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Enhanced Control of Nuclear Lifetimes for Ultra-Energetic Materials Using Electron- Nuclear Processes

by James J Carroll and Christopher J Chiara

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Enhanced Control of Nuclear Lifetimes for Ultra-Energetic Materials Using Electron- Nuclear Processes

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14. ABSTRACT Radioisotopes can store 100,000 times the energy of chemical batteries or fuel cells for durations of many decades or longer. The ability to utilize such ultra-energetic materials for Army power and energy applications would be a truly revolutionary advance. With very long storage lifetimes, the output power achievable with radioisotopes is limited, unless metastable nuclear excited states (isomers) can be used to provide different time scales for energy storage and release. US Army Research Laboratory mission research is underway to investigate methods by which to switch certain radioisotopes from long-lived to short-lived states via direct interactions with nuclei. Supplemental to that program, this special Director's Research Initiative project investigated the possibility of an indirect method via atomic-nuclear interactions, which has been proposed to enhance the efficiency of such switching under specific conditions. The process of nuclear excitation by electron capture was studied in a series of experiments based on a nuclear fusion-evaporation reaction in inverse kinematics. A full analysis is not yet complete, but initial positive indications suggest this approach has provided the first evidence of this new physical effect.					
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1. Introduction

The ability to supply energy as needed is the underlying enabler of everything the Army does and everything the Army could potentially do. At present, the storage and delivery of energy rely heavily on chemical means through fuels or chemical batteries. These materials are available, well-characterized, mass-produced, and transportable; they are ubiquitous within the Army. However, their uses are limited by intrinsic properties of energy storage lifetime and density. As seen in Fig. 1, the specific energies of chemical batteries and fuel cells lie at about 100–1,000 Wh/kg while fuels like gasoline (not shown in the figure) reach the order of 10,000 Wh/kg. These values are governed by the form in which the energy is stored (i.e., in chemical bonds with bond energies on the order of 1–10 eV^{*}). The duration of energy storage is essentially determined by the breakdown of constituent chemical bonds as molecules or compounds degrade spontaneously, or due to interaction with the ambient conditions (e.g., temperature). The energy can be released for usage on a relatively short time scale so that chemical batteries can deliver power at up to about 1,000 W/kg with fuels typically having lower specific power output. One may hope that in the future, advances will increase the energy density of chemical batteries and fuels, their longevity, and their power outputs, but it is most likely that those advances will be more evolutionary than revolutionary in scope. Ultimately, the limitations of chemicals are inherent to the physics underlying the energy storage.

^{*} An electron volt, 1 eV = 1.6×10^{-19} J.

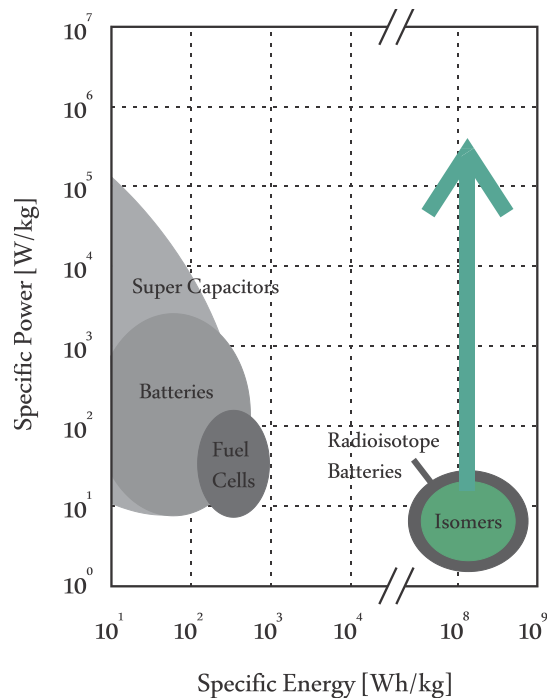


Fig. 1 Specific power vs. specific energy for selected materials (adapted from D'Auria [1])

It is already possible to conceive of Army applications that would benefit from energy storage that greatly exceeds that possible with chemical batteries or fuels. One specific example is that of unattended ground sensors, as discussed in Refs. 2 and 3, which should function for long periods without having to replace the batteries. It has already been recognized that the mission duration for such devices could be extended too many decades by utilizing the energy-storage properties of radioisotopes. As shown in Fig. 1, radioisotopes store energy at specific densities that are on the order of 10^8 Wh/kg, or more than 100,000 times that available from fuel cells. This is simply due to the fundamental storage unit, the atomic nucleus, with energies of 10,000–1,000,000 eV (10 keV–1 MeV). Perhaps more importantly for batteries, the time scale for energy storage can be extremely long. The rate at which radioisotopes release stored energy is given by their characteristic decay half-lives* (e.g., 12.32 years for tritium, 87.7 years for ^{238}Pu , and 100.2 years for ^{63}Ni [4]). Atomic nuclei are well-isolated from the environment by the atomic electrons so that their half-lives are generally insensitive to ambient conditions. A tritium-based radioisotope battery could, in principle, provide power for an unattended ground sensor for many decades; US Army Research Laboratory (ARL) mission-supported research is underway to develop tritium and ^{63}Ni batteries for Army applications. ^{238}Pu is already utilized for deep space probes, for which the limited sunlight would make solar panels infeasibly large.

* A half-life, $T_{1/2}$, is the time required for half of the nuclei to decay, releasing their stored energy.

For the radioisotopes usually considered for energy storage, there is, however, a drawback: there is only one time scale available for both storage and use, so the longevity of radioisotope energy is accompanied by a low specific power density (see Fig. 1). It is the case that for “traditional” radioisotopes, energy applications are based on the decay of the nuclear ground state by transmutation into other elements via charged particle decay (i.e., β^- or α emission). The emitted charge particle leads to electricity by energy deposition in some conversion material. However, all nuclei possess many states of different half-lives at different internal degrees of excitation. In most instances the half-lives of excited nuclear states are quite short, on the scale of 10^{-9} s or less. However, a surprising number of nuclei possess one or more states with much greater half-lives. Two particular examples are ^{178}Hf , with an excited state having $T_{1/2} = 31$ years [4] and storing 3.6×10^8 Wh/kg, and ^{180}Ta , with an excited state having $T_{1/2} > 7.1 \times 10^{15}$ years* [5] and storing 1.5×10^7 Wh/kg. The ^{178}Hf ground state is stable, while that of ^{180}Ta is 8.2 h. When a nucleus possesses multiple states with very long half-lives, it becomes possible to envision applications relying on long energy storage and then a transfer of nuclei into a state of shorter half-life for use. This process is depicted in Fig. 1 for “Isomers”, showing the gains in specific power that can, in principle, be achieved by transitioning a suitable nucleus from a long-lived energy-storing state into a shorter-lived energy-releasing state. One can envision such a process as enabling radioisotopes to be stored in an essentially dormant state and then activated when needed for an Army application. In the extreme, such transitions may support the development of directed energy applications for highly penetrating gamma radiation. In reality, the ability to exert this type of control over nuclear decays would be likely to enable entirely new applications that cannot be imagined today and usher in a revolutionary advance in energy and power for the Army.

ARL mission research is underway into direct mechanisms for transitioning nuclei from long-lived to shorter-lived states (i.e., when the transition is induced via direct excitation of the nucleus using externally produced photons, particles, or ions). Under some instances, an indirect mechanism may prove advantageous but has not yet been demonstrated for any nuclide.† The goal of this Director’s Research Initiative (DRI)-supported project is to provide the first demonstration of such an effect.

* The half-life of that excited state of ^{180}Ta is so long, greater than the age of the universe, that its decay has never been observed.

† A nuclide is a species of atomic nucleus, characterized by a specific atomic number, Z , and atomic mass number, A .

2. Scientific Background

2.1 Nuclear Isomers

Atomic nuclei are dynamic systems consisting of microscopic nucleons (neutrons and protons) whose motion is governed by quantum mechanics. Similar to atomic electrons, nucleons move in a central potential* in quantum orbitals and pair with antiparallel intrinsic spins within those orbitals. The energy, total angular momentum, and parity of each nucleon in its orbital are quantized so that the total energy, angular momentum (J)[†] and parity (π) of the nucleus as a whole are also quantized as a combination of the properties of the constituent nucleons. Different from atomic electrons, the strong nucleon-nucleon interaction leads to additional collective effects. For example, the outer “valence” nucleons may influence the overall nuclear shape by perturbing that of a central “core” so that many nuclei have static spheroidal shapes with axial symmetry. Quantum mechanics then permits quantized collective rotations of the entire nucleus about an axis perpendicular to the symmetry axis. Oscillations of the nuclear shape can also occur, which can be described as quantized vibrations on the nuclear surface. Ultimately, the quantum states of the nucleus may be a combination of orbital (so-called “intrinsic”) and collective motions (for a full discussion, see Casten [6]). A nucleus may possess many different states of excitation, each characterized by quantized energy, angular momentum, and parity.

Each nuclear state is also characterized by a half-life, which is determined by the available decay modes to other nuclear states. For excited nuclear states these decays are typically electromagnetic, wherein a gamma-ray[‡] photon is emitted as the nucleus makes a transition to a different state. Another common decay mode for excited states is that of internal conversion, where nuclear excitation energy is

* The true central potential binding electrons to the atom is due to the Coulomb attraction to the positively charged nucleus, while nucleons move in a central potential of their own creation resulting from the combination of their Coulomb repulsion (protons only) and the near-neighbor attraction (protons and neutrons) due to the strong nuclear force. The combined attractive potential influencing a nucleon is often approximated as a mean field due to the other nucleons, although this neglects some tensor aspects of the nucleon-nucleon force. Because the mean fields experienced by protons and neutrons are different, the nucleus as a whole is often considered as a combination of two largely independent quantum systems for the 2 types of nucleon.

[†] Strictly speaking, the angular momentum quantum number is J and the magnitude of the angular momentum vector is given by $\hbar\sqrt{J(J+1)}$. For convenience, the colloquial usage in nuclear physics is adopted, where the quantum number is referred to as the angular momentum.

[‡] In nuclear physics, the term *gamma ray* is restricted to a photon that is emitted from within a nucleus, due to a nuclear deexcitation. The term does not refer simply to a photon having an energy within a certain range, since it is well-known that some gamma rays have energies as low as to fall within the vacuum ultraviolet range.

carried away by an atomic electron that is ejected from the atom. It is worthwhile to note that internal conversion is a coupled atomic-nuclear process in which the nucleus deexcites by emitting a virtual* photon that is absorbed by the atomic electrons; an electron is then ejected with a kinetic energy given by the difference between the nuclear excitation energy lost and the electron's binding energy prior to ejection. The overall half-life of a particular nuclear excited state corresponds to the inverse of its overall decay rate, which is the sum of the decay rates for all possible decay modes.

The nuclear ground state, the lowest energy state for the nucleus with its specific set of nucleons, may also decay. This is not a result of a rearrangement of motions of the constituent nucleons as for decay of excited states. Instead, it occurs when the proportion of protons to neutrons for a nucleus is not optimum for balancing the attractive (strong nuclear) and repulsive (Coulomb, between protons) forces. When a nucleus has too many neutrons, a neutron will spontaneously decay into an electron, a proton, and an electron antineutrino; the combination conserves charge and other properties of the original neutron. The antineutrino and electron are ejected from the nucleus and atom, carrying energy lost in the decay. This is beta decay; the ejected electron is called a β^- particle for historical reasons. The final nucleus is a different element since the atomic number has increased by 1, while the total number of constituent nucleons has been preserved. The final nucleus may be left in an excited state after the decay so that gamma rays and conversion electrons may subsequently be emitted.

The rates at which nuclear excited states may transition by absorption or emission of gamma rays (electromagnetic radiation) are governed by standard physics principles, but quantitative agreement between measurements and theory is severely lacking. A quantum mechanical transition rate is determined from the matrix element between initial and final nuclear states, mediated by an electromagnetic operator. The present state-of-the-art in theoretical approaches can only provide wavefunctions for nuclear states that describe the bulk properties with reasonable accuracy (general description of the nuclear motion, energy, angular momentum and parity). However, transitions between states represent an overlap between the tails of wave functions that are not well described theoretically. Thus, calculated transition rates between nuclear states can differ by many orders of magnitude from those measured experimentally. As a general rule of thumb, the Weisskopf estimates were developed and assume a very simplified approach: a single nucleon moving in a specific simple potential changes orbital to cause the nuclear transition. Also, the electromagnetic operator is expanded in the standard

* A virtual photon is one that is unobservable outside the system.

multipole expansion for electric and magnetic transition types. The key results are as follows (see deShalit and Feshbach [7]):

- Each nuclear transition is characterized by a change in angular momentum, ΔJ , and change in parity, $\Delta\pi$, determined by the angular momenta and parities of the initial and final nuclear states. Selection rules describe the multipolarity, L , of the allowed transitions for a given angular momentum and parity change. For any transition $L \geq \Delta J$, electric transitions require $\Delta\pi = (-1)^L$ and magnetic transitions require $\Delta\pi = (-1)^{L+1}$. The lowest allowed multipole dominates the transition rate so that a particular transition would be characterized by that multipolarity. For example, a transition between states with $J^\pi = 1^-$ and $J^\pi = 0^+$ would be termed an electric dipole ($E1$) transition, since $\Delta J = 1$ and $\Delta\pi = -1$ (a change of parity).
- Electromagnetic transition rates, λ^{EM} , are proportional to a multipolarity-dependent power of the transition energy so that rates for decays with large L are suppressed relative to rates for lower ΔJ . Also, rates for magnetic transitions are suppressed by a factor of roughly 1/100 relative to electric transitions, for the same multipolarity and for a medium-mass nucleus. A few examples are as follows:

a. Electric dipole ($E1$) radiation:

$$\lambda^{EM}(E1) = (1.03 \times 10^{14} \text{ s}^{-1}) A^{2/3} E_\gamma^3, \quad (1)$$

where A is the atomic mass number of the nucleus and E_γ is the energy of the transition given in MeV.

b. Electric quadrupole ($E2$) radiation:

$$\lambda^{EM}(E2) = (7.44 \times 10^7 \text{ s}^{-1}) A^{4/3} E_\gamma^5. \quad (2)$$

c. Magnetic dipole ($M1$) radiation:

$$\lambda^{EM}(M1) = (3.19 \times 10^{13} \text{ s}^{-1}) E_\gamma^3. \quad (3)$$

Thus, for $A = 100$ and $E_\gamma = 0.1$ MeV, $\lambda^{EM}(E1)/\lambda^{EM}(E2) = 6.4 \times 10^6$ and $\lambda^{EM}(E1)/\lambda^{EM}(M1) = 69.6$. These values only illustrate the general trends: Weisskopf estimates can be orders-of-magnitude different from measured values for transition rates (e.g., collective $E2$ transitions are systematically 100 times larger than the Weisskopf estimates). Experimentally determined measured transition rates are sometimes given, for comparison, in units of the associated Weisskopf estimate (so-called Weisskopf units, or W. u.). Because the lowest term in the expansion of the electromagnetic operator dominates (lowest electric vs.

higher electric, and electric vs. magnetic of the same L), a transition is described as having a specific character of electric dipole, etc. This designation also describes the spatial distribution of gamma rays emitted from identical nuclei undergoing the same transition, analogous to the terminology for antennas.

It is now straightforward to understand qualitatively how some nuclear excited states could have longer half-lives than others. If the angular momentum of a state differed considerably from that of lower-lying states to which it could decay electromagnetically, then the associated transition rates would be suppressed by the large multipolarity—the higher-lying state would then have a relatively long half-life. In some instances this effect can be extreme: for ^{180}Ta , its second excited state has $J^\pi = 9^-$ due to the combination of individual nucleon angular momenta in their orbitals. The ground state has $J^\pi = 1^+$ so that electromagnetic decay from the excited state to the ground state would require $\Delta J = 8$ and $\Delta\pi = -1$ by emission of a photon with extremely low transition rate on the order of 10^{-41} s^{-1} . The rate of decay by internal conversion may be estimated to be more than 10^{19} times larger than the rate of gamma-ray decay, but the total decay rate is still 10^{-22} s^{-1} . Even with a gram of ^{180}Ta in the $J^\pi = 9^-$ state, one would only expect there to occur less than one decay per second. Although this decay rate is based on the Weisskopf estimate, this still illustrates qualitatively why this excited state of ^{180}Ta would have an exceptionally long half-life: experimentally, its decay has never been observed and its half-life is greater than 7.1×10^{15} years [5].

Long-lived nuclear excited states are called isomers, a term borrowed from chemistry for molecules with different properties due to different configurations of the same constituents. The designation “long-lived” is open for interpretation, with some nuclear physicists applying it to states with half-lives greater than about 10 ns (still long-lived compared with most nuclear states). However, truly metastable states with lifetimes greater than a day are of the greatest interest for potential applications in energy storage. Isomers with half-lives greater than about 0.1 s are typically denoted by a superscripted “m” (e.g., $^{180\text{m}}\text{Ta}$). Presently there are 33 known isomers with half-lives greater than 1 day [8].

2.2 Isomer Depletion

While the decay of nuclear isomers is inhibited by the requirement for large changes in angular momentum, there should be some transitions from higher-lying states to isomeric levels that are more frequent. For example, in ^{180}Ta there is a state* at 280 keV having $J^\pi = 10^-$, which can decay to the $J^\pi = 9^-$ isomer by a 203 keV $M1$ transition. The 280-keV state is known to decay only by this transition, but in

* Half-life unmeasured, probably less than 1 ns.

principle, other excited states could decay not only to the isomer, but alternatively to levels whose subsequent decay would lead to the nuclear ground state. Quantum mechanics dictates that transitions are time-reversible so that if a higher-lying state decays to an isomer by gamma-ray emission, then the higher-lying state could be excited from the isomer by resonant absorption of a photon at the same energy. Thus, a process could occur by which an externally produced photon was absorbed to excite such a state from an isomer, followed by decays from the upper state that lead to the ground state. This process is illustrated schematically in Fig. 2 and is called isomer depletion (or isomer switching), as it causes the number of nuclei in an isomeric level to be reduced. The higher-lying state that serves as intermediary for the process is called a depletion level. Isomer depletion results in the release of the excitation energy of the depletion level, although the net energy gain is only the difference between that energy and the amount input from external sources to reach the depletion level from the isomer. If the ground state is unstable, then further energy can be released through its decay (e.g., through β^- decay).

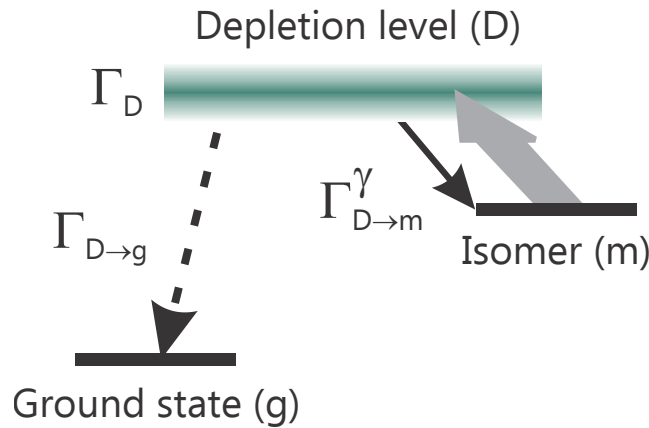


Fig. 2 Schematic depiction of isomer depletion, where a nucleus is initially in an isomer. The upward grey arrow shows a transition from the isomer (m) to a higher-lying depletion level (D), caused by resonant absorption of an externally produced photon. The depletion level may decay back to the isomer or may decay by another path (perhaps requiring multiple decays to lower-lying states) to eventually reach the ground state (g). The Γ s indicate appropriate widths for the states and transitions, as described in the text.

Quantum mechanics dictates that the intrinsic energy uncertainty, $^* \Gamma$, of any state is related to the total half-life of the state due to all decay modes according to

$$\Gamma = \hbar \lambda = \hbar / \tau, \quad (4)$$

* The energy distribution of any state is represented by a Lorentzian, with Γ being its full-width-half-maximum (FWHM).

where $\hbar$ is the reduced Planck constant and $\tau = T_{1/2} / \ln(2)$ is the lifetime of the state, related to its half-life. For the depletion level, its total width would be Γ_D . The absorption of a photon from isomer to depletion level is the inverse of decay by photon emission from the depletion level to the isomer. The partial width corresponding to that gamma-ray transition (the part of the depletion level's width due to gamma-ray decay to the isomer) is denoted by $\Gamma_{D \rightarrow m}^\gamma$. For the decay branch from the depletion level that leads toward the ground state, all decay modes are important (not just the gamma decay mode), with a partial width of $\Gamma_{D \rightarrow g}$. It is assumed that the externally produced photon that initiates the depletion process is part of a broad spectrum, as is typical for bremsstrahlung devices like electron linacs or “x-ray” tubes. In this case, the probability of causing the depletion transition per incident photon per area that is resonant with the absorption transition, and per target nucleus, is given by the integral cross section (ICS) [9]:

$$ICS = \left(\frac{2J_D + 1}{2J_m + 1} \right) \left(\frac{\pi \hbar c}{E_{m \rightarrow D}} \right)^2 \frac{\Gamma_{D \rightarrow m}^\gamma \Gamma_{D \rightarrow g}}{\Gamma_D}, \quad (5)$$

where J_D and J_m are the angular momenta of the depletion level and isomer, respectively, c is the speed of light, and $E_{m \rightarrow D}$ is the energy of the transition between isomer and depletion level. Units of the ICS are typically expressed as eV b.*

Isomer depletion induced by direct nuclear excitation has been investigated, most recently under ARL mission research, by 2 complementary methods:

- Depletion tests, where a sample containing preproduced isomeric nuclei is irradiated. The signal of depletion would be excess energy release from the sample, measured by some radiation detector, and the deduced number of energy-releasing nuclei, N_g , due to isomer depletion to the ground state. The ICS can then be deduced from

$$N_g = N_m ICS \frac{d\Phi(E_{m \rightarrow D})}{dE}, \quad (6)$$

where N_m is the number of nuclei initially in the isomer, and $d\Phi(E_{m \rightarrow D})/dE$ is the spectral flux density of the irradiating spectrum at the appropriate energy to initiate depletion.

* The nuclear unit of barn, b = 10^{-24} cm².

- Spectroscopic studies, where traditional nuclear physics experiments are performed to map the excited states of a nucleus and the transitions between those states. In some instances, it is possible to identify depletion levels from those maps (level schemes) and to measure the angular momenta and widths needed for Eq. 5.

At present, isomer depletion has been demonstrated experimentally for 5 isotopes: $^{180\text{m}}\text{Ta}$ (by bremsstrahlung photons [10] and Coulomb excitation [11]); $^{68\text{m}}\text{Cu}$ (by Coulomb excitation [12]); $^{177\text{m}}\text{Lu}$ (by thermal neutrons [13]); $^{178\text{m}2}\text{Hf}^*$ (by thermal neutrons [14]); and $^{108\text{m}}\text{Ag}$ (by bremsstrahlung [15]). The demonstrations for $^{178\text{m}2}\text{Hf}$ and $^{108\text{m}}\text{Ag}$ were performed as part of ARL mission research. At least partial depletion-level spin, parity, and transition width information has been obtained by spectroscopic measurements for ^{180}Ta [16], ^{68}Cu [4], ^{108}Ag [9], and other nuclides, while ARL mission efforts include the “Rhenium Campaign” of experiments at multiple sites to investigate depletion of the $T_{1/2} = 200,000$ year isomer of ^{186}Re . Considerable research remains to be conducted to determine the physical parameters for depletion of different isomers, prior to any real analysis of those isomers for potential applications.

