Rapid Communications

Spectral anomaly of reactor antineutrinos based on theoretical energy spectra

Tadashi Yoshida,[1,*] Takahiro Tachibana,[2] Shin Okumura,[1] and Satoshi Chiba[1]

[1] *Laboratory for Advanced Nuclear Energy, Institute of Innovative Research, Tokyo Institute of Technology, Tokyo 152-8550, Japan*

[2] *Waseda Research Institute for Science and Engineering, Waseda University, 3-4-1 Ookubo, Shinjyuk-ku, Tokyo 169-8555, Japan*

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There exists a persistent local deviation in the reactor antineutrino spectra between those measured directly at power reactors through the $\bar{\nu}_e + p \to n + e^+$ reaction and the currently used reference data. In order to interpret the origin of this *spectral anomaly* with its peak at 6 MeV, the gross theory of β decay was applied to calculate the antineutrino spectra for about 500 fission products (FPs) where the measured spin parities were introduced into calculations for 178 odd($-N$)–odd($-Z$) FPs in the framework of the gross theory and the recently measured ground-to-ground transition rate for two FPs dominating the spectrum shaping above 5 MeV. The overall consistency between the calculated and the reference spectra is reasonable, and our result supports the direct measurements of the $\bar{\nu}_e$ spectra at power reactors. The anomaly is suggested to originate first with the plutonium isotopes ^{239}Pu and ^{241}Pu, which are closely followed by ^{235}U.

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The Institut Laue-Langevin (ILL) energy spectra of electron-antineutrinos ($\bar{\nu}_e$) emitted from the aggregate fission products (FPs) of ^{235}U, ^{239}Pu, and ^{241}Pu have long been applied as the standards in the fields of neutrino-related science and technology. They were obtained through conversion from the measured electron spectra emitted in fissile samples under neutron irradiation in the high-flux reactor at ILL [1–4]. Later, Mueller *et al.* [5] and Huber [6] refined the conversion procedure. Their revised sets of spectra are widely utilized now. In addition the ^{238}U spectrum was obtained by use of the same methodology as the ILL spectra at the neutron source FRM II in Garchin [7]. Hereafter we call all the lepton spectra mentioned above the BILL spectra because all are based on the measurement using the BILL electron spectrometer [8].

Recently, however, direct measurements of the $\bar{\nu}_e$ spectra from operating power reactors became possible utilizing the $\bar{\nu}_e + p \to n + e^+$ reaction inverse β decay (IBD). In their studies, the Double Chooz [9], RENO [10], Daya Bay [11], and NEOS [12] Collaborations found two types of important deviations from the predicted spectra composed from the ILL ones mentioned above. They are called the *flux* and the *spectral anomalies*, respectively. The former, an integral bias of $\sim$7% from the ILL spectra, initiated the sterile neutrino controversy [13]. Here we deal with the latter, a persistent local deviation from the predicted spectra more than 10% peaking at 6 MeV with a width of about 1 MeV which gave rise to arguments by Huber [14], Buck *et al.* [15], Sonzogni *et al.* [16], and Hayes *et al.* [17].

The summation method was first introduced by several authors around 1980 [18–20] for obtaining the reactor $\bar{\nu}_e$ spectra. Later, Fallot *et al.* [21] and Sonzogni and co-workers [22,23] applied this method making extensive use of the up-to-date experimental decay data of individual FPs. The present theory-based way of the summation calculation is independent of and complementary to the works of these authors.

One of the present authors demonstrated that the gross theory of β decay [24–26] works remarkably well for describing the aggregate decay behavior of a large number of FPs specifying the reactor decay heat [27]. It revealed the real existence of the *pandemonium* problem first addressed on the basis of computer simulation [28]. The calculation reproduced the measured decay heat almost within $\pm5\%$, and the typical experimental error attached to the measured values is also about 5% [29]. Furthermore, it had been tested [30,31] against a lot of decay heat and total absorption γ-ray spectroscopy (TAGS) [32,33] experiments. It is noteworthy that the gross-theory mean β- and the γ-ray energies had been adopted for most of the short-lived FPs in the Japanese evaluated nuclear data library JENDL and in its U.S. counterpart ENDF/B-IV for decades until recently when they were gradually being replaced by TAGS data [34]. Anyway, adoption of the gross-theory mean energies is the reason why both JENDL and ENDF/B-IV reproduce the direct decay-heat measurements much better than the European data files which intentionally exclude theoretical predictions as is seen in Figs. 1–4 in Ref. [29]. From the above consideration we suppose that the error associated with the gross theory prediction for the aggregate FP nuclide is 5%.

