



Experimental Determination of the Antineutrino Spectrum of the Fission Products of ^{238}U

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An experiment was performed at the scientific neutron source FRM II in Garching to determine the cumulative antineutrino spectrum of the fission products of ^{238}U . Target foils of natural uranium were irradiated with a thermal and a fast neutron beam and the emitted β spectra were recorded with a γ -suppressing electron telescope. The obtained β spectrum of the fission products of ^{235}U was normalized to the data of the magnetic spectrometer BILL. This method strongly reduces systematic errors in the ^{238}U measurement. The β spectrum of ^{238}U was converted into the corresponding $\bar{\nu}_e$ spectrum. The final $\bar{\nu}_e$ spectrum is given in 250 keV bins in the range from 2.875 to 7.625 MeV with an energy-dependent error of 3.5% at 3 MeV, 7.6% at 6 MeV, and $\gtrsim 14\%$ at energies $\gtrsim 7$ MeV (68% confidence level). Furthermore, an energy-independent uncertainty of $\sim 3.3\%$ due to the absolute normalization is added. Compared to the generally used summation calculations, the obtained spectrum reveals a spectral distortion of $\sim 10\%$ but returns the same value for the mean cross section per fission for the inverse beta decay.

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Precise predictions of the $\bar{\nu}$ spectra emitted by nuclear reactors are a crucial input for many current and future neutrino experiments. Apart from experiments searching for diffuse supernova neutrino events [1] or geoneutrinos [2,3], where reactor antineutrinos form a substantial background to the expected signal, the knowledge of the spectrum produced by a fuel assembly is of special importance for reactor neutrino disappearance experiments. There exists a variety of reactor neutrino experiments aiming at the determination of neutrino (oscillation) parameters, e.g., by the current Daya Bay [4], RENO [5], and Double Chooz [6] collaborations, as well as plans for the identification of the mass hierarchy with the JUNO [7] or Reno-50 [8] detectors. However, even the setups designed to compare data from near and far detectors are not fully independent of the knowledge of the reactor $\bar{\nu}$ spectrum. Furthermore, there are questions like the possible existence of sterile neutrinos [9] that cannot be studied using this comparative technique. The only possibility to interpret the data of the many short baseline neutrino experiments, performed, so far, in terms of a sterile neutrino analysis, is the accurate prediction of the $\bar{\nu}$ spectrum emitted by the particular fuel assembly. Antineutrino detectors can also be used for the purposes of nonproliferation of nuclear weapons [10]. Monitoring the fuel composition with neutrinos from outside the reactor containment and without input from the reactor staff may give a handle for reducing the undetected manipulation and removal of nuclear fuel. In this case, the neutrino spectrum emitted by the reactor is, again, an important input to the analysis of the data. In a pressurized water reactor (PWR),

four main fuel isotopes contribute to the total power and the neutrino output: ^{235}U , ^{238}U , ^{239}Pu , and ^{241}Pu . The neutron-rich fission products of these isotopes undergo beta decays emitting antineutrinos [11]

$$S_{\bar{\nu},\text{tot}}(E) = \sum_i \text{FR}_i S_{\bar{\nu},i}(E), \quad (1)$$

with $S_{\bar{\nu},\text{tot}}(E)$ being the total $\bar{\nu}$ spectrum emitted by the reactor core, i representing the four main fuel isotopes, FR_i being the fission rate of isotope i and $S_{\bar{\nu},i}(E)$ the total $\bar{\nu}$ spectrum emitted after fission of isotope i , including all decay spectra of the daughter isotopes. There exist two different ways to predict the total $\bar{\nu}$ spectra: (1) The summation method [11–13] uses the databases to build up the β spectra $S_{\beta,i}(E)$ as the sum of the branch-level spectra of all daughter isotopes. This requires accurate knowledge of parameters like fission yields or branching ratios and of the β spectra involved. The conversion of the β spectra into the $\bar{\nu}$ spectra $S_{\bar{\nu},i}(E)$ can be performed on a branch-level with high accuracy [14,15]. (2) In the conversion method, the four β spectra $S_{\beta,i}(E)$ of the main fuel isotopes are measured directly—without knowledge of the branch-level processes. The conversion into the $\bar{\nu}_e$ spectra (see also [15]) is more difficult than in the summation technique and will be described later. Both methods of determining the antineutrino spectrum need the fission rates of the fuel isotopes as an input parameter, which can be calculated with reactor evolution codes like, e.g., MURE [16] or DRAGON [17].