2.3 Nuclear Excitation by Electron Capture (NEEC)

The generation of photons in the range less than about 100 keV is much simpler and more cost-effective than for photons of hundreds to thousands of keV: lower-energy photons can be created by modest-scale “x-ray” tubes or by lasers. So it seems to be clear that a goal would be to find isomers from which depletion levels can be reached with less than 100-keV excitation by externally produced photons. Unfortunately, this conclusion is overly simplistic. The ICS for isomer depletion depends on many factors, with an obvious $(E_{m \rightarrow D})^{-2}$ dependence seen in Eq. 5. However, the transition widths (and therefore the total level width) are also implicitly energy-dependent, following the general trends given by the Weisskopf estimates. Generally, for a fixed angular momentum difference between isomer and depletion levels, the rate and width for transitions between them are reduced for lower-energy transitions compared with higher-energy transitions. For example, for an $M1$ transition (the most common type in nuclei, corresponding to $\Delta J = 1$ and $\Delta \pi = 1$), the width and rate for a 10-keV transition is 1,000 times smaller than for a 100-keV transition. Calculations have shown [17] that for a number of isomer and depletion-level combinations, an indirect excitation mechanism may be more probable than direct excitation by externally produced photons, when the energy

* ^{178}Hf possesses 2 metastable isomers: the m2 designates the higher-lying of the 2, having a half-life of 31 years.

difference between isomer and depletion level is relatively small. The indirect mechanisms rely on the atomic-nuclear interaction.

It is well known that the interaction between atomic electrons and the nucleus can lead to interesting phenomena. The most straightforward is the “isotope effect”, where an atom can demonstrate different atomic spectral lines from a similar atom with a different isotope of the same element [18]. Similarly, the critical temperature of superconductors differs when different isotopes are used in otherwise identical crystals; this was an important clue that electron-phonon interactions were the key to traditional superconductivity [19]. These phenomena are primarily due to the difference in mass between isotopes, as is the kinetic isotope effect, where chemical reaction rates differ when molecules have different isotopes [20]. The different sizes of different isotopes can also play a role. A shift of atomic spectral lines can also occur in the hyperfine interaction [18] due to the coupling between atomic and nuclear magnetic moments that results from angular momenta of the 2 quantum systems. The atomic-nuclear interaction can also influence nuclear properties, as when the energy for resonant photon absorption by Mössbauer nuclei is shifted (chemical or isomer shift) slightly for atoms residing in different atomic environments [21]. As a final example, the rate at which excited nuclear states can decay by internal conversion can be suppressed when atoms are sufficiently stripped for there to be fewer atomic electrons to couple to the nucleus [22].

Two additional, related atomic-nuclear effects were proposed in the 1970s: Nuclear Excitation by Electron Transitions (NEET) and Nuclear Excitation by Electron Capture (NEEC). In NEET [23], energy is transferred from the atomic system to the nucleus when an electron in an outer orbital fills a vacancy in an inner orbital^{*}; the key point is that the difference in atomic binding energies must closely match that required to resonantly excite the nucleus from one state to another. Similarly to the known process of internal conversion[†] described previously, a virtual photon may be modeled as conveying the energy between atomic and nuclear systems.[‡] The probability for the occurrence of NEET, once a suitable atomic vacancy has been created, is proportional to [24]

^{*} NEET is, therefore, a bound-to-bound process for the atomic system.

[†] NEET is the inverse process to bound internal conversion (BIC), where the electron taking the energy from a nuclear deexcitation is not ejected from the atom but is excited to a higher-energy orbital. An observation of BIC was reported in 2000 (Carreyre T, et al. Phys Rev C. 2000;62:024311).

[‡] Another well-known process, emission of Auger electrons, also proceeds via the exchange of a virtual photon within the atomic system; the energy released when an atomic vacancy is filled is conveyed to another atomic electron, which is subsequently ejected from the atom.

$$\left(1 + \frac{\Gamma_i}{\Gamma_f}\right) \frac{E_{\text{int}}^2}{\Delta^2 + \frac{1}{4}(\Gamma_i + \Gamma_f)^2}, \quad (7)$$

where Γ_i and Γ_f are the widths of the initial and final atomic states, and Δ is the energy difference (“detuning”) between the atomic and nuclear transitions. The “interaction energy” E_{int} is a function of the matrix elements for both atomic and nuclear transitions, for the same multipolarity of the virtual photon, so it contains the same nuclear transition energy dependence as expressed in the Weisskopf estimates. A key point is that the detuning should be very small in order to have a large probability for the NEET process, which means nature must be quite kind in providing atomic states differing by almost the same energy as that required to excite the nucleus.

Various experiments have been performed to investigate NEET (see, for example, Tkalya [24]), with a definitive demonstration of the effect being first reported in 2000 for ^{197}Au [25] and a further refinement to the measurement appearing in 2006 [26]. In both cases, the demonstration was performed using narrow-band synchrotron radiation to produce the initial atomic excitation, and subsequent characteristic nuclear decay radiation was detected to show that nuclear excitation had occurred via NEET. The synchrotron radiation was tuned to an energy of 77.300 keV to generate a K-shell vacancy and thus could not directly excite the nuclear transition requiring 77.351 keV. The detuning $\Delta = 51$ eV in Eq. 7 was indeed quite small, yet it was sufficiently large to prevent realization of the theoretical maximum NEET probability; the probability of NEET for this nuclear transition of ^{197}Au was found to be $5.0(6) \times 10^{-8}$ per K-shell vacancy. With the cross section for creating the required K-shell vacancy about 10^{-21} cm^2 , the overall NEET cross section is then about 10^{-28} cm^2 [25]. In contrast, the cross section for direct nuclear excitation approaches 10^{-27} cm^2 . In this case, direct nuclear excitation is about 10 times more probable than nuclear excitation via electron transitions. NEET has not been demonstrated experimentally for any other nuclide (see, for example, Claverie et al. [27]), reflecting the difficulty in finding the right combination of atomic and nuclear bound states.

In 1976, the complementary process of NEEC was proposed [28] as the inverse of internal conversion. The distinction between NEET and NEEC is fundamentally simple in that in NEEC, the nuclear excitation is caused by energy released in the capture of a free electron rather than a bound electron falling into a lower orbital.[†]

* Errors in measured values are expressed in the style of the Evaluated Nuclear Structure Data File (ENSDF) (National Nuclear Data Center, Brookhaven National Laboratory [accessed 2016 Feb 22]. www.nndc.bnl.gov), where $5.0(6) = 5.0 \pm 0.6$.

[†] NEEC is an unbound-to-bound atomic process.

The importance lies in the freedom this permits for matching the energy of the atomic system to that of the nuclear system. A free electron with precapture kinetic energy E_K may be captured into a specific vacancy, with binding energy E_B , so that the total energy available for NEEC is $E_{NEEC} = E_K + E_B$. The vacancy can, to some degree, be selected by the method used to excite the atom, and the binding energy of a vacancy can be different for different overall charge states of the atom. The free electron's kinetic energy can also be adjusted so that E_{NEEC} can effectively be tuned to match the energy required for the nuclear excitation, E_n . Thus, one might expect that it may be possible to find atomic-nuclear systems for which NEEC is more probable at some energies than direct excitation of the nuclear system. If NEEC led to excitation of a depletion level from an isomer, then the cross section (in effect, the efficiency) for isomer depletion could be enhanced compared with depletion using direct excitation of the nucleus.

This possibility has recently been investigated theoretically in several works [17,29,30]. A number of isomeric nuclei were selected for analysis, including ^{242}Am , and comparisons were made between calculated cross sections for isomer depletion via direct nuclear excitation and by NEEC. The isomer $^{242\text{m}}\text{Am}$ has been discussed previously in the context of induced depletion (e.g., in Refs. 5, 31, and 32), and a major spectroscopic study was reported in 2010 [33]. It has many attractive features [6] from the perspective of potential applications as a nuclear battery: the isomer half-life is 141 years, but its excitation energy above the ^{242}Am ground state is modest at 48.6 keV. A potential depletion level lies only 4.10(6) keV above the isomer, although the transition between that state and the isomer has not been observed. Isomer depletion would populate the ^{242}Am ground state, which has a 16.02-h half-life, decaying primarily by β^- emission that releases 664.5(4) keV. The transmutation leads to ^{242}Cm , which decays with $T_{1/2} = 162.8(2)$ days by alpha-particle emission with an energy release of 6.21556(8) MeV. The ^{242}Cm decay produces ^{238}Pu , with a half-life of 87.7(1) years that decays by alpha-particle emission, with an energy release of 5.59320(19) MeV. The decay chain becomes stalled at ^{234}U , which has a half-life of $2.455(6) \times 10^5$ years. Thus, $^{242\text{m}}\text{Am}$ depletion would result in an overall release of more than 12 MeV of energy over $T_{1/2} \sim 87$ years, with about 7 MeV being released over $T_{1/2} \sim 160$ days. Finally, $^{242\text{m}}\text{Am}$ can be produced with a large cross section by thermal neutrons from reactors [8].

An experimental test of NEEC for depletion of $^{242\text{m}}\text{Am}$ was performed prior to 2012 at the Lawrence Livermore National Laboratory (LLNL), using a scheme designed to take advantage of particular expertise and equipment residing there. For NEEC to occur, it is necessary to create a suitable atomic vacancy and then to have a free electron captured by the atom under the NEEC resonance condition $E_{NEEC} = E_n$. Scientists at LLNL were the first to create a research tool called an Electron Beam

Ion Trap (EBIT) [34], as well as a specialized technique for injecting selected atoms into the trap [35]. An EBIT contains ions electromagnetically, while an internal electron beam produces the ionization of trapped atoms. It seems quite reasonable that such a device would be well-suited to a demonstration of NEEC. In the experiment, ^{242}Am atoms were inserted into the EBIT, but in practice their numbers were relatively small, perhaps on the order of only 10^5 atoms. This was one of the practical difficulties encountered. Perhaps more importantly, it was found that the single electron beam was ineffective in producing the NEEC conditions, in that only one kinetic energy could be selected for the electrons. One beam energy might be optimum for providing a target atomic vacancy but might be inappropriate to match the NEEC resonance. Ultimately, the experiment was terminated without any positive indication of isomer depletion via this atomic-nuclear process [36].

Proposals have appeared by which to demonstrate NEEC using other methods, such as in a laser-produced plasma [30,37,38]. Such an environment would indeed contain highly ionized atoms and a spectrum of electron energies but can be expected to produce strong background radiation due to recombination of the plasma electrons. An attempt was apparently made relatively recently using the PHELIX laser [39] at the GSI Helmholtzzentrum für Schwerionenforschung GmbH (GSI) for some nuclide, without success [40]. Other proposals have been made relying on beam-based approaches (e.g., acceleration of ^{57}Fe ions [isomer $T_{1/2} = 98.3(3)$ ns]) into a solid for further stripping and NEEC, prior to moving on to a detector [37], but an experiment at the Grand Accélérateur National d'Ions Lourds failed to observe the effect as reported in Pálffy et al. [17]. Another beam-based approach [41,42] was proposed for uranium isomers that would use yet another unique facility at GSI, the Experimental Storage Ring (ESR) [43]. Uranium ions would be accelerated into the ESR, passed through a solid for further stripping, and then allowed to interact with an electron beam to produce NEEC. Although proposed in 2008, allocation of beam time at GSI is very challenging; it appears that this experiment has finally been scheduled for May 2016 [44,45].

A novel beam-based approach to the demonstration of NEEC was proposed in 2012 [46], and ARL DRI support was obtained in 2014 with which to pursue this concept. The component development and experiments are discussed in the following sections.

3. Experimental Design

3.1 Approach and Challenges

The original scheme was introduced in 2012 [46] and is founded on a novel combination of techniques used for nuclear reaction and spectroscopic studies. First, atoms moving at high velocity through a medium are slowed in a well-known manner [47] due to Coulomb interactions with the charges within the medium, and are also ionized by those interactions. For a specified velocity within a medium, the projectiles will be characterized by a mean charge state [48], although at any given instant the ions may have a different charge state within a distribution about that mean value. This is due to the nearly constant process of removal and capture of electrons by the ion. The effect finds practical use in accelerators, where projectile ions are passed through thin carbon “stripper” foils [49] in order to increase their charge state for maximum acceleration, given a fixed accelerating potential. The effect occurs in all media, including helium gas [50]. The passage of fast projectiles through a medium is a key aspect in this scheme for providing the necessary atomic vacancies for NEEC.

Second, as projectile ions move in a medium, they see atomic electrons of the medium moving toward them relative to the ion’s proper frame of reference. Thus, as the projectiles slow in the medium, they encounter incident electrons with different (decreasing) kinetic energies for potential capture into vacancies in the projectile ions. Both the mean charge state and relative kinetic energy of approaching electrons will decrease but will cover a wide range of values. It is anticipated that the NEEC resonance condition may be met during the transit of projectiles through the medium. The general behavior of the ion velocity, mean charge state, and E_{NEEC} are shown in Fig. 3.

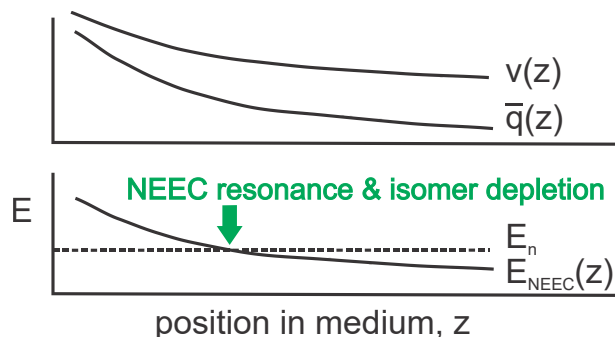


Fig. 3 Qualitative behavior of fast ions in a medium: velocity, $v(z)$, and mean charge state, $\bar{q}(z)$ (top plot) and E_{NEEC} (bottom panel). Isomer depletion would occur when $E_{NEEC} = E_n$, as described in the text.

This scheme requires projectiles with a suitable isomeric nucleus to be injected with high kinetic energy into a medium. The isomer $^{242\text{m}}\text{Am}$, due to its long half-life of 141 years, is available in sample form—such a sample was the target in the experiment described in Hayes et al. [33]. However, its use as a beam is precluded by the concomitant loss of the valuable sample material. There are also few accelerators capable of producing sufficiently energetic beams of such heavy nuclei.

A consideration of the nuclides studied theoretically for NEEC [17] suggested that $^{93\text{m}}\text{Mo}$ would be best matched to the projectile requirements for the proposed experimental scheme. This nuclide has a number of attractive properties [4] for a demonstration experiment (partial level scheme shown in Fig. 4): its isomer* lies at an excitation energy of 2424.95(4) keV and has a half-life of 6.85(7) h; the nucleus is of medium mass, so it and nearby nuclides can be accelerated at a number of facilities; the nuclear structure of ^{93}Mo has been investigated in several works (e.g., Fukuchi et al. [51] and, including quite recently, Hasegawa et al. [52]); and the NEEC probability was estimated [17,46] to be sufficient for observation via depletion of the $^{93\text{m}}\text{Mo}$ isomer and 100 times larger than the probability for direct excitation of the depletion level. Most importantly, the available nuclear structure information identifies a potential depletion transition of only 4.85(9) keV and a gamma-ray transition at 267.93(8) keV that can only occur after population of the isomer if NEEC occurs. That transition is followed by a sequence of rapid transitions, or cascade, that terminates on the ground state having $T_{1/2} = 4.0(8) \times 10^3$ years. Because the half-life of $^{93\text{m}}\text{Mo}$ is 6.85(7) h, it is necessary to produce this isomer near or at the site of NEEC.

* The isomer was discovered in 1950 (Kundu DN, Hult JL, Pool ML. Phys Rev. 1950;77:71).

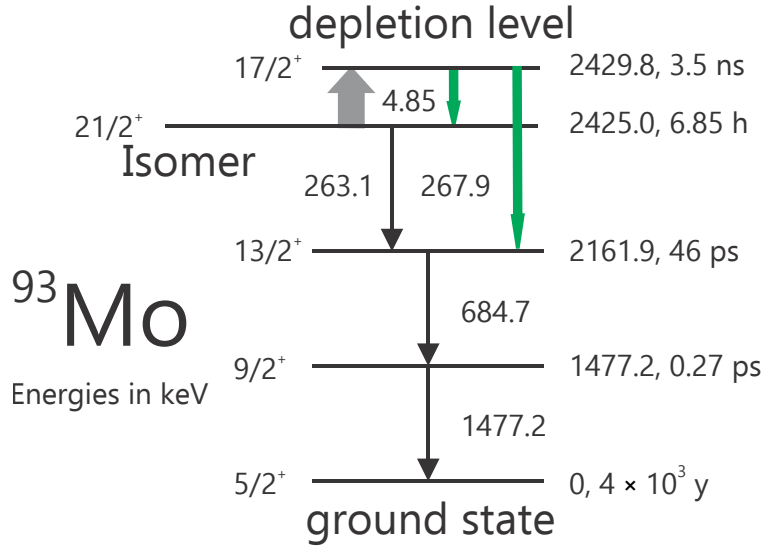


Fig. 4 Partial level scheme for ^{93}Mo [4] (level spacings not to scale). The grey arrow is the potential depletion transition to be excited by NEEC. The solid black arrows show the natural decay cascade from the isomer. The green transitions only occur following NEEC so that the 268-keV gamma ray is a unique signal of the effect (the 4.85-keV transition is likely to completely decay by internal conversion and emit no gamma rays). Angular momentum and parity quantum numbers are given to the left of the levels, and level and transition energies are given in kiloelectron volts (keV).

The third component of the proposed demonstration scheme is in-flight production of $^{93\text{m}}\text{Mo}$ via an inverse-kinematics reaction. Typical nuclear spectroscopic measurements are conducted using gamma-ray detectors to observe decay transitions in highly excited nuclei that are produced by nuclear reactions. It is generally preferred that the gamma rays be emitted without experiencing the Doppler effect. This is because with the Doppler effect, detectors at different angles will measure slightly different energies for identical gamma ray transitions so that when individual spectra are combined from the different detectors, the gamma-ray peaks are broadened and potentially more difficult to identify. For a detector placed at an angle θ with respect to the beam direction, as shown in Fig. 5, the measured gamma-ray energy, E'_γ , is given by [53]

$$E'_\gamma = E_\gamma \frac{\sqrt{1-\beta^2}}{(1-\beta \cos \theta)}, \quad (8)$$

where $\beta = v/c$, with v being the emitting ion velocity, and E_γ being the gamma-ray energy as emitted by a stationary nucleus.

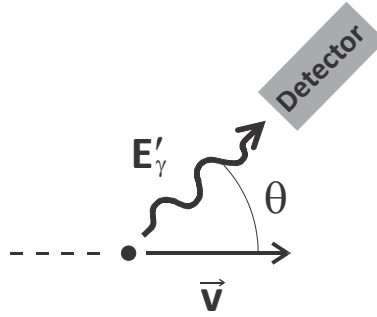


Fig. 5 Gamma-ray emission by a moving nucleus, into a detector located at an angle θ with respect to the beam direction (the example shown is a “forward” angle, with $\theta < 90^\circ$). The velocity vector of the emitting nucleus is $\vec{v}$, and E'_γ is the energy of the gamma ray, including Doppler shift, measured by the detector.

To avoid Doppler shifts for the emitted gamma rays, the nuclear reaction used to produce the excited nuclei is typically performed in “normal” kinematics, where a light ion is accelerated to collide with a heavy target nucleus. Because of conservation of momentum, the final excited nucleus moves with relatively low velocity and quickly comes to a stop in the sample. If this occurs swiftly enough, the gamma rays will be emitted by stopped nuclei and experience no Doppler shift.

For a NEEC test with $^{93\text{m}}\text{Mo}$, it is desired to produce the isomer in a nuclear reaction such that the molybdenum ions retain a high velocity as they move through a medium. This can be accomplished by using a heavy projectile ion incident on a light target nucleus (inverse kinematics) so that momentum conservation will give ^{93}Mo atoms with a large recoil velocity.

The components to the proposed NEEC demonstration experiment may be summarized as follows:

- 1) Molybdenum atoms containing the isotope ^{93}Mo excited to its 6.85-h isomer are produced by a nuclear reaction in inverse kinematics.
- 2) The ions containing $^{93\text{m}}\text{Mo}$, recoiling with high velocity, move through a material and are slowed because of interactions with that stopping medium. The ions are also stripped to a high charge state by those interactions, providing atomic vacancies.
- 3) As the $^{93\text{m}}\text{Mo}$ ions slow, they will capture electrons from the medium with different relative kinetic energies, and into various atomic vacancies of the molybdenum ions. At some point in the ion trajectory within the medium, the conditions suitable for NEEC (i.e., $E_K + E_B = E_{NEEC} = E_n$) may be met.

- 4) NEEC will populate the identified depletion level in ^{93}Mo , causing an energy release toward the ground state that includes a unique gamma ray that is not produced in the natural decay of the $^{93\text{m}}\text{Mo}$ isomer.

The original proposal [46,54] identified the $^4\text{He}(^{91}\text{Zr}, 2\text{n})$ fusion-evaporation reaction^{*†} for production of $^{93\text{m}}\text{Mo}$ using a helium gas target. It was also envisioned that helium gas could serve as the NEEC stopping medium so that there was a certain elegance to the experimental setup, depicted in Fig. 6. Ions of ^{91}Zr would be accelerated to about 6 MeV/A,[‡] or 546 MeV total energy in the lab frame, and enter an extended helium gas cell at standard temperature and pressure (STP). Within the first approximately 15 cm,[§] reactions would produce ^{93}Mo nuclei in its isomer, which would recoil with about 5.5 MeV/A. The well-known Projection Angular-momentum Coupled Evaporation (PACE) fusion-evaporation reaction code [55,56] was used to estimate that about one-third of the ^{93}Mo nuclei would be produced in the isomer, after decay from higher-lying excited states. Taking a cross section of 0.25 b and assuming only 1 particle nA^{**} for the ^{91}Zr beam, $^{93\text{m}}\text{Mo}$ isomers would be produced at a rate of $10^6/\text{s}$. The molybdenum ions will be highly stripped by passage through the gas and exhibit a mean charge as high as +36e [57] near 5.5 MeV/A. For Mo^{36+} , the M_3 ($3p_{3/2}$) subshell (and all outer shells) is vacant such that electrons can be captured into those states. In neutral molybdenum, the M_3 subshell has $E_B = 0.394$ keV [58], but in Mo^{36+} this subshell is more tightly bound and $E_B \approx 2.1$ –2.5 keV has been calculated using modern models [59,60]. As seen by the ^{93}Mo ions when slowed to 4.91–4.33 MeV/A, the relative kinetic energy of approaching atomic electrons from the helium gas will be $E_K = 2.74$ –2.33 keV. Therefore, at some point within the gas, $E_{\text{NEEC}} = 4.85$ keV = E_n and NEEC will occur.

^{*} The standard way of denoting a nuclear reaction is T(P, O)F, where T is the target nucleus, P is the projectile nucleus, and O represents any particles emitted prior to creation of the final nucleus F. The final nucleus may be omitted in the designation if its identity is clear from the context.

[†] In a fusion-evaporation reaction, the incident kinetic energy of projectile and target nuclei in the center-of-mass frame is sufficient to surmount the Coulomb repulsion of the nuclei (the Coulomb “barrier”). The projectile and target nuclei fuse into an intermediate “compound” nucleus that exists (Morinaga H, Yamazaki T. In-beam gamma-ray spectroscopy. New York [NY]: North-Holland; 1976) on the order of 10^{-18} s. As the internal energy of the compound nucleus is large, an effective high internal temperature, it decays most quickly by evaporating nucleons (most often neutrons). After ejection of the hot nucleons, the final nucleus is produced, which can no longer emit particles but can decay by gamma emission and internal conversion.

