Energy Dependence of Prompt Fission Neutron Multiplicity in the ^{239}Pu(n, f) Reaction

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P. Marini[a,b,*], J. Taieb[a,c], D. Neudecker[d], G. Bélier[a,c], A. Chatillon[a,c], D. Etasse[e], B. Laurent[a,c], P. Morfouace[a,c], B. Morillon[a], M. Devlin[d], J. A. Gomez[d], R. C. Haight[d], K. J. Kelly[d], J. M. O'Donnell[d]

[a] *CEA, DAM, DIF, F-91297 Arpajon, France*
[b] *LP2I Bordeaux, UMR5797, Université de Bordeaux, CNRS, F-33170, Gradignan, France*
[c] *Université Paris-Saclay, CEA, LMCE, 91680 Bruyères-le-Châtel, France*
[d] *Los Alamos National Laboratory, Los Alamos, NM-87545, USA*
[e] *Normandie Univ., ENSICAEN, UNICAEN, CNRS/IN2P3, LPC Caen, 14000 Caen, France*

Abstract

Accurate multiplicities of prompt fission neutrons emitted in neutron-induced fission on a large energy range are essential for fundamental and applied nuclear physics. Measuring them to high precision for radioactive fissioning nuclides remains, however, an experimental challenge. In this work, the average prompt-neutron multiplicity emitted in the ^{239}Pu (n, f) reaction was extracted as a function of the incident-neutron energy, over the range 1-700 MeV, with a novel technique, which allowed to minimize and correct for the main sources of bias and thus achieve unprecedented precision.

At low energies, our data validate for the first time the ENDF/B-VIII.0 nuclear data evaluation with an independent measurement and reduce the evaluated uncertainty by up to 60%. This work opens up the possibility of precisely measuring prompt fission neutron multiplicities on highly radioactive nuclei relevant for an essential component of energy production world-wide.

Despite the discovery of nuclear fission being 80 years old, a full understanding of this rich quantum phenomenon is still a challenge for experimentalists and theoreticians. Parallel efforts [1–3], pursued worldwide, carry the promise of a renewed

*Corresponding author
Email address: paola.marini@cnrs.fr (P. Marini)

understanding of this complex phenomenon, and support the development of modern nuclear technologies for energy production effectively complying with the most recent requirements on safety, sustainability, economic competitiveness and proliferation resistance. From an experimental point of view, the most stringent constraints to theoretical models are expected to come from very precise measurements of observables over large energy ranges, as well as from the simultaneous measurement of several observables highlighting their possible correlations. Among them, the number of prompt fission neutrons and their kinetic energy distributions provide valuable information on the amount of excitation energy of the heated fissioning system transferred to the primary fragments. Moreover, these data, for the fissile ^{235}U and ^{239}Pu isotopes and the fertile ^{238}U nuclide, are vital inputs to calculate next-generation nuclear reactor neutronics, which affect projections of the criticality, efficiency, safety, and lifetime of such systems. From a theoretical point of view, a model able to describe the fission process with the requested accuracy is still lacking, therefore nuclear data applications rely, to a large extent, on evaluated data, such as ENDF/B-VIII.0 and JEFF3.3 [2, 3].

Several experimental and evaluation works have been dedicated since the '60-'70s to produce coherent and precise data and evaluations for the average prompt fission neutron multiplicity ($\overline{\nu}_p$) emitted in the neutron-induced fission of ^{239}Pu in the MeV range [4–25]. The importance of these data lies in the fact that the sustainability of the nuclear fission chain reaction in a reactor core, the so-called criticality k_{eff}, depends nearly linearly on the $\overline{\nu}_p$ of the fissioning nuclide [26] and often has the highest sensitivity to the $\overline{\nu}_p$ of the main fuel [27]. The precise measurement of $\overline{\nu}_p$ for a highly radioactive nuclide, as ^{239}Pu, is, however, an experimental challenge as it requires an unambiguous identification of fission events from a very intense α-decay background. Moreover, neutrons need to be detected with good efficiency and discriminated from γ rays emitted in fission. The current widely accepted reference measurement of J. Fréhaut et al. [18], carried out in the 1970's, provides the data reported to be most precise in the incident-neutron energy region between 1 and 30 MeV, with reported uncertainties as low as 0.5% below 15 MeV. Other experimental data [4–25], although with larger uncertainties, are all in good agreement with this measurement. The large majority and the most precise of these measurements [10, 15–22, 24] were realized

detecting neutrons in coincidence with fission events with a close-to-4π scintillator detector tank. Other measurements exploited either proportional counters inside paraffin blocks [5, 7–9, 11–14, 23] or the surrogate-reaction technique [25].

