Photonuclear Reaction of Iodine-129 with Laser-Compton Scattering Gamma-Rays Using Nd:YVO₃ Laser and Electron Storage Ring

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A photonuclear reaction cross section of iodine-129 was measured using a polarized laser-Compton scattering gamma-ray beam in an energy range from 13.9 to 19.7 MeV. The maximum cross section was evaluated to be 220 mbarn ± 50 % at a photon energy of 15.9 MeV ± 4 %. We did not observe any appreciable difference in the cross sections for linearly and circularly polarized gamma-ray beams, considering the limits of experimental error.

Key Words: Polarized gamma-ray, Laser-Compton scattering, Iodine-129, Photonuclear reaction

1. Introduction

The radioactive wastes from nuclear power plants are supposed to be disposed in the Earth’s strata; iodine-129 is one such radioactive waste. Since it is a relatively small fraction of the total radioactive waste and has a very long half-life of 15.7 million years, its radioactivity should be small. However, the geological disposal of iodine-129 is anticipated to seriously affect the environment. Therefore, iodine-129 must be disposed of by nuclear transmutation.

We previously proposed the nuclear transmutation of radioactive nuclei including iodine-129 using a laser-Compton scattering (LCS) gamma-ray beam.¹ To evaluate the process efficiency, we must have an accurate photonuclear reaction cross section of iodine-129. J. Magill, et al.² measured an integral cross section of iodine-129 σₚ by laser-generated bremsstrahlung photons that had a Boltzmann-like distribution with a hot-electron temperature with random polarizations. They assumed a Lorentzian shape for σₚ with a calculated energy threshold of 8.8 MeV and Eₑₓₙ = 12.4 MeV, and obtained a cross section of σₚ(Eₑₓₙ) = 220 mbarn², where Eₑₓₙ is the photon energy which maximizes the σₚ and σₚ(Eₑₓₙ) is the σₚ at Eₑₓₙ. Unfortunately, their measurement was only accurate within a factor of two. In addition, the gamma-ray-polarization dependence of the cross section has not been investigated yet.

Presently, semi-monochromatic LCS gamma-ray beams of a few MeVs to several tens of MeVs with an intensity of 5 × 10⁸ photons sec⁻¹ are available and are steadily being utilized in a beamline of the NewSUBARU synchrotron radiation facility.³ LCS gamma-ray beams are well polarized on the beam axis using a well-polarized incident laser beam⁴.⁵

For this study, we carried out measurements at NewSUBARU to determine the dependence of the photonuclear reaction cross section of iodine-129 on the energy and polarization of incident gamma-ray photons.

2. Measurement of photonuclear reaction cross section

2.1 Experimental

Polarized gamma-rays with maximum energy of 16.7 MeV were generated in the storage ring by the interaction between a 1.064-µm CW laser beam of 4.6 W and a 974 MeV electron beam of 220 mA.⁶ (Fig. 1). The laser polarization was rotated using a combination of λ/4 and λ/2 wave plates in a laser oscillator. The polarized gamma-rays were semi-monochromatized in the 3 % energy spread using a 3-mm-diameter lead collimator. The flux of the gamma-rays incident on the target was measured using a 10-mm-thick plastic scintillator that was installed in front of the target. The scintillator was attached with a 1-mm-thick piece of lead as a signal multiplier on the beam incident side. The flux of the polarized and semi-monochromatized gamma-rays was measured to be 5 × 10⁸ photons sec⁻¹. The polarization of the gamma-rays was

Vol.39, No.6 Photonuclear Reaction of Iodine-129 with Laser-Compton Scattering Gamma-Rays Using Nd:YVO₃ Laser and Electron Storage Ring 445

Fig. 1 Schematic view of NewSUBARU-BL01 including a laser source (1), an interaction region (2), a pair of mirrors for alignments (3), and a gamma-ray beam generated (4).
measured\textsuperscript{11} and confirmed to be controllable by the polarization of the laser beam. Iodine-129 was prepared as a powder of palladous iodide (6 g cm\textsuperscript{-3}). The radioactivity of the target was calibrated to be 36.8 kBq by Isotope Products Laboratories\textsuperscript{5}. The powder of the palladous iodide was sealed in a cone cavity of acrylic whose base and height were 5.15 mm\textsuperscript{2} and 0.77 mm with an apex angle of 118°. The number of iodine-129 atoms in the target was calculated to be 2.63 × 10\textsuperscript{10} from its radioactivity of 36.8 kBq and the half-life of iodine-129 $\tau_{1/2} = 1.57 \times 10\textsuperscript{7}$ year.\textsuperscript{11}

