Photonuclear production of nuclear isomers using bremsstrahlung induced by laser-wakefield electrons

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Abstract

In this study, we theoretically investigate the feasibility of using laser-wakefield accelerated (LWFA) electrons for the photonuclear measurement of nuclear isomers according to the characteristics of the electrons obtained from LWFA experiments conducted at the Compact Laser Plasma Accelerator (CLAPA) laboratory. The experiments at the CLAPA show that a stable electron beam with an energy of 78–135 MeV and a charge of 300-600 pC can be obtained. The bremsstrahlung spectra were simulated using Geant4, which suggests that a bremsstrahlung source with a peak intensity of 1019 photons/s can be generated. Theoretical calculations of isomer production cross-sections from the photonuclear reactions on six target nuclei, 197Au, 180Hf, 159Tb, 115In, 103Rh, and 90Zr were performed and compared with the available experimental data in EXFOR, which suggest that further experiments are required for a series of photonuclear reaction channels. Flux-averaged cross-sections and isomer ratios (IR) resulting from such bremsstrahlung sources are theoretically deduced. The results suggest that IR measurements can be used to constrain nuclear components, such as γ strength function and optical model potential. In addition, the detection of the decay characteristics was evaluated with Geant4 simulations. The use of the LWFA electron beam and its bremsstrahlung for photonuclear studies involving nuclear isomers is anticipated.

Full Text

Preamble

Photonuclear production of nuclear isomers using bremsstrahlung induced by laser-wakefield electrons

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In this study, we theoretically investigate the feasibility of using laser-wakefield accelerated (LWFA) electrons for the photonuclear measurement of nuclear isomers based on electron characteristics obtained from LWFA experiments conducted at the Compact Laser Plasma Accelerator (CLAPA) laboratory. The experiments at CLAPA demonstrate that stable electron beams with energies of 78–135 MeV and charges of 300–600 pC can be obtained. Bremsstrahlung spectra were simulated using Geant4, suggesting that a bremsstrahlung source with a peak intensity of 10^{19} photons/s can be generated. Theoretical calculations of isomer production cross-sections from photonuclear reactions on six target nuclei—^{197}Au, ^{180}Hf, ^{159}Tb, ^{115}In, ^{103}Rh, and ^{90}Zr—were performed and compared with available experimental data in EXFOR, indicating that further experiments are required for a series of photonuclear reaction channels. Flux-averaged cross sections and isomer ratios (IR) resulting from such bremsstrahlung sources were theoretically deduced. The results suggest that IR measurements can be used to constrain nuclear components such as the γ strength function and optical model potential. In addition, the detection of decay characteristics was evaluated with Geant4 simulations. The use of the LWFA electron beam and its bremsstrahlung for photonuclear studies involving nuclear isomers is anticipated.

Keywords: Photonuclear reactions; Laser plasma acceleration; Flux-averaged isomer ratio

INTRODUCTION

Since the theoretical prediction by Soddy [1] in 1917 and the first experimental observation by Hahn [2] in 1921, nuclear isomers have received intensive attention for both fundamental research and application development [3, 4]. In particular, the effective population and manipulation of nuclear isomers has been an active topic in recent years, driven by the prospect of utilizing their intrinsic or depleted decays for developing nuclear clocks, nuclear batteries, and gamma lasers [5–9]. Moreover, exploring nuclei with isomeric states in nuclear astrophysics allows us to determine how they affect element creation in the universe and eventually contribute to the makeup of life in our cosmos [10].