Resulting libraries are then validated with respect to integral experiments such as, e.g., $k_{\rm eff}$ experiments [28] that model the behavior of reactor cores on a small scale. This validation step tests the reliability of entire libraries for applications. ENDF/B-VIII.0 ^{239}Pu (n,f) $\overline{\nu}_p$ were obtained from existing experimental data, but the evaluated data had to be adjusted [2] such that simulated and experimental $k_{\rm eff}$ were in reasonable agreement for application calculations. To illustrate this point, in Fig.1 we show the relative difference between existing experimental and ENDF/B-VIII.0 $\overline{\nu}_p$ values in the fast neutron energy region, with the data normalized to the current ENDF/B-VIII.0 value of ^{252}Cf $\overline{\nu}_p$ [29] and not including its uncertainty. For a more readable figure, the uncertainty on the ENDF/B-VIII.0 values was not propagated in the relative difference and is shown as shaded region around the zero value. Discrepancies as high as 2% below 8 MeV are observed, with data systematically lower than the most recent ENDF/B-VIII.0 evaluation. A different trend and differences up to 1% are also observed for the JEFF3.3 evaluation. As a comparison, it should be kept in mind that a change in $\overline{\nu}_p$ by 0.1% in an energy range as small as 100 keV can modify the computed criticality by about 100 pcm, which is about one third of the range between a controlled and an uncontrolled Pu critical assembly [30, 31].

In this letter we report on high precision experimental data, obtained with a different and novel technique. They validate, below 5 MeV, the ENDF/B-VIII.0 evaluation with an independent measurement, highlight potential shortcomings in existing data, and reduce the evaluated uncertainty of up to 60% while extending the range of the studied energies from 30 to 700 MeV. The method, used here for ^{239}Pu (n, f) $\overline{\nu}_p$ measurement for the first time, consists of detecting neutrons with an ensemble of 17.78 cm-diameter liquid scintillator detectors, located on a half-sphere at about 1 meter distance from a fission detector assuring a good energy resolution for the emitted neutrons. Prompt fission neutron spectra (PFNS) are then measured, as a function of the incident-neutron energy, with a double time-of-flight technique [32]. Values of $\overline{\nu}_p$ are finally extracted from the integration of PFNS. As opposed to tank experiments [10, 15–22, 24], these

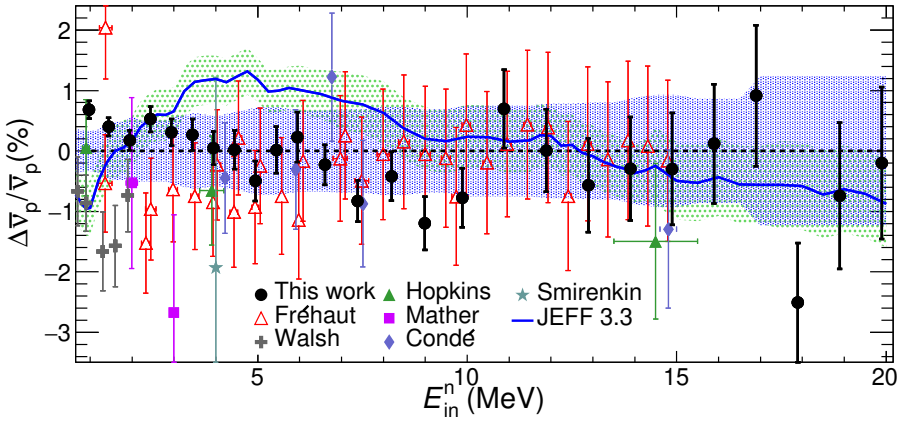

Figure 1: (Color online) Relative difference between experimental and ENDF/B-VIII.0 $\overline{\nu}_p$ values from this work and some previous experiments [10, 13, 15–18]. The difference with the JEFF3.3 evaluation [3] is also shown.

