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RPPR Final Report
as of 21-Oct-2022

Agency Code: 21XD

Proposal Number: 71017PE **Agreement Number: W911NF-17-2-0178**
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DUNS Number: 555456995 EIN: 470049123
Report Date: 30-Sep-2022 Date Received: 12-Oct-2022
Final Report for Period Beginning 01-Oct-2017 and Ending 30-Jun-2022
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Begin Performance Period: 01-Oct-2017 **End Performance Period:** 30-Jun-2022
Report Term: 0-Other

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 2 **STEM Participants:** 8

Major Goals: Goals of the project were divided into four phases, as follows:
Phase I: Optimize x-ray characteristics using photon counting spectroscopy, and to design and implement a system to study photoexcitation of selected isomers as proof-of-principle demonstrators with narrowband x-ray beams.
Phase II: Improve system sensitivity by expanding tests of isomer population to smaller integral cross sections.
Phase III: Preliminary scan of depletion levels for 108mAg, 166mHo, etc. in the 0.1 – 1 MeV range by tuning the energy of the x-ray source while maintaining narrow bandwidth.
Phase IV: Perform detailed scans of depletion levels for selected isomers. And resolve long-standing mystery for depletion of 180mTa near 1 MeV, e.g. inability of standard nuclear spectroscopic experiments to observe back-decay transitions from apparent depletion levels, by scanning across depletion levels with the narrow-line ICS coupled with in-beam measurement of emitted gamma rays

Accomplishments: We investigated the efficacy of laser-driven x-ray radiation for the study of transitions responsible for isomer population or depletion via photon-induced reactions. We achieved this by utilizing a PW-class Ti:sapphire laser system to generate pulsed electron beams with 100 pC charge and energy of 200 MeV that can then generate high energy x-rays by bremsstrahlung emission. The benchmarking of this concept was performed by comparing the photoexcitation of isomeric states in indium isotopes to well-accepted results. The laser-driven isomer irradiation (LDII) scheme was then applied to perform the first known photo-depletion of the promising energy-storage candidate 166mHo.
After the successful benchmarking experiment, we investigated the depletion of 166mHo (m). This isomeric state is typically found as a byproduct in the creation of the radiotherapy isotope of 166gHo (g) through thermal neutron capture by naturally occurring 165Ho (n). If a sample containing both m and g states is allowed to cool for a time much longer than the ground state half-life (on the order of weeks), the remaining radioactivity results solely from the decay of the m state, which is used as an x-ray calibration source. To study the isomeric depletion, we exposed two similar samples of HoCl3 laser-generated bremsstrahlung, one which we reference as “isomeric” that contained a small fraction of isotopes of the m state and another sample which we reference as “natural” that only contained 165Ho. This scheme used relativistic electrons generated by the process of laser-wakefield acceleration, and the interaction of these electrons with a high atomic number (high Z) material to produce high-energy x-rays through the bremsstrahlung mechanism. In this specific irradiation experiment, the goal was to determine whether energy could be released from the long-lived (T1/2 ~1200 year) 166m Ho isomer using these all-laser-driven x-rays. Signatures of a successful depletion of the isomeric state consisted of a prompt measurement looking for

characteristic x-rays emitted directly after the exposure, and a post-irradiation measurement looking for the beta-decay of the ground state of 166Ho.
The experimental results show depletion of the isomeric state of 166Ho is initiated by the absorption of a photon with an energy of 257.8 keV to excite the known depletion level (other unknown depletion levels would also certainly be accessed by the high-energy bremsstrahlung). Accounting for shot-to-shot fluctuations in the electron beam energy, and assuming a pulse duration equivalent to the laser pulse duration, we estimate a spectral flux of 7.3 ± 3.6 x 1017 photons/pC/keV/mm2/sr/s at the known depletion energy for a 2-mm thick Pb converter. While most of the states along the depletion pathway toward the ground state following decay of the 263.8-keV level have half-lives on the order of ns, the Jπ=3+ state has a much longer half-life of 185 us and emits a 136.6 keV x-ray. To isolate this signature, we looked for near-prompt x-rays using the CdTe detector and used the radiation pulse generated by the laser to mark the initial radiation time (t0), as discussed above. Measurements were performed under three distinct conditions: (1) with the isomeric target, (2) with the natural target, and (3) with no target present. In total, 3860 laser shots were taken throughout four separate experimental runs, with an average beam charge of 22.5 pC and an energy of 200 MeV, generating bremsstrahlung with a 200 MeV endpoint. Figure 1(b) shows the difference in signal between the isomeric and natural samples. From this result, the points following the camera dead-time (Figure 1(b-inset)) can be fit to extract a measured half-life of 238 ± 83/112 us which agrees well with the expected half-life of 185 us. Another x-ray transition is observed at 88.6 keV along with the 136.6 keV, with T1/2 -71.4 us as shown in Figure 1 (c). Theoretical relative intensities of these two transitions are expected to be around 1 out of 7000. However, their intensity is observed similar in the experimental results.

Training Opportunities: Throughout the project undergraduate and graduate students in STEM had the opportunity to enhance their skills and knowledge in isomeric nuclear studies. Their performance on tasks of this project supported their professional and academic development. Additional trainings were provided to all students over the course of the project in the areas of high-intensity laser systems, laser-wakefield acceleration, lab safety, and manuscript development for publications.

Results Dissemination: During the project, team members were invited to present at the 2020 Fall Meeting of the APS Division of Nuclear Physics. Team members also presented at the 2021 Nuclear Photonics Meeting.

