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Development of a Multi-reflection Time-of-Flight Device for Separation of Isomeric States

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14. ABSTRACT
The benefits of developing a multi-reflection time-of-flight (MR-ToF) device for future experimental campaigns at the Facility for Rare Isotope Beams (FRIB) are discussed. This kind of device would enable the production of long-lived isomeric beams with unprecedented cleanliness, free of the background of adjacent nuclides and, in most cases, other long-lived states of the same nucleus. This capability would permit more selective measurements of isomer properties and isomer-induced reactions. Development of an MR-ToF for FRIB is particularly motivated by the possibility of studying isomer depletion through the mechanism of nuclear excitation by electron capture, which is an intended focus of future FRIB proposals by members of this collaboration.

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1. Introduction

Nuclear excited states with half-lives longer than the typical sub-nanosecond range of most states arise for a variety of different reasons related to the quantum properties of the nucleus (Walker and Dracoulis 1999). These longer half-lives are caused by differences in the total angular momentum (or *spin*), projection of the spin on a nuclear symmetry axis, nuclear shape, or seniority* between a given state and the lower-energy states to which it can nominally decay. These *isomers* have been identified throughout the nuclear chart and have half-lives that span scales from nanoseconds to far longer than the age of the universe (Jain et al. 2015). The distinctive properties of isomers make them interesting to study in their own right—for example, providing clues to the underlying structure (configurations) of the nuclear states—but also for their roles in the context of astrophysics (so-called *astromers*) (Misch and Mumpower 2024), nuclear medicine (Jensen 2024), and applications (Hemmingsen et al. 2004; Carroll 2018). Developing the tools to provide beams of isomeric ions with reduced—or, ideally, free of—background comprising other nuclides or even the ground state of the isomeric nucleus of interest would be highly beneficial to a breadth of experimental studies.

One area of recent interest is in the phenomenon known as nuclear excitation by electron capture (NEEC). Goldanskii and Namiot (1976) initially proposed NEEC as the inverse of the well-known process of internal electron conversion (Blatt and Weisskopf 1952; Hamilton 1966). NEEC is described as the nucleus of a highly charged ion being excited to a higher-lying intermediate state after the capture of an external electron into an atomic orbital, provided there is a suitable matching of the atomic and nuclear energy differences. Applying this to a nucleus starting in an isomeric state was suggested years later (Zadernovsky and Carroll 2002) and represented a new mechanism by which a long-lived isomer might be depleted (Carroll and Chiara 2024); that is, part of the population of isomeric states can be shifted to the ground state, effectively bypassing the natural decay of the isomer with a much shorter time scale. This process is illustrated in Figure 1. An incident particle may depopulate the isomer through a fast transition to an intermediate, higher-energy state that subsequently bypasses the isomeric state and proceeds to the ground state through a cascade of one or more transitions. This effect could dramatically impact the effective half-life of the nuclear isomeric state. Such mechanisms are known to have potential implications for stellar nucleosynthesis

* The binding energy for a nucleus includes a pairing term, such that the energy is reduced (i.e., the nucleus is more tightly bound) when each given proton (or neutron) is paired in an orbit with a like nucleon having anti-aligned spin. Different arrangements of the nucleons, or configurations, can be created by breaking nucleon pairs that result in excited states. The *seniority* of a particular configuration refers to the number of unpaired nucleons.

and are of interest for practical applications (Carroll 2018; Carroll and Chiara 2024).

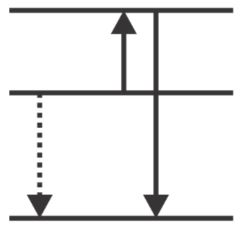

Figure 1. A schematic subset of discrete energy levels in a nuclear system, illustrating isomer depletion. The isomeric state has a slow decay to the ground state (dashed arrow). Faster transitions are indicated by the solid arrows.

Experimental observation of NEEC did not come until more than 40 years after the phenomenon was first proposed, as detailed in Chiara et al. (2018) for the depletion of the 6.85-h isomer in ^{93}Mo. The NEEC probability was deduced to be 1.0(3)% under those experimental conditions. Although concerns that were raised (Guo et al. 2021) regarding the analysis techniques used in Chiara et al. (2018) were successfully rebutted and the results of the measurement were deemed sound by Chiara et al. (2021), this observation has yet to be replicated (Guo et al. 2022) and deserves further attention. The main difficulty of the original measurement was the need to produce the isomeric state and observe the NEEC process in the same physical experimental location in the presence of significant background.

Carrying out an experimental NEEC program at the Facility for Rare Isotope Beams (FRIB) would benefit greatly from the development of high-throughput isomer separation coupled to the ReA ion re-accelerator (Facility for Rare Isotope Beams 2025). It should also be noted that any isomer separation capability would also be beneficial to other basic-science programs, such as studies of the structure of the related nucleus or the investigation of astromers.

