Measurement of neutron total cross sections and resonance parameters analysis of ^{169}Tm in the region from 1 eV to 100 eV*

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Abstract

The neutron total cross section is one of the most fundamental nuclear data. In the low-energy resonance region, discrepancies among measurements arise from experimental backgrounds, sample self-shielding, and instrumental broadening, complicating the precise determination of cross sections.The neutron total cross section of ^{169}Tm was measured at the Back-n facility of the China Spallation Neutron Source (CSNS). Wing-shaped lithium glass detectors were employed to record the transmitted neutron signals from samples with thicknesses of 0.5 mm and 4.5 mm. The raw data were processed by applying time-of-flight corrections and normalizing the spectra to the number of incident protons. The in-beam γ-rays were quantified using the saturated resonance absorption technique with glass scintillators. After background subtraction, transmission and total cross-section were obtained in the region from 1 eV to 100 eV. The measured spectra were analyzed using the SAMMY code within the framework of the ***R***-matrix. Neutron capture yield data are

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incorporated to enhance the reliability of the extracted resonance parameters. For the resonance near 8 eV, the radiative width Γ_γ was was fixed at 86 meV. The resonance energy was determined to be 8.037 eV, consistent with the recently reported resonance by I. Knapová et al. Based on the extracted parameters, the neutron total cross section of ^{169}Tm was reconstructed using the Reich-Moore approximation. The reconstructed cross section shows good agreement with those recommended by the ENDF/B-VIII.1 library, confirming the reliability of the resonance parameters extracted from the capture-transmission measurements.

The datasets presented in this paper, including the neutron transmission, neutron total cross section, and resonance parameters, are openly available at
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1 Introduction

Nuclear data serve as a critical foundation for fundamental nuclear physics research and applications in nuclear engineering and technology. Neutron total cross sections and neutron resonance parameters play pivotal roles in both nuclear physics and nuclear engineering. They are applied not only to validate nuclear level structures and theoretical models of nuclear reactions but also as input parameters for nuclear energy system design and nuclear medicine applications[1-3]. Thulium (Tm) is an important rare-earth element used as a burnable poison in nuclear reactors. Its only naturally occurring isotope is ^{169}Tm. Accurate neutron total cross section data for ^{169}Tm were applied to extract neutron resonance parameters such as resonance energy E_n, neutron width Γ_n, and raditive width Γ_γ. Furthermore, such data help resolve discrepancies in the Experimental Nuclear Reaction Data (EXFOR) database, thereby strengthening the foundation for reactor design and nuclear technology applications.

According to EXFOR records, the earliest measurement of the ^{169}Tm neutron total cross section was reported by Foote et al.[4] in 1953 at Brookhaven National Laboratory (BNL). Using a Be and NaCl crystal spectrometer, they measured cross sections in the 0.1-30 eV energy range and first observed resonance peaks at 3.92, 14.8, and 17.6 eV, estimating an average level spacing of approximately 10 eV for the compound nucleus. Subsequently, Harvey et al.[5] employed a BF_3 counter and the time-of-flight method in 1955 to obtain 318 data points between 3-200 eV. They analyzed the transmission curve using an iterative approximation method to extract E_n and Γ_n. However, limited by instrumental energy resolution and Doppler broadening effects, the resulting parameters lacked high precision. In the 1970s, Tellier et al.[6] performed measurements from 3.9 eV -1 MeV using an electron linear accelerator. By cooling the sample to 77 K with liquid nitrogen to reduce Doppler broadening, they acquired numerous data points and derived neutron resonance parameters. In 1998, Danon et al.[7] conducted measurements at the Rensselaer Polytechnic Institute (RPI) electron linear accelerator over the 0.001-20 eV range and used the SAMMY code to obtain resonance parameters at 3.92, 14.8, and 17.6 eV, providing crucial input for subsequent nuclear data evaluations. In 2010, Wang Wenming et al.[8] measured the total cross section of ^{169}Tm below 100 eV using the Pohang Neutron Facility (PNF) in Korea. Due to the absence of capture experimental data for constraint, their extracted resonance parameters exhibited significant discrepancies with the recommended values of Mughabghab[9].

