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Neutron angular distribution in (γ , n) reactions with linearly polarized γ -ray beam generated by laser Compton scattering



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ABSTRACT

In 1957, Agodi predicted that the neutron angular distribution in (γ , n) reactions with a 100% linearly polarized γ -ray beam for dipole excitation should be anisotropic and universally described by the simple function of $a + b \cdot \cos(2\phi)$ at the polar angle $\theta = 90^{\circ}$, where ϕ is the azimuthal angle. However, this prediction has not been experimentally confirmed in over half a century. We have verified experimentally this angular distribution in the (γ , n) reaction for ¹⁹⁷Au, ¹²⁷I, and natural Cu targets using linearly polarized laser Compton scattering γ -rays. The result suggests that the ($\vec{\gamma}$, n) reaction is a novel tool to study nuclear physics in the giant dipole resonance region.

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Photonuclear reactions have an important role for developing nuclear physics [1] and for various applications such as non-destructive measurements of nuclear materials [2,3]. Recent progress in accelerator and laser physics has led to a new generation of photon beams, based on the technique of laser Compton scattering (LCS). Energy tunable quasi-monochromatic LCS γ -ray beams in the MeV energy region have been used for fundamental science and various applications at Duke University [4], the National Institute of Advanced Industrial Science and Technology in Japan [5], and NewSUBARU in Japan [6]. An advantage of the LCS γ -ray beams comes from the fact that one can generate almost 100% linearly (circularly) polarized beams since the polarization of the laser is directly transferred to the scattered photons.

The polarized γ -rays have been widely used for the study of the nuclear structure via nuclear resonance fluorescence (NRF). With linearly polarized photons, the parity of each excited state in a nucleus is obtained by measuring the angular distribution of the emitted γ -ray yields. As a result, electric-dipole (E1) and

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magnetic-dipole (M1) transition strengths from the ground state can be measured directly (for examples, see Refs. [7,8]). In the photon energy region above the neutron threshold, the (γ , n) reaction is the dominant process in most nuclei and has been studied using non-polarized γ -rays (for example, Refs. [9,10].) Thus, the (γ , n) reaction with linearly polarized γ -ray beams has the potential to be used for studying detailed nuclear structures as NRF.

One important topic in the energy region above the neutron threshold is M1 strength. The "missing" M1 strength still remains an unsolved problem in nuclear physics [1]. A part of the missing M1 strength may be in the giant dipole resonance (GDR) region. The M1 strength is often used to estimate the interaction strength between neutrinos and nuclei in astrophysics. The neutrino-nucleus interaction plays an extremely important role for supernova explosions and syntheses of several rare isotopes [11–13]. However, there is no effective method to measure the M1 strength in the GDR region because of the overlap between the weak M1 strength and the strong E1 strength, although techniques like proton inelastic scattering [14] and total photodisintegration cross-section measurements [15] have been developed.

In the 1950s, angular distribution of cross sections for $(\vec{\gamma}, n)$ and $(\vec{\gamma}, p)$ reactions were calculated [16,17]. Agodi [17] predicted,

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in particular, that the angular distribution of nucleons emitted from states excited via dipole transitions with linearly polarized γ -rays at the polar angle of $\theta = 90^{\circ}$ should follow a simple function: $a + b \cdot \cos(2\phi)$, where ϕ is the azimuthal angle. This formula was derived from the conservation laws for angular momentum and parity without using nuclear models. Agodi presented also that the sign of the parameter b in the function $a + b \cdot \cos(2\phi)$ distinguishes between electric and magnetic transitions when the multipole order is known. This suggests that M1 strengths as well as E1 strengths from the ground state in the GDR region can be measured. However, there are few experimental data with polarized photon beams because the generation of linearly polarized γ -rays is difficult. Kellogg and Stephens measured angular distribution ratios on the ${}^{12}C(\vec{\gamma}, p){}^{11}B$ reaction with linearly polarized γ -rays provided from the ${}^{3}H(p, \gamma){}^{4}He$ reaction [18]. To evaluate M1/E1 ratios, the polarization asymmetry at $\theta = 90^{\circ}$ has been measured for ¹⁶O [19] and ²⁸Si [20] using ($\vec{\gamma}$, p) reactions with bremsstrahlung γ -rays and LCS beams, respectively.

