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Scission Neutrons in Spontaneous and Neutron-Induced Fission: Effect on Prompt Fission Neutron Spectra

Robert C. Haight

Los Alamos National Laboratory Guest Scientist

February 2020

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Report of IAEA-NDS Consultant

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ABSTRACT

This consultant was asked to look into the possibility of so-called "scission neutrons", that is neutrons emitted in the fission process before full acceleration of the two large fragments. Results of new measurements that measure neutron emission relative to the direction of the fragments are available, and the quantification of scission neutron has been derived from these data. More detailed models of the fission process are also new. It is however the conclusion of this consultant that the existence of scission neutrons has not been proven from experimental data. Further, the possibility of some pre-equilibrium process producing high energy neutrons in spontaneous fission or in fission induced by low energy neutrons is also not confirmed. Recommendations are made, with a principal one being that detailed modelling of neutron scattering in the analysis of experimental data is of utmost importance. The data base that pertains to scission neutrons and pre-equilibrium neutrons from the fission process is limited, although the recent experimental data could be mined for more information

February 2020

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Introduction

Knowledge of the number and spectrum of neutrons emitted in nuclear fission is essential for applications in nuclear energy, defence, criticality safety and treaty verification. Much experimental work has been done over nearly 80 years to establish a good data base for these quantities for many isotopes of interest and for many reactions including spontaneous fission, neutron-induced fission, and photo fission. Nuclear models have been developed to predict fission observables for unmeasured isotopes and to interpolate and extrapolate the measured data into unmeasured regions. The models have also been used to understand discrepancies in the experimental results. Finally, specific details of the fission process are of interest for basic physics in the development of models of fission, and at present there is a resurgence in the development of models (not discussed here) based on density-functional approach, time-dependent Hartree-Foch calculations, Langevin descriptions of the fission barrier, and molecular dynamic models.

The specific process that this consultant was asked for comments is "scission neutrons". That is the hypothetical process of neutrons being emitted before the fission fragments are fully accelerated. This process is thought to be small compared with total neutron emission, as the model of neutron emission from the fully accelerated fragments is accepted as the major process of neutron emission. "Scission neutrons", if they exist, would modify the total prompt fission neutron spectrum (PFNS) and give it a different shape from that calculated for neutron emission from fully accelerated fragments.

Three types of experiments are invoked to shed light on the question of what extent, if any, are scission neutrons present in prompt fission neutron spectra.

a) Measurement of the PFNS and comparison with model predictions
b) Measurement of the correlation between a fission fragment (direction, TKE, Z-A split etc.) and one fission neutron (direction, energy)
c) Measurement of the correlation between two or more neutrons emitted in PFNS.

The first and the third are most directly relevant to applications, but the signature, if any, of scission neutrons is small as they are in the background of neutrons emitted from fully accelerated fission fragments. The first is of course essential to the study of critical assemblies. The second is more relevant to the development of models of fission. The third is relevant to model development and also to safeguards applications.

For this consultantship, I was asked also to comment on another possible feature of prompt neutron emission, that is neutron emission in fission in some non-equilibrium process. A signature here would be excess neutrons in the high energy region, say above 8 or 10 MeV. The relationship to scission neutrons is not obvious here.

In addition to archival literature, I was given reports from contracts sponsored by the IAEA-Nuclear Data Section, vugraphs from a recent meeting (THEORY-5) and from a recent presentation, and a recently published paper:

1. IAEA technical report **INDC(NDS)-0808**, Vienna 2019: "Scission neutrons from thermal-neutron induced fission of ^{239}Pu and spontaneous fission of ^{252}Cf" by A.S. Vorobyev and O.A. Shcherbakov, Petersburg Nuclear Physics Institute of National Research Center "Kurchatov Institute", Gatchina, Russia [Vorobyev CR2019].
2. IAEA technical report **INDC(NDS)-0809**, Vienna 2019: "Experimental Investigation of the Properties of Scission Neutrons in Thermal-Neutron Induced Fission of ^{233}U and ^{235}U," by A.S. Vorobyev and O.A. Shcherbakov, Petersburg Nuclear Physics Institute of National Research Center "Kurchatov Institute", Gatchina, Russia [Vorobyev CR2018].
3. "Studies on Prompt Fission Neutron Emission at JRC-Geel," by Alf Göök, F.-J. Hambsch, S. Oberstedt, vugraphs presented at THEORY-5, Castelvecchio Pascoli (Italy), 24-26/09/2019 [Göök 2019].
4. "Non-evaporation neutrons in PFNS at outgoing neutron energies larger than 8 MeV," R. Capote, vugraphs from presentation at THEORY-5, Castelvecchio Pascoli (Italy), 24-26/09/2019 [Capote 2019].
5. "Investigation of the Prompt Neutron Emission Mechanism in Low Energy Fission of ^{233,235}U(nth,f) and ^{252}Cf(sf)," A. S. Vorobyev, O. A. Shcherbakov, A. M. Gagarski, G. V. Val'ski, and G. A. Petrov, vugraphs from EFNUDAT (European Facilities for Nuclear Data Measurements) project workshop held from April, 28-30, 2009 at EC-JRC-IRMM, Geel, Belgium 2009. Full paper is in EPJ 8, 03004 (2010) [Vorobyev 2009].
6. "Prompt Fission Neutron Spectra of Actinides," R. Capote et al., Nuclear Data Sheets 131 (2016) 1–106 [Capote 2016].