The accuracy of the spectra obtained by the summation method suffers from incomplete data of the β spectra, and

there is an intense discussion about the influence of the pandemonium effect [18,19], of weak-magnetism corrections [15], and of the unknown shape of many contributing β spectra to the final calculated spectrum [20]. The direct measurement of $S_{\beta,i}(E)$ for the conversion technique has no dependence on these parameters. In the 1980s, measurements of the cumulative β spectra $S_{\beta,i}(E)$ of the fission products of three of the fuel isotopes (^{235}U , ^{239}Pu , ^{241}Pu) were performed with the BILL magnetic spectrometer at ILL in Grenoble [21–24]. From these β spectra, the particular $\bar{\nu}$ spectra were derived. The BILL spectra can act as benchmark for summation calculations, but, primarily, they are input for the conversion technique. However, as in the BILL measurement only thermal neutrons were used, the spectrum of ^{238}U was not measured. Consequently, until now, the determination of the total $\bar{\nu}$ spectrum from reactors is based on the BILL data for ^{235}U , ^{239}Pu , and ^{241}Pu , but has to rely on summation calculations for ^{238}U . This isotope contributes about 10% to the total $\bar{\nu}_e$ output of a PWR. At the scientific neutron source FRM II in Garching, we performed an experiment to determine—for the first time—the antineutrino spectrum of the fission products of ^{238}U using a fast neutron beam [25]. This Letter describes this experiment and the resulting β spectrum and presents a conversion into the $\bar{\nu}_e$ spectrum.

The ^{238}U experiment.—At the 20 MW scientific neutron source FRM II, the so-called converter facility alternatively provides both a fast and a thermal neutron beam [26]: Plates of highly enriched ^{235}U (~ 500 g) are situated at the inner rim of the moderator tank. A fast neutron spectrum is emitted in the fission processes of this converter, and various filters removed any thermal neutron content from this beam. The converter plates can also be removed from the moderator, resulting in a thermal neutron beam, without the need to change the experimental setup. Two identical target foils from natural uranium (99.3% ^{238}U , 0.7% ^{235}U) were irradiated, one by the thermal and one by the fast neutron beam. In the thermal beam measurement, only fission of ^{235}U was induced, whereas with the fast neutron beam mostly fission of ^{238}U occurred. Thus, it was possible to record β spectra of the fission products of ^{235}U and ^{238}U with the same setup. As described later, our ^{235}U measurement was normalized to the one from the BILL experiment [23] to minimize systematic uncertainties in the ^{238}U analysis.

Similar to the experiment by Carter in 1959 [27], the detector was a γ -suppressing β telescope consisting of two modules: (1) a spectroscopic module for full energy absorption, including a plastic scintillator and a photomultiplier, and (2) a multiwire chamber (MWC), placed in front of the entrance area of the scintillator. These two modules were operated in coincidence as, due to the low density of the counting gas (CF_4), the MWC was not sensitive to γ radiation also emitted from the target foil. This suppression of γ -induced events was determined with γ sources to be better than 99.5%.

For calibration purposes, a ^{207}Bi source and two target foils, one of polyvinylidene chloride and one of $^{\text{nat}}\text{In}$ were used. The ^{207}Bi source was installed in a way that allowed us to place the source either in a passive rest position or directly beneath the target foil. No exchange of a target foil was necessary and any calibration with the monoenergetic conversion lines of ^{207}Bi at ~ 1 MeV could be performed with the same condition as the uranium measurements before and after the calibration. The two calibration foils were consecutively activated by thermal neutrons, producing the beta emitters ^{116}In and ^{38}Cl with endpoints at ~ 3.3 and ~ 4.9 MeV, respectively, and placed at the position of the uranium target foils. The energy response was linear over the whole energy range, and an error of less than one percent on the energy calibration was reached.

The response function of the setup was determined by a GEANT4 simulation [28] and cross-checked at 1 MeV by a comparison of the simulation with the measured results of the ^{207}Bi spectrum. The resolution of the system was $8\% \cdot (1/\sqrt{E[\text{MeV}]})$ (FWHM).

