Experimental Study of Photonuclear Transmutation Driven by Laser-Wakefield Acceleration

Authors: Mr. Yongsheng Xu, Wu, Dr. Di, He, Dr. Chuangye, Cheng, Dr. Hao, Ye, Dr. Shan, Dong, Mr. Bing-Kun, Lu, Mr. Mu-Yang, Liu, Dr. Fu-Long, Could you provide the full Chinese text you' d like translated, wrapped in . . . tags if possible?
For example: 兰浩洋博士

If you only want the name rendered in academic English, a standard form would be:

Lan, Haoyang, Ph.D.
or
Dr. Haoyang Lan, Wang, Dr. Meizhi, Xia, Prof. Yuhui, Prof. Zhenan Wang, Prof. Xinlu Xu, Prof. Xueqing Yan, Dr. Bing Guo, He, Dr. Chuangye

Date: 2026-01-09T20:18:07+00:00

Abstract

The sustainable development of nuclear energy depends greatly on the proper disposal of spent nuclear fuel (SNF). Fast reactors (FRs) and accelerator-driven systems (ADS), based on neutron-induced reactions, show high efficiency in transmuting most actinides and long-lived fission products (LLFPs) in SNF. For a small set of nuclides with relatively low neutron-capture cross-sections, such as 93Zr and 126Sn, complementary strategies are needed. Also, under neutron-induced reactions, successive neutron captures could potentially change stable nuclides into long-lived radioactive ones (e.g., 133Cs to 135Cs). To broaden nuclide transmutation scope and boost overall efficiency, photonuclear transmutation was proposed as a supplement to neutron capture. It uses high-flux γ rays in the giant dipole resonance (GDR) energy region to start photonuclear reactions, converting long-lived nuclides into short-lived or stable ones. In this research, laser-wakefield acceleration (LWFA)-driven photonuclear transmutation was experimentally carried out at Peking University' s Compact Laser Plasma Accelerator (CLAPA). Laser pulses with an intensity of 1019 W/cm2 generated 200 MeV electron beams. These electrons impinge on a Ta converter to produce broadband bremsstrahlung γ rays, which irradiated a CsI target with 133Cs and 127I as LLFP surrogates. Offline activation spectra measured by a high-purity germanium (HPGe) detector showed clear characteristic lines of 132Cs, 126I, and 124I. Combining activation-spectrum analysis with Geant4 simulations, the total transmutation yield normalized to incident laser pulse energy was estimated at 1.35 × 105 J−1. This performance is better than that of laser-induced bremsstrahlung and nuclear-resonance γ-ray sources, and is comparable to electron linac bremsstrahlung. With further setup optimization and laser technology progress, especially in pulse energy and repetition rate, the LWFA-driven γ source is expected to further improve its transmutation ability and serve as an effective complement to FRs and ADS.

Full Text

Preamble

Experimental Study of Photonuclear Transmutation Driven by Laser-Wakefield Acceleration

Yong-Sheng Xu,[1] Di Wu,[2],[3],† Chuang-Ye He,[1],‡ Hao Cheng,[1] Shan Ye,[1] Bing-Kun Dong,[1] Mu-Yang Lu,[1] Fu-Long Liu,[1] Hao-Yang Lan,[2],[3] Mei-Zhi Wang,[2],[3] Yu-Hui Xia,[2],[3] Zhe-Nan Wang,[2],[3] Xin-Lu Xu,[2],[3] Xue-Qing Yan,[2],[3] and Bing Guo[1],§

[1]Institute of Nuclear Physics, China Institute of Atomic Energy, Beijing 102413, China
[2]State Key Laboratory of Nuclear Physics and Technology, School of Physics, CAPT, Peking University, Beijing 100871, China
[3]Beijing Laser Acceleration Innovation Center, Beijing 101407, China

The sustainable development of nuclear energy depends greatly on the proper disposal of spent nuclear fuel (SNF). Fast reactors (FRs) and accelerator-driven systems (ADS), based on neutron-induced reactions, show high efficiency in transmuting most actinides and long-lived fission products (LLFPs) in SNF. For a small set of nuclides with relatively low neutron-capture cross-sections, such as ^{93}Zr and ^{126}Sn, complementary strategies are needed. Also, under neutron-induced reactions, successive neutron captures could potentially change stable nuclides into long-lived radioactive ones (e.g., ^{133}Cs to ^{135}Cs). To broaden nuclide transmutation scope and boost overall efficiency, photonuclear transmutation was proposed as a supplement to neutron capture. It uses high-flux γ rays in the giant dipole resonance (GDR) energy region to start photonuclear reactions, converting long-lived nuclides into short-lived or stable ones.

In this research, laser-wakefield acceleration (LWFA)-driven photonuclear transmutation was experimentally carried out at Peking University' s Compact Laser Plasma Accelerator (CLAPA). Laser pulses with an intensity of 10^{19} W/cm^2 generated 200 MeV electron beams. These electrons impinge on a Ta converter to produce broadband bremsstrahlung γ rays, which irradiated a CsI target with ^{133}Cs and ^{127}I as LLFP surrogates. Offline activation spectra measured by a high-purity germanium (HPGe) detector showed clear characteristic lines

of ^{132}Cs, ^{126}I, and ^{124}I. Combining activation-spectrum analysis with Geant4 simulations, the total transmutation yield normalized to incident laser pulse energy was estimated at 1.35×10^5 J^{-1}. This performance is better than that of laser-induced bremsstrahlung and nuclear-resonance γ-ray sources, and is comparable to electron linac bremsstrahlung. With further setup optimization and laser technology progress, especially in pulse energy and repetition rate, the LWFA-driven γ source is expected to further improve its transmutation ability and serve as an effective complement to FRs and ADS.

