6.86±0.50	155	1.74±0.09	7.83±0.54
165	1.94±0.10	7.72±0.53	175	2.65±0.14	7.05±0.48
185	2.62±0.14	8.39±0.56	195	3.78±0.20	7.38±0.49
205	4.58±0.24	7.12±0.47	215	5.38±0.28	8.39±0.55
225	5.40±0.28	8.02±0.53	235	6.52±0.34	7.76±0.51
245	6.54±0.34	7.83±0.51	255	5.72±0.30	8.02±0.52
265	5.00±0.26	8.95±0.58	275	5.16±0.27	8.47±0.56
285	4.54±0.23	8.35±0.55	295	3.35±0.17	9.55±0.63
305	2.97±0.15	9.21±0.61	315	2.70±0.14	9.19±0.61
325	2.69±0.14	8.64±0.57	335	2.19±0.11	9.38±0.63
345	2.04±0.11	8.88±0.60	355	1.88±0.10	9.32±0.61
365	1.89±0.10	9.14±0.62	375	1.79±0.09	9.17±0.68
385	1.62±0.08	9.40±0.64	395	1.59±0.08	9.06±0.62
410	1.48±0.08	9.70±0.65	430	1.36±0.07	9.99±0.67
450	1.28±0.07	9.43±0.63	470	1.11±0.06	9.73±0.66
490	1.00±0.05	9.39±0.63	510	0.89±0.05	9.85±0.67
530	0.82±0.04	10.14±0.68	550	0.76±0.04	10.03±0.68
570	0.74±0.04	9.66±0.66	590	0.71±0.04	9.85±0.67
610	0.68±0.04	10.10±0.69	630	0.65±0.03	10.03±0.69
650	0.63±0.03	10.07±0.69	670	0.62±0.03	10.03±0.69
690	0.60±0.03	9.99±0.69	710	0.56±0.03	10.37±0.71
735	0.54±0.03	10.41±0.70	765	0.52±0.03	10.55±0.71
795	0.53±0.03	10.22±0.69	825	0.54±0.03	9.99±0.68
855	0.54±0.03	9.66±0.65	885	0.53±0.03	10.07±0.68
915	0.53±0.03	9.73±0.66	945	0.53±0.03	9.70±0.66
975	0.54±0.03	9.47±0.64	1005	0.54±0.03	9.51±0.64
1020	0.53±0.03	9.40±0.63	1060	0.52±0.03	10.03±0.67
1100	0.50±0.03	10.0 ±0.67	1140	0.51±0.03	10.07±0.67
1180	0.50+0.03	9.66+0.63	1220	0.50+0.03	10.0+0.67

