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Fast-neutron-induced fission cross section of ^{242}Pu measured at $n\text{ELBE}$

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The fast-neutron-induced fission cross section of ^{242}Pu was measured at the neutron time-of-flight facility $n\text{ELBE}$. A parallel-plate fission ionization chamber with novel, homogeneous, large-area ^{242}Pu deposits on Si-wafer backings was used to determine this quantity relative to the IAEA neutron cross-section standard $^{235}\text{U}(n,f)$ in the energy range of 0.5 to 10 MeV. The number of target nuclei was determined from the measured spontaneous fission rate of ^{242}Pu . This helps to reduce the influence of the fission fragment detection efficiency on the cross section. Neutron transport simulations performed with Geant4, MCNP6 and Fluka 2011 are used to correct the cross-section data for neutron scattering. In the reported energy range the systematic uncertainty is below 2.7 % and on average the statistical uncertainty is 4.9 %. The determined results show an agreement within 0.67(16) % to recently published data and a good accordance to current evaluated data sets.

I. INTRODUCTION

Future nuclear power concepts with a closed fuel cycle, such as accelerator driven systems and generation IV reactors, targeted to use their fuel more efficiently, will produce less radioactive waste, meet the stringent standards of safety and proliferation resistance, and strive to be more economically competitive [1] compared to current reactor designs. Transmutation of nuclear waste in fast reactors is discussed as a way to reduce the radiotoxicity of the presently existing nuclear fuel. However, the technical realization of such plants is a challenging and expensive endeavour. Accurate nuclear data, especially fast-neutron-induced fission cross sections, are essential for new reactor designs.

^{242}Pu is the longest-lived plutonium isotope in spent nuclear fuel ($T_{1/2} = 375,000$ yr [2]) and hence important for nuclear transmutation, as ^{244}Pu production is negligible [3].

Current uncertainties of the $^{242}\text{Pu}(n,f)$ cross section are of about 21 % in the energy range from 0.5 to 2.23 MeV [4]. For a reliable prediction of the neutron multiplication and other reactor core parameters in these novel reactor concepts, the total uncertainty needs to be

reduced to below 5 % [4, 5]. This task is addressed within the INDEN project [6], where ^{242}Pu is one of the nuclides with the highest priority.

The fast neutron-induced fission of ^{242}Pu is studied since 1960 [7]. A brief summary of the available experimental data acquired since then has already been given in Ref. [8]. In addition, an absolute measurement of the fission cross section was also performed in Dresden in 1983 by using quasi-monoenergetic neutrons with energies of 2.6 MeV, 8.4 MeV and 14.7 MeV [9]. Recently published measurements done at the Los Alamos National Laboratory by Tovesson *et al.* [10], at the Joint Research Center Geel by Salvador-Castñeira *et al.* [8] and at the National Physical Laboratory of the United Kingdom in Teddington by Matei *et al.* [11] and Marini *et al.* [12] tend to be lower than present evaluated nuclear data [13]. To reduce the total uncertainty of the evaluated fission cross section, more accurate and precise nuclear data over a large energy range is needed.

This challenging task was addressed at the neutron time-of-flight (ToF) facility $n\text{ELBE}$ of the Center for High-Power Radiation Sources ELBE¹ at Helmholtz-Zentrum Dresden - Rossendorf. $n\text{ELBE}$ is the first photo-neutron source at a superconducting electron accelerator. It allows operating the electron beam in continuous-wave (cw) mode with more than 100 kHz micropulse repetition rate. Improved neutron beam intensity, experimental conditions, e.g. a low scattering environment, and a suitable spectral fluence for fast neutron-induced reaction studies provided first-rate conditions to achieve this aim [14].

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¹ Electron Linac for beams with high Brilliance and low Emittance

66 The present work reports an experiment on the neu-
67 tron-induced fission cross section of ^{242}Pu relative to the
68 IAEA neutron cross-section standard $^{235}\text{U}(n,f)$ [15].

69 II. EXPERIMENTAL SETUP

70 A. Fast neutrons at ELBE

71 The n ELBE photo-neutron source [16–18] produces
72 fast neutrons with kinetic energies between 10 keV and
73 20 MeV. Electrons impinging on a liquid lead target
74 produce bremsstrahlung during their deceleration. This
75 bremsstrahlung generates the neutrons via (γ, n) reac-
76 tions on the lead nuclei. The neutrons, in turn, are emit-
77 ted almost isotropically from the radiator, while a large
78 part of the electrons and the bremsstrahlung photons
79 mainly emerge in the forward direction. To minimize the
80 photon-to-neutron ratio, only neutrons emitted through
81 under 100° are used in the experimental area passing a
82 dedicated collimator system. The excellent timing of the
83 ELBE electron beam of ~ 5 ps pulse length in combina-
84 tion with the compactness of the neutron source, enables
85 high resolution neutron time-of-flight experiments even
86 at short flight paths of around 6 m.

87 The present experiment was performed with an elec-
88 tron beam energy of 30 MeV and an average bunch charge
89 of 73 pC on the neutron-producing target. The repetition
90 rate was 406.25 kHz. The corresponding pulse separation
91 of $2.46 \mu\text{s}$ prevents neutron pulse overlap while still pro-
92 viding a beam intensity of $3.7 \cdot 10^4 \text{ n}/(\text{cm}^2\text{s})$ which is suf-
93 ficient for the present experiment. An absorber reducing
94 the γ -flash of the electron beam in this experiment was
95 not required.

96 B. Fission chambers

97 A parallel-plate plutonium fission ionization chamber
98 (hereafter PuFC) was constructed at HZDR [19, 20]. It
99 is equipped with eight large area (\varnothing 74 mm), isotopic
100 pure (cf. Tab. I), thin (96(3) to 126(4) $\mu\text{g}/\text{cm}^2$) and
101 homogeneous deposits of ^{242}Pu , which have been pro-
102 duced within the TRAKULA project by Vascon *et al.*
103 [21] at the Institute of Nuclear Chemistry of the Johannes
104 Gutenberg University Mainz. Molecular plating was used
105 to precipitate the fissile material from a nitrate solution
106 on titanium coated silicon wafers of 400 μm thickness.
107 Due to the flatness and minimal surface roughness of the
108 Si-wafers, homogeneous thin layers containing plutonium
109 could be produced. SEM/EDX measurements of the sur-
110 face of the ^{242}Pu layers revealed cracks on a $< 1 \mu\text{m}$ scale,
111 which are due to the drying of the isopropanol solvent
112 used in the molecular plating. Nevertheless, the homo-
113 geneity is still better than for conventional deposition
114 (e.g. painting or electro-deposition) on metallic foils.

115 The incident neutron flux of the reported experiment
116 was determined by the well characterized ^{235}U trans-

TABLE I. Isotopic composition of the used plutonium targets
in the PuFC and uranium targets in the H19. The tabulated
values for uranium have been picked from Ref. [22]. The plu-
tonium composition (Batch I.D. Pu-242-327A1) was given by
the manufacturer, Oak Ridge National Laboratory (ORNL).