kinds of measurements suffer from statistics limitations, due to the limited detector angular coverage, and from the presence of a background due to neutron scattering on surrounding materials. However, they present two main advantages. First, while scintillator tanks are, at most, roughly segmented, the high segmentation of the neutron detector array allows to measure the neutron angular distribution, and therefore to precisely correct for the contribution of regions not covered by the detector. Second, PFNS are precisely measured and an energy-dependent efficiency curve, typically determined with respect to a ^{252}Cf source, can be used. This is not the case in tank experiments, where only the difference in the mean energy of the ^{252}Cf and ^{239}Pu PFNS could be accounted for, based on an empirical parametrization [33], insufficient by today's standards. These two effects lead to systematic biases in the existing $\overline{\nu}_p$ measurements not properly corrected for, thus enlarging further the level of uncertainty on $\overline{\nu}_p$. On the contrary, in our experiment the availability of a high-intensity, pulsed and well-collimated white neutron source, and the described novel technique allowed for the first time to effectively minimize and estimate the sources of possible bias while collecting high counting statistics and providing an independent measurement.

The experiment was performed at the Weapons Nuclear Research facility [34, 35] of Los Alamos Neutron Science Center at the Los Alamos National Laboratory. The

neutron beam was produced by spallation and bombarded a high-purity ^{239}Pu target after a flight path of about 21.5 m. A newly-developed, high-efficiency, light-weight, fast fission chamber, with an improved discrimination capability between fission and α-decay events [36], was coupled to 54 EJ-309 [37] liquid scintillators from the Chi-Nu array [38] to detect neutrons emitted in fission events. The fission chamber housed 47 mg of ^{239}Pu arranged in twenty-two deposits and eleven readout channels, with an α-activity of about 10 MBq per channel, to be compared to a fission rate of about 15 events/s. A fission-fragment detection efficiency of 95% was nevertheless achieved [36]. Such a feature is crucial to avoid any bias of the data associated with the selection of a particular range in angle or kinetic energy of the detected fragments. Neutrons and γ rays were detected in coincidence with a fission-chamber signal in the scintillator cells and identified via the pulse shape discrimination technique down to 200 keV and up to about 14 MeV. The neutron detectors covered nine angles, from 30° to 150°. The use of digital Fast Acquisition SysTem for nuclEar Research [39] allowed the near complete avoidance of numerical dead time. A detailed description of the experimental setup can be found in [36, 38, 40, 41].

The combined setup and the high recorded statistics lead to a precise reconstruction of the PFNS as a function of the incident-neutron energy E_{in}^{n}, from 0.7 to 700 MeV. We stress that the data presented here were collected in the same experiment and under the same experimental conditions as the PFNS data discussed in [40]. The experiment allowed us to access both the PFNS and $\overline{\nu}_p$ observables, the latter with an expanded analysis, as relevant systematic uncertainties are different. The first step to extract $\overline{\nu}_p$ values is therefore common to the two analysis, and is described in Ref. [40], where the PFNS experimental data and the associated uncertainties are reported. Here we only recall that neutron detector efficiencies were obtained by measuring the PFNS of the ^{252}Cf spontaneous fission reaction in the same experimental conditions and with the same analysis procedure as the ^{239}Pu. For each detector, the measured ^{252}Cf(sf) PFNS was divided by the evaluated PFNS standard [42], normalized to the evaluated ENDF/B-VIII.0 $\overline{\nu}_p$ for ^{252}Cf of $(3.759 \pm 0.42\%)$ [2], and used to evaluate the efficiency of every EJ-309 detector. The bias associated with this procedure was carefully evaluated via GEANT4 simulations [43] and found to be negligible. The prompt

fission neutron spectrum for each of the eighty-six E_{in}^{n} bins studied was obtained by combining all the detector spectra corrected for their efficiency. As each of them was corrected by its neutron detector efficiency, the integral of the PFNS is the average number of prompt neutrons emitted per fission ($\overline{\nu}_p$). The so-extracted values are, however, affected by systematic biases which have to be accounted for with a more complex analysis to obtain high-precision data. In the following, results are presented with the absolute statistical and systematic uncertainties, propagated through the data analysis. The latter includes the uncertainty on the evaluated ^{252}Cf PFNS, while the uncertainty of 0.42% on the ^{252}Cf $\overline{\nu}_p$ is not included. This will allow to easier account for more precise future measurements of ^{252}Cf $\overline{\nu}_p$. Tabulated data are provided as supplemental material to the present work.

