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# Precision measurement of the $^{238}$Pu$(n, \gamma)$ cross section

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The neutron-capture cross section for $^{238}$Pu was measured by using the detector for advanced neutron-capture experiments (DANCE) array, which is a highly segmented and highly efficient $4\pi$ $\gamma$-ray calorimeter. The neutron-capture events were recognized by the total $\gamma$-ray energy deposited in DANCE, which is equal 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 performed for the first time by 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.

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## I. INTRODUCTION

The energy released by the $\alpha$ decay of $^{238}$Pu is an important power source for many applications. To optimize strategies for production of this isotope, accurate data for the neutron-capture cross section for $^{238}$Pu are required. In the most recent evaluation performed in ENDF/B-VII.1 [1], the capture cross section for $^{238}$Pu has been changed significantly for incident neutron energy in the 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 are available except for two measurements at the thermal energy by 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 $\alpha$ activity even with a small quantity of this isotope. To the best of our knowledge, only one measurement that uses 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 performed 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 by using the detector for advanced neutron-capture experiments (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 by using the direct method.

## II. EXPERIMENTAL SETUP AND MEASUREMENTS

The neutron-capture cross section for $^{238}$Pu was measured in two experiments by using the DANCE array. The first experiment was fielded over a period of 11 days of beam on target by using a target with a total mass of 395 $\mu$g. The second one was fielded over a period of 21 days of beam on target by using a target with a total mass of 374 $\mu$g, 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) by 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 by 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 $\mu$m. 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 $\sim 2.4 \times 10^8$ $\alpha$/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 hundred 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 was 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 technique, measured by the time difference between the beam pulse and the event detected by either DANCE or PPAC, which has a similar intrinsic time resolution of ~1.2 ns [7]. This detector system has been used for the neutron-capture cross-sectional measurement for many actinide nuclei since its inception, which includes $^{237}$Np [10], $^{241}$Am [11], $^{235}$U [12], as well as the prompt $\gamma$-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 papers.

The $(n,\gamma)$ events are recognized by the total $\gamma$-ray energy deposited in a given calorimeter, which equals the reaction $Q$ value plus the incident neutron energy. This is accomplished by summing the energies of all $\gamma$ 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 $\gamma$-ray energy ($E_{\mathrm{sum}}$), the $\gamma$-ray multiplicity ($M_\gamma$) was derived. To minimize the overcounting of the multiplicity due to the Compton scattering, any $\gamma$ 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 $\gamma$-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 $\gamma$-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 Figs. 1(a)–1(c), respectively. All have the same $M_\gamma$ gate near the peak of the distribution, 3 and 4. Due to the sharp drop in the neutron beam intensity as $1/E_n$, 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 by 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 $\gamma$-ray spectrum, which is obtained by subtracting the background spectrum from the inclusive one as shown in Fig. 1 with the normalization window for the $E_{\mathrm{sum}}$ 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$(n,\alpha)$ reaction rate evaluated in ENDF/B-VII.1.

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 4.91–5.38 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 $E_{\mathrm{sum}}$ vs $M_\gamma$ was derived from the ratio between the efficiency $\epsilon_{\mathrm{Pu}} = 21.22(5)\%$ for the $^{238}$Pu

