RU0110991

Features of the Neutron Spectra Accompanying the Fission of Actinide Nuclei

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Abstract. The spectra of fission neutrons from ^{238}U are measured by the time-of-flight technique at incident-neutron energies E_n=5.0 and 13.2 MeV. The data are compared with those obtained in the previous studies for ^{232}Th, ^{235,238}U, ^{237}Np at E_n=2.9 and 14.7 MeV; for ^{232}Th at E_n=14.6 and 17.7 MeV; for ^{238}U at 16.0 and 17.7 MeV. An excess of soft neutrons, which is observed in comparing experimental spectra for E_n=13.2, 14.7, 16.0, and 17.7 MeV with the results of traditional theoretical calculations, is reproduced fairly well under the assumption that, at high excitation energies of a compound system, some part of postfission neutrons can be emitted by nonaccelerated fragments.

Introduction

A scientific aspect of the investigated problems is connected with the appearance of unknown features (or maybe features that have not received sufficient attention) in the distributions of neutrons as the energy of the bombarding (primary) neutrons E_n is raising above the threshold of the reaction when nuclear fission becomes an emission process. Neutron emission and fission are the dominating types of decay of excited heavy nuclei. When the excitation energy $U= E_n+B_n$ of the initial compound nucleus A satisfies the condition

$$U \geq E_f^{A-\nu} + \sum_{i=1}^{\nu} B_n^{A+1-i} = U_\nu \tag{1}$$

the fission of its lighter isotopes A-ν i.e., of the residual nuclei after the emission of ν neutrons, becomes energetically possible. The quantities E_f and B_n in (1) are, respectively, the height of fission barrier and neutron binding energy for the nuclei. The superscripts are the mass numbers of the nuclei. The entire right side in the case ν=1, i.e., $U_1 = E_f^{A-1} + B_n^A$, is the threshold for emission fission.

The most available data in the region of excitation energy $U \leq U_1$ (first plateau of fission cross sections) do not contradict a hypothesis of the neutron emission from fully accelerated fragments and the neutron-emission process by the evaporation mechanism. This is supported by experimental data on spontaneous nuclear fission and thermal neutron-induced fission. The contribution of other possible mechanisms (such as so-called "scission" neutrons, neutrons emitted during fission fragment acceleration, etc.) is not significant. Although the analysis of numerous experiments has shown that neither Maxwell nor Watt type distributions do not provide the absolutely accurate reproducing of experimental spectra, the deviations are not so remarkable and both of them are commonly used to describe and compare the fission neutron spectra parameters and their behaviour via incident-neutron energy. These deviations do not radically change the effects under discussion.

Initially, these features were observed in comparing the measured spectra of neutrons at two characteristic energies of E_n=2.9 and 14.7 MeV for the target nuclei ^{232}Th, ^{235,238}U, ^{237}Np [1-3]. In the first case, the excitation energy of U=2.9 MeV+B_n is below the energy thresholds U_l for (n,n'f) reactions, and secondary neutrons are emitted from exited fission fragments of compound nuclei. In the second case the excitation energy of U=14.7 MeV+B_n lies much higher than the thresholds U_l for (n,n'f) reactions, and fission process has of an emission character- that is, the fission of the residual nuclei A-1 and A-2 formed after the emission of one or two neutrons is energetically possible. Under such conditions, not only fission fragments but also the fissile nuclei A and A-1 become sources of neutrons. According to the generally adopted terminology, neutrons emitted prior to the fission of $A-\nu$ (ν=1, 2,...) nuclei are referred to as prefission neutrons, whereas emission neutrons from excited fission fragments of $A-\nu$ (ν=0, 1, ...) nuclei are called postfission neutrons. It is usually assumed that postfission neutrons are emitted by fully accelerated fragments.

