Bayesian evaluation of photofission product yields of Th isotopic chains

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Date: 2025-10-10T19:41:08+00:00

Abstract

The fragment yields in photon-induced fission reaction of thorium (Th) isotopes are important in modern nuclear energy applications, as well as in the evolution of the nuclear structure in its isotopic chains. Bayesian neural networks (BNN) models have been constructed to describe fragment yields in the photonuclear fission reactions of thorium isotopes ranging from ^{216}Th to ^{232}Th, including those of ^{232}Th at various incident photon energies. The predicted results of the optimized BNN models show good agreement with the measured data in the reactions. The double-layer BNN models successfully illustrate the systematic transition from asymmetric to symmetric fission in thorium isotopes, including associated odd-even effects, energy dependence, and the leftward shift in mass yield distributions. The developed BNN models provide new tools for predicting fragment yields in thorium photonuclear fission reactions.

Full Text

Preamble

Bayesian evaluation of photofission product yields of Th isotopic chains

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The fragment yields in photon-induced fission reactions of thorium (Th) isotopes are important for modern nuclear energy applications and for understanding the evolution of nuclear structure along isotopic chains. Bayesian neural network (BNN) models have been constructed to describe fragment yields in the photonuclear fission reactions of thorium isotopes ranging from ^{216}Th to ^{232}Th, including those of ^{232}Th at various incident photon energies. The predicted results from the optimized BNN models show good agreement with measured reaction data. The double-layer BNN models successfully illustrate the systematic transition from asymmetric to symmetric fission in thorium isotopes, including associated odd-even effects, energy dependence, and the leftward shift in mass yield distributions. These developed BNN models provide new tools for predicting fragment yields in thorium photonuclear fission reactions.

Keywords: Photonuclear fission reaction, Thorium isotopes, ^{232}Th, Bayesian Neural Network, Total element yield, Mass chain yield, Odd-even staggering phenomenon

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[Figure 1: see original paper]. (Color online) A schematic diagram of a neural network with double hidden layers of 3-3 neurons (H_1=3, H_2=3) and two input variables (I=2).

INTRODUCTION

Photon-induced nuclear fission (PNF) of a heavy nucleus is a process in which high-energy photons interact with the nucleus, causing it to split into two or more lighter hot fragments, followed by decay processes that release neutrons, gamma rays, and energy [1, 2]. The mechanism of PNF differs from other particle-induced nuclear fissions because only electromagnetic interaction exists between the photon and the nucleus [3–5], which can transfer clear angular momentum and directly affect the distribution of fission products, providing a clean probe for further research and understanding of nuclear structure and reaction mechanisms. The PNF process holds significant scientific and applied importance in many fields, including nuclear physics research, energy development, nuclear medicine, and nuclear astrophysics [6, 7]. The PNF reaction also enables unique study of the interaction between macroscopic and microscopic degrees of freedom in the nucleus. At low excitation energy, the reaction mechanism is strongly influenced by nuclear structure and shell effects. The pairing effect plays an important role in the mass distribution and nuclear charge distribution of fission fragments [8–10]. However, existing experimental data can only indirectly explain the influence of nuclear structure on the fission process. Additional data from PNF reactions are important for improving our understanding of this process [11, 12].

Currently, experimental measurements of fission reactions are very difficult. Existing fission data are often incomplete and subject to large uncertainties. Machine learning technologies have demonstrated high quality in accurately predicting residual nucleus cross sections in nuclear spallation reactions [13-15] and projectile fragmentation reactions [16–20]. New theories based on machine learning have also been developed to describe the fissile mechanism of heavy nuclides [21–23], such as extrapolating nuclear mass, fission yield, various nuclear structures, and observed reaction quantities [24, 25]. Due to the difficulties in describing fragment production in PNF reactions, Bayesian Neural Network (BNN) technology is applied to construct models that describe fragment yields in PNF reactions of thorium isotopes to meet requirements for their important applications in modern nuclear reactors, nuclear medicine, and nuclear structure, as well as to evaluate incomplete charge and mass distributions of fragments. The excitation functions of fragments of interest are also investigated.

This article is organized as follows. In Sec. II, the method for building BNN models is described. In Sec. III, the BNN evaluation of total element yield and mass chain yield of PNF fragments is presented. A summary is given in Sec. IV.