In summary, multiple measurements of the ^{169}Tm neutron total cross section using thin samples have been reported. Researchers primarily fitted the Breit-Wigner resonance formula[10] to the neutron transmission to determine resonance parameters such as E_n, Γ_n, and Γ_γ. Notably, Γ_γ was mainly derived indirectly from transmission, despite its higher sensitivity to neutron capture cross sections, which typically requires constraints from neutron radiative capture yield measurements. In 2022, Ren Jie et al.[11] performed radiative capture cross section measurements on ^{169}Tm at the CSNS Back-n facility using C6D6 detectors. They implemented more precise background subtraction and resonance parameter analysis, fitting the neutron radiative capture yield with the SAMMY code to determine Γ_γ in the region from 1 eV to 100 eV. Some of these parameters differ from the recommended values in the Evaluated Nuclear Data File (ENDF). In 2025, Knapová et al.[12] measured the neutron radiative capture cross section of ^{169}Tm at the DANCE facility using two sample thicknesses: 5.08×10^{-3} cm and 2.07×10^{-4} cm. Their study identified several new resonance peaks, including those at 8.037, 22.03, 71.85, and so on; however, further experimental validation is required.

Since the commissioning of the Back-n white neutron beamline at the China Spallation Neutron Source (CSNS Back-n), its broad energy spectrum and high flux have filled a critical gap in China's experimental capabilities for white neutron energy measurements. This facility has become a key platform for nuclear data measurements[17,18], including radiative capture cross sections[13,14] and neutron total cross sections[3,15,16]. A wing-shaped lithium glass detector was employed to measure the neutron transmission and analyzed the cross section and resonance parameters in the region from 1 eV to 100 eV. It contributes new data to the EXFOR database and shows more reliable experimental evidence for evaluating ^{169}Tm resonance parameters. It is benefial for resolving existing discrepancies in resonance parameters, verifying potential new resonances, and improving theoretical models of neutron-induced nuclear reactions.

2 Experimental Measurement

2.1 Measurement Principle

The neutron total cross section was measured using the transmission method combined with the time-of-flight technique. The fundamental principle of the transmission method involves comparing neutron transmission intensities with and without a sample under identical incident neutron conditions to determine the sample's attenuation effect, thereby inferring the total cross section[19]. During sample measurements, particularly in long-duration runs, fluctuations in accelerator beam power can lead to variations in the neutron fluence rate. Additionally, measurement durations for different targets are not strictly equal. To eliminate the influence of beam fluctuations and differing measurement durations on neutron counting statistics, normalization by proton counts is typically applied. Only the normalized time-of-flight spectra can be used to calculate the transmission spectrum. At neutron energy E_i, the experimental transmission (T_{exp}) is defined as the ratio of neutron counts with the sample to those without the sample, after background subtraction, as expressed in Eq. (1):

$$T_{\mathrm{exp}}(E_{\mathrm{i}}) = \frac{C_{\mathrm{in}}(E_{\mathrm{i}})/k_{\mathrm{Cin}} - B_{\mathrm{in}}(E_{\mathrm{i}})/k_{\mathrm{Bin}}}{C_{\mathrm{out}}(E_{\mathrm{i}})/k_{\mathrm{Cout}} - B_{\mathrm{out}}(E_{\mathrm{i}})/k_{\mathrm{Bout}}} \tag{1}$$

where $C_{in}(E_i)$ and $C_{out}(E_i)$ are the neutron counts with and without the sample, respectively; $B_{in}(E_i)$ and $B_{out}(E_i)$ represent the corresponding background counts with and without the sample; and k_{Cin}, k_{Bin}, k_{Cout}, k_{Bout} are the proton normalization parameters. By measuring T_{exp}, the neutron total cross section can be calculated, as shown in Eq. (2):

$$\sigma_{\mathrm{t}}(E_{\mathrm{i}}) = -\frac{1}{n}\ln\, T_{\mathrm{exp}} \tag{2}$$

where n is the areal density of the sample, with units of atoms/b (1 b = 10^{-28} m^2). The time-of-flight method determines neutron energy by measuring the detection time of each neutron. It uses the time-of-flight spectrum to calibrate the neutron's initial emission time and effective flight distance. The non-relativistic formula is given in Eq. (3):

$$E_i = \frac{m_n L^2}{2(t-t_0)^2} \tag{3}$$

where m_n is the neutron mass, L is the effective flight path length, t_0 is the neutron production time, and t represents the time at which neutrons reach the detector.

