New Measurement of 165Ho Neutron Capture Cross Section

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Abstract

The neutron capture cross section for ^{165}Ho was measured at the back streaming white neutron beam line (Back-n) of the China Spallation Neutron Source (CSNS) using total energy detection systems, which are composed of a set of four C_6D_6 scintillator detectors coupled with pulse height weighting techniques. The resonance parameters were extracted using the multi-level multichannel R-matrix code SAMMY, fitting the measured capture yields of the ^{165}Ho(n,γ) reaction in the neutron energy range below 100 eV. Subsequently, the resonance region capture cross sections were reconstructed on the basis of the obtained parameters. Furthermore, the unresolved resonance average cross section of ^{165}Ho(n,γ) reaction was determined relative to the standard sample ^{197}Au 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 the calculations of the TALYS-1.9 code. The comparison indicates that the measured ^{165}Ho(n,γ) cross sections agree well with this data. The present results are significant for the evaluation of the ^{165}Ho neutron capture cross section, enhancing the quality of the evaluated nuclear data libraries, and providing valuable guidance for nuclear theoretical models and nuclear astrophysical studies.

Full Text

New Measurement of 165Ho Neutron Capture Cross Section

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The neutron capture cross section for 165Ho was measured at the back streaming white neutron beam line (Back-n) of the China Spallation Neutron Source (CSNS) using total energy detection systems composed of four C6D6 scintillator detectors coupled with pulse height weighting techniques. Resonance parameters were extracted using the multi-level multichannel R-matrix code SAMMY, fitting the measured capture yields of the 165Ho(n,γ) reaction in the neutron energy range below 100 eV. Subsequently, the resonance region capture cross sections were reconstructed based on the obtained parameters. Furthermore, the unresolved resonance average cross section of the 165Ho(n,γ) reaction was determined relative to the standard sample 197Au within the neutron energy range of 2 keV to 1 MeV. The experimental data were compared with recommended nuclear data from the ENDF/B-VIII.0 library, as well as calculations from the TALYS-1.9 code. The comparison indicates that the measured 165Ho(n,γ) cross sections agree well with these data. The present results are significant for evaluating the 165Ho neutron capture cross section, 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, total energy detection principle, C6D6 scintillator detector, China Spallation Neutron Source

INTRODUCTION

Nuclear data describe the physical properties of atomic nuclei and their interactions. Neutron data play a key role in fundamental nuclear physics research and in the development of nuclear energy and nuclear technology [1, 2]. Neutron capture cross-sectional data are extremely important for stellar nucleosynthesis of heavy elements, medical applications, radiation dosimetry, transmutation of nuclear waste, and advanced nuclear energy systems [3-5].

Many laboratories worldwide, including CERN n_{TOF} [6], Los Alamos National Laboratory DANCE [7], Karlsruhe [8], and GELINA [9], have developed two types of detection systems for online measurement of neutron capture cross sections: gamma-ray total energy detectors and total absorption detectors. Total energy detection systems generally use low-efficiency C6D6 scintillator detectors, which are suitable for measuring stable nuclei with large cross sections and large sample sizes. Total absorption detection systems often use BaF2 crystal detector arrays with high energy resolution, good time resolution, small neutron sensitivity, and high efficiency to carry out neutron capture measurements for small samples, small cross sections, and unstable radionuclides. The China Spallation Neutron Source (CSNS) [10] produces neutrons via a high-intensity proton beam with 1.6 GeV energy bombarding a tungsten target. The back streaming neutron beam line (Back-n) is positioned in the reverse direction of the proton beam at the CSNS with a flight path length of approximately 76 m. [Figure 1: see original paper] shows a schematic view of the experimental setup of the CSNS Back-n.

Currently, five types of spectrometers have been constructed at the Back-n facility for nuclear data measurement. These instruments include a set of four C6D6 detectors [11–13] and a 4π BaF2 detector array, referred to as the Gamma Total Absorption Facility II (GTAF-II) [14], which are utilized for neutron capture measurements. Additionally, a multilayer fast ionization chamber (FIXM) is employed for fission reaction measurements [15], while a neutron total cross section detector (NTOX) is specifically designed for total cross section assessments [16]. Furthermore, light-charged particle detectors (LPDA) are utilized for measuring light charged particle emissions [17]. A detailed description of the spectrometers can be found in Ref. [18].