II. BNN MODELS

To construct a BNN model that reproduces and predicts the cross sections of fragments, the first step begins with establishing the prior distribution of model parameters by observing the given sample data D = (x_1, t_1)···(x_N, t_N), where x_n and t_n (n = 1, 2,···, N) are the input and output datasets, respectively, and N is the total sample size. According to Bayes' theorem, the posterior probability distribution of the model parameters is obtained as follows:

$$p(\theta|D) = \frac{p(D|\theta)p(\theta)}{p(D)}$$

where p(D |) is the likelihood function of the model given parameters , p( ) is the prior probability of model parameter , and p(D) is the normalization constant representing the marginal density of the observed sample, defined as:

$$p(D) = \int f(D|\theta)f(\theta)d\theta$$

For a regression task where the goal is to predict the target variable given an input vector, the likelihood function is typically defined as a Gaussian distribution:

$$p(D|\theta) = \exp\left(-\frac{\chi^2}{2}\right)$$

The objective function ^{2} is defined as:

$$\chi^2 = \sum_{n=1}^{N} \frac{(t_n - f(x;\theta))^2}{\Delta t_n^2}$$

where Δt_n is the noise error. The BNN model typically employs a multilayer perceptron (MLP) network, also known as a "backpropagation" or "feedforward" network. These networks consist of a set of input variables x_i = {Z_{fi}, Z_i, A_i, E_i}, one or several hidden layers with different numbers of neurons, and one or more output variables t_k(x;). The functional equation of a typical MLP network with a single hidden layer is:

$$f(x;\theta) = a + \sum_{j=1}^{H} b_j \tanh\left(c_j + \sum_{i=1}^{I} d_{ji} x_i\right)$$

where H is the number of hidden neurons in the hidden layer, I is the number of input variables, x = {x_i} is the dataset of input variables, and = {a, b_j, c_j, d_{ji}} defines the parameters of the MLP network equation corresponding to the biases and weights of the output layer, the biases and weights of the hidden layer, respectively. The total number of neurons is 1 + (2 + I) × H. In the BNN model, each output variable f(x) is obtained by taking the weighted sum of the hidden unit values and adding the bias of the hidden layer, and each hidden unit value is calculated by taking the weighted sum of the input values and then applying a non-linear activation function. Figure 1 illustrates a schematic diagram of a typical neural network with double hidden layers of 3-3 neurons (H_1=3, H_2=3) and two input variables (I=2).

In this work, two independent BNN models were constructed to predict the yields of two different PNF fragment distributions: one for charge yields (called BNN-CY) for PNF of thorium isotopes (^{216}-^{232}Th) and another for mass yields (called BNN-MY) of PNF fragments for ^{232}Th at different γ incident energies. All calculations in this work adopted 10^5 sampling iterations of BNN, with uncertainty quantification presented using 80% confidence intervals (CI). The optimal number of neurons in each layer was determined by systematically varying the number of neurons per layer and comparing the corresponding standard deviations ^{2}_N to find the best configuration. The ^{2}_N is determined by:

$$\chi_N^2 = \sum_{i=1}^{N} \frac{[t_i - f(x_i)]^2}{N}$$

[Figure 2: see original paper]. (Color online) The photonuclear fission charge yields of γ + ^{217}Th at excitation energy of 11 MeV predicted by the BNN-CY model constructed based on EXFOR data without ^{217}Th. Comparison of charge yields for the ^{217}Th nuclide in this work (full lines) and experimental data from Ref. [32] (solid squares). The shaded region corresponds to the confidence interval (CI) at 80%.

III. RESULTS AND DISCUSSION

A. The fission mechanism of Actinide elements

Before discussing the predicted results of the BNN models and to better understand the evaluation of fragment yields by BNN in γ-induced thorium fission reactions, it is important to review the basic phenomena of fragment yield distribution discovered in experiments.

In this subsection, we list the main experiments and their conclusions. The phenomenological Brosa model [26, 27], based on the mass distribution of fission products, postulates the existence of two asymmetric fission modes. The origin of these distinct asymmetric fission modes is attributable to the influence of fission barriers. The Brosa model further describes the broadening of fission observables through stochastic neck rupture, where longer necks correspond to broader distributions. With the advancement of experimental research, three primary fission modes have been proposed for the nearly stable actinide region: two mass-asymmetric modes standard I (ST1) and standard II (ST2), and a superlong symmetric mode (SL) [28–31].