The measurements performed at E_n=2.9 and 14.7 MeV made it possible to compare in one experiment pure spectra of neutrons from fission fragments (Maxwell type distribution) and spectra involving both pre- and postfission neutrons. The spectra of neutrons at 2.9 MeV served as a background (it is to say a standard of spectrum shape) against which effects associated with emission of prefission neutrons manifested themselves. Experimental results show that the energy distributions at E_n=14.7 MeV are different in shape from the analogous distributions at E_n=2.9 MeV. Additional experimental information obtained for a ^{232}Th target at E_n=14.6 and 17.7 MeV [4], for a ^{238}U target at E_n=16.0 and 17.7 MeV [5] display that the newly measured spectra always differ in shape from the spectra corresponding to E_n=2.9 MeV. In this paper we report the results of our neutron spectra measurements for the target nucleus ^{238}U at the energies of primary neutrons E_n=5.0 and 13.2 MeV and explain the observed change in the shape of neutron spectrum with primary-neutron energy. By way of example, our analysis is performed for the reaction ^{238}U(*n,xn'f*) at E_n=2.9, 5.0, 13.2, 14.7, 16.0, and 17.7 MeV.

Experiment

The first series of measurements (E_n=2.9 and 14.7 MeV) was made at a setup of the Radium Institute (RI) [1,2]. The following researches (E_n=5.0, 13.2, 16.0, and 17.7 MeV) were done at the Institute of Physics and Power Engineering (IPPE) by using a continuous beam from a KG-2.5 neutron generator [4,5]. Background conditions in experimental hall were also different. Neutrons with energies 2.9, 5 MeV and 13.2, 14.7, 16.0, 17.7 MeV were obtained in reactions D(d,n)^{3}He and ^{3}H(d,n)^{4}He.

Energy spectra of neutrons were measured with a time-of-flight spectrometer in coincidence with fission fragments in the energy range 0.25-12 MeV. The measurements were carried out with respect to well-known prompt neutrons spectrum from spontaneous fission of ^{252}Cf, moreover, the investigated $N_i(E,E_n)$ and standard $N_{Cf}(E)$ spectra were studied simultaneously. The experimental setup included the fission fragment detector (ionization chamber), a neutron detector with shielding and instrumentation providing for data acquisition and preliminary experimental data sorting.

The detector of fission fragments was constructed as a four-sectional multilayer ionization chamber, each sections being connected with a separate time-of-flight channel. Three sections of the chamber contained an isotope under investigation with 12 layers per section. The layers of fissile material (2 mg/cm^2 in thickness, 100 mm in diameter) were applied on both sides of aluminum foil backing 0.05 mm in thickness. The total weight of the isotope under investigation in each sections was 1.87 g. The fourth "monitor" section contained two unilateral targets made of the isotope under analysis of the same thickness, with the isotope ^{252}Cf uniformly embedded in them.

The identity of all sections for count and amplitude characteristics was tested by measuring fission fragment spectra.

The detector of neutrons consisted of a stilbene crystal 63 mm in diameter and 39 mm in height coupled to fast photomultiplier tube FEU-30. The characteristics of the detector were as follows: the threshold for neutron detection (with the n-γ-compensation circuit) was equal to 250 keV, and absolute efficiency of neutron detection was about 30% (the latter was determined by measuring the spectrum of prompt neutrons from ^{252}Cf spontaneous fission with the simultaneous detection of fission fragments). The detailed description of the experimental arrangement was presented earlier [1,2].

Experimental results and analysis

Comparison of experimental data represented as the ratios $R_i(E,E_n)=N_i(E,E_n)/N_{Cf}(E)$ of normalized (to unity) spectra of neutrons from the nuclei $i=$ ^{232}Th, ^{235}U, ^{238}U, ^{237}Np to the corresponding spectra as measured for the spontaneous fission of ^{252}Cf revealed [1-3], as it may be seen in Fig.1, that these ratios at E_n=14.7 MeV are totally different in shape from the analogous ratios at E_n=2.9 MeV. In the latter case $R_i(E,E_n)$ is close to linear function. This means that the shape of the distributions observed at E_n=2.9 MeV corresponds to the evaporation mechanism of neutron emission from fully accelerated fragments. The Maxwell distribution $N_M(E,T)$ for the spectrum of postfission neutrons was used in our analysis of experimental data. The slope of calculated ratios (dashed curves in Fig.1 for E_n=2.9 MeV)

$$R_i^c(E,E_n)=\frac{N_M(E,T_i)}{N_M(E,T_{Cf})}=(T_{Cf}/T_i)^{3/2}\exp\left[-\frac{T_{Cf}-T_i}{T_{Cf}T_i}E\right] \qquad (2)$$

are determined by temperature difference $T_{Cf} - T_i$, where T_{Cf} and T_i are the temperatures of the spectra of fission from ^{252}Cf and from the nucleus under investigation. The corresponding values of temperature T_i are shown in Table (the reference value T_{Cf}=1.42 MeV).