The archival literature included a careful study of the high-energy tail of the ^{252}Cf(s.f.) spectrum:

7. "Results of a Low Background Measurement of the Fission Neutron Spectrum from ^{252}Cf in the 9- to 29-MeV Energy Range," by A. Chalupka, L. Malek, S. Tagesen & R. Böttger, Nuclear Science and Engineering, **106**, 367-376, (1990) [Chalupka 1990].

Our work at LANSCE on pre-equilibrium emission in fission induced by fast neutrons has been published [Kelly 2019A]. The process is not treated here as it is for neutron energies above first-chance fission and so pre-equilibrium emission prior to fission can be significant.

I also had access to two submitted but still unpublished works on PFNS measurements of ^{239}Pu(n,f) with fission induced by neutrons in the range 1 to 20 MeV and much higher also in one of the papers. Both experiments were carried out at the Los Alamos Neutron Science Center, both papers were submitted to Physical Review C for publication, and I am a co-author on both. The papers are for different measurements and they have some commonality but also many differences. At the time of writing this report, I did not have numerical data for these experiments.

8. [Marini 2019] "Prompt Fission Neutron Spectra in the ^{239}Pu(n,f) Reaction," by P. Marini, et al., (to be published); preprint available as http://arxiv.org/abs/1902.07951

9. [Kelly 2019B] "Combined 6Li-glass-Liquid Scintillator Measurement of the ^{239}Pu(n,f) Prompt Fission Neutron Spectrum from 10 keV to 10 MeV Induced by 1-20 MeV Neutrons," by K. J. Kelly et al.

Dr. Capote pointed me to experiments at Rez by Košťál et al., [e.g. Kostal 2017] where more high energy fission neutrons were observed than predicted by some models. This is for fission of ^{235}U induced by thermal neutrons.

II. Comment on Relationship to Ternary Fission:

The process of emission of "scission neutrons" is usually considered in the context of "ternary fission" where a light charged particle (LCP) is emitted. Models that account for the observables of energy and direction of the LCP relative to one of the fragment's direction indicate that the LCP is formed between the two large fission fragments, and that accounts for its kinetic energy, from Coulomb repulsion, and its direction, close to 90-degrees relative to the direction of the fission fragments.

Classical analogise to ternary fission are well known in the field of fluid dynamics. I personally have seen a demonstration of pulsed water that result in two large, daughter droplets with a much smaller third droplet in between. Droplet division and separation are important in inkjet printers. Electro-spraying has the phenomenon of fission, and that is interesting in that it involves Coulomb forces as well.

Because light charged particles can be emitted in fission before the larger two fragments separate, it is not much of a stretch of the imagination to conclude that neutrons could also be emitted before the two fragments are fully accelerated. In the case of neutrons, there is, of course, no Coulomb acceleration and therefore no energy boost.

We might use ternary fission, where charged particles are produced, to guide our expectation of scission neutrons. Ternary fission with the emission of light charged particles is an uncommon event. Alpha particles are emitted in ternary fission only about 2 times in 1000 fissions [Wagemans 1991]. The emission of protons is down by another factor of 50 to 100. These values do not encourage one to look for scission neutrons, but perhaps because of charge polarization or some other effect, the probability of scission neutrons could be measurable.

We need to ask, what sort of neutron output in energy and angle could be expected from scission neutrons? There are models and speculations, and they include:

1. Emission of scission neutrons at angles near 90-degrees, similar to light-charged particle emission in ternary fission. These neutrons might have a rather soft distribution in energy. Maxwellian distributions with a temperature of around 1 to 1.3 MeV have been proposed.

2. Other processes that are non-equilibrium, perhaps call them "pre-equilibrium," and these might exist for first-chance fission. The neutron spectrum could be altered at high energies, and, presumably, at low energies as well. The effect might be more visible at high neutron energies where neutron evaporation from fully accelerated fragments is not so large as to obscure any pre-equilibrium emission.

