

New Measurement of ^{165}Ho Neutron Capture Cross Section Data

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Date: 2025-02-18T00:00:00+00:00

Abstract

The neutron capture cross section data for ^{165}Ho were measured at the Back-streaming White neutron beam line (Back-n) of China Spallation Neutron Source (CSNS) using total energy detection systems, which is comprise a set four C_6D_6 scintillator detectors coupled with pulse height weighting techniques. The resonance parameters were extracted using the multilevel, multichannel R-matrix code SAMMY, fitting the measured capture yields of the $^{165}\text{Ho}(\text{n},\gamma)$ reaction in the neutron energy range below 100 eV. Subsequently, the resonance region capture cross sections were reconstructed based on the obtained parameters. Additionally, the unresolved resonance average cross section of $^{165}\text{Ho}(\text{n},\gamma)$ reaction was determined relative to the standard ^{197}Au sample within the neutron energy range of 2 keV to 1 MeV. The experimental data were compared with the recommended nuclear data from the ENDF/B-VIII.0 library, as well as TALYS-1.9 code calculations. The comparison indicates that the measured $^{165}\text{Ho}(\text{n},\gamma)$ cross sections are in good agreements with these data. The present results are significant for evaluating the ^{165}Ho neutron capture cross section data, enhancing the quality of evaluated nuclear data libraries, and providing valuable guidance for nuclear theoretical models and nuclear astrophysical studies.

Full Text

Preamble

New Measurement of ^{165}Ho Neutron Capture Cross Section Data

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The neutron capture cross section data for ^{165}Ho were measured at the Backstreaming White neutron beam line (Back-n) of the China Spallation Neutron Source (CSNS) using total energy detection systems, which comprise a set of four C6D6 scintillator detectors coupled with pulse height weighting techniques. Resonance parameters were extracted using the multilevel, multichannel R-matrix code SAMMY by fitting the measured capture yields of the $^{165}\text{Ho}(n,\gamma)$ reaction in the neutron energy range below 100 eV. Subsequently, the resonance region capture cross sections were reconstructed based on the obtained parameters. Additionally, the unresolved resonance average cross section of the $^{165}\text{Ho}(n,\gamma)$ reaction was determined relative to the standard ^{197}Au sample within the neutron energy range of 2 keV to 1 MeV. The experimental data were compared with the recommended nuclear data from the ENDF/B-VIII.0 library, as well as TALYS-1.9 code calculations. The comparison indicates that the measured $^{165}\text{Ho}(n,\gamma)$ cross sections are in good agreement with these data. The present results are significant for evaluating the ^{165}Ho neutron capture cross section data, enhancing the quality of evaluated nuclear data libraries, and providing valuable guidance for nuclear theoretical models and nuclear astrophysical studies.

Keywords: Holmium, neutron capture reaction, cross section data, total energy detection principle, C6D6 scintillator detector, China Spallation Neutron Source

Introduction

Nuclear data is used to describe the physical properties of atomic nuclei and their interactions. Neutron nuclear data plays a key role in the research of fundamental nuclear physics and in the development of nuclear energy and nuclear technology [?, ?]. Neutron capture cross section data are extremely important for stellar nucleosynthesis of heavy elements, medical applications, radiation dosimetry, transmutation of nuclear waste, and advanced nuclear energy systems [?].

Many laboratories worldwide, including CERN n_{TOF} [?], Los Alamos National Laboratory DANCE [?], Karlsruhe [?], and GELINA [?], have developed two types of detection systems for the online measurement of neutron capture cross sections. These systems include gamma-ray total energy and total absorption detectors. The total energy detection systems generally use a low-efficiency C6D6 scintillator detector, which is suitable for stable nuclide measurement of

high cross-section and large samples. The total absorption detection systems often use a BaF2 crystal detector array with high energy resolution, good time resolution, low neutron sensitivity, and high efficiency to carry out neutron capture measurement for small size samples, low cross sections, and unstable radionuclides.