Table. Temperature values of the fission neutron spectra of the nuclei under investigation

Primary compound nucleus	^{233}Th	^{236}U	^{239}U	^{239}Np
T_i, MeV	1.285±0.018	1.344±0.015	1.332±0.016	1.369±0.015

The observed $R_i(E,E_n)$ and approximating $R_i^c(E,E_n)$ distributions as functions of E reveal small deviations from linear dependencies and approach linear dependencies more closely with decreasing temperature difference $T_{Cf} - T_i$ which quantity determines the tangent of the angle between the dashed curves and the abscissa.

In the case of E_n=14.7 MeV incident neutron energy (Fig.1) the ratios $R_i(E,E_n)$ are also very similar. The contribution of prefission neutrons is clearly identified by the deviation from the Maxwell type distribution corresponding to neutron emission from excited fission fragments. There are some features in the experimental ratios. The maximum at $E_{max}\approx$8.5 MeV as it will be easily understood is connected with the nonequilibrium hard (high energy) component of prefission neutron spectrum. Its right slope of the maximum corresponds to the cut-off in nonequilibrium spectrum by the fission threshold of the residual nuclear A-1 formed after emission of the first neutron. The spectrum has a cut-off at the neutron energy

$$E_{\max} = E_n - E_f^{A-1}. \qquad (3)$$

For $E>E_{max}$, the spectrum of fission neutrons is determined exclusively by postfission neutrons. The effect under discussion could not be observed if the emission of first neutrons were determined only by the equilibrium mechanism characterized by a soft evaporation spectrum such that the yield of neutrons with energies in the vicinity of $E\approx E_{max}$ is small for the E_n values considered here. An admixture of a hard component due to the nonequilibrium mechanism producing neutron with a yield that is greater than that for the evaporation mechanism by several orders of magnitude makes it possible to observe visually the effect associated with the fact that the spectrum of first neutrons is limited by the $(n,n'f)$ threshold. An ascending character of the spectrum with decreasing secondary-neutron energy in the region E<2 MeV was tentatively attributed to the evaporation component of prefission neutrons. As can be seen in Fig.1, our data are in a good agreement with previous measurements [6] carried out for the energy range $E\cong$0.3-5 MeV at the incident neutron energy E_n=14.3 MeV.

Under the assumption of two sources of neutrons (nuclei undergoing fission for prefission neutrons and fully accelerated fragments for postfission neutrons), the traditional statistical description that takes into account the contribution of nonequilibrium neutrons according to the exciton model of preequilibrium decay can reproduce the shape of the observed distributions at E_n=14.7 MeV only in the region of secondary neutrons $E \geq 2$ MeV [2,7]. In the low-energy region (E<2 MeV), experimental spectra show an anomalously large yield of soft neutrons in relation to the results of the calculations. There is a clear tendency toward a decrease in the yield of the anomalous soft component in the spectra of fission neutrons with increasing fissility of nuclei.

The discovery of the anomalous soft component in the spectra of neutrons accompanying the fission of heavy nuclei at E_n=14.7 MeV required further studies. As can be seen from Fig.2(a) the second series of measurements [4] carried out at a setup of IPPE showed that the newly measured spectrum for a ^{232}Th target at E_n=14.6 MeV is in a good agreements with the result of the first series of measurements made at a equipment of RI. Taking into account the scientific and practical significance of the studied effects, we considered it necessary to investigate experimentally the spectra of neutrons from emission fission at higher energies of incident neutrons. It makes possible to establish that anomalously soft neutrons are observed at other excitation energies of the initial fissile nucleus and to carry out a global statistical analysis in order to clarify the physical origin of these neutrons.