Keywords: LWFA, photonuclear transmutation, LLFP, CsI, γ rays

Introduction

Nuclear power plants generate virtually no greenhouse gases or air pollutants during their operation. As of today and for decades to come, the potential of nuclear energy is crucial for achieving a deeply decarbonized energy future in many regions of the world. A challenging task in the widespread utilization of nuclear energy is the management of radioactive spent nuclear fuel (SNF). Minor actinides, including americium, curium, and neptunium, constitute a small fraction of SNF; however, they account for the majority of the long-term decay heat and radiotoxicity of high-level waste (HLW) packages. SNF also contains long-lived fission products (LLFPs), such as Cs, Sr, Tc, and I [?]. Partitioning and transmutation (P&T) is a promising strategy for treating SNF, aiming to reduce its waste volume and toxicity. Transmutation modifies the properties of the waste by converting long-lived radioactive elements into shorter-lived or non-radioactive elements through nuclear reactions in fast reactors (FRs) [?] or accelerator-driven subsystems (ADS) [?]. FRs and ADS can effectively transmute the vast majority of HLW constituents, primarily actinides and LLFPs, via neutron-induced fission or capture reactions. There still remain a small number of HLW constituents that need to be transmuted through other pathways, such as ^{93}Zr and ^{126}Sn \cite{2-5}, because they have relatively small neutron-capture cross sections. Additionally, in some cases, stable nuclides can be transformed into long-lived radioactive nuclides through successive neutron captures. For example, ^{133}Cs can be transmuted into the long-lived nuclide ^{135}Cs [?, ?].

Photonuclear transmutation, proposed as a complement [?] to neutron capture, employs γ-rays in the giant-dipole-resonance (GDR) energy region to convert LLFPs into short-lived or stable nuclides. The transmutation yield is mainly governed by the γ-ray flux and the photonuclear reaction cross section. Transmutation experiments have been conducted for ^{129}I \cite{6-10}, as well as for the surrogate nuclides such as ^{127}I [?], ^{133}Cs [?] and ^{197}Au [?, ?], from which the corresponding yields or reaction cross sections have been obtained. Meanwhile, particle-in-cell (PIC) and Geant4 simulations have been performed for a broad set of LLFPs, including ^{93}Zr [?], ^{107}Pd [?], ^{126}Sn [?, ?], ^{129}I [?, ?], and ^{135}Cs \cite{19,21-24}, as well as ^{90}Sr [?] and ^{137}Cs [?, ?]. These simulations examined how the transmutation yields depend on the laser conditions, the energies and fluxes of the electrons and γ rays, and the target geometries. The results provide guidance that is valuable for optimizing the experimental design.

Based on the experimental and simulation studies above, photonuclear transmutation methods can be classified by the type of γ-ray source into three main categories: first, accelerator-driven, such as electron linac-bremsstrahlung (Linac-BR) [?, ?] and nuclear resonance reaction γ rays (NRγ) [?, ?]; second, accelerator-laser-driven, such as laser Compton scattering (LCS) [?, ?]; and third, purely laser-driven, including laser-induced bremsstrahlung (LIB) using solid targets [?, ?], as well as laser-gas interactions involving direct-laser acceleration (DLA) [?] and laser-wakefield acceleration (LWFA) [?]. In the LWFA mechanism [?], an ultraintense laser pulse propagating through underdense plasma excites a wakefield that traps and accelerates electrons to MeV–GeV energies within mm-cm-scale distances. These high-energy electrons subsequently strike a high-Z converter to generate bremsstrahlung γ rays with high flux and broad spectra, making LWFA a promising way for photonuclear transmutation.

Following these developments, LWFA-driven photonuclear reactions have been investigated in both simulations and experiments. In terms of simulations, Y. C. Wang et al. [?] and X. L. Wang et al. [?] employed PIC and Geant4 to assess the feasibility of LWFA-driven photonuclear transmutation of LLFPs such as ^{135}Cs and ^{126}Sn. Their works provide quantitative parameter references for the design of such experiments. Experimentally, LWFA-driven bremsstrahlung γ rays have been applied to photonuclear reactions. Reed et al. [?, ?] measured photofission in ^{238}U and photoneutron reactions on ^{12}C and ^{63}Cu, reporting reaction yields, while D. Wu et al. \cite{30-32} carried out flux-weighted cross-section measurements. Despite experimental advances, studies focusing on LWFA-driven photonuclear transmutation of LLFP remain scarce, highlighting the need to assess the transmutation performance of the LWFA approach. It is also valuable to compare the LLFP transmutation capability of LWFA with that of other γ-ray sources such as LIB [?], since LWFA can deliver higher-energy and better-collimated bremsstrahlung γ rays.

Therefore, we performed such an experiment at the Compact Laser Plasma Accelerator (CLAPA) facility of Peking University. In this work, a CsI crystal containing the stable nuclides ^{133}Cs and ^{127}I was employed as a surrogate target. This choice was based on the following reasons: Cs and I are typical constituents of LLFPs; direct preparation of targets using LLFPs poses radiological safety risks; in the GDR region, the photonuclear reaction cross sections of ^{133}Cs and ^{127}I are 10^2 mb, comparable to those of LLFPs (IAEA/PD-2019 [?]); the (γ,n) products ^{132}Cs and ^{126}I have intermediate half-lives and emit high branching-ratio, distinct γ rays, making them easy to detect and identify.

The remainder of this paper is organized as follows: Section II describes the experimental setup, Section III presents and discusses the results, Section IV evaluates the transmutation performance, and Section V provides the conclusions and outlook.

II. Experiment

A. Experimental Setup

The CLAPA experimental setup [Figure 1: see original paper] comprises four subsystems: a LWFA electron source, an electron-beam diagnostics station, a bremsstrahlung conversion and photonuclear transmutation target area, and an offline γ-spectroscopy apparatus. Femtosecond laser pulses (200 TW, 800 nm, 0.2 Hz, 30 fs, 2.4 J, 10^{19} W/cm^2) are focused by a lens into a 29 bar helium gas jet to generate an underdense plasma. Real-time plasma density diagnostic is obtained using probe-beam interferometry. In the laser-gas interaction region, electrons are accelerated to several hundred MeV by the LWFA mechanism and then traverse a 40 µm Al foil before exiting the chamber via a Be window [FIGURE:1(a)].

The per-shot electron charge is measured using an integrating current transformer (ICT). The electron beam is dispersed by a 0.8 T dipole magnet according to its energy and imaged on scintillating screens, which are recorded by CCD cameras, enabling simultaneous diagnostics of the beam' s spatial profile and energy distribution [FIGURE:1(b)]. Following the electron acceleration stage, a 2 mm-thick ^{181}Ta converter is installed at the entrance of the magnet, located 1 m downstream of the gas jet. High-energy electrons impinging on the converter generate bremsstrahlung γ rays, and their flux is monitored using activation reactions in Cu and Al [**?**, **?**]. These bremsstrahlung γ rays then irradiate a 5 mm-thick CsI target positioned 33 cm downstream of the converter [FIGURE:1(c)], inducing (γ, n) reactions and producing radioactive daughter nuclides such as ^{126}I and ^{132}Cs.