We start with a calculation of the concentrations of individual FPs making use of the coupled equations,

$$\frac{dN_i(t)}{dt} = -\lambda_i N_i(t) + \sum_j f_{j\to i}\lambda_j N_j(t) + \sum_k y_{ki} F_k, \quad (1)$$

where $N_i(t)$ is the concentration of the ith FP at time t. The symbols λ_i, $f_{j\to i}$, y_{ki}, and F_k are the decay constant, the branching ratio, the independent yield of each FP, and the

*tyoshida@nr.titech.ac.jp

fission rate of the kth fissile, respectively. The actual values for these parameters were taken from JENDL FP decay data file 2011 and JENDL fission yield data file 2011 (hereafter, JENDL) [35] and those from JEFF-3.1 are also used as a reference [36]. For the JENDL ^{235}U yield we used the most recent update from the JAEA Nuclear Data Center (April 19, 2017). By summing up all the contributions, we can calculate the $\bar{\nu}_e$ spectra as

$$I_{\bar{\nu}_e}(E_{\bar{\nu}_e}) = \sum_i N_i \lambda_i I_i^{\bar{\nu}_e}(E_{\bar{\nu}_e}). \tag{2}$$

Here N_i is the solution of Eq. (1) after a certain neutron irradiation whose time duration is typical for each experiment (10–50 h), and $E_{\bar{\nu}_e}$ stands for the energy of antineutrinos. It should be noted that many authors argue the *spectral anomaly* in terms of the so-called prompt energy E_{prompt} instead of $E_{\bar{\nu}_e}$. These two are connected approximately through a relation $E_{\bar{\nu}_e} \approx E_{\text{prompt}} + 0.8$ MeV.

In terms of the gross theory, the $\bar{\nu}_e$ spectrum of the ith nuclide is written as

$$I_i^{\bar{\nu}_e}(E_{\bar{\nu}_e}) = \frac{1}{D}\int_0^{Q_\beta - E_{\bar{\nu}_e}} \left[\sum_\Omega C_\Omega |M_\Omega(E_{\text{ex}})|^2 F(Z,E) S_\Omega(Z,p)\right] E_{\bar{\nu}_e}^2 pE\, dE_{\text{ex}}, \tag{3}$$

where D is the normalization denominator introduced so as to give one antineutrino per decay, namely, $\int_0^{Q_\beta} I_i^{\bar{\nu}_e}(E_{\bar{\nu}_e}) dE_{\bar{\nu}_e} = 1$. The symbol C_Ω stands for the coupling constant multiplied by the multiplicity and a factor depending on the forbiddenness of the type-Ω β transition which covers the Fermi, the Gamow-Teller, and the first-forbidden transitions of ranks 0, -1, and -2. The variable E_{ex} is the excitation energy of the daughter nucleus. Here the electron momentum p and energy E should be written in terms of $E_{\bar{\nu}_e}$, E_{ex}, and several physical constants. The symbols F and S_Ω indicate the Fermi function and the shape factor, respectively, of the Ω-type transition. The strength function $|M_\Omega(E_{\text{ex}})|^2$ is essentially the absolute square of the transition matrix element multiplied by the final level densities expressed as a continuous function of the excitation energy of the daughter nucleus E_{ex}. This important quantity is calculated here based on the gross approximation first introduced by Takahashi and Yamada [24], Koyama *et al.* [25], and Takahashi [26] and later improved by several authors [37,38]. We call this the second generation of the gross theory (GT2) hereafter. We have already mentioned that the gross theory reproduces the experimental data within an error of $\pm 5\%$ in the decay-heat summation calculation. Also, as for the β-decay half-life, we can see quite good agreements between GT2 calculations and experimental data for the neutron-rich nuclides [39,40].

In the practical calculation of $\bar{\nu}_e$ spectra, odd-odd nuclides occupy 60% at 8 MeV and almost 90% at 10 MeV as far as the conventional GT2 being applied [41]. It was known, however, that the gross theory was not good at predicting the half-lives of odd-odd parent nuclides. In order to remedy this, Nakata, Tachibana, and Yamada (NTY) constructed a framework which introduces measured spin parities of the individual odd-odd nuclides [42]. The NTY treatment makes full use of the definite 0^+ and 2^+ structures of the lowest two states of their even-even daughters. As far as the β transitions from an odd-odd nucleus to the ground and to the first excited states in its even-even daughter are concerned, the transition types are clearly classified by the selection rules. In the conventional GT2 model, all the transition types are included on average. In the GT2 + NTY model for odd-odd nuclides, the transition type which satisfies the selection rule is enhanced, and all the other transitions are decreased or cut off in accordance with the selection rules.