As seen in Fig.2(b) the experimental distribution $R(E,E_n)$ measured for the ^{232}Th target nucleus at primary neutron energy E_n=17.7 MeV is also similar in shape to analogous ratios at E_n=14.7 MeV showed in Figs.1, and 2(a). The spectra of fission neutrons were now measured primarily for the ^{238}U target nucleus. Firstly the measurements were carried out for E_n=16.0 and 17.7 MeV [5] after that for E_n=13.2 MeV and finally for E_n=5.0 MeV. Of the four nuclei studied previously, we chose ^{238}U because the attainment of statistical accuracy necessary for observing the above effect (the maximum effect was observed for ^{232}Th and ^{238}U) requires a much less operating time of the accelerator than in experiments with ^{232}Th. This is especially important for long-term measurements. The more detailed picture of the changes in shape of the experimental distributions obtained in researches for the ^{238}U+n system at energy values E_n=2.9, 5.0, 13.2, 14.7, 16.0, and 17.7 MeV are shown in Fig.3.

For E_n=5.0 MeV, the excitation energy $U=E_n+B_n$ is below the energy threshold U_1 for ^{238}U(n,n'f) reaction and secondary neutrons originate only from excited fission fragments of compound nucleus ^{239}U. As in case of E_n=2.9 MeV the observed $R(E,E_n)$ and approximating $R^c(E,E_n)$ (2) distributions for E_n=5.0 MeV (see Fig.3) as functions of E reveal small deviations from linear dependence. The shape of newly measured spectrum of postfission neutrons is close to Maxwell type distribution.

The shape of the experimental distributions $R(E,E_n)$ in Fig.3 at E_n=13.2, 14.7, 16.0, and 17.7 MeV displays special features mentioned above that clearly distinguish these distributions from a nearly linear dependence for E_n=2.9 and 5.0 MeV.

A statistical description that is based on the postulate of two sources of neutrons (the compound system for prefission neutrons and the fully accelerated fission fragments for postfission neutrons) and which takes into account the contribution of prefission neutrons from nonequilibrium decay satisfactory reproduces the shape of the observed distributions (curve 1 in Fig.3) over a larger part of the energy region

($E \geq 2$ MeV), demonstrating that this interpretation of the features associated with nonequilibrium neutron emission is qualitatively correct. However in the soft section of spectra $R(E,E_n)$ (E<2 MeV) the calculated curves 1 are considerably lower than experimental values.

To explain the excess of soft neutrons in the spectra corresponding to the emission fission of nuclei it is assumed that the mechanism according to which postfission neutron emission occurs only from fully accelerated fragments requires refinement. Following [8] we consider fragments at the instant of separation as a two-body system in which the interaction potential involves repulsive Coulomb interaction and attractive nuclear forces. It is also assumed that in this system all degrees of freedom are in statistical equilibrium and its lifetime is such that nonaccelerated fragments may emit neutrons if this is energetically possible. Let $U_0=U_{01}+U_{02}$ is the total excitation energy of internal degrees of freedom (thermal energy) of the complementary fragments that is due to transition of nucleus undergoing fission to the configuration of the fragments in touch. With the excitation energy U_{0i} (i=1,2) we associate neutron emission from nonaccelerated fragments under condition $U_{0i}>B_n^{A_i}$ ($B_n^{A_i}$ is the neutron binding energy in the fragment A_i) and attribute the above low energy anomaly in the spectra to this emission.

The absence of extra neutrons at E_n=2.9 and 5.0 MeV and their appearance at the higher energies of E_n>13 MeV indicate that U_{0i} depends on the energy of bombarding neutrons and, hence, on the excitation energy of an initial compound nucleus. To simplify the problem we assume that this dependence is linear and that the excitation energy is distributed between two fragments in proportion to their masses:

$$U_{0i}=C(E_n+B_n^A)\,A_i/A \tag{4}$$

The factor C shows what fraction of the excitation energy of a compound nucleus has been transferred to the intrinsic degrees of freedom of fragments at the instant of their separation. In estimating the neutron spectrum from nonaccelerated fragments the distribution of the excitation energy for an individual fragment is assumed to have a Gaussian form with mean value $\langle U_{0i}\rangle$ according to (4). The factor C affects the hardness of the spectrum and the yield of neutrons from nonaccelerated fragments. All the details of this theoretical approach were presented in [9].