The main breakthrough with respect to previous measurements is the possibility of effectively estimating the sources of possible systematic bias. Data were corrected for four different experimental biases: the neutron detection energy range, the presence of a slower incident neutron background (wrap-around) [44], the limited detector angular coverage and the detector dead time. The correction on the $\overline{\nu}_p$ values related to each of these physical effects ($\epsilon_{\overline{\nu}_p}$), as well as the uncertainty introduced on the final $\overline{\nu}_p$ value by each correction ($\sigma_{\overline{\nu}_p}$), are plotted in Figs. 2a and b, respectively, as a function of E_{in}^{n} .

First, the detection limits of 0.2 and 14 MeV for the fission neutrons were considered. The lower limit was set by the threshold for discriminating neutrons from γ-rays, while the high-energy one was related to the dynamic range of the electronics. For corrections needed by these detection limits, the two regions were handled separately. The contribution to $\overline{\nu}_p$ of neutrons below the 200 keV detection limit was estimated assuming a simple theoretical description of the low-energy region of the PFNS based on a Maxwellian spectrum [45, 46] and found to be as high as 1%, as shown in Fig. 2a (red dots). The measured spectra were therefore extrapolated at energies from 200 keV downwards and $\overline{\nu}_p$ corrected for it. The bias introduced by the arbitrary choice of the fitting range on the extracted $\overline{\nu}_p$ found to be smaller than 0.2% (cyan stars in Fig. 2a). The uncertainty introduced on the $\overline{\nu}_p$ values by this procedure is negligible with respect to the statistical and systematic uncertainty of the uncorrected value (compare red dots

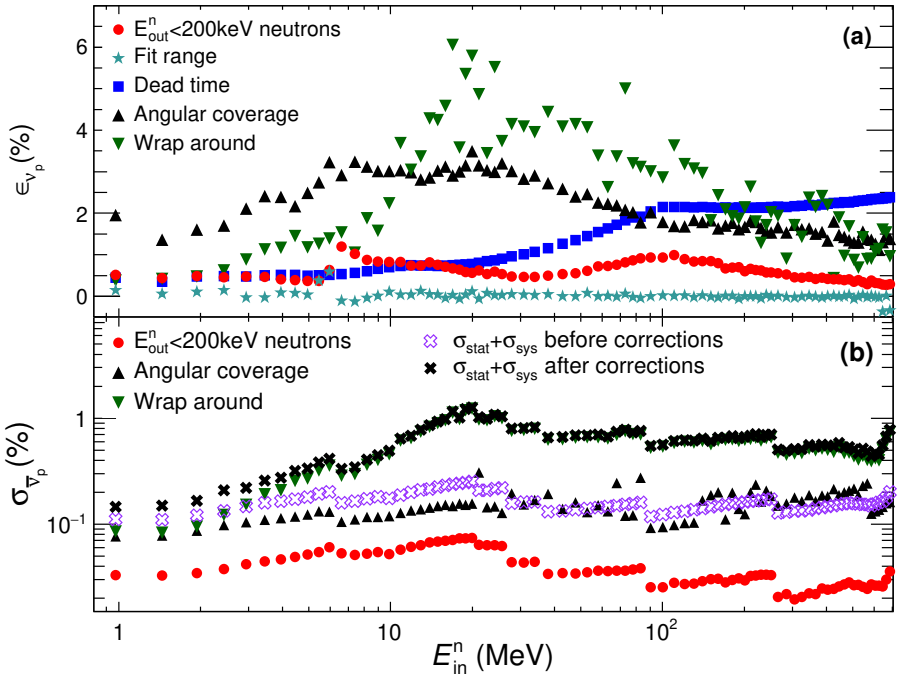

Figure 2: (Color online) (a) Bias of $\overline{\nu}_p$ due to different experimental effects. (b) Uncertainty on $\overline{\nu}_p$ ($\sigma_{\overline{\nu}_p}$) introduced by the correction of the experimental bias and the sum of the $\sigma_{\overline{\nu}_p}$ before and after the corrections. The "fit range" and the "dead time" corrections $\sigma_{\overline{\nu}_p}$ are negligible (see text).

and open crosses in Fig. 2b). For the high-energy limit, we can reasonably expect neutrons above 14 MeV to be emitted during a pre-equilibrium, pre-fission process for incident energies above about 24 MeV. A TALYS calculation [47] estimates their contribution to be about 0.9% of $\overline{\nu}_p$ at this E^n_{in}. It should be noted that this process can contribute significantly to $\overline{\nu}_p$, but it cannot be estimated quantitatively as available pre-equilibrium emission models have been validated on limited experimental data [48]. Therefore, $\overline{\nu}_p$ values for E^n_{in} above 24 MeV should be considered as a lower limit.

