Probing ^{93m}Mo Isomer Depletion with an Isomer Beam

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The isomer depletion of ^{93m}Mo was recently reported [Chiara *et al.*, Nature (London) **554**, 216 (2018)] as the first direct observation of nuclear excitation by electron capture (NEEC). However, the measured excitation probability of 1.0(3)% is far beyond the theoretical expectation. In order to understand the inconsistency between theory and experiment, we produce the ^{93m}Mo nuclei using the ^{12}C(^{86}Kr, $5n$) reaction at a beam energy of 559 MeV and transport the reaction residues to a detection station far away from the target area employing a secondary beam line. The isomer depletion is expected to occur during the slowdown process of the ions in the stopping material. In such a low γ-ray background environment, the signature of isomer depletion is not observed, and an upper limit of 2×10^{-5} is estimated for the excitation probability. This is consistent with the theoretical expectation. Our findings shed doubt on the previously reported NEEC phenomenon and highlight the necessity and feasibility of further experimental investigations for reexamining the isomer depletion under low γ-ray background.

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In the last decades, extensive discussions on the potential use of a long-lived nuclear isomeric state as a form of clean and renewable energy storage with high energy density are ongoing [1]. Several million electron volts (MeV) can be released by switching an atomic nucleus from an isomeric state to its ground state, while only photons and electrons are emitted. However, there exists a critical challenge to artificially control the energy release process. As a consensus, the rapid release of the isomeric energy is expected to be achieved by "isomer depletion," that is, by exciting the isomer to an adjacent excited state (triggering state) which then decays to the ground state promptly [2].

Isomer depletion can be triggered by several mechanisms, such as Coulomb excitation [3,4], photoabsorption [5,6], inelastic scattering [7,8], nuclear excitation by electron transfer [9], and nuclear excitation by electron capture (NEEC) [10]. Among them NEEC was predicted to be an efficient mechanism [11]. As the inverse process of internal conversion, NEEC is a process in which a nuclear excitation is driven by capturing a free electron into a vacancy of an ion. Its occurrence requires the coexistence of free electrons and highly charged ions, and therefore, NEEC is expected to occur in astrophysical plasmas [12–14]. In terrestrial laboratories, this condition can be fulfilled by various means, such as generating plasma using lasers [14–19] or stripping electrons from energetic ions in certain materials [20–22].

Recently, the first experimental observation of NEEC was reported from a beam-based experiment [23]. In that

experiment, the ^{93m}Mo nuclei with a half-life of 6.85 h were produced using a fusion-evaporation reaction and implanted into a carbon foil 3 mm behind the reaction target. It was proposed that, during the slowdown process in the stopping foil, the ^{93m}Mo isomer was excited to a 5-keV higher-lying triggering state with a half-life of 3.5 ns, which decays via a 268-keV γ transition. Therefore, the NEEC events were stamped by the coincidence of the 268-keV γ ray with the transitions directly feeding the ^{93m}Mo isomeric state. A surprisingly large excitation probability 1.0(3)% was extracted. However, this value could not be reproduced by a subsequent theoretical calculation based on state-of-the-art atomic theory [24]; i.e., the theoretical value was about 9 orders of magnitude smaller than the experimental one. Very recently, a new theoretical study aimed to resolve this disagreement [25], but only slightly shifted the upper limit and the dramatic gap still remains.

It is worth noting that in the previous work [23] the triggering state was populated not only by isomer depletion, but also by the fusion-evaporation reactions. A bunch of γ rays including statistical γ rays and unidentified cascade γ rays were emitted in the process from the entrance states to the triggering state. If they overlapped with those populating the ^{93m}Mo isomer, it might induce false coincidences into the extraction of the isomer depletion events. The possible false coincidences, together with random coincidences, nonlinear components of γ-spectra background, and enhanced overlapping due to the Doppler effect, could have impeded an unambiguous identification of the isomer depletion [26]. A note was presented to clarify the background consideration [27]; nonetheless, it was agreed that another independent experimental approach is of critical importance to verify the long-sought NEEC phenomenon.

In the present work, a secondary beam line was used to deliver the ^{93m}Mo residues to the implantation and detection station, so that the primary reactions and the isomer depletion were well separated, as illustrated in Fig. 1. Meanwhile, the intense primary beam avoided hitting the stopping material directly. Therefore, the observed decay from the triggering state would be unambiguously attributed to the ^{93m}Mo isomer depletion.