After 20 minutes of γ irradiation, the CsI target is placed offline in a low-background chamber equipped with an HPGe detector shielded by 10 cm of lead [FIGURE:1(d)]. The measurement starts after a cooling time of 1.5 h and continues for 24.5 h, during which the CsI γ-decay characteristic spectrum is recorded to identify the transmutation products.

The transmutation yields depend on the geometric dimensions of both the converter and the CsI target. Therefore, a transverse size of 20\$×20*mm*^{2}\$ is adopted for both targets, taking into account the electron-beam divergence and their relative positions to ensure full coverage of the irradiation area. Their thicknesses are optimized independently. For the converter, high-Z metals are efficient bremsstrahlung emitters [**?**], and Ta is commonly employed in similar setups due to its favorable γ-ray yield, thermomechanical properties, and machinability. According to the Particle Data Group [**?**], the radiation length of Ta is $X_0 = 0.4094$ cm, which is the mean distance for a high-energy electron to lose all but 1/e of its energy via bremsstrahlung. A converter that is significantly thinner than the radiation length leads to incomplete electron-to-photon energy conversion, whereas an excessively thick converter results in pronounced self-absorption of the emitted γ rays [**?**]. At CLAPA, a 2 mm Ta converter (0.5 X_0) is adopted as a practical compromise \cite{30–32,37}.

For the CsI target, a thickness of 5 mm is selected by balancing the daughter-nuclide yield and the escape efficiency of the characteristic γ rays from them. Using the 388.6 keV line of the dominant (γ,n) product ^{126}I as the reference energy, the mass attenuation coefficient of CsI is interpolated from NIST-XCOM [?] to be (μ/ρ) 0.129 cm^2 g^{-1}. This value corresponds to a linear attenuation coefficient of μ 0.58 cm^{-1} at a density of 4.51 g cm^{-3}. According to the uniform-volume self-absorption expression $R_0 = R\ \mu t/(1-e^{-}\mu\ )$ [?], where R_0 and R denote the emitted and detected γ-ray rates, respectively, a target thickness of $t = 5$ mm corresponds to a self-absorption loss of about 13%, which keeps the characteristic γ-ray signals strong enough for HPGe measurements.

B. Detector Calibration

The characteristic γ rays from CsI transmutation products lie primarily in the range of a few hundred keV to about 1 MeV. Standard γ rays from ^{152}Eu, ^{137}Cs, and ^{60}Co, which span this energy region, are used for HPGe efficiency calibration. The measured efficiencies are empirically fitted with a double-exponential function, giving a coefficient of determination of $R^2 > 0.98$ [Figure 2: see original paper]. From this fit, the efficiencies at 388.6 keV, 667.7 keV, and 602.7 keV are determined to be 4.46(2)%, 1.78(8)%, and 1.95(6)%, respectively, and these values are subsequently applied in the yield calculations. For energy calibration of the offline HPGe measurements, the natural background γ lines of ^{40}K (1460.8 keV) and ^{208}Tl (2614.5 keV) provide two reference points for determining the linear energy scale of the irradiated CsI spectra.

C. Geant4 Simulation

Geant4 [?] is used to simulate the bremsstrahlung generation and the corresponding transmutation yields, with the electrons in PrimaryGeneratorAction sampled from the measured LWFA energy spectrum and angular distribution. The simulation apparatus geometry reproduces the experimental layout. The physics list employs FTFP_{BERT} with G4eBremsstrahlung in the converter and G4PhotoNuclearProcess in the CsI. Radioactive decays follow the built-in data, including branching ratios.

Irradiation of the Ta converter by the incident electrons generates bremsstrahlung γ rays, which then illuminate the CsI target and induce photonuclear reactions, forming the activation products that make up the end-of-irradiation (EOI) inventory. In the simulation, the activation products undergo post-irradiation decay under the same cooling and measurement times as those used experimentally, emitting the characteristic γ rays of the daughter nuclides. When treating these emitted γ rays, the simulated spectrum is obtained by considering the HPGe efficiency [Figure 2: see original paper] and the energy-dependent transmittance of CsI. The simulated yields are then extracted from this spectrum using the same analysis chain as in the measurement.

The γ-ray transmittance is obtained from a separate Geant4 transport simulation that uses a spatially uniform and isotropic γ source implemented by emitting a sufficiently large number of photons at selected energies throughout the CsI volume. A virtual spherical detector enclosing the target records the detected photons, and the ratio of detected to emitted photons as a function of energy directly represents the γ-ray transmittance used in the yield calculations.

III. Results and Discussion

A. LWFA Electrons

In LWFA, prior studies indicate that the mean electron energy increases as the gas jet pressure decreases [**?**, **?**, **?**], so a 29 bar helium jet was used in this experiment to obtain higher electron energies. At this pressure, the measured spectrum shows a quasi-monoenergetic peak at (200 ± 20) MeV with a high-energy tail extending to 290 MeV [FIGURE:3(a)]. The beam has a transverse spot of 10 mm and a 5 mrad half-angle divergence measured 1 m downstream from the source [FIGURE:3(b)]. The ICT-measured charge showed a shot-to-shot jitter of about 30%, which would introduce noticeable uncertainty into the quantitative evaluation of transmutation yields if only a single shot were used. To reduce this effect, 240 shots (0.2 Hz, 20 min) were accumulated, giving an average charge of 300 pC per shot and reducing its uncertainty to a few percent, thereby improving the statistical precision of the resulting yields.

