Photonuclear reaction study with the (p, γ) resonance γ-source

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Abstract.
The (p, γ) resonance is a good way to produce monoenergetic γ rays. It becomes an important tool for studying photonuclear reactions. In this work, 13C(p, γ)14N resonance is studied to produce 9.17 MeV γ ray using the 2×1.7 MV tandem accelerator at CIAE. The flux of 9.17 MeV γ was determined to be 2.3×10^7/s. 197Au photoneutron cross section was measured to be 45.4±6.9 mb under the irradiation of 13C(p, γ)14N resonance γ-source. The value is close to the previous results. It certifies that we have developed an experimental method for photonuclear reaction study.

1 Introduction

Experimental studies of photonuclear reactions have opened the way for a variety of important applications. Here we will mainly focus the applications on nuclear reactor design [1] and nuclear astrophysics [2–4]. In nuclear reactor design, accurate photonuclear data are needed in many areas as photonuclear reaction plays a non-neglectable role in nuclear reactors, such as reactor in-core dosimetry, radiation damage estimates in reactor structural materials, safeguards, and fast reactor calculations etc. In nuclear astrophysics, one of the unanswered questions is to determine how the heavy elements were synthesized during the evolution of universe [5]. It is widely accepted that the bulk of the nuclei heavier than iron have been synthesized by neutron capture in the astrophysical r-process. Those neutron-capture processes cannot be accounted for the synthesis of some of the heavy (A ≥ 100) neutron-deficient nuclei. These nuclei are shielded from the chain of β− decays by other stable isotobars. The production mechanism for these so-called p-nuclei [6] is photodisintegration in the astrophysical γ process by successive (γ, n), (γ, p), and (γ, α) reactions.

The difficulty among these studies discussed above is that photonuclear cross-section data are scarce in Giant Dipole Resonance (GDR) energy range [7, 8], and especially there is a lack of data in some important cases. On one hand, most of the existing experimental data are from bremsstrahlung photon sources. But only measurements from monoeenergetic sources give emission spectra directly useful for cross-section evaluations. On the other hand, the energies of astrophysical interest for (γ, n) cross sections practically no data exist for the p-nuclei because of their low abundance. Reliable experimental data would be a great improvement to reduce the nuclear physics uncertainties of astrophysical model calculations. Under this background, the experimental study of photonuclear reaction is increasingly important.

The photonuclear data were reported to have quite large discrepancy using traditional measurements [9, 10], most of which were using bremsstrahlung unfolding techniques [11] or the in-flight annihilation of monochromatic positrons [12]. The (γ, n) cross sections measured by using bremsstrahlung γ sources could create systematic errors due to source uncertainties, which would lead to increase the errors of absolute cross section values. Photoneutron data from in-flight annihilation of monochromatic positrons source also have considerable systematic errors even at the peak values of the GDR mainly owing to the difficulty in calibrating the incident quasi-monoenergetic photon beam accurately. By comparison with traditional methods, the experiments on monoenergetic γ sources have definite advantages in obtaining accurate cross sections, since they are free from a low-energy tail. (p, γ) resonance is a good way to produce monoeenergetic γ rays. There are several different reactions to be chosen to produce high energy γ rays ranging from 6 MeV to 20 MeV. The energy range just fits in the GDR region. In this work, 13C(p, γ)14N resonance [13–15], which emits 9.17 MeV γ rays, was used for (γ, n) measurements.

197Au(γ, n) reaction is well suited as a standard for (γ, n) experiments [16] and regarded as one of the most fundamental data in nuclear physics, since gold has only one stable isotope. Its (γ, n) cross section has been widely measured around the GDR by several different methods in the last decades [17–23], and recently a very precise measurement was made in the entire energy range using a quasi-monoenergetic γ-source produced in laser-induced Compton backscattering with relativistic electrons [21]. In this work, we measure the 197Au(γ, n) cross section at
γ-ray energy of 9.17 MeV using $^{13}$C(p, γ)$^{14}$N resonance in order to develop a photoneutron experimental setup.