III. Status of our knowledge of scission neutrons:

Experimental situation: The importance of scission neutrons, and even their existence, has been summarized recently in the reports by Vorobyev et al. [Vorobyev 2018, Vorobyev 2019] and for fission of ^{233,235}U and ^{239}Pu induced by thermal neutrons and for spontaneous fission of ^{252}Cf (see tables). They give the following assessment: "It can be concluded … that the yield of "scission" neutrons is the less, the more sophisticated becomes an experimental technique and the more detailed becomes an analysis of all possible systematic effects."

The range of experimental data is unfortunately not large. It comprises:

1. Experiments that detect two fission fragments and the associated fission neutrons as a function of angle relative to the fission axis. In principle, these measurements could also investigate the dependence on the split into fission products, Z1-A1 and Z2-A2. The systems studied are:

a. Spontaneous fission of ^{252}Cf.
b. Thermal neutron fission of ^{233}U, ^{235}U and ^{239}Pu
c. Fission of ^{235}U and ^{239}Pu by neutrons in the resonance regions

2. Fission neutron spectra

a. Spontaneous fission of ^{252}Cf and a few other actinides
b. Neutron-induced fission from thermal to over 100 MeV. (We will limit the range to energies where only first-chance fission is allowed.) These data can include angular distributions of the fission neutrons relative to the incident neutron direction. For thermal neutrons, this is not an issue. But for fast neutrons incident, in the keV range, only recently has one group measured also the neutrons relative to both the incident neutron direction and the fission axis.

3. Correlated neutron emission (n-n) from fission.

IV. Situation as seen from the modelling perspective:

This consultant is not at all an expert in the models. However, it seems to be accepted that "scission neutrons", if they exist, will be emitted preferentially toward 90-degrees with respect to the fission axis.

Even if they are emitted isotopically, then the best place to look for them is where neutrons emitted from the fully accelerated fragments (principal component) are the least, which is near 90-degrees, as the kinematic boost will to some extent carry the neutrons emitted from the fully accelerated fission fragments in the direction of those fragments. In either case, the angular dependence of the principal component needs to be well understood from the models. This is a subject of active interest at present.

The status of models was summarized recently by Schmidt and Jurado [Schmidt 2018]: "Therefore, the deexcitation process does not carry so much information on the fission process, if we disregard the quest for scission neutrons, which are neutrons of non-statistical nature emitted at scission and whose existence has been controversially discussed, as well as efforts to better comprehend the generation of angular momentum of the fission fragments, which is still not well understood."

The principal mechanism of neutron emission from fully accelerated fission fragments involves the angular momentum of these fragments. The angular distribution of neutrons emitted from them must be known in order to identify the other process of scission neutrons. At present, the identification of scission neutrons depends on models that have different physics or different parameters. It is my understanding that modelling efforts in the US are making good progress in incorporating angular momentum in the fission fragments. Such work might guide a better understanding of the angular distribution of neutrons emitted from the fully accelerated fragments.

Table 1. Main results of the investigations of neutron emission mechanism for $^{252}Cf(sf)$.

Authors, References, Experimental Set-up	Yield of "scission" neutrons	Average energy of "scission" neutrons	Anisotropy of PFN emission in the centre-of-mass system of fission fragment, A_2
Investigation of (n,f)-angular correlation			
H.R. Bowman *et al* [5, 6] (1962). Two plastic scint. for FFs spectroscopy (TOF 100cm);Two neutron detectors (plastic scint.) were used, TOF(100cm). One of them was placed at 11.25° relative to FFs direction. The position of the second detector was varied from 22.5° to 90° in step 11.25°.	10%	2.6 MeV	0.016 ± 0.012
V.M. Piksaikin *et al* [7] (1977). Two Si-surface barrier detectors for FFs spectroscopy; Single-crystal (stilbene) proton-recoil spectrometer was placed interchangeably at 8.7° and 90° relative to FFs direction, n/γ pulse shape discrimination.	20%	1.52 MeV	Not investigated
Yu.S. Zamyatnin *et al* [8] (1979). IC with collimator used for FFs spectroscopy; One neutron detector (plastic scint.) was placed interchangeably at *0° and 90° relative to FFs* direction, TOF (40cm).	15 ± 8%	---	Not investigated
P. Riehs [9] (1981). Two Si-surface barrier detectors for FFs spectroscopy; Two neutron detectors (NE213) were placed at 0° and 90° relative to FFs direction, n/γ pulse shape discrimination, TOF(75cm).	5.9 ± 1.3 % (13.2 ± 3.1%)	2.0 MeV	Not investigated
D. Ward *et al* [10] (1983). Three Si-surface barrier detectors to set direction of FFs (0°, 45° and 90°) relative to axis of neutron detector (NE213), n/γ pulse shape discrimination, TOF(34cm)	10 ± 5%	---	Not investigated
E.A. Seregina *et al* [11, 12] (1985). Six Si-surface barrier detectors to set direction of FFs (from 0° to 90° in interval 10°) relative to axis of neutron detector (Single-crystal stilbene proton-recoil spectrometer), n/γ pulse shape discrimination.	10.6 ± 2.1 %	1.5 ± 0.3 MeV	0.07
H. Marten *et al* [13] (1991). Two parallel-plate avalanche counters for selection of FFs group; Two neutron detectors NE912(*6-Li glass*) and NE213, TOF(35, 160cm	< 5%	---	0.06
O.I. Batenkov *et al* [14] (1989). Micro channel plates used for FFs spectroscopy (TOF with base 9 cm); Two neutron detectors (stilbene) arranged collinearly were used, n/γ pulse shape discrimination, TOF (37.5, 75, 150cm).	2 - 3 %	---	0.04