B. Bremsstrahlung γ-rays

After measuring the LWFA electron beam, the Geant4 simulation was performed using the electron source and a 2 mm Ta converter to produce the corresponding bremsstrahlung spectrum. As seen in Fig. 3 Figure 3: see original paper, the photon counts per MeV decrease monotonically with photon energy and extend to about 280 MeV. Within the GDR region (9–30 MeV), it reaches the order of 10^7 MeV^{-1}, corresponding to more than 10^8 photons above 9 MeV. Taking the 2 mm Ta thickness as the effective emission length gives a γ-burst duration of 6.7 ps, implying an instantaneous flux exceeding 10^{19} s^{-1} on the CsI target. Figure 3(d) displays the angular distribution of the bremsstrahlung photons, which is strongly forward-peaked, with most of the photons confined within 0.1 rad.

C. Transmutation Products

With the experimental setup and parameters described above, activation γ-ray spectra of the irradiated CsI target and the natural background were measured offline with the HPGe detector [FIGURE:4(a), main portion], where the nuclide assignments of the characteristic γ lines are indicated. The background spectrum arises mainly from naturally occurring radionuclides ^{40}K, ^{208}Tl, ^{214}Pb, ^{214}Bi, and ^{228}Ac. Relative to the background, the CsI activation spectrum exhibits γ lines attributable to the daughter nuclides ^{126}I, ^{124}I and ^{132}Cs. Their decay schemes are shown in [FIGURE:4(b)-4(d)], where the transition lines labeled with energies and branching ratios represent the measured γ lines; all nuclear data are taken from the National Nuclear Data Center (NNDC) [?].

Transmutation of Iodine. ^{127}I(γ, n)^{126}I. The reaction product ^{126}I ($T_1/_2$ = 12.93 d) decays via two branches, β^- to ^{126}Xe and ϵ to ^{126}Te, as shown in [FIGURE:4(b)]. For the β^- branch, the decay scheme on the left of [FIGURE:4(b)] shows two γ-ray transitions at 388.6 and 491.2 keV with γ branching ratios of 35.6% and 2.88%, respectively. In [FIGURE:4(a), the 388.6 keV line is well resolved and appears as an isolated line without contamination, while the 491.2 keV line, despite its low branching ratio, is still observed in the spectrum. For the ϵ branch, the decay scheme on the right of [FIGURE:4(b)] identifies two γ-ray transitions at 666.3 and 753.8 keV, with γ branching ratios of 32.9% and 4.15%, respectively. In [FIGURE:4(b), the 666.3 keV transition, although its γ branching ratio (32.9%) is comparable to that (35.6%) of the 388.6 keV line, appears with higher intensity because it overlaps with the 667.7 keV line of ^{132}Cs, forming a composite peak in the spectrum. The 753.8 keV transition, though it has a smaller branching ratio, is clearly visible above the background. Therefore, the agreement between the decay scheme in [FIGURE:4(b) and the four γ-ray lines in the spectrum confirms the presence of ^{126}I.

^{127}I(γ, 3n)^{124}I. In [FIGURE:4(a), the 602.7 keV peak shows slight tail overlap from the 609.3 keV ^{214}Bi line but remains clearly resolved. In addition, the decay scheme in [FIGURE:4(c)] indicates that ^{124}I ($T_1/_2$ = 4.176 d) decays entirely to ^{124}Te by ϵ branch and that the 602.7 keV transition is dominant (62.9%). These observations confirm the presence of ^{124}I.

Transmutation of Cesium. ^{133}Cs(γ, n)^{132}Cs. The reaction product ^{132}Cs ($T_1/_2$ = 6.48 d) decays predominantly by ϵ (98.13%) to ^{132}Xe, while a minor β^- branch (1.87%) leads to ^{132}Ba, as shown in [FIGURE:4(d)]. For the ϵ branch, the right part of [FIGURE:4(d)] shows the dominant 667.7 keV transition with a γ branching ratio of 97.59%. For this reason, the 667.7 keV line in [FIGURE:4(a)] appears very strong, with some contribution from 666.3 keV of ^{126}I. The 630.2 keV line is also observed, though with only a 0.95% γ branching ratio. For the β^- branch, the left part of [FIGURE:4(d)] shows the 464.5 keV transition with a γ branching ratio of 1.58%, which is visible in [FIGURE:4(a)] but is merged with the 463.0 keV ^{228}Ac peak. The three lines at 667.7, 630.2, and 464.5 keV, together with the decay scheme in [FIGURE:4(d)], indicate the presence of ^{132}Cs in the irradiated CsI target.

Overall, the consistency between the measured γ peaks and the NNDC decay schemes confirms the identification of ^{126}I, ^{124}I, and ^{132}Cs, which are used for subsequent yield evaluation. Notably, although the production of ^{124}I via the ^{127}I(γ, 3n) channel confirms that the energies of the incident γ rays exceed the multi-neutron emission thresholds, nuclides such as ^{125}I, ^{131}Cs, and ^{130}Cs are not observed. Their absence is attributable to the strong self-absorption of decay X-rays below 40 keV accompanying ^{125}I and ^{131}Cs, and to the short half-life of ^{130}Cs (29.21 min), which caused substantial decay before the offline counting.

D. Transmutation Yields

Simulated Yields. Efficient photonuclear transmutation requires that a high γ-ray flux coincide with the large photonuclear reaction cross section in the GDR energy region. The transmutation yield is accordingly expressed as

$$Y = N_t \int_{E_{th}}^{E_{max}} \Phi(E_\gamma)\sigma(E_\gamma)dE_\gamma,$$

where N_t is the number of target nuclei, E_{th} is the reaction threshold, and E_{max} is the upper limit of the GDR energy region. The quantity $\Phi(E_\gamma)$ denotes the photon flux density and $\sigma(E_\gamma)$ is the photoneutron reaction cross section.

[Figure 5: see original paper] shows the photon flux and the photonuclear reaction cross sections of ^{133}Cs and ^{127}I. They are from the Geant4 bremsstrahlung γ-ray simulation and from the TENDL-2023 database [**?**], respectively. The threshold energies for the (γ, n) reactions are 8.99 MeV for ^{133}Cs and 9.14 MeV for ^{127}I, and this channel dominates in the 9-25 MeV energy region. Within this region, the peak cross sections of ^{133}Cs and ^{127}I are of comparable magnitude, so their (γ, n) products ^{132}Cs and ^{126}I are expected to be obtained in similar amounts. At higher energies (25-30 MeV), the threshold for the (γ, 3n) reaction of ^{127}I is 25.83 MeV, and its cross section reaches only about 10% of the (γ, n) peak, so the yield of ^{124}I is anticipated to be the lowest among the three products.