Agodi's prediction is considered to be correct because it is derived from the fundamental spin-algebra, namely the conservation laws of angular momentum and parity. However, it is not clear whether the predicted angular distribution in nuclei in the medium-heavy and heavy mass regions can be observed. This is questionable since for heavy nuclei the level density at high excitation energies becomes large [21]. Because the sign of the parameter b for the M1 transitions is different from that of E1 [17], mixing of E1 and M1 transitions via many excited states can thus lead to a situation where the measured neutron angular distribution may be isotropic. In general, (ν, n) reaction cross sections in heavy nuclei are substantially larger than those of (γ, p) reactions by several factors because of the Coulomb barrier. In addition, in the case of $(\vec{\gamma}, n)$ reactions, there is an advantage that we can use relatively thick targets to obtain high statistics. However, there is no experimental data of $(\vec{\gamma}, n)$ reactions except some light nuclei such as the deuteron [22,23]. As the initial step for future applications of the $(\vec{\gamma}, n)$ reactions, we should demonstrate whether a neutron anisotropy can be measured for heavy nuclei or not.

The main purpose of the present paper is to show experimental results of angular distributions from $(\vec{\gamma}, n)$ reactions on three targets using a time-of-flight (TOF) method. The second aim is to revive the pioneering work by Agodi. Agodi's paper had been cited by previous papers in the 50–60's (for example, see Refs. [24,25,18]) but it has been almost forgotten at present. The progress of the LCS gamma-ray beam may enable us to study new physics using the $(\vec{\gamma}, n)$ reactions in the near future. Therefore, Agodi's work should be reevaluated as the pioneering work.

The LCS γ -ray beam was generated by scattering of laser photons and electron bunches stored at the electron storage ring New-SUBARU [6]. The details of the γ -ray source and basic experimental procedures are described in previous papers [26–28]. In order to verify the systematic behavior of the neutron angular distribution, we irradiated three Au, NaI, and natural Cu targets. Their sizes were \emptyset 10 mm \times 40 mm, \emptyset 10 mm \times 50 mm, and \emptyset 5 mm \times 50 mm for Au, NaI, and Cu, respectively.

The storage ring was operated by a single bunch mode. A pulsed electron bunch with an energy of 974 MeV was stored. The repetition rate was 2.5 MHz. A Q-switch Nd:YVO₄ laser system provided laser photons with a wavelength of 1064 nm. The laser power was 4 W and the pulse width was 8 ns. The maximum energy of the generated γ -ray beam was about 16.7 MeV, which was determined by the electron energy and the laser wavelength. On the other hand, the lowest energy of the γ -ray beam depends on the collimator size and the emittance of the electron beam. An extra collimator with a diameter of 6 mm was located before the target position. The diameter of the incident photon



Fig. 1. Schematic view of (γ, n) reactions with laser Compton scattering gamma-rays on ¹⁹⁷Au. The excited states on ¹⁹⁷Au are populated with linearly polarized photons and subsequently these exited states decay to the ground state and excited states on ¹⁹⁶Au through neutron emission. The solid arrows indicate high energy neutrons correspond to transitions from highly excited states on ¹⁹⁷Au to the ground state or a low excited state on ¹⁹⁶Au. The dashed arrows show low energy neutrons. The number of transitions with low energies is more than the number of the high energy neutrons, a smaller subset of transitions are selectively measured.

beam was about 6 mm on the Nal and Cu targets. This collimator was not used for the Au measurement to obtain high statistics and the beam diameter was larger than the Au target diameter. The energy width of the incident γ -ray beam was about 5 MeV for Au or 3 MeV for Nal and Cu. The flux of the incident γ -rays was measured by a Nal detector with a size of $\emptyset 6'' \times 5''$. The evaluated γ -ray flux was (1–2) $\times 10^6$ photons/s in an energy range from 12 MeV to 16.7 MeV.