Among the four U and Pu isotopes we start with the $\bar{\nu}_e$ spectrum from the ^{235}U sample irradiated at ILL which provides us with the data for the widest range of energy. The blue (gray) dot-dashed curve in Fig. 1 shows the ^{235}U $\bar{\nu}_e$ spectrum calculated fully based on GT2 which is essentially the same as reported previously by the present authors [41]. This is compared with three versions of the ILL spectra, namely, those converted by the original authors (Schreckenbach *et al.*) [2], by Mueller *et al.* [5], and by Huber [6]. Although the overall agreement is not bad, the calculation divided by experimental (C-over-E) value varies from 0.85 (6 to 8 MeV) to 1.25 (2 to 4 MeV). Furthermore, overestimation becomes conspicuous above 8 MeV.

In order to improve the spectrum calculation, the NTY treatment was applied for 177 odd-odd FPs for which definite spin parities are assigned in JENDL [35]. In addition, it was also applied to ^{98m}Y, one of the most important contributors in the 7–9-MeV region of the spectra. Singh and Hu assigned spin 4 or 5 to this isomer, but its parity is unknown [43]. We assumed here $J^\pi = 5^+$. Other possible selections (4^+, 4^-,

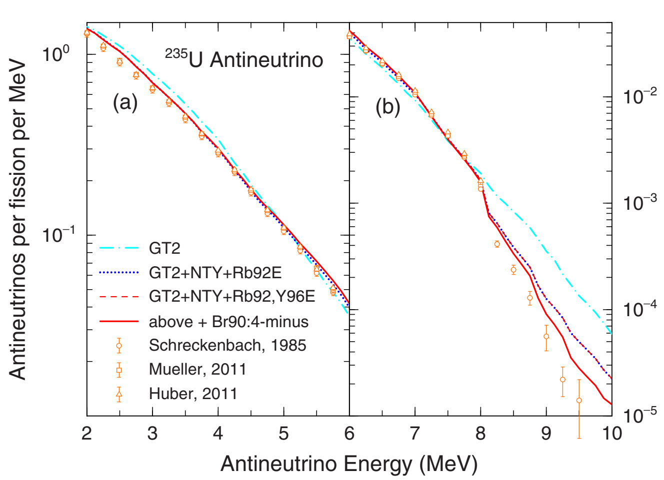

FIG. 1. Calculated antineutrino spectra from ^{235}U before (dot-dashed curve) and after the NTY treatment and the enhancement of ground-to-ground transition rates (see the text). The thick solid curve shows a calculation where a spin parity of 4^- is assumed for ^{90}Br. In the legend, E stands for ground-to-ground transition enhanced. Panel (a) refers to the left axis, and panel (b) refers to the right axis.

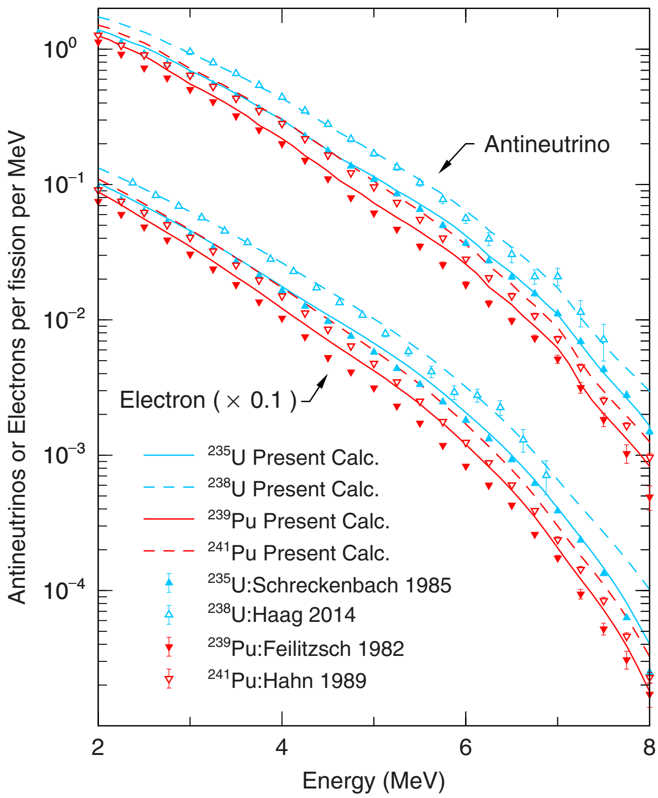

FIG. 2. Calculated and experimental lepton spectra of U and Pu isotopes. All the curves for antineutrinos and for electrons are the GT2 calculations with the NTY treatment for 178 FPs and the ground-to-ground enhancement for ^{92}Rb and ^{96}Y (see the text). Conversion of experimental data from electron to antineutrino spectra is due to Mueller *et al.* [5] for ^{235}U and Pu and to Haag *et al.* [7] for ^{238}U. The electron spectra are multiplied by 0.1 for concise presentation.