To minimize the effects of not having reached radioactive equilibrium, data of the first 11 hours of irradiation was not included in the analysis (BILL experiment: 12 h). Only data from the following 42 h were used. The error due to off-equilibrium effects cannot be measured, but the calculations [11] show a difference of the $\bar{\nu}$ spectrum recorded after 12 h and 450 d irradiation time of less than 1% at 3 MeV, even decreasing towards higher energies. Thus, no error was included due to this effect in the final error budget of the data presented.

Background in fast neutron beam data.—The composition of the β spectrum recorded with a uranium target irradiated by fast neutrons was analyzed by background measurements. Figure 1 illustrates the results of this analysis. The main background to the uranium β spectrum was generated by diffuse background, which is the spectrum one obtains with the fast neutron beam on-line but

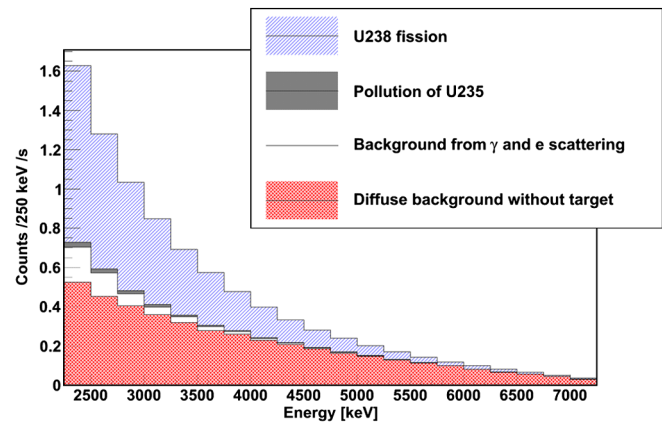


FIG. 1 (color online). Stacked plot of the different contributions to the β spectrum emitted by the uranium target foil under irradiation with fast neutrons. See text for explanation.

without any target foil. The events in the diffuse background mainly stem from (a) gammas and electrons present in the neutron beam that scatter off the material near the detector, leading to electrons reaching the detector, (b) captured neutrons producing unstable isotopes and high-energy gamma radiation, and (c) cosmic muons. This diffuse target-independent background had an intensity similar to the β spectrum from the fission products of ^{238}U but could be determined with high accuracy. In addition, scattered electrons and gammas converted into electrons at the target foils cause a target-dependent background. To determine this background in the uranium data, measurements with dummy targets of Pb and Ni were performed. With the help of GEANT4 simulations, the background spectra recorded with the dummy targets were used to deduce the background spectra present in the uranium data. Finally, as the uranium targets consisted of natural uranium, fission of ^{235}U contributed to the fast neutron beam spectrum. Because of the knowledge of the neutron beam spectrum, of the fission cross sections and the measurement of the ^{235}U in the thermal beam, this small contribution (2.6% in total) could be corrected for.

Background in thermal neutron beam data.—With removed converter, the γ and electron content in the neutron beam—and thus, the diffuse background—was significantly reduced. The background in the thermal beam was dominated by captures of neutrons by the material surrounding the detector, leading to electrons and gammas that were partly converted into electrons. This contribution, as well as the remaining background induced by gammas from the beam, could again be determined with the help of dummy-target measurements (Pb and Ni). The signal-to-background ratio was 6.5 at 4 MeV and better than 1 up to energies of 6.5 MeV.

Normalization to the BILL measurement.—The advantage of the experiment described herein was the possibility to normalize the data to the very accurate BILL measurement of ^{235}U . This is of importance as it greatly suppresses systematic uncertainties connected to, e.g., the unknown detector efficiency, unexpected energy dependence of the detector response function, and the barely known neutron-beam profile and intensity. A normalization function NF is defined bin-wise as the ratio of the β spectra of ^{235}U measured in the present experiment (U235) and the BILL experiment (BILL). NF is then applied to the ^{238}U spectrum

$$\text{NF} := \frac{\text{U235}}{\text{BILL}} = F_\gamma \frac{\text{U238}}{\text{U238}_{\text{final}}} \quad (2)$$

$$\Rightarrow \text{U238}_{\text{final}} = F_\gamma \text{U238} \frac{\text{BILL}}{\text{U235}}. \quad (3)$$

