LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Precision measurement of 238Pu(n,gamma) cross section

A. Chyzh, C. Y. Wu, E. Kwan, R. A. Henderson, J. M. Gostic, T. A. Bredeweg, A. Couture, R. C. Haight, H. Y. Lee, J. M. O'Donnell, J. L. Ullmann

July 17, 2013

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

Precision measurement of ^{238}Pu(n,γ) cross section.

A. Chyzh[1], C. Y. Wu[1], E. Kwan[1,3], R. A. Henderson[1], J. M. Gostic[1], T. A. Bredeweg[2],
A. Couture[2], R. C. Haight[2], H. Y. Lee[2], J. M. O'Donnell[2], and J. L. Ullmann[2]

[1] *Lawrence Livermore National Laboratory, Livermore, California 94550 USA*
[2] *Los Alamos National Laboratory, Los Alamos, New Mexico 87545 USA and*
[3] *National Superconducting Cyclotron Laboratory, East Lansing, Michigan, 48824, USA*

The neutron capture cross section for ^{238}Pu was measured using the Detector for Advanced Neutron Capture Experiments (DANCE) array, which is a highly segmented and highly efficient 4π γ-ray calorimeter. The neutron-capture events were recognized by the total γ-ray energy deposited in DANCE, which equals to the reaction Q-value plus the incident neutron energy. The absolute neutron capture cross section was derived as a function of incident neutron energy from thermal to about 30 keV. The measured cross section for incident neutron energy below 18 eV was made for the first time using the direct method and does not support the most recently adopted changes in ENDF/B-VII.1, where the neutron capture cross section was lowered by as much as a factor of ~ 3 in the neighborhood of 0.3 eV from those evaluated in ENDF/B-VII.0.

PACS numbers: 25.40.Lw,27.90.+b,29.30.Kv

I. INTRODUCTION

The energy released by the α decay of ^{238}Pu is an important power source for many applications. To optimize strategies for production of this isotope, accurate data for neutron capture cross section for ^{238}Pu is required. In the most recent evaluation made in ENDF/B-VII.1 [1], the capture cross section for ^{238}Pu has been changed significantly for incident neutron energy in thermal to epithermal region from those evaluated in ENDF/B-VII.0 [2]. For instance, the cross section is a factor of ~ 3 lower for incident neutron energy in the neighborhood of 0.3 eV. The validation of this drastic change requires new measurements of the capture cross section with incident neutron energy below 18 eV, where no high fidelity experimental data is available except for two measurements at the thermal energy using the integral method with reactor neutrons [3, 4].

Measurement of the neutron-induced reactions for ^{238}Pu is difficult because its relatively short lifetime leads to an extreme α activity even with a small quantity of this isotope. To the best of our knowledge, only one measurement using the direct method was fielded for the neutron capture cross section above thermal energies and this was accomplished by using the intense neutron flux from an underground nuclear detonation [5]. The measurement was made for incident neutron energy from 18 eV to 200 keV. No measurement was ever attempted at the laboratory environment. Here, we report the measurement of the capture cross section for incident neutron energy from thermal to ~ 30 keV using the DANCE array [6] located at the Los Alamos Neutron Science Center(LANSCE). This represents the first such measurement for incident neutron energy below 18 eV using the direct method.

II. EXPERIMENTAL SETUP AND MEASUREMENTS

The neutron capture cross section for ^{238}Pu was measured in two experiments using the DANCE array. The first experiment was fielded over a period of 11 days of beam-on-target using a target with a total mass of 395 μg. The second one was fielded over a period of 21 days of beam-on-target using a target with a total mass of 374 μg, inserted into a parallel-plate avalanche counter (PPAC) [7] in an attempt to measure the neutron-induced fission of ^{238}Pu.

