Substitution measurement of the 65Cu(γ, n)64Cu reaction cross section using natCu and 63Cu targets with quasi-monoenergetic γ beams at SLEGS*

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Date: 2025-07-09T19:31:07+00:00

Abstract

To overcome the difficulty and expensive cost for some specific isotopic targets, a substitution method was proposed to measure the cross section of (γ, n) reactions. Considering that the natural copper element (natCu) only has 63Cu and 65Cu isotopes, the 65Cu(γ, n)64Cu reaction was taken as an example to test the substitution method. Using quasi-monoenergetic γ beams provided by the Shanghai Laser Electron Gamma Source (SLEGS) of the Shanghai Synchrotron Radiation Facility (SSRF), the natCu(γ, n) was measured from Eγ= 11.09 to 17.87 MeV. Furthermore, based on the 63Cu(γ, n) reaction measured using the same experimental setup at SLEGS, the 65Cu(γ, n)64Cu was extracted using the substitution method. The abundance variation of natural copper, showing a significant influence in the cross section, is also investigated. The results were compared to the existing experimental data measured by bremsstrahlung and positron annihilation in-flight sources, and the TALYS 2.0 predictions. The γ strength function (γSF) of 65Cu is obtained from the 65Cu(γ, n) data, and the reaction cross section of 64Cu(n, γ) was further calculated.

Full Text

A Substitution Measurement for the Cross Section of the 65Cu(γ, n)64Cu Reaction Using natCu and 63Cu Targets by Quasi-Monoenergetic γ Beams at SLEGS

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To overcome the difficulty and high cost associated with specific isotopic targets, a substitution method was proposed to measure the cross section of (γ, n) reactions. Considering that natural copper (natCu) contains only 63Cu and 65Cu isotopes, the 65Cu(γ, n)64Cu reaction was selected as an example to test this substitution method. Using quasi-monoenergetic γ beams provided by the Shanghai Laser Electron Gamma Source (SLEGS) at the Shanghai Synchrotron Radiation Facility (SSRF), the natCu(γ, n) reaction was measured from $E\gamma$ = 11.09 to 17.87 MeV. Furthermore, based on the 63Cu(γ, n) reaction measured using the same experimental setup at SLEGS, the 65Cu(γ, n)64Cu cross section was extracted using the substitution method. The influence of natural copper's isotopic abundance variation, which significantly affects the cross section, was also investigated. The results were compared with existing experimental data obtained using bremsstrahlung and positron annihilation in-flight sources, as well as TALYS 2.0 predictions. The γ strength function (γSF) of 65Cu was obtained from the 65Cu(γ, n) data, and the cross section for the inverse 64Cu(n, γ) reaction was subsequently calculated.

Keywords: photoneutron cross section, flat efficiency detector, laser Compton scattering, γ rays, SLEGS, substitution measurement, copper isotopes

INTRODUCTION

The 65Cu(γ, n)64Cu reaction has important applications in both medical and scientific research. In medicine, 64Cu is a short-lived $\beta+$ emitter (T1/2 = 12.7 h; $\beta+$ with a mean energy of 278 keV and branching ratio of 61.5%, and $\beta-$ with a mean energy of 191 keV and branching ratio of 38.5%) that is widely used in nuclear medical imaging techniques such as positron emission tomography (PET) and single photon emission computed tomography (SPECT). For example, 64Cu-labeled peptides such as 64Cu-DOTATATE are used in the diagnosis of neuroendocrine tumors, 64Cu-labeled oxygen depletion probes such as 64Cu-ATSM are used to detect hypoxic regions in tumors, and 64Cu-labeled prostate specific membrane antigen (PSMA) ligands are employed in PET imaging of prostate cancer to precisely localize tumor cells. In addition, 64Cu can be used in radionuclide therapy to destroy tumor cells through its $\beta+$ and $\beta-$ decay properties.