Fig. 1 shows a schematic view of photonuclear reactions on ¹⁹⁷Au. Excited states in ¹⁹⁷Au are populated via photon absorption and subsequently each populated state decays predominantly to the ground state or an excited state in the neutron deficient isotope¹⁹⁶Au by neutron emission. The neutron energy was measured using the time-of-flight (TOF) method. The neutrons were measured using a plastic scintillation detector of $\varnothing100~\text{mm} \times$ 100 mm with a Ø78 mm photomultiplier (ET enterprises Co., type 9305). A lead shield with a thickness of 2 mm was located in front of the detector. The detector was located at the polar angle of $\theta = 90^{\circ}$ outside of the irradiation room with a concrete shield with a thickness of 540 mm, whereas individual targets were located inside of the room. Neutrons were emitted from the targets to the detector through a hole with a diameter of 80 mm. The length of the flight path was 970 mm. A time-to-amplitude converter (TAC) was used to measure the time interval between a start pulse from the detector and a stop pulse from the electron storage ring as a synchronization signal. The TAC signals were recorded using a multi-channel analyzer (MCA). The dead time of the MCA was below 3% and the neutron counting rate was 0.7-1.0 count/s.

To measure the angular distribution of neutrons, we changed the linear polarization plane angle of the LCS γ -rays. We used the same detector system without any rotation. The advantage of this method is that it deduces the systematic error. The angle of the linear polarization plane of the γ -ray beam was tuned by changing the linear polarization plane of the incident laser. It should be noted that there is a possibility that reflection mirrors to guide the laser beam to the LCS γ -ray generation point may change the angle of the laser polarization plane. Therefore, we should know the polarization plane angle at the collision point. For this purpose,



Fig. 2. Time-of-flight spectrum on the ¹⁹⁷Au(polarized γ , n)¹⁹⁶Au reaction at $\phi = 90^{\circ}$. The peaks of prompt γ -rays and neutrons are clearly separated. Natural background by radioactivity and cosmic rays are much lower than these signals.

the laser beam was extracted to the outside of the electron storage ring after the γ -ray generation without additional mirrors and the polarization angle was measured by a combination of a laser power meter and a Glan–Thompson prism. The polarization angle was determined by changing the angle of the Glan–Thompson prism until the angle was found which maximized the transmitted laser power. We measured the neutron energy spectra as a function of the laser polarization angle in a range from $\phi = 0^{\circ}$ to 360° in 30° steps for NaI and Cu, where the $\phi = 0^{\circ}$ was defined as the electric polarization vector being in the plane of the detector. We measured only 9 angles for the Au measurement to obtain high statistics.

Fig. 2 shows a TOF spectrum from the ¹⁹⁷Au measurement. The neutrons and prompt γ -rays are clearly separated. The energy spectra of the neutrons were derived from the TOF signals. Neutrons with energies lower than 2 MeV were not measured because of the detection efficiency. The measured maximum energy of the neutrons is about 8 MeV for Au. This energy is consistent with an expected energy of 8.6 MeV, the difference between the maximum photon energy of 16.7 MeV and the neutron separation energy of 8.1 MeV (see Fig. 1).

One of the excellent features of the LCS γ -ray beam is its short pulse length. The duration of the generated LCS γ -ray pulse is equal to the width of the shorter pulse of the electron and laser in the case of "head-on collision." The widths of the laser and the electron were 8 ns and 60 ps, respectively. Thus, the pulse width of the LCS γ -ray beam should be only 60 ps. The measured time width of the prompt γ -rays is, however, about 3 ns because of time fluctuation of the slow rise-time photomultiplier and the time jitter between the laser generation time and the external trigger signal from the electron storage ring. On the other hand, the time resolution, originating from the ratio of the detector thickness to the flight path length, was about 5%. Furthermore, the final energy resolution in the present setup was determined by the energy spread of the incident beam, since the incident beam energy spread of 3-5 MeV were wider than the energy resolution of about 5%, for example about 0.4 MeV at 8 MeV. These energy spreads of 3-5 MeV are much wider than the energy level spacing in the target nuclei. Thus, the level structures of the residual nuclei cannot be observed in the TOF spectra.