or 5$^-$) do not change the calculated spectrum appreciably. In addition to this, we enhanced the ground-to-ground transition rate of ^{92}Rb from 40% (as calculated by NTY) to 91% in the framework of GT2 because this strong ground-state first-forbidden transition (87.5% after Ref. [44] or 91% [45]) is not well reproduced even by the NTY treatment. The difference between 87.5% and 91% is small in terms of the calculated $\bar{\nu}_e$ spectrum. The same enhancement was performed for ^{96}Y (from 49% to 95.5% [45]). In this way, minimum but important experimental information was incorporated into our theoretical calculation for influential odd-odd FPs.

The results are shown as the dotted and the dashed curves in Fig. 1. These two are consistent with the experimental data in the whole energy range except less than 4 MeV. This discrepancy in the low-energy region is a problem left to be solved. A possible reason is that the gross theory is hard to be applied to low-Q_β decays from its own nature. In reality, the theory was applied only to decays with Q_β values larger than 5 MeV for the successful reactor decay-heat calculations [27]. In this point of view, we do not try to pursue further the origin of this discrepancy below 4 MeV at the present stage of the investigation. The remaining overestimation above 8 MeV is expected to be mitigated by further application of the NTY

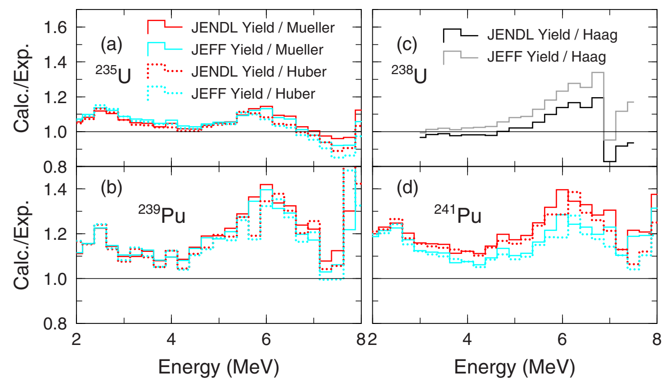

FIG. 3. Ratio of calculated-to-experimental values of the antineutrino spectra. The experimental data are identified by the name of the converter from the electron to the $\bar{\nu}_e$ spectrum. Both results based on the JENDL and the JEFF-3.1 fission yields are shown here. The legend in panel (a) applies also to panels (b) and (d).

treatment, for example, to ^{90}Br, the biggest contributor here having a large Q_β value of 10.4 MeV. The solid curve is a result where 4$^-$ is assigned to this nuclide after Nordheim's rule [46]. This trial does not, however, affect the following argument dealing with the energy range below 8 MeV.Then we check the applicability of our methodology for fissioning nuclides other than ^{235}U. As far as the $\bar{\nu}_e$ detection based on the IBD reaction is concerned, antineutrinos with energies higher than 8 MeV play only a negligible role. Then we concentrate on the energy range less than 8 MeV hereafter. Figure 2 shows the comparison between the calculated and the BILL lepton spectra from ^{235,238}U and ^{239,241}Pu. It is noteworthy that the present calculation reproduces well both the overall shapes of the spectra and the large variations reaching a factor of 3 depending on the fissioning nuclide. The local deviations in each nuclide spectrum cannot be overlooked. Next, we will focus our discussion on these deviations. A persistent overestimation is seen especially for the plutonium isotopes (^{239}Pu and ^{241}Pu) between 5 and 7 MeV both in the electron and in the antineutrino spectra. The peak position of this deviation from the BILL spectra for electrons seems to shift toward the lower-energy side compared to the $\bar{\nu}_e$ spectra. This shift may correspond to the electron rest mass of ∼0.5 MeV. Including this point, the overall consistency between the $\bar{\nu}_e$ and the electron spectra is acceptable.