The number of iodine-128 atoms $N(t)$ is derived as a function of time from the following equation:

$$\frac{d}{dt} N(t) = Y - \lambda N(t),$$

(1)

where $Y$ is the yield rate of iodine-128, $\lambda = ln 2/\tau_{1/2} = 0.028$ min\textsuperscript{-1} is the decay rate coefficient, which is constant, and $\tau_{1/2} = 25$ min is the half-life of iodine-128.\textsuperscript{11} By solving Eq. (1) with an initial condition of $N(0) = 0$, $N(t)$ is expressed as follows:

$$N(t) = \frac{Y}{\lambda} \left(1 - e^{-\lambda t}\right).$$

(2)

We found that as the irradiation period increases, $N(t)$ approaches $Y\lambda$, and has a value of 0.5 $\times$ $Y\lambda$ at a irradiation period of $\tau_{1/2}$, but its value increases to 0.996 $\times$ $Y\lambda$ when the irradiation time increases to 8 $\times$ $\tau_{1/2}$. Therefore, the irradiation period of gamma-rays $T$ was chosen as $\tau_{1/2} = 25$ min, giving accuracy as sufficient as that by $8 \times \tau_{1/2}$.

After the irradiation of the gamma-rays, the number of decayed gamma-rays with an energy of 443 keV, which were emitted from the radioactive iodine-128, was measured using a Ge detector in a typical measurement period of 50 min. The signal level in the Ge detector due to the 443-keV photons from the radioactive iodine-128 was about 70 cwt with irradiation of 16.7-MeV gamma-rays. The peak detection efficiency of the Ge detector used in this experiment was calibrated to be 5% using the 384-keV gamma-ray photons emitted by barium-133. The probability of one iodine-128 atom emitting a 443-keV photon in a spontaneous decay process was considered to be 12.6%\textsuperscript{11}; thus, the number of decayed iodine-128 atoms $N_T$ was calculated to be 1.1 $\times$ 10\textsuperscript{11}.

The number of iodine-128 atoms $N_i = N(T)$ was calculated to be 1.5 $\times$ 10\textsuperscript{11} assuming $T = 50$ min, using the following equation:

$$N_i = \frac{N_T}{1 - e^{-\lambda T}},$$

(3)

where $T$ is a time period for measuring the 443-keV photons.

2.2 Derivation of reaction cross section

The photonuclear reaction cross section of iodine-129 $\sigma_i$ was evaluated as follows using $N_i$ [Eq. (3)]. The variation of the yield rate of iodine-128 $dY$ for distance $dx$ along the direction of the LCS gamma-ray beam is expressed as follows:

$$dY = n \cdot f(x) \cdot \sigma_i \cdot dx \cdot ds,$$

(4)

where $n$ is the atomic density of iodine-129 and $f(x)$ is the flux of the gamma-ray photons per unit area at position $x$. The atomic density was calculated to be 7.4 $\times$ 10\textsuperscript{17} mm\textsuperscript{-3} from the size of the target. The 3-mm-diameter collimator passes an axial portion of the incident gamma-ray beam so that the spatial distribution of the transmitted beam with a 3-mm diameter is uniform, and the target has rotational symmetry whose axis is coaxial with the beam axis. $f(x)$ is obtained as follows:

$$f(x) = f(0) e^{-\phi x},$$

(5)

where $\phi$ is the cross section for all processes except the photonuclear reaction and $f(0)$ is the value of the incident flux at the target entrance. $f(0)$ was measured as $f(0) = 4.7 \times 10\textsuperscript{7}$ photon mm\textsuperscript{-2} sec\textsuperscript{-1} $\pm$ 2%. By evaluating Eq. (4), $Y$ is described as follows:

$$Y = 2\pi \cdot n \cdot \sigma_i \cdot f(0) \int_0^x \int_0^{\theta(x)} e^{-\phi \rho(x) \sigma_i \rho(x)} dx \cdot dr,$$