Holmium (Ho), comprised entirely of the isotope 165Ho with 100% natural abundance, has been proposed as a standard for neutron capture cross section measurements. This is attributed to its favorable radioactive half-life and the substantial capture cross-section observed at the first resonance peak, which is essential for "saturation resonance" calibration [19–21]. Previous studies on 165Ho(n,γ) reveal marked discrepancies: Poenitz et al. [22, 23] reported divergent keV–MeV cross-sections using time-of-flight and statistical models, while Voignier et al. [24] identified data scarcity for holmium. Recent Oslo-method analyses [25] reduced MACS uncertainties but highlighted unresolved impacts of low-energy isomers on s-process abundances. Therefore, many more neutron capture measurements of 165Ho should be carried out to test the accuracy of evaluated nuclear data and available experimental data. This work presents a new dataset of the 165Ho(n,γ)166Ho reaction in the neutron energy range from 1 eV to 1 MeV, 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 [26] relevant to this study.

METHODS

A. Measurement

In May 2022, measurement of the 165Ho(n,γ)166Ho reaction was performed at the CSNS Back-n experimental area (#ES2), located at a flight path length of about 76 m. The neutron beam with beam spots of 30 mm was delivered at ES2 with about 6.92$×10^{5}$ $\text{cm}^{-2}\text{s}^{-1}$ neutrons per nominal pulse of 1.6$×10^{13}$ protons in the energy range of 0.3 eV to 200 MeV. The neutron energy spectrum was determined using two different detectors: a Li-Si detector and a calibrated fission chamber, based on the reactions ^{6}Li(n,t) and ^{235}U(n,f), respectively [27, 28]. The neutron flux in the experiment was monitored by a silicon flux monitor (SiMon), consisting of a thin ^{6}LiF conversion layer and eight silicon detectors, approximately 20 meters upstream from the sample location.

The γ-rays from the 165Ho 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° relative to the neutron beam, as shown in [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 . A natural metallic holmium sample was employed to determine the neutron capture cross section of 165Ho. A gold sample was utilized for measuring the neutron flux and normalizing the neutron capture data. Measurements of background 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 [29], 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. Dead time corrections were disregarded due to the lower event statistics observed in the present experiment. 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 165Ho sample with the ^{181}Ta and ^{59}Co neutron filters was also measured 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 was assessed carefully by analyzing the neutron flux and the energy distribution of the in-beam γ-rays [30]. Energy spectra of neutrons and γ-rays produced at the spallation target were sampled randomly for the incident particle energy spectra of the GEANT4 Monte Carlo code [31], allowing for simulations both with and without filters. The counts of scattered neutrons and γ-rays were recorded at the detector position. The reduced attenuation factors for neutrons and γ-rays were found to be 0.92 and 0.68 [12], respectively, which are applied as corrections to f and f . More information on the evaluation method used in this work can be found in Ref. [12].

The experimental neutron capture yield as a function of neutron energy can be calculated as Eq.(4), where E is the incident neutron energy converted from the neutron time-of-flight (TOF) spectra using the relativistic relation. S(E) is the 165Ho sample counting, B(E) is the evaluated background, and Φ(E) is the neutron flux. 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 _c is the detection efficiency of a capture event. The total energy detection principle was used, combining the abovementioned C6D6 detection system with the pulse height weighting technique (PHWT) [32, 33], to achieve proportionality between _c and the total γ-ray energy (E_c) released in the capture event. Hence, _c = kE_c = k(S_n + E_{cm}), where S_n is the neutron separation energy (i.e., 6.24 MeV) of the compound nucleus, E_{cm} is the center-of-mass energy of the incident neutron, and the proportionality factor k is taken as 1 MeV^{-1}.

In the analysis of the 165Ho(n,γ)166Ho measurement, the thresholds established are E_{min}^{dep} = 250 keV and E_{max}^{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) and B(E) spectra for subsequent data analysis.