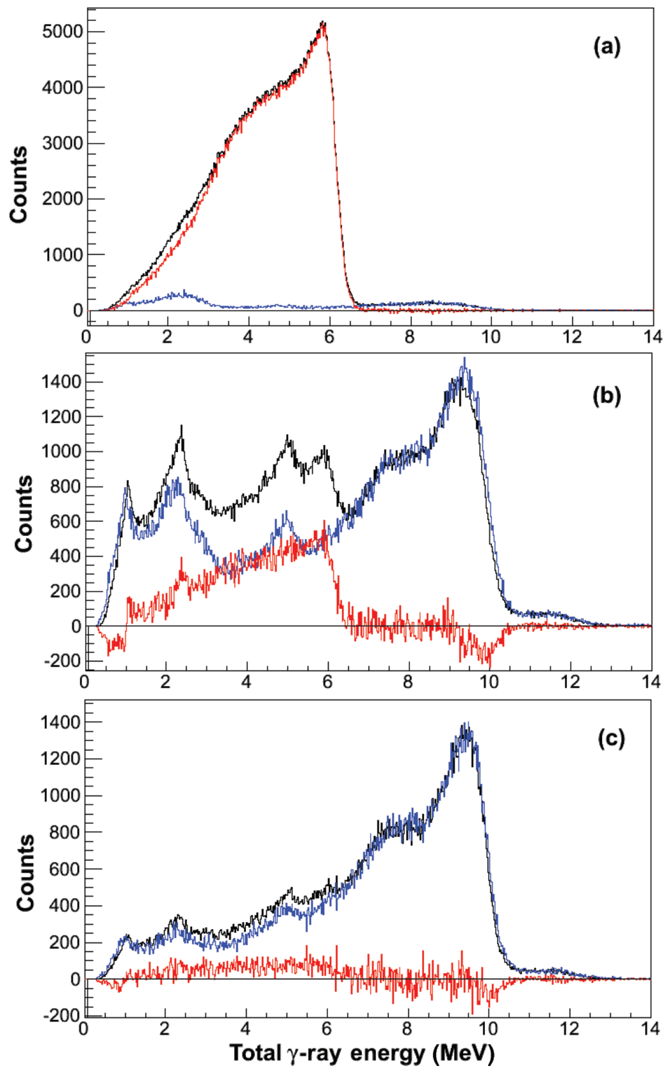


FIG. 1. (Color) The summed $\gamma$-ray energy spectra with a gate on the $M_\gamma = 3$ and 4 as well as the incident neutron energy at (a) 18.6 eV, (b) 1.0 keV, and (c) 10.0 keV. The spectra of the inclusive and background measurements are shown in black and blue, respectively. The background subtracted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

resonance at 18.6 eV and the efficiency $\epsilon_{\mathrm{U}} = 10.24(16)\%$ for the $^{234}$U resonance at 5.16 eV and was 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 17.5–19.0 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$(n,\gamma)$ cross section was determined by using the following formula:

$$\sigma_{\mathrm{Pu}} = \sigma_{\mathrm{U}} \frac{\epsilon_{\mathrm{U}}}{\epsilon_{\mathrm{Pu}}} \frac{N_{\mathrm{Pu}}}{N_{\mathrm{U}}} R_{\mathrm{U/Pu}}, \quad (1)$$

where $\sigma_{\mathrm{Pu}}$ is the absolute $^{238}$Pu$(n,\gamma)$ cross section, $\sigma_{\mathrm{U}}$ is the absolute $^{234}$U$(n,\gamma)$ cross section, $R_{\mathrm{U/Pu}}$ is the ratio of $^{234}$U to $^{238}$Pu, $N_{\mathrm{Pu}}$ and $N_{\mathrm{U}}$ are the counts of the $^{238}$Pu$(n,\gamma)$ and $^{234}$U$(n,\gamma)$ 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 $(n, \gamma)$ resonances in $^{234}$U were intentionally omitted in the $^{238}$Pu$(n, \gamma)$ cross-sectional spectrum. The fission contribution is not subtracted and is estimated according to the ratio between our measured $(n, \gamma)$ cross section and the fission data evaluated in ENDF/B-VII.1 [1]. It is given in Fig. 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 1 b, 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 and 10 keV.

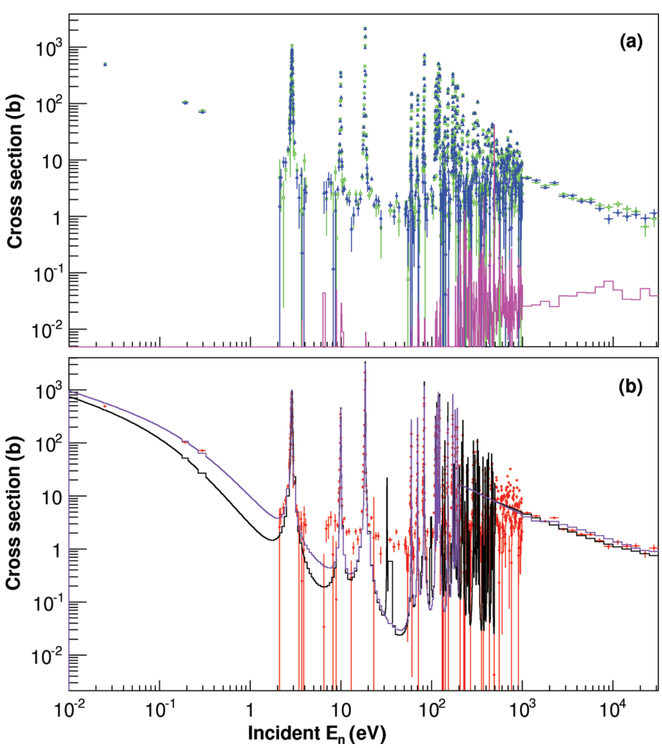


FIG. 2. (Color) Comparison of the derived $^{238}$Pu$(n, \gamma)$ cross sections as a function of incident neutron energy between two current measurements is shown in (a) with the first measurement labeled in green and the second one labeled in blue. The horizontal bar indicates the bin size but not the uncertainty. The contribution from the daughter nucleus $^{234}$U was subtracted. The fission contribution is not subtracted and is 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).