The measurement was performed in the Heavy Ion Research Facility in Lanzhou [28]. As shown in Fig. 1, the ^{93m}Mo residues were produced by the ^{12}C(^{86}Kr, $5n$) fusion-evaporation reaction at a beam energy of 559 MeV and delivered to a low γ-background detection station via the radioactive ion beam line in Lanzhou (RIBLL) [29] with a length of 35 m. We used a stack of two carbon foils as the primary target, which were installed at the two sides of the target frame located at the primary target position of RIBLL. Each carbon foil has a thickness of 100 μg/cm^2, and the two foils are ~2 mm apart from each other. The magnetic fields of RIBLL were set to select highly charged

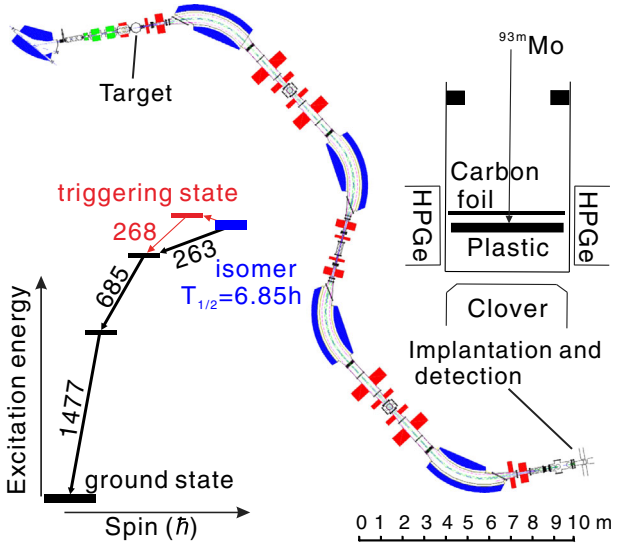

FIG. 1. Experimental setup in the present Letter. The secondary beam line RIBLL is shown with the corresponding distance scale. ^{93m}Mo residues were produced at the primary target position and transported to the end of RIBLL to study the isomer depletion. In the lower left area, the isomer depletion of ^{93}Mo is sketched together with the spontaneous decay of the long-lived isomer. The setup for implantation and detection is shown in the upper right area.

^{93m}Mo^{36+} ions with an energy of 460 MeV. After 1.14 μs transportation, ^{93m}Mo^{36+} ions were slowed down and finally stopped in a plastic detector covered by a 20-μm-thick carbon foil. The purity of ^{93m}Mo was estimated to be ~0.6%. However, few γ rays were emitted at the detection station from the contaminating ions since most of them arrived in the stopping material in their ground states. The γ rays were detected by five high-purity germanium (HPGe) detectors equipped with anti-Compton shields. Among them, four detectors of coaxial type were located perpendicular to the implantation direction, while one segmented clover detector faced it. The signals from the plastic and germanium detectors were recorded using a XIA digital data acquisition system (14 bit, 100 million samples/s, Pixie-16).

The data were collected for 93 h with beam on target, for 18.4 h with beam off, and for 24 h for environmental background measurements. In Fig. 2(a), the total γ-ray spectrum in the beam-off period is shown in purple, together with that of the environmental background in black. It is clear that the main difference between the two spectra is the presence of γ peaks from the spontaneous decay of ^{93m}Mo. As shown in Fig. 2(b), a few γ peaks from other evaporation residues delivered to the detection station are also observed in the total beam-on γ-ray spectrum. Nevertheless, the γ rays from the ^{93m}Mo decays dominate the spectra, and hence it provides an excellent opportunity to investigate the isomer depletion under low γ-ray background.