[‡] Projectile and recoil kinetic energies may be given per nucleon (A = total number of nucleons within a nucleus) or by the total energy.

[§] After 15 cm, the ^{91}Zr ions would be traveling too slowly for the $^4\text{He}(^{91}\text{Zr}, 2\text{n})$ reaction to occur.

^{**} 1 particle nanoampere = 6.24×10^9 particles/s.

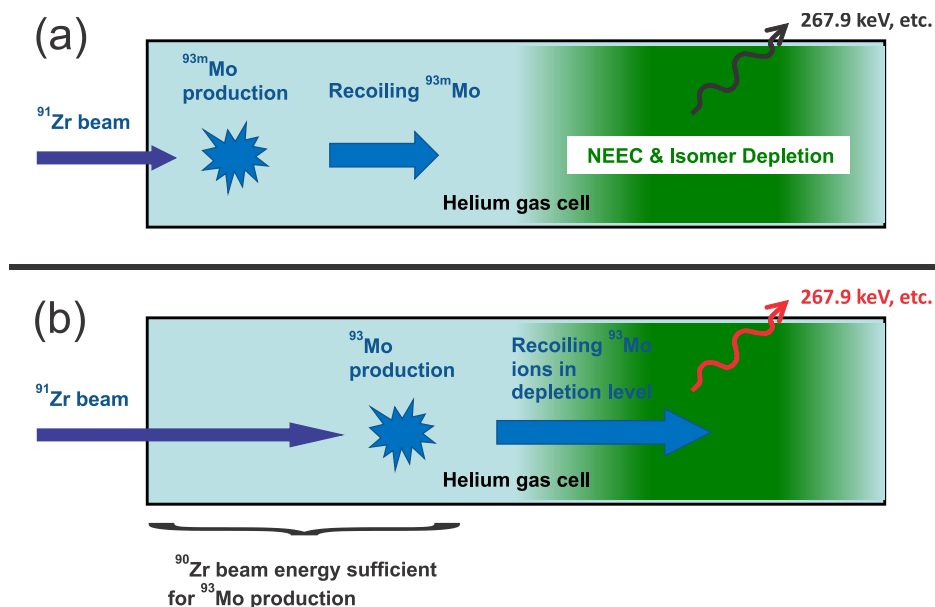


Fig. 6 Original helium gas experimental configuration, the gas providing both production of $^{93\text{m}}\text{Mo}$ via the $^4\text{He}(^{91}\text{Zr}, 2\text{n})$ reaction and stopping medium for NEEC. (a) Gamma emission at 267.9 keV, after population of the 6.85-h isomer, from later parts of the ^{93}Mo ion track would indicate isomer depletion. (b) Gamma emission at 267.9 keV without population of the 6.85-h isomer because of decay of the 3.5-ns depletion state after direct population from higher-lying levels. This would serve as a form of background to the measurement.

The probability of NEEC was estimated for these conditions to be 10^{-4} [46], giving a rate of isomer depletion of 100/s. As the charge states and velocities of the ^{93}Mo ions change along their track in the gas, there would likely be many opportunities for NEEC resonance to occur so that the signal estimate was presumed to be conservative. An increase in the ^{91}Zr beam energy or current would increase the signal. Background radiation would be emitted from within the medium, both in the form of gamma rays, and characteristic X-rays from molybdenum and helium atoms due to radiative recombination. The X-rays, however, will have energies less than 20 keV (the $\text{K}_{\beta 1}$ line of molybdenum lies at 19.6 keV and other lines are less energetic) so that the walls of the helium cell would likely shield any detector from that radiation. Gamma rays would come from competing nuclear reactions, such as $^4\text{He}(^{91}\text{Zr}, \text{n})^{94}\text{Mo}$ or $^4\text{He}(^{91}\text{Zr}, 3\text{n})^{92}\text{Mo}$, and Coulomb excitation of excited states in the ^{91}Zr projectiles. The available nuclear data [4] suggests that characteristic gamma rays due to other reactions could be differentiated from the NEEC signal by their energies if they were different from those of the depletion-level cascade shown in Fig. 4. If any other product nuclei have previously unmeasured gamma-ray transitions at the energies signaling NEEC in ^{93}Mo , that would be a significant source of experimental background.

There are noteworthy challenges, however, to the use of this scheme in the helium-based form that are primarily due to the low density of the medium. At STP, the

production reactions to create $^{93\text{m}}\text{Mo}$ will take place over a spatial range of tens of centimeters, while NEEC would occur over an even longer distance. Gamma rays would be emitted from nuclei moving with different velocities, and at different positions within the gas, so that detectors at different locations would record different Doppler-shifted gamma-ray energies. This precludes the use of state-of-the-art gamma detector arrays, which have physical configurations designed for studies of excited nuclei confined to a rather small volume (i.e., typically stopped within a thin target foil). The inability to use such detector arrays, with their large efficiencies and ability to record multi-gamma-ray events, makes it more difficult to isolate gamma rays signaling NEEC from those resulting from other reactions. Also, the 268-keV gamma ray that would signal NEEC after the ^{93}Mo nucleus reached the 6.85-h isomer could also occur in the latter part of the gas cell (see Fig. 6b) by decay of the depletion level without the nucleus ever reaching the long-lived isomer because of direct feeding from states above the depletion level (such states are not shown in Fig. 4, but a number are known [4,52]), and multi-gamma-ray events would be needed to differentiate between the 2 situations. Differing Doppler shifts could be eliminated by using detectors in a ring around the ion trajectory, collimated so that they could only see gamma rays emitted from a narrow range along the track and only within a narrow angular window. However, this would greatly reduce the data acquisition rate, as gamma rays emitted from other positions along the track and in other directions would not be detected.

Finally, there are practical issues in the use of a gas target. Accelerators deliver ions along evacuated beamlines so that either a thin window would be required prior to entering the helium volume or differential pumping would be used for a windowless gas cell. Both approaches are used in experiments (see, for example, Rehm et al. [61] and references therein) but would introduce new challenges to the present experiment by adding competing nuclear reactions with the window material or by limiting the stopping cell to low pressure to permit differential pumping. Cryogenic gas targets are possible but are difficult to construct and exist at only a few facilities.

3.2 Concept Refinement

Potential variations to the core concept—use of inverse kinematics and high-recoil velocities of $^{93\text{m}}\text{Mo}$ ions through a medium—were considered. These could be characterized as the evaluation of other producing reactions and as the consideration of modifications to the physical configuration of the experiment.

3.2.1 Reaction Possibilities

The originally proposed ${}^4\text{He}({}^{91}\text{Zr}, 2n)$ reaction has a number of attractive features, which can be seen from the PACE calculations* shown in Fig. 7. A total of 100,000 excited-state cascades were simulated for each choice of beam energy. As mentioned previously, about one-third of the total production of ${}^{93}\text{Mo}$ was estimated to produce excited states that would lead to population of the 6.85-h isomer. This occurs with a cross section peaking at nearly 500 mb, and production occurs over a relatively wide range of beam energies. This means that if particles enter the production medium at, for example, 800 MeV,[†] they will continue to have an opportunity to produce ${}^{93\text{m}}\text{Mo}$ for some distance as the projectiles slow. Despite improving production of the isomer, it also widens the length of the track along which ${}^{93\text{m}}\text{Mo}$ ions are created and broadens the distribution of their recoil velocities. Within the range of beam energies of 550–750 MeV, the production of other nuclides is relatively small, which is important since they will also be produced in excited states and emit gamma rays.

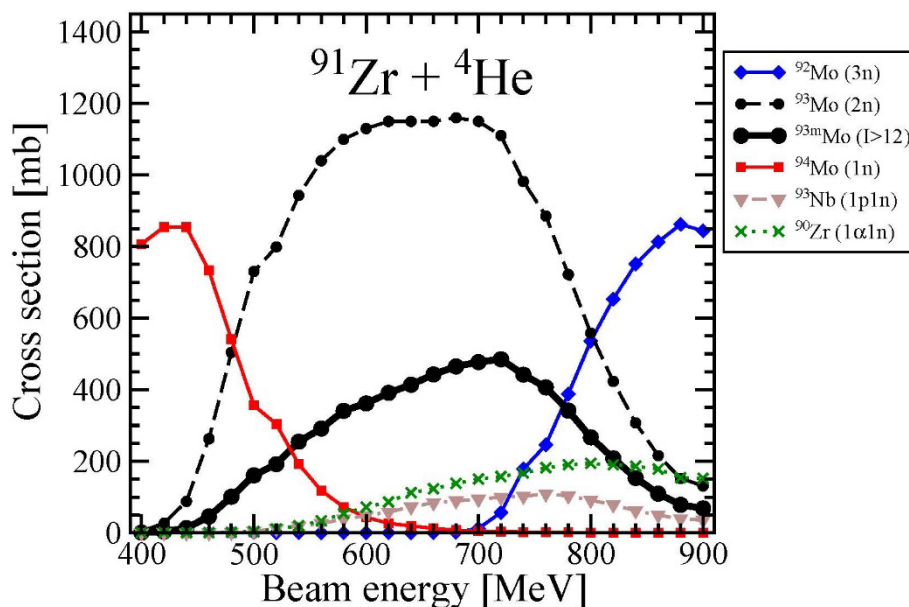


Fig. 7 Estimates of cross sections for reactions with ${}^{91}\text{Zr}$ projectiles on ${}^4\text{He}$ target nuclei, showing cross sections for specific product nuclei as a function of ${}^{91}\text{Zr}$ beam energy. The legend identifies product nuclei and the corresponding evaporated nucleons.

* The PACE code can be used to estimate the cross section for population of all states having a minimum angular momentum. The population of the $J^\pi = 21/2^+$ isomer was estimated from the PACE results, assuming it would be reached by decay cascades from levels with angular momentum larger than $J = 12$. It was assumed that each evaporated nucleon would carry away 1–2 units of angular momentum.

[†] When kinetic energies are given in the context of cross section estimates or measurements, units of megaelectron volts (MeV) will be used. When kinetic energies are given for recoiling ions or in reference to NEEC resonances, units of megaelectron volts per nucleon (MeV/A) will be used.

The $^1\text{H}(^{93}\text{Nb}, n)$ reaction was simulated as well, with PACE results shown in Fig. 8. This reaction is attractive in that, compared to the production of ^{93}Mo , there is relatively little production of competing products. However, the reaction imparts little angular momentum to the ^{93}Mo nucleus so that there is little production of the 6.85-h isomer. The small angular momentum is due to the maximally asymmetric masses of the reaction pair and the small number of evaporated nucleons (one neutron) to produce ^{93}Mo .

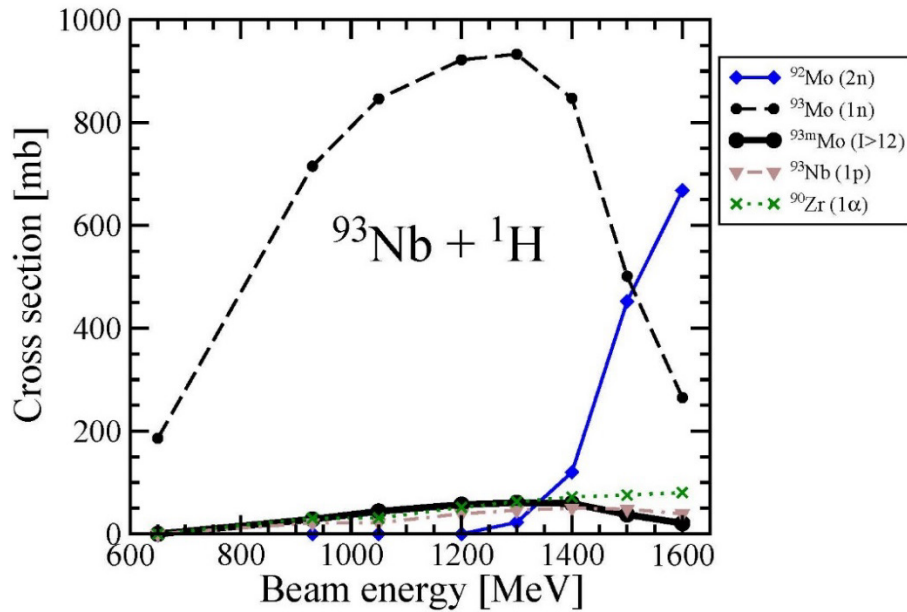


Fig. 8 Estimates of cross sections for reactions with ^{93}Nb projectiles on ^1H target nuclei, showing cross sections for specific product nuclei as a function of ^{93}Nb beam energy. The legend identifies product nuclei and the corresponding evaporated particles.

The $^7\text{Li}(^{89}\text{Y}, 3n)$ reaction was investigated, as shown in Fig. 9. Production of ^{93m}Mo is significant but at lower beam energies than for the above reactions. There is also strong production of other nuclides whose gamma emission may compete with the NEEC signal.

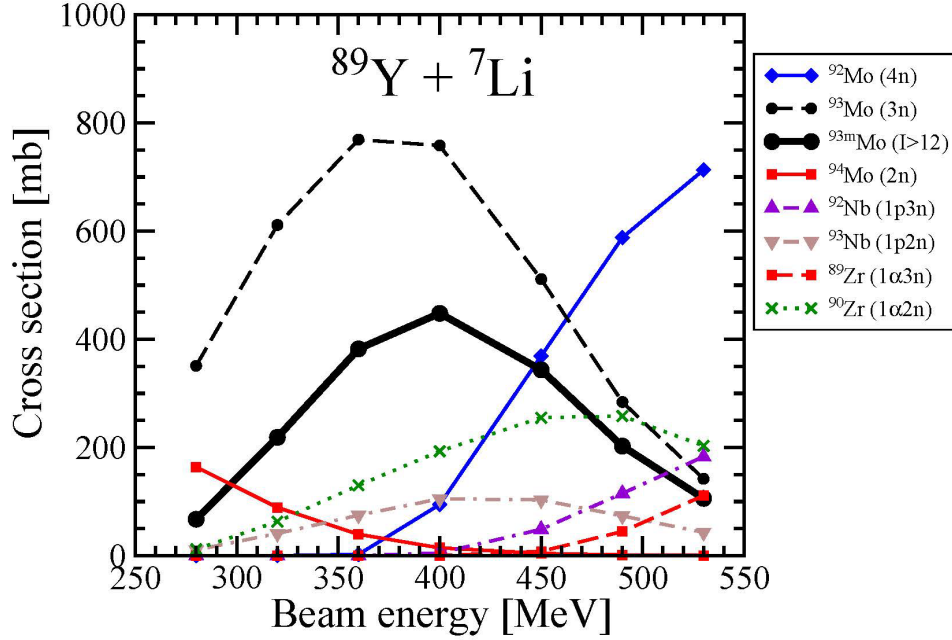


Fig. 9 Estimates of cross sections for reactions with ^{89}Y projectiles on ^7Li target nuclei, showing cross sections for specific product nuclei as a function of ^{89}Y beam energy. The legend identifies product nuclei and the corresponding evaporated nucleons.

A number of other reactions were analyzed, including $^{13}\text{C}(^{84}\text{Kr}, 4n)$ and $^7\text{Li}(^{90}\text{Zr}, p3n)$, with PACE results for the latter shown in Fig. 10. This reaction has a good cross section for production of $^{93\text{m}}\text{Mo}$ over a range of energies, but there are a number of competing product nuclei from this reaction pair.

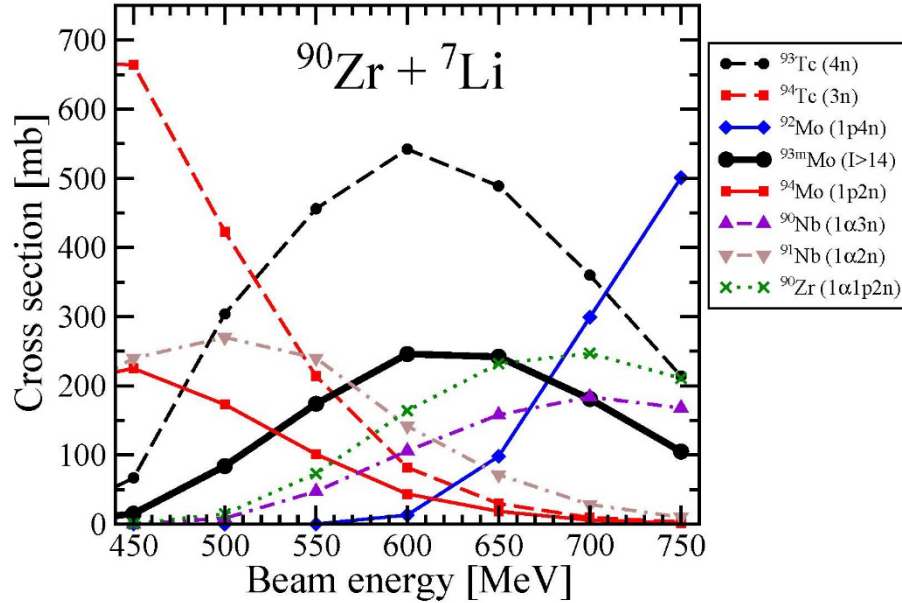


Fig. 10 Estimates of cross sections for reactions with ^{90}Zr projectiles on ^7Li target nuclei, showing cross sections for specific product nuclei as a function of ^{90}Zr beam energy. The legend identifies product nuclei and the corresponding evaporated nucleons.

In addition to the cross section for the production of $^{93\text{m}}\text{Mo}$, it is critical that recoiling molybdenum ions have sufficient kinetic energy for NEEC to occur in a subsequent stopping medium. The recoil velocities of $^{93\text{m}}\text{Mo}$ ions after the reaction was calculated for the reaction pairs of Figs. 7–10 from momentum conservation,* assuming a projectile energy near the peak of the cross section. These values are listed in the following Table. (In an actual experiment, the recoil energies will be characterized by a distribution due to production of ^{93}Mo over a range of energies as projectiles slow in a material and due to random angular distribution in the emission of evaporated nucleons.)

Table. ^{93}Mo ion recoil energy for selected reactions

Reaction	Projectile energy (MeV)	Recoil energy (MeV/ <i>A</i>)
$^1\text{H}(^{93}\text{Nb}, \text{n})$	1300	13.7
$^4\text{He}(^{91}\text{Zr}, 2\text{n})$	720	7.3
$^7\text{Li}(^{89}\text{Y}, 3\text{n})$	400	3.9
$^{13}\text{C}(^{84}\text{Kr}, 4\text{n})$	350	3.1
$^7\text{Li}(^{90}\text{Zr}, \text{p}3\text{n})$	650	6.2

The analyses of production cross sections and recoil energies for these reactions support the following conclusions:

- More symmetric reactions (smaller mass difference between the reaction pair nuclides) produce higher angular-momentum states in ^{93}Mo product nuclei, leading to a higher cross section for creating $^{93\text{m}}\text{Mo}$. However, they generate lower recoil energies for those ions in inverse kinematics.
- More asymmetric reactions (larger mass difference between the reaction pair nuclides) lead to larger recoil energies for ^{93}Mo ions in inverse kinematics.
- The cleanest reaction is $^1\text{H}(^{93}\text{Nb}, \text{n})$, which produces the fewest other products. The ^{93}Mo recoil energy is high, but overall isomer production is low.
- The originally proposed $^4\text{He}(^{91}\text{Zr}, 2\text{n})$ reaction is optimum from the perspective of $^{93\text{m}}\text{Mo}$ production and recoil.

These cross-section estimates are only a guide in experiment planning and cannot replace actual measurements.

* Even for projectile kinetic energies approaching 1,000 MeV, this is much smaller than the rest mass for $A \sim 90$ nuclei ($> 87,000$ MeV). Thus, the recoil can be calculated classically rather than relativistically.

3.2.2 Configuration Options

Although ${}^4\text{He}({}^{91}\text{Zr}, 2n)$ is optimum from the pure perspective of the reaction, there are significant difficulties introduced by the use of a gas target, as discussed previously. A more detailed understanding of the causes for these difficulties is necessary to facilitate consideration of alternative physical configurations, including the use of other targets.

A successful NEEC demonstration experiment with ${}^{93\text{m}}\text{Mo}$ must unambiguously identify events where the depletion level has been excited from the 6.85-h isomer by the atomic-nuclear process. In practice, this means accurate measurement of the 268-keV gamma ray emitted by decay of the depletion level, as well as of gamma rays at 685 and 1478 keV that are emitted in the remainder of the cascade to the ground state, as shown in Fig. 4. However, those same gamma rays can be emitted without the nucleus ever reaching the long-lived isomer and, thus, without NEEC, since the producing reaction may result in direct population of the depletion level by decay cascades from higher-lying states. An experiment must be able to differentiate between gamma-ray emission events due to NEEC and those which are not. In general, this differentiation could be accomplished in 2 ways:

- Directly identifying on an event-by-event (nucleus-by-nucleus) basis which gamma rays emitted after depletion-level decay occurred after population of the 6.85-h isomer. This would require use of state-of-the-art multidetector arrays with their ability to record the energies of many gamma rays emitted by a nucleus within a very short time window.
- Physically separate the region in which the signature gamma rays are emitted from the depletion level after direct population of that level and the region in which those gamma rays are emitted after NEEC.

To consider the feasibility of using a multidetector array with a gas target, it is illustrative to consider the case where the half-lives are extremely short (i.e., $\text{fs} = 10^{-15} \text{ s}$) for all states whose decay cascades feed either the depletion level directly or the 6.85-h isomer. The depletion level is also assumed to be similarly short-lived. For the ${}^4\text{He}({}^{91}\text{Zr}, 2n)$ reaction, production of ${}^{93}\text{Mo}$ can occur over a spatial range within a helium gas cell. A zirconium ion energy of 800 MeV provides a significant cross section for population of ${}^{93}\text{Mo}$, as shown in Fig. 7, and the reaction can occur probabilistically just after the ${}^{91}\text{Zr}$ enters the helium gas. Taking place at this energy, the ${}^{93}\text{Mo}$ would recoil after the reaction with a mean energy of 8.06 MeV/A (the evaporated neutrons could be emitted isotropically, so on average, they do not change the momentum and energy of the recoiling nuclei). Under the half-life assumptions taken presently, some ${}^{93}\text{Mo}$ nuclei would immediately reach the depletion level and some would immediately reach the 6.85-h isomer. Those

reaching the depletion level directly would also immediately decay, emitting the 268-keV gamma ray from that location just inside the helium gas. In contrast, ^{93m}Mo nuclei could move for a relatively long distance within the helium gas before NEEC occurred. Assuming that the NEEC resonance would occur near $5.5 \text{ MeV}/A$, as originally estimated [54], the ^{93m}Mo nuclei would travel a distance of 21 cm in helium at STP, as determined from the SRIM (Stopping Range of Ions in Matter) code [47]*. Occurrence of NEEC at that position would lead to immediate gamma-ray emission at 268 keV and is spatially separated from the same emission from the depletion level after its direct population.