The target with highly enriched ^{238}Pu was fabricated at Lawrence Livermore National Laboratory (LLNL) using the electroplating cell described in Ref. [8]. It also contains 0.30% of ^{239}Pu and 0.34% of ^{240}Pu relative to ^{238}Pu, provided by the vendor, Oak Ridge National Laboratory. In addition, 12.45(11)% of ^{234}U, the daughter nucleus of ^{238}Pu, relative to ^{238}Pu as of March, 2011 was determined using the mass spectrometer at LLNL. The target material was deposited over an area of ~ 7 mm in diameter on both sides of the titanium foil with a thickness of 3 μm. The usefulness of the PPAC employed in the second experiment for the fission measurement by detecting fission fragments is limited due to the target radioactivity of $\sim 2.4 \times 10^8$ α/s, however, the capture events were still collected. As a result, the neutron capture cross section for ^{238}Pu is determined from the data collected over a period of 32 days of beam-on-target in two separated measurements.

Incident neutrons with energies from thermal up to several hundreds keV were produced by bombarding 800 MeV protons, at a repetition rate of 20 Hz, on a tungsten target to yield a spectrum of spallation neutrons that then slowed in a water moderator. This target assembly was installed in 2010 and described in detail in Ref. [9]. DANCE is located in the Lujan Center of LANSCE at a station with a 20.23-m flight path. The incident neutron energy is determined by the time-of-flight tech-

nique, measured by the time difference between the beam pulse and the event detected by either DANCE or PPAC, which has similar intrinsic time resolution of ~ 1.2 ns [7]. This detector system has been used for the neutron capture cross section measurement for many actinide nuclei since its inception, which includes ^{237}Np [10], ^{241}Am [11], and ^{235}U [12] as well as the prompt γ-ray emission for the spontaneous fission of ^{252}Cf [13] and the neutron-induced fission of ^{235}U and ^{239,241}Pu [14, 15]. Detailed descriptions of the experimental setup, the data acquisition system, and the beam monitoring system were given in those publications.

The (n,γ) events are recognized by the total γ-ray energy deposited in a given calorimeter, which equals to the reaction Q-value plus the incident neutron energy. This is accomplished by summing the energies of all γ rays detected by DANCE over a coincident time window of 100 ns in the preliminary analysis. This time window is narrowed to 40 ns in the final analysis after the time alignment was applied for all elements of DANCE, which consists of 160 equal-volume BaF$_2$ scintillation crystals. In addition to the summed γ-ray energy (E_{sum}), the γ-ray multiplicity (M_γ) also was derived. To minimize the overcounting of the multiplicity due to the Compton scattering, any γ ray detected by DANCE with adjacent crystals triggered in a given time window is counted as one. This event-by-event built spectrum of summed γ-ray energy vs. multiplicity can be gated to optimize the true-to-background ratio for events selected in the determination of the neutron-capture cross section as a function of incident neutron energy.

To display the quality of data, the gated summed γ-ray energy spectra with incident neutron energy (E_n) at 18.6 eV (resonance) with a range from 17.9 to 18.9 eV, 1.0 keV with a range from 0.8 to 1.5 keV, and 10.0 keV with a range from 8 to 10 keV, are shown in Fig. 1 (a), (b), and (c), respectively. All have the same M_γ gate near the peak of the distribution, 3 and 4. Due to the sharp drop of the neutron beam intensity as a function of $1/E_n$, the true-to-background ratio deteriorates quickly for the incident neutron energy above 1 keV despite the optimum gate applied. Therefore, the background subtraction is essential for an accurate determination of the cross section. Background spectra were measured after each experiment using a duplicated container without the target material. Obviously, the cross section can be determined with high accuracy for incident neutron energy at resonances, shown in Fig. 1(a), and reasonably well for the neutron energy up to 1 keV, shown in Fig. 1(b). It may reach the detection limit with the current setup for the incident neutron energy in tens of keV.