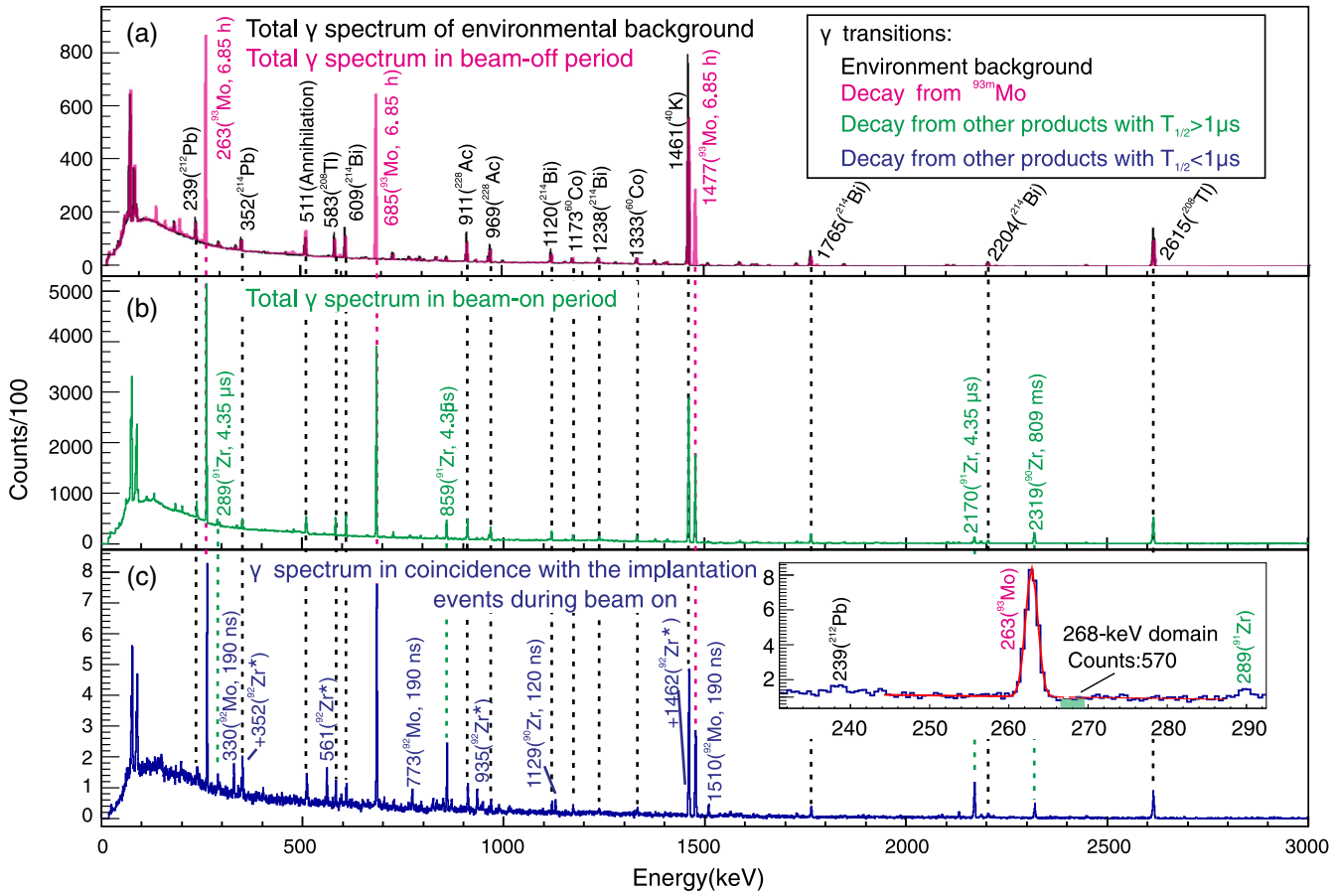

FIG. 2. Spectra acquired by germanium detectors in this measurement. (a) The γ spectra of decay events (in purple) and environmental background (in black). (b) The total γ spectrum recorded by the germanium detectors during the collection of products. (c) The γ spectrum in coincidence with implantation events. The detailed spectrum for the 263- and 268-keV transitions is shown in the inset and fitted by a combination of Gaussian and linear functions. The half-lives of the known isomers are marked for corresponding γ peaks, and the lines assigned to an unidentified isomer of ^{92}Zr are marked with asterisks. Two γ rays with nearly the same energy of 352 keV were identified to originate from ^{214}Pb and ^{92}Zr, respectively. The 352-keV line in (a) and (b) is attributed to ^{214}Pb, and in (c) this line has two components corresponding to ^{214}Pb and ^{92}Zr.

While slowing down in the carbon foil and the plastic detector, the ^{93m}Mo isomer can be excited to the triggering state with a half-life of 3.5 ns. The 268-keV transition depopulating the triggering state is expected to be observed within a few nanoseconds after the implantation in the plastic counter. The γ-ray spectrum in coincidence with the implantation events is shown in Fig. 2(c). The coincidence time window between the signals of the germanium and plastic detectors was set to be 50 ns, and the lower threshold of the plastic detector was 110 MeV. The implantation rate on the plastic detector was about 61 kHz. In the spectrum correlated with the implantation, four coincident 935-, 561-, 1462-, and 352-keV transitions appear. They were identified as the first four transitions of the yrast band of ^{92}Zr [30]. Their intensities are almost identical after correction for their detection efficiencies. These findings strongly suggest that the observed transitions come from the sequential decay of the 8^+ state in ^{92}Zr, which is fed by the decay of an unidentified short-lived isomer. If such an isomer has similar excitation energy to the 8^+ state, its decay to this state would be dominated by internal conversion, a process that is considerably delayed for highly charged ions. Such delay would explain the survival of the isomer during the transportation time and its fast decay after the implantation, when electrons are reabsorbed. Because of the coincidence with the implantation events, the relative heights of the γ peaks from the short-lived products are significantly enhanced. A 268-keV peak depopulating the triggering state would be observed if the isomer depletion occurs.

