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Unifying measurement of ^{239}Pu(n,γ) in the keV to MeV regime

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A single, unifying measurement of the ^{239}Pu capture cross section from 1 keV to 1.3 MeV has been performed for the first time using the Detector for Advanced Neutron Capture Experiments (DANCE) at the Los Alamos Neutron Science Center (LANSCE). The experimental method combines a prior experiment's characterization of prompt fission γ-rays in conjunction with a fission tagging detector with a separate experiment using a thick ^{239}Pu sample to extract the neutron capture cross section in ratio to ^{239}Pu(n,f). We have made new predictions of the capture cross section taking into account recent results for the M1 scissors mode present in other actinides. The results show deviations from current evaluations which are 30% higher at the highest energies, and will be used to improve calculations relevant for several applications.

Neutron capture on ^{239}Pu above 1 keV was first published six decades ago and remeasured a number of times since [1–7], yet experimental constraints on this quantity are still inadequate for the needs of modern application in nuclear technology and discrepancies exist between the major nuclear data evaluations [8–10]. This is largely caused by the difficulty of separating fission decay γ-rays from capture γ-rays on fissile nuclei combined with ^{239}Pu's unique challenges as a spontaneous α-emitter and a small capture to fission ratio above 100 keV. Data points reported by prior work are relatively sparse and widely discrepant, and uncertainties across the entire region need to be reduced to meet the needs of advanced reactor design studies as well as transmutation in the actinides [11, 12].

A new method of determining capture cross sections on fissile nuclei has been developed using the Detector for Advanced Neutron Capture Experiments (DANCE) at LANSCE [13, 14] and successfully demonstrated on ^{235}U [15]. The method uses a set of three different measurements to extract the final capture cross section. First, a thin sample ($\sim$1 mg) of the fissile nucleus is measured inside a fission tagging Parallel Plate Avalanche Counter (PPAC) to characterize the prompt fission γ-ray spectrum. In the ^{239}Pu case we also extracted a capture cross section from the thin target dataset below 1 keV which was published previously [16]. Second, a measurement on a thick target (50 mg total mass in this case) is conducted to achieve sufficient counting statistics at high energies. Finally, scattered neutron background from the thick target is characterized using a ^{208}Pb sample - in this case Ni foils were added to each side of the ^{208}Pb to represent thick (200 mg/cm^2) cladding which surrounded the ^{239}Pu. Figure 1 (a) and (b) illustrate the ^{239}Pu thick target and background target geometries respectively. The ^{239}Pu isotopic purity was unknown, so incident neutron energies below 100 eV were analyzed to determine the presence of isotopic contaminants by observing characteristic resonances which are sensitive to contamination at the sub-percent level. We did not observe any resonances from other Plutonium isotopes which limited any contamination to a negligible level.

The thick target did not have explicit fission tagging, so the prompt fission γ shape was measured with the thin target and used to define a fission background line shape for the thick target dataset. Using the fission γ-ray profile to characterize the amount of background within our capture gate required a normalization to set its amplitude. Neutron induced fission emits many more γ-rays than capture, so a region of high multiplicity in DANCE has been used to provide a clean fission normalization. The fission spectrum can then be subtracted from the capture region and remove what is the most dominant background in measurements with no explicit fission tag. Observed γ-rays are grouped into physics events by a software-defined coincidence gate, and a clusterization algorithm is used to group γ-rays which scatter between adjacent detectors into one "cluster". In the following, we use crystal multiplicity M_{crys} to define the number of crystals which participate in a given event, and cluster multiplicity M_{cl} to define the number of participating clusters.

The clean fission region is also used to define a fission yield which makes the measurement of capture relative to fission possible. This serves to remove systematic uncertainties associated with target mass and spatial distribution, as well as neutron flux related uncertainties from the measurement. A detailed description of DANCE and the ratio-to-fission technique can be found elsewhere [15–21], and details unique to or important for the current work on ^{239}Pu are reported below.