The cross section for production of ^{93}Mo in the $^4\text{He}(^{91}\text{Zr}, 2n)$ reaction is considerably reduced, but still non-negligible, by the time the zirconium ion energy has dropped to 545 MeV. A ^{91}Zr ion could travel 26 cm in helium at STP before slowing to this energy: ^{93}Mo produced thereby would have a recoil energy of $5.5 \text{ MeV}/A$, so that NEEC could occur at that position after immediate population of the 6.85-h isomer. Likewise, emission of 268-keV gamma rays following direct population of the depletion level would occur from that same position. The production reactions would occur over a length of about 26 cm in helium at STP, starting at the entrance of the zirconium ions to the gas. The region in which NEEC could occur starts at about 21 cm into the gas and extends over a range of about 5 cm thereafter, which overlaps part of the ^{93}Mo production range. Emission of 268-keV gamma rays due to direct population of the depletion level will occur over the entire range over which ^{93}Mo could be produced, as depicted in Fig. 6. With no ability to physically separate all NEEC and non-NEEC events that emit this gamma ray, a multidetector array would be needed. The physical extent of the NEEC range, 5 cm, is small enough to be accommodated within the central cavity of most arrays. The full extent of the helium gas cell would be larger but could also be accommodated by removing some detectors from the array (with an accompanying loss of some detection efficiency) toward the end of the cell into which the ^{91}Zr beam is incident. The principal difficulty lies in the Doppler shifts introduced by emission of 268-keV gamma rays by moving nuclei within the NEEC region.

In addition to different nuclei emitting at different velocities, giving different amounts of energy shifts, they will also emit from different positions within the extended 5-cm NEEC range. This means a variation of the angle θ shown in Fig. 5, which will introduce variations of the shifts. Not only will the spectra measured by each detector in the array show 268-keV gamma-ray peaks at different locations, but within each spectrum the 268-keV peak will be broadened as it is an accumulation of separate events with different measured gamma-ray energies. The

* The original proposal envisioned a ^{91}Zr beam energy of only $6 \text{ MeV}/A$, corresponding to a ^{93}Mo production range of approximately 15 cm in helium at STP.

situation is even worse if the negligible half-life assumption is removed for feeding of the depletion level and 6.85-h isomer, as the real half-lives will further extend the regions for $^{93\text{m}}\text{Mo}$ production, NEEC, and emission from the depletion level. Overall, the ability to differentiate between NEEC and non-NEEC emission at 268 keV with multidetector arrays would be severely degraded if using helium (or other gas) media for both isomer production and NEEC. The issue is created simply by the low density for gases and only mitigated slightly by the use of a cryogenic target; even for a helium cell near 3 atm at 80 K, the ^{93}Mo production range would still be 12 cm.

The use of a solid material for both $^{93\text{m}}\text{Mo}$ production and for NEEC would greatly reduce the length of the corresponding regions (e.g., to on the order of 50 μm for a lithium target with the $^7\text{Li}(^{90}\text{Zr}, \text{p}3\text{n})$ reaction). However, the reactions with hydrogen and helium were identified as promising. Thus, alternative configurations were considered using a gas target for ^{93}Mo production and a separated solid target for NEEC. A solid target for NEEC has the additional advantage of providing higher mean charge states for fast ions, compared with a gas [57]. This would enhance creation of the needed atomic vacancies.

Configurations consisting of a gas production medium and subsequent solid NEEC target generally fell into the 2 variations shown in Fig. 11. In the variant shown in Fig. 11a, a cell of helium serves to produce ^{93}Mo ions through the $^4\text{He}(^{91}\text{Zr}, 2\text{n})$ reaction. The gas cell is relatively thin so that it would use only a small region of the cross section curve shown in Fig. 7, and ions of ^{93}Mo created within the gas cell would recoil with little loss of energy prior to exiting into the vacuum region. There, they would travel without energy loss for a period sufficient for any directly produced depletion levels to decay. The half-life of the depletion level is 3.5 ns, and the recoil velocities of ^{93}Mo ions will be on the order of 8 MeV/A, which corresponds to a speed of about $4 \times 10^9 \text{ cm/s} = 4 \text{ cm/ns}$.^{*} Thus, a transit length in vacuum of about 1.4 m would be necessary for 10 half-lives of the depletion level to have elapsed, reducing the emission rate of 268-keV gamma rays due to direct depletion-level population by about 1,000 times. The cumulative half-life for population of the 6.85-h isomer must be significantly shorter than 800 ps (the shortest half-life observed for excited states of ^{93}Mo) or measured half-lives should have been reported in previous spectroscopic works [51,52]. Thus, population of the $^{93\text{m}}\text{Mo}$ isomer will be complete before those ions enter the solid NEEC target. The solid target will produce high charge states for those ions as they are slowed in that medium to permit NEEC to occur. Once the depletion level is excited by NEEC, the ions would continue to move in the solid target until stopped so that it

^{*} This is about 13% the speed of light in a vacuum.

is likely that many gamma-ray emissions will experience Doppler shifts. One or more detectors viewing the NEEC medium would record those gamma rays. A multidetector array could be used to increase overall detection efficiency, but more advanced features of such a system would not be needed.

A major difficulty, however, would be a large number of ^{91}Zr ions that passed through the thin gas cell without causing a reaction. They would traverse the vacuum region without slowing and strike the NEEC medium, producing additional reactions with that medium with considerable prompt gamma-ray emission from the resulting nuclei. To prevent ^{91}Zr ions from reaching the NEEC target, a mass separator could be used as part of the vacuum region shown in Fig. 11a. Unfortunately, the capabilities of state-of-the-art separators may be insufficient for this approach; the Fragment Mass Analyzer at Argonne National Laboratory (ANL) cannot effectively steer ions with the anticipated charge states at these high recoil energies [62], while the Momentum Achromat Recoil Spectrometer separator at Texas A&M University is used for lower masses but higher kinetic energies [63]. Foils are often used to degrade ion kinetic energies prior to entering a mass separator but could not be used here as the velocity of $^{93\text{m}}\text{Mo}$ ions entering the NEEC medium must be large.

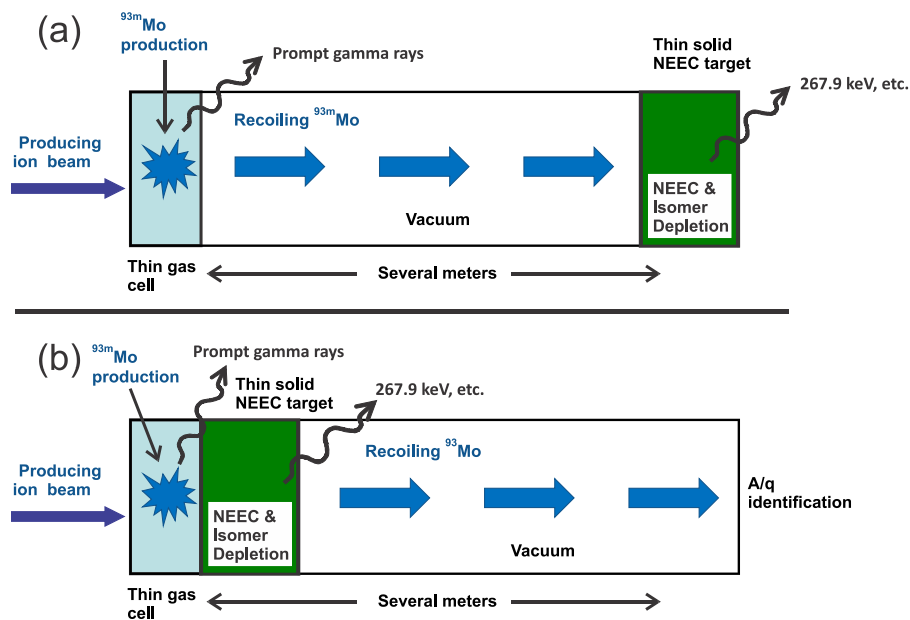


Fig. 11 Alternative physical configurations based on separation of gas production and solid NEEC media. (a) Introduction of a physical gap of several meters in vacuum between media to allow sufficient delay for all directly produced depletion levels to decay. Gamma rays emitted from within the solid NEEC medium would be observed by a multidetector array. (b) Adjacent production and NEEC targets, with both production and NEEC media observed by a multidetector array. Along a subsequent long vacuum path in a mass separator, the corresponding A/q of the ions would be determined, identifying the nuclide responsible for emitted gamma rays.

Another alternative configuration is shown in Fig. 11b. There, the helium production and solid NEEC media would also be separate but placed back-to-back. A multidetector array would need to observe the NEEC target but would also see the production medium due to its proximity so that the advanced features of such arrays would be required. Passage of ions of ^{93}Mo and other product nuclei, as well as of unreacted ^{91}Zr projectiles, through the NEEC target (thinner in this configuration than that in Fig. 11a) would slow them sufficiently to be suitable for mass separation. Identification of ^{93}Mo ions in the mass separator would be correlated in time with gamma rays observed by the multidetector array to distinguish those events from events resulting from other nuclides. Again, the 3.5-ns half-life of the depletion level is problematic in that with a thinner solid NEEC target, nuclei will not have come to rest before emission of the characteristic gamma rays. Spread over a distance of several centimeters, the emission of those gamma rays would be subject to Doppler shifts due to different velocities and emission positions.

Finally, the use of a gas target was abandoned in favor of one solid target that would serve as both production and NEEC media. Considering the potential reactions analyzed previously, the $^7\text{Li}(^{90}\text{Zr}, p3n)$ reaction was selected as most attractive, as solid ^7Li targets have previously been made [64].

4. Experiments and Results

Although there have been numerous prior studies of ^{93}Mo , no experimental information about its production following the fusion of ^{90}Zr and ^7Li nuclei has been reported in the literature. As a precursor to the NEEC experiment, it was important to first explore this reaction with a benchmark experiment under conditions in which NEEC could not occur, ensuring that the reaction and possible background were well understood. A simple way to achieve this would be to run the same reaction in normal kinematics—namely, use a ^7Li beam directed onto a ^{90}Zr target. In the center-of-mass (CM) frame, this reaction is identical to the inverse-kinematics $^{90}\text{Zr} + ^7\text{Li}$ reaction described earlier, provided the CM energies are the same. In the laboratory frame, this amounts to the same projectile energy per nucleon in either case. One key difference between the normal- and inverse-kinematics reactions is that the ^7Li beam in the former case would impart considerably less momentum to the reaction products than in the latter case. The correspondingly small kinetic energy (and velocity) of the ^{93}Mo recoils would fall below the threshold for NEEC to occur. Thus, the normal kinematics reaction can be used to study the $^{90}\text{Zr} + ^7\text{Li}$ reaction without possible contributions from the NEEC process, providing a control with which to compare the later experiment.

An extensive search was made to identify facilities [65] capable of performing the benchmark and NEEC experiments. For the NEEC experiment, the Argonne Tandem Linear Accelerator System (ATLAS) was clearly optimal because of its ability to accelerate the required ^{90}Zr ions with sufficient energy and beam current, and established expertise to construct the needed ^7Li target. The unique capabilities of the Gammasphere multidetector array (described later) were also indispensable, into which beams from ATLAS can be delivered.

In principle, the benchmark experiment could also have been performed at ANL. This would have been the best option to provide an environment and detector setup that matched the NEEC experiment as closely as possible, thus giving even better control over background and instrumentation response. However, beam time at ATLAS is in high demand, and all beam requests must be approved by a Program Advisory Committee (PAC) that meets only once or twice a year. Even if an experiment has been approved, it can sometimes be many months (or, rarely, years) before the experiment is scheduled. Also, although a necessary component to the eventual NEEC experiment, the benchmark study is not by itself of sufficient scientific import to make likely its acceptance for ATLAS. The Heavy Ion Accelerator Facility (HIAF) at Australian National University (ANU) [65,66], on the other hand, operates with a more informal process, so the benchmark experiment could be proposed, approved, and executed on a much shorter time scale. This was valuable since the results of the benchmark experiment would be needed before the NEEC test could be conducted. Despite there being differences between the 2 facilities, the information gathered at ANU successfully met all goals set forth for the benchmark experiment.

4.1 Benchmark Experiment (Normal Kinematics)

There were 3 main goals of a normal kinematics benchmark experiment. One was to determine the appropriate experimental conditions for the NEEC test, such as the optimal beam energy. This could be achieved by determining an excitation function (i.e., a measurement of the relative yields of isomeric $^{93\text{m}}\text{Mo}$ and other reaction products as a function of beam energy). The excitation function is the experimental counterpart to the estimates shown in Fig. 10. The best choice of beam energy, from a production point of view, would maximize the $^{93\text{m}}\text{Mo}$ cross section while keeping the production of other nuclei to a minimum.

The second goal of such an experiment was to further develop the ^{93}Mo level scheme. Although some excited states at energies above the 6.85-h isomer have been previously established in ^{93}Mo using, for example, the $^{82}\text{Se}(^{16}\text{O}, 5n)$ [51] and $^{93}\text{Nb}(p, n)$ [67] reactions, the states accessed by these reactions likely differ from

those that would be populated by the ${}^7\text{Li}({}^{90}\text{Zr}, \text{p}3\text{n})$ reaction. More symmetric reactions, such as ${}^{16}\text{O} + {}^{82}\text{Se}$, can bring significant angular momentum into the compound system and favor high-angular-momentum states, typically populating only the few lowest-energy states for each value of angular momentum; conversely, highly asymmetric reactions like ${}^1\text{H} + {}^{93}\text{Nb}$ populate multiple states of a given angular momentum across a broad range of excitation energies, but only for relatively low values of angular momentum. The ${}^7\text{Li} + {}^{90}\text{Zr}$ reaction falls between these limits and may access some subset of states identified in both of the other reactions, as well as previously unobserved states. Performing the measurement in normal kinematics first would provide information on the coincident* gamma rays that would be expected in the later inverse-kinematics NEEC experiment. The absence of Doppler effects in normal kinematics, due to stopping the slowly recoiling excited nuclei within a sufficiently thick target, results in narrow peaks in the gamma-ray energy spectra, simplifying the analysis.

The third goal was to ensure that there will be a clear signature for when NEEC has occurred by demonstrating that expected NEEC-related coincidences are not present in the normal kinematics experiment (i.e., no false positives). Since NEEC would occur as an excitation out of the 2425-keV, 6.85-h isomer, the first step is to verify that it was populated in the reaction. This can be achieved by requiring that one of the gamma rays known to directly feed the isomer is observed in the NEEC experiment. As can be seen from the partial ${}^{93}\text{Mo}$ level scheme in Fig. 12, the 2475-keV gamma ray would be an appropriate choice. Under normal circumstances, after emission of the 2475-keV gamma ray, the nucleus would remain “trapped” in the isomeric state, decaying, on average, hours later by the emission of the 263–685–1478 keV sequence. However, if NEEC occurs, indicated by the dashed red arrow in Fig. 12, the nucleus can be excited to the 2430-keV level on a much shorter time scale (picoseconds), followed by a slightly delayed emission of the 268–685–1478 keV sequence due to the half-life of the depletion level at 3.5 ns.

* Two gamma rays are considered to be in prompt time coincidence if they are successively emitted within a small time window of each other, typically on a microsecond or shorter time scale.

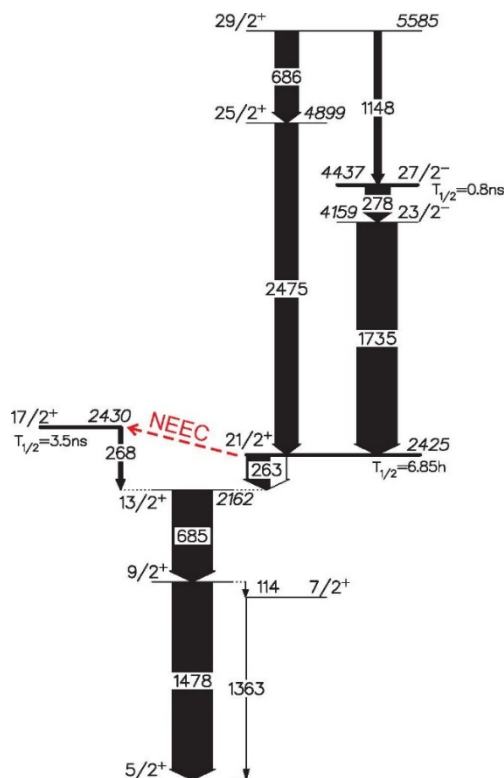


Fig. 12 Partial level scheme of ^{93}Mo showing transitions feeding the 6.85-h isomer, adapted from Fukuchi et al. [51] and Baglin [69]. The dashed red arrow illustrates the transition to higher excitation energy via the NEEC process. The separation between the $21/2^+$ isomer and $17/2^+$ depletion state is exaggerated for clarity. Transition intensities, represented by the arrow widths, are taken from the $^{82}\text{Se}(^{16}\text{O},5n)$ reaction [51]. The white part of the arrow for the 263-keV transition corresponds to the fraction of the decays occurring by internal conversion while the black part represents the gamma-ray fraction.

It should be noted that the 2475-keV gamma ray should not be observed in prompt time coincidence with the 685- and 1478-keV gamma rays below the 6.85-h isomer, since the coincidence time window would be far too narrow. In fact, the 2475-keV gamma ray could never be coincident with the 268-keV transition unless NEEC has occurred, allowing the long half-life of the isomer to be circumvented. Thus, the signature coincidences to be sought are those involving gamma rays above the isomer, such as the one at 2475 keV, and the 268-, 685-, and 1477-keV transitions. The aim is to show that such coincidences, possibly originating from previously unknown coincident gamma rays in ^{93}Mo or other background reaction products, are *not* present in the normal kinematics benchmark reaction, where the recoil energies are too low for NEEC to occur.

The benchmark $^7\text{Li} + ^{90}\text{Zr}$ experiment was performed at HIAF during May 2015 [68]. There, singly negatively charged ^7Li ions were produced by an ion source and

injected into a Pelletron electrostatic tandem accelerator* having a “terminal” voltage V_{term} . The maximum achievable terminal voltage at HIAF is 15 MV. Ultimately, fully stripped ${}^7\text{Li}^{3+}$ ions were delivered by the accelerator to the target position, and the terminal voltage was adjusted throughout the experiment to provide various energies between 30 and 56 MeV. The beam was “chopped” rather than continuous so that the irradiations occurred in cycles of 1-ms-duration beam on target followed by 1 ms with no beam on target. This provided the opportunity to monitor long-lived activity (build-up of long-lived states) that contributes to background during the beam-on periods through random coincidences.

The ${}^7\text{Li}$ beam was directed onto zirconium target material enriched to 99.36% in ${}^{90}\text{Zr}$ (the natural abundance is 51.45%). Three targets were prepared at Oak Ridge National Laboratory in the form of 13- × 15-mm rectangular foils, approximately 18 μm thick. These foils were used either individually or in a stack of 3. The foils were tilted by 20° with respect to the beam direction, increasing their effective thickness by about 6%.

The target position was surrounded by the CAESAR multidetector array, a set of 11 high-purity germanium (HPGe) detectors used for recording gamma-ray emissions (Fig. 13). This type of detector is preferred for nuclear spectroscopy because of its combination of excellent energy resolution† on the order of 0.1% of the gamma-ray energy, modest time resolution (the time of gamma ray detection can be defined with an uncertainty of a few nanoseconds), and detection efficiency ($\sim 1.5\%$ for the CAESAR array as a whole, for 1-MeV gamma rays) [70]. For example, the energy resolution is important for distinguishing discrete peaks in complex gamma-ray spectra where multiple peaks can be in close proximity to each other.

* A tandem van de Graaff accelerator establishes the terminal voltage between the midpoint of the device and either end—both ends are at ground potential. A singly negatively charged ion is injected and accelerated to an energy given by qV_{term} , where $q = e$, the charge of an electron. Near the midpoint of the accelerator, the ions pass through a thin foil that strips electrons from the projectile. In the case of lithium, the electrons were fully stripped so the charge of the ions was $q = +3e$. The ions are accelerated further from the midpoint to the end of the device, again adding qV_{term} , but with this different q . The overall energy reached by the lithium ions would then be $4V_{term}$, higher than could have been achieved from the terminal voltage if the ions were always singly charged. A Pelletron uses a segmented chain to charge the accelerator, which is otherwise based on the van de Graaff approach.

† A spectral peak is largely symmetric, represented by a Gaussian, with a width characterized by its FWHM. These instrumental widths are much larger than the intrinsic widths, Γ , as shown in Fig. 2.

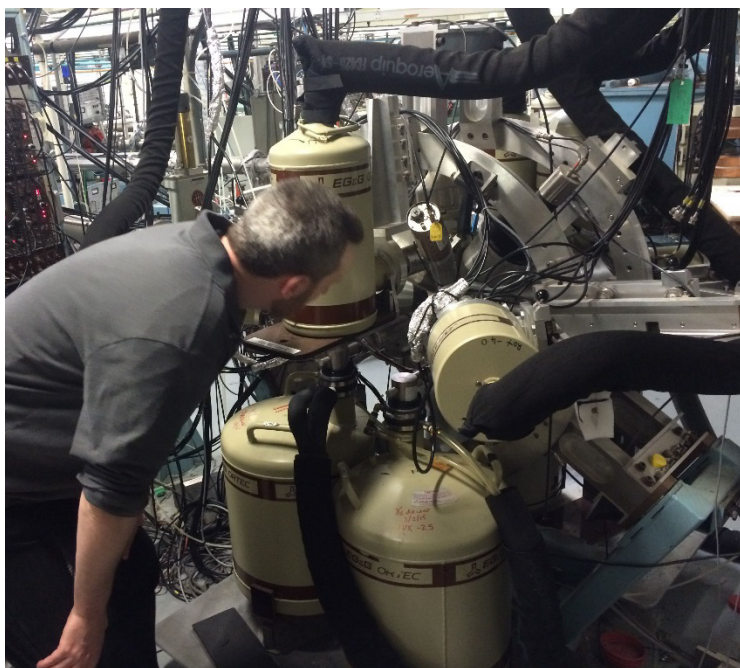


Fig. 13 Photograph of the CAESAR array at Australian National University. The tan/brown cylinders attached to thick black fill hoses are the LN_2 dewars for each HPGe detector, which must operate at 80 K. The HPGe detectors themselves are not visible in this view, as they face inward toward the target chamber.

Two of the 11 detectors in CAESAR were smaller-volume planar Low-Energy Photon Spectrometers, which have excellent performance (energy resolution, timing, and efficiency) for low-energy gamma rays but do not contribute much efficiency to the array for higher energies (greater than a few hundred kiloelectron volts). Thus, most of the analysis of the ANU data was on signals from the 9 larger-volume HPGe detectors, which had coaxial crystal geometries.

The use of a multidetector array with reasonable timing resolution provides a means of studying gamma-gamma coincidence relationships. That is, when a sequence of gamma rays is emitted in rapid succession from an excited nucleus, one gamma ray in the sequence can be recorded in one detector and a subsequent gamma ray in a different detector; the corresponding data are then recorded as a correlated event that includes energy, timing, and detector-identification information for each gamma ray. These coincidence data can then be analyzed offline (after the experiment), as described later in this section.

CAESAR uses a standard analog-digital electronics system. Analog tail pulses* from the HPGe detectors are split into timing and energy channels. The analog signals in the timing channel may be used to generate square STOP pulses, while the

* Tail pulses have sharp, nearly linear rising edges, with falling edges that follow an exponential decay of the charge through the equivalent resistor–capacitor circuit of the detector’s preamplifier.

accelerator pulses may be used to generate START pulses. A time-to-analog converter (TAC) then produces voltage pulses whose heights are proportional to the time between START and STOP pulses, and the TAC output is digitized by an analog-to-digital converter (ADC). In the energy channel, tail pulses from the detectors pass through a linear shaping amplifier whose output is sent to an ADC; the heights of the amplified pulses are proportional to the energy deposited in the detector by the gamma ray. The data are then recorded as a stream of consecutive packets, each of which contains the identification of the detector hit by a gamma-ray, the time of the hit, and the height of the amplified detector pulse. While a subset of the data can be viewed in real time to ensure the acquisition of the proper data and correct functioning of the data acquisition (DAQ) system, most data analysis is performed offline. This is accomplished by “sorting” the data files to extract the information of particular interest. For the analog-digital system, the overall DAQ rate is limited by the maximum rate of events from any individual detector, which cannot be so large that tail pulses occur so frequently that they pile up on each other. Such pile-up would cause the effective height of the pulses to not accurately reflect the energy deposited in the detector, as the pulses were shifted upward by riding on the tails of previous pulses, introducing a variable voltage offset that cannot be discerned by the ADC. The maximum single-detector rate achievable was about 5 kHz.