Fig. 3 shows the angular distributions of the ($\vec{\gamma}$, n) reactions for the three targets. The neutron yields are presented as a function of the azimuthal angle ϕ . The solid lines show the function of $a + b \cdot \cos(2\phi)$ obtained by χ^2 -fitting. The natural Cu consists of two stable isotopes ⁶³Cu (69%) and ⁶⁵Cu (31%). The sum for the two isotopes is also reproduced by this function. The NaI target consists of ²³Na and ¹²⁷I. Since the neutron separation energies of these nuclides are 12.4 MeV (²³Na) and 9.1 MeV (¹²⁷I) the contribution



Fig. 3. Angular distributions of neutrons of (γ, n) reactions on (a) Au, (b) Nal, and (c) natural Cu targets. The solid lines are obtained by χ^2 -fitting with a function of $a + b \cdot \cos(2\phi)$ predicted by Agodi in 1957. This function reproduces well these three data and thereby this prediction is experimentally examined for the first time over the wide range.

of ¹²⁷I is dominant for the NaI target. We would like to stress that the three neutron angular distributions are well described as the function of $a + b \cdot \cos(2\phi)$ independent of nuclides. Therefore, the theoretical prediction by Agodi is for the first time verified over the wide mass region.

A question we have to address here is why the anisotropy, b/a, increases with increasing atomic number of the targets. The GDR peak energy decreases, in general, with increasing atomic number. The level density in heavy nuclei is relatively high and the wave function becomes more complex. There are many types of transitions of which the sign of the parameter *b* are different from each other. As a result, the anisotropy of the neutron angular distribu-



Fig. 4. Plot of the anisotropy, b/a, of the neutron angular distribution as a function of neutron energy for ¹⁹⁷Au. The dashed line is a guideline. The anisotropy increases with increasing the neutron energy. This can be explained by the number of the transitions (see the main text).

tion in heavy nuclei is expected to decrease. However, the present result is opposite to this expectation.

For a more detailed investigation of the relatively strong anisotropy measured in the $^{197}\text{Au}(\gamma, n)^{196}\text{Au}$ reaction, we plot the anisotropy, b/a, as a function of the neutron energy in Fig. 4. It is clearly shown that the b/a ratio increases with increasing neutron energy. We also evaluate b/a values for NaI and Cu. The NaI anisotropy is $0.77\pm0.08~(0.17\pm0.09)$ for 4–8 MeV (2–4 MeV). The Cu anisotropy is $0.30\pm0.09~(-0.05\pm0.07)$ for 4–7 MeV (2–4 MeV). In the cases of NaI and Cu, the anisotropy also increases with increasing neutron energy. These three results lead to the conclusion that the anisotropy for all transitions as shown in Fig. 4 originates from the contribution of the high energy neutrons.

The trend that the anisotropy increases with increasing neutron energy can be understood by taking into account the number of combinations of an initial state before neutron emission and a final state. Since the incident γ -ray beam has an energy spread of about 5 MeV for Au, many excited states in ¹⁹⁷Au with an energy range of 12-17 MeV are populated. High energy neutrons decay from highly excited states near the maximum energy 17 MeV in ¹⁹⁷Au to the ground state or a low excited state on the residual nucleus ¹⁹⁶Au (see the solid arrows in Fig. 1). Thus, the anisotropy of these specific transitions can be selectively observed with the gate on high neutron energy. In contrast, in the case of the low energy neutron gate, there are a large number of transitions (see the dashed arrows in Fig. 1). The angular distribution of the low energy neutrons is the summation of these multiple transitions. The sign of a part of the transitions should be opposite to that of the other and thus the anisotropy of the summation decreases.