In scientific research, the 65Cu(γ, n)64Cu reaction is also of great importance. In nuclear physics, this reaction enables studies of nuclear structure, properties, and reaction mechanisms. The photoneutron reaction for producing 64Cu offers advantages over traditional methods by avoiding the use of rare and expensive 64Ni targets and complex chemical separation procedures. Furthermore, the 64Cu(n, γ)65Cu reaction plays a key role in quality control of the medical isotope 64Cu, helping assess potential 64Cu loss during neutron irradiation and providing valuable data for understanding nucleosynthesis of medium-mass elements in stars through neutron capture processes.

During the last century, laboratories worldwide conducted experimental studies on the photoneutron reaction of 65Cu using bremsstrahlung (BR) sources and positron annihilation in flight (PAIF) sources with the activation method. Varlamov et al. evaluated existing 65Cu(γ,n) experimental data, revealing considerable discrepancies. The evaluated and experimental cross sections show that BR and PAIF results are close within the low incident γ energy range but differ significantly at high γ energies, reflecting systematic errors caused by misclassification of neutron channels. The quasimonochromatic γ-ray source generated by laser Compton scattering provides an opportunity to measure the (γ, n) reaction with improved precision, helping to resolve differences in existing data.

In this work, cross sections for the natCu(γ, n) reaction were measured within the giant dipole resonance (GDR) energy region using the SLEGS beamline at SSRF. The 65Cu(γ, n) cross sections were determined via the substitution method using previously measured 63Cu(γ, n) reaction data. Furthermore, neutron capture cross sections for 64Cu were also extracted. The article is organized as follows: Section II describes the experimental procedure for measuring natCu(γ, n) cross sections. Section III presents the methods for processing experimental data and the results for quasi-monoenergetic and monochromatic cross sections of 65Cu(γ, n) obtained by the subtraction method. Section IV discusses discrepancies between measured data and existing experimental results, as well as extraction of the radiative neutron capture cross section of 64Cu. A brief conclusion is given in Section V.

II. EXPERIMENT

This experiment was carried out at the SLEGS beamline station in SSRF. The beamline uses inverse Compton scattering technology: 3.5 GeV electrons in the SSRF storage ring collide with photons from a 10.64 µm-wavelength, 100 W CO2 laser, generating quasimonochromatic gamma rays with tunable energies from 0.66 to 21.7 MeV. The energy of the γ beam is adjusted in slant-scattering mode with a minimum step of 10 keV. For details on (γ, n) reaction measurements at SLEGS, see Refs. [20–23].

The cross sections for the natCu(γ, n) reaction were measured at 37 energy points ranging from 10.9 MeV (= 90°) to 17.8 MeV (= 130°). For each angle, the measurement time and neutron statistics were as follows: 2 hours with neutron statistics exceeding 1.0×10^4 for $\leq 98°$, 1 hour with neutron statistics exceeding 4.3×10^4 for $99° \leq \ \leq 105°$, and 0.5 hours with neutron statistics exceeding 4.8×10^4 for $\geq 106°$. The laser Compton scattering (LCS) γ beam, after passing through the collimation system, irradiated the experimental target positioned at the center of the 3He flat efficiency detector (FED) array. The in-beam gamma flux was monitored by a large-volume BGO detector downstream of the FED. The incident γ spectrum was reconstructed using the direct unfolding method combined with a Geant4-simulated detector response matrix [Figure 2: see original paper]; see Refs. [25–27] for details.

A. Targets

The natCu target of 3.15 g was placed in polyethylene target holders and irradiated using LCS γ beams. The alignment of the target and the FED with the LCS γ-ray beam was adjusted using a MiniPIX X-ray pixel detector for collimation. Detailed specifications can be found in Table 1 .

The target holder has a 10-mm diameter window. Considering that the size of the LCS γ-ray beams was approximately 4 mm in diameter at the target position, the 10 mm diameter of the window was sufficient for the measurement while avoiding the influence of neutrons from polyethylene.