Presentation:
Applications of a Novel Laser-Driven X-ray source to Nuclear Science
D. Haden, S. Banerjee, J. Carroll, C. Chiara, C. Fruhling, G. Golovin and D. Umstadter
2nd International Conference on Nuclear Photonics, June 24-29, Brasov, Romania, 2018

Papers Published in Peer Reviewed Journals
Golovin, G., Yan, W., Luo, J., Fruhling, C., Haden, D., Zhao, B., … Umstadter, D. (2018). Electron Trapping from Interactions between Laser-Driven Relativistic Plasma Waves. Phys. Rev. Lett., 121, 104801. doi:10.1103 /PhysRevLett.121.104801

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: Faculty
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Person Months Worked: 6.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Tianna Barth
Person Months Worked: 3.00 **Funding Support:**

Project Contribution:
National Academy Member: N

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Project Contribution:
National Academy Member: N

Participant Type: PD/PI
Participant: Donald Umstadter
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Technician
Participant: Shao Qin
Person Months Worked: 2.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Undergraduate Student
Participant: Benjamin Mattingly
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Project Contribution:
National Academy Member: N

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National Academy Member: N

Participant Type: Technician
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Project Contribution:
National Academy Member: N

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Project Contribution:
National Academy Member: N

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Participant: Bhavya Muvya
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Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Rafeeq Syed
Person Months Worked: 1.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Junzhi Wang
Person Months Worked: 4.00 **Funding Support:**
Project Contribution:
National Academy Member: N

RPPR Final Report
as of 21-Oct-2022

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Title: Research and Applications of Laser-Driven-Radiation
Authors: Daniel, Haden
Acknowledged Federal Support: **Y**

Partners

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I certify that the information in the report is complete and accurate:
Signature: Donald Umstadter
Signature Date: 10/12/22 2:21PM

Report documentation page

Period covered by the report: October 01, 2017 – June 30, 2022

Proposal Title: Controlled release of energy stored in nuclear isomers by laser-driven x-rays

Contract/Grant number: 71017-PH

Agreement number: W911NF-17-2-0178

Author(s) of the report: M. Ashiq Fareed and Donald Umstadter

Performing organization Name(s) and Address(es): Board of Regents of the University of Nebraska Lincoln, 2200 Vine Street, box 830861, Lincoln, NE 68503

Abstract

Controlled release of substantial amounts of stored energy is important for the instantaneous global operations of the United States Army. Sources of chemically stored energy are currently limited by maximum amount of stored energy, cost, complexity, and safety hazard. Controlled release of energy from nuclear isomers has been proposed as a viable alternative. In this concept, specific nuclei are excited to long-lived metastable isomeric states. When the nuclei are irradiated with x-rays, the stored energy can be released. The isomeric state becomes excited to a different level, from which it decays to an intermediate or ground state. The major advantage of this approach is that the energy storage capacity of a nuclear system is potentially a million-fold higher than a chemical system, due to comparatively higher energy-excitation levels (MeV versus eV). A primary goal of the project is to perform measurements of the decay of nuclear isomeric states from excited levels with laser-driven x-rays in the 0.1-1.0 MeV spectral range. In collaboration with nuclear scientists from the Army Research Laboratory, we use a compact high-power laser system located at the University of Nebraska-Lincoln (UNL).

Submissions or publications under ARO sponsorship

- "Research and Applications of High-Intensity-Laser-Driven Radiation," Daniel J. Haden, ISBN: 9798535572284, (University of Nebraska PhD Thesis, 2021) http://libproxy.unl.edu/login?url=https://www.proquest.com/dissertations-theses/research-applications-high-intensity-laser-driven/docview/2572607961/se-2?accountid=8116.
- "Transient Relativistic Plasma Grating to Tailor High-Power Laser Fields, Wakefield Plasma Waves, and Electron Injection," Qiang Chen, Dominika Maslarova, Junzhi Wang, Shao Xian Lee, Vojtech Horný, and Donald Umstadter, Phys. Rev. Lett. 128, 164801 (2022) https://doi-org.libproxy.unl.edu/10.1103/PhysRevLett.128.164801.
- "Laser-Driven Photoexcitation of 115*m* In and 166*m* Ho," Daniel J. Haden et. Al., *Review of Scientific Instruments* (to be submitted).

Student/Supported Personnel Metrics for this reporting period

Number of Undergraduates STEM students

1

Number of Graduate STEM students

7

Number of students that received a STEM degree

2

Technology Transfer

N/A

Scientific Progress and Accomplishments

Summarize the overall goals/milestones of the project

We investigated the efficacy of laser-driven x-ray radiation for the study of transitions responsible for isomer population or depletion via photon-induced reactions. We achieved this by utilizing a PW-class Ti:sapphire laser system to generate pulsed electron beams with 100 pC charge and energy of 200 MeV that can then generate high energy x-rays by bremsstrahlung emission. The benchmarking of this concept was performed by comparing the photoexcitation of isomeric states in indium isotopes to well-accepted results. The laser-driven isomer irradiation (LDII) scheme was then applied to perform the first known photo-depletion of the promising energy-storage candidate ^{166m}Ho.

After the successful benchmarking experiment, we investigated the depletion of ^{166m}Ho (m). This isomeric state is typically found as a byproduct in the creation of the radiotherapy isotope of ^{166g}Ho (g) through thermal neutron capture by naturally occurring ^{165}Ho (n). If a sample containing both m and g states is allowed to cool for a time much longer than the ground state half-life (on the order of weeks), the remaining radioactivity results solely from the decay of the m state, which is used as an x-ray calibration source. To study the isomeric depletion, we exposed two similar samples of $HoCl_3$ laser-generated bremsstrahlung, one which we reference as “isomeric” that contained a small fraction of isotopes of the m state and another sample which we reference as “natural” that only contained 165Ho.

What was completed this year

The major milestone of this reporting period was data analysis, understanding results, and writing manuscript of the experimental run using laser-driven irradiation scheme developed last year at Extreme Light Laboratory. This scheme used relativistic electrons generated by the

process of laser-wakefield acceleration, and the interaction of these electrons with a high atomic number (high Z) material to produce high-energy x-rays through the bremsstrahlung mechanism.

In this specific irradiation experiment, the goal was to determine whether energy could be released from the long-lived ($T^{1/2}$ ~1200 year) 166m Ho isomer using these all-laser-driven x-rays. Signatures of a successful depletion of the isomeric state consisted of a prompt measurement looking for characteristic x-rays emitted directly after the exposure, and a post-irradiation measurement looking for the beta-decay of the ground state of 166Ho.