The China Spallation Neutron Source (CSNS) [?] provides neutrons via a high-intensity 1.6 GeV proton beam bombarding a tungsten target. The back-streaming neutron beam line (Back-n) facility is positioned in the reverse direction of the proton beam at the CSNS with a flight path length of approximately 80 meters. Figure 1 [Figure 1: see original paper] shows the schematic view of the experimental setup of the CSNS Back-n. At present, five types of spectrometers are constructed for nuclear data measurement at Back-n, such as a set of four C6D6 detectors [?, ?] and a 4π BaF2 detector array (called Gamma Total Absorption Facility II, GTAF-II) [?] for neutron capture measurement, a multilayer fast ionization chamber (FIXM) [?] for fission reaction measurement, neutron total cross section detectors (NTOX) [?] for total cross section measurement, and light-charged particle detectors (LPDA) [?] for light charged particle emission measurement. A detailed description of the spectrometers can be found in Refs [?].

Holmium has been proposed as a standard for neutron capture cross section measurements due to its suitable radioactive half-life and the large capture cross section at the first resonance peak for “saturation resonance” calibration [16–18]. Only a few experimental studies have been performed in the past for the low-energy resonance region, and large discrepancies are observed between the reported data [19–22]. Therefore, many more neutron capture measurements of ^{165}Ho should be carried out for testing the accuracy of evaluated nuclear data and the available experimental data. This work presents a new dataset of the $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ reaction in the neutron energy range from 1 eV to 1 MeV, which was measured using the C6D6 detection system at the CSNS Back-n white neutron source. In the following sections, we outline the methods employed in the experiment and data analysis, discuss the reliability of results, and provide detailed information about the CERN ROOT code [?] relevant to this study.

Methods

A. Measurement

In May 2022, measurement of the $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ reaction was performed at the CSNS Back-n experimental area (#ES2), which is located at a flight path length of about 76 meters. The neutron beam was delivered at ES2 with about $6.92 \times 10^5 \text{ cm}^{-2}\text{s}^{-1}$ neutrons per nominal pulse of 1.6×10^{13} protons in the energy range of 0.3 eV to 200 MeV of beam spots in 30 mm. The neutron energy spectrum was determined using two different detectors, one being a Li-Si detector and the other being a calibrated fission chamber, which were based on

the reactions of ${}^6\text{Li}(\text{n},\text{t})$ and ${}^{235}\text{U}(\text{n},\text{f})$, respectively [?, ?]. The neutron flux in the experiment was monitored by a silicon flux monitor (SiMon), consisting of a thin ${}^6\text{LiF}$ conversion layer and eight silicon detectors, approximately 20 meters upstream from the sample location.

The γ -rays from the ${}^{165}\text{Ho}$ capture reaction were detected by a set of four C6D6 scintillators. The detectors were positioned approximately 17 cm from the target at an angle of 125 degrees relative to the neutron beam, as shown in figure 2 [Figure 2: see original paper]. The characteristics of the samples used in this experiment, provided by the China Institute of Atomic Energy, are detailed in Table 1. A natural metallic holmium sample was employed to determine the neutron capture cross section of ${}^{165}\text{Ho}$. A gold sample of the same dimensions as the holmium sample was utilized for measuring the neutron flux and normalizing the neutron capture data. Background measurements due to scattered neutrons and in-beam γ -rays were conducted using a lead sample. Additionally, an empty sample run was performed to evaluate the sample-independent background. Detector signals were recorded by the CSNS Back-n general-purpose Data Acquisition System [?], which operates at a sampling rate of 1 GS/s with 12-bit full-waveform digitizers. Data acquisition was triggered by the pickup signal from the proton beam. Total beam measurement time was 100 hours, and the CERN ROOT code was utilized for offline analysis.

B. Data Analysis

The natPb sample data are parameterized as eq.(1) to evaluate the in-beam γ -rays and scattered-neutron background contributions for this experiment. The ${}^{165}\text{Ho}$ sample with the ${}^{181}\text{Ta}$ and ${}^{59}\text{Co}$ neutron filters were also performed and used to determine the normalization factors f and f for B and B components by matching the dips of the filtered spectra. The influence of the filters on in-beam γ -rays and neutrons is assessed carefully by analyzing the neutron flux and the energy distribution of the in-beam γ -rays [?]. Energy spectra of neutrons and γ -rays produced at the spallation target are sampled randomly for the incident particle energy spectra of the GEANT4 Monte Carlo code [?], allowing for simulations both with and without filters. The counts of scattered neutrons and γ -rays are recorded at the detector position. The reduced attenuation factors for neutrons and γ -rays are found to be 0.92 and 0.68, respectively, which are applied as corrections to f and f . Much more information on the evaluation method used in this work can be found in Ref. [?].