The resonance shape analysis code SAMMY [34] was used to fit the measured neutron capture yield calculated by Eq.(4). In the SAMMY code, the reaction cross section 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 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 [35]. In the fitting process, the initial resonance parameters were taken from the evaluated nuclear data library ENDF/B-VIII.0 [36] and iteratively refined until convergence. The resonance parameters, including resonance energy and capture kernels, were determined using SAMMY fitting 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 165Ho(n,γ)166Ho 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 the formula (5), where N_s 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 165Ho neutron capture cross section was determined relative to the standard ^{197}Au sample [37] within the neutron energy range of 2 keV to 1 MeV.

The uncertainties in the present experimental data arise from both statistical and systematic errors. The systematic errors were evaluated based on contributions from various sources including the energy-dependent neutron flux shape (4.5% below 150 keV and 8.0% above this threshold), normalization (1%), background subtraction using filters (about 8.6%), sample impurities (0.01%), and calculation of PHWT (3%). 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 includes 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 [29], with event-by-event connectivity based on 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. 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.

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).

TECHNICAL VALIDATION

[Figure 3: see original paper] presents the SAMMY fitting (red lines) compared to the measured capture yields (black circles) for the reaction 165Ho(n,γ)166Ho across neutron energy ranges below 100 eV. Good agreement is observed between measurement and SAMMY fitting, 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 = gΓ_nΓ_γ/(Γ_n + Γ_γ), where g is the statistical factor) obtained in this study are compared with data from the ENDF/B-VIII.0 library [36], as detailed in . 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 the experimental resolution function encountered at the CSNS Back-n facility during this measurement.

[Figure 4: see original paper] presents the comparison of the 165Ho(n,γ)166Ho cross sections between the TALYS-1.9 [38] calculations and the results reconstructed from SAMMY resonance fitting for neutron energies below 100 eV. The measured neutron capture cross sections in the energy range from 2 keV to 1 MeV are displayed together with calculated results from TALYS-1.9 and evaluated data derived from the ENDF/B-VIII.0 library [36] in [Figure 5: see original paper]. These comparisons demonstrate that 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 performed reliably and effectively.

USAGE NOTES

The dataset publication presents newly measured cross sections for the 165Ho(n,γ)166Ho 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 166Ho, an odd-odd deformed nucleus, plays a significant role. The 165Ho(n,γ)166Ho 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 in 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 166Ho is a significant branching nucleus, characterized by a ground state half-life of 26.9 hours and an isomeric state (7^-) with a half-life of 1200 years, primarily formed by neutron capture of 165Ho. Consequently, the 165Ho(n,γ)166Ho reaction not only depletes the abundance of 165Ho but also influences the abundance of 166Ho 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 166Ho [$T_1/_2$ = 26.9 h, E_β = 1.77 MeV (48%) and 1.85 MeV (51%), E_γ = 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 165Ho is the only naturally stable isotope of holmium (natHo) and is used to produce 166Ho through the (n,γ) reaction. Accurate data on the neutron capture cross section and resonance integral for the 165Ho(n,γ)166Ho reaction are essential for evaluating neutron irradiation time, activity, and yield of 166Ho 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 [26], 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}^{dep} = 250 keV and E_{max}^{dep} = 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, with the parameters being as follows:

- LiSi #1: LiSiData1.BCid = 5
- LiSi #2: LiSiData1.BCid = 6

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}Li primarily generates helium nucleus (α) and tritium nucleus (T), resulting in a bimodal structure in the energy spectrum. Due to the high energy peak (2.73 MeV) of T, 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).

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AUTHOR CONTRIBUTIONS STATEMENT

Su-Ya-La-Tu Zhang: Conceptualization, Methodology, Writing–review & editing
Yong-Shun Huang: Investigation, Data curation, Writing–original draft
Wei Jiang, Jie Ren, and Rui-Rui Fan: Measurement, Methodology
De-Xin Wang: Formal analysis
Guo-Li and Dan-Dan Niu: Measurement
Chun-Lei Zhang: Editing & English grammar correction
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.