Second, the obtained values of $\overline{\nu}_p$ were corrected for neutrons emitted in fissions induced by slower-than-measured neutrons, the wrap-around background [44]. The fraction of wrap-around background, k_{WA}, in each E^n_{in} bin could be analytically determined from the time-of-flight spectra of incident neutrons as described in [40]. The procedure was here validated by the observation of dips in the evolution of k_{WA} with

E_{in}^{n} at energies corresponding to absorption resonances in ^{16}O, ^{14}N (i.e. air) and ^{11}B (boron material present in the beam hardener). The k_{WA} fraction varies from about 10% to about 3% below 20 MeV and above 200 MeV, respectively, of the impinging neutron flux and modifies the $\overline{\nu}_p$ value up to 6%, pointing out its importance (green triangles in Fig. 2a). The relative uncertainty introduced on the $\overline{\nu}_p$ values by the correction of this effect, which reaches up to 1%, arises from the statistics available for the estimation of k_{WA} (green triangles in Fig. 2b).

Third, the limited detector angular coverage was considered. The high segmentation of the Chi-Nu array allowed for the reconstruction of the $\overline{\nu}_p$ angular distribution and the correction for those angles that were not covered by detectors. Nine spectra, one for each measured θ_{lab}, were obtained by combining the spectra from the six detectors at the considered angle and $\overline{\nu}_p\,(\theta, E_{in})$ extracted. Their uncertainty is close to 0.3 %. The angular distributions, $\overline{\nu}_p\,(\theta, E_{in})$ vs cos(θ), exhibit two main characteristics: first, they are not isotropic, even at low incident energies, with a $\overline{\nu}_p\,(0^\circ)/\overline{\nu}_p\,(90^\circ)$ of about 1.05 below 10 MeV, and a trend similar to the one observed in the data for fission-fragment anisotropy [49–51]. Second, they are characterized by a certain degree of forward/backward asymmetry which increases with E_{in}^{n} , reflecting the increase in the kinematical boost and the pre-equilibrium emission. Angular distributions were fitted with up to 4^{th}-order polynomial functions and $\overline{\nu}_p\,(E_{in})$ was taken as the sum of the experimental values and the values deduced from the fitted distributions, for those angles that were not covered by detectors during the experiment. The systematic uncertainty on $\overline{\nu}_p$ due to the arbitrary choice of the functions was found to be negligible with respect to its final uncertainty. Accounting for the neutron angular distribution modifies up to 4% the $\overline{\nu}_p$ values and it mainly arises from the contribution of the most forward/backward angles (black triangles in Fig. 2a). This implies that the assumption of a flat angular distribution or a non-accurate knowledge of it likely leads, even at low energies, to an underestimation of $\overline{\nu}_p$. Interestingly, existing literature data are generally lower than the present results, and this could be a source of discrepancy (see Figs.1 and 3). The uncertainty on the $\overline{\nu}_p$ values introduced by the described correction is shown as black triangles in Fig.2b.