The experimental neutron capture yield as a function of neutron energy can be calculated as eq.(4):

$$Y_{\text{exp}}(E_n) = S(E_n) - B(E_n) / (c \times \Phi(E_n))$$

where E is the incident neutron energy converted from the neutron time-of-flight (TOF) spectra using the relativistic relation, $S(E_n)$ is the count spectrum of the ${}^{165}\text{Ho}$ sample, $B(E_n)$ is the evaluated background, and $\Phi(E_n)$ is the neutron flux spectrum. The normalization factor f , determined by self-normalizing the

measured capture yield of the 4.9 eV resonance of ^{197}Au , accounts for the absolute incident neutron flux. The ϵ is the detection efficiency of a capture event. The total energy detection principle was used, combining the above-mentioned C6D6 detection system with the pulse height weighting technique (PHWT) [?], to achieve proportionality between ϵ and the total γ -ray energy (E_{γ}) released in the capture event. Hence, $\epsilon = kE_{\gamma} = k(S + E_{\text{cm}})$, where S is the neutron separation energy (i.e., 6.24 MeV) of the compound nucleus, and E_{cm} is the center-of-mass energy of the incident neutron.

In the analysis of the $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ measurement, the thresholds established are $E_{\{\min\}} - E_{\{\text{dep}\}} = 7$ MeV, corresponding to the Compton edges for γ -ray energies of 124 keV and 6.75 MeV, respectively. The weighted function (WF) as a 5th polynomial function is fitted with Geant4 Monte Carlo code, simulating the C6D6 detector response for 27 different monoenergetic γ -rays from 0.1 MeV to 10 MeV. This WF was then applied to all the $S(E_n)$ and $B(E_n)$ spectra for following data analysis.

The resonance shape analysis code SAMMY [?] was used to fit the measured neutron capture yield calculated by eq.(4). In the SAMMY code, the reaction cross section data is described by a multi-level Reich-Moore formalism, which only depends on the properties of the nuclear excitation. This code takes into account all the experimental conditions such as multiple interacting events, sample characteristics, self-shielding, the broadening of resonances due to thermal motion, and experimental resolution of the CSNS Back-n facility [?]. In the fitting process, the initial resonance parameters were taken from the evaluated nuclear data library ENDF/B-VIII.0 [?] and iteratively refined until convergence. The resonance parameters, including resonance energy and capture kernels, were determined using SAMMY fits within the resonance region up to 100 eV, where individual neutron resonances were fitted with high precision. Based on these resonance parameters, the resonance cross sections for the $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ reaction were reconstructed. However, resonance structures could not be adequately resolved beyond approximately 100 eV due to the deterioration of experimental resolution and reduced event statistics as the neutron energy increased. Consequently, an averaged neutron capture cross section was directly derived from the measured neutron capture yield in the unresolved resonance range using formula (5), where N represents the sample's areal density.

In this case, the measured capture yield was corrected for multiple scattering and self-shielding effects through Geant4 code that accounts for the sample composition, geometry, and both neutron scattering and capture cross sections. Subsequently, the ^{165}Ho neutron capture cross section was determined relative to the standard ^{197}Au sample within the neutron energy range of 2 keV to 1 MeV.

The systematic uncertainty for the measured cross sections was determined from various sources. The energy-dependent neutron flux shape contributes an uncertainty of 4.5% below 150 keV and 8.0% above this threshold. Additionally, the uncertainty related to the normalization accounts for approximately 1%,

while the calculation of PHWT introduces a contribution of 3% to the overall uncertainties. Furthermore, the uncertainty from background subtraction using filters is about 8.6%, and an additional uncertainty of 0.01% due to sample impurities was also considered. The sum of these components yields an overall systematic uncertainty of 10.2% (12.2%) for the capture cross section.