In recent decades, worldwide development of high-intensity and high-repetition-rate laser systems has opened new research opportunities for particle acceleration [11–17] and nuclear photonics [18]. The ultra-short pulse radiation source and plasma environment generated by laser-target interaction provide unique physical conditions for studying nuclear isomers. Using a table-top hundred TW laser, Feng et al. [19] demonstrated that femtosecond pumping of ^{83} Kr with a peak efficiency of 2.34×10^{15} s^{-1} can be achieved through Coulomb excitation of ions with quivering electrons during laser pulse-cluster interactions at nearly solid densities. Wang et al. [20] proposed a novel approach to populate the nuclear clock isomer ^{229} Th by combining laser-driven electron recollision and electronic nuclear excitation (e.g., Coulomb excitation), which does not require precise knowledge of the isomeric energy. The common idea of these schemes is to utilize the dense low-energy electrons produced in laser-plasma interactions and the relatively high electronic excitation cross sections of ionized atoms in plasma. A comprehensive comparison of possible physical processes for isomer population and manipulation, including nuclear excitation by electron capture, excitation by electron transition, electron bridge, and photoactivation, was also given in Ref. [21], indicating that nuclear excitations assisted by electrons would be far more efficient than straightforward photon excitation. This is reasonable for isomer manipulation or triggered isomer depletion scenarios where the target nucleus must remain unchanged and the excitation energy required is as low as the difference between atomic electron shells. However, this is not the case for isomer population, where the isomer can be populated without extra scruples on target nucleus disintegration. Specifically, the cross-section of photonuclear reactions can be significantly enhanced by giant dipole resonances (GDR) to hundreds of millibarns above the neutron separation threshold, whereas only narrow discrete resonances from isolated bound states are generally mentioned in most discussions of photoactivation of nuclear isomers. Utilizing (γ,γ') scattering of the collective excitation mode, Pan et al. [22] depicted the prospect of accumulating medically interesting isomers to clinically applicable activity with an intense monochromatic γ-ray beam. Meanwhile, quasimonochromatic high-intensity electron pulses generated by laser wakefield acceleration (LWFA) in gas jets are commonly available at high-power laser facilities and can be converted into bremsstrahlung radiation, thus providing vast research opportunities for photonuclear studies. Although previous studies have utilized laser-driven photonuclear reactions for electron diagnosis [23], nuclear waste transmutation [24], and medical isotope production [25], little attention has been paid to nuclear isomers. Consequently, a systematic investigation is required to evaluate the potential of isomer population by laser-driven photonuclear excitation.

In this work, we theoretically investigate the potential to accumulate nuclear isomers by laser-driven photonuclear excitation through Monte Carlo simulations based on electron characteristics obtained from LWFA experiments. LWFA experiments at the Compact Laser Plasma Accelerator (CLAPA) laboratory of Peking University are introduced, and the conversion from electrons to bremsstrahlung is simulated with Geant4 [26]. Theoretical calculations of photonuclear cross-sections are then performed. A series of nuclear isomers were selected as calculation candidates, considering both the nuclear flow of photonuclear reactions and the availability of stable natural target nuclei. The corresponding photonuclear cross-sections for isomer generation were evaluated using TALYS [27, 28]. To verify the reliability of the TALYS inputs, a benchmark of theoretical predictions against available experimental data extracted from the EXFOR database [29] was performed. The flux-averaged cross-sections and flux-averaged isomer ratios were deduced according to theoretical photonuclear cross-sections and simulated bremsstrahlung spectra. Furthermore, photonuclear cross-sections of different reaction channels were incorporated into the simulation toolkit Geant4-GENBOD [30–32] to estimate the feasibility of detecting the nuclear isomers of interest. In the simulations, typical laser-driven electron beam parameters extracted from experiments are considered, along with physical processes of radioactive decay of various reaction residuals and energy deposition in commercially available $LaBr_3$ scintillation detectors. A comprehensive discussion is provided in the context of production and detection of the nuclear isomers of interest.

II. LWFA ELECTRONS AND BREMSSTRAHLUNG CONVERSION AT CLAPA

We conducted experiments to accelerate electrons for bremsstrahlung generation with the 200 TW laser facility at CLAPA, Peking University, which delivers laser pulses with 30 fs duration, 4 J energy, and 5 Hz repetition rate. The laser beam was focused to a spot of 21 × 23 μm full width at maximum onto the leading edge of a 4 × 1 mm supersonic gas jet. The effective energy on target was approximately 1.8 J with a laser intensity of approximately 1.5×10^{19} W/cm^2. In the interaction between laser pulses and mixed gas composed of 5% N_2 and 95% He, monochromatic energetic electrons can be produced by LWFA. The electrons propagate through a thin Be window, exit the vacuum chamber, and reach a magnetic spectrometer placed 100 cm downstream of the gas jet, where deflected electron signals are diagnosed using fluorescent screens and CCD cameras. The electron charge was measured by a Turbo Integrating Current Transformer (Turbo-ICT) assembled right after the Be window.