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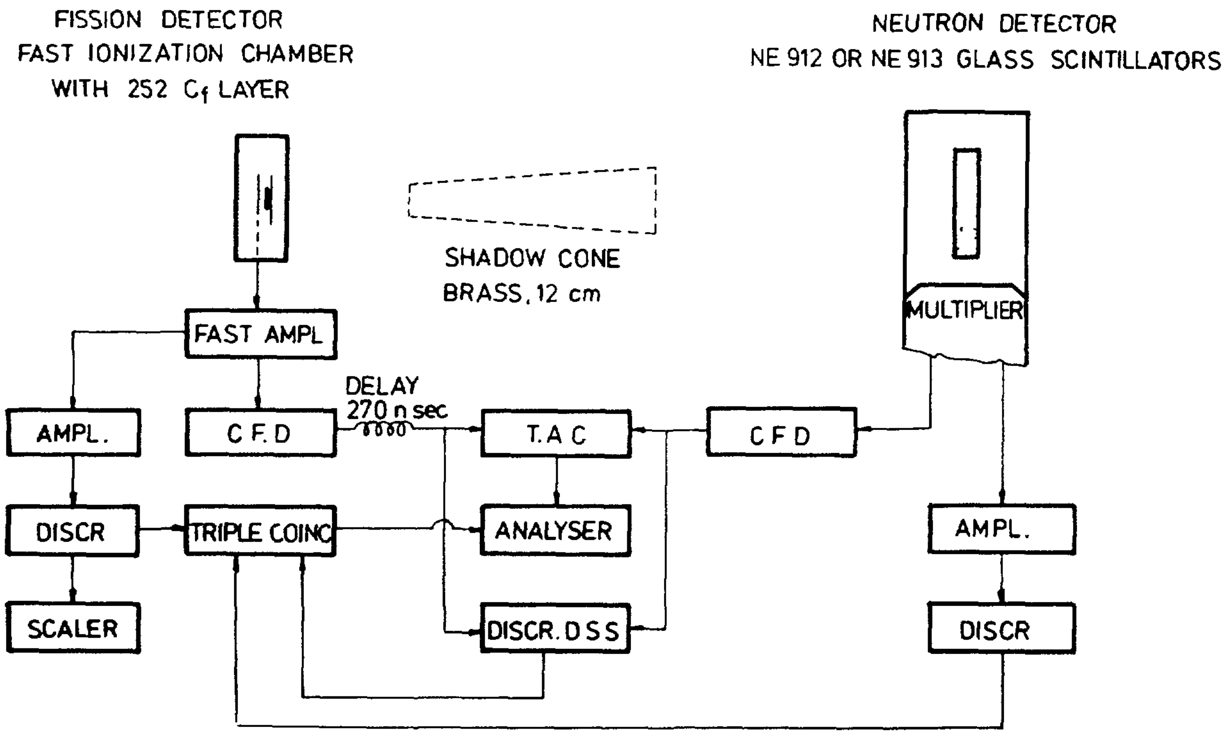
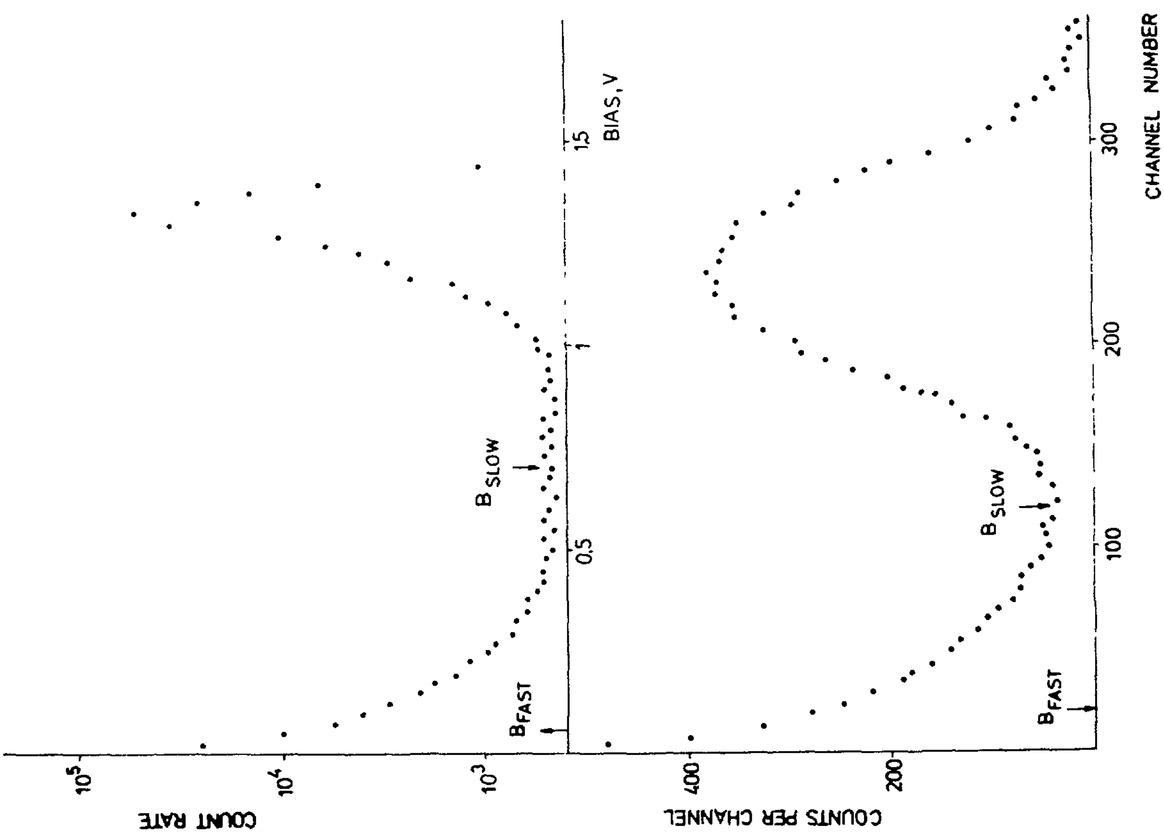


Fig. 1. Schematic drawing of experimental arrangement.

Fig. 2. Fission fragment amplitude spectrum of ionization chamber /top/ and thermal-neutron spectrum of NE-921 scintillator /bottom/.