(6)

where $R$ is the radius of the target, $L(r)$ is the penetration distance of the incident gamma-rays into the target, which is a function of radius $r$. From Eqs. (2) and (6), $N_i$ is expressed as:

$$N_i = \frac{Y}{\lambda} \left(1 - e^{-\lambda T}\right) \cdot 2\pi \cdot n \cdot \sigma_i \cdot f(0) \int_0^x \int_0^{\theta(x)} e^{-\phi \rho(x) \sigma_i \rho(x)} dx \cdot dr.$$

(7)

2.3 Dependence of reaction cross section on polarization and energy of incident gamma-rays

Figure 2 shows photonuclear reaction cross section $\sigma_i$ derived from Eq. (7) by assuming the value of $\sigma_i$ proposed by J. Berger, et al.\textsuperscript{12} in an energy range from 13.9 to 19.7 MeV for both linear (crossed) and circular (circled) polarizations of incident gamma-rays. The gamma-ray photon energy was scanned by changing the energy of the electron beam from 890 to 1060 MeV.

The probability distribution of the signal intensity against the count rate was assumed to be a Poisson distribution. From its standard deviation, the error bars in Fig. 2 were calculated to be 12% at respective gamma-ray energy. The error in the absolute value of the cross section was evaluated to be ±50% by taking into account the errors due to the alignment accuracy between the gamma-ray beam and the target axes; this accuracy was a major factor compared

\textsuperscript{11} J. K. Tuli: "Nuclear Decay Data in the MIRD Format," <http://www.nndc.bnl.gov/mird/> (2010).

\textsuperscript{12} M. J. Berger, J. H. Hubbell, S. M. Seltzer, J. Chang, J. S. Coursey, R. Sukumar, D. S. Zucker, and K. Olsen: "XCOM: Photon Cross Sections Database," <http://www.nist.gov/pml/data/xcom/index.cfm> (2010).
with the errors in the number of iodine-129 atoms (< ± 1 %),
the flux of the gamma-rays (± 2 %), their energy spread
(− 3 %), and the peak detection efficiency of the Ge detector
(± 10 %).

An appreciable difference in the cross sections determined
for linear and circular polarizations was not observed because
the data of both polarizations remained within the dispersion
of the measurements (Fig. 2).

Therefore, all data of the cross section of the iodine-129
target were treated as a set and fitted by a Lorentzian curve to
derive the dependence of the cross section on the photon energy.\(^6\)
From the above evaluation, the energy dependence of
the cross section was determined by a fitting curve with a
maximum value of 220 mbarn ± 50 % at 15.9 MeV ± 4 %
with a half-width at half-maximum of 2.55 MeV ± 20 %. In
this measurement the accuracy of the cross section obtained
was improved compared with that of the data by J. Magill, et
al.\(^5\) because the polarized gamma-rays were semi-monochro-
matized in the 3 % energy spread so that the cross section was
directly obtained for each energy point of the gamma-ray pho-
ton. The photon energy dependence of the iodine-129 \(\sigma_s\)
was obtained experimentally by scanning the energy of the semi-
monochromatized gamma-ray photons.

These values are in good agreement with the data of
iodine-127 by R. L. Bramblett, et al.\(^6\) who used monoenergetic
photons with a photon energy resolution of approximately 170
keV. They obtained a maximum value of 231 mbarn at 15.2
MeV.

The \((\gamma, n)\) reaction is caused by the giant resonance which
is due to the gamma-ray photon field in nucleus. No apprecia-
table difference observed in the cross sections determined for
linear and circular polarizations suggests that the helicity of
the photon due to the polarization does not affect the photon reaction
by the giant resonance. The theoretical analysis to
explain the above result will be done in further study.

3. Conclusions

We determined the maximum photonuclear reaction cross
section of iodine-129 to be 220 mbarn ± 50 % at a photon en-
ergy of 15.9 MeV ± 4 % with a half-width at half-maximum of
2.55 MeV ± 20 %. We did not observe any appreciable dif-
ference in the cross sections determined for linearly and cir-
cularly polarized gamma-ray beams considering the limits of ex-
perimental error.

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