In the simulation of transmutation inside the CsI target, Geant4 records all residual nuclei in an inventory. The results show that transmutation is dominated by the (γ, n) channel, producing ^{132}Cs and ^{126}I, which together account for about 73% of all products. The (γ, 2n) and (γ, 3n) channels contribute 11% and 4%, respectively, generating nuclides such as ^{131}Cs, ^{125}I, ^{130}Cs, and ^{124}I. Beyond the reaction channels discussed above, knock-out and spallation reactions also occur in the CsI target, induced by the high-energy bremsstrahlung tail extending up to 280 MeV. The reaction products, including light ejectiles (H, He) and heavier residues (Sn, Te, Xe, etc.), constitute the remaining 12% of the inventory.

The inventory is then processed by Geant4 to generate the simulated activation spectrum, from which the transmutation yields are obtained. As listed in , the simulated per-shot yields are 1.18 $\times$ 10^5 for ^{126}I, 1.94 $\times$ 10^5 for ^{132}Cs, and 9.57 $\times$ 10^2 for ^{124}I, and their sum amounts to 3.13 $\times$ 10^5 nuclei per shot.

Experimental Yields. The experimental transmutation yields are determined from the characteristic γ-ray signals in the HPGe spectra together with the relevant factors. For a transition with energy E, the EOI yield Y_E, corresponding to the total yield accumulated over the 240 laser shots, is calculated as

$$Y_E = \frac{C_E}{I_\gamma(E)\varepsilon_E\eta_E e^{-\lambda t_c}(1-e^{-\lambda t_m})},$$

where C_E is the net peak count, $I_\gamma(E)$ is the branching ratio of the transition, ε_E is the transmittance of the characteristic γ ray through the CsI target, η_E is the detection efficiency of the HPGe detector, and λ is the decay constant of the product nuclide. The exponential terms provide the decay corrections for the cooling time t_c and the measurement time t_m.

For ^{126}I, the counts $C_{388.6}$ are obtained directly from the 388.6 keV line. For ^{132}Cs, the counts $C_{667.7}$ are derived from the 667.7 keV transition. However, this line is blended with the 666.3 keV from ^{126}I. To extract $C_{667.7}$, the 666.3 keV contribution should be subtracted from the total blended-peak counts, $C_{total} = 8.09(4) \times 10^4$. The 666.3 keV contribution is obtained from

$$C_{666.3} = C_{388.6}\frac{I_\gamma(666.3)}{I_\gamma(388.6)}\frac{\eta_{666.3}}{\eta_{388.6}}\frac{\varepsilon_{666.3}}{\varepsilon_{388.6}}.$$

Thus, the net 667.7 keV counts are $C_{667.7} = C_{total} - C_{666.3}$. For ^{124}I, the counts $C_{602.7}$ are determined from the 602.7 keV line.

From the yield equation, together with the above counting results and the relevant parameters in , which include the γ-ray branching ratios and decay constants taken from the NNDC database, the γ-ray transmittance derived from an independent Geant4 transport simulation, and the HPGe detection efficiency obtained from the calibration fit, the average experimental per-shot yields of the three nuclides are 1.21(4) × 10^5 for ^{126}I, 1.99(10) × 10^5 for ^{132}Cs, and 3.15(36) × 10^3 for ^{124}I, which sum to 3.23(11) × 10^5 nuclei per shot.

As summarized in , the yield uncertainties arise from four main sources: counting statistics, detector efficiency, transmittance, and γ-ray branching ratios. In addition, the per-shot charge fluctuation contributes a uniform 1.9% systematic uncertainty to all three nuclides. For ^{126}I (388.6 keV), the relative uncertainty is about 3.3%, and it is mainly determined by the branching-ratio and charge uncertainties. For ^{132}Cs (667.7 keV), the combined uncertainty is roughly 5.0%, dominated by the HPGe efficiency and the charge contribution; the additional uncertainty associated with decomposing the 666.3/667.7 keV blended peak is negligible because the ^{126}I contribution at 666.3 keV is small. For ^{124}I (602.7 keV), the relative uncertainty reaches about 11.4% and is dominated by counting statistics due to the low net peak counts, while the charge uncertainty is relatively small.

Comparing the experimental per-shot yields with the Geant4 predictions, the differences amount to 2.5% for both ^{126}I and ^{132}Cs and to 69.6% for ^{124}I. For the (γ, n) products ^{132}Cs and ^{126}I, this small deviation indicates good agreement between experiment and simulation. For the (γ, 3n) product ^{124}I, the large deviation reflects the low statistics of its 602.7 keV line.

IV. Transmutation Performance

Photonuclear transmutation is known to be achievable with a variety of γ-ray sources, including LIB, Linac-BR, LCS, and NRγ configurations. In this work, the transmutation performance of LWFA is compared with these representative sources using two transmutation metrics, namely the energy-normalized efficiency and the transmutation rate, as summarized in .

The energy-normalized efficiency is defined as the number of transmuted nuclei per unit of the incident-beam energy. It is noted that this definition does not refer to the wall-plug efficiency. For the LWFA experiment, a transmutation yield of 3.23×10^5 nuclei per shot together with a laser pulse energy of 2.4 J corresponds to 1.35\$×10^{5}\$ J^{-1}. The transmutation rate is defined as the number of transmuted nuclei per unit irradiation time. Using the per-shot yield and the 0.2 Hz repetition rate, the corresponding transmutation rate is $6.46 \times 10^4\ \text{s}^{-1}$.