Finally, the impact of the neutron detector dead time was investigated. Once a

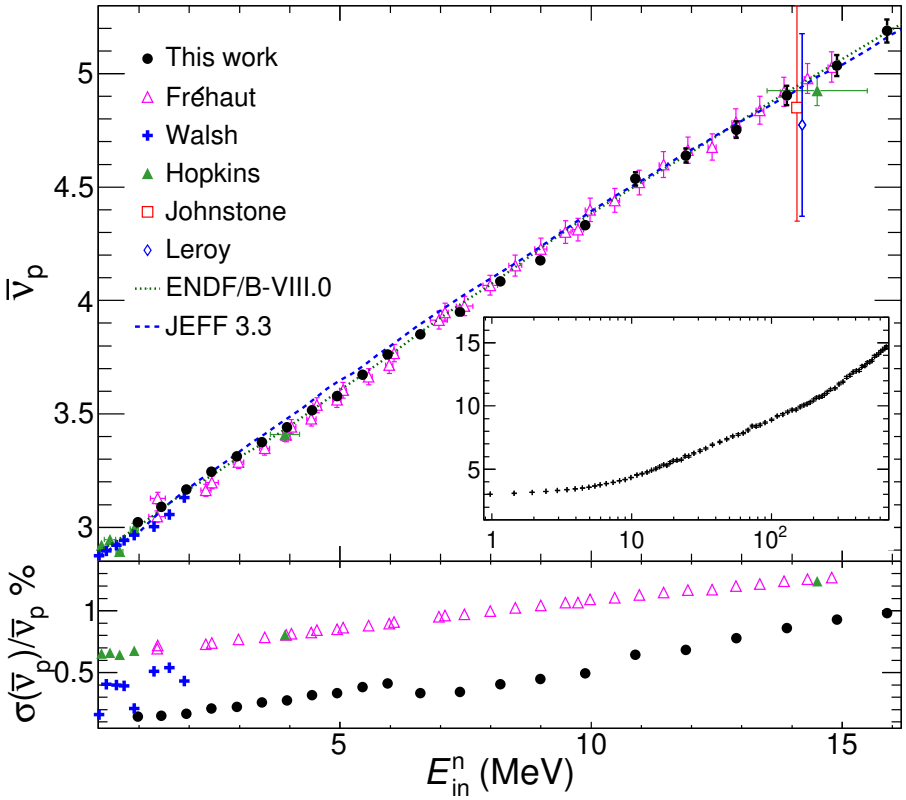

Figure 3: (Color online) Measured $\overline{\nu}_p$ and its uncertainty as a function of incident neutron energy up to 16 MeV. Some data from previous experiments are also shown [11, 13–15, 18]. The ^{252}Cf $\overline{\nu}_p$ uncertainty was removed from existing data. Dotted and dashed lines are ENDF/B-VIII.0 and JEFF3.3 evaluations, respectively. The insert shows the measured $\overline{\nu}_p$ over the whole studied E^n_{in} energy range.

particle (neutron or γ ray) fires a scintillator detector of the Chi-Nu array, a charge-integration window of 200 ns is opened, during which any other impinging particle is not recorded separately, but its charge signal adds to that of the first one. Therefore particles impinging with a time difference smaller than 200 ns in the same detector can be mis-identified and neutrons can be "lost" or "gained". The net amount of "lost" neutrons was estimated with a full Monte Carlo simulation based on experimental distributions. Measured neutron and γ-ray multiplicities, as well as time-of-flight distributions for each E^n_{in} bin were sampled and used as input. A similar procedure was undertaken

for the fast-to-total signal charge ratio vs total signal charge correlation of each detector. The same simulation, with the appropriate inputs, was run for the ^{252}Cf(sf) data, as part of the detector dead time distortion is accounted for when correcting the PFNS for the detector efficiency. The net correction varies from $\sim 0.5\%$ to above 2% for energies below 10 MeV and above 100 MeV, respectively (squares in Fig. 2a), due to the increase of γ and neutron multiplicities as the incident-neutron energy increases. The number of simulated events is high enough so that the statistical uncertainty introduced by this correction is negligible.

The data corrected as described above are shown as a function of E_{in}^{n} in Figs. 1 and 3. Our data exhibit the expected constant increase up to 700 MeV with no obvious structure. As mentioned, $\overline{\nu}_p$ values for energies above about 24 MeV, where the contribution of high-energy (> 14MeV) pre-equilibrium neutrons becomes non negligible, should be considered as a lower limit. Below about 14 MeV, $\overline{\nu}_p$ exhibits a linear dependence with the neutron energy. A linear extrapolation below 3 MeV provides an estimated value of $\overline{\nu}_p$ at thermal neutron energies of (2.879 ± 0.010) neutrons/fission, in agreement with the evaluated value of (2.868 ± 0.012) [2] and with comparable uncertainty.