B. Measurements

The SLEGS facility features a new FED with 26 proportional counters arranged in three concentric radii within a polyethylene moderator shielded by a 2 mm Cd sheet [23]. The counters, with an effective length of 500 mm and filled with

3He gas at 2 atm pressure, were read out through Mesytec MDPP-16 digitizers and MVME DAQ. Fig. 3 [Figure 3: see original paper] shows the efficiency curves of each ring and the total efficiency curve simulated by GEANT4 using the real detector configuration. For the neutron evaporation spectrum, the total detector efficiency increases from 35.64% at 50 keV to 42.32% at 1.65 MeV, then decreases slowly to 39.05% at 4 MeV [28]. The efficiency calibrated using the 252Cf source is 42.10 ± 1.25%, corresponding to an average neutron energy of 2.13 MeV. In our experiment, we used the ring-ratio technique to obtain the average energy of neutrons produced by the (γ, n) reaction and then estimated the detector efficiency using its calibration curve [29, 30].

III. DATA ANALYSIS AND SUBSTITUTION MEASUREMENT METHOD

A. Data Analysis Method

In the monochromatic approximation, the photoneutron cross section can be expressed by the integral equation [31]:

$$\int_{E_{\text{threshold}}}^{E_{\max}} n_\gamma(E)\sigma(E)dE = N_\gamma N_t \xi \epsilon_n$$

where $n_\gamma(E)$ is the energy distribution of LCS γ-ray beams normalized in the integration region, $\sigma(E)$ represents the photoneutron cross section, N_n the number of detected neutrons, N_t the number of target nuclei per unit area, and N_γ the number of incident γ-rays with energies above the neutron threshold. The self-attenuation coefficient ξ is given by [32] as $(1-e^{-\mu t})/(\mu t)$, where μ is the linear attenuation coefficient of the sample and t the thickness of the sample. The photoneutron cross section in the monochromatic approximation is calculated by:

$$\sigma_{E_{\max}}(\gamma, n) = \frac{N_n}{N_\gamma N_t \xi \epsilon_n}$$

In the experiment, the laser pulse cycle was 1000 µs (50 µs on + 950 µs off). Due to the energy dispersion of LCS γ-ray beams, the monochromatic approximation is insufficient to determine photoneutron cross sections. When neutrons were counted with the FED array, flat-efficiency regions for each detector ring were determined, considering neutron energy and detector parameters. The median method was used to establish optimal efficiency points and improve neutron count statistics.

To solve the unfolding problem, the integral in Eq. (1) is approximated as a summation for each γ beam profile, resulting in a system of linear equations $\sigma_f = D\sigma$ [33, 34]. The folding iteration method was used to solve this underdetermined system. Starting with a constant trial function σ_0, the folded vector $\sigma_0^f = D\sigma_0$ was calculated [35–38]. The next trial input function σ_1 was obtained by adding the difference between the experimental spectrum σ_{exp} and the folded spectrum σ_0^f to σ_0, after spline interpolation to match dimensions. The iteration proceeds with:

$$\sigma_{i+1} = \sigma_i + (\sigma_{\text{exp}} - \sigma_i^f)$$

with typical convergence achieved in about three iterations, making the reduced χ^2 value approximate 1.

B. Substitution Measurement Method

In this experiment, the natural copper target (natCu) has an isotopic abundance of 69.15% for 63Cu and 30.85% for 65Cu. The one-neutron (S_n) and two-neutron (S_{2n}) separation energies for 63Cu are 10.86 MeV and 19.74 MeV, respectively, while for 65Cu they are 9.91 MeV and 17.83 MeV, respectively [42, 43]. Within the energy range where the one-neutron separation energy thresholds of 63Cu and 65Cu overlap, the FED detector measures neutrons from both the 63Cu(γ, n) and 65Cu(γ, n) reactions as:

$$N_{\text{natCu}} = N_{\text{63Cu}} + N_{\text{65Cu}},$$

where N_{natCu} represents the result obtained from direct measurement, while N_{63Cu} needs to be calculated by combining the monoenergy cross section $\sigma(\gamma, n)$ with the current energy spectrum $n_\gamma(E_\gamma)$. The detailed calculation process is shown in Eq. (6):

$$N_{\text{63Cu}} = N_t \xi \epsilon_n \int_{E_{\text{threshold}}}^{E_{\text{max}}} n_\gamma(E_\gamma) \sigma_{\text{63Cu}}(\gamma, n)(E_\gamma) dE_\gamma.$$

By substituting the neutron count of N_{63Cu} into Eq. (7), the quasi-monoenergetic photoneutron cross section data of 65Cu can be obtained as:

$$\sigma_{\text{65Cu}}(\gamma, n) = \frac{N_{\text{natCu}} - N_{\text{63Cu}}}{N_\gamma N_t \xi \epsilon_n}.$$

The monochromatic cross section of the 65Cu(γ, n)64Cu reaction was derived using a deconvolution iteration method. Fig. 4 [Figure 4: see original paper] compares the quasi-monoenergetic and monochromatic cross sections of 65Cu.