The back pressure of the gas jet was varied from 33 bar to 39 bar, obtaining LWFA electrons with peak energies of 78–135 MeV. One hundred continuous shots were recorded for each gas pressure to test the stability of accelerated electrons. The repetition rate remained at 0.25 Hz to ensure the vacuum level of the laser-gas interaction chamber could recover for LWFA. Typical electron distributions recorded by the magnetic spectrometer are shown in Fig. 1(a)–(c). Electron energies of 78 ± 10, 103 ± 14, and 135 ± 20 MeV were obtained at 39, 36, and 33 bar, respectively. The divergence of the electrons was within 4.3 mrad and the directional stability was better than 5.2 mrad. The electron charge was measured to be 300–600 pC.

The averaged energy and divergence distributions of electrons obtained at 39, 36, and 33 bar were incorporated into Geant4 to simulate the resulting bremsstrahlung characteristics. In the simulations, electrons were directed to a 2-mm-thick Ta converter 1 m away from the electron generation point, inducing bremsstrahlung radiation. The electron charge was set to 300 pC, which is a conservative estimate for the number of LWFA electrons produced per shot. The bremsstrahlung photons were recorded in a plane perpendicular to the propagation direction assembled 20 cm downstream from the converter. The simulated bremsstrahlung energy distributions are shown in Fig. 1(d). The photon intensity shows a typical decaying trend with increasing photon energy, which can fully cover the GDR region of the selected target nuclei. The conversion efficiency from electrons to photons increases with increasing electron energy. The total photon flux can reach 4.35×10^8 photons/shot, corresponding to a peak photon flux of 1.24×10^{19} photons/s considering a pulse duration spread of 35 ps inside the converter. The correlation between photon energy and polar angle for incident electrons at 33 bar is shown in Fig. 1(e). The angular spread of the photons tends to narrow with increasing photon energy, with most photons within 0.4 rad. With such a photon beam featuring high peak flux and ultrashort pulse duration, one can potentially measure nuclear isomers with short half-lives produced in photonuclear reactions. Accordingly, reaction data covering the GDR and quasi-deuteron regions were probed.

III. PRODUCTION ROUTE AND PHOTONUCLEAR CROSS-SECTIONS

A. Theory

The nuclear flow of photonuclear reactions and mechanism for isomer generation are depicted in Fig. 2. When a target nucleus is excited above the particle separation energy, the excited compound subsequently decays through emission of particles (such as neutrons, protons, and alpha particles) or photons to different energy levels of the residual nucleus, including the isomer state. At energies of 10–40 MeV, the GDR resulting from out-of-phase oscillation between proton and neutron fluids clearly enhances the probability of photon absorption and compound nucleus formation. At 40–130 MeV, the photon interacts with the nucleus via the quasi-deuteron mechanism (QD), where the compound nucleus decays predominantly via emission of several nucleons, mostly neutrons. The decay of the compound nucleus leading to isomer population can be well described by the Hauser-Feshbach reaction model, which is based on Bohr's fundamental hypothesis that reactions occur through intermediary formation of a compound nucleus that can reach thermodynamic equilibrium. The Hauser-Feshbach model averages over many resonances in compound nucleus formation and can be applied when the nuclear level density (NLD) in the compound nucleus is sufficiently high at the compound formation energy (the excitation energy at which the compound nucleus is formed).

B. Verification of photonuclear cross-sections

Generally, photonuclear reactions tend to push the involved nucleus toward the lower-left side of the nuclear chart, placing a clear limit on possible populated isomers when considering target nucleus stability. According to the nuclear flow of photonuclear reactions and target nucleus stability, we selected a series of stable nuclides—^{197}Au, ^{180}Hf, ^{159}Tb, ^{115}In, ^{103}Rh, and ^{90}Zr—as candidates for theoretical calculations of reaction cross-sections. Cross sections for typical photonuclear reactions such as (γ, γ'), (γ, xn), (γ, p), and (γ, α) were calculated using the software package TALYS, which provides a complete description of all reaction channels and observables and includes many state-of-the-art nuclear models covering all main reaction mechanisms encountered in light-particle-induced nuclear reactions. The nuclear structure components used in TALYS calculations were as follows:

Nuclear masses were taken from the Atomic Mass Evaluation 2016 (AME2016) [67] whenever available, while HFB-27 nuclear masses [68] were used when AME2016 mass data were unavailable. Discrete experimental levels compiled in the RIPL-3 library [69] and the continuum level spectrum represented by nuclear level densities (NLDs) were both considered in calculations. NLDs were obtained from predictions in the Fermi gas model plus a constant temperature. Photon strength functions (SFs) from the Brink-Axel Lorentzian model were used to calculate electromagnetic transmission coefficients for the photon channel. Phenomenological optical model potentials (OMPs)—specifically, the local and global parameterizations of Koning and Delaroche—were employed to determine transmission coefficients for particle channels.

Theoretical photonuclear reaction cross-sections based on TALYS for selected targets and corresponding experimental data available in the EXFOR database are shown in Fig. 3. The (γ, n) reaction cross-section is generally dominant in the GDR region, with a maximum of hundreds of mb. As excitation energy continues to increase, multiple emission reactions take over, with maximum cross-section decreasing as the number of emitted particles increases. Theoretical calculations agree with EXFOR data obtained from previous measurements using photon sources such as (tagged) bremsstrahlung [33–39], laser-Compton γ rays [40–44], and positron annihilation in flight [45–53], indicating the reliability of TALYS inputs and calculations. However, previous measurements have not fully covered all photonuclear reaction channels, although (γ,xn) reaction data for the most studied nucleus ^{197}Au are rather abundant. Future experiments are expected to provide new data for these reaction channels.

Isomer production cross-sections from photonuclear reactions on the six selected target nuclei are shown in Fig. 4. Isomer production from (γ, n) reactions is generally greater than from other reaction channels, with maximum cross-sections of approximately 100 mb. Experimental isomer production cross-sections are very rare and could potentially be measured using state-of-the-art LCS facilities [70–72] in the future.

IV. PRODUCTION YIELDS AND FLUX-AVERAGED ISOMER RATIOS

A. Isomer production yields

In bremsstrahlung-driven photonuclear measurements, isomers and their decay have long been interesting topics because they can provide constraints for nuclear structures. Under bremsstrahlung radiation, the flux-averaged cross sections (FACS) of a given photonuclear reaction can be given by:

$$\sigma_{FA}(E_e) = \frac{\int_{E_{th}}^{E_e} \sigma(E)\Phi(E)dE}{\int_{E_{th}}^{E_e} \Phi(E)dE}$$

where $\sigma(E)$ is the reaction cross-section at photon energy E, $\Phi(E)$ is the photon flux, E_e is the electron energy, and E_{th} is the photonuclear reaction threshold.

Using the bremsstrahlung driven by laser-plasma-accelerated electron beams described in Section II, we conducted experimental measurements on the (γ,xn) cross sections of ^{197}Au and compared them against theoretical estimations based on Eq. 1. Experimental details and data analysis can be found in Ref. [73]. Agreement between experimental results and theory demonstrates that measuring FACS with laser-driven bremsstrahlung is feasible. With this premise, we further evaluated expected FACS for possible isomer residuals when laser-driven bremsstrahlung beams were used for photoactivation of ^{197}Au, ^{180}Hf, ^{159}Tb, ^{115}In, ^{103}Rh, and ^{90}Zr, as shown in Fig. 5. Notably, FACS for producing nuclear isomers $^{196\ 1}$Au, ^{195} Au, $^{179\ 1}$Hf, ^{158} Tb, ^{114} In, ^{101} Rh, and ^{89} Zr were relatively larger than others, exceeding 10 mb. Moreover, available experimental FACS for producing nuclear isomers for these target nuclei are very limited. The evaluated FACS for producing $^{196\ 2}$Au from the (γ, n) reaction on ^{197}Au agrees well with experimental measurements by Naik et al. [61], while those for producing ^{102} ,^{101} ,^{100} ,^{99} Rh from (γ,xn) reactions on ^{103}Rh are all lower than experimental values obtained by Rahman et al. [62]. FACS for isomers of other isotopes produced from reactions such as (γ, p) and (γ, α) are also shown in the Supplementary Material, which are less abundant in experimental data. These results show that experimental efforts to measure FACS involving isomer residuals are expected to resolve discrepancies and deficiencies in photonuclear data in the QD region.