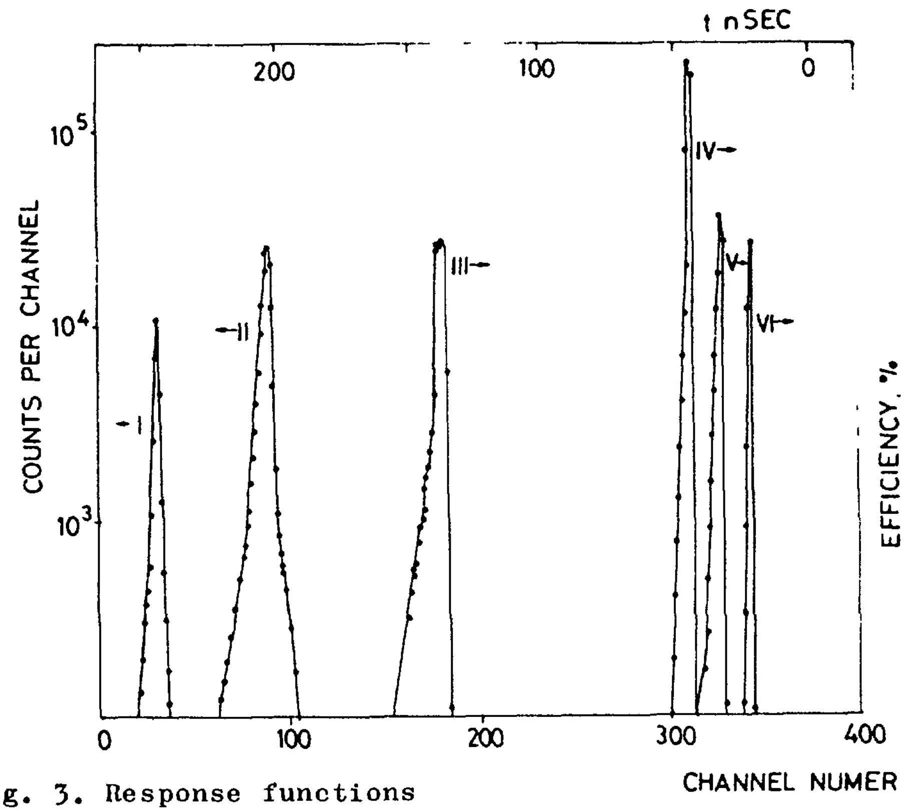
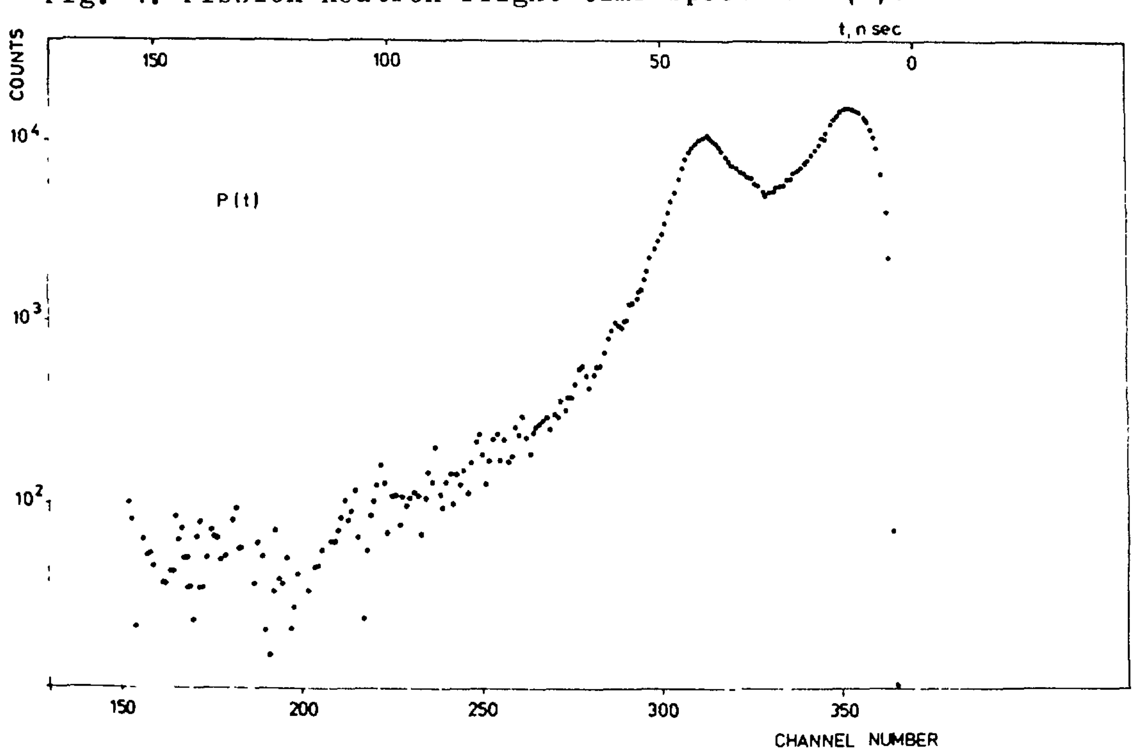


Fig. 3. Response functions
I-II the measured $F_f(t,t')$ and $F_n(t,t')$ response function
III-IV the calculated $F''(E,t)$ response function at neutron energies 0.0025, 0.245, 0.445 and 1.005 MeV.