Certain aspects of the analysis differed from [15] due to the unique challenges facing the ^{239}Pu experiment. One such detail is shown in Figure 2, which shows the time ΔT_{event} between the first and last γ-ray signal in

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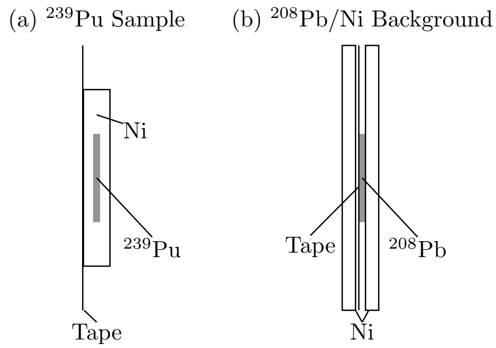

FIG. 1. Schematic of target geometry for (a) the ^{239}Pu sample and (b) the ^{208}Pb/Ni background.

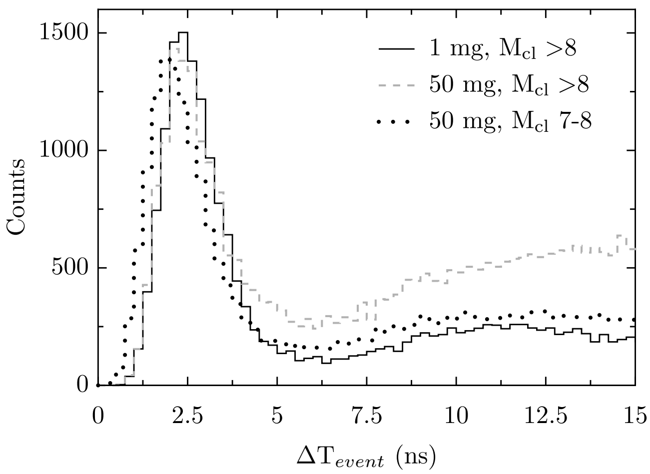

FIG. 2. Time separation between the first and last γ-ray participating in DANCE events.

a DANCE event for neutron kinetic energies ≥ 100 keV. The thin target analysis reported in [16] used a gate of $M_{cl} > 8$ to identify fission events for the purposes of PPAC characterization (solid black line). It is clear that most events lie below an event width of 6 ns, but the dashed gray curve showing the same spectrum for the present work indicates a significantly larger fraction of events which are too wide. This was caused by the high instantaneous event rate for early in the beam pulse. The target mass was chosen to achieve reasonable capture counting rates at the highest energies where the cross section is small, and the much larger fission cross section was able to produce non-negligible pile-up. The fission multiplicity distribution for $M_{cl} > 8$ rapidly decreases, emphasizing the background due to pile-up. The dotted black curve represents the distribution which only uses $M_{cl} = 7, 8$ for a fission tag, significantly reducing the fraction of wide events. Therefore, only $M_{cl} = 7, 8$ were used in the fission tag and the coincidence window was required to be 6 ns.

The fission tagging cut came closer to the (n,γ) part of the energy / multiplicity distribution in DANCE than the work of [16], so we investigated the possibility of cross-

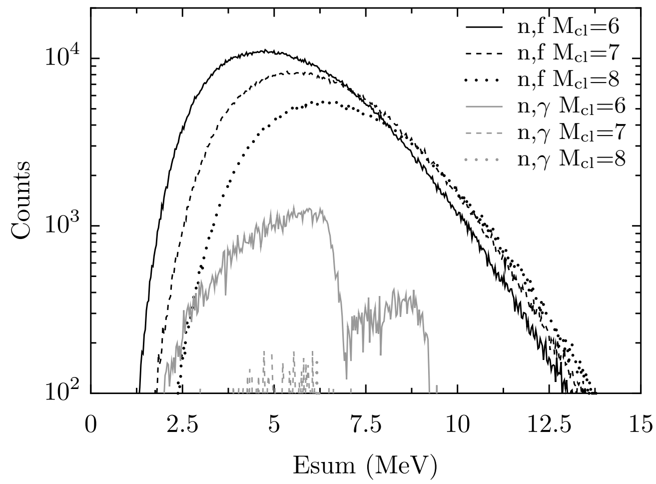

FIG. 3. Relative strength of fission, capture in M_{cl} 6-8 region. Using $M_{cl} = 6$ in addition to M_{cl} 7,8 raises the capture contamination of fission signals from 0.3% to 3.6%.