In the determination of the neutron capture cross section as a function of incident neutron energy, one needs to determine the relative scale first. This is accomplished by placing a gate with the summed energy between 5.55 and 6.55 MeV, near the reaction Q-value of 5.65 MeV, on the background subtracted γ-ray spectrum which is obtained by subtracting the background spectrum from
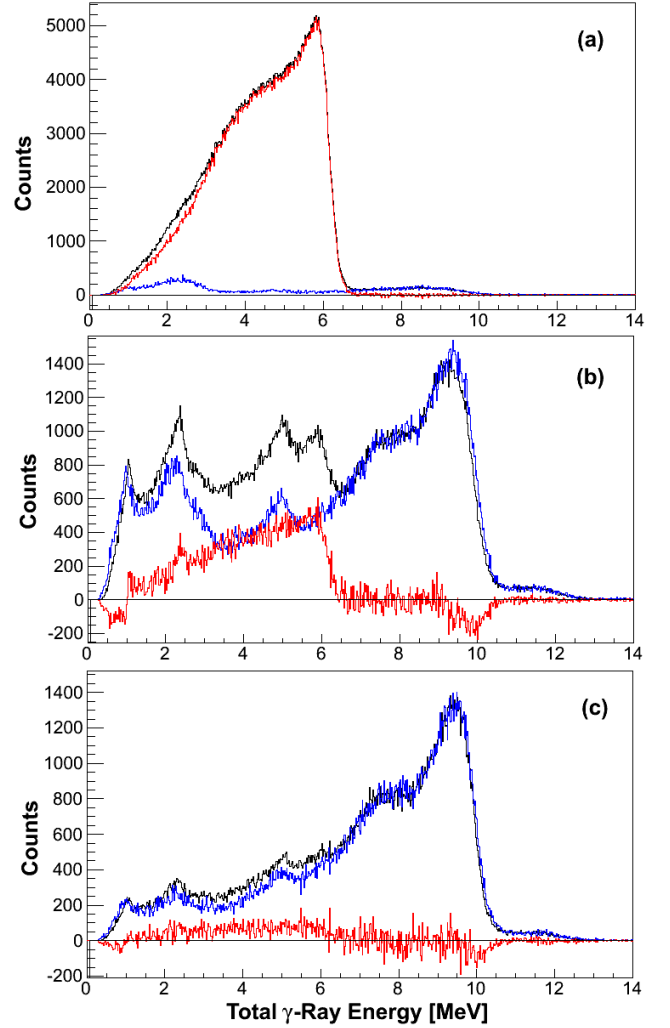

FIG. 1: The summed γ-ray energy spectra with a gate on the $M_\gamma = 3$ and 4 as well as the incident neutron energy at (a) 18.6 eV, (b) 1.0 keV, and (c) 10.0 keV. The spectra of the inclusive and background measurements are shown in black and blue, respectively. The background subtracted spectrum is shown in red.

the inclusive one, as shown in Fig. 1, with the normalization window for the E_{sum} between 7 and 9 MeV. An additional correction was applied for the neutron flux as a function of incident neutron energy, which was measured by monitoring the ^{6}Li(n, α) reaction rate.

The absolute scale of the cross section is set according to the measured ^{234}U resonance at 5.16 eV, which has an integrated cross section of 2640 b·eV in the energy range of 4.91 to 5.38 eV from ENDF/B-VII.1 [1]. As mentioned earlier, the amount of ^{234}U relative to that of ^{238}Pu was 12.45(11)% as of March, 2011. This daughter nucleus was not fully removed chemically before the target was fabricated. The relative efficiency for gates applied to the spectrum of E_{sum} vs M_γ was derived from the ratio between the efficiency ϵ_{Pu}=21.22(5)% for the ^{238}Pu resonance at 18.6 eV and the efficiency ϵ_U=10.24(16)%

for the ^{234}U resonance at 5.16 eV, and assumed to be the same for all incident neutron energy. The integrated cross section for this resonance was determined to be 645(19) b·eV with the energy range of 17.5 to 19.0 eV according to Eq. 1, which is compared to 783 b·eV from ENDF/B-VII.1 [1], 667 b·eV from ENDF/B-VII.0 [2], and 804(166) b·eV from Ref. [5]. The absolute scale of the ^{238}Pu(n,γ) cross section was determined using the following formula:

$$\sigma_{Pu} = \sigma_U \frac{\epsilon_U}{\epsilon_{Pu}} \frac{N_{Pu}}{N_U} R_{U/Pu} \qquad (1)$$

where σ_{Pu} is the absolute ^{238}Pu(n,γ) cross section, σ_U is the absolute ^{234}U(n,γ) cross section, $R_{U/Pu}$ is the ratio of ^{234}U to ^{238}Pu, N_{Pu} and N_U are the counts of the ^{238}Pu(n,γ) and ^{234}U(n,γ) events correspondingly corrected by the neutron flux.

III. RESULTS AND DISCUSSIONS

The absolute neutron capture cross section as a function of incident neutron energy for ^{238}Pu derived from two current measurements is shown in Fig. 2(a). The contribution from ^{234}U was subtracted according to the data evaluated in ENDF/B-VII.1 [1] without broadening the energy resolution. The residual data points from the (n,γ) resonances in ^{234}U were intentionally omitted in the ^{238}Pu(n,γ) cross section spectrum. The fission contribution is not subtracted and is estimated according to the ratio between our measured (n,γ) cross section and the fission data evaluated in ENDF/B-VII.1 [1]. It is given in 2(a) and generally is negligible until E_n reaches about 10 keV, where the correction is about 6%. The error bar plotted in the figure is the statistical uncertainty only. Good agreement is reached between the two sets of data except for those with the cross section near or below one barn, which we believe is the sensitivity limit for the current setup with DANCE. The systematic uncertainty is estimated to about 5% for E_n below 1 eV attributed to the uncertainty of the neutron flux measurement [11] and about 30% for E_n above 10 keV attributed to the uncertainty of the background subtraction. It is better than 3% for E_n between 1 eV and 1 keV, and better than 10% for E_n between 1 keV and 10 keV.

Shown in Fig. 2(b) is the (n,γ) cross section with the weighted average of two current measurements together with those evaluated in ENDF/B-VII.0 [2] and ENDF/B-VII.1 [1]. The cross section for incident neutron energy from thermal to epithermal range was lowered significantly for the latter from those evaluated in ENDF/B-VII.0 [2]. Again, only the statistical uncertainty is plotted. The expanded view of this comparison is shown in Fig. 3. The evaluated cross section for incident neutron energy at thermal energy ($E_n = 0.0253$ eV) is 418 b in ENDF/B-VII.1 [1] and 567 b in ENDF/B-VII.0 [2], compared to 403(8) b measured by Butler *et al.* [3] and 476(33) b by Bringer *et al.* [4] using the integral method

with reactor neutrons. Our measured value is 491(25) b (systematic uncertainty included), which is $\sim 19\%$ higher than the evaluated data in ENDF/B-VII.1 [1] and $\sim 15\%$ lower than the evaluated data in ENDF/B-VII.0 [2] but agrees with the latest measurement [4]. With the increasing incident neutron energy, the discrepancy between the evaluated cross section in ENDF/B-VII.1 [1] and our measurement increases to as much as a factor of ~ 3 until the incident neutron energy reaches the first resonance at 2.87 eV, where the agreement is reasonable. Obviously, the data does not support the most recently adopted changes for the neutron capture cross section with incident neutron energy below the first resonance. The agreement is reasonable well for incident neutron energy above the first resonance until $\sim$30 keV, the current measurement limit.

A comprehensive evaluation of the neutron-induced reaction cross sections for ^{238}Pu was performed by Derrien in 1982 [16] and was available in ENDF/B-V format. The evaluated capture cross section remained the same until the version of ENDF/B-VII.1 [1], where a drastic change was made for the non-resonant cross section with incident neutron energy in thermal to epithermal range by as much as a factor of ~ 3. This may be due to the fact that there is no direct measurement for the incident neutron energy below 18 eV except for two measurements at the thermal energy. The current effort provides a crucial measurement to fill this gap and has important impacts on many applications.