2 Experiment

Two separate experiments were carried out with the 2×1.7 MV tandem accelerator at China Institute of Atomic Energy (CIAE). The first measurement was focused on γ-ray intensity calibration for the $^{13}$C(p, γ)$^{14}$N resonance γ-source, while the second one was concerned on $^{197}$Au(γ, n) cross section measurement.

2.1 The $^{13}$C(p, γ)$^{14}$N resonance γ-source

The experimental arrangement used in the measurement of the γ-ray intensity calibration is shown in Fig.1. The proton beam extracted from the tandem accelerator was focused onto a $^{13}$C thick-target. In the experiment, the proton beam intensity was about 8 µA with the beam energy of 1.750 MeV. The target consists of a 100 µg/cm² foil of $^{13}$C (isotopically enriched to 97%) onto an air-cooled Ta disk with the thickness of 300 µm.

![Figure 1. Scheme of detector setup](image)

The gamma-ray spectrum for the p+$^{13}$C reaction has been measured with a HPGe (35% relative efficiency), a φ3-inch × 3-inch LaBr₃(Ce) and a φ5-inch × 5-inch NaI(Tl). Different detectors played different roles in the measurement. HPGe is used for the thick target yield measurement. LaBr₃(Ce) is used for angular distribution measurement. NaI(Tl) is used for γ beam monitor. Under these conditions the γ-ray spectrum consisting of 9.17 MeV line is shown in Fig 2. The full-energy peak and its escape peaks are displayed clearly.

![Figure 2. Sample spectrum of $^{13}$C(p,γ)$^{14}$N resonance detected by HPGe detector](image)

Offline measurements were taken in a low background shielding system including a HPGe detector. The detector relative efficiency is 105% for the 1.332 MeV gamma ray. The counting rate for the low background system is around 0.1 Hz within the energy region of 50–3000 keV. The details of the low background shielding system can be seen in ref [24].

2.2 The $^{197}$Au(γ, n) reaction measurement

Two air-cooled disks of $^{197}$Au were used, which were coated with 100 µg/cm² foil of $^{13}$C. The gold disks were enriched to 99% with a diameter of 10 mm. Experimental setup is shown in Fig. 3. The $^{13}$C coated side of Au is facing to the beam, and the proton beam is impinged on the $^{13}$C coating directly. Two Au targets were irradiated under the same condition for several hours respectively. A HPGe detector was also used at the same time as a γ beam monitor. In order to eliminate the influence from short-lived nuclei, the irradiated targets were both cooled down for 3 to 4 hours.

![Figure 3. Experiment setup of $^{197}$Au(γ, n) reaction measurement](image)

γ-ray energies from $^{13}$C(p, γ)$^{14}$N resonance range from hundreds of keV to 9.17 MeV, but the standard radioactive γ-source can only provide the γ-ray energies up to ~3.5 MeV. Then the detector calibration has to use other method. Geant4 [25] is a commendable tool, and it has been widely tested and verified. In some previous (p, γ) resonance experiments, they used the Geant4 codes for the detector relative efficiency simulation. It was found that the simulations agreed well with the experiment results [26]. In our work, standard γ-sources $^{56}$Co and $^{152}$Eu
were used for the absolute efficiency calibration. For the high energy region up to 9 MeV, a extrapolation by using Geant4 simulation was used, as shown in Fig.4. The determined absolute efficiency calibrated by standard sources is shown together with the Geant4 simulation. Furthermore, the relative efficiencies, which were deduced from the previous data measured by Kiener [26] in company with our data, for 2143, 2313, 2726, 6445, 7027 and 9170 keV are also compared with the simulations. It can be found that the simulation fits the relative efficiency with different energies very well. Finally, the absolute efficiency of HPGe at 9.17 MeV is obtained to be $\epsilon = 7.21 \times 10^{-6}$.

![Figure 4](image_url1)

**Figure 4.** Absolute full-energy peak efficiency of the HPGe detector determined with Geant4 simulation and the standard $\gamma$-sources.