2.2 Experimental Setup

This experiment was performed to measure the neutron total cross section of ^{169}Tm at the CSNS Back-n facility. Fig. 1 shows the neutron beam path, which sequentially passes from left to right through the beam pipe window, collimator 1, the sample, collimator 2, the detector, and finally into the dump. During the measurement, the neutron source, sample, and detector were carefully aligned along the same central axis. Specifically, the detector surface was positioned approximately 21 m from the sample center, and the sample center was located about 56 m from the spallation target core. The detector was placed directly in the neutron beam path, while two photomultiplier tubes (PMTs) were offset from the beam center and mounted on opposite sides of the detector. This configuration significantly reduces the impact of scattered neutrons and γ-flash on the PMTs. Two sample thicknesses, 0.5 mm and 4.5 mm were utilized in the experiment, with detailed parameters provided in Table 1.

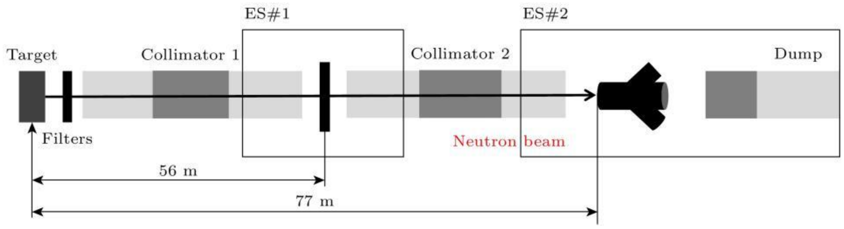

Fig. 1 Schematic diagram of measurement device.

Table 1 Parameters of the ^{169}Tm sample.

Sample	Length/mm	Width/mm	Weight/mg	Areal density/(atom·b^{-1})
1#	60.34 ± 0.05	60.98 ± 0.08	17542.48 ± 0.30	1.694×10^{-3}
2#	60.37 ± 0.11	60.62 ± 0.02	151679.48 ± 0.46	1.472×10^{-2}

3 Data Analysis

Signal waveforms were acquired using a CAEN DT5730B digitizer. The raw data were corrected for dead time and normalized under all conditions. Background subtraction for the sample-in and sample-out spectra was then performed using ^{6}Li and ^{7}Li glass scintillators (^{6}Li-GS and ^{7}Li-GS) with the saturated-resonance absorption technique. Finally, the neutron transmission and total cross section were determined.

3.1 Data Correction

During pulse signal processing by the data acquisition system, the maximum readout rate is constrained by the system's processing capacity. This limitation determines whether the system can promptly respond to the next pulse before completing the processing of the current one. When the pulse processing time exceeds the time interval between successive pulses, the signals from these consecutive pulses cannot be recorded, resulting in data loss. The data acquisition system used in this study recorded nearly all triggered signals. To distinguish pulse pile-up events, the data analysis program implemented a time window τ (τ = 380 ns), within which all detected signals were treated as a single pulse. The dead-time correction factor $f_{\mathrm{dt}}(E_{\mathrm{i}})$ employed in this work is based on the fundamental model described in Ref. [20]:

$$f_{\mathrm{dt}}(E_{\mathrm{i}})=\frac{1}{1-\tau N_{i}(E_{\mathrm{i}})} \tag{4}$$

where the count rate of the experimental spectrum $N_{\mathrm{i}}(E_{i})$ represents the number of neutron counts per proton pulse. In the energy range of 1-100 eV, the dead-time correction was less than 0.2%.