The experimental results show depletion of the isomeric state of ^{166}Ho is initiated by the absorption of a photon with an energy of 257.8 keV to excite the known depletion level (other unknown depletion levels would also certainly be accessed by the high-energy bremsstrahlung). Shown in Figure 1(a), and accounting for shot-to-shot fluctuations in the electron beam energy, and assuming a pulse duration equivalent to the laser pulse duration, we estimate a spectral flux of 7.3 ± 3.6 x 10^{17} photons/pC/keV/mm2/sr/s at the known depletion energy for a 2-mm thick Pb converter. While most of the states along the depletion pathway toward the ground state following decay of the 263.8-keV level have half-lives on the order of ns, the J^{π}=3+ state has a much longer half-life of 185 μs and emits a 136.6 keV x-ray.

To isolate this signature, we looked for near-prompt x-rays using the CdTe detector and used the radiation pulse generated by the laser to mark the initial radiation time (t_0), as discussed above. Measurements were performed under three distinct conditions: (1) with the isomeric target, (2) with the natural target, and (3) with no target present. In total, 3860 laser shots were taken throughout four separate experimental runs, with an average beam charge of 22.5 pC and an energy of 200 MeV, generating bremsstrahlung with a 200 MeV endpoint. Figure 1(b) shows the difference in signal between the isomeric and natural samples. From this result, the points following the camera dead-time (Figure 1(b-inset)) can be fit to extract a measured half-life of 238 ± 83/112 μs which agrees well with the expected half-life of 185 μs.

Another x-ray transition is observed at 88.6 keV along with the 136.6 keV, with $T^{1/2}$ ~71.4 μs as shown in Figure 1 (c). Theoretical relative intensities of these two transitions are expected to be around 1/7000. However, their intensity is observed similar in the experimental results (Figure 1(c)). This is probably because of the limited time resolution of our x-ray camera. An x-ray camera with a faster time scale can resolve these structures accurately.

Summary of progress

- Experiment involving the irradiation of ^{166m}Ho sample using laser-generated bremsstrahlung x-rays
- Data analysis involving the prompt signature of isomer depletion using a CdTe detector and post-irradiation using gamma and beta particle detectors.

- Invited presentation at the *2020 Fall Meeting of the APS Division of Nuclear Physics* (http://meetings.aps.org/Meeting/DNP20/Session/LA.3).
- Presentation at *2021 Nuclear Photonics Meeting* (https://www.photon.osaka-u.ac.jp/NP2020Kurashiki/index.html#program)
- Preparation of manuscript for Review of Scientific Instruments
- Successful completion of a PhD program by Daniel Haden

Data from experiments

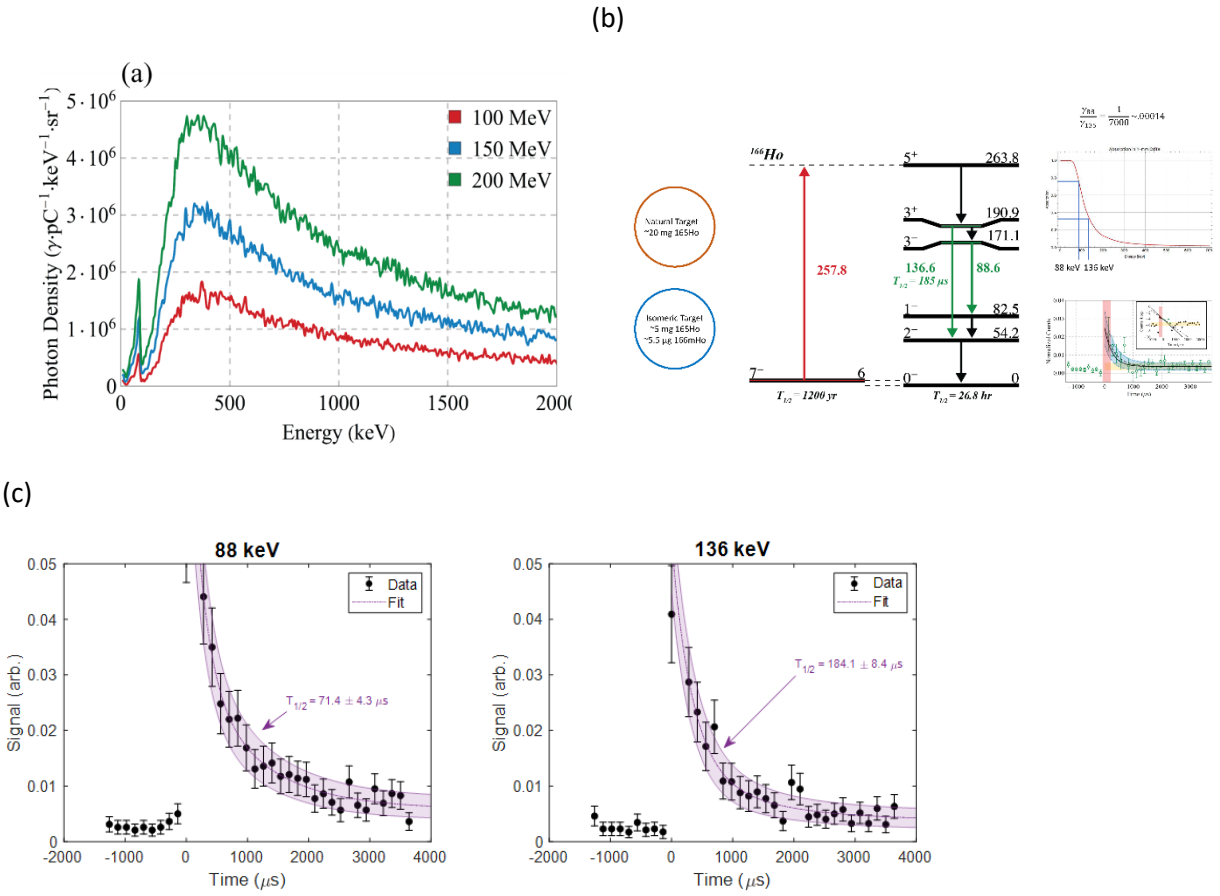

Figure 1: (a) Simulated x-ray spectrum used to irradiate the holmium target. (b) Level diagram for the depletion of the 166mHo 7- isomer at 6 keV and Integrated difference between natural and isomeric holmium targets at the resonance energy of 136 keV. (b-inset) Fitting parameters of the points following the radiation pulse. (c) Comparison of x-ray transitions at 88 keV and 136 keV.