![Figure 5](image_url2)

**Figure 5.** Excitation curve for 9.17 MeV $\gamma$ ray from the reaction $^{13}$C(p, $\gamma$)$^{14}$N.

### 3 Experimental results

#### 3.1 Thick-target yield of 9.17 MeV $\gamma$-ray

The yield of 9.17 MeV $\gamma$-ray from $^{13}$C(p, $\gamma$)$^{14}$N resonance was measured in a quite broad range of proton energy from 1.740 MeV to 1.780 MeV, as shown in Fig. 5. A Saturation appears at the energy region from 1.750 to 1.758 MeV for the thick target yield measurement. In order to get accurate thick target yield data, some simulations have also been done using Geant4. It shows in Fig. 5 that the simulation agrees with the experimental data very well. At last, we measured the thick target yield at the beam energy of 1.750 MeV. The calculation of thick-target yield can be written as:

$$ Y = \int \int_{0}^{\pi} Y(\theta) \sin(\theta) d\theta d\phi = 2\pi \int_{0}^{\infty} Y(\theta) \sin(\theta) d\theta $$  \hspace{1cm} (1)

where $Y(\theta)$ is the 9.17 MeV $\gamma$ ray yield as the function of $\theta$. Finally, the thick-target yield of 9.17 MeV $\gamma$-ray is determined to be $(4.7 \pm 0.4) \times 10^{-5}$ $\gamma$/proton. The $\gamma$-flux will be $2.3 \times 10^{3}$/s when the proton beam intensity is $8 \mu$A.

![Figure 6](image_url3)

**Figure 6.** $^{196}$Au decay paths.

![Figure 7](image_url4)

**Figure 7.** Sample spectrum of $^{196}$Au $\beta$ decay detected by the low background shielding system.

| Target No. | Detected $\gamma$ number | Cross section(nb) |
|------------|--------------------------|------------------|
| 1          | 1566                     | 45.2$\pm$6.9     |
| 2          | 6610                     | 45.6$\pm$6.9     |

### 3.2 Cross section of $^{197}$Au($\gamma$, n) reaction

Two gold targets were irradiated by $\gamma$ rays with the energy of 9.17 MeV for 6 hours and 5.5 hours respectively, and then they were both placed in a lead chamber to cool down respectively for 3.6 hours and 3.1 hours. After about three hours, the Au targets were then put into the low background shielding chamber for $\beta$ decay measurement. The
half-life of $^{196}$Au is 6.18 days. It has two decay paths as shown in Fig. 6, one is changing to $^{196}$Pt by electron capture and the other is changing to $^{196}$Hg by $\beta^-$ decay [27]. 355.7 keV $\gamma$-ray from $^{196}$Au electron capture has the largest emission probability with the value of 87%. It is better to use this $\gamma$-ray to determine the yield of $^{196}$Au. A HPGe detector was used to detect the $\gamma$-rays from $^{196}$Au beta decay. The efficiency of this detector was calibrated before the beta decay measurement. The efficiency calibration was using a standard point source $^{133}$Ba since its 356.0 keV characteristic energy as the 355.7 keV $\gamma$-ray in the Au target were considered. The cross sections of $^{197}$Au($\gamma$, n) at 9.17 MeV from two rounds of measurement are obtained as listed in Table 1. The total experimental uncertainties including statistical errors (91%) and systematic errors(9%) are indicated in Table 1.

The duration of the detection for the first target was 20.6 hours, and the second one was 94.2 hours. A sample of the measured decay spectrum from $^{196}$Au electron capture is shown in Fig. 7. In order to get accurate absolute intensity of 355.7 keV $\gamma$-ray, both the attenuation of 9.17 MeV $\gamma$-ray and the self-absorption for 355.7 keV $\gamma$-ray in the Au target were considered. The cross sections of $^{197}$Au($\gamma$, n) at 9.17 MeV from two rounds of measurement are obtained as listed in Table 1. The total experimental uncertainties including statistical errors (91%) and systematic errors(9%) are indicated in Table 1.