Fig. 4. Fission neutron flight time spectrum P(t).



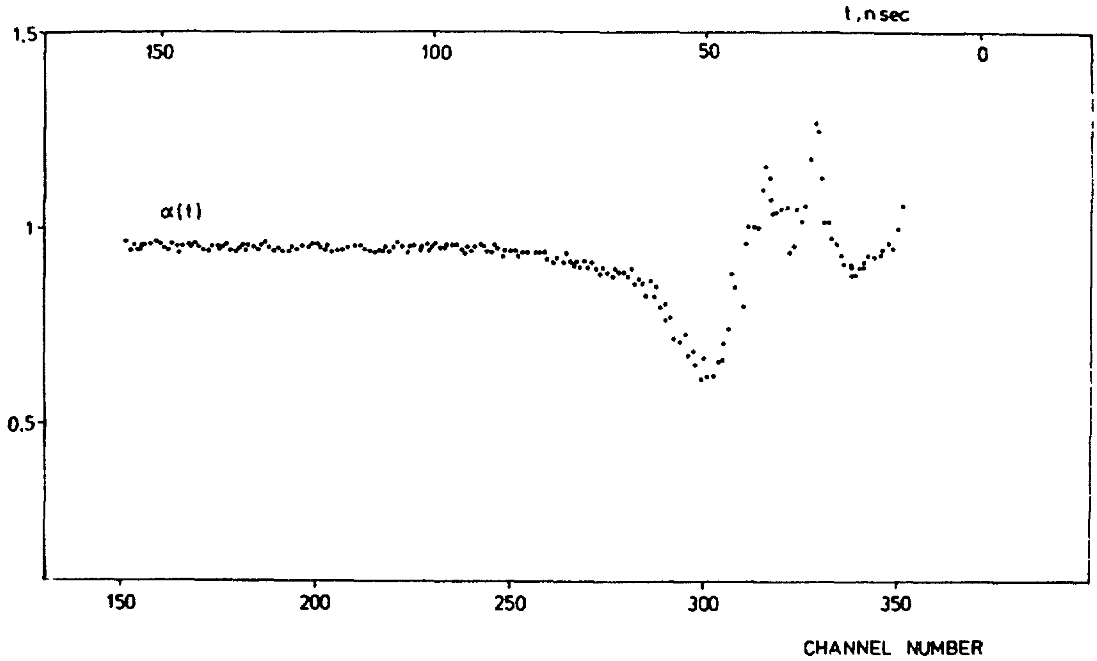


Fig. 5. $\alpha(t)$ response function correction

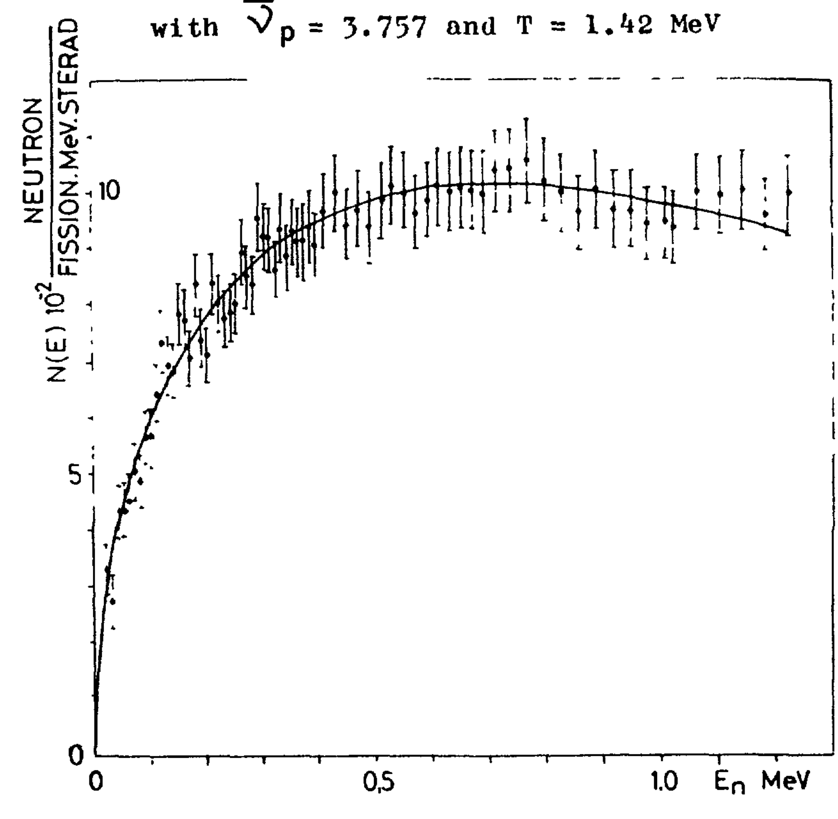
Fig. 6. Prompt fission neutron spectrum N(E) of ^{252}Cf