For the resolved resonances with the incident neutron energy between 2.87 and 407 eV, we have performed an analysis using the R-matrix code SAMMY [17] assuming all have the spin 1/2 and a broadening function for the energy resolution given by Koehler [20]. The comparison of the determined resonance energies with the ones given in ENDF/B-VII.1 [1] is made in Table I. The resonance energies in the current measurement have an uncertainty varied from 0.3% to 0.7% for the neutron energy ranging from 2.87 to 407 eV. The average deviation in the resonance energies is 0.2% lower than those in ENDF/B-VII.1 [1]. Both gamma and neutron widths of 32 resonances were extracted using SAMMY by fixing the fission widths according to those evaluated in ENDF/B-VII.1 [1]. Results are given in Table I together with those quoted in ENDF/B-VII.1 [1]. The quoted uncertainty is statistical only. The general agreement for the neutron width between the current measurement and those quoted in ENDF/B-VII.1 [1] is reasonable and the average gamma width of all the resonances listed in Table I is 32 meV compared to 34 meV adopted by ENDF/B-VII.1 [1].

IV. SUMMARY

Precision measurement of the neutron capture reaction for ^{238}Pu was carried out at the Lujan Center of LANSCE using the DANCE array, which is a highly segmented, highly efficient γ-ray calorimeter. The absolute neutron

TABLE I: Comparison of the ^{238}Pu(n,γ) resonance energies and both gamma and neutron widths in the range of 2.87 to 407 eV.

Energy [eV]		Γ_γ [meV]		Γ_n [meV]	
Current	ENDF/B-VII.1	Current	Ref. [16]	Current	ENDF/B-VII.1
2.87	2.89	25.9(14)	36.8	0.063(1)	0.0747
9.91	9.98	27.0(10)	30.3	0.190(1)	0.208
18.4	18.6	45.7(5)	35.2	3.09(1)	4.14
59.5	59.8	25.4(27)		1.32(10)	1.31
69.8	70.1	24.1(26)		2.15(10)	2.51
109.7	110.1	38.0(32)		7.08(30)	5.67
112	111.2	38.3(36)		0.43(2)	0.127
118.1	118.6	36.7(31)		31.6(10)	30.8
121.8	122.4	17.8(11)		29.2(30)	30
131.9	132.4	34.4(33)		0.90(10)	0.851
139.3	139.7	37.1(35)		3.47(30)	2.84
150.4	151.1	13.5(7)		20.2(23)	29.4
165.2	165	34.1(34)		0.22(2)	0.218
168	168	34.4(34)		23.1(30)	46.8
182	182.9	35.3(16)		29.4(26)	28.5
203	203	42.8(38)		7.81(60)	4.99
215	216	28.9(29)		14.9(15)	17.6
220	221	24.1(26)		53.9(50)	59
232	232	33.8(34)		0.71(7)	0.716
244	245	35.6(35)		7.12(70)	6.73
251	252	31.2(32)		14.3(15)	15.6
260	261	33.7(34)		0.26(3)	0.258
288	285	33.4(34)		36.2(37)	26
291	289	35.7(35)		27.5(27)	37.6
300	300	32.8(28)		53.6(47)	55.4
303	305	27.1(27)		6.30(60)	8.03
320	320	31.6(7)		184(17)	179
336	337	34.4(34)		15.6(15)	15.4
367	368	32.6(33)		16.6(20)	17.3
384	382	34.0(34)		0.39(3)	0.39
390	391	33.7(34)		13.0(13)	13.1
407	408	30.5(30)		16.6(20)	18.6

capture cross section is derived as a function of incident neutron energy from thermal to $\sim$ 30 keV. The cross section for the neutron energy below 18 eV is determined for the first time using the direct method and does not support the most recent evaluation made in ENDF/B-VII.1 [1], where the non-resonant cross section in thermal and epithermal energy range was lowered by as much as a factor of $\sim$ 3 from those evaluated in ENDF/B-VII.0 [2]. This finding has important impacts on many applications such as the production of ^{238}Pu as an energy source and the Science-Based Stockpile Stewardship Program. A reevaluation of the (n,γ) cross section of ^{238}Pu is necessary. In addition, the report of both gamma and neutron widths for 32 resonances for incident neutron energy from 2.87 to 407 eV would improve the model predictability of the cross section for higher incident neutron energy.