B. Dependence of isomer ratio on nuclear structure ingredients

The ratio of probabilities of forming the isomer and ground state is called the isomeric ratio, given by:

$$IR_{m_i/g} = \frac{\sigma_{FA}^{m_i}(E_e)}{\sigma_{FA}^{g}(E_e)} = \frac{\int_{E'_{th}}^{E_e} \sigma_{m_i}(E)\Phi(E)dE / \int_{E'_{th}}^{E_e} \Phi(E)dE}{\int_{E_{th}}^{E_e} \sigma_g(E)\Phi(E)dE / \int_{E_{th}}^{E_e} \Phi(E)dE}$$

where $\sigma_{FA}^{m_i}(E_e)$ and $\sigma_{FA}^{g}(E_e)$ are FACS to produce the i-th isomer and ground state, respectively; $\sigma_{m_i}(E)$ and $\sigma_g(E)$ are production cross-sections for the i-th isomer and ground state; and the production threshold for the i-th isomer E'_{th} differs slightly from that for the ground state E_{th}.

In this study, we theoretically investigated the sensitivity of IR to three nuclear structure ingredients: NLD, OMP, and SF. Specifically, IRs of $^{196\ 2}$Au to ^{196} Au ($IR_{m2/g}$), $^{196\ 1}$Au to ^{196} Au ($IR_{m1/g}$), and $^{196\ 2}$Au to the sum of ^{196} Au and $^{196\ 1}$Au ($IR_{m2/(m1+g)}$) produced from (γ, n) reactions of Au were evaluated using different sets of NLDs, OMPs, and SFs available in TALYS.

Results predicted using six sets of NLDs (1: Constant temperature Fermi gas model [74]; 2: Back-shifted Fermi gas model [75]; 3: Generalised superfluid model [76]; 4: HFB-Skyrme model [77]; 5: HFB-Skyrme model with combinatorial method [78]; 6: Temperature-dependent HFB-Gogny model [79]), two sets of OMPs (0: phenomenological Wood-Saxon potential [80]; 1: semi-microscopic JLMB potential with HFB-Skyrme matter density [68, 81–83]), and eight sets of PSFs (1: Generalized Lorentzian [84]; 2: Brink-Axel Lorentzian [85, 86]; 3: HFBCS-QRPA model [87]; 4: HFB-Skyrme-QRPA model [88]; 5: Hybrid model [89]; 6: T-dependent HFB-Skyrme-QRPA model [88, 90]; 7: T-dependent RMF model [91]; 8: HFB-D1M-QRPA model [92]) are shown in Fig. 6. IRs grow rapidly with increasing incident electron energy at low energies and tend to saturate at higher energies of tens to hundreds of MeV, where discrepancies from different models emerge. Specifically, $IR_{m1/g}$ is not sensitive to the three nuclear ingredients, whereas both $IR_{m2/(m1+g)}$ and $IR_{m2/g}$ show relatively strong sensitivity to SFs and NLDs. This suggests that existing experimental data and further measurements aimed at IRs, especially in high-energy regions, can be used to constrain these nuclear-structure ingredients.

V. DETECTION OF CHARACTERISTIC DECAYS

In photonuclear experiments, there are two ways to measure reaction cross-sections: online detection of photo-induced particle emissions (neutrons, protons, and α particles) and offline detection of characteristic γ decays of radioactive residuals. The former is generally used in experiments with energy-tunable, quasi-monochromatic probes such as laser-Compton γ-ray sources and positron annihilation in flight, because energy spectra of emitted particles exhibit easily identifiable reaction kinematics. In bremsstrahlung-driven photonuclear experiments, offline detection is typically performed. Detection of residuals depends not only on their decay characteristics and production yields but also on detector energy resolution.