- ST1 is predominantly governed by doubly magic shell closure around ^{132}Sn, resulting in a nearly spherical heavy fragment.
- ST2 is characterized by the stabilization of the heavy fragment near Z_f = 54.
- SL exhibits a symmetric mass split, forming two fragments with comparable masses, both undergoing significant deformation.

Schmidt et al. [30, 31] investigated fission reactions of 70 short-lived radioactive isotopes in 2000 using the secondary beam facility at GSI Darmstadt. The primary beam of 1 A GeV ^{238}U was irradiated on a beryllium target. The mass and charge of the fragments produced were analyzed using the Fragment Separator (FRS). These products were utilized as a secondary beam, which was excited electromagnetically by interacting with a secondary lead target, leading to fission of the nucleus within an excitation energy range of approximately 11 MeV, mostly by the giant dipole resonance. This provided the first experimental characterization of the transition from asymmetric fission in actinides to symmetric fission in pre-actinides. Chatillon et al. [32] also conducted a similar experiment at GSI in 2019, employing the R3B/SOFIA (Reactions with Relativistic Radioactive Beams/Studies On FIssion with Aladin) setup [33–35]. The mass and nuclear charge of the fission fragments were measured in coincidence with the total prompt neutron multiplicity, which infers evidence of the existence of a new compact symmetric fission mode in the light thorium isotopic chain [36]. Based on the predictions of the BNN, a detailed discussion of the photofission mechanism of Th isotopes will be presented in Sec. III B and Sec. III C.

[Figure 3: see original paper]. (Color online) The charge yield distribution of fragments from γ-induced fission of thorium isotopes at excitation energy of 11 MeV. Comparison of one-layer (green lines) and two-layer (pink lines) BNN-CY learning results of fragment charge yields from Ref. [30]. Open circles represent experimental data. The shaded region corresponds to CI at 80%.

[Figure 4: see original paper]. (Color online) The charge yield distribution of fragments from γ-induced fission of thorium isotopes at different excitation energies. Comparison of one-layer (green lines) and two-layer (pink lines) BNN-CY learning results of fragment charge yields from Ref. [32]. Open circles represent experimental data. The shaded region corresponds to the CI at 80%.

B. Total element yield of fission products

The BNN-CY model was constructed to predict PNF fragment charge yields based on learning experimental charge yield data in the EXFOR database, which includes 1,437 experimental data points from PNF products of 14 nuclides (^{217}-^{230}Th). All measured data are taken from Refs. [30, 32]. The input dataset of BNN-CY consists of $x_i = \{Z_{fi}, Z_i, N_i, E_i\}$, which refer to the charge number of fission fragments, the charge and neutron numbers of the fissioning nucleus, and the excitation energy (E_i) of the compound nucleus, respectively. The output dataset t_i is the charge yield of fission fragments. The fission charge yields of the compound nucleus ^{217}Th predicted by the BNN-CY model are illustrated in Fig. 2. For clarity, prediction results from neural networks without odd-even parameters display only central values without confidence intervals. Predictive results from single-layer neural networks excluding odd-even parameters demonstrate two notable anomalies: negative values emerge in the vicinity of fission fragment charge $Z_f = 35$, while an abnormal upward tail becomes apparent for $Z_f > 60$. Moreover, double-layer neural networks without odd-even parameters generate aberrant negative predictions near $Z_f = 54$. The production of fission fragments with even numbers of protons is usually enhanced because the fully paired proton configuration tends to survive at scission with high probability. To describe the odd-even effect in the reaction system, an additional input $\delta = \pm 0.1$ was added to the BNN-CY learning collection to represent even and odd fragments, respectively. The input parameter set becomes $x_i = \{Z_{fi}, Z_i, A_i, E_i, \delta_i\}$. As shown in Fig. 2, BNN-CY evaluations without the odd-even parameter δ failed to capture the odd-even effect in the charge yield distribution of PNF fragments, while inclusion of the odd-even parameter δ allowed BNN-CY to accurately reproduce this effect.