LWFA vs. LIB. In the LIB, bremsstrahlung γ rays are produced in laser-solid interactions and then irradiate the transmutation target. Ledingham et al. [?] used the VULCAN laser (360 J per shot, 0.7 ps) to generate 2.9×10^6 ^{128}I nuclei per shot, corresponding to an energy-normalized efficiency of $8.06 \times 10^3\ \text{J}^{-1}$. This efficiency is lower than that of the LWFA by about two orders of magnitude, as explained by the electron and γ-ray characteristics discussed below. The LWFA electrons are quasi-monoenergetic at approximately 200 MeV with a divergence of about 5 mrad. They produce a bremsstrahlung flux in which the majority of photons are confined within 0.1 rad (5.7°) and which covers the 9-30 MeV GDR window, with a high-energy tail that maintains a considerable flux. In contrast, the LIB case generates hot electrons within the Au target following a near-Boltzmann distribution with an effective temperature of kT   6.4 MeV. The resulting γ rays have kT   5.5 MeV and a broad angular spread ($\geq$ 45°), as indicated by similar ^{128}I peak intensities measured both along the laser axis and along the target normal. Their broader angular distribution and lower-energy characteristics reduce the photon flux density within the GDR window, thereby lowering the per-joule transmutation yield. In terms of transmutation rate, the LIB method produces $1.45 \times 10^3\ \text{s}^{-1}$ at a repetition rate of 5×10^{-4} Hz [?], whereas the LWFA method achieves a rate higher by a factor of about 45.

LWFA vs. Linac-BR. Linac-BR refers to a linac-driven bremsstrahlung γ-ray source, in which high-energy electrons strike a high-Z converter to generate bremsstrahlung γ rays for photonuclear transmutation. As a representative experiment, Mamtimin et al. [?] operated the electron linac (40 MeV, 2.5 µA, 100 W) at the Idaho Accelerator Center to transmute LLFPs. The electron beam was directed onto two W converters, which were immersed in water, to produce a bremsstrahlung-driven mixed γ-neutron field that irradiated six Cs_2MoO_4 samples placed at different positions in the field. Each sample contained ^{133}Cs as a surrogate for ^{129}I. After 3 h of irradiation and 1 h of cooling, the sample placed between the two W converters produced 21 µCi of ^{132}Cs, corresponding to an EOI yield of about 6.30$×10^{11}$ nuclei. Under these irradiation conditions, this EOI yield gives an energy-normalized efficiency of about 5.81×10^5 J^{-1} and an average transmutation rate of about 5.81×10^7 s^{-1}. From , the Linac-BR transmutation rate exceeds that of the LWFA experiment by about three orders of magnitude. This disparity arises mainly from the much higher average input power of the linac (100 W) compared with that of the LWFA laser (0.48 W), the latter being limited primarily by its low repetition rate (0.2 Hz). When normalized to the input energy, LWFA shows a slightly lower efficiency than the Linac-BR method, while it remains in the same order of magnitude as Linac-BR at 10^5 J^{-1}. This small difference can be reduced by increasing the laser pulse energy and the laser-to-electron conversion efficiency.

LWFA vs. LCS. Among different γ-ray sources, the LCS method generates γ rays through inverse laser Compton scattering. These γ rays are quasi-monochromatic and continuously energy-tunable, making them suited for photonuclear transmutation. Li et al. [?] conducted an LCS transmutation experiment on the BL01 beamline of the NewSUBARU storage ring at the University of Hyogo in Japan. In that work, a 1 GeV stored electron beam and a continuous-wave Nd:YVO$_4$ laser at 1.064 µm with a rated power of 5 W [?] were used to generate LCS γ rays that irradiated a ^{23}Na^{127}I target, in which ^{127}I acted as a surrogate for ^{129}I. The irradiation produced 1.15×10^9 transmuted nuclei over 8 h, corresponding to an average rate of 3.99×10^4 s^{-1}, which is of the same order as that obtained in the LWFA experiment. In a storage ring, circulating electrons continuously lose energy through synchrotron radiation, amounting to 33.4 keV per turn [?]. The electron beam remains stored and is reused on every turn, with its energy periodically compensated. This synchrotron-radiation loss represents the necessary energy investment required to sustain LCS operation for photonuclear transmutation. The beam current of 117 mA is inferred from the parameters reported in Ref. [?]. Using the beam current and the radiative loss, together with the transmuted nuclei over 8 h, the energy-normalized efficiency is estimated to be about 10 J^{-1}, which is lower than that achieved by the LWFA method. Although the LCS method exhibits lower energy-normalized transmutation efficiency, its well-defined photon spectrum enables accurate measurement of photonuclear reaction cross-sections, providing a reliable platform for elucidating transmutation mechanisms.

LWFA vs. NRγ. The NRγ is generated by bombarding target nuclei with protons of specific energies to induce the resonant (p, γ) reaction. This process can also provide γ rays within the GDR energy region. At CIAE, Ye et al. [?] employed the ^{7}Li(p, γ)^{8}Be reaction using a 441 keV, 0.81 µA proton beam from a 2 × 1.7 MV tandem accelerator to generate 14.8 and 17.6 MeV γ rays. These γ rays irradiated a ^{197}Au target for 3.85 h, giving a transmutation rate of 5.87×10^2 s^{-1} and an energy-normalized yield of 1.64×10^3 J^{-1}, both of which are lower than those of the LWFA.

Overall, shows that, in terms of energy-normalized efficiency, the LWFA exceeds

LIB, NRγ, and LCS, and is at the same order as the Linac-BR. For the transmutation rate, the LWFA is higher than LIB and NRγ, close to LCS, and lower than the Linac-BR.

V. Conclusion

In this work, we experimentally investigated LWFA-driven photonuclear transmutation using a CsI target containing ^{133}Cs and ^{127}I as LLFP surrogates. The CLAPA laser system (200 TW, 2.4 J, 0.2 Hz) generated 200 MeV electrons, which in turn produced high-flux, high-energy bremsstrahlung γ rays in a Ta converter that covered the GDR region. Offline HPGe measurement identified the daughter nuclides ^{132}Cs, ^{126}I, and ^{124}I, confirming that photonuclear transmutation occurred in the target. The measured energy-normalized efficiency is 1.35×10^5 J^{-1}, corresponding to a rate of 6.46×10^4 s^{-1}. Geant4 simulation is consistent with the experiment, supporting the reliability of the measured yields.

This performance could be further enhanced with high-intensity laser systems such as ELI-ALPS (700 TW, 34 J, 10 Hz) [**?**], which can deliver higher-energy LWFA electron beams and therefore more intense bremsstrahlung γ rays. With further improvements to the overall experimental setup, the LWFA method is anticipated to achieve energy efficiencies approaching 10^8 J^{-1} [**?**], while maintaining higher transmutation rates. In summary, this study validates the photonuclear transmutation mechanism under LWFA-driven bremsstrahlung irradiation and indicates that it may offer a complementary approach to neutron-induced transmutation.