3.2 Experimental Background Analysis

The background in neutron total cross-section measurements originates primarily from two sources. The first is time-independent background, such as environmental activation background and high-energy cosmic rays. This background remains constant over time and constitutes the flat background B_0. The second source is time-dependent background, which can be further divided into sample-related and sample-unrelated components. These backgrounds mainly arise from in-beam γ-rays, with energies spanning from keV to over 10 MeV[21]. During data analysis, the experimental background comprises the time-invariant flat background and the γ-rays, as expressed in Eq. (5):

$$B(E_i)=B_0+B_\gamma(E_i)\times k \tag{5}$$

where $B(E_i)$ is the experimental background under conditions with or without a sample, B_0 represents the flat background under the given experimental conditions, and $B_\gamma(E_i)$ signifies the in-beam γ-ray background. The parameter k is the normalization factor for the in-beam γ-ray background. Since the ^{6}Li-GS and ^{7}Li-GS detectors perform identical responses to γ-rays, the experiment employed the ^{7}Li-GS detector combined with the saturated resonance technique to measure the in-beam γ-ray background and determine k. Fig. 2 shows the composition of the experimental background measured by the ^{6}Li-GS detector for a 0.5 mm sample. Counts below 0.3 eV (corresponding to a time-of-flight exceeding 10^7 ns) in the TOF spectrum consist almost entirely of the flat background. The in-beam γ-ray background is distributed across the entire measured energy range and becomes dominant above the keV energy region. The contribution of the in-beam γ-ray background measured by the ^{7}Li-GS detector was determined using the resonance absorption dip at 3.90 eV in the time-of-flight spectrum, yielding a normalization coefficient k = 0.43.

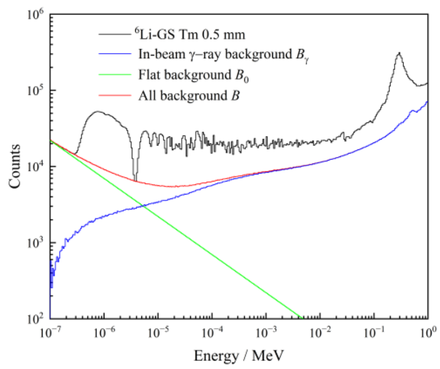

Fig. 2 Background of a 0.5 mm sample.

For background measurements without a sample, filters with strong resonance absorption, such as Co, Ta, and Cd, were used to fully absorb neutrons at specific energies, producing characteristic dips in the time-of-flight spectrum[22,23]. During data processing, the resonance peak of ^{181}Ta (4.28 eV) was selected to calculate the normalization factor for the ^{6}Li-GS detector with filters, yielding a value of 0.42. The energy distribution of in-beam γ-rays remains largely consistent across different energy regions[21]. Using the Geant4 toolkit[24-26], γ-ray spectra were simulated both with and without the filters, resulting in an average attenuation coefficient of 0.91 for in-beam γ-rays due to the filters. The in-beam γ-ray contribution coefficients under sample and no-sample conditions are nearly identical, with deviations about 3%.

3.3 Neutron Total Cross Section Calculation

After background subtraction, neutron transmission can be calculated using Eq. (1), as shown in Fig. 3. Black and red data points correspond to sample thicknesses of 0.5 mm and 4.5 mm, respectively. The transmission curve for the thicker sample shows more pronounced broadening, primarily attributable to self-shielding and multiple scattering effects. To enable finer analysis of resonance peak shapes within the 1-100 eV energy range, this experiment utilized a denser energy grid. However, this approach reduced the counting statistics per energy point, leading to transmission uncertainties exceeding 10% in certain energy regions. The neutron total cross section was derived using Eq. (2). As illustrated in Fig. 4, our experimental results show overall agreement with multiple datasets from the EXFOR database. The broadening of resonance absorption peaks is largely consistent, although some discrepancies in peak heights persist. These differences are primarily due to sample thickness: thinner samples are less prone to saturation effects at strong resonance peaks, and self-absorption as well as multiple scattering effects are comparatively reduced.

