Prompt-fission-neutron average energy for $^{238}\text{U}(n, f)$ from threshold to 200 MeV

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Abstract

Energy distributions of prompt neutrons in coincidence with fission induced on $^{238}\text{U}$ were measured for incident neutron energies up to 200 MeV. The double time-of-flight technique was used to deduce incident and emitted neutron energies. The experimental average and standard deviations of the fission neutron spectra (FNS), for emitted neutron energies from 0.65 to 7.5 MeV, are reported. The results compare well to predictive calculations with the improved Los Alamos model below 20 MeV incident neutron energy. The observed dip at the opening of the second chance fission channel at 6 MeV is confirmed and analyzed. Above 20 MeV, the experimental results of the FNS are smaller than the calculated ones. At 50 MeV and higher, the data suggest a slight increase of the temperature and the kinetic energy of the fission fragments.

Keywords: Neutron-induced fission; FIGARO; Double time-of-flight technique; Los Alamos model; Watt parametrization

1. Introduction

Interest in obtaining a more detailed understanding of prompt neutron emission in fission is now high. To date, there are about half a dozen fission neutron spectra (FNS) compiled in nuclear data libraries for neutron induced fission on $^{238}\text{U}$ at various energies below and at 14 MeV. No data are available on a broad, continuous range of incident neutron energies. Such measurements are now possible. The data are of great importance in the connection of accelerator-coupled nuclear reactor systems (ADS) burning and incinerating actinides, with $^{238}\text{U}$ considered as a prototype actinide. The spallation reactions used to relax the neutron economy of fast nuclear reactors reach to high energy neutrons, which are investigated for the first time in the experiment presented. Moreover, these data would provide valuable information to improve our understanding of fission at high excitation energy. In particular, it is interesting to investigate in which proportions the heated system releases the excess of...
energy with pre-fission neutron and charged particle emission and increase of excitation energy and angular momentum of the primary fragments. As a matter of fact, a theoretical effort has been pursued recently [1,2], to predict the properties of prompt neutron emission in fission at high excitation energies. Prompt fission neutrons are characterized by two basic quantities, the average number of prompt neutrons emitted per fission, which is known up to 30 MeV [3], and the neutron energy spectrum which is not nearly as well known. However, it was shown for a few cases that not only the average energy but also the shape of the FNS depends on the incident neutron energy [4–6]. The fission neutrons are emitted from various sources. At low energies, the main source of neutrons is their evaporation from fission fragments (FFs) from first chance fission. With increasing energy the mixture of fissioning systems with different masses complicates the overall pattern. Also, the emission of pre-equilibrium neutrons prior to fission can contribute to a high-energy tail of the FNS [7,8]. In our measurement we do not distinguish between neutrons emitted before and after fission.

In order to analyze and discuss the data in the next sections, we used the Los Alamos model to calculate the FNS for comparison with data. The starting point of the model is the Weisskopf theory for neutron evaporation from a fragment at a given excitation energy calculated using the triangular prescription by Terrell [9] as a function of nuclear temperature. The neutron energies in the center of mass are transposed to the laboratory frame by assuming isotropic emission from average heavy and light fragments. As the incident neutron energy goes up, higher-chance fission channels open. The laboratory spectra are then weighted by the recently evaluated partial fission cross sections [10]. Pre-fission neutrons, statistically emitted from the compound nucleus, are also accounted for in the FNS. A primary advantage of the Los Alamos model is its predictive power [11].

A simplified approach, but one which represents both experiment and models, is to fit the FNS with a Watt parameterization [12]. In the center of mass, a Maxwellian distribution is assumed for the energy of the neutrons emitted from the decaying fragments with a temperature $T_e$ and an average kinetic energy per nucleon $E_f$. Then in the laboratory frame, the FNS is simply described by a two parameter function

$$N_w(E) = \frac{2A^{3/2}}{(\pi B)^{1/2}} \exp\left(-\frac{B}{4A}\right) \exp(-AE) \times \sinh(BE)^{1/2},$$

where the Watt parameters are related to the physical quantities by the relations $A = 1/T_e$ and $B = 4E_f/T_e^2$. The average neutron energy of the Watt distribution is simply given by

$$\overline{E} = \frac{3}{2}T_e + E_f.$$

The parametrization has a limited accuracy, but it is a useful approximation to describe FNS with low statistics. Also, the average neutron energy is derived analytically, and therefore is not sensitive to details of the spectral shape near the chosen upper and lower thresholds.