	Abundance / %		
	PuFC		H19
^{238}Pu	0.0020(3)	^{234}U	0.03620(20)
^{239}Pu	0.0050(3)	^{235}U	99.9183(3)
^{240}Pu	0.0220(3)	^{236}U	0.00940(10)
^{241}Pu	0.0020(3)	^{238}U	0.03610(20)
^{242}Pu	99.9670(3)		
^{244}Pu	0.0020(3)		

117 fer instrument H19 of PTB Braunschweig [22, 23]. An
118 overview of the key-properties of the fission targets of
119 both fission chambers is given in Tab. II.

TABLE II. Key parameters of the PuFC and H19 fission de-
posits. The areal densities and total activity of the n ELBE
targets have been calculated from their individual sponta-
neous fission rates, which have been measured in situ and
reduce the systematic uncertainties compared to conventional
 α -spectroscopy (see Sec. II E for more details.) Their homo-
geneity was derived from radiographic images. The properties
of the H19 fission deposits were taken from Refs. [22, 23].

	PuFC (^{242}Pu)	H19 (^{235}U)
type of deposition	molecular plating	painting
no. of deposits	8 (single-sided)	5 (double-sided)
deposited area / cm^2	43.0(5)	45.4(5)
enrichment / %	99.9670(3)	99.9183(3)
total mass / mg	37.24(22)	201.4(5)
areal density / $\mu\text{g}/\text{cm}^2$	96(3)-126(4)	444(5)
total activity ^a / kBq	8,317.60	32.91
homogeneity / %	96.7 ^b	>96

^a Including contaminants.

^b Homogeneity means 1 minus the ratio of the standard deviation
and the mean of the summed intensities of the radiographic
images.

120 Both H19 and PuFC were operated in the forward
121 biasing mode. This means that the five double-sided
122 fission samples of the H19 and the eight single-sided
123 samples of the PuFC were cathodes on ground poten-
124 tial. Compared to the H19 electrode spacing of 5 mm,
125 the distance between the anodes and cathodes of the
126 PuFC was doubled, to increase the signal-to-noise ratio
127 (charge of fission fragment induced signals compared to
128 the charge of α -particle induced signals). The electric
129 field strength of both chambers, $|\vec{E}|^{\text{H19}} = 240 \text{ V cm}^{-1}$
130 and $|\vec{E}|^{\text{PuFC}} = 300 \text{ V cm}^{-1}$, was chosen to ensure fast
131 signals and good timing properties.

132 The induced charges on the anodes of the PuFC were
133 read out by in-house developed charge-sensitive pream-
134 plifiers. Short rise times of approximately 80 ns and a
135 signal length in the order of 400 ns reduce the pile-up

136 probability by a factor of 5 in comparison to the com- 159
 137 monly used combination of a spectroscopic amplifier and 160
 138 a conventional preamplifier with μs -shaping time. Fur- 161
 139 ther details of the $n\text{ELBE}$ fission chamber can be found
 140 in Ref. [24].

141 C. Setup

142 H19 and PuFC were placed at a distance of 5.95 to
 143 6.35 m with respect to the photo-neutron source and a
 144 distance of 10 cm between each other. The neutron beam
 145 diameter in this region is between 52 and 56 mm and,
 146 therefore, always smaller than the fission targets. The
 147 beam profile was measured at different points along the
 148 neutron beam axis by using horizontally and vertically
 149 scanning plastic scintillators and linearly interpolated to
 150 the region of interest (see Ref. [18]).

151 A sketch of the whole experimental setup is shown in
 152 Fig. 1. With the beam parameters chosen, the average
 153 neutron-induced fission rate of the H19 was about 31 s^{-1} .
 154 The respective photo-fission rate was the nearly the same.
 155 For the PuFC, the neutron-induced fission rate was 5 s^{-1} .

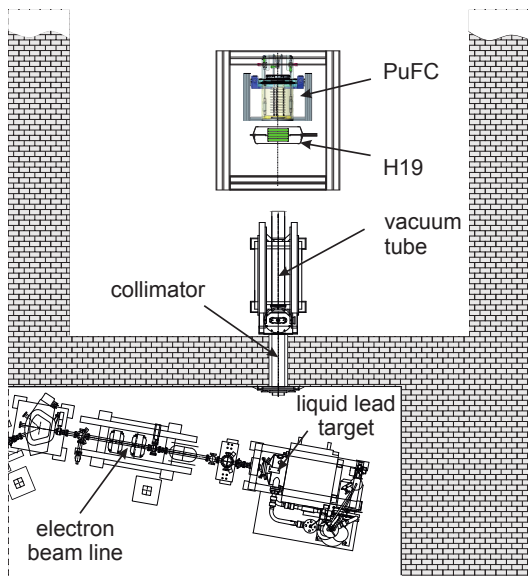


FIG. 1. Experimental setup. The ELBE electron beam comes from the lower left side and is guided to the photo-neutron source. A fraction of the isotropically emitted neutrons passes a collimator and enters the low-scattering experimental area. The incident neutron flux was measured with the ^{235}U fission chamber H19. Fast neutron-induced fission events of ^{242}Pu were recorded with the fission chamber PuFC. Picture not to scale.

156 D. Data acquisition

157 The timing and energy information of both fission
 158 chambers was registered in list mode by the MBS data

159 acquisition software developed at GSI, Darmstadt [25].
 160 A scheme of the VME-based data acquisition electronics
 161 is shown in Fig. 2.

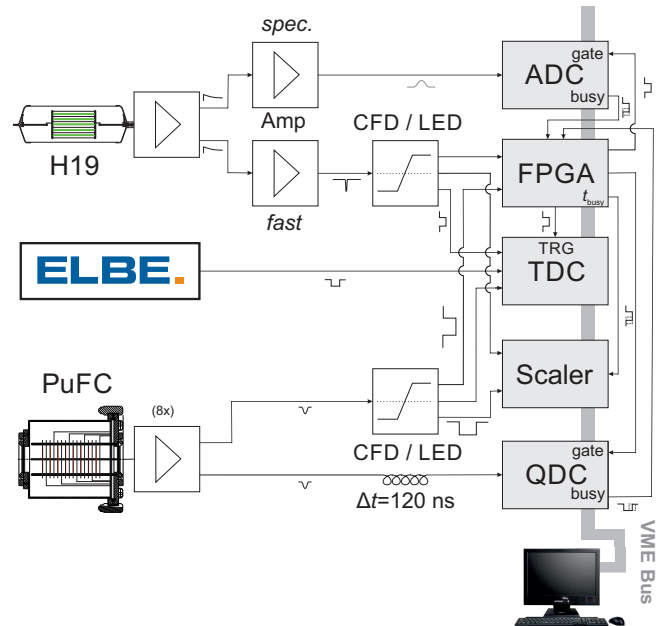


FIG. 2. Scheme of the electronic setup and the data acquisition system. The output signals of the charge-sensitive (ns-) preamplifiers are split to determine the timing and the collected charge. Pulse heights of the H19 signals are acquired with an ADC after getting shaped by a spectroscopic amplifier, whereas the charge of the eight PuFC channels (only one is shown here) is determined by a QDC. The production of a fast trigger makes the use of a timing-filter-amplifier in the timing branch of the H19 necessary. The second output signal is converted to a logical signal by an in-house developed discriminator (CFD/LED). The logical signals are used to determine the timing in a time-to-digital converter (TDC) and to produce a trigger for the data acquisition in an FPGA.