The maximum energy of the incident LCS beam was fixed to 16.7 MeV, whereas the neutron separation energy decreases with increasing the atomic number (197 Au: 8.1 MeV, 127 I: 9.1 MeV, 63 Cu: 10 MeV, 65 Cu: 9.9 MeV). Thus the energy difference increases with increasing the atomic number. This suggests that the percentage of high energy neutrons is high in heavy nuclei. Thus, the result that the strongest anisotropy is observed for the heaviest nucleus, 197 Au, can be understood in terms of the neutron separation energy.

The anisotropy for all the transitions originates from the contribution of the high energy neutrons as discussed above. This indicates that the M1/E1 mixing in heavy nuclei can be evaluated from a polarization asymmetry at $\theta = 90^{\circ}$ (or other angles) by gating with the highest neutron energy to select the transition to the ground state of the residual nucleus as the previous studies using $(\vec{\gamma}, p)$ reactions [19,20]. Ogata et al. show that the parameters, *a* and *b*, depend on the charge transition density for both $(\vec{\gamma}, n)$ and $(\vec{\gamma}, p)$ reactions [29]. With theoretical calculations for the charged transition density, we can study the detailed nuclear structure in the GDR region.

For such a purpose, the key point is the energy resolution of neutron measurements. In the present experiment, the energy resolution is finally determined by the energy width of the incident γ -ray beam. The typical energy spread of the photon beam is 3–10% in the present available LCS γ -ray facilities. With 17-MeV γ -rays, the neutron energy resolution is about 0.5–1.7 MeV. This suggests that we should select a target nucleus in which the energy of the first excited state is higher than 0.5 MeV (if the energy spread of the incident beam is 3%) or that we should measure the summation of some transitions to the ground state and low excited states. The progress in laser and accelerator physics enables us to realize the next generation of the LCS γ -ray sources including ELI-NP [30], MEGA-ray [31], and the ERL-LCS [32]. The extended plan of the HI γ S can be included in them [33]. The energy spread of these γ -ray beams is expected to be lower than $dE/E \sim 10^{-3}$. If we couple the present experimental method with a high energy resolution γ -beam, it is possible to study the detailed nuclear structure of the GDR with an excellent resolving power of the order of keV.

Finally we would like to point out that the present method is applicable in the energy region above the 2 neutron separation energy. Even when (γ, xn) $(x \ge 2)$ reaction channels open, the (γ, n) channel is also still open and we can selectively measure the 1n reaction channel by gating the maximum neutron energy, which corresponds to the transition from excited states near the highest populated level to the ground state of the residual nucleus.

In summary, we measured the neutron angular distribution at the polar angle $\theta = 90^{\circ}$ from (γ , n) reactions on three targets of Au, NaI, and Cu with a linearly polarized laser Compton scattering γ -ray beam at NewSUBARU. In 1957, Agodi predicted that angular distributions in (γ, n) and (γ, p) reactions with a 100% linearly polarized γ -ray beam should be anisotropic and described as a simple function of $a + b \cdot \cos(2\phi)$ at $\theta = 90^{\circ}$ from the conservation laws of angular momentum and parity. However, this prediction has not been experimentally confirmed in over half a century. The anisotropy may vanish in heavy nuclei because of their complex nuclear structures. However, the present experimental result clearly shows that the angular distributions are well described by the function $a + b \cdot \cos(2\phi)$ for the three nuclei. In addition, we found that the anisotropy originated from high energy neutrons, corresponding to the transition from highly excited levels to the ground state or a low excited state in the residual nucleus. In the near future, the next generation of the LCS γ -rays will be available. The lost Agodi's prediction will have a more precious role to study nuclear physics in the GDR region and applications using photonuclear reactions.

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