2. Experiment and data selection

The experiment was performed on the FIGARO beam line [13] installed at the WNR spallation source of fast neutrons [14] at the Los Alamos National Laboratory. The neutrons are created by an 800 MeV proton beam on an unmoderated tungsten target, with an average proton current of 5 µA. The beam structure consisted of short micro-pulses 1.8 µs apart, grouped in a 625 µs beam gate at a repetition rate of 100 Hz. The incident neutron spectrum was similar to a Maxwellian distribution with a temperature of about 2 MeV and a high energy tail. The FIGARO setup was situated at 22 m from the neutron source and 30 deg with respect to the incident proton beam. The beam was collimated to a spot size of 1.3 cm diameter at the sample position. The double time-of-flight (TOF) method was used. The incident neutron energy was derived from the time difference between the proton pulse in the production target and the fission pulse in a fission chamber. The energy of fission neutrons was derived from the time difference between the fission chamber pulse and a neutron detector pulse.

The 94-layer ionization fission chamber, containing $\sim$ 380 mg of pure $^{238}$U, was $\sim$ 10 cm long. Each layer consisted of a stainless steel backing with deposits of
2 mg of $^{238}$U on both sides. The fission rate was typically 300 s$^{-1}$. But, because of the 6.25% duty factor of the proton beam, the instantaneous fission rate rose higher than 10 000 s$^{-1}$. Discrimination between fission fragments, $\alpha$ particles and noise was excellent, and no bias to the detected fragment distribution was assumed. The time resolution of 7 ns (FWHM) of the chamber was deduced from the photo-fission peak induced by the $\gamma$-ray flash accompanying the proton pulse. The time-resolution associated error on the incident neutron energy ranged from 17 keV at 1.6 MeV to 29 MeV at 200 MeV.

Neutron detectors were used for the experiment. They consisted of scintillation-liquid-filled cells that are 12.5 cm in diameter and 5 cm thick coupled to photo-multiplier tubes. Three detectors were put at 112 cm from the fission chamber at laboratory angles 90, 105 and 120 deg, and two at 218 cm and angles around 90 deg on the opposite side. The detection angles were chosen to minimize the sensitivity of the results to pre-equilibrium neutrons, which are emitted preferentially in the forward direction.

A classical pulse-shape discrimination, based on short-time and long-time scintillation light components, was applied to the data off-line to eliminate $\gamma$-ray events. This discrimination sets a lower limit for the detection of neutrons to several hundreds of keV. However, the residual background in the emitted neutron TOF spectrum had to be reduced by applying an additional requirement on selected events. Indeed, a group of events exhibited a large light deposition but a low emitted neutron energy calculated from the TOF. These unusable events were attributed to accidental coincidences of fission and scattered neutrons. This selection set the FNS threshold to 0.65 MeV for our experiment. The spectra recorded with the detectors at 218 cm could not be used because of the remaining background level. The time resolution of the neutron detectors was much less than that of the fission chamber, i.e., 2 to 3 ns (FWHM). An overall time resolution of 7.5 ns (FWHM) was deduced from the fission prompt $\gamma$-ray peak in the raw TOF spectrum. The associated error on the detected neutron energy ranged from 15% at 0.65 MeV (low threshold) to 50% at 7.5 MeV (high threshold). A total of 650 000 neutron-fission coincidences were recorded during 14 days of actual beam time and for incident neutron energies up to 200 MeV.

A set of 59 FNS spectra was extracted from the TOF spectra. The statistics were summed over the detectors. In the analysis process, the digital value of the TOF of an event was replaced with a real number chosen at random between the two consecutive digits defining the bin. The weight of the event was taken from a normalized trapezoidal interpolation with a slope determined from the local derivative of the distribution. Then, the energy was calculated analytically from the TOF real value and the distribution was stored in a 50 keV per bin spectrum. It was checked that the procedure did not bias the values of the average and standard deviation of the FNS.

The incident neutron energy bin definitions were chosen to get similar statistics in each bin. With about 11 000 counts in each bin the statistical error of the FNS average is less than $\pm1.5\%$.

### 3. Data analysis and results

The efficiency of the neutron detectors was determined from the analysis of the energy spectrum around 2.9 MeV incident neutron energy, where good reference data are available. The distribution was divided by the precision measurement by Boykov et al. [15] and fitted to a 6th order polynomial. The shape of the relative efficiency obtained with this procedure resembles the typical intrinsic efficiency expected for a proton recoil scintillator [16], i.e., a sharp threshold at 0.65 MeV, a maximum at 1.9 MeV and a roughly constant value above 3.2 MeV. The experimental spectrum without and with the normalized efficiency correction is plotted in Fig. 1. The reference spectrum, normalized to the raw data spectrum, is also plotted on the same figure. The efficiency corrected spectrum matches well the reference spectrum within the statistical uncertainties. However, it underestimates slightly the reference below 0.8 MeV and overestimates it from 0.85 to 1.1 MeV. This systematic mismatch is due to the choice of the efficiency function. Since the FNS statistics are rather low, this simple function was preferred over a more sophisticated one. Therefore, the confidence gained over the determination of the efficiency in the range 0.65–7.5 MeV was at the price of a slight overestimate of the average FNS energy.