The energy calibration for CEASAR was determined by placing a standard ^{152}Eu gamma-ray source, with beam off, in place of the target at the center of the array. Peaks corresponding to well-known transitions from the decay of ^{152}Eu were fitted* in the energy range from 122 to 1408 keV. The measured peak positions were compared with the known energies of those transitions to establish a linear energy calibration that could then be applied to the in-beam data.

A standard ^{133}Ba source covering energies 53 to 384 keV was used, in addition to the ^{152}Eu source, in a relative-efficiency calibration. Here, the peak areas were determined from fits and compared with the known gamma-ray intensities,[†] I_γ , for each peak; the ratio of events recorded in the peak, N , to the known intensity yields the relative efficiency of the array at that gamma-ray energy, $\varepsilon_\gamma = N/I_\gamma$. As the absolute activity (number of decays per second) of each source was not well defined, only a relative efficiency was determined; that is, the fraction of emitted gamma rays that would be recorded at one energy compared to the fraction at another energy was established but not the absolute probability of a gamma ray of

* The fit uses a Gaussian shape atop a (locally) linear background.

† Gamma-ray intensities give the fraction of nuclear decays for which that gamma ray is emitted, thus reflecting the probability of a specific cascade.

a specific energy being recorded. The relative efficiency data were fitted to a well-known [71] empirical function, written here as

$$\ln(\varepsilon_\gamma) = \left[(A + Bx + Cx^2)^{-G} + (D + Ex + Fx^2)^{-G} \right]^{1/G}, \quad (9)$$

where $x = \ln(E_\gamma)$, and A through G are parameters of the fit. The efficiency curve for the HPGe detectors is a maximum around 130 keV and falls off gradually at higher energies due to the increasing probability that the gamma ray scatters out of the crystal, depositing only part of its energy. This Compton scattering will be discussed in more detail later. At energies below the maximum efficiency, the curve drops precipitously as there is increased likelihood that low-energy gamma rays will be attenuated by absorption in the outer casing of the detector, in the vacuum chamber containing the target, and even within the target itself. The efficiency curve for CAESAR determined from the current experiment is shown in Fig. 14. The calibration sources did not provide gamma-ray peaks beyond 1408 keV, but the efficiency is a smooth function at higher energies and its behavior is sufficiently well-known to permit extrapolation for this experiment, particularly as absolute measurements of gamma-ray yields were precluded for other reasons mentioned later.

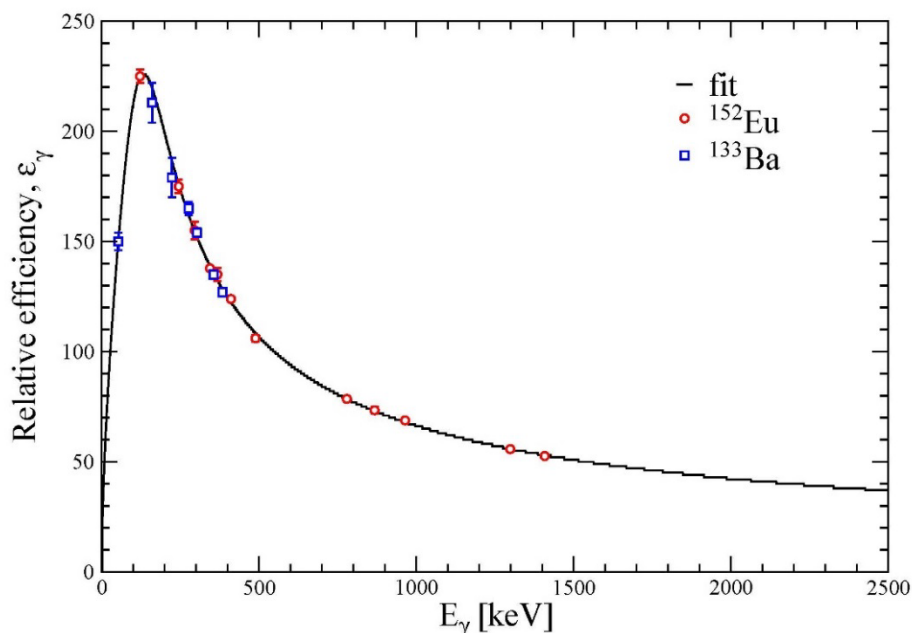


Fig. 14 Relative efficiency as a function of gamma-ray energy for the CAESAR array, determined from ^{152}Eu and ^{133}Ba calibration sources

The experiment was performed in 2 primary modes:

- Excitation function: The beam energy was varied from 56 to 35 MeV in 3.5-MeV steps. A single target foil was used to sample a modest energy

range of the excitation function. Data were recorded in “singles” (i.e., no gamma-ray coincidence requirement was imposed*).

- Coincidence measurements: Two beam-energy settings were used, 52 MeV and then 30 MeV. The target comprised a stack of 3 foils. Data were recorded with the requirement that at least 2 gamma rays were observed in separate coaxial HPGe detectors within a sub-microsecond time window. The data acquisition rate for this condition was limited to about 2 kHz due the single-detector rate limit.

The detected fold[†], F , is the number of HPGe detectors registering gamma rays emitted within a coincidence time window. A fold $F = 1$ corresponds to singles, while $F = 2$ corresponds to “doubles”.[‡]

In the first mode, data were recorded for 1.5 to 2 h at each of the 7 beam-energy settings. A current monitor provided a value proportional to the total number of ^7Li beam ions that were incident on the target, but the monitor was not calibrated so the absolute number of ions could not be determined. For each beam setting, a singles gamma-ray spectrum was constructed from the 9 coaxial HPGe detectors cumulatively from the beam-on periods. Singles spectra were also created from the cumulative beam-off periods, normalized such that the 1363-keV peaks in those spectra, from the cascade beginning with decay out of the 6.85-h $^{93\text{m}}\text{Mo}$ isomer (see Fig. 12), were the same height as in the corresponding beam-on spectra. The normalized beam-off spectra were then subtracted from the beam-on spectra, greatly reducing the background contributions from long-lived activity that built up during the course of the entire experiment. The background-subtracted spectra for beam energies 52.5 and 38.5 MeV are provided in Fig. 15. (While many of the same peaks are common to both spectra, their relative heights are different, sometimes drastically.)

* A singles spectrum is produced by combining separate spectra obtained from individual detectors. Prior to being combined, the individual spectra must be “gain matched.” All detectors and their individual signal-processing electronics are unique, such that without gain matching the gamma rays with the same energy may appear at slightly different locations in the spectra. The response of the detector and electronics may also drift with time, although the likelihood of this effect is reduced by achieving good climate control for the system. Gain matching is performed by examining the individual spectra and determining what modification to a linear energy calibration function is necessary so that all peaks from the same gamma rays appear at the same location. Without gain matching, peaks in the combined spectrum would be smeared out, and the overall energy resolution degraded. Because a singles spectrum may be stored as a vector, with each cell therein containing the number of counts in that channel of the spectrum, this is often referred to as 1-D data.

[†] The detected fold is related to the physical multiplicity, or number of gamma rays emitted by a single nucleus in a rapid sequence of decays (a cascade).

[‡] Doubles is often referred to as γ^2 or $\gamma\text{-}\gamma$ data. A doubles spectrum can be stored as a matrix and thus may be called 2-D data.

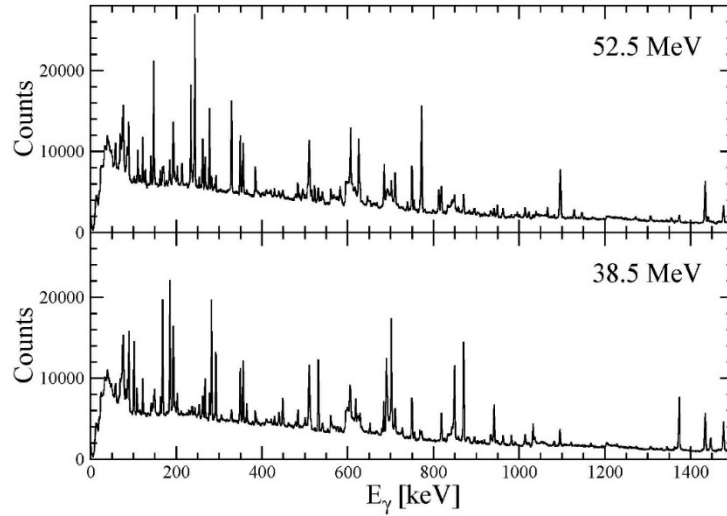


Fig. 15 Gamma-ray singles spectra from the CAESAR array at beam energies 52.5 MeV (top) and 38.5 MeV (bottom) during beam-on periods, after background subtraction of normalized beam-off spectra

The background-subtracted singles spectra were examined to determine the overall yield of each nucleus produced in the reaction. If a particular nucleus was too weakly produced for even its strongest known gamma-ray emission to be clearly visible in the singles spectra at any of the beam energies, it was left out of the analysis. For ^{92}Mo and ^{94}Mo , where all of the excited state decay cascades feed through a single gamma ray to the ground state (1510 and 871 keV, respectively), the yield determination was straightforward. This simply required fitting the areas of the observed peaks for the sole transitions terminating at the respective ground states to determine the corresponding number of events, N , and dividing that value by the array efficiency at that gamma-ray energy. The corresponding intensity, $I_\gamma = N/\epsilon_\gamma$, reflects the relative number of those nuclides produced per total ^7Li charge delivered to the target in a given irradiation. Other nuclei have multiple paths to the ground state, in which case the intensities of all transitions terminating at the ground state were summed. The presence of a long-lived isomer prevents decay cascades from reaching the ground state on the time scale of the beam-on periods; in these cases, the total yield being “trapped” in the isomer was additionally determined. For example, ^{91}Nb has 3 strong ground-state transitions with energies 1791, 1985, and 2291 keV, as well as a weak 1082-keV transition feeding a 60.86-day isomer; the combined intensity of all these branches was summed to determine the overall ^{91}Nb yield. For ^{93}Mo , the yield of most interest for this project is that of the 2425-keV, 6.85-h isomer rather than the yield of the ground state. The $^{93\text{m}}\text{Mo}$ yield was determined by the sum of the intensities for the 2475 and 1735 keV transitions, although only the former (accounting for about one-third of the sum) would be useful as a signature of NEEC, as discussed later.

The gamma-ray intensities reflecting production of specific nuclei (and for production of the $^{93\text{m}}\text{Mo}$ isomer) were normalized by the relative number of beam ions delivered to the target, determined by the current monitor at each beam energy. This put all measured yields on the same footing by removing the effect of varying beam intensities during different irradiations. These normalized values are plotted in the top panel of Fig. 16, in comparison with the PACE estimations described earlier (bottom panel). Overall, the gross features of the experimental and simulated curves are in good agreement. For the most part, the yield for each reaction channel peaks as predicted by PACE; for example, ^{92}Mo (formed by the emission of a proton and 4 neutrons from the compound nucleus, or “p4n channel”) rises in cross section at high beam energies and ^{94}Mo (p2n channel) rises for lower values, both peaking outside the measured range. The $^{93\text{m}}\text{Mo}$ isomer reaches a maximum around 7 MeV/A although, surprisingly, the yield does not fall off by much at higher energies. This latter observation was noteworthy, as it meant that the final NEEC experiment could be run at a higher beam energy and still have a substantial cross section for $^{93\text{m}}\text{Mo}$ production.

There are other clear differences between the measured and simulated yields: ^{91}Nb is a stronger channel than predicted, with a yield comparable to or greater than that of $^{93\text{m}}\text{Mo}$, whereas ^{94}Tc never rises any higher than ^{94}Mo in the data. The conclusions that can be drawn from these results are that beam energies above 7 MeV/A would be rather effective for $^{93\text{m}}\text{Mo}$ production and, although there are differences between the data and calculations, the expected nuclei are present, just with somewhat different relative contributions to the background for ^{93}Mo than predicted.

In the second mode of the experiment, events with 2 or more coincident gamma rays detected following reactions between a 52-MeV ^7Li beam and a ^{90}Zr target stack with an effective thickness of 58 μm were recorded for approximately 42 h.

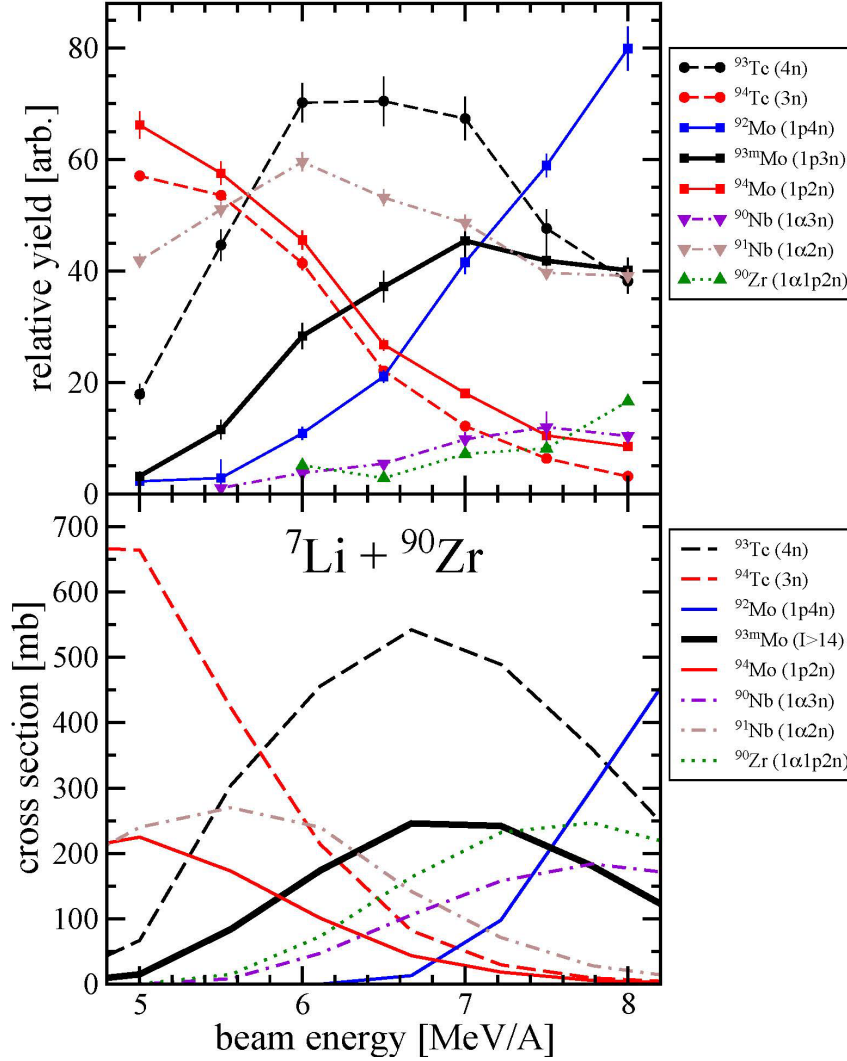


Fig. 16 Measured vs. estimated excitation functions for products of ${}^7\text{Li} + {}^{90}\text{Zr}$ reactions. (Top) Measured excitation functions for the most strongly produced nuclides. The numbers of protons (p), neutrons (n), and alpha particles (α) evaporated from the compound nucleus for each reaction channel are indicated in parentheses in the legend. (Bottom) Cross sections for the same reactions, estimated by PACE (similar to Fig. 10).

In the offline analysis, events with $F = 2$ were extracted (“unfolded”*) from the $F \geq 2$ recorded data and stored into a matrix. Rows and columns of this γ - γ matrix are identified by indexed coordinates (i, j) that correspond to gamma-ray energies. Thus, a specific event with coincident gamma rays having energies $E_{\gamma 1}$ and $E_{\gamma 2}$, being registered by 2 separate detectors, increments the number of events stored in the matrix at indices corresponding to $(E_{\gamma 1}, E_{\gamma 2})$. The matrix is also, by convention, incremented at the symmetric location $(E_{\gamma 2}, E_{\gamma 1})$. A column of the matrix, labeled

* If an event has n gamma rays, it can be unfolded into $n(n-1)/2$ unique pairs. All possible pairs will appear in a γ - γ matrix.

by a specific value of index $j = J$ and having coordinates (i, J) , contains the energy spectrum of recorded gamma rays that were detected in coincidence with gamma rays whose energy corresponds to that value of $j = J$. Likewise, due to the symmetry of the matrix, row $i = J$ in the matrix contains a spectrum identical to that contained in column $j = J$. By selecting, or gating, a specific row (or column) corresponding to a certain gamma-ray energy, it is then possible to extract from the matrix a spectrum containing all other gamma rays that occur in coincidence with that gamma ray. Of course, the detector resolution causes peaks to be spread over a Gaussian shape so that in practice, one selects a small range of channels to capture all events coincident with that gamma ray.

Summing the spectra for all rows (or all columns) produces what is called the total projection of the matrix, effectively collapsing the 2-D information into a 1-D spectrum. This 1-D spectrum will bear some similarity to the singles spectrum for the entire detector array, with many of the same peaks being present, but is not truly a singles spectrum. There will be differences in relative peak heights because of the imposed requirement that at least 2 gamma rays were detected here, whereas no such condition exists for a true singles measurement. For example, suppose a particular nuclide possessed a high-lying excited state that emitted a single gamma ray in a transition to the ground state. That gamma ray would appear in the singles spectrum from a multidetector array but would not appear in the projection of a γ - γ matrix from the array, since that gamma ray would never be in coincidence with another gamma ray. The total projection of the γ - γ matrix for beam energy 52 MeV is shown in Fig. 17a.

The total projection is used to identify gamma-ray peaks of interest and to gate on them to generate coincidence spectra. For example, the total projection from the CAESAR data is shown in Fig. 17a. Gating on the 1478-keV peak (off scale in Fig. 17a) produces the 1-D (“gated”) spectrum shown in Fig. 17b, which reveals coincident gamma rays at 685, 268, and 263 keV, as would be expected for the level scheme shown in Fig. 12. Also as expected, no gamma-ray peak from the 1363-keV transition that is parallel to the 1478 keV in the decay scheme is found in this spectrum. The data are recorded on an event-by-event basis, so a coincidence between 1363- and 1478-keV gamma rays cannot occur, because one nucleus cannot emit both those gamma rays.

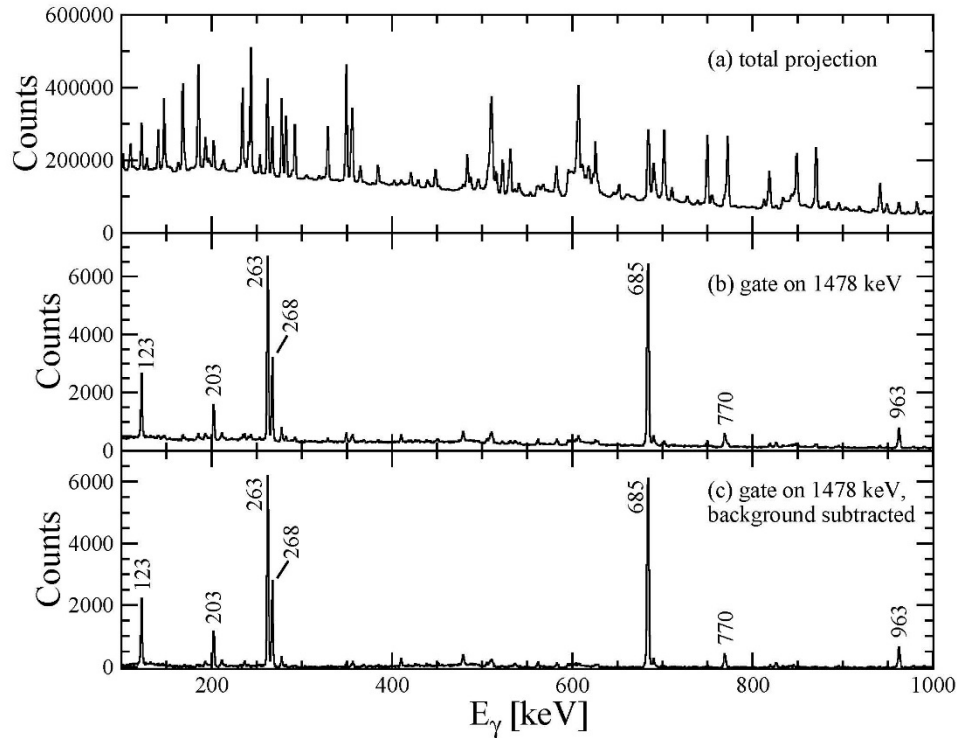


Fig. 17 Projected spectra from a γ - γ matrix obtained with CAESAR. (a) Total projected spectrum. (b) Projected spectrum after gating on the 1478-keV transition in the total projection. (c) Projected spectrum after gating on the 1478-keV transition and after background subtraction.

Beneath and beside the discrete peaks in the spectra of Figs. 17a and 17b lies a smooth background arising largely from gamma rays that scattered and deposited only part of their energy within the HPGe detector (this type of event will be discussed in more detail later). A gate on a peak in the total projection produces a 1-D spectrum that includes all events that are in coincidence with the peak upon which the gate was made, whether they were due to full- or partial-energy deposition by coincident gamma rays. A well-known procedure exists by which to remove the background contribution in the gated spectrum [72] and was used to produce the background-subtracted gated spectrum of Fig. 17c.

Analysis of the γ - γ matrix was performed using the ESCL8R program within the RadWare suite [71,73]. This program provided a convenient means by which to apply gamma-ray energy gates, perform background subtraction, fit energies and efficiency-corrected intensities, and construct level schemes. The latter was accomplished by examining all combinations of 2-gamma-ray coincidences, considering energy sums (if the decay between 2 states can occur via 2 or more different paths, the sum of the gamma-ray energies for each path must be consistent), and intensity balances (the amount of intensity, or number of

transitions, feeding into a state should balance that which exits the state*). All of these factors must be considered when deciding where a particular gamma-ray transition should be placed in a level scheme.

Construction of the ^{93}Mo level scheme proceeded initially by searching for and verifying coincidence relationships as presented in previous work, particularly that of Fukuchi et al. [51]. For the most part, the CAESAR data confirmed the level scheme proposed in that work, with slight differences in gamma-ray energies that are typically within the experimental uncertainties. There was insufficient intensity in the present data to verify a few of the highest-lying transitions. The level scheme deduced from the 52-MeV $^7\text{Li} + ^{90}\text{Zr}$ matrix is shown in Fig. 18 (the partial level scheme of Fig. 12, emphasizing the transitions feeding the 6.85-h isomer and related to NEEC, was confirmed). The green lines enclose those portions that are new to this work.

Despite the observation of several new states and transitions in the ANU data, no new paths directly feeding the isomer were identified. Thus, the known 2475-keV transition remains the best signal that the 6.85-h isomer has been populated. Although the 1735-keV gamma ray would similarly guarantee the isomer has been fed, the presence of a 0.8-ns short-lived isomer above it at the $J^\pi = (27/2^-)$ state delays that decay path sufficiently such that the recoiling ^{93}Mo ions would already come to rest before reaching the $21/2^+$ isomer. That would prevent any possibility that NEEC could occur for nuclei reaching the 6.85-h isomer via the 1735-keV transition.