162 The signals from the 10 deposits of the H19 fission
 163 chamber were summed, amplified by one charge-sensitive
 164 preamplifier and afterwards measured by a conventional
 165 spectroscopic amplifier (Ortec 671) in the energy branch
 166 and by a timing-filter-amplifier (Ortec 474) in the timing
 167 branch. The signals of the eight ^{242}Pu deposits were reg-
 168 istered separately with the fast charge-sensitive pream-
 169 plifiers mentioned in Sec. II B, to reduce possible pile-up
 170 of α -radioactivity of the ^{242}Pu even further.

171 The short signal length of the ns-preamplifier allows
 172 a charge-to-digital converter (QDC, CAEN V965A) to
 173 be used. The energy information of the H19 was deter-
 174 mined by a peak-sensing analog-to-digital converter
 175 (ADC, CAEN V1785N).

176 The timing of the recorded signals was extracted by
 177 an in-house developed discriminator (CFD/LED), which
 178 combines a constant-fraction and a leading-edge discrim-
 179 inator. The neutron time-of-flight was measured relative
 180 to the ELBE radio frequency using a multi-event/multi-

181 hit time-to-digital converter (TDC, CAEN V1290A). The
 182 trigger for the whole data acquisition was a logical OR of
 183 all fission chamber channels generated by a multi-purpose
 184 board (FPGA, CAEN V1495). The leading-edge out-
 185 puts of the discriminator as input for the FPGA prevents
 186 losing valid signals with slow rise-time, which otherwise
 187 will not be registered due to imperfect ARC-timing [26].
 188 This was investigated to be especially important for small
 189 amplitude signals, mainly by α -particles. The trigger
 190 thresholds and the delays of the CFD were chosen in a
 191 way, that the loss of fission fragments above the threshold
 192 was minimal, for both chambers integrally below 0.3%.
 193 Further details of the acquisition electronics can be found
 194 in Ref. [27].

195 E. Analysis

196 The pulse-height information of the recorded list-mode
 197 data was used to separate time-independent background
 198 resulting due to the natural α -decay of the target isotopes
 199 from the interesting fission events of interest. The charge
 200 spectra show the excellent quality of the ^{242}Pu samples
 201 (cf. Fig. 3), which is expressed in a peak-to-valley ratio
 202 of 20 to 21 for all Pu-deposits.

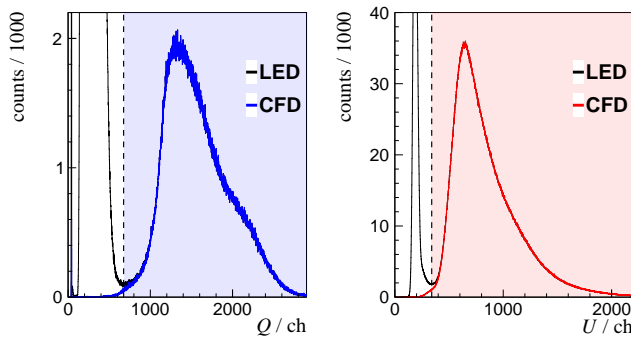
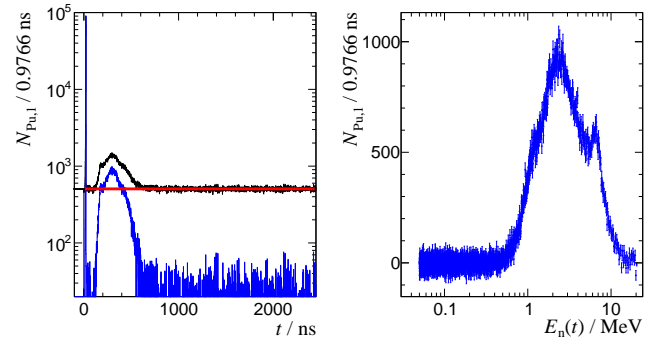


FIG. 3. Charge spectrum of channel #1 in the PuFC (left) and pulse height spectrum of the H19 (right). The leading-edge triggered QDC- and ADC values of the chambers are shown in black, the constant-fraction triggered ones in blue or red, respectively. The coloured areas indicate regions of pulse heights and charges related to fission fragments.

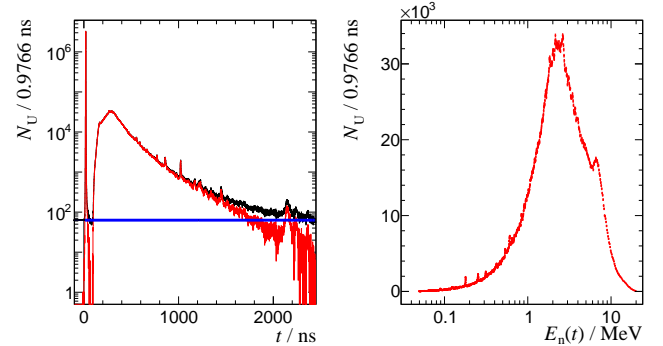
203 The time-of-flight spectra of each PuFC channel (e.g.
 204 channel #1, see Fig. 4(a)) and of the H19 (Fig. 4(b))
 205 were calibrated with photo-fission events. The full width
 206 at half maximum of the photo-fission peak corresponds
 207 to the time resolution of the fission chambers. For the
 208 summed signal of all H19 deposits, this value is slightly
 209 higher (2.3 ns) than the single-readout PuFC (1.7 ns).

210 After subtraction of a constant spontaneous fission
 211 background, the $^{242}\text{Pu}(n,f)$ fission rate $\dot{N}_{\text{Pu},i}$ could be
 212 determined as a function of neutron kinetic energy shown
 213 in Fig. 4(a) on the right-hand side.

214 A consistent energy binning for all fission targets is
 215 chosen to combine the counts of individual channels of



(a) PuFC ch. 1.



(b) H19

FIG. 4. Left: Detected time-of-flight spectrum N before (in black) and after (in blue for PuFC ch.1 and in red for H19) background subtraction. The horizontal red and blue lines indicate a constant extrapolation of the background induced by spontaneous fission events and room-return neutrons. Right: Background subtracted energy spectrum calculated using the time-of-flight spectrum shown on the left side.