Table 1. Main results of the investigations of neutron emission mechanism for $^{252}Cf(sf)$ (continued).

Authors, References, Experimental Set-up	Yield of "scission" neutrons	Average energy of "scission" neutrons	Anisotropy of PFN emission in the centre-of-mass system of fission fragment, A_2
Investigation of (n,f)-angular correlation			
H.-H. Knitter *et al* [15, 16] (1992). IC used for FFs spectroscopy and measuring FF angle; One neutron detector (NE213), n/γ pulse shape discrimination, TOF(50 cm).	1.1 ± 0.3 %	0.4 MeV	0.01 ± 0.02
Yu.D. Kamarzhnov *et al* [17] (1998). Two Si-surface barrier for FFs spectroscopy; Two neutron detectors (NE213) were placed at 0° and 90° relative to FFs direction, n/γ pulse shape discrimination, TOF(75cm).	2.5 ± 1.0 %	---	Not investigated
A.S. Vorobyev *et al* [18] (2010). 16 MWPC for FFs spectroscopy (TOF with base 14cm); Two stilbene neutron detectors were used, n/γ pulse shape discrimination, TOF (~50cm). The neutron spectra measurements were done simultaneously for angles ranging from 0° to 180° in interval 18° relative to the light FFs direction.	≤ 5%	---	0.04
Investigation of (n,n)-angular correlation			
J.S. Pringle and F.D. Brooks [19] (1975). Two neutron detectors (NE213), n/γ pulse shape discrimination, TOF (30, 40 and 50cm).	< 10 %	---	Not investigated
A.M. Gagarski *et al* [20] (2008). Two stilbene neutron detectors, n/γ pulse shape discrimination, TOF (40-70 cm). V.E. Sokolov *et al* [21] (2010) reanalysis of [20].	10 - 11 % 8 ± 3%	1.6 MeV 2.0 ± 0.2 MeV	Weakly sensitive
Compilation of (n,f) data from refs [5], [12] *and* [15]			
N.V.Kornilov *et al* [22] (2001).	~ 10%	Two components with average energy 0.9 MeV and 3.0 MeV.	Not investigated

Table 1. Main results of the investigations of neutron emission mechanism for $^{235}U(n,f)$.

Authors, References Experimental Set-up	Yield of "scission" neutrons	Average energy of "scission" neutrons	Anisotropy of PFN emission in the centre-of-mass system of fission fragment, A_2
Investigation of (n,f)-angular correlation			
K. Skarsvag *et.al.* [4] (1963). A gas scintillation counter with collimator was used as FFs detector; One neutron detector (plastic scintillator), TOF (90cm). The angle between the direction of FFs and neutrons was varied with 15° step by pivoting the FFs counter about an axis.	15%	~ 1.8 MeV	"There is a little or no indication of any anisotropy …"
S.S. Kapoor *et.al.* [6] (1963). Ion chamber scintillation detector used for measuring FF angle and kinetic energy of fission fragment; One neutron detector (plastic scintillator), TOF(103cm).	10%	3.2 MeV	≤ 0.09±0.06 [5]
J.S. Fraser *et.al.* [7] (1966). Two plastic scint. for FFs spectroscopy (TOF with base 125cm and 99cm); Four neutron detectors (plastic scintillator) were used, TOF (106cm). The neutron spectra measurements were done simultaneously at 10°, 25°, 45° and 80° relative to FFs direction.	20%	1.9 MeV	"… all results are consistent with A = 0."
M.S. Samant *et.al.* [23] (1995). IC used for FFs spectroscopy and measuring FF angle; One neutron detector (NE213), n/γ pulse shape discrimination, TOF (70 cm).	10 ± 2%	---	Not investigated
N.V.Kornilov *et.al.* [24] (2001). Compilation of Skarsvag's data [4].	15%	Two components with average energy 0.9 MeV and 3.0 MeV.	
A.S. Vorobyev *et.al.* [25] (2010). 16 MWPC for FFs spectroscopy (TOF with base 14cm); Two stilbene neutron detectors were used, n/γ pulse shape discrimination, TOF (~50cm). The neutron spectra measurements were done simultaneously for angles ranging from 0° to 180° in interval 18° relative to the light FFs direction.	≤ 5%	---	0.04±0.02
Investigation of (n,n)-angular correlation			
C.B. Franklyn *et.al.* [26] (1978). Two stilbene neutron detectors. The measurements were done for angles ranging from 9° to 180° in 9° interval between pairs of fission neutrons.	20%	---	Not investigated
I.S. Guseva *et.al.* [27] (2018). Two stilbene neutron detectors, n/γ pulse shape discrimination.	2.0 ± 1.5%	1.8 ± 0.2 MeV	Weakly sensitive