Data Records

For each neutron pulse, data from three different types of detectors are simultaneously recorded and stored. One type is a proton beam counter, monitored by the pick-up detector of the Proton Synchrotron accelerator. The second type is a neutron flux counter, composed of eight SiMon detectors. These two data types are utilized for cross-validation and normalization of various measurements. The third type of data includes the event information for radiative neutron capture, recorded by four C6D6 scintillator detectors. All the aforementioned data are acquired using a fully digital data acquisition (DAQ) system of the CSNS Back-n [?], with event-by-event connectivity on the basis of CERN ROOT code. For each neutron capture event, both the deposited energy, represented as a pulse height spectrum in the C6D6 detector, and TOF information of the incident neutrons are recorded. Table 2 lists a comprehensive summary of the event information for all samples utilized in the measurement. The data records are organized in Tree format according to the CERN ROOT version 5.34 and consist of two branch datasets: “NeuDataTree” and “SiDataTree”. “NeuDataTree” recorded data from four C6D6 detectors, including nine leaves: GPSsec (triggering time in seconds), GPSnsec (triggering time in nanoseconds), T0id (trigger T0 identification number), BCid (detector identification number), Energy (neutron energy spectrum), ToF (time-of-flight spectrum), Ph (pulse height spectrum), PeakValue (pulse amplitude), and PeakPoint (pulse timing information). “SiDataTree”, on the other hand, contained data from eight SiMon detectors, organized into six leaves: GPSsec (triggering time in seconds), GPSnsec (triggering time in nanoseconds), T0id (trigger T0 identification number), BCid (detector identification number), SiToF (time-of-flight spectrum), and SiPeakValue (pulse amplitude). This structured organization of data enables efficient storage and facilitates detailed analysis of time, energy, and spectral characteristics captured by the detectors.

Technical Validation

Figure 3 [Figure 3: see original paper] presents the SAMMY fits (red lines) compared to the measured capture yields (black circles) for the reaction $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ across neutron energy ranges below 100 eV. A good agreement is observed between measurement and SAMMY fits, both in terms of resonance energy and spectral shape. The resonance energies E_R , radiative width Γ_γ , neutron width Γ_n , and capture kernels k ($k = g\Gamma_n\Gamma_\gamma/(\Gamma_n + \Gamma_\gamma)$, where g is the statistical factor) obtained in this study are compared with the data from the ENDF/B-VIII.0 library [?], as detailed in Table 3. The

experimental capture resonance parameters exhibit significant agreement with the ENDF/B-VIII.0 evaluations in the energy range below 100 eV. However, disparities are observed in the energy range between 100 eV and 2 keV, which can be ascribed to the degradation of experimental resolution function encountered at the CSNS Back-n facility during this measurement.

Figure 4 [Figure 4: see original paper] and 5 show the measured neutron capture cross sections obtained in this study, along with the calculated results from TALYS code version 1.9 [?] and the evaluated data derived from the ENDF/B-VIII.0 library [?]. The cross sections determined in this work are accurately reproduced by both the TALYS-1.9 calculations and the evaluated data across the full neutron energy range investigated. These results indicate that the experimental apparatus and data analysis methodologies have functioned effectively.

All raw data described in this paper have been uploaded to the Science Data Bank. A direct link to the dataset is available at (<https://doi.org/10.57760/sciencedb.21041>).

Usage Notes

The dataset publication presents newly measured cross sections for the $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ reaction obtained with the CSNS Back-n facility. Our objective is to comprehensively document the data analysis procedures in a dedicated publication, thereby providing access to the neutron capture data for both the nuclear physics community and researchers in related disciplines for future studies. This dataset has numerous applications in nuclear physics, particularly in the following areas:

- (1) The spectroscopic information of heavy nuclei is challenging to obtain experimentally due to the rapid increase in nuclear level density (NLD) with rising excitation energies. To address this, statistical models provide a framework for understanding the internal structure of these nuclei at higher energies, relying on key parameters such as the NLD and the γ -ray strength function (γSF). These parameters are essential for a wide array of calculations in nuclear reactions, particularly in determining neutron capture reaction cross sections. The accuracy of these calculations is vital for evaluating the reliability of nuclear models. In this context, the case of ^{166}Ho , an odd-odd deformed nucleus, plays a significant role. The $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ neutron capture reaction serves as a crucial tool for validating theoretical descriptions of the NLD and γSF . By examining this reaction, researchers can test and refine the predictive power of nuclear models, thereby enhancing our understanding of the underlying nuclear structure and reaction dynamics.
- (2) The nucleosynthesis of elements heavier than iron is considered one of the “11 Biggest Unsolved Mysteries in Physics” that require urgent attention this century. Nuclear astrophysicists generally agree that the slow neutron capture process (s-process) and the rapid neutron capture process (r-process) are the primary mechanisms responsible for the production of

these heavier elements. Holmium, an important rare earth element, is primarily produced through explosive r-process nucleosynthesis, with approximately 9% of its abundance synthesized in the main s-process during the evolution of intermediate-mass stars. The isotope ^{166}Ho is a significant branching nucleus, characterized by a ground state half-life of 26.9 hours and an isomeric state (7^-) half-life of 1200 years, primarily formed by the capture of a neutron by ^{165}Ho . Consequently, the $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ reaction not only depletes the abundance of ^{165}Ho but also influences the abundance of ^{166}Ho and subsequent s-process nucleosynthesis products. Therefore, this reaction cross section is critically important for the study of nucleosynthesis in nuclear astrophysics.