All spectra were then corrected for efficiency using the above prescription. The raw spectrum statis-
tical error was propagated in each bin. The average neutron energy and standard deviations were simply computed from the statistical analysis of the distribution, applying upper and lower thresholds. The experimental values and the values calculated from the Los Alamos model are plotted in Fig. 2 for comparison. The shape of the distribution up to 20 MeV is very similar to that given by the Los Alamos model. The main features of the FNS average are a dip at 6 MeV, corresponding to the opening of the second chance fission, and a mismatch above 20 MeV, as the calculated values strongly overestimate the data.

In order to reduce the sensitivity of the FNS average to the choice of the efficiency function and the values of the thresholds, the Watt parametrization was used. Both experimental and calculated (Los Alamos model) FNS were fitted with the function of Eq. (1). Then, Eq. (2) was simply used to calculate the FNS average. The error bars were calculated from the propagation of the errors of the fitted parameters $A$ and $B$, typically 5 and 20% for the experimental spectra. The experimental and calculated values are plotted in Fig. 3. The absolute values confirm the trends discussed for Fig. 2, but the error bars are three times larger, because statistical and systematic uncertainties are added. The dominant term, i.e., the partial error with respect to $A$, sets the precision of the FNS average to ±5%.

It is also worth noting that the confidence level of the fit of the FNS with a Watt function was comparable at all incident neutron energies. This means that, for this study of the FNS average, low and high

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**Fig. 1.** Fission neutron spectrum for the system $^{238}\text{U}(n, f)$ measured with the FIGARO setup. Data are for incident neutron energies from 2.1 to 4.0 MeV and are compared to a precision measurement at 2.9 MeV incident neutron energy [15].
incident neutron energy spectra can be approximated by Watt functions with the same precision, i.e., the impact of the higher energy portion of the spectrum is statistically negligible.

4. Discussion and conclusion

The average value provides less detailed information than precise measurements of FNS, but it was measured and calculated with good precision over a wide range of incident neutron energy for $^{238}\text{U}(n,f)$. The results are not sensitive to pre-equilibrium neutrons and high-energy neutrons because of the fitting procedure and because the angles at which neutron detectors were placed biased against pre-equilibrium neutrons. The agreement within errors with the well-tested Los Alamos model calculations up to 20 MeV incident neutron energy validates the experimental procedure. Looking in closer detail at the calculations, one can interpret the dip at 6 MeV as the observation of a transition from first to second chance fission, resulting in a net effect of a 10% decrease of the average energy. Indeed, the relative fission cross-section for the first chance drops from 100% to 30% between 5 and 7 MeV, whereas it rises from 0% to 70% for the second chance in the same range according to a recent evaluation [10]. Above 20 MeV, the calculation overestimates the data. It is also interesting to analyze the gross trend of the FNS av-
Fig. 3. Fission neutron spectrum average for the system $^{238}\text{U}(n, f)$ as a function of the incident neutron energy. Experimental and calculated spectra were fitted with a two parameter Watt function (see Eq. (1)). The average energy is analytically calculated from Eq. (2). Points represent the experimental data and a curve is given for the calculated spectra.

In conclusion, it was shown that experimental tools have recently become available to investigate fission neutron spectra for a broad range of incident neutron energies. The first results with a $^{238}\text{U}$ target are reported in this Letter. The results were interpreted with the Los Alamos model which is well constrained and validated at low incident neutron energies. The observed sharp transition from first to second chance fission results in a 10% decrease in the average energy. Finally, a slight increase of the fission fragment temperature and kinetic energy from threshold to 200 MeV is deduced from the analysis. The results presented in this Letter are the starting point of a more comprehensive future program. In order to measure the missing portions of the FNS, where previous
Fig. 4. Temperature (upper plot) and kinetic energy per nucleon (lower plot) of the fission fragments for the system $^{238}\text{U}(n, f)$ as a function of the incident neutron energy. The values were obtained from the fit of the experimental and calculated (Los Alamos model) FNS with a Watt function. The Watt parameters are represented with points for the experimental data and as a curve for the calculations.

experiments have shown significant deviations from a pure Maxwellian distribution, many more detectors will be mounted. They will be positioned farther off to measure neutron energies above 7.5 MeV, shielded to reduce the reported experimental threshold of 0.65 MeV, and set at various angles to investigate the emission of pre-equilibrium neutrons. Also, these experiments will be carried out with other targets of importance, e.g., $^{235}\text{U}$, $^{239}\text{Pu}$, $^{237}\text{Np}$, which are readily available.

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References