Herein, U238 is the β spectrum of the fission products of ^{238}U measured in the present experiment, and $\text{U238}_{\text{final}}$ is

the ^{238}U spectrum quoted as final result. As this normalization did not take into account the different number of fissions in the two target foils during irradiation, a factor F_γ (44.4 ± 0.3) had to be introduced. F_γ was determined by γ spectrometry of the irradiated foils after the β measurement by measuring the peak areas of selected γ lines emitted by the fission products with a high-resolution Ge spectrometer [29].

This normalization fully correlates the ^{238}U spectrum obtained in the present experiment with the BILL spectrum of ^{235}U [see Eq. (3)]. The final β spectrum of ^{238}U is given in Table I.

Error content.—The error ϵ quoted in the third column of Table I is the energy-dependent, bin-to-bin uncorrelated, combined statistical and systematic error. ϵ is dominated by the error introduced by the statistical subtraction of the diffuse background. At energies below ~ 3.5 MeV, systematic uncertainties in the background model enlarge this error by $\sim 1\%$. Furthermore, an error of $\sim 0.5\%$ is caused by the energy calibration. In addition to ϵ , there is an almost energy-independent uncertainty of the absolute normalization $\epsilon_{\text{norm}} \approx 2.8\%$, calculated as the quadratic sum of $\epsilon_{\text{exp,norm}}$ and $\epsilon_{\text{BILL}} \cdot \epsilon_{\text{exp,norm}}$ is due to inaccuracies of the γ spectrometry and different dead-time corrections for the two uranium measurements (2.1%) and ϵ_{BILL} is the error of absolute scale in the BILL experiment ($\sim 1.8\%$). The latter

TABLE I. The final β spectrum of the fission products of ^{238}U . N_β is the spectrum given in units of betas per fission and MeV. The relative combined statistical and systematic error ϵ is given in column 3. Column 4 gives the error $\epsilon_{\text{exp,norm}}$ which is due to uncertainties in the absolute normalization of the present experiment. The last column shows the error ϵ_{BILL} on the absolute rate in the BILL measurement. All errors given at 68% confidence level (1σ).

E [keV]	N_β $\left[\frac{\text{betas}}{\text{fission} \cdot \text{MeV}} \right]$	ϵ [%]	$\epsilon_{\text{exp,norm}}$ [%]	ϵ_{BILL} [%]
2250–2500	1.032	3.2	2.1	1.7
2500–2750	8.302×10^{-1}	3.0	2.1	1.7
2750–3000	6.922×10^{-1}	2.4	2.1	1.7
3000–3250	5.698×10^{-1}	2.3	2.1	1.7
3250–3500	4.533×10^{-1}	2.4	2.1	1.7
3500–3750	3.740×10^{-1}	2.4	2.1	1.7
3750–4000	2.807×10^{-1}	2.7	2.1	1.7
4000–4250	2.279×10^{-1}	2.9	2.1	1.7
4250–4500	1.725×10^{-1}	3.5	2.1	1.8
4500–4750	1.343×10^{-1}	3.9	2.1	1.8
4750–5000	1.084×10^{-1}	4.5	2.1	1.8
5000–5250	7.891×10^{-2}	5.5	2.1	1.8
5250–5500	5.831×10^{-2}	6.8	2.1	1.8
5500–5750	4.137×10^{-2}	9.7	2.1	1.8
5750–6000	2.909×10^{-2}	11.7	2.1	1.8
6000–6250	2.765×10^{-2}	11.1	2.1	1.8
6250–6500	2.248×10^{-2}	12.7	2.1	1.8
6500–6750	1.296×10^{-2}	18.9	2.1	1.9
6750–7000	7.078×10^{-3}	28.1	2.1	1.9

adds a slight energy dependence to the absolute normalization error and is listed separately to be able to disentangle the different contributions if using another normalization than the BILL spectrum would be desired.

A lower threshold of 2250 keV had to be set to avoid an additional, in the present experiment indeterminable, background of the 2.2 MeV gammas from the capture of neutrons by the scintillation detector itself [25].