According to Eq. (7), the statistical uncertainty is mainly caused by N_n. The methodological uncertainty arises from the extraction algorithm for N_n and the deconvolution method incorporating the simulated BGO response matrix.

Systematic uncertainty, the dominant source of total error, includes the γ-flux uncertainty from the copper attenuator, target thickness uncertainty, and FED efficiency uncertainty. Table 2 summarizes the systematic and methodological uncertainties.

IV. RESULTS AND DISCUSSION

65Cu Photoneutron Reaction Cross Section

The 65Cu(γ, n)64Cu photoneutron cross section data measured at SLEGS are compared with existing experimental and evaluated data in Fig. 5 [Figure 5: see original paper]. Although the overall trends are consistent, significant discrepancies are observed in absolute values. The experiment by Fultz et al. [13] at Lawrence Livermore National Laboratory (LLNL) is closest to the results of this work. They used BF3 neutron detectors and measured in the energy range of 9.34 to 27.78 MeV. Data from Katz et al. [11] using a BR source at the BETAT accelerator in Canada with a 22 MeV endpoint energy are notably higher than the LLNL data. Antonov's [12] measurements using the BR source at JINR are significantly higher than other datasets in both neutron threshold and cross-section values.

Isotopic abundance variations influence cross sections and cause result discrepancies. The photoneutron cross sections change systematically with alterations in 65Cu abundance. Based on data from the 2025 SLEGS experiment, comparing cross sections of 65Cu at 35.85%, natural abundance (30.85%), and 25.85%, the photoneutron cross sections decrease as the abundance of 65Cu increases (e.g., from 25.85% to 35.85%) and increase as the abundance decreases (see the inset figure in Fig. 5). This occurs across all energy ranges and is most noticeable near the threshold and peak position in the cross section distribution. It is suggested that isotopic abundances in target materials should be determined prior to analysis of substitution data. The sensitive dependence on isotopic abundance also suggests that enhanced isotopic targets could be used to determine (γ, n) cross sections in addition to pure isotopic targets.

As discussed in Ref. [45], ratios of integral cross sections provide a clear indication of systematic differences among various data compilations. The integral cross sections in the S_n and $S_{\max}$ regions are given by:

$$\sigma_{\text{int}} = \int_{S_n}^{S_{\max}} \sigma(E)dE.$$

Based on these experimental data, integral ratios of the photoneutron reaction cross section were calculated for energy ranges from S_n to 15 MeV, 15 MeV to S_{2n}, and S_n to S_{2n}, with results presented in Table 4 . In the energy range of S_n to 15 MeV, the measured results show a difference of only 0.8% from Fultz data and less than 0.4% discrepancy from TENDL-2021 theoretical calculations, while discrepancies with other datasets exceed 40%. In the 15 MeV to S_{2n} range, the minimum difference between this work and Katz data [11] is 0.4%, with differences exceeding 20% for other datasets. In general, the neutron threshold and peak position of the cross section demonstrate good consistency with measurements by Fultz [13] and the TENDL-2021 evaluation [46]. The neutron threshold exhibits favorable agreement with Katz data [11]. However, notable differences in both neutron threshold and peak position are observed compared to Antonov's data [12].