[1] M.B. Chadwick *et al.*, Nucl. Data Sheets 112, 2887 (2011).

[2] M.B. Chadwick *et al.*, Nucl. Data Sheets 107, 2931

(2006).

[3] J.P. Butler, M. Lounsbury, and J.S. Morritt, Can. J. Phys. 35, 146 (1957).

[4] O. Bringer *et al.*, Proceedings of the Int. Conf. On Nuclear Data for Science and Technology, April 22-27, 2007, Nice, France, editors O.Bersillon, F.Gunsing, E.Bauge, R.Jacqmin, and S.Leray, EDP Sciences, 2008, DOI:10.105/ndata:07612.

[5] M.G. Silbert and J.R. Berreth, Nucl. Sci. Eng. 52, 187 (1973).

[6] M. Heil, R. Reifarth, M. M. Fowler, R. C. Haight, F. Kappeler, R. S. Rundberg, E. H. Seabury, J. L. Ullmann, and K. Wisshak, Nucl. Instrum. Methods Phys. Res. A 459, 229 (2001).

[7] C.Y. Wu, A. Chyzh, E. Kwan, R. Henderson, J. Gostic, D. Carter, T. Bredeweg, A. Couture, M. Jandel, J. Ullmann. Nucl. Instrum. Methods Phys. Res. A 694, 78 (2012).

[8] R.A. Henderson, J.M. Gostic, J.T. Burke, S.E. Fisher, and C.Y. Wu, Nucl. Instrum. Methods Phys. Res. A 655, 66 (2011).

[9] M. Mocko and G. Muhrer, Nucl. Instru. Methods Phys. Res. A 704, 27 (2013).

[10] E.-I. Esch *et al.*, Phys. Rev. C 77, 034309 (2008).

[11] M. Jandel *et al.*, Phys. Rev. C 78, 034609 (2008).

[12] M. Jandel *et al.*, Phys. Rev. Lett. 109, 202506 (2012).

[13] A. Chyzh *et al.*, Phys. Rev C 85, 021601(R) (2012).

[14] A. Chyzh *et al.*, Phys. Rev. C 87, 034620 (2013).

[15] J.L. Ullmann *et al.*, Phys. Rev. C 87, 044607 (2013).

[16] H. Derrien, INDC(FR)-57/l (1982).

[17] N.M. Larson, Oak Ridge National Laboratory, ORNL/TM-9179/R8, (2008).

[18] T.E. Young, F.B. Simpson, J.R. Berreth, and M.S. Coops, Nucl. Sci. Eng. 30, 355 (1967).

[19] W.F. Stubbins, C.D. Bowman, G.F. Auchampaugh, and M.S. Coops, Phys. Rev. 154, 1111 (1967).

[20] P.E. Koehler, Phys Rev C 76, 025804 (2007).