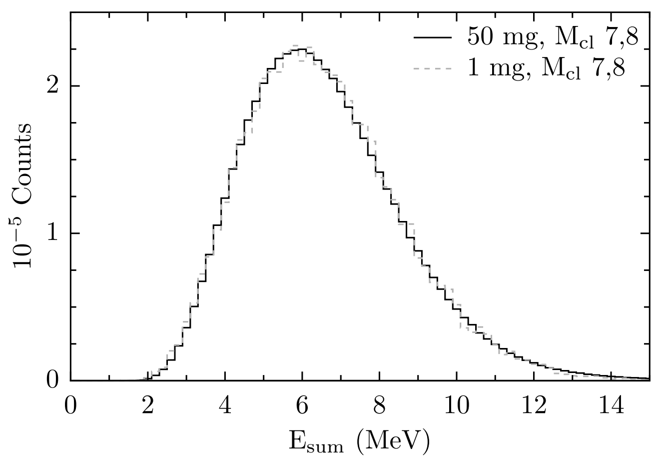

FIG. 4. Consistency of fission γ-ray spectrum between thin and thick target in M_{cl} 7,8 region and neutron energies 10 keV to 1 MeV. Spectra are normalized to equal area for comparison.

talk between capture and fission channels. Figure 3 shows a diagnostic for cross-talk between capture and fission as a function of multiplicity, where solid, dashed, and dotted lines represent $M_{cl} = 6, 7, 8$ respectively. The black curves represent the spectrum for PPAC tagged data, while the gray lines show fission-subtracted spectra. The capture contribution at $M_{cl} = 6$ is noticeable and would result in a contamination of 3.6% to the fission normalization. Using only $M_{cl} = 7, 8$ results in a contamination of ≤0.3% which is much smaller than other sources of uncertainty and therefore negligible.

Performing a ratio measurement with the massive target required a fission tag resulting from γ-rays rather than fission fragments, so the consistency of the fission γ-ray spectrum between the thin and thick targets was an important check on the robustness of the analysis

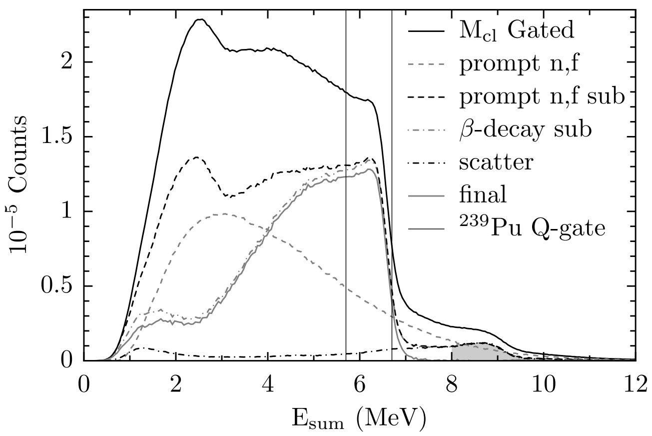

FIG. 5. Total γ-ray spectrum for incident neutrons between 200 eV and 10 keV and $M_{cl} = 4$ illustrating the background subtraction process. See text for details.