Table 2. Main results of the investigations of neutron emission mechanism for $^{239}Pu(n_{th},f)$.

Authors, References, Experimental Set-up	Yield of "scission" neutrons	Average energy of "scission" neutrons	Anisotropy of PFN emission in the centre-of-mass system of fission fragment, A_2
Investigation of (n,f)-angular correlation			
J.S. Fraser *et al* [23] (1965). Two plastic scint. for FFs spectroscopy (TOF with the base of 125cm and 99cm). Four neutron detectors (plastic scint.) were used, TOF (106cm). The neutron spectra measurements have been done simultaneously at 10°, 25°, 45° and 80° relative to FFs direction.	30%	~ 2 MeV	"... all results are consistent with A = 0."
Yu.S. Zamyatnin *et al* [8] (1979). IC with collimator used for FFs spectroscopy; One neutron detector (plastic scint.) was placed interchangeably at 0° and 90° relative to FFs direction, TOF (40cm).	20 ± 12 %	---	Not investigated
A.S. Vorobyev *et al* [24] (2018). 8 MWPC for FFs separation (TOF with base 14cm); Two stilbene neutron detectors were used, n/γ pulse shape discrimination, TOF (~50cm). The neutron spectra measurements were done simultaneously for angles ranging from 0° to 180° in interval 18° relative to the light FFs direction.	4.5 ± 0.9%	1.6 ± 0.2 MeV	0.04 ± 0.2
Investigation of (n,n)-angular correlation			
V.E. Sokolov *et al* [25] (2010). Two stilbene neutron detectors, n/γ pulse shape discrimination.	14 ± 3%	1.8 ± 0.2 MeV	Weakly sensitive
I.S. Guseva *et al* [26] (2018) reanalysis of [25].	4.0 ± 1.5%	1.8 ± 0.2 MeV	

V. Some general comments on the experiments measuring neutron emission spectra

These comments apply to experiments using active neutron detectors based on organic scintillators:

1. The contribution of scission neutrons is now generally agreed to be small, on the order of a few percent and certainly not 30% as some authors claimed for many years. Thus, to detect and better quantify the effect, experiments need to be as good as possible and designed from the beginning to reduce systematic effects that would make the results difficult to analyse. Thorough documentation is essential for continuing analyses of the data.
2. Many of the neutron spectrum measurements take the PFNS of ^{252}Cf(s.f.) as a reference. Although this spectrum has been measured many times, there are still uncertainties. If the signature of scission neutrons is in this standard but not recognized as such, the ratio measurements of any other PFNS, spontaneous or neutron-induced, will be interpreted incorrectly and the signatures of any scission neutrons in these other systems could be obscured.
3. Accurate measurements of neutrons around 1 MeV are important as a signature of scission neutrons if the model of a Maxwellian distribution of scission neutrons at T = 1 to 1.3 MeV is accepted. This energy range is difficult experimentally for three reasons:
    a. It is close to the threshold for detecting neutrons with organic scintillation detectors and therefore there are uncertainties in the detector efficiency.
    b. Neutron-gamma-ray separation is more difficult at these neutron energies than at higher energies.
    c. Scattering of neutrons from the primary mechanism of neutron emission by fully accelerated fragments produces neutrons in this energy range as a background.

4. Almost none of the measurements, including several recent ones, take into account neutron scattering with detailed Monte Carlo calculations. One exception is a measurement of PFNS for neutron-induced fission ^{239}Pu at LANSCE, submitted for publication [Kelly 2019B]. For a separate experiment at LANSCE led by researcher from the CEA, the reference ^{252}Cf(s.f.) is used as a reference [Marini 2019] and it can mockup the scattering to first order. However, because the PFNS of ^{252}Cf (s.f.) is different from fission in the U and Pu isotopes, the first is not an exact standard and corrections will still need to be made for neutron scattering.