Figure 3 shows the same results as Fig. 2 in terms of the linear-scale C-over-E values. From here the *spectral anomaly* mentioned at the beginning of this Rapid Communication comes into sight. We see bumps with a peak at 6 MeV for ^{235}U and two plutonium isotopes. These bumps suggest that the BILL spectral values, which appear as the denominator in these figures, are too small around 6 MeV. In this way, the present calculations support the recent IBD measurement which is more than 10% larger than the BILL prediction at 6 MeV. It should be noted that the nuclidewise variation of the calculated spectra in Fig. 3 comes solely from the differences in fission yields of each fissile providing the weight of each spectrum in the summation method. The spectrum of each FP is calculated by the GT2-NTY procedure with the special

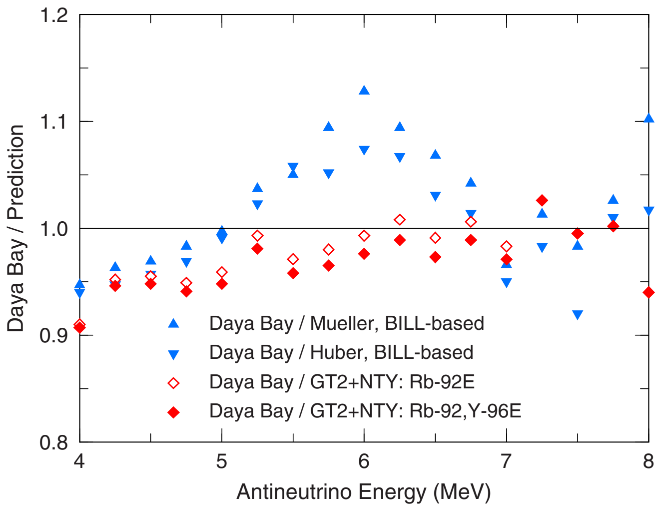

FIG. 4. Comparison of the BILL-based and the present predictions of the Daya Bay IBD reaction rate [11]. The upward triangle was calculated from the conversion of Mueller *et al.* [5] of the BILL data for ^{235}U and ^{239,241}Pu with the data and conversion of Haag *et al.* [7] for ^{238}U. The downward triangle is from Huber's conversion [6] and the ^{238}U data of Mueller *et al.* [5]. In obtaining the reaction rate, the IBD cross section was taken from Ref. [47]. As for symbol E, see the caption for Fig. 1.

treatment for ^{92}Rb and ^{96}Y. The applicability of this treatment has been checked for the case of ^{92}Rb by direct comparison with the experimental spectrum of Zakari-Issoufou *et al.* [44].

Figure 4 compares the two types of $\bar{\nu}_e$ spectra predictions, the BILL based and the GT2 based, in terms of ratios of the IBD reaction rates measured at Daya Bay [11] to them. The bump seen in the triangles (the BILL prediction) corresponds to the *spectral anomaly* mentioned at the beginning of this Rapid Communication. On the contrary, the diamonds (the present prediction) exhibit no bump. The height of the bump is 13% at 6 MeV at most from the horizontal axis. It reaches 16% when measured from the average of the present calculation (solid diamonds). We can break down this 16% into the fissilewise contributions using the Daya Bay fission fractions ^{235}U: ^{238}U: ^{239}Pu: ^{241}Pu $= 58:8:29:5$ [11] and the peak height shown in Fig. 3. As a result, ^{235}U, ^{239}Pu, and ^{241}Pu are responsible for this deviation by 7%, 8%, and 1%, respectively. If we ignore the difference between 13% (measured from the horizontal axis) and 16% (from the present calculation average), these values are regarded as the fractional contributions to the anomaly from these three fissioning nuclides. We withhold here drawing any conclusion about the role of ^{238}U in causing the *anomaly*. One reason is that the ^{238}U experimental data behave in a strange way above 5.5 MeV as we see in Fig. 2 and another is the fact that the ^{238}U data based on the BILL spectrum is not yet widely used for applications. Anyway, the ambiguity due to ^{238}U seems to be a few percent as far as we judge from its small fission fraction ($\sim$8%) and the deviations seen in Fig. 3. It is worth noting that the deviations discussed here are much larger than 5%, almost within which the gross theory reproduces the measured FP decay heat as mentioned at the beginning of this Rapid Communication. This seems to support the reliability of the argument here.

The present calculation, which exhibits no bump in Fig. 4, seems to support the recent IBD measurement against the currently prevailing BILL spectra and suggests that the plutonium isotopes ^{239}Pu and ^{241}Pu are primarily responsible for more than a half of the reactor spectral anomaly closely followed by ^{235}U. This conclusion is consistent with the recent argument by Huber [14] that the two Pu isotopes are disfavored as the sole source of the anomaly. Antineutrino spectrum measurements are underway or planned with the high-flux reactors at Grenoble (the STEREO Collaboration) [48], Oak Ridge (the PROSPECT Collaboration) [49], and at Mol (the SoLid Collaboration) [50]. The results from these pure-uranium-fueled reactors are expected to make these things clearer in the near future.

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