The obtained $\overline{\nu}_p$ total uncertainties (excluding the 0.42% uncertainty associated to the reference data ^{252}Cf (s.f.) $\overline{\nu}_p$ [2]) span from 0.15 to 1.3%, and are smaller than 1% below 14 MeV E_{in}^{n} (see Figs. 2b and 3). Such low uncertainties on a broad energy range were never reached before, not even with different experimental techniques ([15–24] and [5, 9, 13]) as shown in the bottom panel of Fig. 3. The relative difference between our data and ENDF/B-VIII.0 values is shown in Fig. 1. At low E_{in}^{n} our data show a different trend than the reference experimental data of J. Fréhaut et al. [18], but the difference between our data and the recent ENDF/B-VIII.0 evaluation [2], averaged over the points measured below 5 MeV, is of 0.3%. A significant discrepancy is observed at the opening of the second-chance fission. The observed overall agreement with the ENDF/B-VIII.0 evaluation shows that the data presented here agree with the general trend of the bulk of the data considered to be reliable enough for the evaluation.

Complex phenomenological fission models [52–54] are nowadays available, which allow for a reasonable and cross-dependent description of many fission observables.

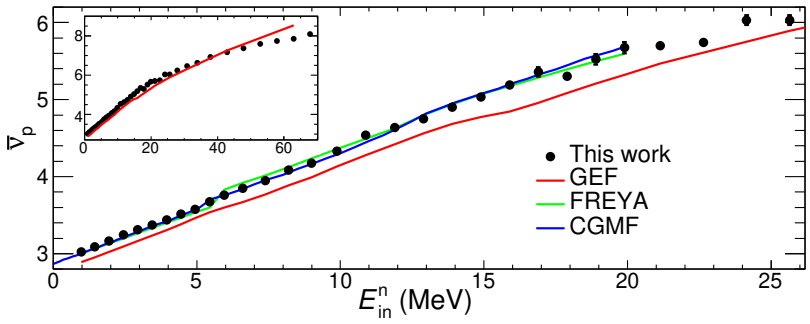

Figure 4: (Color online) Comparison of our data to GEF [55], FREYA [53] and CGMF [52] predictions.

Accurate values of $\overline{\nu}_p$ are essential constraints to these models. Notably, the sharing of energy between kinetic and excitation energy together with the energy sharing between the two fragments are crucial components of the models sensitive to $\overline{\nu}_p$. However, it should be noted that $\overline{\nu}_p$ alone cannot constrain the models. Our data are compared to the semi-empirical model GEF [55] in Fig.4. We observe that GEF predictions reproduce our $\overline{\nu}_p$ values within 0.15 (4.5%) and 0.4 (8%) neutrons per fission below 8 MeV and over the full energy range [1 − 25] MeV, respectively. The observed difference of 0.15 (0.4) neutrons per fission corresponds, in the GEF model, to a "wrong" sharing between fission-fragment excitation and kinetic energies of about 1 (2.8) MeV, to be compared to about 200 MeV released in fission. Although these model results are too far from experimental data to be used in evaluations, it should be noted that GEF is not tuned to these experimental data. Our data are also compared to the fission-event generators FREYA [53] and CGMF [52], up to 20 MeV, which is the limit of the models [52, 56]. FREYA and CGMF values systematically underestimate our data below 5 MeV, with differences up to 1.5% and 1%, respectively. While this deviation is smaller than for GEF values, it should be noted that for FREYA and CGMF results the fission-fragment total kinetic energy is adjusted to reproduce available $\overline{\nu}_p$ data. The obtained values are therefore not $\overline{\nu}_p$ predictions. Moreover, structures commensurate with second- and third-chance fission can be seen in all model values and our experimental data; however, model and experimental values do not fully agree on the E^n_{in} where multiple-chance fission structures are observed. This is the case because these struc-

tures could not be observed in many other experimental data before given that a high precision is needed to resolve them. Hence, the data presented here can inform complex fission models on the absolute value and shape of $\overline{\nu}_p$ more conclusively. Indeed, our data were used as an input to an evaluation with the CGMF model [52]. In this case, they could be fitted well within the small experimental uncertainties. However, it should be noted that parameters underlying these models need to be constrained and/or validated with respect to precise experimental data to be able to provide precise enough nuclear data to meet the requirements demanded by nuclear energy applications. As mentioned before, above 25 MeV the contribution of neutrons with energies above 14 MeV becomes significant, making of the measured $\overline{\nu}_p$ values a lower limit. For E^n_{in} greater than 60 MeV, the GEF pre-equilibrium neutron spectra become unphysical.