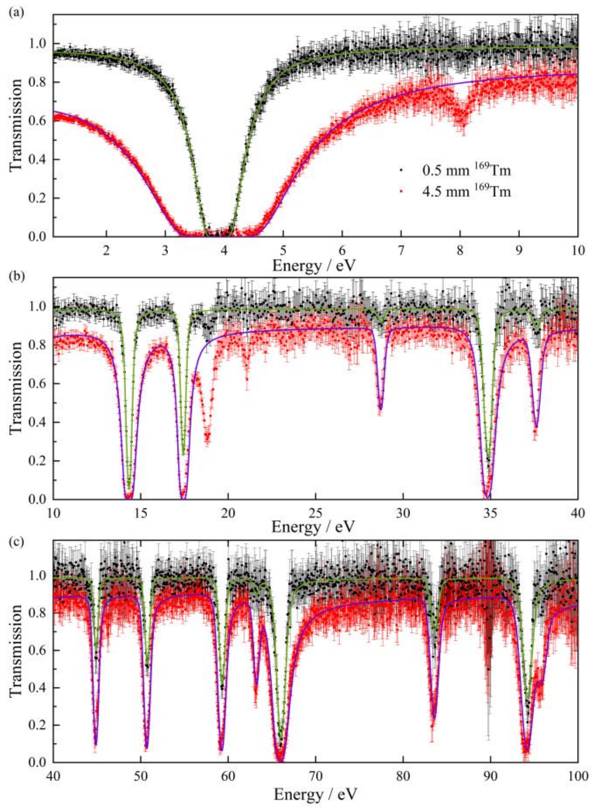

Fig. 3 Neutron transmission and SAMMY fitting results.

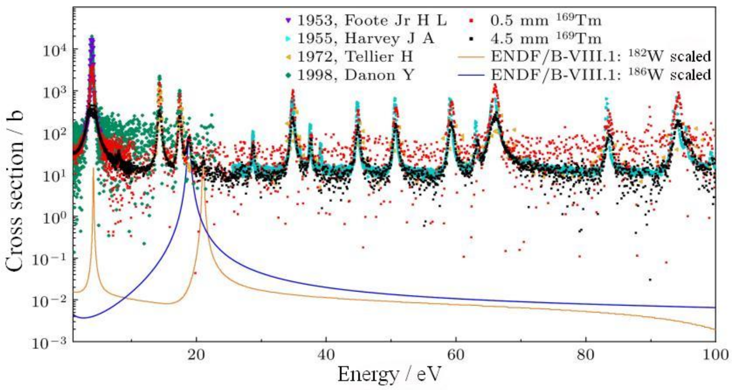

Fig. 4 Comparison of the ^{169}Tm neutron total cross section with EXFOR and ENDF libraries.

Previously, Knapová et al.[12] reported new resonance peaks at 8.037, 22.03, 71.85, and 75.57 eV in capture experiments. In this study, a pronounced resonance was observed near 8 eV. This peak is not included in the ENDF library and may be attributed to the relatively thin samples used in prior transmission experiments. No resonance structure was reproduced in the 38-40 eV energy interval, as reported in reference [11]; this feature is likely caused by impurities. The resonance structures observed near 18.8 eV and 21.1 eV primarily originate from resonant absorption by tungsten (W) in the spallation target material. The main tungsten isotopes are ^{182}W and ^{186}W. After normalizing their evaluated cross sections, the strongest resonance peak of ^{182}W aligns with 21.1 eV (yellow curve in Fig. 4), while that of ^{186}W corresponds to 18.8 eV (blue curve in Fig. 4). The W-related structures are localized and have few influence on the extracted ^{169}Tm resonance parameters.

4 Neutron Resonance Parameter Analysis

Neutron total cross-section data obtained experimentally cannot be directly used as input for nuclear energy system design and analysis. Instead, resonance parameters must be extracted by fitting neutron transmission data. SAMMY is a code package developed by Oak Ridge National Laboratory (ORNL) for neutron resonance parameter analysis. It is based on the multilevel, multichannel ***R***-matrix theory and employs Bayesian methods to fit experimental data. SAMMY accounts for multiple experimental effects, including internal scattering due to sample geometry, sample impurities, experimental background, Doppler broadening from temperature, and experimental resolution[27]. Upon convergence, a complete parameter covariance matrix is generated by SAMMY, from which the parameter uncertainties are derived. Using the extracted resonance parameters, cross sections can be reconstructed via theoretical formulas. In the resolved resonance region (RRR), SAMMY offers several theoretical models, including the Reich-Moore ***R***-matrix approximation, the multilevel Breit-Wigner approximation, and the single-level Breit-Wigner approximation.