* Not all intensity feeding into a state may necessarily be observed. Multiple feeding transitions, each too weak to observe as identifiable peaks, may cumulatively contribute a significant amount. Therefore, it is typically assumed that the intensity out of a state is at least as much as what is observed feeding into it. An exception is when a state is an isomer; for long enough half-lives, the decay out may not be observed at all in an experiment.

background contributions (from ^{93}Mo itself or other reaction products) that would lead to a false indication of NEEC in the final experiment. The 685-keV line above the 6.85-h isomer is problematic because of the proximity of its energy to that of the 686-keV transition lying below that isomer and immediately above the 1478-keV transition (see Fig. 12); a gate on the 2475-keV gamma ray will pick up the upper 685-keV transition rather strongly, giving a false positive if confused with the 686-keV transition. However, the other combinations, 2475–268 keV and 2475–1478 keV can be considered. Figure 19 shows the background-subtracted spectrum gated on 2475 keV, from the γ - γ matrix for the 52-MeV beam energy. The expected peaks are present, as indicated by their energy labels. The region where a peak corresponding to the 268-keV gamma ray from depletion-level decay only shows statistical fluctuations around zero events. Thus, the 2475–268-keV coincidence would be a “clean” signature for NEEC. In the same spectrum, there is a slight excess of counts above background at 1478 keV, but not so substantial an amount as to consider there to be a 2475–1478-keV coincidence. The NEEC experiment will require a triple, or γ - γ - γ , coincidence to look for these signals, to further reduce the likelihood of any non-NEEC contributions as a false positive in the measurement.

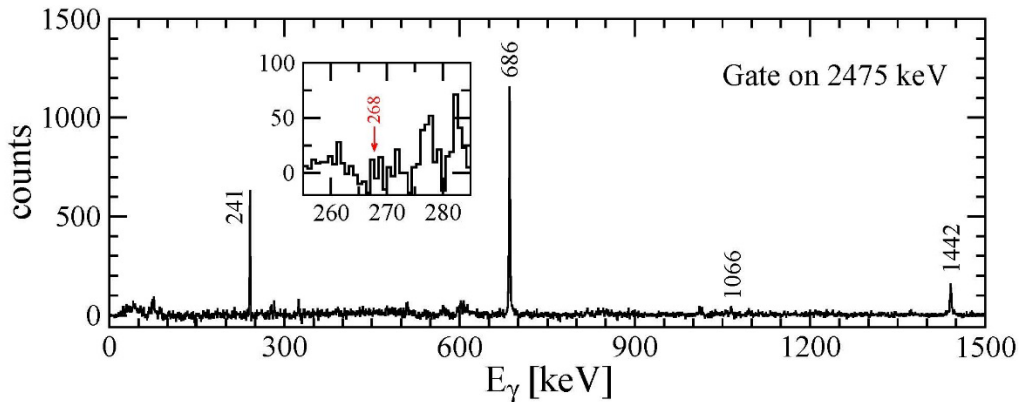


Fig. 19 Background-subtracted spectrum gated on the 2475-keV transition from CAESAR. The data were taken at a beam energy of 52 MeV. The expanded region in the insert demonstrates the absence of a 2475–268-keV coincidence.

The above analysis was performed for the 52-MeV data. As the NEEC experiment was expected to span a broad range of beam energies, due to slowing of ^{90}Zr ions over a spatial range as discussed in Section 3.2.2, the 30-MeV normal-kinematics data were similarly examined to check for possible interference with the NEEC-signature coincidences. In that data set, there was minimal yield for the 2475-keV gamma ray, as can be inferred from the excitation function in Fig. 16, so a gate in the matrix at that energy resulted in only statistical fluctuations at 263 and 1478 keV, with net counts consistent with zero.

To summarize the results of the ANU measurements, all 3 primary goals for the benchmark experiment were achieved. The expected yields were measured over a range of beam energies to provide excitation functions, and these confirmed that the ${}^7\text{Li} + {}^{90}\text{Zr}$ reaction would be a suitable choice for populating ${}^{93\text{m}}\text{Mo}$. Surprisingly, the yield of the long-lived isomer persists to higher beam energies than estimated from PACE simulations. The level scheme for ${}^{93}\text{Mo}$ populated with this beam–target combination was developed, identifying several new levels and gamma rays. A newly discovered 262-keV transition lies close in energy to one of the characteristic NEEC lines, 268 keV, but is not coincident with the primary gating transition, 2475 keV, and thus would not interfere with the NEEC signature. Finally, there is no evidence that the NEEC-signature coincidences would be contaminated by background from other sources. These results provided the benchmark information needed to run the NEEC experiment.

A final point was analyzed from the CAESAR data, in addition to the primary aspects. As was argued in Section 3.1, one reason for running an experiment in normal kinematics is that Doppler broadening of gamma-ray peaks can generally be avoided because the recoiling nucleus comes to rest before decaying. However, stopping of a fast ion takes a finite time, albeit short for solid targets. If the cumulative time for a specific decay cascade to occur is sufficiently fast (less than a picosecond), a nucleus may still be in motion when a gamma ray is emitted. The recorded energy is a function of how fast the nucleus was moving at the time of emission as well as the location of the detector relative to the recoil direction (see Eq. 8 and associated discussion in Section 3.1). There are a variety of experimental techniques that exploit this relationship between level half-lives and Doppler-shifted energies for particular purposes (e.g., Dewald et al. [74]). For the NEEC experiment, the time scale for feeding the 6.85-h isomer could be a complicating factor.

The half-life for the state emitting the 2475-keV transition is not known, nor are those of the higher-lying levels that feed it from above [4,51]. In order for NEEC to occur for ${}^{93}\text{Mo}$, the 6.85-h isomer must be populated before the recoiling ions slow too much in the medium, since at some point in the medium E_{NEEC} will become too small. It is the cumulative time from when ${}^{93}\text{Mo}$ was produced in a high-energy state until the cascade of gamma rays terminates at the long-lived isomer via the 2475-keV transition that is of concern. This is only a potential problem for the final NEEC experiment, but the normal kinematics reaction at ANU could be analyzed to estimate the effective half-life for the 2475-keV decay path. To accomplish this, data were extracted into nonstandard matrices that were asymmetric, where rows identified gamma rays recorded by either of 2 detectors in CAESAR that were located at a specific angle with respect to the beam direction (the “ θ ” dimension of

the matrix), and columns were identified with gamma rays observed in any other detector (the “any” dimension of the matrix). Three matrices were created in this way, with “ θ ” dimensions for specific angles of 145° , 97° , and 49° .

A gate was placed on the 686-keV peak on the “any” dimension of each matrix, and background subtraction was performed on the resulting 1-D spectra. If the cumulative decay time was short enough, the 2475-keV peak in the 97° spectrum would be near its true energy (see Eq. 8), with $\theta = 97^\circ$; CAESAR did not have detectors at exactly $\theta = 90^\circ$, while those at 49° or 145° would be shifted to, respectively, higher or lower energies. No such shifts were observed in the data. This provided an estimate of about 0.5 ps for the lower limit of the effective lifetime for the 2475-keV decay, based on the calculated stopping time of ^{93}Mo recoils in zirconium (the target used at ANU) and for an initial recoil energy of 3.6 MeV (corresponding to a 52-MeV beam energy). If the cumulative time for transitions feeding the 6.85-h isomer was any shorter than this, Doppler shifts would have been observable in the ANU data. Unfortunately, it was impossible to determine from these data the precise cumulative feeding time. For the inverse-kinematics NEEC experiment, where the ^{93}Mo recoils will initially move at a higher velocity than at ANU, it would take longer for those ions to slow down in a ^7Li target, but within a few picoseconds, the ion kinetic energy would drop below the NEEC threshold. Thus, only if the cumulative feeding time is in the range of approximately 0.5 ps to a few picoseconds would NEEC be able to occur in the inverse-kinematics experiment while being consistent with the observations in the ANU data.

4.2 NEEC Experiment (Inverse Kinematics)

A proposal for a NEEC demonstration experiment using a solid ^7Li target and the $^7\text{Li}(^{90}\text{Zr}, p3n)$ reaction was submitted for consideration by the ATLAS PAC in February 2015 and was designated as “Priority I” by the PAC during its March 2015 meeting. This priority is reserved for “those experiments that must be run at all costs” and indicates the potential scientific impact of a demonstration of NEEC, a new physical effect that has never previously been observed. Due to the need for the results of the benchmark experiment conducted at ANU, the NEEC test at ANL was scheduled for December 2015. A typical duration of 5 continuous days, plus 1 day for beam tuning,^{*} was allocated for the experiment.

^{*} The accelerator must be adjusted for each different isotope used as beam projectiles, plus the beam must be directed through various focusing and guiding magnets into the Gammasphere target chamber. This is collectively called *beam tuning*.

Gammasphere* at ANL is the premier instrument for gamma-ray spectroscopy, shown in Fig. 20, being an array of typically 101† HPGe detectors in a geometry that covers 46% of 4π solid angle around a central cavity [75–77]. The central cavity typically encloses a vacuum chamber in which targets are placed for irradiation by particle beams from ATLAS. Each detector is almost completely surrounded by a secondary bismuth germanate (BGO) scintillator crystal detector. When gamma rays enter an HPGe detector, some may deposit all their energy by the photoelectric effect and be recorded in a spectral peak.‡ However, some gamma rays may enter the detector and interact through the Compton effect [78], where a scattered photon takes some of the incident energy out of the detector. Those events will not contribute to a full-energy peak but instead will contribute to a continuous background within the spectrum at lower energies than that of the incident gamma ray. This background not only reduces the number of events in a particular gamma-ray peak, but also makes more difficult the identification of other peaks that sit on that background. The energy deposited by this type of interaction, E_{dep} , is related to the incident gamma-ray energy, E_γ , and the angle, θ , with which the outgoing photon is scattered with respect to the original gamma-ray direction:

$$E_{dep} = E_\gamma \left(\frac{E_\gamma}{E_0} \right) (1 - \cos \theta) \left(1 + \left(\frac{E_\gamma}{E_0} \right) (1 - \cos \theta) \right)^{-1}, \quad (10)$$

where $E_0 = 511$ keV is the rest mass of the electron. Since different Compton-scattering events occur with different directions for the outgoing photon, a continuous background is accumulated called the Compton continuum. The BGO crystal surrounding the HPGe detector serves to reduce the size of the Compton continuum, improving the signal-to-noise ratio for full-energy peaks. When Compton scattering occurs in an HPGe detector, the outgoing photon may enter the associated BGO crystal and interact with it by depositing energy. In this case, both the HPGe detector and its BGO “shield” produce electrical signals within a small window of time, on the order of 10–20 ns, and such events are discarded [78]. The process is not perfect, in that some scattered photons may not interact with the BGO shield, but it nevertheless significantly lowers the Compton background.

* Gammasphere is considered to be a third-generation multidetector array. Fourth generation arrays are under development in the United States and Europe, but only partial systems have been constructed as prototypes.

† Gammasphere can physically accommodate 110 detectors, although 9 locations are typically empty at any time due to detector maintenance.

‡ Referred to as the full-energy peak for that gamma ray.

Performance of the Compton shields for Gammasphere detectors is described in Carpenter et al. [79].*

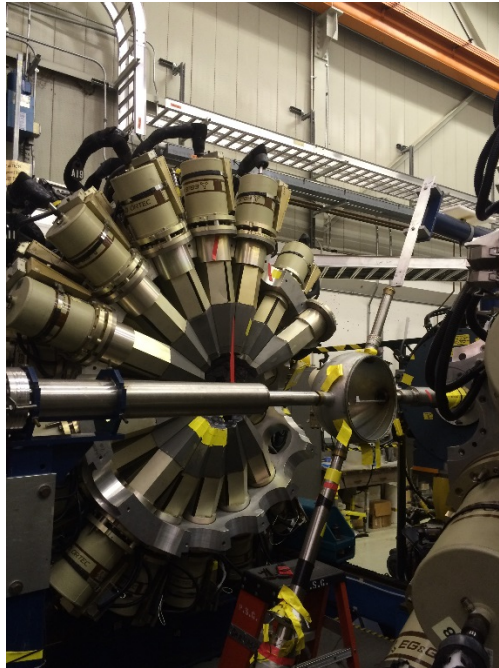


Fig. 20 Photograph of Gammasphere at Argonne National Laboratory. The 2 hemispheres of the array are separated for access to the target chamber, which is also open. The array is closed around the target chamber prior to measurements. As in Fig. 13 for CAESAR at ANU, the tan/brown cylinders are LN₂ dewars used to keep the HPGe detectors at operating temperature. The BGO shield scintillators are the faceted surroundings of the HPGe detectors themselves, which are aimed toward the central cavity. The beam enters from the left into the target chamber through a vacuum pipe (the target chamber is also evacuated during measurements).

The great strength of Gammasphere lies in its ability to record large numbers of higher-fold events. The efficacy for recording high-fold events is strongly correlated with the number of detectors and their solid-angle coverage, their energy resolution, and the total efficiency of the array considered as a single effective detector [80]. Gammasphere is characterized by a total efficiency of about 10% for detection of 1-MeV gamma rays. Considering all factors, the array then provides the best signal-to-noise ratio for gamma-ray peaks corresponding to a detected fold $F \sim 5$ [81], assuming that nuclei exhibit cascades with a physical multiplicity of 30. In contrast, CAESAR has an efficiency of about 1.5% at 1 MeV, providing its best signal-to-noise ratio at $F \sim 3$. The cascades for the product nuclei observed in the ANU experiment have a smaller physical multiplicity so that the optimum detected folds for those nuclei with Gammasphere and CAESAR are estimated to be closer

* Compton shields were available for detectors in the CAESAR array but were not operational during the ANU benchmark experiment.

to 4 and 2, respectively. The sensitivity limit, or limit of observation, for an array reflects the fraction of all gamma-ray-emitting events for which a gamma-ray cascade of given physical multiplicity could be detected with a specified fold. For Gammasphere and CAESAR, the sensitivity limits at their optimum detected folds are 5×10^{-5} and about 10^{-3} , respectively. This illustrates that Gammasphere is capable of detecting events that occur much more rarely, even when a requirement for more coincident gamma-ray detections is imposed. CAESAR is most typically used to acquire doubles data ($F = 2$), since the number of events recorded at higher folds is greatly reduced. Gammasphere is typically used to acquire γ - γ - γ “triples” or higher-fold data.

In this experiment, Gammasphere was operated in the “Digital Gammasphere” (DGS) mode, a relatively recent development [82]. With DGS, the analog tail pulses from the HPGe detectors (if not vetoed by the associated BGO shield detectors) and some of their preceding background are immediately sampled at a rate of 100 MHz, thus 10 ns between sample points. The data stream includes the identification of which HPGe detector received a gamma ray, a time stamp that is generated online from the HPGe detector’s signal, and a partial record of its sampled waveform, as well as similar information for the associated BGO shield. It is more difficult to analyze data in real time, since considerable data processing is necessary to determine the energy deposited in a detector from the waveform. Also, the size of the data from an experiment becomes much larger: about 11 TB from this experiment, compared to about 15 GB for the ANU experiment. However, the great advantage to using DGS is that it greatly increases the rate at which useful data can be acquired. The key point is the ability to allow some pile-up of pulses, as depicted in Fig. 21.

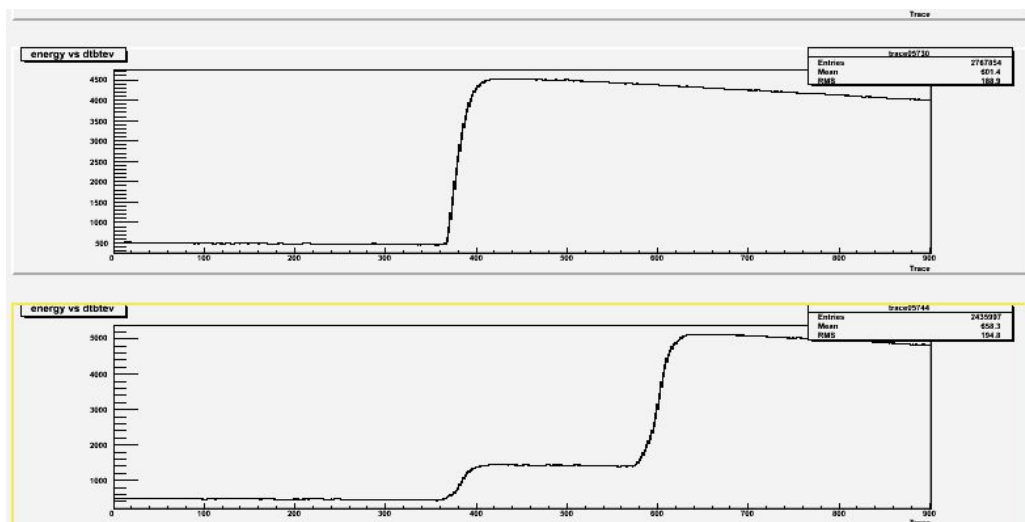


Fig. 21 Gammasphere HPGe detector tail pulses without pileup (top) and with pileup (bottom), from Carpenter [83]

The top panel of Fig. 21 shows part of a single Gammasphere HPGe detector pulse with a long exponentially falling tail (truncated in the figure). The background is established by the level to the left of the pulse (corresponding to early times before the detector pulse is generated), which is at ground so that the height at the peak of the tail pulse can be accurately determined by a peak-sensing ADC [78]. This is not the case for the second (right-most) pulse shown in the lower panel of Fig. 21. The peak voltage of that second pulse above ground is not the correct height of the pulse, as it rides on the tail of the earlier (left-most) pulse; typically, both pulses would be discarded in an analog-digital system. In the DGS system, both pulses can be used, provided that it is possible in post- (offline) processing to determine the height of the background just before the second pulse and then subtract that from the height of the second pulse. While the single-detector rate for Gammasphere in the DGS mode was only used up to 5.5 kHz, this could have been as high as 20 kHz. More importantly, data were acquired with the condition that 3 or more gamma rays were coincident, at a rate near 50 kHz. Thus, the much larger size of the recorded data from DGS was due to a higher event rate and the greater length of the data packets.

The precise target configuration and beam energy for the experiment were determined from the NEEC requirements. Figure 22 shows the calculated recoil energies of ^{93}Mo ions after production by the $^7\text{Li}(^{90}\text{Zr}, \text{p}3\text{n})$ reaction in comparison with the kinetic energies at which those resonances could, in principle, participate in NEEC. If a ^{90}Zr beam energy of 6.7 MeV/A was used in the experiment, this would mean that, at best, only those atomic resonances accessible at recoil energies below about 6 MeV/A could contribute. Of course, this assumes that the ^{93}Mo was produced immediately after the zirconium ions entered the lithium and that the 6.85-h isomer was populated immediately. Zirconium ions traveling into the medium farther before reacting would have been slowed so that the ^{93}Mo recoil energy might be too small for NEEC to occur. Also, if ^{93}Mo was produced immediately upon ^{90}Zr entering the medium, but the molybdenum traveled too far before the long-lived isomer was populated, the ^{93}Mo ions might be moving too slowly for NEEC to occur. These types of scenarios were discussed previously. Ultimately, it was decided to select a ^{90}Zr beam energy of 8.3 MeV/A. At this energy, ^{93}Mo ions would have the optimum production cross section over a spatial and energy range, the best chance of populating the isomer, and the opportunity to access more of the NEEC resonances during slowing in the medium.

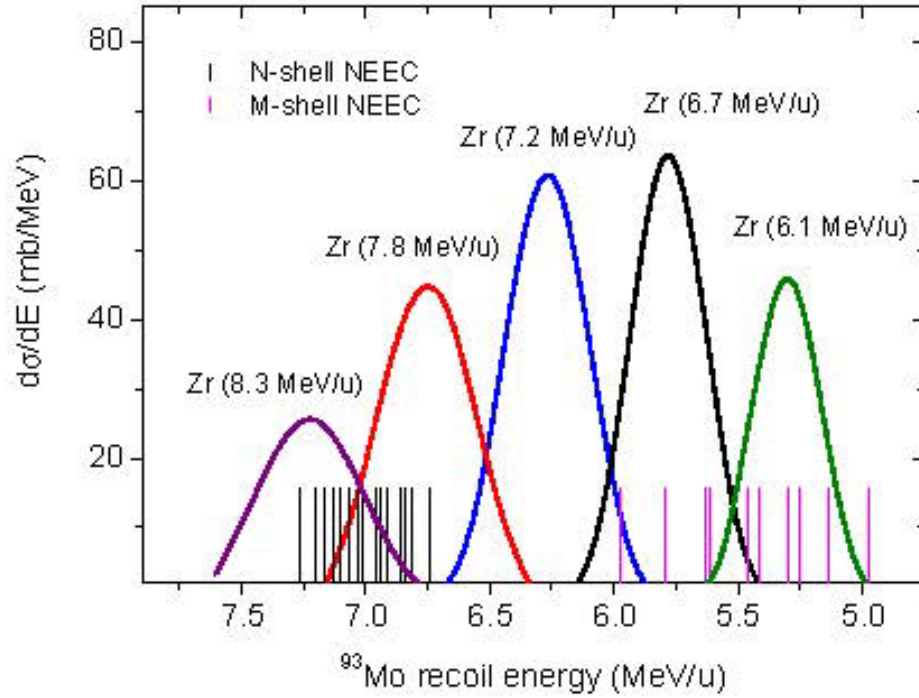


Fig. 22 Energies of ^{93}Mo ions recoiling after production in a ^7Li target in inverse kinematics for different choices of incident ^{90}Zr energies given in parentheses. Note that “MeV/u” is used equivalently to “MeV/A”. The distributions of recoil energies are due to the possibility that evaporated nucleons from the reaction could be emitted isotropically, with those emitted backward adding to the recoil energy of the ion, etc. Also shown are ^{93}Mo kinetic energies (indicated by the vertical bars) for which NEEC resonances are possible, if the mean charge state of the ions is appropriate (see Fig. 23 and discussion in the text). The $d\sigma/dE$ reflects the probability of ^{93}Mo production.

Figure 23 shows the mean charge state of ^{93}Mo ions, as well as the distribution of charge states around that mean value, as a function of recoil kinetic energy. The kinetic energies at which NEEC resonances could occur are indicated by the vertical bars, while the upper termination of the bar indicates the charge state for which the necessary atomic vacancy would exist for NEEC to occur. For a ^{90}Zr beam energy of 8.3 MeV/A, the ^{93}Mo recoil at about 7.25 MeV/A would permit NEEC through several vacancies in the atomic N subshell, provided ^{93m}Mo was populated before the ions slowed below about 6.75 MeV/A. As the ions slowed further, if NEEC had not already occurred, this process could take place via the atomic vacancies in the M subshell between about 5.8 and 5.25 MeV/A. Again, 8.3 MeV/A for the ^{90}Zr beam energy appeared to provide the greatest chance of observing NEEC.