Conversion to $\bar{\nu}$ spectrum.—Finally, the obtained β spectrum was converted into an $\bar{\nu}_e$ spectrum. A standard conversion procedure applied to the BILL spectra is based on introducing hypothetical beta branches which are used to fit the experimental β spectra and are converted on the branch level. From these, the total neutrino spectrum is built up. Because of low statistics in the high-energy regime, this technique could not be used for the present ^{238}U measurement. Instead, an empirical method proposed in [23], was chosen: Being the sum spectra of many decaying isotopes, the cumulative β and $\bar{\nu}_e$ spectra emitted are very similar. The main differences can be corrected by shifting the electron spectrum by 511 keV (the mass of the electron) and additional 50 keV to account for an average Coulomb attraction of nucleus and electron [13]. All further corrections—which are of the order of 5% [25]—can be described by a factor $k(E)$

$$N_{\bar{\nu}}(E) = N_{\beta}(E - 511 \text{ keV} - 50 \text{ keV})k(E). \quad (4)$$

$k(E)$ was extracted from the BILL experiment and the summation method: By inserting in Eq. (4), e.g., the measured ^{235}U β spectrum from the BILL experiment and the $\bar{\nu}$ spectrum obtained from this β spectrum by the standard conversion procedure, one can extract $k(E)$. This determination of $k(E)$ can be performed with any known pair of (measured or predicted) β and corresponding $\bar{\nu}$ spectra. The factor $k(E)$ extracted from the ^{235}U spectra of the BILL experiment and the one obtained from the summation calculations of ^{238}U [11] agrees within $\sim 1\%$. Thus, the same $k(E)$ is valid for both ^{235}U and ^{238}U . In the present Letter, the spectra calculated in [11] were used to extract $k(E)$. Note that even though the summation predictions result in relatively large errors of the final spectra—partly due to unknown β spectra—the factor $k(E)$ extracted from these is more reliable as it is only sensitive to errors in the branch-level conversion. A detailed discussion of the conversion will be performed in an upcoming paper [30]. Table II gives the final antineutrino spectrum of the fission products of ^{238}U . The conversion introduced an additional 2% to the error of the absolute normalization, which was quadratically added to the uncertainty ($\epsilon_{\text{exp,norm}}$, ϵ_{BILL}) described above.

Discussion.—The experimental $\bar{\nu}_e$ spectrum of the fission products of ^{238}U reveals a slight spectral distortion in the currently assumed shape. Figure 2 plots the ratio of the experimental spectrum and the results of the summation

TABLE II. The $\bar{\nu}_e$ spectrum of the fission products of ^{238}U in units of $\bar{\nu}_e$ per fission and MeV. The energies E represent the center of the 250 keV wide bins. ϵ is the combined inaccuracy of all error sources (stat. + syst.), except for the global absolute normalization uncertainty ϵ_{norm} which is quoted in the last column and includes the uncertainty of the BILL measurement. All errors given at 68% confidence level (1σ).

E [keV]	$N_{\bar{\nu}} \left[\frac{\bar{\nu}}{\text{fission} \cdot \text{MeV}} \right]$	ϵ [%]	ϵ_{norm} [%]
3000	9.586×10^{-1}	3.5	3.3
3250	7.952×10^{-1}	3.1	3.3
3500	6.603×10^{-1}	2.6	3.3
3750	5.406×10^{-1}	2.6	3.3
4000	4.433×10^{-1}	2.6	3.3
4250	3.498×10^{-1}	2.8	3.3
4500	2.787×10^{-1}	2.9	3.3
4750	2.171×10^{-1}	3.3	3.3
5000	1.700×10^{-1}	3.7	3.4
5250	1.341×10^{-1}	4.1	3.4
5500	1.032×10^{-1}	5.0	3.4
5750	7.737×10^{-2}	5.9	3.4
6000	5.618×10^{-2}	7.6	3.4
6250	3.973×10^{-2}	10.6	3.4
6500	3.048×10^{-2}	12.6	3.4
6750	2.085×10^{-2}	11.7	3.4
7000	2.093×10^{-2}	14.1	3.4
7250	1.139×10^{-2}	21.9	3.4
7500	7.132×10^{-3}	30.0	3.4