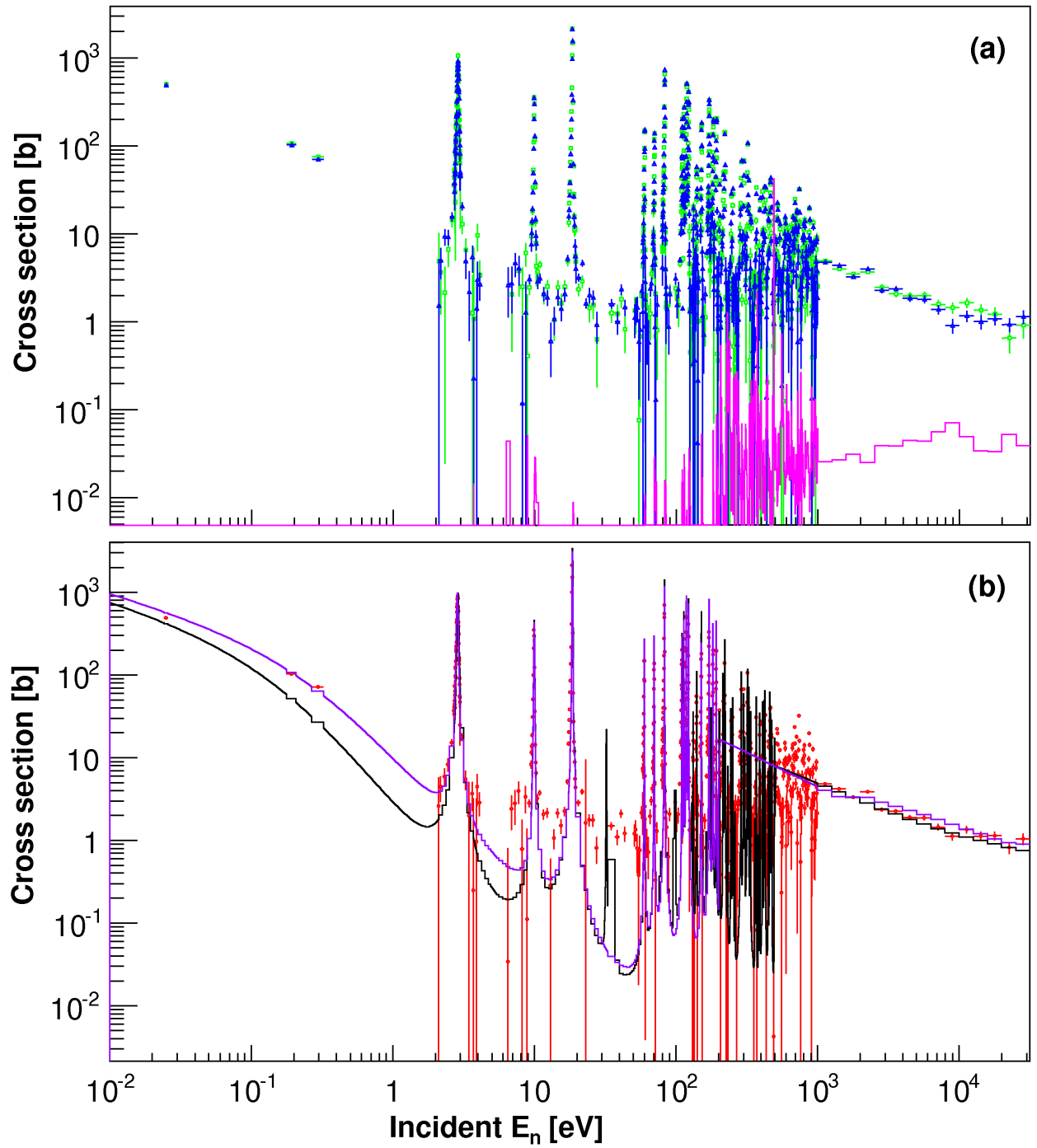**FIG. 2:** Comparison of the derived ^{238}Pu(n,γ) cross sections as a function of incident neutron energy between two current measurements is shown in (a) with the first measurement labeled in green and the second one in blue. The horizontal bar indicates the bin size but not the uncertainty. The contribution from the daughter nucleus, ^{234}U was subtracted. The fission contribution is not subtracted and shown in