Novelty of results

- The work accomplished over this year establishes a new method for photoexcitation-based perturbation of isomeric nuclear states using laser-plasma accelerator.

- The single-pulse irradiation scheme affords the ability to “cleanly” excite population/depletion pathways without exciting intermediate states that have lifetimes > ns.
- The results from the prompt measurement potentially demonstrate the first ever induced depletion of ^{166m}Ho.

Plans for the coming year

- Reduction of x-ray source bandwidth and increase in x-ray spectral brightness.
- Use Geant4 Monte Carlo simulations to study irradiation of other atoms and molecules with laser-generated bremsstrahlung x-rays.
- Expand this irradiation technique to general atoms and molecules for broader applications.

Training for students

Transition replacement graduate student.

Future of the ARO project after 2022

Experiments demonstrating advantage of ultrashort duration x-ray pulses for isomer depletion with improved x-ray source.

Copies of technical reports

Laser-Driven Photoexcitation of ^{115m}In and ^{166m}Ho

Daniel Haden,[1] James J. Carroll,[2] Christopher J. Chiara,[2] Sudeep Banerjee,[1,3] Donald Umstadter[1]

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Abstract: *Laser-driven accelerators can generate high-peak-brightness x-rays with energies relevant to nuclear processes. Nuclear isomers have been identified as potential candidates for future energy-storage applications, but the ability to extract energy in these states remains elusive. In our work, we investigate the efficacy of laser-driven radiation for the study of transitions responsible for isomer population or depletion via photon-induced reactions. We achieved this by utilizing a PW-class Ti:sapphire laser system to generate pulsed electron beams with 100 pC charge and energy of 200 MeV that can then generate high energy x-rays by bremsstrahlung emission. The benchmarking of this scheme was performed by comparing the photoexcitation of isomeric states in indium isotopes to well-accepted results. The laser-driven isomer irradiation scheme was then applied to perform the first known photo-depletion of the promising energy-storage candidate ^{166m}Ho. These results, paired with cutting-edge advancements in the generation of high-peak brightness x-rays using lasers, can enable the future of energy storage in nuclear isomers.*

I. INTRODUCTION

Nuclear isomers have long been proposed [1–3] as an application for long-term energy storage due in part to their ability to store energy in a long-lived state that is not the ground state of the nucleus. Over 2400 isomeric states with lifetimes >10 ns have been identified [4], and, of those states, only 106 have lifetimes on the order of hours and, further, only 12 known states have lifetimes on the order of years (see Table 1 of [1] plus ^{210}Bi [5]). The most extreme example of energy storage in an isomeric state is the case of ^{180m}Ta, which has a lifetime >4.5x10^{16} years [6] and exists at an energy level that is 77.2 keV above the ground state [5]. While this energy seems ripe for harvesting, the ability to selectively release the energy from the nucleus through switching the isomer population to the ground state remains a challenge that currently prevents its usage in practical applications. For example, experiments have shown that photon-induced depletion of ^{180m}Ta can be achieved, but requires a minimum excitation energy of ~ 1 MeV [7,8].

Isomeric states can exist due to three types of barriers to their decay [9]: (1) "spin isomers", where the decays (isomeric transitions) are characterized by selection rule prohibited changes in the total angular momentum of the nucleus along with low transition energies; (2) "*K* isomers", where there is a large change in the projection *K* of the total angular momentum on a symmetry axis of the nucleus; and (3) "shape isomers" that arise from transitions between local minima in the potential energy surface of the nucleus corresponding to different shapes. Selection rules are typically used to express the inhibition of spin and K isomers to lower-lying states. Direct irradiation [1], nuclear excitation by electronic transition (NEET) [10], and nuclear excitation by electron capture (NEEC) [10,11] are all methods that have been studied as potential pathways to initiate switching of isomers. We note that early claims of isomer depletion of ^{178m2}Hf have not been supported by rigorous, sensitive measurements (see [12] and references therein).

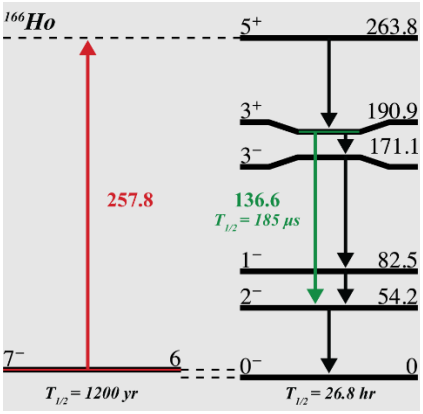

FIG 2. Level diagram for the depletion of the ^{166m}Ho 7$^-$ isomer at 6 keV [13]. Half-lives < 5ns are not labelled, and all energies are listed in keV. The initial ^{166m}Ho state can be excited to the $J^\pi=5^+$ state by absorbing a 257.8 keV photon. There is a non-zero branching ratio of the decay of this state that enables a series of transitions that end at the ground state: Thus, the state at 263.8 keV can serve as a depletion level, the excitation of which can lead to transfer of population from isomer to ground state.

The ^{166m}Ho isomer is a very long-lived ($T_{1/2}$ = 1200 yr) spin-isomer with $J^\pi=7^-$ that is at 6 keV above the ground state. This particular isomer is very interesting for energy storage purposes, not only because it has a very long half-life but also because its ground state is naturally unstable. Instead of only the energy difference between the isomer and ground state being released, more energy can be obtained from the subsequent beta decay of ^{166}Ho, with energies of up to 1.85 or 1.77 MeV (via 48.8% or 49.9% decay branches to the ^{166}Er ground or first-excited states, respectively), and a half-life of 26.8 hr. Full depletion of the isomer followed by ^{166g}Ho beta decay would provide a power density of 7.7 x 10^4 W/g, compared to .57 W/g for ^{238}Pu. A candidate direct irradiation depletion pathway for ^{166m}Ho was identified by Carroll et al. [5] and is shown in FIG 2. Unfortunately, previous isomer depletion via direct photon irradiation [7,8,14], which might be explored for ^{166m}Ho, have relied on bremsstrahlung produced from RF linear accelerators with accelerating gradients of a few MeV/m; the physical size of such facilities would seem to limit their practical application for the release of isomer energy that might need to be portable.