Acknowledgments

We are grateful to the CLAPA operations team for support with the overall operation of the experimental apparatus.

References

[1] International Atomic Energy Agency, *Status of Minor Actinide Fuel Development*, IAEA Nuclear Energy Series No. NF-T-4.6 (IAEA, Vienna, 2009).

[2] X. Y. Sun, L. H. Han, X. X. Li et al., Transmutation of MAs and LLFPs with a lead-cooled fast reactor. *Sci. Rep.* **13**, 1693 (2023). doi: 10.1038/s41598-023-29002-3

[3] A. Herrera-Martínez, Y. Kadi, G. Parks, Transmutation of nuclear waste in accelerator-driven systems: Thermal spectrum. *Ann. Nucl. Energy* **34**, 550-563 (2007). doi: 10.1016/j.anucene.2007.02.009

[4] S. Chiba, T. Wakabayashi, Y. Tachi et al., Method to reduce long-lived fission products by nuclear transmutations with fast spectrum reactors. *Sci. Rep.* **7**, 13961 (2017). doi: 10.1038/s41598-017-14319-7

[5] W. S. Yang, Y. Kim, R. N. Hill et al., Long-Lived Fission Product Transmutation Studies. *Nucl. Sci. Eng.* **146**, 291–318 (2004). doi: 10.13182/NSE04-A2411

[6] J. Magill, H. Schwoerer, F. Ewald et al., Laser transmutation of iodine-129. *Appl. Phys. B* **77**, 387–390 (2003). doi: 10.1007/s00340-003-1306-4

[7] K. W. D. Ledingham, J. Magill, P. McKenna et al., Laser-driven photo-transmutation of ^{129}I—a long-lived nuclear waste product. *J. Phys. D: Appl. Phys.* **36**, L79–L82 (2003). doi: 10.1088/0022-3727/36/18/L01

[8] A. K. M. L. Rahman, S. Kuwabara, K. Kato et al., Measurement of the photonuclear (γ, n) reaction cross section for ^{129}I using bremsstrahlung photons. *Nucl. Sci. Eng.* **160**, 363–369 (2008). doi: 10.13182/NSE160-363

[9] K. Horikawa, T. Mochizuki, S. Miyamoto et al., Photonuclear reaction of iodine-129 with Laser-Compton scattering gamma-rays using Nd:YVO$_4$ laser and electron storage ring. *Laser Res.* **39**, 445–447 (2011). doi: 10.2184/lsj.39.445

[10] B. Liesfeld, K.-U. Amthor, F. Ewald et al., Nuclear reactions triggered by laser-accelerated relativistic electron jets. *Appl. Phys. B* **79**, 1047–1052 (2004). doi: 10.1007/s00340-004-1652-9

[11] D. Li, K. Imasaki, K. Horikawa et al., Iodine transmutation through Laser Compton scattering gamma rays. *J. Nucl. Sci. Technol.* **46**, 831–835 (2009). doi: 10.1080/18811248.2007.9711592

[12] M. Mamtimin, F. Harmon, V. N. Starovoitova et al., Electron linac based mixed-field transmutation of ^{129}I. *Nucl. Instrum. Methods Phys. Res. B* **344**, 11–15 (2015). doi: 10.1016/j.nimb.2014.11.076

[13] G.-Y. Fu, Y.-L. Dang, F.-L. Liu et al., Photo-transmutation based on resonance γ-ray source. *Chin. Phys. B* **28**, 060707 (2019). doi: 10.1088/1674-1056/28/6/060707

[14] S. Ye, F. Liu, N. Bo et al., Proof-of-principle study of photon-nuclear transmutation based on ^{7}Li(p, γ)^{8}Be reaction. *Atomic Energy Science and Technology* **58**, 714–720 (2024). doi: 10.7538/yzk.2023.youxian.0552

[15] R. Sadighi-Bonabi, E. Irani, B. Safaie et al., Possibility of ultra-intense laser transmutation of ^{93}Zr (γ, n) ^{92}Zr a long-lived nuclear waste into a stable isotope. *Energy Convers. Manage.* **51**, 636–639 (2010). doi: 10.1016/j.enconman.2009.09.017

[16] E. Irani, H. Omidvar, R. Sadighi-Bonabi, Gamma rays transmutation of Palladium by bremsstrahlung and laser inverse Compton scattering. *Energy Convers. Manage.* **77**, 558–563 (2014). doi: 10.1016/j.enconman.2013.09.029

[17] E. Irani, S. K. Sadighi, S. Zare et al., Laser-induced ^{126}Sn - A hazardous nuclear photo transmutation of waste product-into short-lived nuclear

medicine of ^{125}Sn. *Energy Convers. Manage.* **64**, 466–472 (2012). doi: 10.1016/j.enconman.2012.05.024

[18] X. L. Wang, Z. Y. Xu, W. Luo et al., Transmutation prospect of long-lived nuclear waste induced by high-charge electron beam from laser plasma accelerator. *Phys. Plasmas* **24**, 093105 (2017). doi: 10.1063/1.4998470

[19] D. Li, K. Imasaki, M. Aoki, Analysis on Coupling Gamma-ray to Nuclear Giant Resonance. *J. Nucl. Sci. Technol.* **39**, 1247–1249 (2002). doi: 10.1080/18811248.2002.9715317

[20] J.-G. Chen, W. Xu, H.-W. Wang et al., Transmutation of nuclear wastes using photonuclear reactions triggered by Compton backscattering photons at laser electron gamma source. *Chin. Phys. C* **32**, 677-680 (2008). doi: 10.1088/1674-1137/32/8/019

[21] R. Takashima, S. Hasegawa, K. Kato et al., Possibility of transmutation of ^{135}Cs by ultraintense laser. *Appl. Phys. Lett.* **86**, 011501 (2005). doi: 10.1063/1.1847715

[22] Z.-C. Zhu, W. Luo, Z.-C. Li et al., Photo-transmutation of long-lived nuclear waste ^{135}Cs by intense Compton γ-ray source. *Ann. Nucl. Energy* **89**, 109–114 (2016). doi: 10.1016/j.anucene.2015.11.017