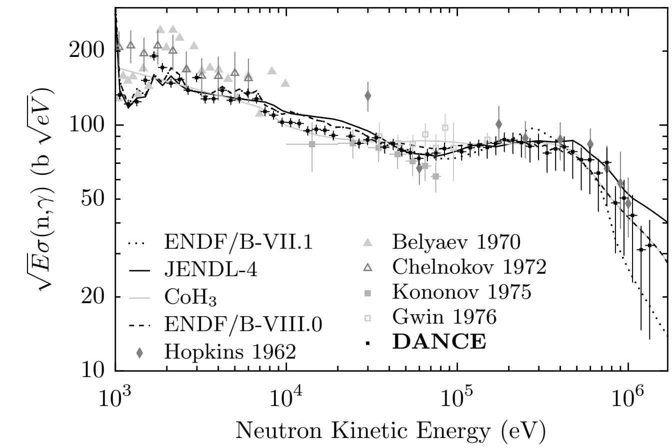

FIG. 6. DANCE ^{239}Pu(n,γ) cross section compared to model predictions, historical data, and evaluation in the 1 keV to 1 MeV region.

method. Figure 4 shows the thick target spectrum for $M_{cl} = 7, 8$ in solid black and the thin target in dashed gray for neutron energies of 10 keV to 1 MeV. The two spectra are consistent with each other which indicates that our fission tagging procedure is robust.

The capture yield was obtained by gating on M_{cl}=4 and total γ-ray energy (E_{sum}) 5.7 - 6.7 MeV and subtracting backgrounds. The background subtraction procedure proceeded in the manner described in Ref. [15, 16], with a brief description provided below. Figure 5 shows the procedure in terms of the E_{sum} spectrum. The solid black curve represents the yield of events which passed a $M_{cl} = 4$ gate and contains capture events mixed with fission and scattered neutron background. The dashed gray line indicates the background prompt fission spectrum, where the spectral shape was defined by the work reported in Ref. [16] and normalized at M_{cl} 7,8 as discussed above. Since there was no explicit fission tag, this background comes in at a level ~3x that of Ref. [16] and represents the most significant background.

Neutron induced fission populates a broad distribution of unstable nuclei which β-decay back toward stability with lifetimes much longer than the DANCE coincidence window. Partial feeding to excited states result in γ-rays which cause an additional fission-related source of background with a decay time of $\sim$ 40 min. The lineshape of this background comes from data taken between beam pulses, and its normalization was calculated from the relative time width of each neutron energy bin relative to the background bin as discussed in [16]. The effect is shown as the move from the dashed black line to the gray dash-dotted line (β-decay sub) in Figure 5 and contributes most heavily below 5 MeV.

Finally, scattered neutron and γ-ray backgrounds were characterized by loading a Ni-Pb-Ni sample into DANCE. This resulted in the dot-dashed black line. The primary background results from neutrons moderating in and capturing on Barium in the DANCE crystals. The shaded region from 8 to 10 MeV is populated cleanly by capture on Barium in DANCE and serves as a reference point to normalize the scattered background from the fission subtracted spectrum. The result is shown as the solid gray line which should be representative of the radiative capture spectrum. The background subtraction procedure appears to work well above 3 MeV, and the ^{239}Pu capture Q-value is 6.5 MeV. Below 3 MeV, the backgrounds appear to be under subtracted. This does not affect the capture cross section measurement in any way.

The Q-value gate used in this analysis was narrower than what was reported in [16] (5.7 to 6.7 MeV vs. 5.0 to 6.7 MeV). Several Q-value gates were evaluated, and the narrower gate achieved the most favorable signal-to-noise levels at high incident neutron energy. The driving consideration was the dominance of the fission background above 100 keV, where the capture cross section rapidly decreases while fission remains comparatively flat. The analysis assumed a constant capture detection efficiency for all incident neutron energies, justified by analyzing the background subtracted capture total γ-ray energy spectrum as a function of neutron energy. The ratio of events within the gate to the total events in the spectrum did not change in a significant way as a function of neutron energy.