Based on the values of $^{2}_{N}$ in Eq. (6), to optimize the number of neurons in the hidden layers, a double-layer network with 30 neurons was ultimately adopted for this study. Generally, a double-layer network significantly enhances learning performance for data points due to its larger number of connection parameters compared to a single-layer network, even when both have the same total number of neurons. To verify this point and better describe the odd-even effect in the reaction system, we conducted a detailed comparison of evaluation results between single-layer neural networks with odd-even parameters and double-layer neural networks with odd-even parameters. Specifically, a single-layer network with 30 neurons and a double-layer network with 15-15 neurons were compared, with 10^5 BNN-CY sampling iterations performed for both. The standard deviations ^{2}_N obtained from Eq. (6) were 1.7$×10^{2}$ and 1.3$×10^{2}$, respectively, indicating similar uncertainties. In Figs. 3 and 4, the single-layer network can effectively reproduce the odd-even effect in the charge distribution of fission fragments, similar to the double-layer network. However, the confidence interval of the double-layer network is slightly larger than that of the single-layer network.

Figure 3 presents a comparison between BNN-CY evaluation results and experimental charge yield data for PNF fragments from targets ^{217}-^{229}Th at an average excitation energy of 11 MeV, as reported by Schmidt et al. [30]. The BNN-CY results are in good agreement with experimental data. Notably, neural networks incorporating odd-even parameters successfully reproduce the characteristic odd-even staggering in the charge distribution of the reaction system. Furthermore, these calculations accurately capture the transition from asymmetric to symmetric fission in the PNF mechanism across the thorium isotopic chain. The number of peaks in the fission fragment charge distribution evolves with increasing neutron number of the target nuclei: single-peak distributions for neutron-deficient thorium targets transition through triple-peak patterns in intermediate-mass thorium nuclei, eventually becoming double-peak distributions for heavier thorium isotope targets. This systematic evolution demonstrates the strong dependence of fission mechanisms on neutron number along the Th isotopic chain. Experimental data on charge yields of ^{221}-^{230}Th fission fragments at an average excitation energy of around 14 MeV by Chatillon et al. [32] in Fig. 4 show a similar phenomenon.

The results predicted by the double-layer network with the smallest ^{2}_N value were selected for further evaluation and prediction, focusing on charge yield values associated with the three prominent peaks observed in the fission fragment charge distribution across the thorium isotopic chain. Fragments with Z_f = 36 (Kr), 45 (Rh), and 54 (Xe) were particularly analyzed because of their critical role in quantifying competition between symmetric and asymmetric fission modes. These yields serve as direct indicators of the evolving fission mechanism. Furthermore, the fission product ^{135}Xe has a substantial neutron absorption cross section, acting as a reactor "poison" that significantly reduces operational power, making evaluation of its production amount highly important.

[Figure 5: see original paper]. (Color online) The BNN-CY predicted fission charge yields for Z_f = 45 of γ + ^{216}-^{232}Th at excitation energies of 11 MeV (full line) and 14 MeV (dotted line). Comparison of charge yields for the Th isotopic chain in this work (lines) and experimental data from Refs. [30, 32] (solid dots). Squares represent experimental charge yield data at 11 MeV excitation energy, while circular dots represent experimental charge yield data in the 13-15 MeV excitation energy range. The shaded region corresponds to the CI at 80%.

[Figure 6: see original paper]. (Color online) The BNN-CY predicted fission charge yields for Z_f = 36 and 54 of γ + ^{216}-^{232}Th at excitation energies of 11 MeV (red full line and dotted line) and 14 MeV (blue full line and dotted line). Comparison of charge yields for the Th isotopic chain in this work (full lines) and experimental data (solid dots) from Refs. [30, 32]. Squares represent experimental charge yield data at 11 MeV excitation energy, while circular dots represent experimental charge yield data in the 13-15 MeV excitation energy range. For clarity, CIs are not shown in these BNN-CY results.