- (3) Holmium (Ho) has extensive applications in nuclear medicine, particularly the β^- and γ -emitting isotope ^{166}Ho [$T_{1/2} = 26.9$ h, $E_{\beta} = 1.77$ MeV (48%) and 1.85 MeV (51%), $E_{\gamma} = 81$ keV (6.7%)], which has been developed for radionuclide therapy and single photon emission computed tomography (SPECT) imaging due to its favorable decay properties. The isotope ^{165}Ho is the only naturally stable isotope of holmium (natHo) and is used to produce ^{166}Ho through the (n,γ) reaction. Accurate data on the neutron capture cross section and resonance integral for the $^{165}\text{Ho}(n,\gamma)^{166}\text{Ho}$ reaction are essential for evaluating neutron irradiation time, activity, and yield of ^{166}Ho produced in nuclear reactors.

Code Availability

The publication of the dataset is accompanied by a software package based on CERN ROOT version 5.34/34 [?], which includes examples for reading the data, generating pulse height spectra of neutrons, performing background subtraction, analyzing neutron resonances, and deriving neutron capture cross sections.

In the dataset, the neutron energy range from 0.2 eV to 2 MeV was logarithmically divided into 3500 bins. The bin intervals and quantities can be reallocated according to the user's range of interest. Two classes, C6D6Data and LiSiData, are defined to read the corresponding data from the ROOT file. The TChain function is used to read all ROOT files under the same experimental conditions. For the C6D6 data, the neutron energy thresholds were set to $E_{\min} = 250$ keV and $E_{\max} = 7$ MeV, with the following detector parameters:

- C6D6 #1: C6D6Data1.BCid = 1, Min bin = 2822, Max bin = 78865
- C6D6 #2: C6D6Data1.BCid = 2, Min bin = 2822, Max bin = 78865
- C6D6 #3: C6D6Data1.BCid = 3, Min bin = 2807, Max bin = 78342
- C6D6 #4: C6D6Data1.BCid = 4, Min bin = 2890, Max bin = 80629

For the LiSi data, the signals from eight LiSi detectors were divided into two paths for storage. During the in-beam experiment, there was an issue with the second signal from the LiSi detector (LiSiData1.BCid = 6), so the first signal (LiSiData1.BCid = 5) was chosen for data processing. The reaction

between neutrons and ${}^6\text{Li}$ primarily generates helium nuclei (α) and tritium nuclei (T), resulting in a bimodal structure in the energy spectrum. Due to the high energy of the T peak (2.73 MeV), saturation may occur during the experiment, leading to poor statistical performance. Therefore, the α peak is selected as the effective neutron counting of the LiSi detector to determine the neutron flux. More detailed information about the data analysis code can be accessed as a notebook on the Science Data Bank, where the complete dataset for this work has been uploaded. A direct link to the dataset is available at: (<https://doi.org/10.57760/sciencedb.21041>).

Acknowledgements

The authors thank the efforts of the CSNS Back-n operators for the skilled operation of the facility. We would like to express thanks to Hong-Wei Wang, Xi-Chao Ruan, and Jing-Yu Tang for their helpful comments on the measurement. We also thank Qi-Wen Fan for preparing the samples.

Author Contributions Statement

Su-Ya-La-Tu Zhang: Conceptualization, Methodology, Writing-review & editing. Yong-Shun Huang: Investigation, Data curation, Writing-original draft. Wei Jiang and Rui-Rui Fan: Measurement, Methodology. De-Xin Wang: Formal analysis. Guo-Li and Dan-Dan Niu: Measurement. Mei-Rong Huang: Writing-review & editing. All authors reviewed the manuscript.

Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Note: Figure translations are in progress. See original paper for figures.

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