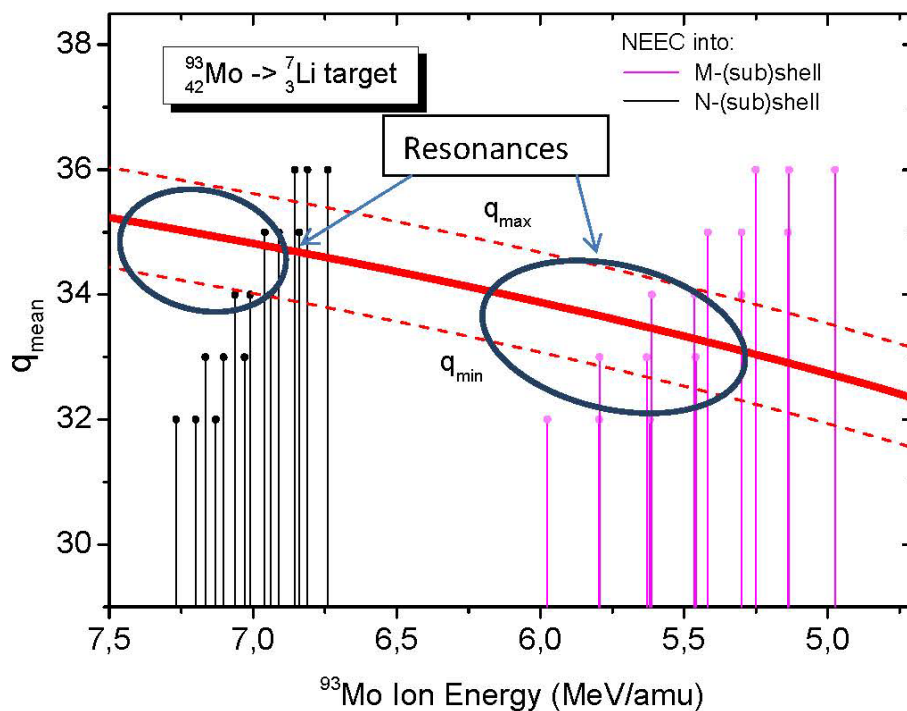


Fig. 23 Mean charge state for ^{93}Mo ions and locations of NEEC resonances in lithium as a function of the ion kinetic energy per nucleon. The mean value is indicated by the solid red line as the ions slow in the medium, while the minimum and maximum charges states are denoted by the dashed red lines. As the ions slow, the mean charge state diminishes; when fully stopped, the charge state would be zero and the atoms neutral. The vertical bars show at which kinetic energies NEEC resonances could occur, provided the mean charge state given by the top of the bars lies within the values achieved by the ions at those energies.

A technique for production of lithium targets was previously developed at ANL [64], taking into account the need to avoid exposure to oxygen to prevent degradation of the material. The method involved evaporation of isotopically enriched pure lithium from a heated crucible within a large vacuum chamber. The evaporated lithium can be deposited on a thin foil of another material, which is held within a standard Gammasphere target frame. The frame is made of 2-mm aluminum and has a 1.2-cm-diameter hole, with the outer square dimension being about 2 cm. The frame is attached to the end of a long rod which enters the large vacuum chamber from a smaller, portable chamber that is temporarily attached during target preparation. After evaporation of lithium on the foil held by the frame, the rod is extracted from the large chamber back into the small chamber, past a ball valve. The valve is closed, and the small chamber is unbolted from the evaporation chamber and moved (to minimize the time under which the target is not being pumped) to Gammasphere. There the small chamber is attached, the ball valve opened, and the target extended down into the target chamber, which is being pumped. Figure 24 shows a photograph of this latter part of the process. Thickness

of the lithium was measured in the deposition chamber as described in McCutchan et al. [64]. However, the deposition scheme limited the maximum thickness* to that which was achieved here, 5 mg/cm^2 ,† or $93 \text{ }\mu\text{m}$.



Fig. 24 Photograph showing installation of the target transfer vacuum chamber to the Gammasphere target chamber, as described in the text

The effective thickness of the lithium as seen by the accelerated beam would be larger in the Gammasphere target chamber, since the target rod extends into the target chamber at a 27° angle with respect to the vertical direction. Also, the rod could be rotated around its axis so that an additional 45° could be introduced. This required a modification to the internal support structure within the target chamber. Together, these 2 angles could be used to increase the effective thickness by a factor of 1.6 over the true thickness, giving about 8 mg/cm^2 . The slowing of ions within this thickness should be sufficient for NEEC to occur, based on the chosen ^{90}Zr energy, but insufficient to completely stop the ^{93}Mo ions. Since it was desired that transitions emitted after decay of the depletion level were from stopped nuclei, so their peaks would not be Doppler shifted or broadened (or those ions simply lost down the beamline after passing through the lithium), a 33 mg/cm^2 foil of enriched ^{208}Pb was used as backing on which the lithium would be deposited. The isotope

* After one deposition the target could be withdrawn into the attached transfer chamber, the intervening ball valve closed, the deposition chamber opened, and then more raw lithium added. This was done to achieve the maximum thickness of lithium used in the experiment but was generally to be avoided as this process unavoidably introduced oxygen into the chambers.

† Expressing thicknesses as mass per area is typical in nuclear spectroscopy, as it is easy to determine the mass under irradiation by multiplying by the beam spot size. The mass in these units is easily converted to thickness in length dimensions by dividing by the mass density of the material.

^{208}Pb was selected due to its high atomic number, $Z = 82$, which would mean that molybdenum or zirconium ions could not cause nuclear reactions with the backing foil*, provided those ions were sufficiently slowed by the lithium, and because the first excited state of that nuclide was at 2615 keV; Coulomb excitation† of that state would not interfere with measurement of the gamma rays from ^{93}Mo . The target configuration is shown in Fig. 25. The challenges to achieving this lithium thickness and modifying the target chamber should not be minimized. The heat load on the lithium/Pb target was analyzed but not found to be an issue at the anticipated beam currents.

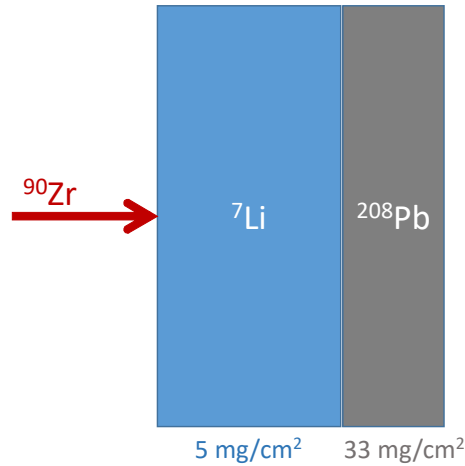


Fig. 25 Schematic of Pb-backed lithium target for the NEEC experiment. Thicknesses are given in units of mass/area, typical in nuclear spectroscopy. Note that the ^{90}Zr beam did not enter at normal incidence during the experiment.

The $^7\text{Li}/^{208}\text{Pb}$ target was prepared just before the start of the experiment to minimize exposure to oxygen, while ATLAS was being tuned for the required ^{90}Zr beam energy of 8.3 MeV/ A . The beam current varied between 1 to 2 nA which, since the ^{90}Zr ions were stripped to a charge of 19^+ for acceleration, corresponded to $(3-6) \times 10^8$ particles/s. The standard ATLAS beam structure was used, in which subnanosecond beam pulses were delivered every 82.47 ns. Gammasphere was operated in DGS mode, with events recorded for $F \geq 3$ and this beam current at a rate of about 50 kHz. A total of 92 HPGe detectors were in place because of unexpected maintenance caused by previous experiments at ATLAS by other

* A nuclear reaction can only occur if the projectile and target nuclei can approach each other sufficiently closely. This is not possible if the energy required to overcome their electrostatic repulsion (the “Coulomb barrier”), which depends on the atomic numbers of both projectile and target nuclei, is greater than the kinetic energy of the projectile. For the ^{90}Zr beam, ^{93}Mo and all other nuclides produced by reactions in the lithium, by the time they could reach the backing foil their energies would be less than their individual Coulomb barriers with Pb.

† In Coulomb excitation, a projectile and target exchange energy through their electric and magnetic fields as they pass each other. This is not strictly a nuclear reaction but can result in excitation of states in both projectile and target nuclei.

groups. In DGS, signals from groups of 5 HPGe detectors and their associated BGO shields are processed in a digitizer module and recorded in files on disk. Individual files were restricted to about 1 h of data acquisition.

To analyze the data, it is first necessary to merge the separate files from each of these modules into a file that contains all information recorded from the entire detector array. Merging takes about the same amount of time as the original data acquisition, since all events must be read from and written to disk. Then the relevant data for a singles spectrum or matrices must be extracted (sorted) from the merged data. As mentioned previously, real-time data analysis is much more challenging with DGS, but this mode had the advantage of providing a much higher rate of data acquisition. Data were stored in raw (unmerged) form on 1-TB disks, whose capacity corresponded to about 10 h of the experiment; larger disks were not used to minimize the danger of data loss should a single disk fail. When a raw data disk was full, DAQ was halted briefly and a new disk was inserted prior to restarting the experiment. In the interest of time, merging of the raw data on the just-removed disk began immediately on another system so that partial analysis of the data (construction of singles spectra and matrices) could be done in as close to real time as possible to assess the quality of the data and make on-the-fly modifications to the experiment. Data were merged on a set of 4-TB disks so that a backup copy of all the data was available on different disks from the raw data. The raw and merged data occupied about 11 TB each. It was also possible during parts of the experiment to have detector signals processed simultaneously and independently by the Analog Gammasphere (AGS) analog-to-digital system. The AGS system could not process the full rate of events but permitted some degree of real-time monitoring.

During the first approximately 24 h of the experiment, data were merged and then analyzed to ensure that recorded gamma rays did indeed correspond to nuclides produced via $^{90}\text{Zr} + ^7\text{Li}$ reactions. Singles spectra were also obtained by summing spectra from specific rings of HPGe detectors in Gammasphere, where each ring corresponded to a specific angle θ with respect to the beam direction in order to assess which gamma rays had been Doppler shifted. The shift observed by detectors at $\theta = 90^\circ$ would be minimal, while forward (backward) detectors would register higher (lower) energies. The velocity of ions emitting those gamma rays could be determined using Eq. 8 and the corresponding ion kinetic energy calculated. Figure 26 shows singles spectra (in red) obtained from rings of detectors at $\theta = 90^\circ$ (top panel) and $\theta = 79^\circ$ (bottom panel), focusing attention on the region containing the 2475-keV gamma ray that feeds the 6.85-h isomer. Analysis of these spectra indicated a significant problem with the experimental setup.

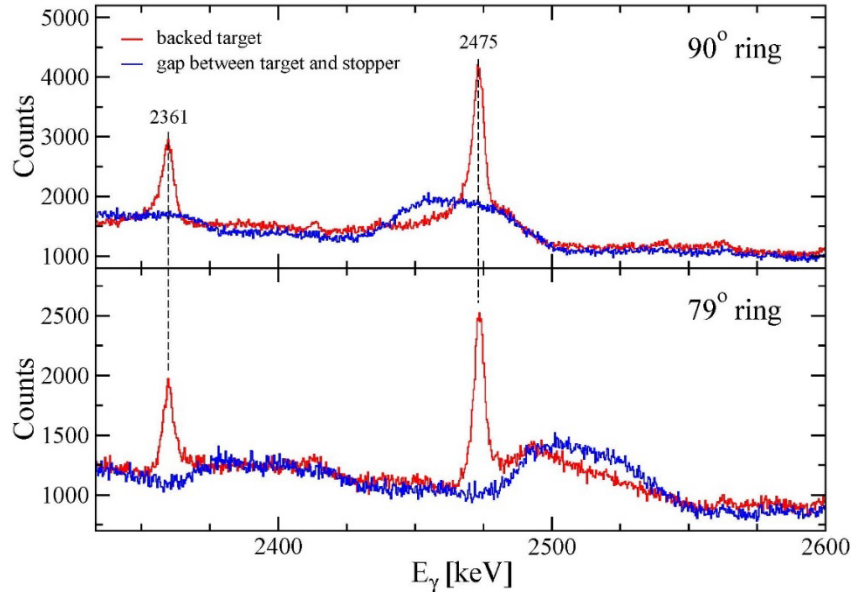


Fig. 26 Singles spectra for rings of detectors in Gammasphere showing Doppler shifts for the 2475-keV gamma ray at $\theta = 90^\circ$ (top) and $\theta = 79^\circ$ (bottom). Spectra in red were obtained from the Pb-backed target, while those in blue were obtained from the gap target.

Doppler broadening and shifting of some 2475-keV gamma rays are most apparent in the forward ($\theta = 79^\circ$) ring in the bottom panel of Fig. 26. The broad structure appearing between about 2475 and 2550 keV corresponds to 2475-keV gamma rays emitted from moving ^{93}Mo nuclei, while the narrow peak at 2475 keV corresponds to gamma rays emitted from stopped nuclei. The broadening of the peak is due to the angular width $\Delta\theta$ of the detectors within the ring and cannot be reduced as long as gamma rays are emitted by moving nuclei. The amount of the shift could be estimated from the distance from the centroid of the broadened structure to the narrow “stopped” peak at 2475 keV. However, the main conclusion is reached simply by noting that a significant number of 2475-keV gamma rays were recorded in the stopped peak. For those events, ^{93}Mo nuclei had come to rest in the Pb prior to population of the 6.85-h isomer. Recall that in order for NEEC to occur, ^{93}Mo nuclei must be moving with kinetic energies large enough to access the atomic vacancies shown in Fig. 23 after population of the 6.85-h isomer, which means after emission of the 2475-keV gamma ray. The presence of a stopped peak at this energy completely precludes NEEC for the corresponding emitting nuclei. The numbers of events in the stopped peak and those in the broadened/shifted structure for 2475 keV were estimated to be approximately equal, meaning that one cumulative half-life for the transitions feeding the isomer had occurred prior to reaching the Pb backing. Based on the transit time for ions through the effective 8 mg/cm^2 lithium layer, the cumulative feeding half-life was estimated to be on the order of 5–10 ps, consistent with the lower limit of 0.5 ps estimated from the ANU experiment. With

this half-life, NEEC would not be able to occur under this beam/target configuration.*

A new target configuration was designed and constructed to compensate for the previously unknown slowness of the transitions feeding the $^{93\text{m}}\text{Mo}$ isomer but still using the $^{90}\text{Zr} + ^7\text{Li}$ combination for its production. The essential point was the introduction of a gap within the target that would allow produced ^{93}Mo to coast in vacuum so that there was more time to feed the 6.85-h isomer without concurrently slowing beyond the kinetic energy needed for NEEC. As shown in the left panel of Fig. 27, a 1.55 mg/cm^2 thickness of lithium was created for production of ^{93}Mo . This layer was evaporatively deposited on a thin ($\sim 500 \text{ } \mu\text{g/cm}^2$) ^{12}C foil. In this configuration, the backing foil behind the lithium production target should not stop the recoiling ^{93}Mo ions, and such thin carbon foils were available and within the experience of the ANL target maker. A vacuum gap of approximately 3 mm was arranged behind this production target by using 2 consecutive Gammasphere frames back-to-back. The NEEC target on the back was prepared from a 4.2 mg/cm^2 ^{12}C foil backed by a 33 mg/cm^2 ^{208}Pb foil. A dual deposition of lithium for both production and NEEC media was infeasible for the “gap” target, but calculations indicated that carbon would provide NEEC resonance conditions similar to lithium (see Fig. 28, compared to Fig. 23). Construction required some time, including testing different options, so DAQ was continued with the original target during that time to avoid ATLAS/Gammasphere being idle. A total of 34 h of data was obtained with the original target. After construction of the new target, it was transferred to the Gammasphere target chamber in the small vacuum chamber. A 45° twist of the target rod was no longer required to achieve the necessary effective thickness, so the standard internal support structure was returned to the target chamber. This retained the 27° target angle with respect to the beam direction, with a corresponding 12% increase in effective thicknesses and size of the gap. A photograph of this second target within the target chamber is shown in the right panel of Fig. 27.

* It is also possible that the gamma rays emitted by stopped ^{93}Mo may have been from nuclei produced deeper within the lithium, and thus there was less time for them to reach the 6.85-h isomer than for those nuclei produced early in the lithium. However, in either case those ions would not be traveling fast enough for NEEC to occur.

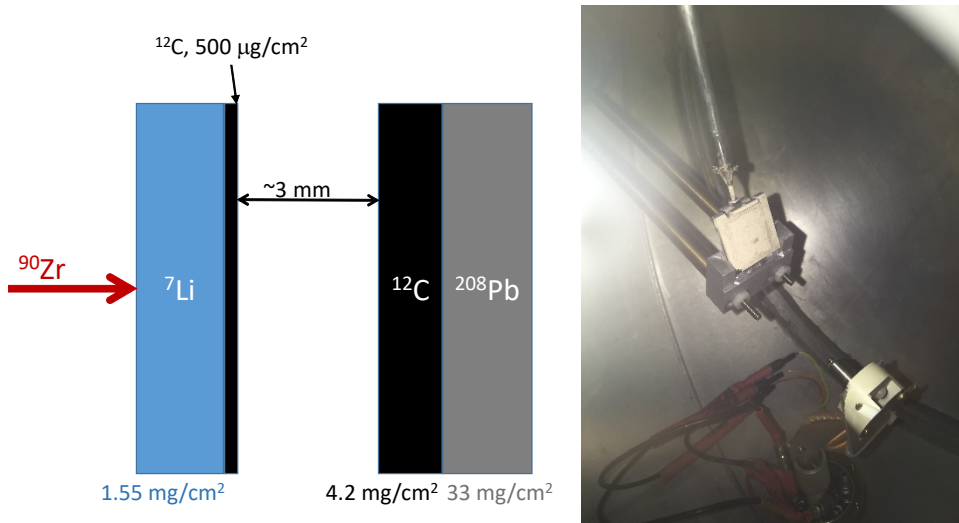


Fig. 27 Gap target configuration and position inside the Gammasphere target chamber. The configuration is shown schematically in the left panel. Note that the ^{90}Zr beam did not enter at normal incidence during the experiment. The right panel shows a photograph of the target, taken through a small viewing port, when positioned within the Gammasphere target chamber. The beam enters from the lower right and, since taken after the experiment, the target shows a discoloration from the beam spot. The target rod, entering from the top of the photograph, introduced a 27° angle with respect to the perpendicular to the beam direction; no 45° twist of the rod was used.

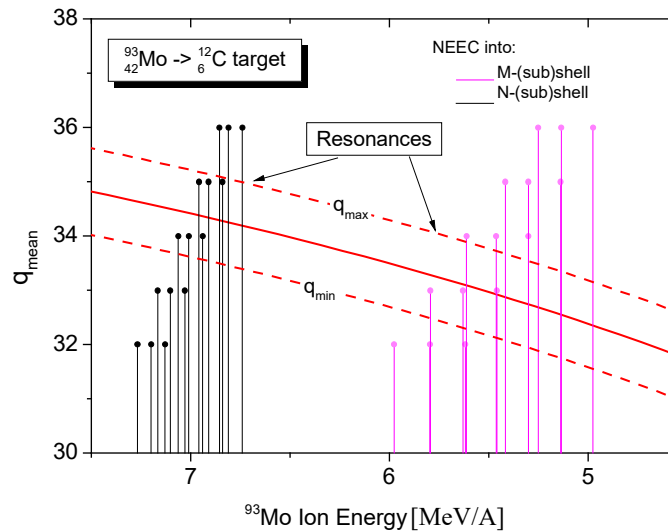


Fig. 28 Mean charge state for ^{93}Mo ions and locations of NEEC resonances in carbon as a function of the ion kinetic energy per nucleon. The mean value is indicated by the solid red line as the ions slow in the medium, while the minimum and maximum charge states are denoted by the dashed red lines. As the ions slow, the mean charge state diminishes; when fully stopped, the charge state would be zero and the atoms neutral. The vertical bars show at which kinetic energies NEEC resonances could occur, provided the mean charge state given by the top of the bars lies within the values achieved by the ions at those energies.

Irradiation began on the new target, with the previous ^{90}Zr beam energy of 8.3 MeV/A, and data were collected for 12 h. Those data were merged and sorted to produce spectra for analysis, including those shown in blue in Fig. 26. No stopped 2475-keV peak is evident in spectra obtained from those detector rings, and only Doppler broadened and shifted structures are seen that correspond to this gamma ray. The Doppler shift was estimated to correspond to an average ^{93}Mo ion kinetic energy of 4.66 MeV/A ($\beta \approx 0.100$), too low to access any atomic vacancies for NEEC as seen in Fig. 28 [84]. This was surprising, since the lithium thickness, measured within the deposition chamber, and that of its carbon backing, should not have been sufficient to slow the ^{93}Mo ions to that degree. The effective thickness of lithium that would produce this slowing would be about twice that measured for the actual lithium layer within the evaporation chamber. An error of more than about 10% in that measurement was unlikely. Thus, the consensus conclusion was that oxygen had either entered into the transfer chamber during transport, or the target chamber when the transfer chamber was attached or when the target rod was extended. Lithium and oxygen form Li_2O , which has a larger stopping power* for ions compared with lithium [47]. The color of Li_2O is described as “grey”, compared to “silver-white” for pure lithium. It is difficult to assess the target color accurately from the photograph in Fig. 27, but it may be closer to grey. To accommodate the unexpectedly larger energy loss within the “lithium” layer, the ^{90}Zr beam energy was raised to 9.3 MeV/A, the highest available from ATLAS during the experiment. With this energy, recoiling ^{93}Mo ions entering the gap should have a velocity $\beta \approx 0.109$ for a kinetic energy of 5.54 MeV/A. This would enable access to atomic vacancies in the M subshell but not any of the N subshell vacancies (see Fig. 28). A problem with ATLAS required repair, plus there was some retuning necessary so that 10 h of beam time were lost before the experiment could resume with the higher energy and gap target. The remaining 62 h of beam time acquired data under these conditions.

The analysis of the coincidence data from the inverse-kinematics reaction at ANL is more complex than that of the normal-kinematics reaction at ANU. In the latter, the slow ^{93}Mo recoils typically were stopped in the target before emitting most of the identified gamma rays, thus the peaks in the gamma-ray energy spectra had narrow widths (stopped peaks). In the Gammasphere data, there was a mix of narrow peaks and broadened structures. Those gamma rays that were emitted promptly, on the order of picoseconds or less, exhibited large Doppler shifts from recoil velocities up to about 11% the speed of light, with the measured peak shifting according to Eq. 8. In principle, knowing the recoil velocity and the angle θ to the

* Stopping power is defined as the differential change in ion kinetic energy per differential distance traversed in a medium, dE/dx .

detector, as shown in Fig. 5, it is possible to invert Eq. 8 to obtain the true energy of the gamma ray from the measured Doppler-shifted energy. Applying such a “Doppler correction” can correct the centroids of shifted peaks separately for each ring of detectors, at a specific θ , returning them to their true energies. For a 2475-keV gamma ray emitted from a nucleus moving at $\beta = 0.1$, the energies that would be recorded in detectors at $\theta = 50^\circ$ and at $\theta = 130^\circ$, for example, are 2632 and 2314 keV, respectively; summing the spectra for all angles from 50° to 130° would thus result in a peak smeared out by more than 300 keV. Applying a Doppler correction greatly reduces this spread but does not eliminate it completely.

The intrinsic resolution of the detector is typically not recovered in the Doppler correction, however, because of 2 other aspects related to the Doppler shift. One is that the detectors are not point-like but have physical size, covering a range of angles θ . It is not known event-by-event where in the crystal the gamma ray entered a detector, so θ is taken to be the value at the center of the detector. The spread in θ values over the face of a given detector translates to a spread in the measured gamma-ray energies (see Eq. 8), and thus even in a spectrum from a single detector, a Doppler-shifted peak would be broader than the intrinsic resolution. It is also clear from Eq. 8 that a nonuniform velocity, such as that experienced by a recoiling ion slowing down in a target medium, will result in Doppler shifts of different magnitudes. However, the variation in β is not very large in the case of gamma decays from recoils that have entered the gap in the target. These effects explain why in the spectra observed at 90° (top panel of Fig. 26) the 2475-keV peak is substantially broader when there is a gap in the target (blue spectrum), which allows the ^{93}Mo recoil ions to maintain a large velocity, than when decays can occur after the nucleus has come to rest (narrow peak in the red spectrum).