[23] X.-L. Wang, Z.-Y. Tan, W. Luo et al., Photo-transmutation of long-lived radionuclide ^{135}Cs by laser–plasma driven electron source. *Laser Part. Beams* **34**, 433–439 (2016). doi: 10.1017/S0263034616000318

[24] Y.-C. Wang, Z.-W. Cao, X.-Y. Sun et al., Study of photo-transmutation induced by laser wakefield accelerated electrons. *High Power Laser and Particle Beams* **35**, 091006 (2023). doi: 10.11884/HPLPB202335.230079

[25] S. K. Sadighi, R. Sadighi-Bonabi, The evaluation of transmutation of hazardous nuclear waste of ^{90}Sr, into valuable nuclear medicine of ^{89}Sr by ultraintense lasers. *Laser Part. Beams* **28**, 269–276 (2010). doi: 10.1017/S0263034610000145

[26] R. Sadighi-Bonabi, O. Kokabee, Evaluation of Transmutation of ^{137}Cs(γ,n)^{136}Cs Using Ultra-Intense Lasers in Solid Targets. *Chin. Phys. Lett.* **23**, 1434–1436 (2006). doi: 10.1088/0256-307X/23/6/021

[27] H. T. Kim, V. B. Pathak, C. I. Hojbota, et al., Multi-GeV laser wakefield electron acceleration with PW lasers. *Appl. Sci.* **11**(13), 5831 (2021). doi: 10.3390/app11135831

[28] S. A. Reed, V. Chvykov, G. Kalintchenko et al., Photonuclear fission with quasimonoenergetic electron beams from laser-wakefields. *Appl. Phys. Lett.* **89**, 231107 (2006). doi: 10.1063/1.2400400

[29] S. A. Reed, V. Chvykov, G. Kalintchenko, et al., Efficient initiation of photonuclear reactions using quasimonoenergetic electron beams from laser wakefield acceleration. *J. Appl. Phys.* **102**, 073103 (2007). doi: 10.1063/1.2787159

[30] D. Wu, H. Y. Lan, J. Y. Zhang et al., New measurements of ^{92}Mo(γ,n) and (γ,3n) reactions using laser-driven bremsstrahlung γ-ray. *Front. Phys.* **11**, 1178257 (2023). doi: 10.3389/fphy.2023.1178257

[31] D. Wu, H. Y. Lan, J. Y. Zhang et al., ^{197}Au(γ,xn; x=1-9) reaction cross section measurements using laser-driven ultra-intense γ-ray source. arXiv:2209.13947v2 [nucl-ex] (2023). doi: 10.48550/arXiv.2209.13947

[32] M. Z. Wang, D. Wu, H. Y. Lan et al., Cross section measurements of ^{27}Al(γ,x)^{24}Na reactions as monitors for laser-driven bremsstrahlung γ-ray. *Nucl. Phys. A* **1043**, 122834 (2024). doi: 10.1016/j.nuclphysa.2024.122834

[33] T. Kawano, Y. S. Cho, P. Dimitriou et al., IAEA Photonuclear Data Library 2019. *Nucl. Data Sheets* **163**, 109–162 (2020). doi: 10.1016/j.nds.2019.12.002

[34] P. Tavana, N. Bukharskii, M. Gyrdymov et al., Ultra-high efficiency bremsstrahlung production in the interaction of direct laser-accelerated electrons with high-Z material. *Front. Phys.* **11**, 1178967 (2023). doi: 10.3389/fphy.2023.1178967

[35] Particle Data Group, *Tantalum (Ta) -Atomic and Nuclear Properties* (2025). https://pdg.lbl.gov/2025/AtomicNuclearProperties/HTML/tantalum_{Ta}.html (accessed 2025-09-10).

[36] V. C. Petwal, S. C. Bapna, R. S. Sandha et al., Bremsstrahlung converter for high power EB radiation processing facility. In *Proc. APAC'07*, Indore, India (2007), THPMA088, pp. 767–769. https://jacow.org/APAC2007/papers/thpma088.pdf

[37] H. Y. Lan, D. Wu, J. X. Liu et al., Photonuclear production of nuclear isomers using bremsstrahlung induced by laser-wakefield electrons. *Nucl. Sci. Tech.* **34**, 74 (2023). doi: 10.1007/s41365-023-01219-x

[38] National Institute of Standards and Technology, *X-Ray mass attenuation coefficients and mass energy-absorption coefficients for cesium iodide element (Cesium Iodide)*. https://physics.nist.gov/PhysRefData/XrayMassCoef/ComTab/cesium.html (accessed 2025-09-10).

[39] G. R. Gilmore, *Practical Gamma-ray Spectrometry*, 2nd ed. (John Wiley & Sons, Chichester, 2008), p. 156.

[40] S. Agostinelli, J. Allison, K. A. Amako et al., GEANT4—a simulation toolkit. *Nucl. Instrum. Methods Phys. Res., Sect. A* **506**, 250 (2003). doi: 10.1016/S0168-9002(03)01368-8

[41] National Nuclear Data Center, *Evaluated Nuclear Structure Data File (ENSDF)*, Brookhaven National Laboratory. https://www.nndc.bnl.gov/ensdf/ (accessed 2025-09-10).

[42] TALYS-based evaluated nuclear data library (TENDL-2023). https://tendl.web.psi.ch/tendl_{2023}/tendl (accessed 2025-11-11).

[43] Wayforlight, *Vulcan Laser System*, Central Laser Facility, Rutherford Appleton Laboratory. https://www.wayforlight.eu/beamline/27195 (accessed 2025-09-10).

[44] LASTI, University of Hyogo, *LCS (γ, n)-ray Beamline BL01 Facility*. https://www.lasti.u-hyogo.ac.jp/NS/facility/bl01/ (accessed 2025-09-10).

[45] LASTI, University of Hyogo, *LASTI Annual Report*, Vol. 24 (University of Hyogo, 2022), p. 9. https://www.lasti.u-hyogo.ac.jp/annualreport/LASTI_{{{Annual}}*{{Report}}}*{{{vol}}}{

[46] R. S. Nagymihály, F. Falcoz, B. Bussière et al., The petawatt laser of ELI ALPS: reaching the 700 TW level at 10 Hz repetition rate. *Opt. Express* **31**, 44160–44176 (2023). doi: 10.1364/OE.509615