216 the PuFC, which have slightly different flight paths [28].
 217 After rebinning and background subtraction, the relative
 218 fission cross section is determined by:

$$\frac{\sigma_{\text{Pu}}}{\sigma_{\text{U}}} = K \frac{\sum_i C_{\text{Pu},i} \dot{N}_{\text{Pu},i}}{\langle C_{\text{U}} \rangle \dot{N}_{\text{U}}} \frac{1}{I}. \quad (1)$$

219 Eq. (1) is the ratio of the detected fission count rates
 220 \dot{N} of both fission chambers, taking into account a neutron
 221 scattering correction C between individual fission targets
 222 for the PuFC or $\langle C_{\text{U}} \rangle$ averaged over all fission layers in
 223 the case of H19. This correction factor is discussed in detail
 224 in section IIF. The constant factor K is the ratio of the
 225 effective total areal densities εn of both fission cham-
 226 bers. Here, ε is the fission fragment detection efficiency
 227 which is in general difficult to determine. For the H19,
 228 $\varepsilon_{\text{U}} n_{\text{U}} = 107.5(16) \cdot 10^{-17} \text{ cm}^{-2}$ was taken from Ref. [23],
 229 whereas for the PuFC, $\varepsilon_{\text{Pu}} n_{\text{Pu}}$ was determined using the
 230 measured spontaneous fission rate of ^{242}Pu . This method
 231 was already introduced by Weigmann *et al.* in Ref. [29],
 232 feasible because the total uncertainty of the spontaneous
 233 fission partial decay constant λ_{SF} is smaller than 2%

234 [30, 31]. Taking into account the recent measurement
 235 of Salvador-Castiñeira *et al.* from Ref. [32], the weighted
 236 average (weighting according to Ref. [31]) of all avail-
 237 able data is $\lambda_{\text{SF}} = 3.25(4) \cdot 10^{-19} \text{ s}^{-1}$. An overview of
 238 all present data (expressed as $\ln 2/\lambda_{\text{SF}}$) is given together
 239 with the evaluated values in Fig. 5.

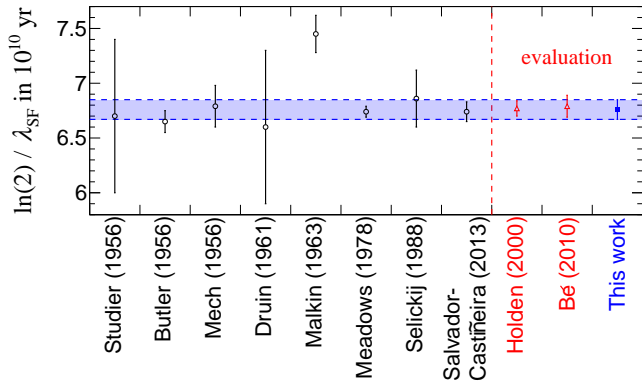


FIG. 5. Compilation of measured and evaluated partial half-lives for the spontaneous fission (SF) of ^{242}Pu . The experimental data were taken from [30] and [32]. The re-evaluation of this data by Bé *et al.* [31] has a slightly higher uncertainty. In blue, the weighted average of all listed values is shown including the latest measurement of Salvador-Castiñeira *et al.* [32]. The blue shaded area marks the combined standard uncertainty of this value (1.3%).

240 As the area of the plutonium deposits $A_{\text{Pu},i}$ is constant
 241 for all eight channels of the PuFC, the effective total areal
 242 density $\varepsilon_{\text{Pu}} n_{\text{Pu}}$ is determined by:

$$\begin{aligned} \varepsilon_{\text{Pu}} n_{\text{Pu}} &= \sum_i \varepsilon_{\text{Pu},i} n_{\text{Pu},i} \\ &= \frac{\alpha}{A \lambda_{\text{SF}}} \sum_i \dot{N}_{(\text{SF}),i}. \end{aligned} \quad (2)$$

243 In Eq. (2) a small dead time correction ($\approx 1\%$) of the
 244 DAQ is introduced denoted by α . Using this relation, the
 245 normalization factor K can be written as follows:

$$K = \frac{\varepsilon_{\text{U}} n_{\text{U}}}{\varepsilon_{\text{Pu}} n_{\text{Pu}}} = A \frac{\lambda_{\text{SF}}}{\alpha} \frac{\varepsilon_{\text{U}} n_{\text{U}}}{\sum_i \dot{N}_{(\text{SF}),i}}. \quad (3)$$

246 Inserting K into Eq. (1) shows that the relative cross
 247 section is independent from the fission-fragment detec-
 248 tion efficiency of the PuFC. This only holds for small
 249 neutron energies below 10 MeV because the higher the
 250 linear and angular momentum induced by the incident
 251 neutrons, the larger is the fission fragment anisotropy.
 252 This anisotropy lowers the detection efficiency. A model
 253 to calculate this effect was proposed by Carlson *et al.* in
 254 Ref. [33]. Due to the lack of experimental data for the
 255 fission fragment anisotropy and the barely known specific
 256 energy loss of fission fragments in the deposits, this in-
 257 efficiency I is not an accurate value. An estimate based

258 on the angular correlation data of Simmons *et al.* [34], a
 259 GEF 2016.1.2 calculation [35] to determine the ratio of the
 260 target nuclei velocity to the average fission fragment
 261 velocity and a Geant 4.10.1 [36] transport calculation to
 262 determine the specific energy loss of the fission fragments
 263 in the deposit is shown in Fig. 6.

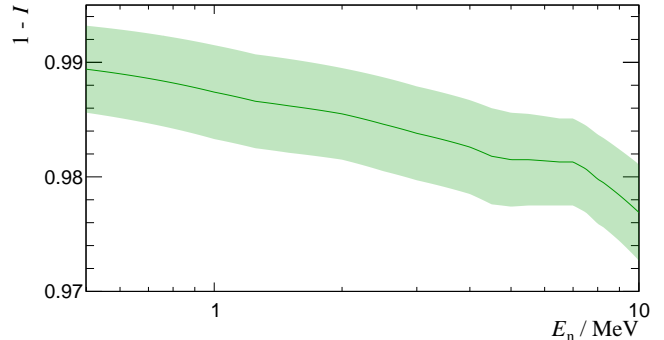


FIG. 6. Correction factor for the detection inefficiency I of fission fragments in the PuFC due to linear and angular momentum transfer according to the Carlson model [33].

F. Neutron scattering corrections

265 Corrections for neutron scattering are an important is-
 266 sue in analyzing neutron time-of-flight experiments. Two
 267 major effects play an important role, the attenuation of
 268 the neutron beam in every passed material and the loss of
 269 the correlation between neutron kinetic energy and their
 270 corresponding time-of-flight.