To search for evidence of the 2475–268–1478 keV triple coincidence, a signature for NEEC in this experiment, the following method was used for a preliminary analysis on a subset of the full data set. As the 2475-keV transition is prompt, it experiences a Doppler shift, as discussed previously. The 268- and 1478-keV gamma rays, however, lie beneath the 3.5-ns depletion level and would be emitted after the recoil ion has stopped in the target. Therefore, a matrix was created from the recorded $F \geq 3$ data from pairs of gamma rays coincident with the broad 2475-keV structure, after Doppler correcting the positron of that structure. The Doppler correction was based on Eq. 8 for the 2475-keV structure using the specific angle of the detector with respect to the beam and using $\beta = 0.109$.* No Doppler correction was applied to gamma rays in the matrix. Gamma-ray peaks due to

* This average value of β was obtained by comparing forward and backward rings of detectors at angles θ and $180^\circ - \theta$, measuring amount by which the centroids of the peaks for the 2475-keV gamma ray were shifted, and calculating the value of β that would be required in order to line up the peaks at the same energy.

stopped nuclei would be narrow while any corresponding to moving nuclei would be broadened. This γ - γ matrix could then be analyzed in a fashion similar to that described for the ANU experiment.

The spectrum from a gate on 1478 keV was projected from the matrix. For this preliminary analysis, a simpler form of background subtraction than found in Radford [72] was employed. A gate of similar width was placed in a region of the total projection where the background was flat, at slightly higher energy than 1478 keV. The latter spectrum was subtracted from the one gated on 1478 keV. This background-subtracted spectrum is shown in Fig. 29a. Several peaks for coincident gamma rays are observed. As can be seen in the level scheme in Fig. 18, these are all in coincidence with the 1478-keV transition, but many are not expected to be found in a 2475-keV gated spectrum, even if NEEC has occurred. This is because the background for the 1478-keV gate has been subtracted but not the background for the 2475-keV gate performed by the data sort. To account for the latter component, another matrix was sorted with the condition that a gamma ray with a Doppler-corrected energy from a region above 2475 keV (on a flat background, in an energy window the same width as used for the 2475-keV gate) was present in the event. Placing the same gates on the peak and background regions for the 1478-keV transition in the second matrix as were used for the first matrix, another background-subtracted spectrum was produced. This spectrum is shown in Fig. 29b, plotted with the same vertical scale as Fig. 29a. The same peaks found in the first spectrum are also seen here with similar peak heights. Finally, the background-subtracted spectrum from the second matrix was subtracted from the first spectrum to remove the contributions from background associated with the 2475-keV gate. The resulting spectrum is provided in Fig. 29c. Most of that spectrum contains only statistical fluctuations about zero, with key exceptions for the 268- and 685-keV lines, which have a clear excess of counts above background. These are exactly the transitions that are expected if NEEC has occurred, in coincidence with each other and with Doppler-shifted 2475-keV gamma rays.

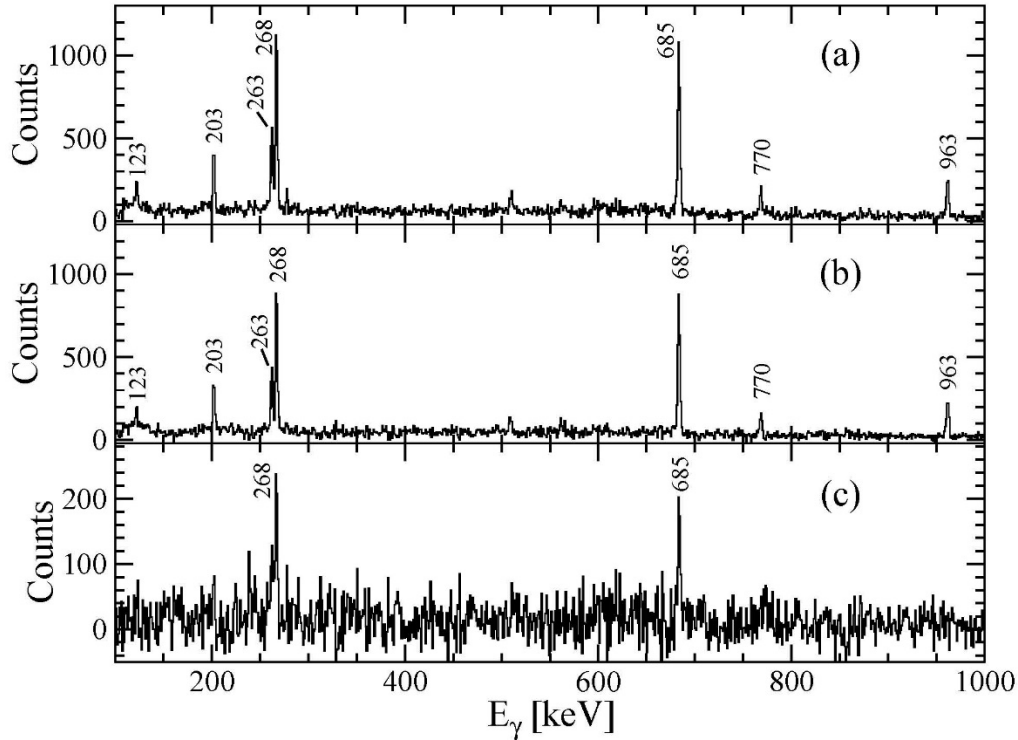


Fig. 29 Spectra projected from gamma-gated γ - γ matrices obtained with Gammasphere. (a) Projected spectrum after gating on the 1478-keV transition in a matrix that was created with the condition that all pairs of coincident gamma rays were also coincident with a 2475-keV transition in the corresponding event. A Doppler correction with $\beta = 0.109$ was applied for the gate on 2475 keV, while the energies in the matrix had $\beta = 0$. Background for the gate on 1478 keV was subtracted but not for the gate on 2475 keV. (b) Projected spectrum similar to (a) but from a matrix created with a required coincidence with a gamma ray in a background region at energies somewhat higher than the 2475-keV peak. (c) The spectrum formed by subtracting the spectrum in (b) from the one in (a). The observed peaks indicate that both 268- and 685-keV gamma rays below the depletion level of ^{93}Mo are coincident with both the 1478- and 2475-keV transitions. Although there are several sources of background that could give a false positive, this is the first evidence of NEEC, demonstrated through isomer depletion.

The apparent coincident 2475–(268 and 685)–1478 keV NEEC signatures represented by Fig. 29c are promising, but caution is in order. These triple coincidences may arise from various sources of background. There may be erroneous correlations (so-called random coincidences) between unrelated events due to the high DAQ rates or due to the manner in which the coincidence time window can be defined for DGS data. This definition may be inadequate to distinguish signals originating from successive beam bursts. Also, additional background events can enter the gate on the shifted 2475-keV peak when detectors at backward angles are included, since the Doppler shift may move the broad 2475-keV structure to lower energies such that it overlaps with peaks from stopped gamma rays. Each of these potential issues can be addressed. Ultimately, a more rigorous treatment of the background is needed, along the lines of that given by

Starosta et al. [85]. A narrower gamma–gamma time-difference window (~ 40 ns compared to the 200 ns used here) can also be imposed to avoid improper correlations between unrelated events from separate beam bursts. To prevent the inclusion of events from the stopped peaks in the 2475-keV gate, only detectors at those angles where there is clear separation between the 2475-keV peak and the contaminants would be used for gating. These are planned for the next stage of the analysis, plus the full data set will be examined.

In addition to having the ANU benchmark experiment as a control for the NEEC measurements, there is also a type of “internal control” that can be exploited to test the veracity of a claimed observation of NEEC. Parallel to the 2475-keV decay to the 6.85-h isomer is a path through the 1735-keV transition that lies beneath the 0.8 ns, $J^\pi = (27/2^-)$ state (see Figs. 12 and 18). The delay in this decay sequence is long enough for the ^{93}Mo recoils to come to rest before the isomer is fed by the 1735-keV transition. Therefore, if the above analysis of gamma-gated matrices is repeated but with the 1735-keV gamma ray replacing 2475 keV, there should be *no* evidence of NEEC. If the origin of the 268- and 685-keV lines in Fig. 29c is due to random correlations with other events rather than true coincidences, this should be evident in the 1735–1478 keV double gate as well.

If evidence for the 2475–(268 and 685)–1478 keV coincidences persists under further intense scrutiny, it can be claimed with some confidence that the NEEC phenomenon has been observed for the first time. For a direct test of the theoretical predictions, the probability for NEEC can be quantified by double gating on the 1442- and 686-keV transitions, which lie immediately above the 2475-keV transition (see Fig. 18) and would be broadened by Doppler shifts, and measuring the efficiency-corrected areas of the 2475- and 268-keV peaks. The intensity of the 2475-keV transition would indicate how many times the $^{93\text{m}}\text{Mo}$ isomer was populated, and the 268-keV gamma-ray intensity would give the fraction of those that subsequently followed the NEEC decay path, resulting in isomer depletion.

Future efforts related to this experiment will include a determination of the Gammasphere efficiency, similarly to that done for CAESAR. After the end of the experiment, while Gammasphere was otherwise idle during beam tuning for the next scheduled experiment by another group, measurements were taken with a ^{56}Co source from which to determine the efficiency. That source provides gamma rays with energies up to 3.547 MeV [86]. The Gammasphere efficiency will be similar to that shown in Fig. 14 for CAESAR but with the peak shifted to slightly higher energies due to the use of larger detectors. Considerable additional analysis will be performed to examine the issues of random coincidences and other contributions to background and potential false positives. A “presort” program has very recently been constructed, with which the merged full data (containing detector pulse

samples) can be transformed into collapsed data containing only the final necessary form of detector identification, time stamp, and pulse height. Data files presorted in this way will be about 17 times smaller and, thus, more amenable to extracting many different coincident gamma sets (cubes or matrices) for analysis. Also, collaborating scientists from ANL have suggested that following maintenance to ATLAS, it will be possible to provide sufficient beam energy to consider use of a different reaction involving $^{89}\text{Y} + ^9\text{Be}$ for a further test of NEEC with Gammasphere. A gap target using beryllium would be easier to construct than the present lithium target, with less susceptibility to contamination, and the higher-energy ^{89}Y beam may provide recoiling ^{93}Mo nuclei with sufficient kinetic energy to access all available atomic vacancies for NEEC.

5. Summary

The ability to utilize nuclear-scale energy storage for Army applications would represent a revolutionary advance. It would also require significant strides in the understanding of the process by which external agents can interact with nuclei to control the rate at which energy is released. Under some conditions, indirect energy-release mechanisms mediated by atomic electrons may prove more efficient in comparison with direct interaction with nuclei. This project represents an attempt to demonstrate, for the first time, the process called Nuclear Excitation by Electron Capture (NEEC), and obtaining a signal of that process through an induced energy release from a nuclear metastable state (isomer). Extensive effort was conducted to evaluate potential experimental approaches and to design and conduct benchmark and NEEC-demonstration studies at state-of-the-art facilities. The NEEC experiment was conducted in December 2015 so that only preliminary analysis of a subset of that data (about 11 TB, in toto) has been performed thus far. Initial results show the first evidence of this previously unobserved physical effect, although there are several potential sources of false positives that must be eliminated in a complete, detailed analysis. This report describes the development of the experimental approach and the analysis to date. In any case, this type of high-risk discovery research will be necessary to determine the feasibility of achieving revolutionary advances in energy density for applications.

6. References

1. D'Auria JM. The nation's needs for isotopes: present and future [presentation]. Washington (DC): Office of Nuclear Physics, Department of Energy (US); 2008.
2. Morgan JS. USASOC S&T Gaps. Ver. 4.1. Army Special Operations Command, Department of Science and Technology (US); 2012.
3. Office of the Under Secretary of Defense for Acquisition, Technology, and Logistics. Defense science board report on technology and innovation enablers for superiority in 2030. Washington (DC): Defense Science Board; 2013 [accessed 2016 Feb 29]. www.acq.osd.mil/dsb/reports/DSB2030.pdf
4. Evaluated nuclear structure data file (ENSDF). National Nuclear Data Center, Brookhaven National Laboratory [accessed 2016 Feb 22]. www.nndc.bnl.gov.
5. Hult M, Gasparro J, Marissens G, Lindahl P, Wätjen U, Johnston PN, Wagemans C, Köhler M. Phys Rev C. 2006;74:054311.
6. Casten RF. Nuclear structure from a simple perspective. 2nd ed. New York (NY): Oxford University Press; 2001.
7. deShalit A, Feshbach H. Theoretical nuclear physics. New York (NY): Wiley; 1990. (Nuclear structure; vol. 1).
8. Carroll JJ, Karamian SA, Rivlin LA, Zadernovsky AA. Hyperfine Int. 2001;135:3.
9. Sethi J, et al. J Phys G. 2015;43:015103.
10. Belic D, et al. Phys Rev Lett. 1999;83:5242.
11. Schumann M, Käppeler F, Böttger R, Schäölermann H. Phys Rev C. 1998;58:1790.
12. Stefanescu I, et al. Phys Rev Lett. 2007;98:122701.
13. Roig O, Méot V, Rossé B, Bélier G, Daugas J-M, Letourneau A, Menelle A, Morel P. Phys Rev C. 2011;83:064617.
14. Karamian SA, Carroll JJ. Phys Rev C. 2011;83:024604.
15. Carroll JJ, Litz MS, Netherton KA, Henriquez SL, Pereira NR, Burns DA, Karamian SA. AIP Conf Proc. 2013;1525:586.
16. Walker PM, Dracoulis GD, Carroll JJ. Phys Rev C. 2001;64:061302.

17. Pálffy A, Evers J, Keitel CH. Phys Rev Lett. 2007;99:172502.
18. Herzberg G. Atomic spectra and atomic structure. Mineola (NY): Dover Publications; 1945.
19. Eisberg R, Resnick R. Quantum physics of atoms, molecules, solids, nuclei, and particles. New York (NY): John Wiley & Sons; 1985.
20. Bigeleisen J, Mayer MG. J Chem Phys. 1947;15:261.
21. Wertheim GK. Mössbauer effect: principles and applications. New York (NY): Academic Press; 1964.
22. Akber A, et al. Phys Rev C. 2015;91:031301(R).
23. Morita M. Theor Phys. 1973;49:1574.
24. Tkalya EV. Nucl Phys. 1992;A539:209.
25. Kishimoto S, Yoda Y, Seto M, Kobayashi Y, Kitao S, Haruki R, Kawauchi T, Fukutani K, Okano T. Phys Rev Lett. 2000;85:1831.
26. Kishimoto S, Yoda Y, Kobayashi Y, Kitao S, Haruki R, Masuda R, Seto M. Phys Rev C. 2006;74:031301(R).
27. Claverie G, Aleonard MM, Chemin JF, Gobet F, Hannachi F, Harston MR, Malka G, Scheurer JN. Phys Rev C. 2004;70:044303.
28. Goldanskii V, Namiot VA. Phys Lett B. 1976;62:393.
29. Pálffy A, Scheid W, Harman Z. Phys Rev A. 2006;73:012715.
30. Gosselin G, Morel P. Phys Rev. 2004;C 70:064603.
31. Karamian SA. Phys Part Nucl (in Russian). 2008;39:950.
32. Zadernovsky AA, Carroll JJ. Hyperfine Int. 2002;143:153.
33. Hayes AB, et al. Laser Phys. 2007;17:745.
34. Levine MA, Marrs RE, Henderson JR, Knapp DA, Schneider M. Phys Scr. 1988;T22:157.
35. Elliott SR, Marrs RE. Nucl Inst Meth B. 1995;100:529.
36. Bernstein L. Lawrence Livermore National Laboratory, Livermore, CA. Personal communication, 2012.
37. Morel P, Daugas JM, Gosselin G, Meot V, Gogny D. AIP Conf Proc. 2005;769:1085.

38. Gosselin G, Meot V, Morel P. Phys Rev C. 2007;76:044611.
39. PHELIX laser facility [accessed 2016 Feb 22]. https://www.gsi.de/en/start/research/forschungsgebiete_und_experimente/appa_pni_gesundheit/plasma_physicsphelix/phelix.htm?nr=%2Fproc%2Fself%2Fenviron%27%22.
40. Roth M. GSI Helmholtzzentrum für Schwerionenforschung GmbH, Darmstadt, Germany. Personal communication, 2015.
41. Palffy A, Harman Z, Kozhuharov C, Brandau C, Keitel CH, Scheid W, Stohlker T. Phys Lett B. 2008;661:330.
42. Atanasov D et al. J Phys B. 2015;48:144024.
43. Storage Ring ESR [accessed 2016 Feb 22]. https://www.gsi.de/en/research/accelerator_facility/storage_ring.htm.
44. Litvinov Y. GSI Helmholtzzentrum für Schwerionenforschung GmbH, Darmstadt, Germany. Personal communication, 2016.
45. Beam Time at GSI (GSI Helmholtzzentrum für Schwerionenforschung GmbH) [accessed 2016 Feb 22]. https://www.gsi.de/de/work/organisation/wissenschaftliche_gremien/user/beam_time.htm?nr=%27%22.
46. Karamian SA, Carroll JJ. Phys Atomic Nucl. 2012;75:1362.
47. Ziegler JF. SRIM – the stopping and range of ions in matter [accessed 2016 29 Feb]. <http://www.srim.org>.
48. Betz HD. Rev Mod Phys. 1972;44:465.
49. Shima K, Kuno N, Yamanouchi M, Taware A. At Data Nucl Data Tables. 1992;51:173.
50. Liu W, et al. Nucl Inst Meth A. 2003;496:198.
51. Fukuchi T, et al. Eur Phys J A. 2005;24:249.
52. Hasegawa M, Sun Y, Tazaki S, Kaneko K, Mizusaki T. Phys Lett B. 2011; 696:197.
53. Richtmyer FK, Kennard EH, Cooper JN. Introduction to modern physics. 6th ed. New York (NY): McGraw-Hill; 1955.
54. Carroll JJ. FY14 Director's Research Initiative (DRI) proposal for enhanced control of nuclear lifetimes for ultra-energetic materials using electron-nuclear processes [unpublished]. Adelphi (MD): Army Research Laboratory (US); 2014.

55. PACE4 fusion-evaporation reaction code. Lansing (MI): National Superconducting Cyclotron Laboratory, Michigan State University; 2016 [accessed 29 Feb 2016]. <http://lise.nsl.msui.edu/pace4.html>.
56. Tarasov OB, Bazin D. Nucl Inst Meth B. 2003;204:174.
57. Schiwietz G, Grande PL. Nucl Inst Meth B. 2001;125:175–177.
58. Thompson A, et al. X-Ray Data Booklet. Berkeley (CA): Lawrence Berkeley National Laboratory; 2001.
59. Polasik M. Nicolaus Copernicus University, Toruń, Poland. Personal communication, 2013.
60. Safronova UI, Namba C, Murakami I, Johnson WR, Safronova MS. Phys Rev A. 2001;64:012507.
61. Rehm KE, Greene JP, Harss B, Henderson D, Jiang CL, Pardo RC, Zabransky B, Paul M. Nucl Inst Meth A. 2011;647:3.
62. Seweryniak D. Argonne National Laboratory, Lemont, IL. Personal communication, 2014.
63. Roeder B. Texas A&M University, College Station, TX. Personal communication, 2014.
64. McCutchan EA, Lister CJ, Greene JC. Nucl Instrum Meth A. 2009;607:564.
65. Research facilities in nuclear physics. Singapore: International Union of Pure and Applied Physics (IUPAP); 2007. IUPAP Report 41.
66. Heavy Ion Accelerator Facility [accessed 2016 Feb 22]. <https://physics.anu.edu.au/nuclear/hiaf.php>.
67. Günther C, Banaschik MB, Hübel H, Schneider WD, Gonsior KH, Herzog P. Nucl Phys A. 1975;244:287.
68. Chiara CJ, Carroll JJ, Lane GJ, Reed MW, Mitchell AJ, Hota S, Palalani N. Phys Rev C (in preparation for 2016).
69. Baglin CM. Nucl Data Sheets. 2011;112:1163.
70. Lane GJ. Australian National University, Canberra, Australia. Personal Communication, 2015.
71. Radford DC. Nucl Inst Meth A. 1995;361:297.
72. Radford DC. Nucl Inst Meth A. 1995;361:306.

73. RadWare [accessed 2016 Feb 22]. <http://radware.phy.ornl.gov/main.html>.
74. Dewald A, Harissopulos S, von Brentano P. Z Phys A. 1989;334:163.
75. Lee IY. Nucl Phys. 1990;A520:641c.
76. Lee IY, Deleplanque MA, Vetter K. Rep Prog Phys. 2003;66:1095.
77. Gammasphere [accessed 2016 Feb 22].
<http://www.phy.anl.gov/gammasphere/doc/index.html>.
78. Knoll GF. Radiation detection and measurement. 2nd ed. New York (NY): Wiley; 1989.
79. Carpenter MP, et al. Nucl Inst Meth A. 1994;353:234.
80. Beausang CW, Simpson J. J Phys G. 1996;22:527.
81. Nolan PJ, Beck FA, Fossan DB. Ann Rev Nucl Part Sci. 1994;44:561.
82. Anderson JT, et al. In: Proceedings of the Nuclear Science Symposium and Medical Imaging Conference (NSS/MIC). 2012 Oct 27 – 3 Nov; Anaheim, CA. p. 1082.
83. Carpenter MP. In: Workshop on Decay Spectroscopy at CARIBU: Advanced Fuel Cycle Applications, Nuclear Structure and Astrophysics. Lemont (IL): Argonne National Laboratory; 2011.
84. Polasik M, Rzadkiewicz J, Chiara CJ, Carroll JJ. Phys Rev A (in preparation for 2016).
85. Starosta K, Fossan DB, Koike T, Vaman C, Radford DC, Chiara CJ. Nucl Inst Meth A. 2003;515:771.
86. Barker PH, Connor RD. Nucl Instr Meth. 1967;57:147.

List of Symbols, Abbreviations, and Acronyms

ADC	analog-to-digital converter
AGS	Analog Gammasphere [mode]
ANL	Argonne National Laboratory
ANU	Australian National University
ARL	US Army Research Laboratory
ATLAS	Argonne Tandem Linear Accelerator System
BGO	bismuth germanate
BIC	bound internal conversion
CM	center of mass
DAQ	data acquisition
DGS	Digital Gammasphere [mode]
DRI	Director's Research Initiative
EBIT	Electron Beam Ion Trap
ESR	Experimental Storage Ring
FWHM	full-width-half-maximum
GSI	GSI Helmholtzzentrum für Schwerionenforschung GmbH
HIAF	Heavy Ion Accelerator Facility
HPGe	high-purity germanium
ICS	integral cross section
LLNL	Lawrence Livermore National Laboratory
NEEC	Nuclear Excitation by Electron Capture
NEET	Nuclear Excitation by Electron Transitions
ORAU	Oak Ridge Associated Universities
PAC	Program Advisory Committee
PACE	Projection Angular-momentum coupled Evaporation

SRIM	Stopping Range of Ions in Matter
STP	standard temperature and pressure
TAC	time-to-analog converter

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(PDF) IMAL HRA
RECORDS MGMT
RDRL DCL
TECH LIB

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K KRAPELS
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