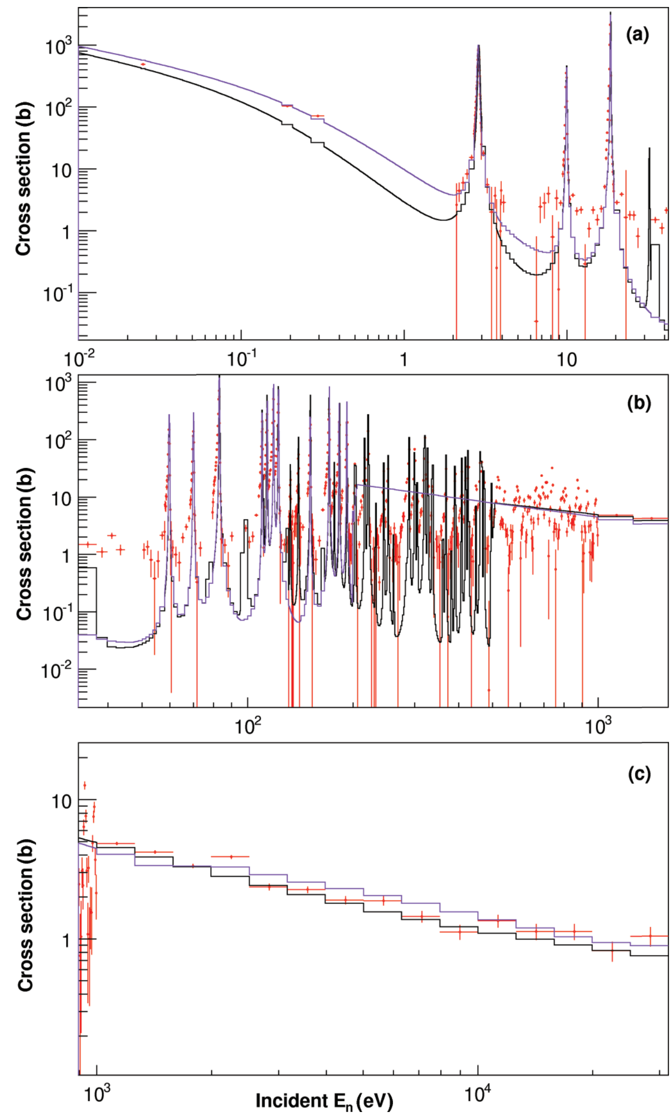


FIG. 3. (Color) Comparison of the weighted average $^{238}$Pu$(n, \gamma)$ cross sections (red) to those evaluated in ENDF/B-VII.0 (violet) and ENDF/B-VII.1 (black) is shown as a function of incident neutron energy with the neutron energy from (a) 0.01 to 40 eV, (b) 30 eV to 2 keV, and (c) 1 to 30 keV.

Shown in Fig. 2(b) is the $(n, \gamma)$ 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 the thermal to the 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] by using the integral method with reactor neutrons. Our measured value is 491(25) b (systematic uncertainty included), which is ∼19% higher than the evaluated data in ENDF/B-VII.1 [1] and ∼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 do not support the most recently adopted changes for the neutron-capture cross section with incident neutron energy below the first resonance. The agreement is reasonably good for incident neutron energy above the first resonance until ∼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 nonresonant cross section with incident neutron energy in the 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 by using the $R$-matrix code SAMMY [17] (see Refs. [18,19]) that assumes all have the spin 1/2 and a broadening function for the energy resolution given by Koehler *et al.* [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 that ranges 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 by 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].

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

| Energy (eV) | | $\Gamma_\gamma$ (meV) | | $\Gamma_n$ (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 |

## IV. SUMMARY

Precision measurement of the neutron-capture reaction for $^{238}$Pu was carried out at the Lujan Center of LANSCE by using the DANCE array, which is a highly segmented highly efficient $\gamma$-ray calorimeter. The absolute neutron-capture cross section is derived as a function of incident neutron energy from thermal to ∼30 keV. The cross section for the neutron energy below 18 eV is determined for the first time by using the direct method and does not support the most recent evaluation performed in ENDF/B-VII.1 [1] where the nonresonant cross section in thermal and epithermal energy ranges was lowered by as much as a factor of ∼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 ($n,\gamma$) 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.

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