To describe the spectra of pre- and postfission neutrons the observed fission cross section $\sigma_f(E_n)$ was represented as the sum $\sigma_f(E_n)=\sum_{\nu=0}^{\nu_{max}(E_n)}\sigma_{fA-\nu}(E_n)$ of partial components $\sigma_{fA-\nu}(E_n)$ [2, 9] for a chain of the fissile nuclei A, A-1, ..., $A-\nu_{max}$ corresponding to ν=0, 1, ..., $\nu_{max}(E_n)$ neutrons emitted prior to fission. Isolating the contributions of prefission neutrons, neutrons from nonaccelerated fragments, and neutrons from fully accelerated fragments the calculated spectrum can be represented as

$$\frac{d\bar{\nu}_c(E,E_n)}{dE}=\frac{d\bar{\nu}_{pre}(E,E_n)}{dE}+\frac{d\bar{\nu}_{naf}(E,E_n,C)}{dE}+\alpha\sum_{\nu=0}^{\nu_{max}(E_n)}\bar{\nu}_{fA-\nu}N_M(E,\beta T_\nu)\frac{\sigma_{fA-\nu}}{\sigma_f}, \tag{5}$$

where α is a constant introduced to fit experimental data and to compensate for inevitable uncertainties in describing $\bar{\nu}_{f A-\nu}$ on the basis of extrapolating the systematics of $\bar{\nu}_f(E_n)$ from [10] to the region $E_n>6$ MeV. Similar uncertainties are inherent in using the systematics of $T(E_n)$ from [11]; because of this, the quantities T_ν were varied within 3% with the aid of the constant β.

By varying the fitting parameter C there was achieved a correct description of the experimental spectra in the low-energy region $E<2$ MeV. The calculated neutron spectrum (5) with parameters fitted to experimental data corresponds to the calculated ratio $R_c(E,E_n)=\frac{d\bar{\nu}_c(E,E_n)/dE}{\bar{\nu}(E_n)N_M(E,T_{Cf})}$. At one value of the coefficient C (C=0.53) the shape of neutron distributions observed at E_n=13.2, 14.7, 16.0, and 17.7 MeV is satisfactorily reproduced by the results of the calculations (curve *2* in Fig.3) over the entire range of measured secondary neutron energies including the low-energy section $E<2$ MeV.

Conclusion

The experimental ratios $R(E,E_n)$ of the spectra $N(E,E_n)$ of prompt neutrons for ^{238}U fission induced by neutrons with energies of E_n=5.0 and 13.2 MeV to the spectrum $N_{Cf}(E)$ for the spontaneous fission of ^{252}Cf were measured. At least in the measured energy range of secondary neutrons 0.25-12 MeV for the reaction (n,f) induced by primary neutrons of energy E_n=5.0 MeV the behaviour of experimental distribution shape was found to be analogous to those for neutrons from the induced fission of ^{238}U at E_n=2.9 MeV and the spontaneous fission of ^{252}Cf. As well as the well-studied spectrum of spontaneous fission of ^{252}Cf spectra $N(E,E_n)$ at E_n=2.9 and 5 MeV are close to Maxwell distribution $N_M(E,T)$ with an accuracy of the correction which takes into account some small deviations from $N_M(E,T)$ [12].

At E_n=13.2, 14.7, 16.0, and 17.7 MeV the distributions have a maximum at $E=E_{max}$ (3) and rise with decreasing energy E in the region $E<2$ MeV. Under the assumption of two sources of neutrons (nuclei undergoing fission for prefission neutrons and fully accelerated fragments for postfission neutrons) the results of calculations can reproduce the shape of the observed distributions only in the region $E\geq 2$ MeV. In the low-energy region ($E<2$ MeV) the experimental spectra display an anomalously large yield of soft neutrons in relation to the results of calculations. If the third source of neutron emission from nonaccelerated fragments is incorporated into the model calculations the results agree well with experimental data over entire range of measured secondary neutron energies, including the anomalous region $E<2$ MeV.