271 The latter is important especially for inelastically scat-
 272 tered neutrons, because they lose a large amount of their
 273 kinetic energy within a single interaction. If such an
 274 event occurs close to a fission target, the kinetic energy
 275 of the scattered neutron determined from the measured
 276 time-of-flight will be much higher than the true kinetic
 277 energy and the cross section at high neutron energies
 278 will be overestimated. Particle transport calculations al-
 279 low for correcting the influence of scattering as in these
 280 calculations both the true kinetic energy and the time-
 281 of-flight of the neutrons are accessible at once, which
 282 cannot be determined experimentally with the present
 283 setup. Such calculations have been performed using
 284 Geant 4.10.1 [36, 37], MCNP 6.1.1 [38] and Fluka 2011
 285 [39, 40]. The geometry has been implemented identi-
 286 cally in all three simulations with special attention to all
 287 materials close to the neutron beam. The outcome of
 288 all event-by-event calculations is a correlation matrix of
 289 the true kinetic energy E_n and the kinetic energy $E_n(t)$
 290 derived from their time-of-flight and the assumed undis-
 291 turbed flight path. An example of such a correlation
 292 matrix for the last target in the beam (PuFC channel
 293 #1) is shown in Fig. 7.

294 Because scattering cross sections are energy-depend-
 295 ent, it is necessary to use a realistic input spectrum in

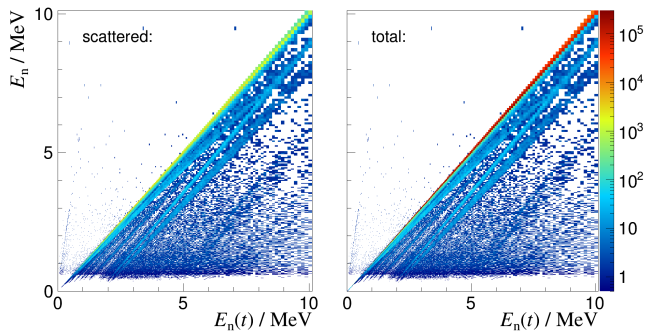


FIG. 7. Energy to time-of-flight-correlation of the last PuFC deposit in the neutron beam calculated using Geant 4.10.1. On the right-hand side, all neutrons passing the actinide target are shown, whereas on the left-hand side, only events are drawn, which have been scattered at least once. The bin content of each histogram was multiplied column-wise with the ^{242}Pu fission cross section at the respective neutron energy E_n to be proportional to the fission rate. Structures off the diagonal are caused by elastic and inelastic scattering on the target backing (mostly ^{28}Si) and stainless steel windows of the fission chamber (mostly ^{56}Fe).

the simulations. The measured neutron fluence detected by the H19 was used for this purpose. The influence of neutron scattering within the H19 itself is negligible.

To correct the attenuation of the neutron beam, one can define a transmission factor

$$T_i(E_n(t)) = \frac{N_i(E_n = E_n(t))}{N_0(E_n)}, \quad (4)$$

which is the ratio of all counted neutrons N_i in the i -th actinide target, that have not been scattered on their way to the target (on the main diagonal on the right of Fig. 7), and the total number of neutrons N_0 started from the neutron source. The average loss of neutrons between the first and last fission target is in the order of 15%, which is a consequence of the thickness of the Si-backings and the $200\ \mu\text{m}$ stainless steel windows of the PuFC.

For the loss of the energy to time-of-flight correlation, a similar correction factor k_i is defined.

$$k_i(E_n(t)) = \frac{N_i(E_n = E_n(t), E_n(t))\sigma(E_n)}{\int N_i(E'_n, E_n(t))\sigma(E'_n)dE'_n} \quad (5)$$

Scattered neutrons could still contribute to fission, so that k_i is the ratio of the detected fission rate of unscattered neutrons and the total detected fission rate. Because the fission rate depends on the cross section, the correlation matrices have been multiplied column-wise with the evaluated fission cross section of ^{242}Pu taken from ENDF/B-VIII.0. [41], which is for this particular reaction identical to its predecessor ENDF/B-VII.1 [13].

With Eq. (4) and Eq. (5), the neutron scattering correction factor $C_i(E_n(t))$ is defined in the following way:

$$C_i = \frac{k_i}{T_i} \quad (6)$$

As only the sum of all H19 fission targets is available, the arithmetic mean (C_U) was calculated to take the neutron scattering within this chamber into account. The average total correction factor is in the order of 9% and shown for all three simulations in Fig. 8. While Geant 4 and MCNP 6 provide identical results within their statistical fluctuations, the Fluka 2011 results show a negligible shift towards a higher correction factor.

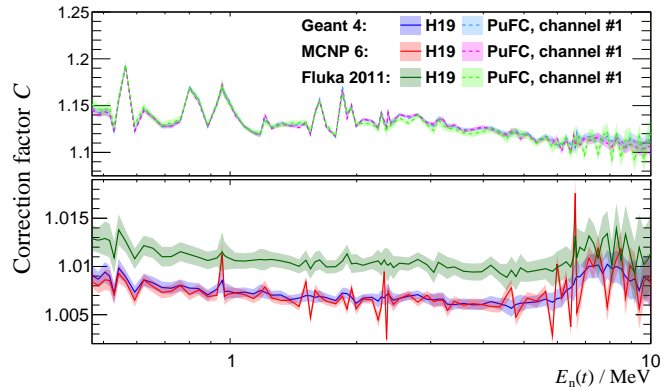


FIG. 8. Correction factor C for neutron scattering derived from Geant 4 (blue), MCNP 6 (red) and Fluka 2011 (green). This plot shows the maximum effect by comparing the first target of the H19 (lower panel) with the last target (PuFC, channel #1, upper panel) in the neutron beam. The confidence intervals shown here correspond to the 1σ statistical uncertainty.

The correction procedure was verified by evaluating the ratio of neutron-induced and spontaneous fission rate of the PuFC. Whereas the spontaneous fission is completely independent from any scattering, the neutron-induced is not. The ratio shows an exponential decrease along the plutonium chamber (red line in Fig. 9) and becomes constant after applying the neutron scattering correction (blue line in Fig. 9).

III. RESULTS AND DISCUSSION

With the scattering corrections in Sec. II F and the normalization constants listed in Tab. III, we are now able to calculate the relative fission cross section according to Eq. (1). The result is shown in Fig. 10 and compared to the measurements of Tovesson *et al.* [10], Staples *et al.* [42] and Weigmann *et al.* [29].

Because only the relative data of Staples *et al.* were included in the EXFOR database [43], the absolute cross section of the other two has been divided by their reported reference cross section to fit into this plot.