The input file incorporated room-temperature Doppler broadening, the energy resolution function, and instrumental broadening parameters[28]. It also integrated results from the ^{169}Tm neutron capture experiment[11], adopting the experimentally determined Γ_γ in the parameter file. As shown in Fig. 3, the green and purple curves represent SAMMY fits to the transmission data for 0.5 mm and 4.5 mm thick samples, respectively. Both fits agree well with the experimental data, confirming the applicability of the ***R***-matrix model. Table 2 shows the resonance parameters and their uncertainties extracted from the transmission data. The results indicate that the standard deviations of the resonance energies obtained from SAMMY fitting are within 0.07%, with deviations from recommended values ranging from 0.05% to 0.60%. The standard deviations of the neutron widths range from 0.35% to 7.53%, with deviations from 0% -20%. The derived parameter k $(k = \frac{g\Gamma_n \Gamma_\gamma}{\Gamma_n + \Gamma_\gamma})$ closely matches the values recommended in evaluated libraries. This work validates the set of Γ_γ values reported in reference [11] and provides more reliable experimental input for nuclear data library evaluations.

Table 2 Comparison of ^{169}Tm neutron reaction resonance parameters with ENDF/B-VIII.1 recommended values.

E_n /eV		Γ_n /meV		Γ_γ /meV		k		J
This work	ENDF/B-VIII.1	This work	ENDF/B-VIII.1	This work	ENDF/B-VIII.1	This work	ENDF/B-VIII.1	
3.908±0.002	3.906	7.38±0.24	7.47	105.70±7.40	102.4	5.17±0.21	5.22	1
14.33±0.01	14.32	9.13±0.16	9.12	98.80±6.92	97.1	2.09±0.14	2.08	0
17.43±0.01	17.42	5.46±0.04	5.74	81.40±5.70	81.4	1.28±0.04	1.34	0
28.73±0.26	28.90	0.21±0.01	0.21	93.70±6.56	95.0	0.16±0.01	0.16	1
34.85±0.01	34.79	6.86±0.06	5.87	86.00±6.02	86.0	4.76±0.06	4.12	1
37.64±0.06	37.51	0.45±0.02	0.46	123.96±8.68	86.0	0.34±0.02	0.34	1
44.88±0.01	44.79	2.91±0.18	3.47	99.00±6.93	93.0	2.12±0.17	2.51	1
50.74±0.07	50.58	5.06±0.37	5.40	89.01±6.23	79.0	3.59±0.33	3.79	1
59.29±0.02	59.07	10.13±0.21	10.0	86.00±6.02	84.0	6.80±0.18	6.70	1
63.17±0.01	62.97	0.96±0.06	1.00	78.02±5.46	78.0	0.71±0.06	0.74	1
65.98±0.01	65.75	139.18±0.49	113	83.00±5.81	83.0	13.00±2.28	11.96	0
83.55±0.13	83.18	7.50±0.09	7.53	85.00±5.95	85.0	5.17±0.09	5.19	1
94.16±0.06	94.10	35.08±1.92	32.0	119.12±8.14	86.0	18.56±1.19	17.49	1
95.95±0.02	95.40	4.25±0.32	3.80	71.00±4.97	71.0	1.00±0.29	0.90	0

As shown in Fig. 5, a resonance structure exists in the energy range of 7.2-8.8 eV. We performed ***R***-matrix fitting on this resonance using SAMMY, setting the raditive width of the resonance peak to the average value from the evaluated library (86 meV). The resulting fitted resonance parameters are listed in Table 3. The fitted resonance energy is 8.037 eV, consistent with the newly reported resonance energy in reference [12]. However, due to the small cross section of this resonance peak and the influence of experimental resolution and statistical fluctuations, the neutron width extracted in this work is larger than that reported in reference [12]. Nevertheless, we can preliminarily assign this resonance peak to ^{169}Tm.

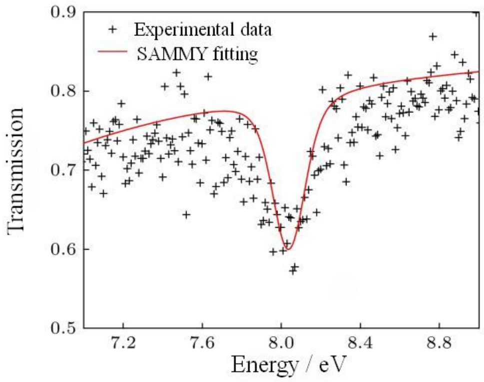

Fig. 5 The results of fitting transmission.