Further experiments to study this effect are necessary for obtaining deeper insight into the mechanism that is responsible for the emergence of soft neutrons. It should be noted that the additional experimental information about the neutron distributions especially in the low-energy region 10 keV-2 MeV would be useful.

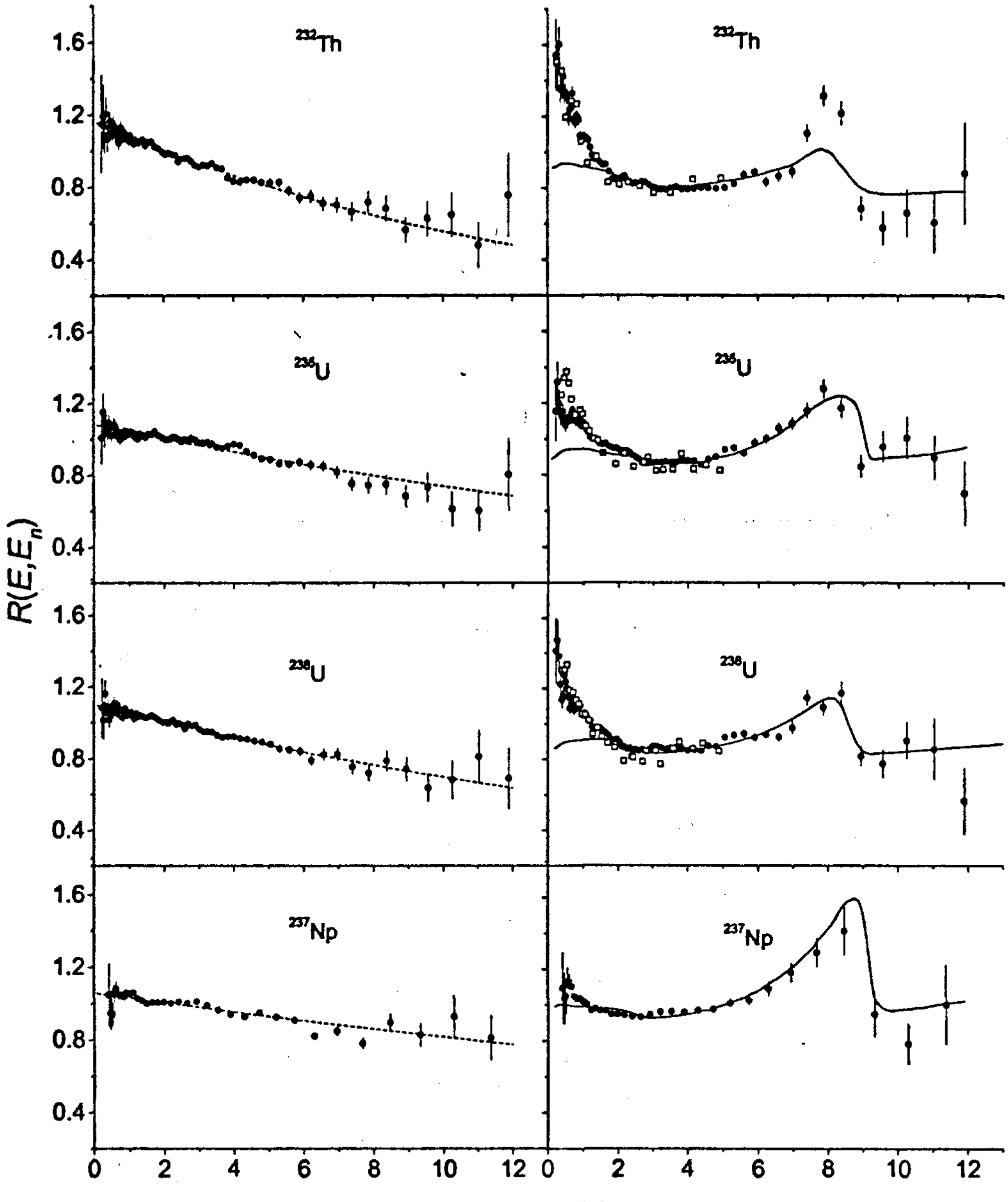

Fig. 1. Ratios of the energy distributions of fission neutrons $R_i(E,E_n)$ for target nuclei $i={}^{232}$Th, $^{235,\,238}$U, and ^{237}Np versus energy E for E_n=2.9 MeV (left-hand column) and E_n=14.7 MeV (right-hand column); the closed points are our experimental values. The open points are the results obtained in [6]; dashed curves show the ratios of the Maxwell distribution with temperatures T_i presented in Table to one with T_{Cf}=1.42 MeV; the solid curves are the results of the calculations for E_n=14.7 MeV