To compare our measurement with other recent data sets as well, the absolute cross section was determined

TABLE III. Normalization constants

λ_{SF}	$= 3.25(4) \cdot 10^{-19} \text{ s}^{-1}$	SF partial decay constant, cf. Fig. 5
A	$= 43.0(5) \text{ cm}^2$	actinide area, from deposition cell
ε_{U}	$= 0.945(14)$	fission fragment detection efficiency H19, Ref. [23]
n_{U}	$= 113.8(3) \cdot 10^{17} \text{ cm}^{-2}$	atomic areal density H19, Ref. [23]
$\alpha \sum_i \varepsilon_{\text{Pu},i} \dot{N}_{(\text{SF}),i}$	$= 29.688(4) \text{ s}^{-1}$	measured SF-rate
K	$= 5.04(12)$	total normalization

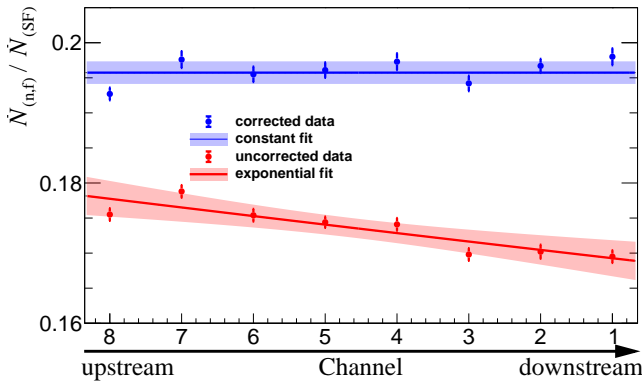


FIG. 9. Ratio of neutron-induced and spontaneous fission rate without (red) and with (blue) correction for neutron scattering. Without the correction, the fraction of the neutron-induced fission rate drops exponentially between the fission chamber channels, whereas the spontaneous fission rate stays constant. Note that channel #8 corresponds to the deposit closest to the neutron source while channel 1 is the farthest from it, thus having the largest absorption correction.

351 using the ^{235}U -IAEA Neutron Cross Section Standard
 352 from Ref. [44]. This is shown in Fig. 11. All data shown
 353 in this plot were re-normalized with the same standard.

354 One can see that there is a good overall agreement of
 355 the $n\text{ELBE}$ data compared to the other selected data
 356 sets presented here. While the ratio of the $n\text{ELBE}$ and
 357 the Tovesson *et al.* and Salvador-Castiñeira *et al.* data
 358 is about 0.99, larger discrepancies to the Matei *et al.*
 359 and Weigmann *et al.* data were observed especially in
 360 the plateau region between 1.2 - 5 MeV. This is of spe-
 361 cial interest, because the current European evaluation
 362 JEFF-3.3 [45] relies mainly on the latter one [29]. A
 363 comparison of shape and scale parameters of the other
 364 experimental data sets with respect to the $n\text{ELBE}$ data
 365 is listed in Tab. IV. The average deviations with respect
 366 to ENDF/B-VIII.0 (shown in Fig. 12) are presented in
 367 Fig. 13, where the residuals of the EXFOR data are ap-
 368 proximated by a constant. All experiments shown are
 369 on average in good agreement within their total uncer-
 370 tainties. The experimental data of [8, 10, 12, 42] and this
 371 work on the average tend to be 4% lower than ENDF/B-
 372 VIII.0. In these experiments different neutron sources
 373 (spallation, photo-neutron and quasi-monoenergetic neu-
 374 trons) with different reference reactions as well as differ-

375 ent target-beam combinations were used. It seems that
 376 the systematic effects in these experiment were taken
 377 into account in a realistic way resulting in a consistent
 378 weighted average with less than 2% uncertainty.

TABLE IV. Average deviations $\Delta = \frac{\sigma^{\text{EXFOR}}}{\sigma^{n\text{ELBE}}} - 1$ of the measured $^{242}\text{Pu}(n,f)$ cross sections with respect to selected EXFOR data in the energy range of 0.5-10 MeV. The listed reduced chi square (χ^2/n) and the p -value are a measure for the agreement in shape.

Measurement	Δ in %	$n\text{ELBE}$ χ^2/n	p in %
Weigmann <i>et al.</i> , 1984	3.82(16)	229.76 / 161 = 1.43	0
Staples <i>et al.</i> , 1998	-2.59(20)	164.68 / 100 = 1.65	0
Tovesson <i>et al.</i> , 2009	0.67(16)	178.69 / 259 = 0.69	100
Salvador-Castiñeira <i>et al.</i> , 2015	0.7(3)	179.46 / 23 = 7.80	0
Matei <i>et al.</i> , 2017	4.6(8)	5.43 / 4 = 1.36	25
Marini <i>et al.</i> , 2017	2(1)	8.19 / 3 = 2.73	4

A. Uncertainties

380 Table V gives an overview of the respective contribu-
 381 tions to the statistical and systematic uncertainties.

382 For an energy range of 0.87 to 8.5 MeV the sta-
 383 tistical uncertainty of the background-corrected counts
 384 within a 2 ns time-of-flight binning is below 3%. The
 385 highest significance is reached in the plateau region,
 386 whereas the largest uncertainties are in the threshold and
 387 second-chance fission region, where the neutron fluence
 388 of $n\text{ELBE}$ is too low to achieve better statistics within
 389 the available measuring time of 80 h.

390 The systematic contributions from the reference cross
 391 section and the scattering corrections described in sec-
 392 tion IIF are always below 1% over the whole energy
 393 range. The effect of fission fragment detection ineffi-
 394 ciency caused by the fragment anisotropy at high neu-
 395 tron energies (discussed in section IIE) increases with
 396 increasing neutron energy and is 1.6% on average for
 397 the included energy range. The largest contribution to
 398 the combined averaged systematic uncertainty of 2.9%,
 399 though, results from the uncertainty on the target area
 400 ($\sigma_A/A \approx 1.1\%$). Although radiographic images show a