We performed Geant4 simulations to evaluate the feasibility of detecting considered isomers produced in photonuclear reactions. The simulation setup is arranged as follows: The energy distribution and angular spread of bremsstrahlung produced by LWFA electrons were set according to experimental results obtained at a gas pressure of 36 bar. The electron charge was set as 100 shots at 300 pC. The bremsstrahlung photons irradiated activation targets with 5 mm thickness, placed 20 cm downstream of the converter, inducing photonuclear reactions and producing radioactive residuals. The γ decays of radioactive residuals, including isomers, were recorded by a $LaBr_3$ detector with dimensions of ϕ 8 cm × 10 cm assembled at 90° with respect to beam propagation. Energy resolution was set to 3%, which is achievable for commercially available $LaBr_3$ detectors. To simulate isomer production and decay radiation detection in a one-stop manner, we adopted the Geant4-GENBOD [30–32] toolkit and implemented the radioactive decay process of radioactive residuals (including isomers). Photonuclear reaction cross-sections of related isotopes were calculated with a routine similar to Section III B.

An exemplary correlation of energy deposition versus detection time for radioactive residuals induced by photonuclear reactions on ^{90}Zr is given in Fig. 7. In activation experiments based on laser-plasma acceleration, there is a period (cooling time t_c) for opening the reaction chamber before activated samples are transferred to the detector for data acquisition. If t_c exceeds 1 h and 10 s, decay information for products ^{89} Zr ($T_{1/2} = 4.16$ m) and ^{90} Zr ($T_{1/2} = 809$ ms) will be lost. To detect isomers with short half-lives such as ^{90} Zr, the detector must be placed near the activation target in an "online" manner. Backgrounds induced by backscattered bremsstrahlung, electrons, and electromagnetic pulses can be cut off by applying a time window with a delay signal generator. The correlation of energy deposition versus detection time for radioactive residuals induced by photonuclear reactions on all six selected targets is presented in the Supplementary Material. Principal decay characteristics and expected counts within three half-lives for isomers produced from photonuclear reactions of the six selected isotopes are shown in Table 1. Photonuclear products $^{196\ 1}$Au, $^{194\ 1}$Au, ^{157}Tb, $^{156\ 1}$Tb, and ^{103} Rh are not measurable with the $LaBr_3$ detector because their decay energy is too low and attenuation in the target is strong. Measurement of $^{195\ 2}$Au is unlikely because its decay energy and intensity have not yet been determined. For ^{180}Hf, decay of ^{180} Hf is difficult to measure because the (γ, γ') reaction cross-section is very low. Most isomer residuals can be measured with net counts of 10^2–10^6 within 14 days of data acquisition. Decay energies of some products such as ^{193}Au and ^{193} Au are very close and cannot be disentangled from the recorded energy spectrum of a detector with poor energy resolution, though this can be remedied by fitting recorded counts across several data-acquisition cycles.

VI. CONCLUSION

In this study, we calculated production cross-sections of nuclear isomers from photonuclear reactions on ^{197}Au, ^{180}Hf, ^{159}Tb, ^{115}In, ^{103}Rh, and ^{90}Zr with TALYS. Deficiencies in corresponding experimental data were found in comparisons between theoretical estimations and available EXFOR data. Laser-plasma electron acceleration experiments at CLAPA were introduced. Stable LWFA electrons with energies of 78 ± 10, 103 ± 14, and 135 ± 20 MeV can be obtained with charges of 300–600 pC per shot. The resulting peak bremsstrahlung flux is expected to be 1.24×10^{19} photons/s. To evaluate the prospect of using such a photon beam for photonuclear studies regarding nuclear isomers, FACSs and IRs for isomer production were deduced and compared with EXFOR data. The sensitivity of IRs to nuclear ingredients SF and NLD was determined, highlighting the importance of measuring photonuclear reactions involving isomers. Moreover, the prospect of photonuclear measurement of nuclear isomers using LWFA electrons at CLAPA is foreseen through Geant4 simulations.

VII. ACKNOWLEDGEMENT

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AUTHOR CONTRIBUTIONS

All authors contributed to study conception and design. Material preparation, data collection, and analysis were performed by Hao-Yang Lan, Di Wu, Jia-Xin Liu, Jian-Yao Zhang, Huan-Gang Lu, Jian-Feng Lv, and Xue-Zhi Wu. The first draft was written by Hao-Yang Lan and all authors commented on previous versions. All authors read and approved the final manuscript.

DATA AVAILABILITY STATEMENT

The data supporting the findings of this study are openly available at https://doi.org/10.57760/sciencedb.j00186.00066 and https://cstr.cn/31253.11.sciencedb.j00186.00066.

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