5. Documentation of recent experiments is far from complete so that effects of neutron scattering from materials in the experimental area cannot be modelled by an outside analyst. Information on the size of the target room, the distance of the experiment from the floor and walls, the presence of scattering material, all are important in modelling.

6. Detector responses are often not documented, and this failing again makes it virtually impossible for the experiment to be modelled. This includes:
    a. What is the detector response to neutrons for the full range of neutron energy? Near threshold, there are usually significant systematic uncertainties. Are they included in the results?
    b. How was the reported threshold determined? Generally, for experiments where a neutron is detected, the threshold is often by reference to a gamma-ray source, in MeV-electron equivalent. But the pulse-height (or pulse area) resolution also needs to be include here. Without more details, one is left wondering how the experiment was analysed.

7. The discussions of uncertainties, for example systematic errors, are often minimal.

VI. Some suggestions for future experiments on fragment – neutron correlation

1. Address the problems listed above.

2. The addition of a rather thin sheet of lead in front of the neutron detectors can reduce the effects of low energy gamma and x-rays but be almost transparent to fission neutrons. Our French colleagues [Marini 2019] have chosen a thickness of 2 mm.

3. Resolution of masses of the fast fission products is clearly an issue. I am not sure I understand it as the details are often lacking. But clearly there are some problems (e.g. Vorobyev vugraph #16 [Vorobyev 2009], comparison of data with those of Nishio and Maslin).

4. Mining existing event-mode data (see below).

VII. Neutron-neutron correlations

As shown in the above tables by Vorobyev, there are not so many measurements of neutron-neutron correlations that have been analysed in the search for scission neutrons. Correlations come from a coincidence between an energetic fission neutron that carries information on the direction of the fission axis (by its kinematically boosted energy) and another neutron. The information on the fission axis therefore has poorer angular resolution than if a fission fragment were detected directly and so the angular distributions from n-n coincidences are not as sharply defined as those from n-fission coincidences. Deviations from the model calculations are then even more difficult in the analyses.

The analysis of n-n correlations by Guseva et al. [Guseva 2018] for thermal neutron fission of ^{235}U does include modern Monte Carlo calculations to address the problem of neutron crosstalk between detectors. These detectors were placed only 51cm from the fissionable sample and so, even though the detectors were shielded, the possibility of crosstalk was still certainly there. The authors made an experimental test of crosstalk with a radioactive Pu-Be neutron source that emits only one neutron at a time. The energy spectrum of this source is not stated and probably is different from a fission neutron spectrum. The Pu-Be neutron spectrum can vary with the composition of the materials. This is because the neutron production mechanism for this source is $^{9}Be(alpha,n)^{12}C$, which does produce neutrons. The Q-value for this reaction is 5.7 MeV and together with the alpha particle energy of up to 5 MeV, neutrons above the mean energy of fission neutrons are produced. The spectrum of neutrons from the Pu-Be neutron source depends on the alpha particle energy as it acts on ^{9}Be, and the alpha particle energies depend on their slowing-down in the material. There is thus a significant uncertainty in the produced neutron spectrum. It is not known if the authors included this uncertainty in their analysis of neutron crosstalk.

If, for example with this harder neutron spectrum from Pu-Be, a greater cross talk correction is applied, then the angular correlation corrected for this effect would reduce the curves at the very forward and very backward correlation and then the mid-range angles would appear to have more events, which could be interpreted as scission neutrons. (Note: this consultant has had experience with crosstalk of neutron detectors [Schuster 2019] and can say with confidence that it is "tricky business" and deserves extensive Monte Carlo modelling where even the uncertainties in the input nuclear data need to be included.)

New measurements on n-n correlations have been made from photo fission of ^{238}U [Burggraf 2020] and they show a dip in the correlation between 165 and 180-degrees. This dip is seen, within statistics, in ^{252}Cf(s.f.) when rather low energy neutrons (e.g. E_{n1} and E_{n2} both being between 0.4 and 1.2 MeV) are selected. The observation that the dip is seen rather generally in ^{238}U(gamma,f) but only for one neutron energy cut in ^{252}Cf(s.f.). This observation needs to be studied further. There is however a suggestion that if two neutrons are emitted from one fragment at, respectively, 0 and 180 degrees to the fission axis, then the other fragment could possibly shadow (or scatter) the neutron emitted toward that fragment. This would be an additional physical process and it could lead to (scattered) neutrons at other angles and could increase the probability of neutrons scattered at 90-degrees to the fission axis. The authors note also that there is disagreement between the data and FREYA for the correlations in this angular range.