4 Conclusion

Experimental study of $^{13}$C(p, $\gamma$)$^{14}$N resonance as a $\gamma$-source has been performed with the 2x1.7 MV tandem accelerator at China Institute of Atomic Energy. The flux of 9.17 MeV $\gamma$ was deduced to be 2.3x$10^7$/s measured by HPGe, LaBr$_3$(Ce) and NaI(Tl) detectors. Two Au targets were irradiated with the $^{13}$C(p, $\gamma$)$^{14}$N resonance $\gamma$-source. The $^{197}$Au($\gamma$, n) cross section was measured to be 45.4±6.9 mb. It is very close to the value 48.3±2.1 mb [21] measured by laser Compton scatter $\gamma$-source, and even more closer to the value of 45±5 mb [20] measure by the in-flight positron annihilation $\gamma$-source. It certifies that we have successfully established the experimental method for photonuclear reaction study.

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References

[1] IAEA, Handbook on photonuclear data for applications: Cross-Sections and spectra (International Atomic Energy Agency, Vienna, 2000) IAEA Report No. 1178.
[2] D. L. Lambet, Astron. Astrophys. 3, 201 (1992)
[3] M. Arnould, K. Takahashi, Rep. Prog. Phys. 62, 395 (1999)
[4] S. E. Woosley, W. M. Howard, Astrophys. J. Suppl. 36, 285 (1978)
[5] E. Haseltine, Discovery. 23, 37 (2002)
[6] M. Arnould, S. Goriely, Phys Rep. 384, 1 (2003)
[7] Kaoru. Y. Haru, Hideo. Harada, Fumito. Kitatani et al., J. Nucl. Sci. Technol. 44, 938 (2007)
[8] Fumito. Kitatani, Hideo. Harada, Shinji. Goko et al., J. Nucl. Sci. Technol. 47, 367 (2010)
[9] Fumito. Kitatani, Hideo. Harada, Shinji. GOKO et al., Phys Rep. 48, 1017 (2011)
[10] B. L. Berman, R. E. Pywell, S. S. Dietrich et al., Phys. Rev. C. 36, 1286 (1987)
[11] S. Costa, F. Ferrero, C. Manfredotti, L. Pasqualini et al., Nuovo. Cim. 51B, 199 (1967)
[12] P. Carlos, H. Beil, R. Bergere et al., Nucl. Phys. A. 258, 365 (1976)
[13] W. Biesiot and Ph. B. Smith, Phys. Rev. C. 24, 2443 (1981)
[14] D. Vartsky, and M. B. Goldberg, G. Engler et al., Nucl. Phys. A. 505, 328 (1989)
[15] S. S. Hanna and L. Meyerschitzmeister, Phys. Rev. C. 115, 986 (1959)
[16] K. Vogt, M. Babilon, W. Bayer et al., AIP Conf. Proc. 561, 137 (2000)
[17] B. L. Berman, R. E. Pywell, S. S. Dietrich, Phys. Rev. C. 36, 1286 (1987)
[18] G. M. Gurevich, L. E. Lazareva, V. M. Mazur et al.,Nucl. Phys. A. 351, 257 (1981)
[19] S. C. Fultz, R. L. Bramblett, T. J. Caldwell et al., Phys. Rev. 127, 1273 (1962)
[20] A. Veyssiere, H. Bell, R. Bergere, Nucl. Phys. A. 159, 561 (1970)
[21] Osamu. Itoh, Hiroaki. Utsunomiya, Hideto. Akimune et al., J. Nucl. Sci. Technol. 48, 834 (2011)
[22] C. Nair, M. Erhard, A. R. Junghans et al., Phys. Rev. C. 78, 055802 (2008)
[23] K. Vogt, P. Mohr, M. Babilon, Nucl. Phys. A. 707, 241 (2002)
[24] L. C. He, L. J. Diao, B. H. Sun et al., Nucl. Instrum. Methods. Phys. Res, A. 880, 22 (2018)
[25] S. Agostinelli, J. Allison, K. Amako et al., Nucl. Instrum. Methods. Phys. Res, A. 506, 250 (2003)
[26] J. Kiener, M. Gros, V. Tatischeff et al., Nucl. Instrum. Methods. Phys. Res, A. 519, 623 (2004)
[27] www.nndc.bnl.gov