We then turn to laser-plasma accelerators (LPA). LPA's have been identified as strong candidates for applications involving nuclear interactions ranging from the use of nuclear resonance fluorescence (NRF) [15,16] for the identification of materials to photodissociation using (γ,xn) and (γ,f) reactions for the transmutation of nuclear waste [17,18]. This is because they can generate high-peak-brightness (>10^{20} photons s^{-1} mm^{-2} $mrad^{-2}$ 0.1% BW^{-1}) x-rays with energies >1 MeV using bremsstrahlung [17,19] and inverse-Compton scattering (ICS) [20–24] techniques. Such a feat is accomplished by first producing low-emittance, relativistic electron bunches using laser-wakefield acceleration (LWFA), [25] followed by the perturbation of these electron bunches by either the introduction of a high-Z material in the case of bremsstrahlung or by a colliding laser field in the case of ICS. This is all done in a small device footprint thanks to the GV/cm acceleration gradient achieved by LWFA. Because of these advantages, LPA-driven x-ray sources are also promising candidates for direct isomer photoexcitation and photodepletion experiments. The ICS radiation is particularly attractive as it is pseudo-monoenergetic, which would serve as a more precise source of irradiating photons compared with the broad spectrum of bremsstrahlung, albeit not so precise as the 10^{-4} bandwidth typical from monochromatized synchrotron radiation at much lower energies. The narrow spectral width would enable isomer measurements similar to those described in Ref. [26], yet in a much more compact setup due to the LPA approach. Furthermore, the polarization of the ICS radiation [27,28] and the ability to generate short pulse duration, MeV x-rays [29] can provide new capabilities for the investigation of new excitation/depletion schemes [30,31].

This paper will present the laser-driven isomer irradiation (LDII) scheme to show the population or depletion of isomers. To start, we will describe the experimental layout and the measurement techniques. Following that, we will present results that benchmark this method by showing the successful population of isomeric states in naturally occurring indium and comparing it to known measurements. Then we will show the potential of this method to investigate new systems by showing the first evidence of photo-induced isomer depletion in holmium by accessing the depletion scheme depicted in FIG 2. Finally, we will conclude by discussing the implications of advancements in the LDII scheme to improve future isomeric transition triggering.

II. METHODS

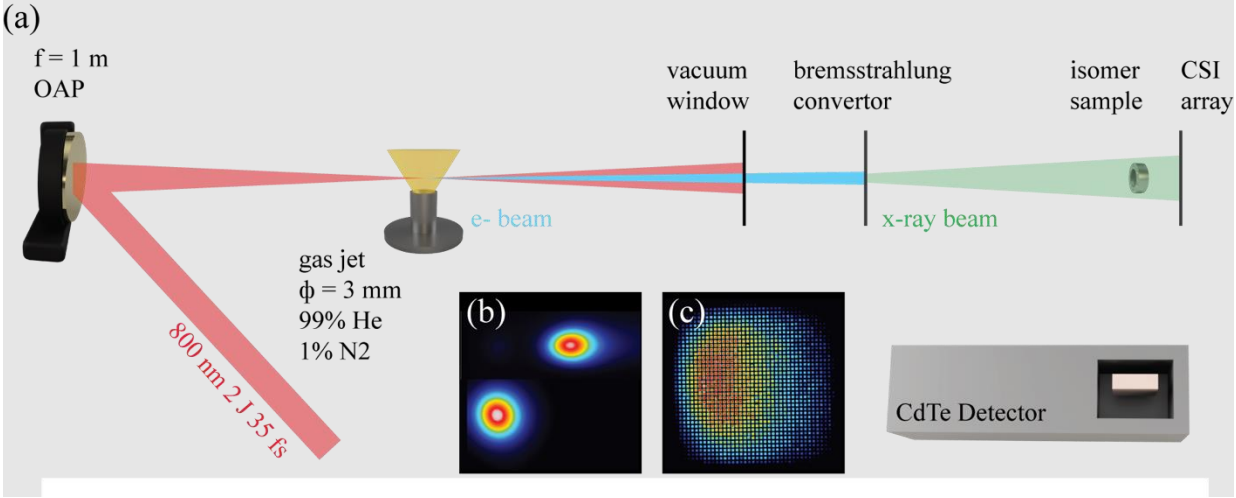

FIG 3. (a) MeV x-ray source driven by a Ti:Sapphire laser system which produces 200-MeV electrons by the process of laser-wakefield acceleration and converts the electron beam to an x-ray beam by transmission through a bremsstrahlung converter. Relativistic electrons are deflected by a 0.8 T permanent dipole magnet (not pictured) after the converter so that no direct impact between the electron beam and the sample occurs. A CdTe detector placed off-axis from the beam measures the photon flux from the sample post-irradiation. (b) Typical undeflected and deflected electron beams as captured by the Lanex imaging system. (c) Bremsstrahlung x-ray spatial profile measured by the CsI(Tl) imaging system.

FIG 3 shows the general layout for LDII. In this experiment, the Diocles [32] laser in the Extreme Light Laboratory at the University of Nebraska-Lincoln was used to produce 200 MeV electrons by LWFA. In this acceleration scheme, 2 J of 800-nm laser light, with a pulse duration of 35 fs, was focused using a 1 m off-axis paraboloid (OAP) to a 15 μm (FWHM) spot size into a gas target. The gas target consisted of a 3 mm diameter supersonic nozzle driven by a pulsed valve with an open time of 20 ms and was backed by a gas mixture of 99% He, 1% N_2 at pressures ranging from 50 – 150 psi. Under optimal conditions, we could generate up to 200 MeV electron beams with a total charge of ~100 pC in each pulse. These low-divergence, high-charge, high-energy electron beams were then propagated outside of the vacuum chamber through an 8 mm thick glass window.