magenta. The weighted average cross sections together with those evaluated in ENDF/B-VII.0 (violet) ENDF/B-VII.1 (black) are given in (b).
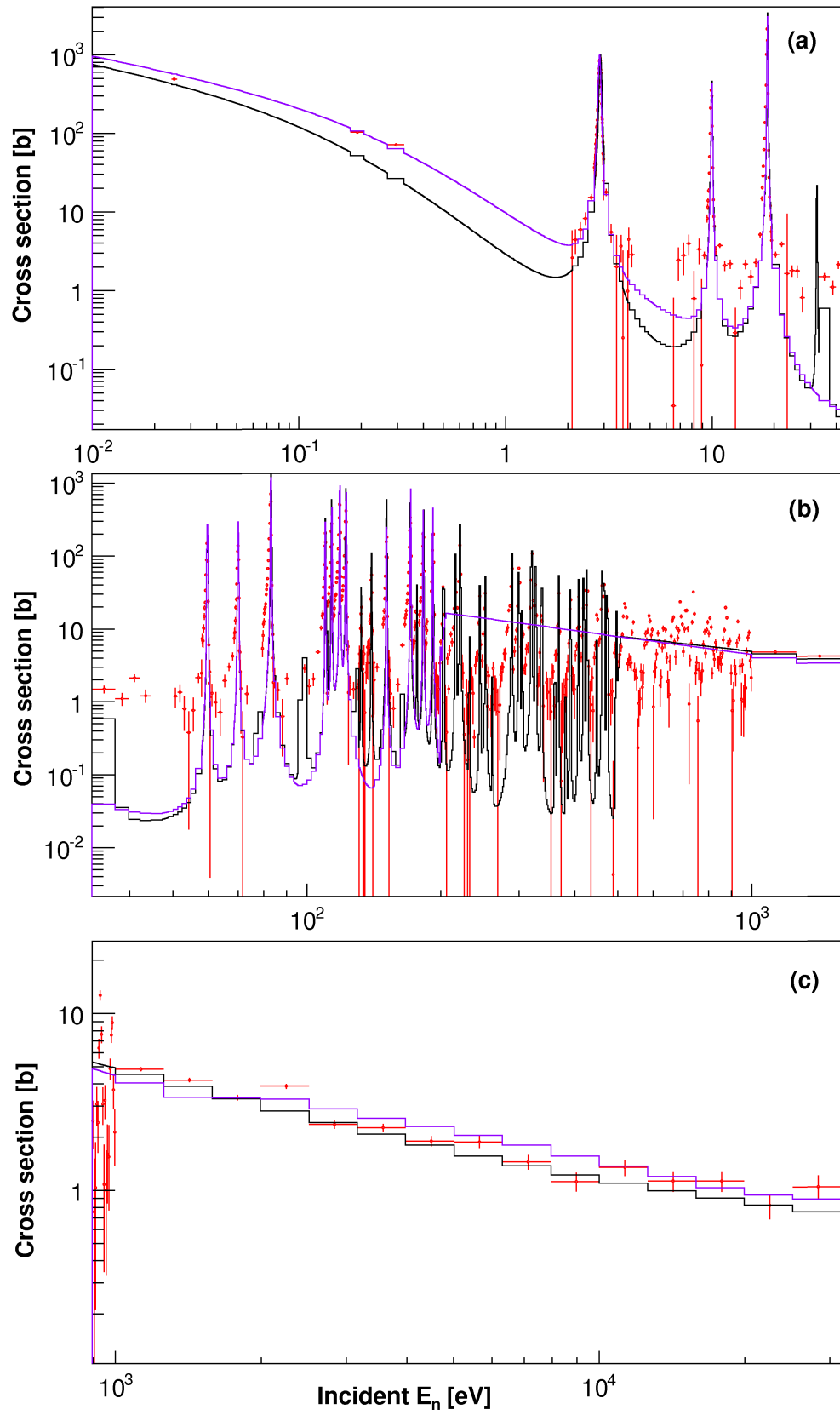
FIG. 3: Comparison of the weighted average ^{238}Pu(n,γ) cross sections (red) to those evaluated in ENDF/B-VII.0 (violet) and ENDF/B-VII.1 (black) is shown as a function of incident neutron energy with the neutron energy from (a) 0.01 to 40 eV, (b) 30 eV to 2 keV, and (c) 1 to 30 keV.