We also performed a theoretical model calculation using the statistical Hauser-Feshbach code, CoH_3, where the M1 scissors mode contribution was estimated from our previous study on ^{238}U [22]. The optical potential of Soukhovitskii [23] was employed, and the first 7 levels in the rotational band were coupled. The Engelbrecht-Weidenmüller transformation [24] was applied to correctly take the direct reaction mechanism into account in the Hauser-Feshbach formalism. The fission barrier parameters were adjusted to reproduce the evaluated fission cross sections in the energy range of experiment.

Figure 6 shows the quantity $\sqrt{E} \times \sigma$ vs. neutron energy to most clearly see differences between the DANCE result, our new model predictions, and historical data and evaluation. At all incident neutron energies, systematic uncertainty from the background subtraction is the dominant uncertainty. As described in [15, 16], the capture cross section was measured relative to the ENDF/B-VII.1 fission cross section and a region must be chosen to normalize the capture to fission ratio. In this case, the region was 37 - 100 eV, and the code SAMMY was used to broaden the evaluated cross sections according to the known resolution function [25, 26]. This work is consistent with results already reported below 1 keV in [16]. Since the calculated fission and capture cross sections are very sensitive to the fission barrier parameters, together with the fact that the fission model in the Hauser-Feshbach code is still rather crude, it is imperfect to reproduce the evaluated fission cross section in this energy range, hence the calculated capture cross section is influenced by the fission channel.

From 10 to 30 keV our experimental results are consistently lower than both ENDF/B-VII.1 and JENDL evaluations and are consistent with CoH_3 calculations, while in the region from 40 keV to 200 keV we are in general agreement with the evaluations and lower than predictions. Our measured and predicted cross sections above 400 keV are on the low side of the Hopkins confidence interval, and support a cross section roll-off between the extremes of ENDF/B-VII.1 and JENDL. Indeed, the newly released ENDF/B-VIII.0 uses our predicted cross section above 400 keV to achieve consistency with benchmark calculations [27].

Our measurement considerably improves the constraints on the ^{239}Pu capture cross section in the regions important for nuclear technology applications. Nevertheless, improvements at the highest incident neutron energies (above 700 keV) are still desirable. Future work using DANCE for this purpose would need to consider the three limiting factors for this measurement. First, the fission background yield inside this work's capture gate at 1 MeV incident neutron energy was approximately 4 times larger than the capture yield, so that even with detailed background subtraction the uncertainty associated with the fission channel became large. A new mechanism for tagging fission events in the presence of a massive target would remove fission events from the spectrum and reduce the total fission background uncertainty.

Second, the target mass could be optimized to lower the instantaneous rate on the DANCE crystals. A neutron source redesign effort which is currently underway would significantly alter the neutron beam flux above 1 keV and provide the necessary room to optimize in this area. Finally, the 200 mg/cm^2 Ni plating which served as containment for the ^{239}Pu sample could be made thinner to reduce the scattered neutron background. This would be particularly important if the fission background were reduced through fission tagging, as scattered neutrons would become the dominant background.

We have measured the ^{239}Pu cross section from 1 keV to 1.3 MeV and performed theoretical calculations which unify the entire region with a single measurement with improved uncertainties, confirm prior work in many cases, and extend the measurement to higher energies. The results have already impacted upcoming nuclear data evaluation, will significantly improve future calculations for nuclear applications, and will provide insight for future experiments seeking to address uncertainties above 700 keV.

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[1] P. E. Spivak, B. G. Erozolimskij, G. A. Dorofeev, V. N. Lavrenchik, I. E. Kutikov, and P. Dobrynin, Atomnaya Energiya **1**, 21 (1956).

[2] V. N. Andreev, Atomnaya Energiya **4**, 185 (1958).

[3] J. C. Hopkins and B. C. Diven, Nuclear Science and Engineering **12**, 169 (1962).