recent experimental data

– calculated Maxwell spectrum

$$N(E) = \bar{\nu}_p (2\pi)^{-3/2} T^{-3/2} \sqrt{E} \exp(-E/T)$$

with $\bar{\nu}_p = 3.757$ and T = 1.42 MeV



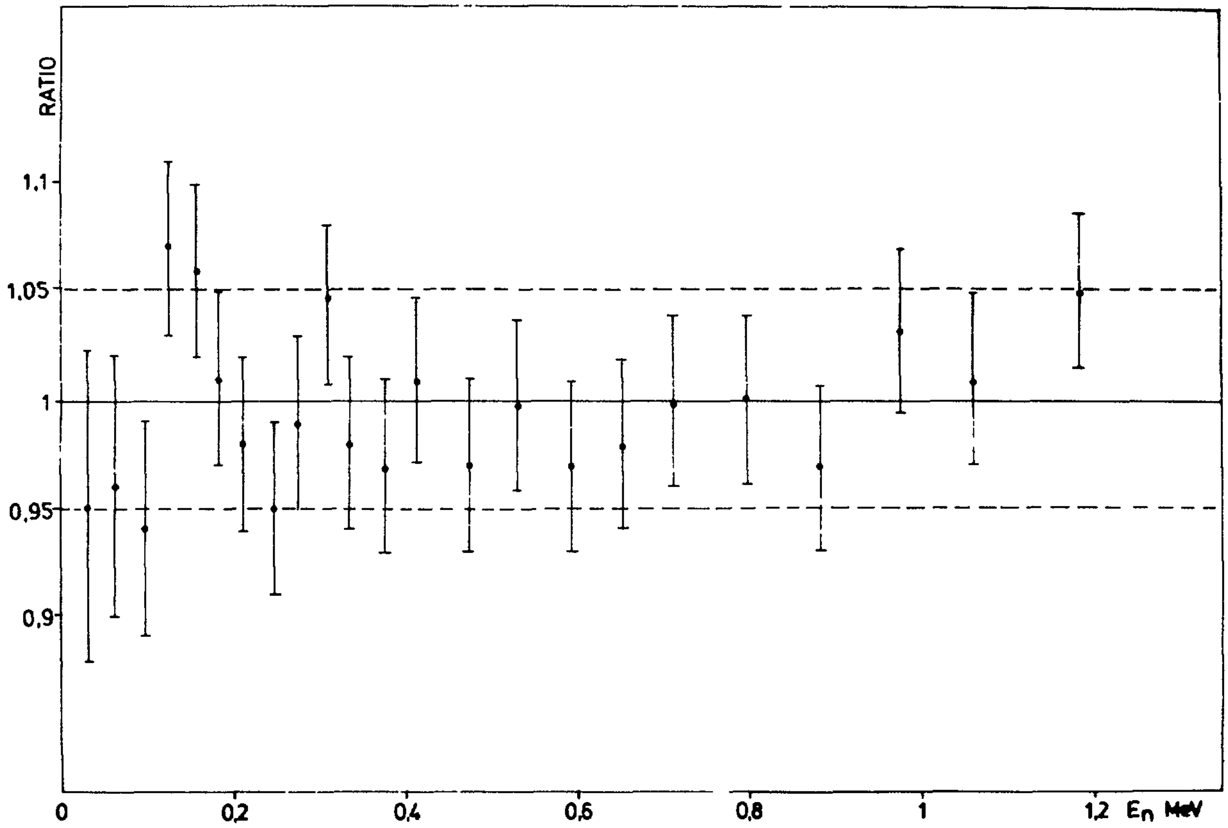


Fig. 7. Ratio of the present results to Maxwellian with a temperature of 1.42 MeV.