64Cu Radiative Neutron Capture Cross Section

The gamma strength function (γSF) [47] describes the average probabilities of gamma decay and absorption in nuclear reactions and is a crucial parameter for characterizing nuclear reaction processes. When research involves reactions with gamma rays, such as (n, γ) and (γ, n), the precision of the γSF is particularly important. According to the principle of detailed balance [48] and the generalized Brink assumption [49, 50], the upward strength function $f_{X\ell}(E_\gamma)$ is approximately equal to the downward strength function. Therefore, the (upward) photoneutron cross section $\sigma_{\gamma n}$ is connected to the (downward) γSF [52] by:

$$f_{X\ell}(E_\gamma) \approx f_{X1}(E_\gamma) = \frac{3\pi^2\hbar^{2c^2}}{\sigma_{\gamma n}(E_\gamma)},$$

where the constant $1/(3\pi^2\hbar^{2c^{2)}} = 8.674 \times 10^{-8}$ mb^{-1}MeV^{-2}. Using this relationship, the γSF can be obtained from the photoneutron reaction cross section.

The γSF model in TALYS is compared with the experimentally constrained γSF. The model prediction closest to the experimentally obtained γSF is selected. Furthermore, the γSF model is constrained using the normalization parameter G_{norm} in the TALYS 2.0 toolkit, with optimization achieved by minimizing the χ^2 value, thus making theoretical calculations more consistent with experimental γSF values. The γSF of 65Cu(γ, n) constrained by measured (γ, n) data is shown in Fig. 6 [Figure 6: see original paper], with χ^2 determined by:

$$\chi^2 = \sum_{i=1}^{N} \left(\frac{\sigma_{\text{th},i} - \sigma_{\text{exp},i}}{\sigma_{\text{err},i}} \right)^2,$$

where N is the total number of experimental data points, and $\sigma_{\text{th},i}$, $\sigma_{\text{exp},i}$, and $\sigma_{\text{err},i}$ denote the theoretical value, experimental measured value, and experimental error of the γSF at the i-th data point, respectively. The neutron capture cross section of 64Cu, after adjusting the optimal G_{norm} value, is shown in Fig. 7 [Figure 7: see original paper]. Specifically, G_{norm} is set to 1.2 in the Kopecky-Uhl generalized Lorentzian model [53] and 1.4 in the Goriely hybrid model [54].

When constrained by G_{norm}, the hybrid model [54] yields the smallest χ^2 value among all investigated models, showing the best agreement with this experimental dataset. Similarly, Utsunomiya et al. [39] and Li et al. [44] previously conducted related research and measured (n, γ) radiative reaction cross sections for 136,137Ba and 62Cu isotopes. In this study, due to the lack of experimental data on low-lying excited states and neutron resonance spacings of 65Cu, constraints on the nuclear level density (NLD) model are limited, resulting in substantial theoretical uncertainties.

The study of the 64Cu(n, γ) cross section is valuable for improving nuclear data and optimizing medical isotope production. It is closely related to the 65Cu(γ, n)64Cu cross section measurement through the principle of detailed balance, and the former can verify the reliability of the latter, collectively highlighting the overall significance of this research.

V. SUMMARY

The reaction cross sections of natCu(γ, n) were measured in the incident energy range of 11.09 to 17.87 MeV using the 3He FED detector array developed at SLEGS. Based on the measured photoneutron cross section data and previously measured results for 63Cu(γ, n) at SLEGS, the reaction cross sections of 65Cu(γ, n)64Cu were obtained using the cross section substitution method. Compared with existing experimental data, the reliability of this method was demonstrated, providing a new approach for photoneutron cross section measurements. Given the extensive applications of 64Cu in medical fields such as nuclear medicine imaging and tumor therapy, clarifying existing discrepancies in the reaction cross sections of 65Cu(γ, n)64Cu is likely to play an important role in these fields. The sensitivity to isotopic abundance changes in natural copper targets shows that enhanced purity of specific isotopes could be used to measure (γ, n) cross sections via the substitution method proposed in this work. The experimentally constrained γSF of 65Cu was extracted from the 65Cu(γ, n)64Cu cross section distribution. Additionally, the cross section curve for its inverse reaction, 64Cu(n, γ), was calculated, providing a new approach for extracting (n, γ) cross sections for some unstable nuclides.

VI. ACKNOWLEDGMENT

The authors thank the SSRF accelerator team for stable maintenance of the electron beam during experiments and the SLEGS team (https://cstr.cn/31124.02.SSRF.BL03SSID) for assistance in measurement and data analysis.

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