[4] F.N.Belyaev, K.G.Ignat'ev, S.I.Sukhoruchkin, S.P.Borovlev, V.V.Pavlov, M.B.Polozov, and A.N.Soldatov, in *Nuclear Data for Reactors Conf., Helsinki* (1970) p. 339.

[5] V.B.Chelnokov, V.A.Tolstikov, Ju.Ja.Stavisskij, A.A.Bergman, and A.E.Samsonov, *Measurements of radiation capture and fission cross-sections for some heavy nuclei by slowing-down- time method in lead*, Tech. Rep. (Yaderno-Fizicheskie Issledovaniya, 1972).

[6] V. N. Kononov, E. D. Poletayev, and B. D. Yurlov, Atomnaya Energiya **38**, 82 (1975).

[7] R.Gwin, E.G.Silver, R.W.Ingle, and H.Weaver, Nuclear Science and Engineering **59**, 79 (1976).

[8] M. Chadwick, M. Herman, P. Obloinsk, M. Dunn, Y. Danon, A. Kahler, D. Smith, B. Pritychenko, G. Arbanas, R. Arcilla, R. Brewer, D. Brown, R. Capote, A. Carlson, Y. Cho, H. Derrien, K. Guber, G. Hale, S. Hoblit, S. Holloway, T. Johnson, T. Kawano, B. Kiedrowski, H. Kim, S. Kunieda, N. Larson, L. Leal, J. Lestone, R. Little, E. McCutchan, R. MacFarlane, M. MacInnes, C. Mattoon, R. McKnight, S. Mughabghab, G. Nobre, G. Palmiotti, A. Palumbo, M. Pigni, V. Pronyaev, R. Sayer, A. Sonzogni, N. Summers, P. Talou, I. Thompson, A. Trkov, R. Vogt, S. van der Marck, A. Wallner, M. White, D. Wiarda, and P. Young, Nuclear Data Sheets **112**, 2887 (2011), ISSN 0090-3752, `http://www.sciencedirect.com/science/article/pii/S009037521100113X`.

[9] K. Shibata, O. Iwamoto, T. Nakagawa, N. Iwamoto, A. Ichihara, S. Kunieda, S. Chiba, K. Furutaka, N. Otuka, T. Ohsawa, T. Murata, H. Matsunobu, A. Zukeran, S. Kamada, and J. . Katakura, Journal of Nuclear

Science and Technology **48**, 1 (2011), cited By (since 1996):167, www.scopus.com.

[10] T. J. team, "Jeff-3.2: Evaluated nuclear data library," online, http://www.oecd-nea.org/dbdata/jeff.

[11] G. Aliberti, G. Palmiotti, M. Salvatores, T. Kim, T. Taiwo, M. Anitescu, I. Kodeli, E. Sartori, J. Bosq, and J. Tommasi, Annals of Nuclear Energy **33**, 700 (2006), ISSN 0306-4549, http://www.sciencedirect.com/science/article/pii/S0306454906000296.

[12] M. Salvatores, G. Albierti, M. Dunn, A. Hogenbirk, A. Ignatyuk, M. Ishikawa, I. Kodeli, A. J. Koning, R. McKnight, R. W. Mills, P. Oblozinsky, G. Palmiotti, A. plompen, G. Rimpault, Y. Rugama, P. Talou, and W. S. Yang, *Uncertainty and Target Accuracy Assessment for Innovative Systems Using Recent Covariance Data Evaluations*, Tech. Rep. (Nuclear Energy Agency Organization for Economic Co-Operation and Development, 2008) nEA/WPEC-26.

[13] P. W. Lisowski, C. D. Bowman, G. J. Russell, and S. A. Wender, Nucl. Sci. Eng. **106**, 208 (1990).

[14] M. Mocko and G. Muhrer, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment **704**, 27 (2013).