Lestone comes to the same conclusion: "As in the ^{235}U(nth,f) reaction, the predicted ^{252}Cf n-n angular correlations (solid curves) are a little stronger than the experimental results. This discrepancy can be significantly reduced by the inclusion of a neutron source that is not kinematically boosted by the motion of the fission fragments. However, at this point in time, we cannot rule out that a small amount of neutron scattering has led to a slight washing out of the measured n-n angular correlations. In our judgement, the inclusion of a scission-neutron source is not warranted, at the present time, until additional well documented experiments and the analysis thereof become available." [Lestone 2016]

VIII. Search for high-energy neutrons and gamma rays:

The fission process has plenty of energy (~ 180 MeV) and it is conceivable that, in some unknown pre-equilibrium process, a good fraction of that energy could go into emission of a high energy neutrons or gamma rays.

For ^{252}Cf(s.f.) there appears to be no excess of fission neutrons in the high energy region. For example, Browne and Dietrich [Browne 1974] found that literature data on the neutron spectrum could be accounted for with a Hauser-Feshbach calculation, and they found no deviation from a Maxwellian up to 15 MeV emitted neutron energy.

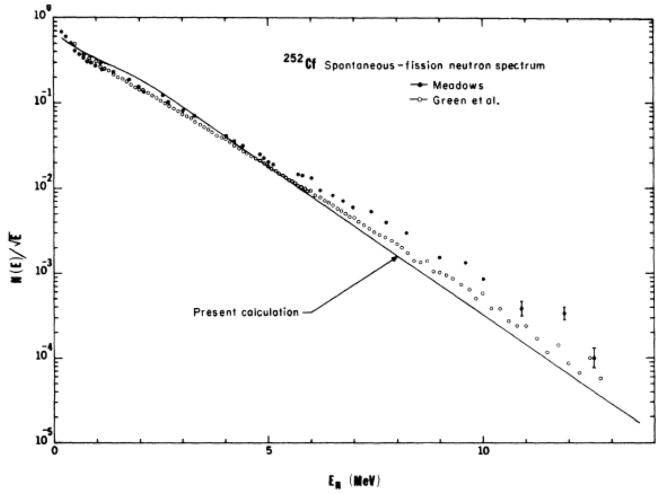

FIG. 2. Experimental and calculated neutron spectra (laboratory energy) for ^{252}Cf spontaneous fission. The solid line represents the results obtained by dividing the calculated $N(E)$ by the calculated value of $\bar{\nu} = 3.64$ so that the integral of $N(E)$ is equal to one as is the case for Meadows's data. The Green *et al.* data are normalized to Meadows's data at 2 MeV.

Figure: "Hauser-Feshbach calculation of the ^{252}Cf spontaneous-fission neutron spectrum," [Browne 1974]

The real clincher (in the opinion of this consultant) for the high end of the ^{252}Cf(s.f.) spectrum was done in a low-background environment. Neutrons from ^{252}Cf(s.f.) were investigated in a very clean experiment done underground: "Results of a Low Background Measurement of the Fission Neutron Spectrum from ^{252}Cf in the 9- to 29-MeV Energy Range," by A. Chalupka et al. [Chalupka 1990]. They say: "The results of our neutron energy spectrum measurement especially designed to cope with the problems in the high-energy region do not show any significant deviation from a Maxwellian shape with T = 1.42 MeV in the region between 20- and 29-MeV neutron energy."

My conclusion is that there is no credible evidence that high energy neutrons are produced in spontaneous fission of ^{252}Cf. Should there be high energy neutrons from fission of other isotopes induced by thermal or fast neutrons? The question is open.

Spectrum-average measurements of the high energy tail of the fission neutrons from thermal neutron fission has been investigated in several works summarized in the International Reactor Dosimetry File, IRDFF-II [Trkov 2019]. New measurements of with fission neutrons from thermal neutron fission of ^{235}U have been reported by Košťál et al. [Kostal 2017] at the Rez reactor near Prague for thermal neutron induced fission of ^{235}U. The experiments were by activation foils with thresholds in the 10.5-13.0 MeV energy range. These new results confirm the harder prompt fission neutron spectrum in the CIELO evaluation than in the ENDF/B-VII evaluation. The new evaluation of the PFNS for thermal-neutron-induced fission of ^{235}U was made without the need for some new mechanism for production of high energy fission neutrons.

Pre-equilibrium neutrons, emitted prior to fission, have been observed and quantified by my colleagues at Los Alamos for fission induced by neutrons above first-chance fission.

These results do not pertain to pre-equilibrium mechanisms of the fission process itself. [Kelly 2019A]. Instead, they can be explained in the well-known preequilibrium process of neutron emission seen in reactions on non-fissionable nuclei. The angular distribution of pre-equilibrium neutron emission and the threshold for this effect (incident neutron energies ~ 6 MeV) are the key observables in this case.