The relevant geometries after the beams exit the chamber varied depending on three modes of operation; electron beam charge (1) and energy (2) measurements and sample irradiation (3). For beam charge measurements, a Kodak fast Lanex screen is placed in the beam and imaged by a 12 bit CCD (example shown in FIG 3(b)), for which the charge/signal has been calibrated. To obtain the beam energy, we used a 0.8 T, 10 cm long permanent dipole magnet with the exit of the magnet placed 30 cm from the Lanex imaging system used in mode (1) to disperse the beam according to its energy. Finally, the sample

irradiation mode starts with the components of mode (2) and adds a second Lanex imaging system for charge monitoring and a bremsstrahlung converter before the magnet. After the magnet, the isomer target is placed (~1.5 m from the electron beam source), followed by a CSI(Tl) array imaged by a 16 bit, high-gain electron multiplying charge coupled device (EMCCD) to monitor the x-ray beam profile and fluence (FIG 3(c)).

Post-irradiation analysis was performed by two different methods: a near-prompt x-ray measurement using a pixelated CdTe detector and delayed γ/β measurements where the sample is moved to a separate room to count for multiple days. The latter of these methods is the less complex, where high-purity germanium (HPGe) detectors were used to measure γ-rays with energies in the range of 0.05-2 MeV, and a thin CaF_2 crystal coupled with a photomultiplier tube (PMT) was used to measure β particles up to 3 MeV. Samples were transferred to the counting room <10 minutes after the last beam shot (from a sequence of shots to increase the total photon fluence on the samples), and data were acquired in 15-minute intervals for a total time of 72 hours.

The near-prompt measurement consisted of a HEXITEC CdTe detector [33,34] placed off the beam axis at a distance of 5-10 cm from the isomeric sample. This system was chosen in order to avoid the effects of the electromagnetic pulse associated with the laser-plasma interaction that typically contributes to a large deadtime in HPGe and NaI detectors. The sensitive detector material in this device is a CdTe crystal that is 1 mm thick and has 80x80 pixels with a size of 250x250 μm^2/px that can measure photons with energies in the range of 4-200 keV. We operated this detector in the free-run mode at an acquisition frame rate of 7.154 kHz, where 1 frame is the collection of the information from each pixel across the entirety of the array. This frame rate is equivalent to a duration of 140 μs/frame.

III. RESULTS

The population of isomeric states of ^{115}In and ^{113}In

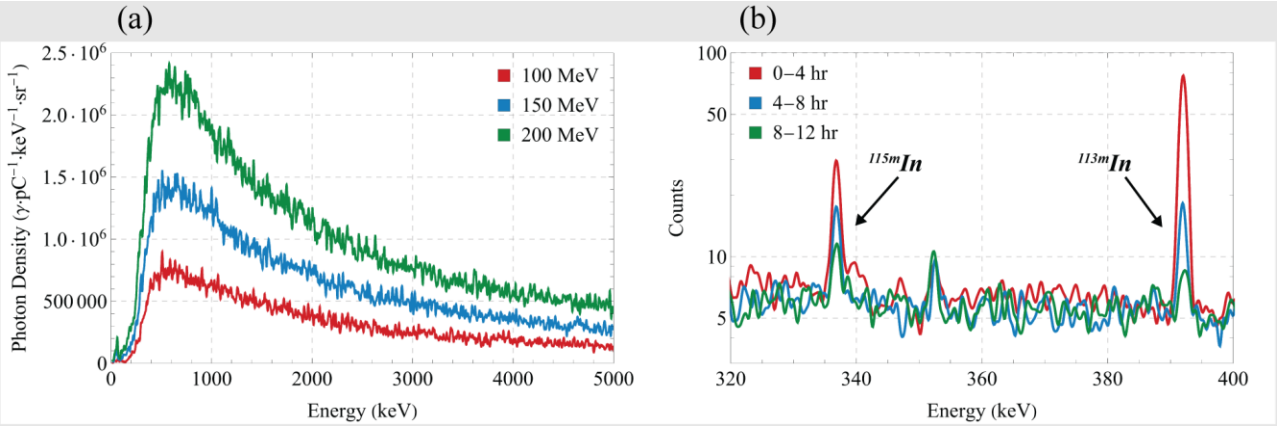

FIG 4. (a) Bremsstrahlung x-ray photon fluence calculated in Geant4 for various electron beam energies used in the indium exposure. (b) Post-irradiation gamma-ray spectrum measured with the HPGe detector in consecutive four-hour incremental periods, showing the decay of ^{115m}In and ^{113m}In following their known half-lives.

To benchmark the LDII method, we turn first to the study of indium. Indium has two naturally occurring isotopes, ^{115}In and ^{113}In, with abundances of 95.71% and 4.29%, respectively. Both isotopes have long-lived isomeric states. In ^{115}In, this is the $J^{\pi}=1/2^-$ state at 336.2 keV, which has a half-life of 4.49 hr , and in ^{113}In the isomer is the $J^{\pi}=1/2^-$ state at 391.7 keV with a half-life of 1.66 hr. The photoexcitation of the ^{115m}In isomer, first discovered in 1935 by Szilard and Chalmers [35], has been well studied since this process was first measured in 1939 [36]. Recent studies, which include Belic et al. [37] and Tornow et al. [26], have precisely measured the ^{115}In(γ,γ') ^{115m}In cross-section for photoexcitation energies ranging

from 1.8-3.7 MeV, showing resonant excitations with resolution typical for bremsstrahlung and ICS irradiations, respectively.

The large energy range of the excitation energies in combination with the available range of electron beam energies led us to choose a 6 mm tantalum bremsstrahlung converter to produce a high photon density in this region. From the Geant4 simulations shown in FIG 4(a) we estimate the bremsstrahlung photon spectral flux across the excitation energies to range from $2.2 \pm 0.9 \times 10^{19}$ γ/pC/keV/sr/s at 1.8 MeV, to $1.1 \pm 0.7 \times 10^{19}$ γ/pC/keV/sr/s at 3.7 MeV. This source was used to irradiate a 4.74 g, 99.99% pure indium foil folded into a target with dimensions of 10.5 mm x 10.75 mm x 6.8 mm. The geometry was chosen to maximize the absorption of the x-rays along the long axis during the exposure and to minimize the self-absorption along the short axis for the post-exposure measurements.