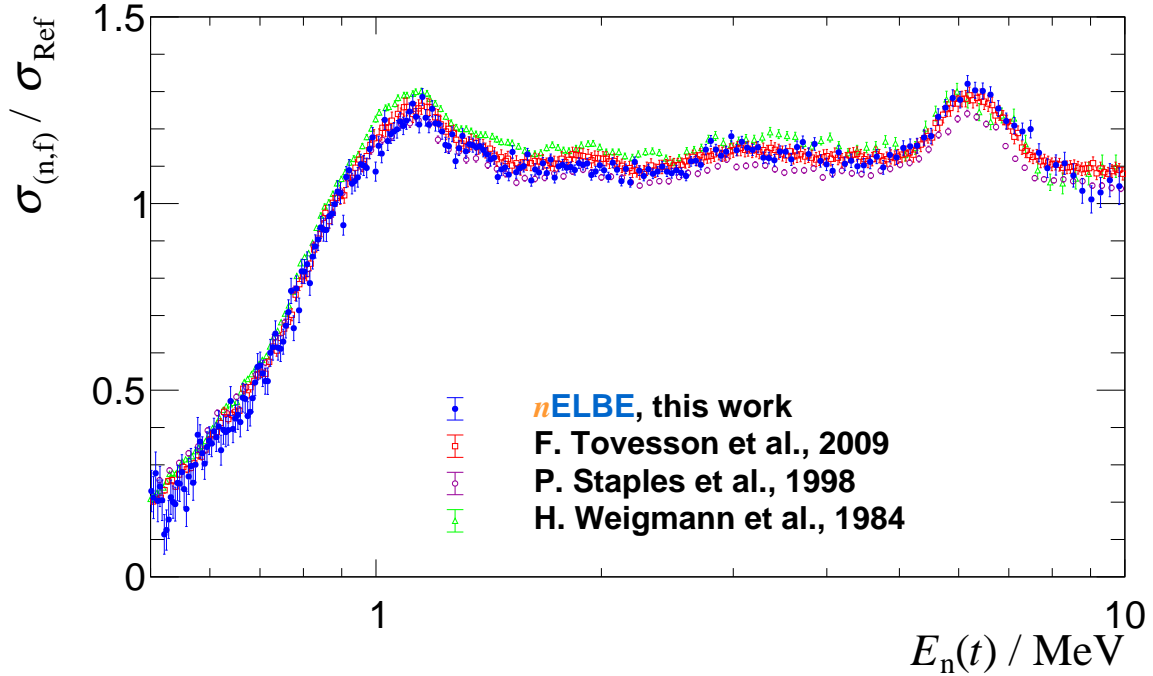


FIG. 10. Neutron-induced fission cross section of ^{242}Pu relative to the one of ^{235}U . The $n\text{ELBE}$ data are shown in blue together with selected EXFOR-data of Tovesson *et al.* [10], Staples *et al.* [42] and Weigmann *et al.* [29]. Within their statistical uncertainties, there is a good agreement of the presented data set with the data of Tovesson. Small deviations from the Weigmann and Staples data are clearly visible.

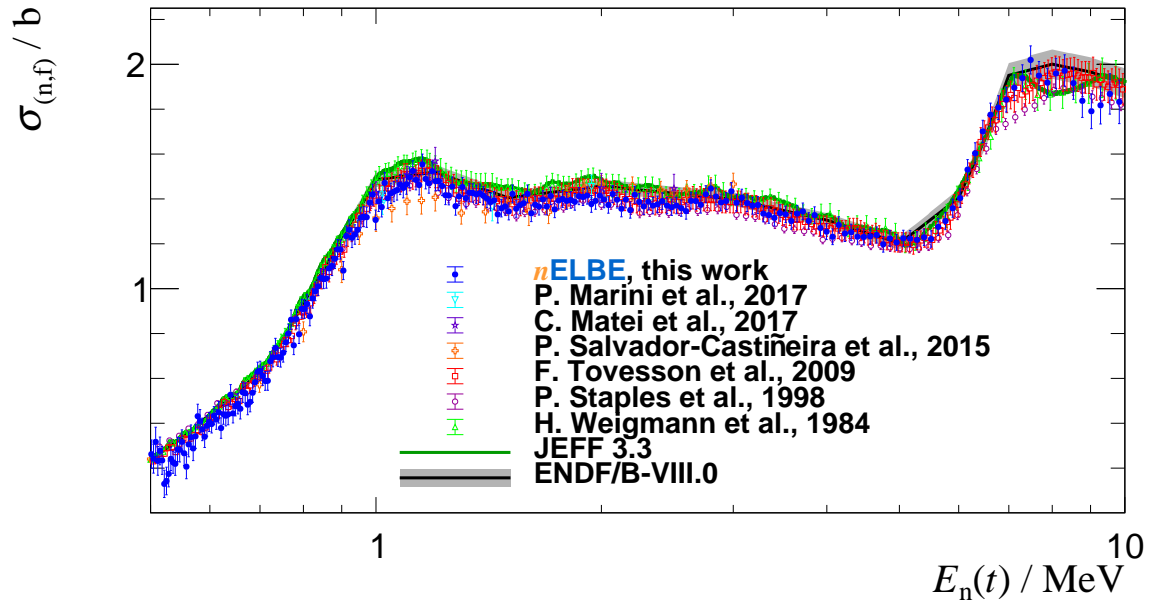


FIG. 11. Neutron-induced fission cross section of ^{242}Pu . The $n\text{ELBE}$ data are shown in blue together with selected EXFOR-data of Tovesson *et al.* [10], Staples *et al.* [42], Weigmann *et al.* [29] and Salvador-Castiñeira *et al.* [8]. Within their total uncertainties, there is a good agreement of the presented data set with the data of Tovesson. Small deviations from the Weigmann data and the measurement of Salvador-Castiñeira are clearly visible.

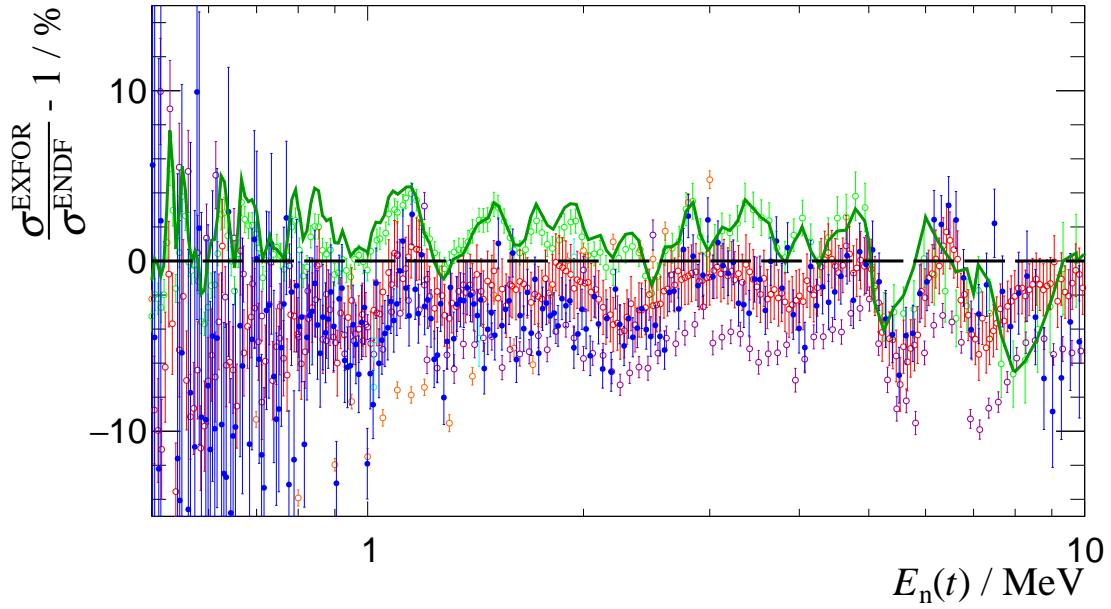


FIG. 12. Residuals of the discussed data sets shown in Fig. 11 with respect to the ENDF/B-VIII.0 evaluation. The error bars plotted here only represent the statistical uncertainty of the measurements. The used color code is identical to that in Fig. 11