Figure 5 presents the evolution of yields for fission fragments with Z_f = 45 (Rh) as a function of target mass number. The observed systematic decrease in Rh fragment yields with increasing target neutron number clearly demonstrates the diminishing contribution of symmetric fission channels. This trend culminates in near-zero fragment charge yields for ^{230}-^{232}Th, establishing asymmetric fission as the dominant mechanism in heavier thorium isotopes, while symmetric fission prevails in lighter Th isotopes. Higher charge yields were observed at 11 MeV excitation energy in heavier Th isotopes compared to the 13-15 MeV range. Figure 6 presents the evolution of fission fragment yields for Z_f = 36 (Kr) and 54 (Xe) as a function of target mass number. The BNN-CY results successfully reproduce the symmetry characteristics of charge yield distributions. Experimental data reveal a perfectly symmetric charge yield distribution where Kr and Xe fragment yields are identical, a feature accurately reproduced by BNN-CY predictions. Moreover, the charge yields of fragments representing asymmetric fission show a systematic increase with the neutron number of the target nucleus, with a more pronounced growth rate observed at 14 MeV excitation energy. These results provide clear evidence for the transition from symmetric fission in neutron-deficient thorium isotopes to asymmetric fission in their heavier counterparts.

C. Mass chain yield distribution

The BNN-CY prediction results in Sec. III B indicate that the fission mechanism of ^{232}Th is dominated by asymmetric fission. In this section, to explore the energy dependence of the mass yield distribution of fission fragments, the BNN-MY model was constructed based on experimental mass yield data in the EXFOR database, which includes 771 data points within an energy range of 8-80 MeV for PNF products of ^{232}Th [37-44]. The mass yields of ^{232}Th fission fragments were measured by Naik et al. [39-44] using the activation method. The charge distribution exhibits a pronounced odd-even effect, whereas this effect appears obscured in the mass distribution. Consequently, there is no need to introduce δ parameters as input corrections for mass yield data. The network consists of input variables x_i = {A_{fi}, Z_i, N_i, E_i}, which denote the mass number of fission fragments, the charge and neutron numbers of the fissioning nucleus, and the incident γ energy (E_i), respectively. The output data t_i is the mass yield of fission fragments. We compared a single-layer network with 38 neurons and a double-layer network with 18-20 neurons, performing 10^5 BNN-MY sampling iterations. The standard deviations obtained for the single-layer and double-layer networks were 6.4$×10^{3}$ and 3.8$×10^{3}$, respectively. As shown in Fig. 7, the CI of the double-layer network is significantly narrower than that of the single-layer network. The double-layer network successfully reproduces the peak structures, whereas results from the single-layer network appear smoother. Based on these considerations, we selected the double-layer network as the more suitable model for further data evaluation in this study.

[Figure 7: see original paper]. (Color online) The mass yield distribution of fragments from γ-induced fission of ^{232}Th at different energies. Comparison of one-layer (blue lines) and two-layer (pink lines) BNN-MY learning results of fragment mass yields of γ+^{232}Th in the γ energy range of 8 MeV to 80 MeV from Naik et al. [39–44]. The shaded region corresponds to the CI at 80%.

[Figure 8: see original paper]. (Color online) The excitation function of the mass yield of photonuclear fission fragments within the incident γ energy range of 5 MeV to 80 MeV. The predicted results of BNN-MY (A_f = 91 as red line and A_f = 140 as blue line) are compared with experimental data for the light asymmetric peak (A_f = 91 as square points) and the heavy asymmetric peak (A_f = 140 as circle points) [37–44]. The shaded region corresponds to the CI at 80%.

The results in Fig. 7 show that fission yield exhibits a monotonic increase with energy in the valley region (fragment mass number A_f 100-130), whereas the twin peaks corresponding to asymmetric fission modes diminish in prominence; the same conclusion is shown in Fig. 8. Generally, the fragment yields at A_f = 91 and 140, representing asymmetric fission, increase gradually within the incident γ energy range of 5-40 MeV. However, when the γ energy exceeds 40 MeV, the yield decreases significantly rather than continuing to increase. In 2010, Demekhina et al. [28] compared PNF fragment mass yields of ^{232}Th at γ energies of 50 MeV and 3500 MeV, observing that symmetric fission dominated as energy increased, with its contribution increasing by almost an order of magnitude. Moreover, due to additional neutron emission from the fissioning nucleus under higher excitation energy, the mass numbers of fission fragments decrease, leading to a leftward shift in the mass-yield distribution. Figure 9 displays the BNN-MY evaluated mass yield distributions of ^{232}Th PNF fragments at incident γ energies of 8 MeV, 25 MeV, and 80 MeV. It can be seen that the yield of symmetric fission (A_f = 100-130) increases with energy. In contrast, contributions from the light fragment peak (A_f = 80-100) and the heavy fragment peak (A_f = 130-150) decrease progressively, representing the symmetric fission mechanism. This suggests that with increasing excitation energy of the nucleus, particle evaporation (predominantly neutron emission) from the excited nucleus opens a new decay channel leading to the formation of neutron-deficient fission fragments (primarily through symmetric fission) [45, 46]. However, no obvious phenomenon of the light fragment peak moving toward lower mass numbers was observed. This indicates that the PNF mechanism of ^{232}Th changes with incident γ energy: in the low-energy region, the PNF mechanism of ^{232}Th mainly involves asymmetric fission, while symmetric fission dominates in the high-energy region.