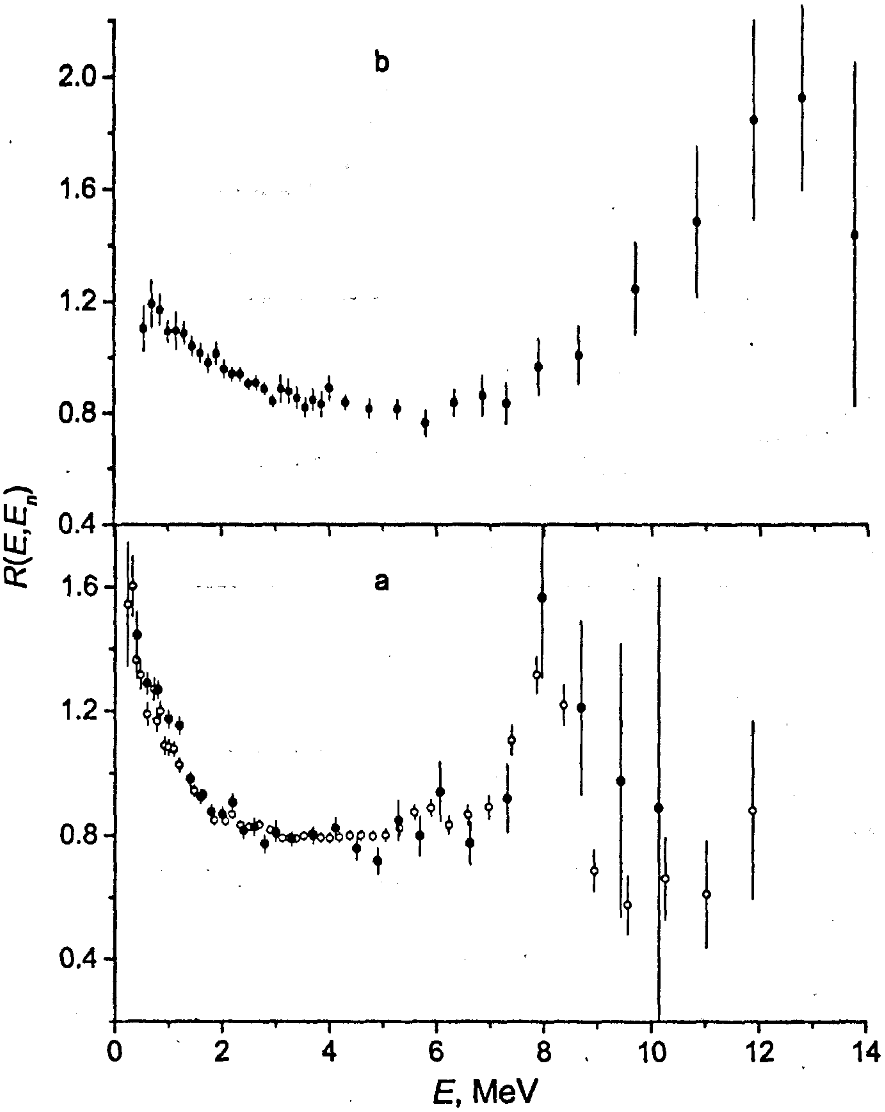

Fig. 2. Exsperimental ratios $R(E,E_n)$ for target nucleus ^{232}Th versus energy E for E_n=14.6 MeV (a) and E_n=17.7 MeV (b). Closed points - IPPE experiment, open points - RI experiment E_n=14.7 MeV