Table 3 Resonance parameters of neutron nuclear reactions for two spin states.

J	E_n /eV	Γ_n /meV	Γ_γ /meV
0	8.037±0.009	0.036±0.002	86
1	8.037±0.009	0.015±0.001	86

After obtaining the resonance parameters, the ***R***-matrix theory can be employed to calculate the neutron total cross section. In this work, the resonance parameters listed in Tables 2 and 3 were input into the SAMMY code. Using the Reich-Moore approximation, we reconstructed the neutron total cross section of ^{169}Tm in the energy range of 1-100 eV and applied Doppler broadening at room temperature (293 K). As shown in Fig. 6, the reconstructed cross section agrees well with that derived from evaluated nuclear data libraries, except at the 8.037 eV resonance peak. The discrepancies in peak height and width between the two datasets correlate with the differences in their respective resonance parameters, thereby validating the reliability of the capture-transmission experiment for extracting resonance parameters.

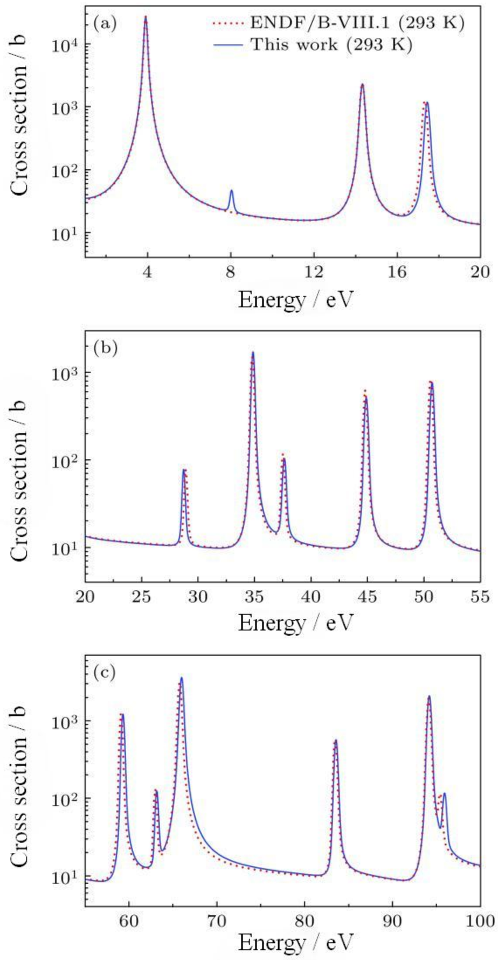

Fig. 6 Comparison of the reconstructed cross section of the resonance parameters in this work with those recommended by the ENDF/B-VIII.1 library.

5 Conclusion

This work employed the transmission and time-of-flight methods to measure the neutron total cross section of ^{169}Tm with a wing-shaped lithium glass detector. The ^{7}Li ground state (^{7}Li-GS) technique combined with saturated resonance absorption effectively subtracted the experimental background. Transmission and cross section data were obtained in the energy range of 1-100 eV, showing good agreement with experimental values in the EXFOR database. Building upon these results and incorporating capture experimental data, we performed ***R***-matrix analysis of the transmission data with the SAMMY code. The fitting results agree well with the experimental measurements, yielding a set of neutron resonance parameters in the 1-100 eV range. This analysis also confirmed the resonance peak at 8.037 eV observed by Knapová et al.[12] in the DANCE experiment. The height and the width of these resonances reconstructed from this work agree well with those derived from the ENDF/B-VIII.1 recommended parameters. More reliable experimental data are provided for nuclear data evaluation, thereby contributing to the further refinement of evaluated nuclear data libraries. Future work should focus on the following: (1) conducting transmission measurements with samples of varying thicknesses to improve the precision and stability of resonance parameter extraction; (2) performing more detailed simulations of the in-beam gamma-ray background to enhance the accuracy of the normalization coefficients; (3) calibrating the detector energy resolution function with greater accuracy; (4) extending resonance parameter analysis to a broader energy range.

Data Availability Statement

The dataset supporting the findings of this study is accessible at the Science Data Bank: https://www.doi.org/10.57760/sciencedb.j00213.00192.

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