[15] M. Jandel, T. A. Bredeweg, E. M. Bond, M. B. Chadwick, A. Couture, J. M. O'Donnell, M. Fowler, R. C. Haight, T. Kawano, R. Reifarth, R. S. Rundberg, J. L. Ullmann, D. J. Vieira, J. M. Wouters, J. B. Wilhelmy, C. Y. Wu, and J. A. Becker, Phys. Rev. Lett. **109**, 202506 (Nov 2012), http://link.aps.org/doi/10.1103/PhysRevLett.109.202506.

[16] S. Mosby, T. A. Bredeweg, A. Chyzh, A. Couture, R. Henderson, M. Jandel, E. Kwan, J. M. O'Donnell, J. Ullmann, and C. Y. Wu, Phys. Rev. C **89**, 034610 (Mar 2014), http://link.aps.org/doi/10.1103/PhysRevC.89.034610.

[17] M. Heil, R. Reifarth, M. Fowler, R. Haight, F. Kaeppeler, R. Rundberg, E. Seabury, J. Ullmann, J. Wilhelmy, and K. Wisshak, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment **459**, 229 (2001), ISSN 0168-9002, http://www.sciencedirect.com/science/article/pii/S0168900200009931.

[18] M. Jandel, T. Bredeweg, A. Couture, M. Fowler, E. Bond, M. Chadwick, R. Clement, E.-I. Esch, J. ODonnell, R. Reifarth, R. Rundberg, J. Ullmann, D. Vieira, J. Wilhelmy, J. Wouters, R. Macri, C. Wu, and J. Becker, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **261**, 1117 (2007), ISSN 0168-583X, http://www.sciencedirect.com/science/article/pii/S0168583X07009160.

[19] C. Wu, A. Chyzh, E. Kwan, R. Henderson, J. Gostic, D. Carter, T. Bredeweg, A. Couture, M. Jandel, and J. Ullmann, Nuc. Instr. Meth. in Phys. A **694**, 78 (2012), ISSN 0168-9002, http://www.sciencedirect.com/science/article/pii/S0168900212008480.

[20] J. Wouters, A. Vicente, T. Bredeweg, E. Esch, R. Haight, R. Hatarik, J. O'Donnell, R. Reifarth, R. Rundberg, J. Schwantes, J. Ullmann, D. Vieira, and J. Wilhelmy, Nuclear Science, IEEE Transactions on **53**, 880 (2006), ISSN 0018-9499.

[21] M. Jandel, T. A. Bredeweg, E. M. Bond, M. B. Chadwick, R. R. Clement, A. Couture, J. M. O'Donnell, R. C. Haight, T. Kawano, R. Reifarth, R. S. Rundberg, J. L. Ullmann, D. J. Vieira, J. B. Wilhelmy, J. M. Wouters, U. Agvaanluvsan, W. E. Parker, C. Y. Wu, and J. A. Becker, Phys. Rev. C **78**, 034609 (Sep 2008), http://link.aps.org/doi/10.1103/PhysRevC.78.034609.

[22] J. Ullmann, T. Kawano, T. A. Bredeweg, A. Couture, R. C. Haight, and M. Jandel, Phys. Rev. C **89**, 034603 (2014).

[23] E. S. Soukhovitskii, R. Capote, J. M. Quesada, and S. Chiba, Phys. Rev. C **72**, 024604 (Aug 2005), https://link.aps.org/doi/10.1103/PhysRevC.72.024604.

[24] T. Kawano, R. Capote, S. Hilaire, and P. Chau Huu-Tai, Phys. Rev. C **94**, 014612 (Jul 2016), https://link.aps.org/doi/10.1103/PhysRevC.94.014612.

[25] N. M. Larson, *Updated Users Guide for SAMMY: Multilevel R-Matrix Fits to Neutron Data Using Bayes Equations*, Tech. Rep. ORNL/TM-9179/R7 (ORNL, 2006) (unpublished).

[26] P. E. Koehler(2007), private communication.

[27] "Endf/b-viii.0," https://ndclx4.bnl.gov/gf/project/endf/.