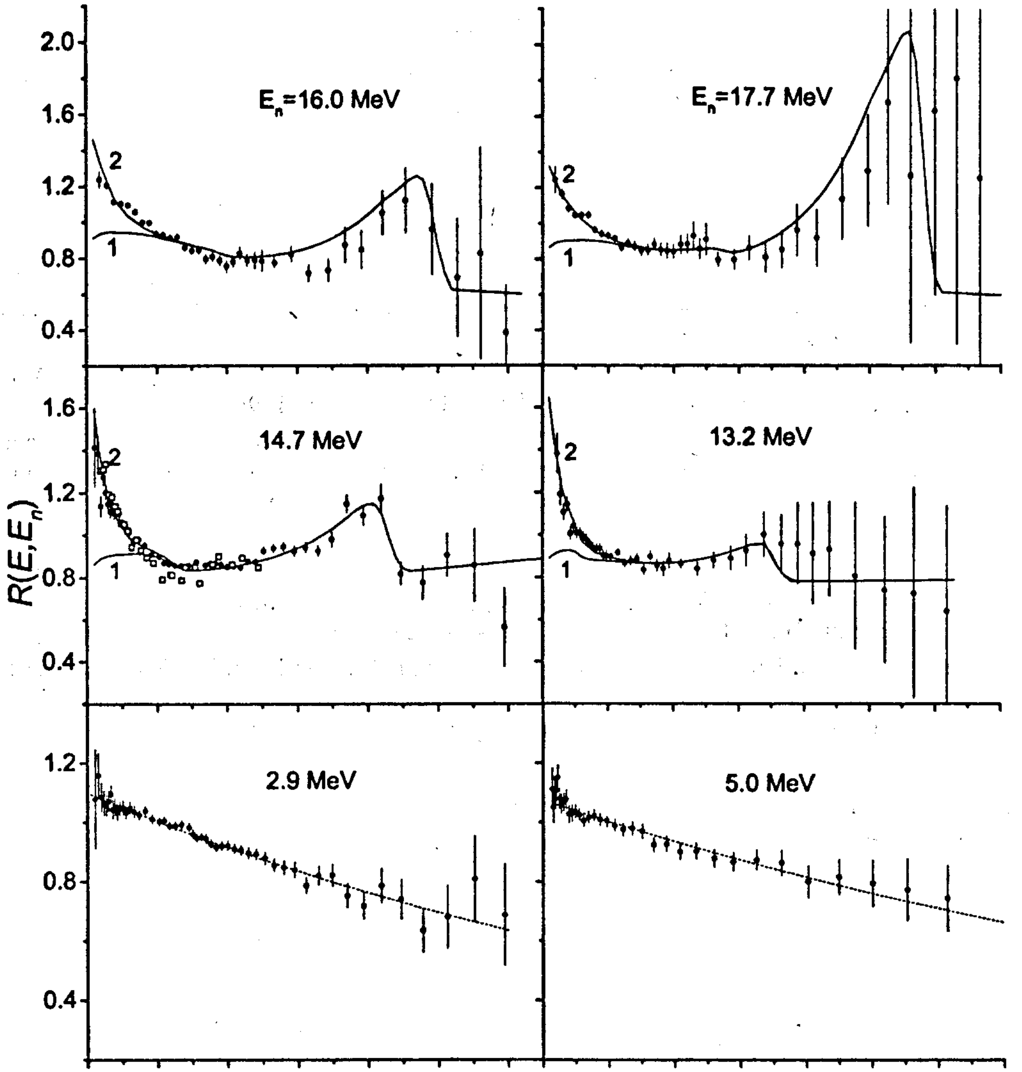

Fig. 3. Ratios $R(E,E_n)$ of the spectra of fission neutrons from the reaction ^{238}U(n,xn'f) to the spectrum of neutrons from the spontaneous fission of ^{252}Cf: (points -solid circles [1-3], - open squares [4]) experimental values, (solid curves 1) results of the calculation without allowance for the contribution of neutrons from nonaccelerated fragments, (solid curves 2) results of the calculation with allowance for the contribution of neutrons from nonaccelerated fragments, and (dashed lines) ratios of Maxwell distributions with temperatures T_U=1.332 MeV for E_n=2.9 MeV, T_U=1.353 MeV for E_n=5.0 MeV and T_{Cf}=1.42 MeV

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