method applied in [11]. This was chosen, as it is the one in use in the actual reactor neutrino disappearance experiments. Furthermore, the reduced mean cross section per fission $\langle \sigma_f \rangle$ for the inverse beta decay (IBD) can be calculated

$$\begin{aligned} \langle \sigma_{f,\text{U238}} \rangle_{\text{red}} &= \int_{2.875\text{MeV}}^{7.625\text{MeV}} S_{\text{U238}}(E) \sigma_{\text{IBD}} dE \\ &= 8.51 \times 10^{-43} \pm 9.07 \times 10^{-45} (\text{stat.} + \text{syst.}) \\ &\pm 2.80 \times 10^{-44} (\text{norm.}) \frac{\text{cm}^2}{\text{fiss.}}, \end{aligned} \quad (5)$$

with $S_{\text{U238}}(E)$ being the $\bar{\nu}_e$ spectrum of the fission products of ^{238}U and σ_{IBD} the cross section for the inverse beta decay. The label “red” indicates that the mean cross section could only be calculated for the reduced energy range covered by the experiment. The ratio between the reduced mean cross section determined by experiment (Expt.) and summation method (Sum.) from [11] is

$$\begin{aligned} \frac{\langle \sigma_{f,\text{U238}} \rangle_{\text{red}}(\text{Expt.})}{\langle \sigma_{f,\text{U238}} \rangle_{\text{red}}(\text{Sum.})} &= 0.97 \pm 0.08 (\text{syst.} + \text{stat.}) \\ &\pm 0.03 (\text{norm.}) \end{aligned} \quad (6)$$

The error of 0.08 (sys. + stat.) comprises the uncertainties of the experiment and summation approach and is

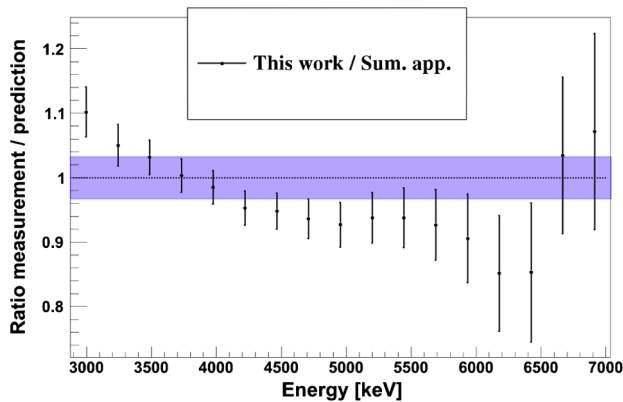


FIG. 2 (color online). The ratio of the measured $\bar{\nu}_e$ spectrum of ^{238}U and the result of the summation approach [11]. The shaded band represents the error of 3.3% of the absolute normalization and the error bars illustrate the combined syst. and stat. error of the experiment only. No error was included from the summation calculations. Note a spectral distortion of $\sim 10\%$.

highly dominated by the error of the theoretical calculations. The uncertainty of 0.03 represents the error of the absolute scale of the experiment. Thus, the ratio is compatible with 1, and the experiment confirms the currently assumed value for the mean cross section per fission.

Conclusion.— For the first time, the $\bar{\nu}_e$ spectrum of the fission products of ^{238}U was determined experimentally. The experiment covers a range from 2.875 to 7.625 MeV and has a relative energy-dependent error of 3.5% at 3 MeV, 7.6% at 6 MeV and $\gtrsim 14\%$ at energies $\gtrsim 7$ MeV (68% C.L.). The uncertainty of the absolute scale of $\sim 3.3\%$ is almost energy independent. Thus, the spectrum has an error significantly lower than the one quoted for the summation calculations [11]: At energies around 4 MeV, which is most interesting for neutrino oscillation experiments, the uncertainty of the measurement is about a factor of 2 lower. The results reveal spectral distortions of $\sim 10\%$ in the currently used spectrum achieved with summation calculations. The value of the mean cross section per fission in the energy range covered by the experiment (2.875–7.625 MeV) matches the one determined by the summation approach. With this spectrum, it is now possible to determine reactor antineutrino spectra without the use of theoretical spectra of the contributing fission isotopes.

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