2. Current Isomer Separation Capabilities at FRIB

Rare isotopes in multiple beta-decaying ground and isomeric states are routinely produced at FRIB and delivered to experimental stations (see examples in Suchyta et al. 2014 and Chester et al. 2021). The Low-Energy Beam and Ion Trap (LEBIT) (Ringle et al. 2009) group has already delivered isomerically pure beams of ^{70}Cu to an experimental end station using the mass-spectroscopy capabilities of a Penning trap. There are three beta-decaying states in ^{70}Cu: the 6^- ground state, a 3^- isomeric state, and a 1^+ isomeric state at 0, 101, and 247 keV, respectively. All three have been separately isolated and characterized. Although the separated isomeric states were successfully used, the throughput of the overall system was low at approximately 10 pps at most. Furthermore, the separated isotopes cannot be independently counted. These two deficiencies make a similar experimental setup inadequate for NEEC observation, and a new method needs to be developed.

3. Multi-reflection Time-of-Flight Separation

Multi-reflection time-of-flight (MR-ToF) devices have gained remarkable attention at radioactive-ion-beam facilities in the last 20 years for high-precision mass measurements and for mass separation. These devices have mass resolving powers $R = m/\delta m$ exceeding 5×10^5 within a few milliseconds of processing time, which enables resolving approximately half of all known nuclear isomers with half-lives of 10 ms or longer (Dickel and Mollaebrahimi 2024). If a mass resolving power of $R > 3 \times 10^6$ could be achieved, 90% of those isomers could be separated by ToF. The best experimentally achieved mass resolving power that has been reported so far is $R \sim 1.2 \times 10^6$ for $^{36}Ar^+$ within a storage time of 12 ms (Rosenbusch et al. 2023). Using an MR-ToF device at FRIB to separate an isomeric state from its ground state and deliver the clean isomeric beam to an experimental station offers an improved ability to study the NEEC process, enabling high-throughput isomer separation for the experimental program. Examples of some of the nuclides containing isomers that are of interest for exploring NEEC are provided in Table 1.

Table 1. Subset of isomeric states of interest for NEEC studies via MR-ToF.

Nucleus	Isomeric state			Intermediate state		Ground state	
	$t_{1/2}$	E_m (keV)	J^π	E_i (keV)	J^π	$t_{1/2}$	J^π
^{93}Mo	6.85 h	2425.0	$21/2^+$	2439.8	$17/2^+$	4000 y	$5/2^+$
^{84}Rb	20.3 min	463.6	6^-	466.6	5^-	32.8 d	2^-
^{109}In	210 ms	2101.9	$19/2^+$	2102.2	$17/2^+$	4.2 h	$9/2^+$

Note: Data are from the National Nuclear Data Center (n.d.).

4. Experimental NEEC Program

FRIB will produce a nucleus through fragmentation reactions* in both the ground and isomeric states. The nuclide of interest will be separated from other fragmentation products using the Advanced Rare Isotope Separator (ARIS) (Hausmann et al. 2013; Portillo et al. 2023), then slowed down to thermal energies in a linear gas cell filled with approximately 100 mbar of helium gas (Lund et al. 2020; Sumithrarachchi et al. 2020). On the other side of the helium gas cell, the

* Fragmentation is a type of nuclear reaction that can occur when a beam of ions is incident upon a target at energies well above that needed to overcome the coulomb repulsion between the positively charged projectile and target nuclei, leading to breakup of a heavy-mass nucleus into several lighter nuclides in highly excited states (Feshbach 1992).

ions will be delivered at thermal energies to an MR-ToF to separate the isomeric and ground states. Only the isomer will be delivered to an experimental station consisting of a silicon stopping detector surrounded by a high-purity germanium (HPGe) detector array. The silicon detector will serve as both the “NEEC target”—that is, the medium that supplies electrons to potentially initiate the NEEC process—and as an ion counting system.

Since the production of the isomeric state is physically separated from its characterization, there will be no complications related to possible contamination of experimental spectra from the presence of the ground state. Furthermore, the observation of any potential NEEC process is performed in a clean environment well separated from any potential background that could come from the production process. Observation of the NEEC process could be observed in an experiment on the order of 3 days, assuming a rate of 100 pps of isomeric states delivered to the end station.

5. Conclusion

Development of an MR-ToF device for precision mass separation of ions at FRIB will open new possibilities for future experimental studies involving isomeric beams. A clear majority of isomers with half-lives of 10 ms or longer could be separated from the same nuclear species in the respective ground state, leading to isomer beams cleanly separated from contaminant ions. This capability offers the opportunity for a new design of future NEEC experimental studies at FRIB.

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List of Symbols, Abbreviations, and Acronyms

$^{36}Ar^{+}$	argon-36
ARIS	Advanced Rare Isotope Separator
^{70}Cu	copper-70
FRIB	Facility for Rare Isotope Beams
HPGe	high-purity germanium
LEBIT	Low-Energy Beam and Ion Trap
^{93}Mo	molybdenum-93
MR-ToF	multi-reflection time-of-flight
NEEC	nuclear excitation by electron capture

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