A total of 406 shots with an average beam charge of 19.7 pC were taken on the target at a repetition rate of ~ 0.1 Hz, for a total exposure time of 1.6 hr. For this exposure, the shots were taken with the sample inside the vacuum chamber, so it took approximately 20 min to vent the chamber and move the sample to the counting room. In Fig. 3(b), the gamma-ray energy spectra recorded with the HPGe detector clearly indicate the emission of the 336.2 keV and 391.7 keV isomeric transitions in ^{115}In and ^{113}In, respectively, with their yields decreasing with time according to decay rates that corresponded with the literature values for the isomer half-lives. [5,26] Thus, the benchmarking of this laser-driven system for the photoexcitation of isomers has been successfully demonstrated. The integrated net counts (above background) for the two peaks in the first 30 min of counting were 84 and 209, respectively. After correcting for detector efficiency and the collection angle, this corresponded to a rate of incident gamma-rays of 0.39 and 1.10 Hz. The use of such broad-band irradiation from bremsstrahlung having an endpoint $\geq$ 100 MeV would produce isomer population, in different proportions, by photon absorption through all of the known [26] and likely unknown intermediate states. Some isomer population could also be due to other reactions.

The depletion of ^{166m}Ho

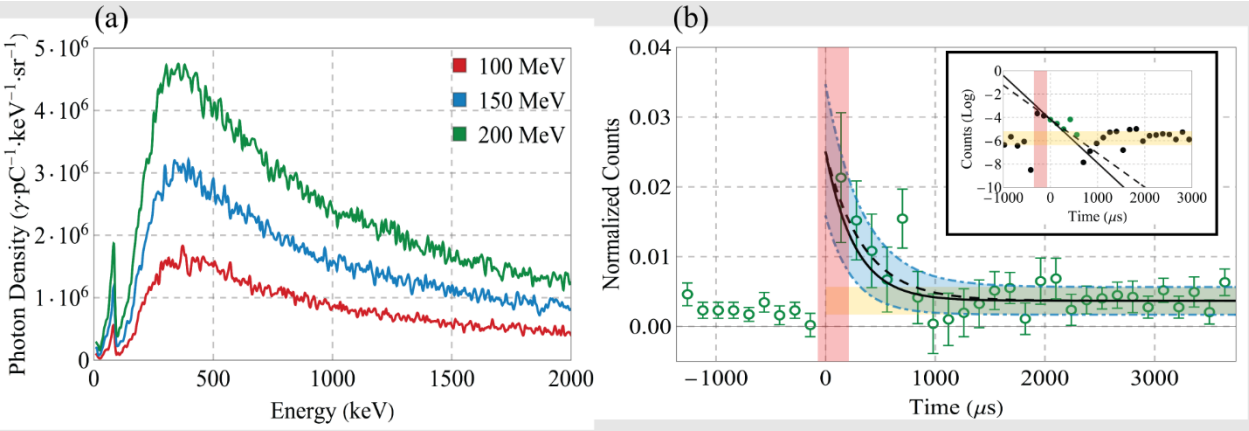

FIG 5. (a) Bremsstrahlung x-ray photon fluence from a Pb converter calculated in Geant4 for various electron beam energies used in the holmium exposure. (b) Integrated difference signal at 136 keV for the isomer compared to the natural target as a function of time, with T_0 established as described earlier: The red band indicates the dead-time of the detector due to the high count rates from background radiation, the yellow band is the long-lived background level that the sample relaxes to, the blue bands indicate the extrema of the fit of the half-life, the black dashed line is the half-life taken from the fit of the log-scale data (inset), and the solid black line is the expected half-life for the decay of the $J^{\pi}=3^{+}$ state.

After the successful benchmarking experiment, we then investigated the depletion of ^{166m}Ho (*m*). This isomeric state is typically found as a byproduct in the creation of the radiotherapy isotope of ^{166g}Ho (*g*)

through thermal neutron capture by naturally occurring ^{165}Ho (*n*). If a sample containing both *m* and *g* states is allowed to cool for a time much longer than the ground state half-life (on the order of weeks), the remaining radioactivity results solely from the decay of the *m* state, which is used as a γ-ray calibration source. To study the isomeric depletion, we exposed two similar samples of $HoCl_3$ laser-generated bremsstrahlung, one which we reference as "*isomeric*" that contained a small fraction of isotopes of the *m* state and another sample which we reference as "*natural*" that only contained the *n* isotope.

The *isomeric* sample that we used was created at the Missouri University Research Reactor (MURR) in 2010 by the exposure of a solution of ^{165}Ho by a thermal neutron flux of ~$3x10^{14}$ n/cm^2/s for one week. [38] By knowing these details of the exposure, the initial mass of *n* in the sample, the final activity of the sample (2.2 Ci/mg) dominated by the decay of *g*, and the thermal neutron capture cross-sections from $n \rightarrow g$ and $n \rightarrow m$ ($\sigma_{n\rightarrow g} \sim 60$ b, $\sigma_{n\rightarrow m} \sim 3.4$ b [39–41]), we can estimate that this yielded a post-irradiation solution of $HoCl_3$ with the percent mass ratio of the Ho species being 99.46% *n*, 0.43% *g*, and 0.11% *m*. This solution was then allowed to cool for several weeks; once the *g* decayed away, the solution was dripped onto a 500 μm thick Be disk where the drop was allowed to evaporate, and the process was repeated until 10 μCi of activity was achieved. The source was sealed by placing another Be disk on top and then secured in an Al housing. Using the measured activity of *m* and the total age of the sample (~10 years), the total masses of each species of holmium can be calculated and are shown in Table 1. For comparison, we enclosed powdered *natural* $HoCl_3$ in Al foil and sealed the target in an Al housing nearly identical to the housing used for the *isomeric* target. The total mass of the chloride in the *natural* target was measured to be 33 ± 5 mg, which results in 19.8 ± 3.0 mg of ^{165}Ho therein, a factor of 3.9 ± 0.6 times the amount in the *isomeric* target.