To assess the impact of our data compared to existing data sets, we performed two new evaluations of ^{239}Pu $\overline{\nu}_p$, with ($Ev^{w/\,this\,work}$) and without the data presented here ($Ev^{w/o\,this\,work}$), using the same methodology. The uncertainties of the all data [8, 9, 13, 15–22, 24] were carefully reviewed and increased according to [57] in cases where uncertainties were missing. Our data reduce the ENDF/B-VIII.0 and $Ev^{w/o\,this\,work}$ $\overline{\nu}_p$ evaluated relative uncertainty, $\sigma^{ev}_{\overline{\nu}_p}$, by up to 50% and 60%, respectively, in the 1 to 15 MeV range (see dashed lines in Fig.5). This is of high importance for nuclear applications as it reduces the uncertainties and increases the predictive power of neutronics calculations. In addition to the high-impact due to low uncertainties, our data offer two additional benefits. Below 5 MeV the $\overline{\nu}_p$ evaluated mean value, $\overline{\nu}^{ev}_p$, is only sligthly modified ($< 0.15\%$) by our data (Fig. 5 green full line), which therefore validate an evaluation obtained by an average over previous data measured all by the same-but different than here technique. That is of high importance, given that this validation was missing so far. Furthermore, above 5 MeV, where no integral data exist and experimental data are scarce, our results modify ENDF/B-VIII.0 and $Ev^{w/o\,this\,work}$ $\overline{\nu}^{ev}_p$ by up to 0.7% and 0.9%, respectively. This is consistent with an increasing importance of physics effects leading to biases as E^n_{in} increases, which were carefully accounted for in this work. Our data provide therefore a more solid ground for future evaluations.

In conclusion, previously unattained precise and accurate new data on ^{239}Pu $\overline{\nu}_p$ are reported, which extend the studied range from 1 up to 700 MeV. The data were obtained

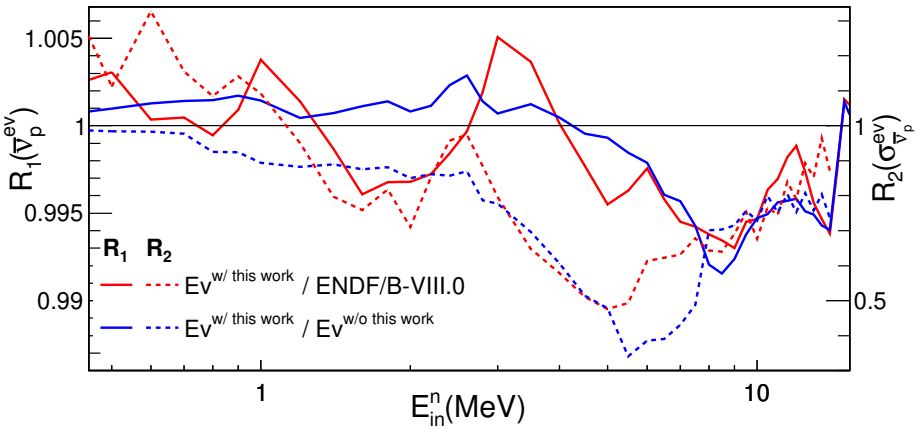

Figure 5: (Color online) Ratio of $\overline{\nu}_p$ evaluated mean value (R_1, full lines, left axis) and relative uncertainty (R_2, dashed lines, right axis) for different evaluations (see text).

with the double time-of-flight technique and an innovative setup. It allowed to explicitly account for experimental systematic bias, which have hampered the precision and accuracy of existing experimental results, thus providing more reliable-than-existing data. Below 5 MeV a good agreement with the recent ENDF/B-VIII.0 evaluation is observed validating it, for the very first time, with an independent measurement. A new evaluation performed here with these data shows that they significantly reduce the uncertainty on evaluated nuclear-data libraries for a nuclide, the ^{239}Pu , crucial for nuclear energy applications.

With this measurement the experimental challenge of precisely measuring prompt fission-neutron multiplicity on highly radioactive nuclei has been taken up thanks to the innovative setup and experimental technique. New high-precision ^{239}Pu $\overline{\nu}_p$ measurements could be realized at incident-neutron energies from 200 keV to 2 MeV, where existing data are highly spread, and even down to 1 keV where no data exist. Moreover these results open up the possibility of precisely investigating other high-activity actinide nuclei to contribute to a better understanding of the fission process while providing key elements for the development of new technologies relevant for society.

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