[Figure 9: see original paper]. (Color online) The BNN-MY predicted fission mass yield of γ+^{232}Th at γ energies of 8 MeV, 25 MeV, and 80 MeV. For clarity, CIs are not shown in these BNN-MY results.

[Figure 10: see original paper]. (Color online) The BNN-MY predicted fission mass yield of γ + ^{232}Th at γ energies of 7.64 MeV and 17.5 MeV, where experimental data for these specific reactions were not included in the training dataset of BNN-MY. Comparison of mass yields for the ^{232}Th nuclide in this work (full lines) and experimental data (solid points) from Refs. [47, 48]. The shaded region corresponds to the CI at 80%.

Figure 10 evaluates the predictive capability of BNN-MY using a test set that excludes PNF fragment mass yield data for ^{232}Th at γ energies of 7.64 MeV and 17.5 MeV. Comparison between predicted results and experimental measurements [47, 48] shows that BNN-MY can effectively predict mass yields of PNF fragments for unstudied reactions.

IV. SUMMARY

Nuclear fission is an extremely complex non-equilibrium quantum many-body dynamical process, and gaining deeper insights into fission remains a well-recognized challenge in nuclear physics. There remains strong motivation for studying nuclear fission, driven by expanding nuclear applications in energy production and rare isotope generation, as well as in fundamental physics domains including superheavy element synthesis and constraints on the r-process. This work presents the relationship between yield and target mass number and the yield-energy correlation of fission fragments of interest, enabling an energy-dependent two-dimensional distribution of fission yields. The results reasonably reflect the evolution of fission modes with increasing energy.

In lighter-mass fission nuclei, charge yields of PNF fragments tend to be symmetric. In heavier-mass regions, charge yields begin to exhibit asymmetric components that gradually dominate, most notably in isotopes of Th and Pa. The proton number of PNF fragments carries information about scission directly. Experimentally, it has been observed that production of PNF fragments with even proton numbers is typically enhanced, which is one of the most prominent features of fission fragment yields. To describe the odd-even effect in the reaction system, we added an additional input $\delta = \$\pm\$0.1$ in the BNN-CY learning collection to represent even and odd fragments, respectively. Uncertainty decreases after adding the odd-even effect, reflected in the PNF fragment charge distribution of the BNN-CY model, which also shows a regular transition from symmetric fission of lighter Th isotopes to asymmetric fission of heavier Th isotopes. The fission mechanism of fissile nuclei evolves with the excitation energy of the reaction system. The mass distribution of PNF fragments predicted by the BNN-MY model indicates that asymmetric fission prevails at low excitation energies, whereas symmetric fission becomes the dominant mode at higher energies. For ^{232}Th, asymmetric fission dominates at incident γ energies below 40 MeV, while the contribution of symmetric fission gradually increases at energies above 40 MeV. Furthermore, no leftward shift in fragment mass distribution was observed within the γ energy range of several tens of MeV.

The BNN serves as a powerful and reliable tool for predicting photofission fragment yields. It has successfully revealed the evolution of fission mechanisms and associated odd-even effects, providing critical theoretical support for nuclear databases and reactor design. Currently, numerous facilities have begun conducting research on PNF reactions, such as the High Intensity γ-ray Source (HIγS) [49–51], GSI [33, 52, 53], and Extreme Light Infrastructure Nuclear Physics (ELI-NP) [54–56]. Building upon the theoretical work presented in this paper, future photofission experiments could be carried out using laser Compton scattering (LCS) γ rays at the Shanghai Laser Electron Gamma Source (SLEGS) at the Shanghai Synchrotron Radiation Facility (SSRF), which provides monoenergetic γ beams.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (Nos. 12375123 and 12247126), and the Natural Science Foundation of Henan Province (No. 242300421048).

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