Table 1: Mass of the isotopes of holmium present in the *isomeric* sample after the sample was initially prepared in a neutron flux trap.

Species of Ho	Mass
166m	5.5 μg
166g	22.0 ± 0.4 μg
165	5.1 ± 0.1 mg

The depletion of the isomeric state of ^{166}Ho is initiated by the absorption of a photon with an energy of 257.8 keV, as shown in FIG 2, to excite the known depletion level (other hitherto unknown depletion levels would also certainly be accessed by the high-energy bremsstrahlung). To access the known pathway with laser-generated bremsstrahlung, we again used Geant4 to choose the ideal converter material type and thickness, to maximize the peak spectral brightness near the absorption energy. The result of the simulations are shown in FIG 5(a), and accounting for shot-to-shot fluctuations in the electron beam energy and assuming a pulse duration equivalent to the laser pulse duration, we estimate a spectral flux of 7.3 ± 3.6 x 10^{19} γ/pC/keV/sr/s at the known depletion energy for a 2 mm thick Pb converter. FIG 2 also displays the states along the depletion pathway toward the ground state following decay of the 263.8-keV level. Though most of those states have half-lives on the order of ns, the $J^{\pi}=3^{+}$ state has a much longer half-life of 185 μs and emits a 136.6 keV γ-ray.

To isolate this signature, we looked for near-prompt γ-rays using the CdTe detector and used the radiation pulse generated by the laser to mark the initial radiation time (t_0), as discussed above. Measurements were performed under three distinct conditions; (1) with the *isomeric* target, (2) with the

natural target, and (3) with no target present. In total, 3860 laser shots were taken throughout four separate experimental runs, with an average beam charge of 22.5 pC and an energy of 200 MeV, generating bremsstrahlung with a 200 MeV endpoint. The distribution of those shots is shown below in Table 2.

Table 2: Total number of shots taken on each type of target

Target	# of shots
isomeric	1737
natural	1428
no target	695

The performance of the CdTe detector in the laser-generated radiation environment is heavily affected by a significant background signal that comes from radiation scattered into the detector from every object that the beam hits. We estimate the duration of this background signal to be no longer than 280 μs (or 2 frames). During sample exposure, the acquisition window was 300 s which was enough time to produce 40-50 radiation shots on target. Each shot was isolated in the data in post-processing by identifying the frame in which the significant background signal was present. After identifying the exposure frame t_0, the total spectrum as a function of the frame (within a window of 35 frames around t_0) was recorded for each shot, and a 5x5 charge sharing addition algorithm [42–45] was applied to correct for the small-pixel effects in the camera. From this point, spectra from all shots are summed across their corresponding frames, where further integration at specific energy windows is performed.

In each of the measurement cases, the integration window had 5 keV width centered around 136.6 keV and the results were plotted as a function of time for each shot. Then, the results from all laser shots were added together and normalized to the total number of shots for each specific target. FIG 5(b) shows the difference in signal between the *isomeric* and *natural* samples. From this result, the points following the camera dead-time (FIG 5(b-inset)) can be fit to extract a measured half-life of 238 ± 83/112 μs which agrees well with the expected half-life of 185 μs.

IV. FUTURE OUTLOOK

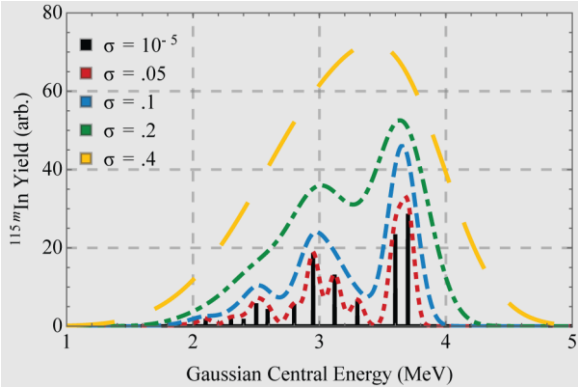

FIG 6. ^{115m}In yield for a narrow-band laser-driven x-ray source as a function of central beam energy and spectral width σ.

This LDII scheme is dependent on bremsstrahlung radiation, which is a relatively inefficient process as only a small fraction of the photon yield comes at the energy(ies) of interest, within narrow absorption bandwidths. Current advances in the stability of LWFA and the all-optical generation of narrow-band x-rays by inverse-Compton scattering can increase the relative efficiency for the utilization of irradiating photons. Of course, the total number of photons/laser pulse is much less for inverse-Compton

scattering vs. bremsstrahlung. For example, consider the case where the x-ray spectrum from inverse-Compton scattering has an ideal Gaussian distribution. In this situation, the central energy and spectral width can be adjusted to match the transition energy requirements for absorption by an intermediate state for isomer population or a depletion level for isomer depopulation. An example of this is shown in FIG 6 for the population of ^{115m}In, though it should be noted that this is using an unnormalized Gaussian distribution to simply show the shape of the yield curve as a function of central energy. If instead, the total photon number is kept constant and the spectral width is reduced, it is logical that the efficiency will increase nonlinearly to the limit of the width of the resonance. In the simplified example of ^{115m}In, for an x-ray beam with central energy of 3.6 MeV, the yield is increased by a factor of 3x when the width is reduced from 40% to 5%. This potential to achieve high-efficiency photon sources capable of isomer depletion through precise photoexcitation of nuclei within a small device footprint like the LDII scheme presented in this work can open up the future of energy storage and release with nuclear isomers.

V. CONCLUSION

This paper has presented a scheme for photoexcitation of nuclei using an all laser-driven bremsstrahlung x-ray source. After the successful benchmarking of this method by photoactivating the isomers in the well-studied systems of ^{113m}In and ^{115m}In, we then successfully applied this method to ^{166}Ho, where we demonstrated for the first time, by this or any technique, the induced depopulation (depletion) of its long-lived 7^- isomer.

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