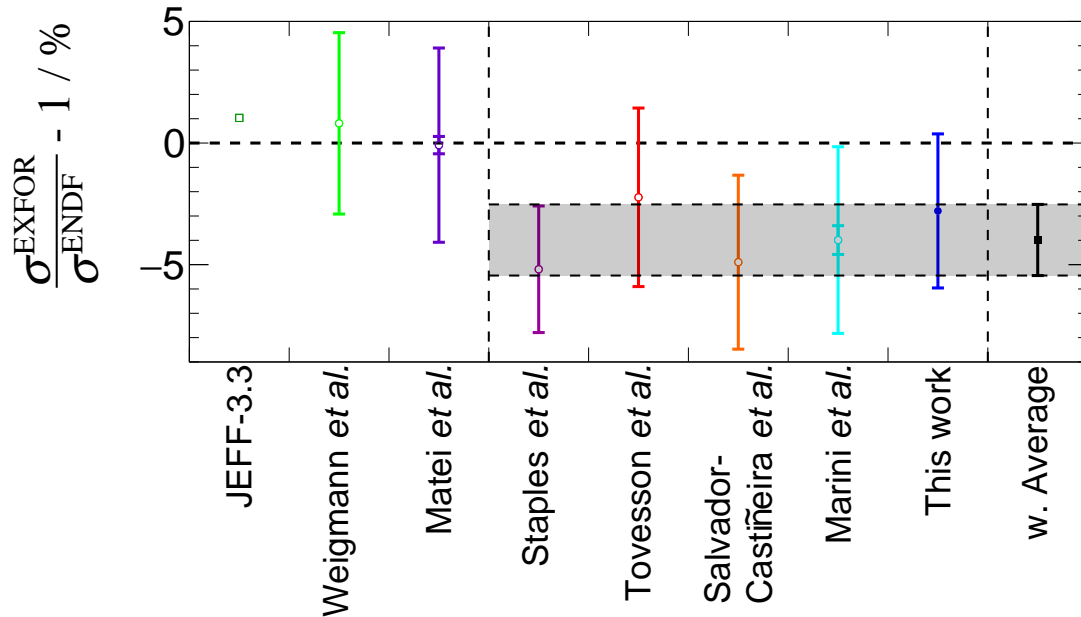


FIG. 13. Average deviations of JEFF-3.3 and selected EXFOR data sets with respect to ENDF/B-VIII.0. The weighted average has been determined by fitting a constant to the residuals shown in Fig. 12. Error bars indicating both, the statistical and the total uncertainty, are drawn for each data point. With the exception of the Weigmann *et al.* and Matei *et al.* data, all recent measurements tend on average to 4.1(15)% smaller cross-sections compared to ENDF/B-VIII.0.

TABLE V. Contributions to the 1σ -uncertainty of the determined cross section for neutron energies between 0.5-10 MeV and a time-of-flight binning of 2 ns.

Contribution	$\Delta x/x$ in %		
	min	max	mean
statistical			
counting statistics	1.2	47.4	4.9
scattering correction C^a	0.17	0.93	0.21
systematic			
normalization K			2.3
reference cross-section σ_{Ref}	0.6	0.8	0.7
scattering correction C^b	0.17	0.93	0.21
Inefficiency I , cf. Fig. 6	0.38	0.43	0.41
combined:			2.9

^a The uncertainty given here only reflects the counting statistics of the simulation

^b From the propagated uncertainty of the underlying total cross sections

very homogeneous activity distribution along the whole surface (cf. Fig. 4.1.4. and 4.1.5 in Ref. [27]), the distribution at the target edges is not assessable. A conservative assumption was taken here to consider edge effects in the order of 0.4 mm with respect to the target diameter.

B. Comparison with state of the art nuclear model codes

Recent nuclear model calculations show substantial deviations in comparison to all experimental neutron-induced fission cross section data of ^{242}Pu . This is exemplarily demonstrated in Fig. 14.

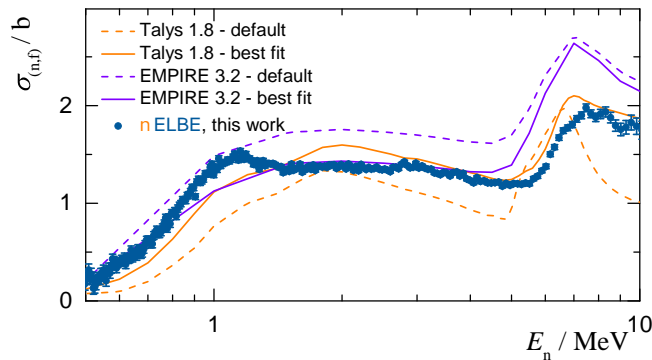


FIG. 14. Comparison of the $n\text{ELBE}$ data with nuclear model calculations from Talys 1.8 and EMPIRE 3.2.

Here the $n\text{ELBE}$ data are shown together with results from calculations performed with the nuclear model code Talys 1.8 [46] and with EMPIRE 3.2 [47, 48]. For both

one calculation was performed with the default settings of the code and one with an improved set of parameters. For EMPIRE, the fission barrier heights and widths have been adjusted to fit with the data. The same was also done in Talys, but here widths, heights and additional parameters of the “Adjusted Input-Parameters” of TENDL 2017 [49] were used.

The results demonstrate that nuclear fission is one of the most complex nuclear reactions and that current nuclear model codes cannot yet predict fission cross sections with the accuracy required for some technological applications.

IV. CONCLUSIONS

The fast neutron-induced fission cross section of ^{242}Pu has been measured in the range of 0.5-10 MeV at $n\text{ELBE}$. It is in good agreement to recent experimental data from different neutron facilities. The $n\text{ELBE}$ data shows a smaller cross section compared to recently evaluated data. In the plateau region (1.3 to 5.0 MeV), the agreement with the Staples *et al.* ($\Delta = -2.51(24)\%$, $\chi^2/n = 1.12$) and Tovesson *et al.* ($\Delta = 0.83(20)\%$, $\chi^2/n = 0.54$) data is excellent. We encountered deviations from the data of Weigmann *et al.* ($\Delta = 3.81(19)\%$, $\chi^2/n = 1.16$), which the JEFF-3.3 evaluation is mainly based on. At the plateau, where $n\text{ELBE}$ has the largest neutron fluence, we achieved a statistical uncertainty of 1.1%. The systematic uncertainty is dominated by edge effects of the actinide targets and is in the order of 2.9% on average over the measured energy range.

It has been shown that neutron scattering corrections are crucial in analyzing neutron time-of-flight experiments. For the present data, the average correction was around 9%.

In comparison to state of the art nuclear model codes like Talys 1.8 and EMPIRE 3.2, deviations of about 20% to 30% from all experiments are observed. This might be indicative of the predictive power of such codes on an absolute scale for neutron-induced fission cross sections of the minor actinides. Precise measurements remain the basis for nuclear data evaluation of fission cross sections.

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