For high energy gamma rays from ^{252}Cf(s.f.), Dietrich et al. [Dietrich 1974] detected gamma rays up to 17 MeV. With excitation energy in this range, the implication is that a fission fragment could emit a high energy neutron. The neutron energy would of course be less than the excitation energy due to the binding energy of that neutron. Any gamma rays above 17 MeV were of such low abundance that they were not reported. A more modern measurement quantified gamma-rays up to about 20 MeV in thermal neutron fission of 235U [Makii 2019]. Again, a pre-equilibrium process does not seem to be required to reproduce the measured data.

From Makii et al. [Makii 2019].

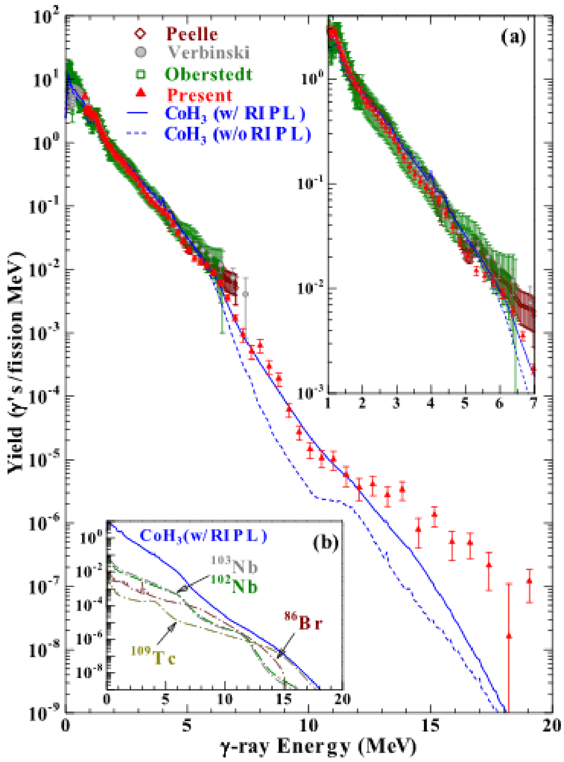

FIG. 2. Prompt γ-ray spectra obtained in the present study (filled triangles) are compared to those by Peelle *et al.* (open diamonds) [7], Verbinski *et al.* (filled circles) [9], and Oberstedt *et al.* (open rectangles) [4]. The spectra are compared with the calculation by the CoH_3 code [44] (solid and dashed lines); see text for details. Expansion of the spectra in the energy range $E_\gamma = 1$–7 MeV is shown in the inset (a). The inset (b) shows the γ-ray spectra for ^{86}Br, ^{102,103}Nb, and ^{109}Tc with the CoH_3 code [44] (dashed-dotted lines).

IX. Mining data from recent experiments for more results:

Data from the work of Göök et al. and Vorobyev et al. could be mined for more information relevant to the possibility of scission neutrons. They have large sets of event-mode data where fragment masses and energies are measured together with the energy and direction of associated neutrons. The question is, are their regions where scission neutrons might be better identified? Here are some suggestions.

- Focus on different regions of the PFNS spectrum (my guess – look at the angular distribution of fission neutrons relative to the fission axis in the 0.2 to 1.5 MeV range (and perhaps other ranges) of fission neutrons instead of the full integral of the neutron spectrum).
- Focus on different regions of total kinetic energy (my guess would be that scission neutrons – long neck – would be more clearly seen at low TKE).
- Focus on symmetric versus asymmetric fission (no guess here).
- Combinations of the above.

Of course, when one takes smaller and smaller slices of the event-mode data, there are fewer and fewer counts, and these suggestions might end up with insufficient statistics for any conclusions. As is written above, good analyses of neutron scattering by Monte Carlo simulation of the experimental environments is still essential.

X. Unknown unknowns:

Given the recent experimental progress, the present data barely scratch the surface of the subject of scission neutrons. Significant gaps exist in the following for possible measurements where fission fragment properties, including direction, are measured along with neutron emission. These include:

- Fission induced by fast neutrons, even for the common $^{233,235,238}U$ and ^{239}Pu.
- Fission of light actinides, e.g. thorium
- Fission of minor actinides induced by thermal as well as fast neutrons.

XI. Applications:

Nearly all applications do not care what the direction of the fission fragments is with respect to the emitted neutron. This observable is of use in validating nuclear models so that they might account more accurately to the gross PFNS. One approach is simply to parameterize measured data for the PFNS with respect to emitted energy spectra versus angle. A model of emission from fully accelerated fragments should be used in this approach. Given the present question about even the existence of scission neutrons, that approach might be the best now.

There are applications, e.g. in safeguards, where neutron-neutron correlations in energy and angle are important, and these should be kept in mind in the development of improved models and in the design of experiments.

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