It is evident that the 268-keV γ ray is not observed in this Letter, as shown in the inset of Fig. 2(c). Actually, the counts around the expected 268-keV peak are sightly lower than the average background. To extract the upper limit of the yield of the 268-keV transition, we checked the counts between 266.5 and 269.5 keV and assumed that the total number (570) was due to both isomer depletion and Compton background. The maximum yield of the 268-keV

transition thus corresponds to the minimum of the background. The estimated number of Compton background counts is about 596, and its 3σ lower limit is 523. Therefore, the upper limit for the area of a possible 268-keV transition is about (570–523)/95% ~50, where the 95% is the confidence interval within the central 3-keV range for the presumed 268-keV peak with a Gaussian shape. The maximum number of isomer depletion events is

$$\mathrm{Max_{ID}} = \frac{50(1+a_{268})}{\varepsilon_{268} P_E P_T (1-P_{dp})}, \tag{1}$$

where ε_{268} and a_{268} are the detection efficiency and the internal conversion coefficient for the 268-keV transition, P_E is the proportion of selected ^{93m}Mo implantation events observed with a 110-MeV threshold of the plastic detector, P_T is the proportion of selected 268-keV transitions in coincidence with the ^{93m}Mo implantation events within a time window of 50 ns, and P_{dp} is the proportion of dead time or pileup events detected by the plastic detector.

The $T_{1/2} = 6.85$ h ^{93m}Mo isomer decays naturally via the 263-, 685-, and 1477-keV cascade. From Figs. 2(b) and 2(c), we extracted $1.763(1) \times 10^6$ counts for the 263-keV peak during beam on and $2.78(1) \times 10^5$ counts in the following 18.4 h with beam off. Therefore, the total number of the implanted ^{93m}Mo ions is

$$N_{\mathrm{isomer}} = \left[1.763(1)\times 10^6 + \frac{2.78(1)\times 10^5}{1-(1/2)^{\frac{18.4}{6.85}}}\right](1+a_{263})/\varepsilon_{263}, \tag{2}$$

where ε_{263} and a_{263} are the detection efficiency and the internal conversion coefficient for the 263-keV transition, respectively. Using the coefficients listed in Table I, the upper limit of the excitation probability is estimated to be

$$P_{\mathrm{max}} = \frac{\mathrm{Max_{ID}}}{N_{\mathrm{isomer}}} \approx 2 \times 10^{-5}. \tag{3}$$

The excitation probability is at most 0.2% of that determined by Chiara *et al.* [23]. In the proposed NEEC process, an isomer is excited to a nearby state by capturing a free electron into a vacancy of the atom, and the energy released from the capture should match the energy difference between the isomeric and triggering states. Given the fact that both Chiara *et al.* [23] and the authors of this Letter used carbon as the main stopping material, the energy distributions are determined by the charge state and recoil energy of the ion of interest. Limited by the primary beam energy in the present Letter, the recoil energy of ^{93m}Mo before injecting into the carbon foil is 460 MeV, which is lower than that in the previous work [23] (a distribution with a center of 674 MeV estimated using LISE++ [32]). During the slowing down process in the stopping material, the charge-state distribution of the ^{93m}Mo ions evolves toward lower mean values, which are ~32.7 at 674 MeV and ~31.5 at 460 MeV [24]. The profile of kinetic energy and its associated charge states set up the NEEC resonant points and consequently determine the excitation probability, which can be calculated by the theoretical approaches as in Refs. [24,25].

In Ref. [24], 648 NEEC channels for charge states from Mo^{14+} to Mo^{42+} and recombination into all possible atomic orbitals of the L, M, N, and O shells have been considered. Taking into account the momentum distribution of the target electrons, the NEEC probability within the impulse approximation [25] is given by

$$P = n_c \sum_{q,\alpha} \int \frac{f_q \sigma_q^\alpha}{-dE/dx} dE, \tag{4}$$

where n_c is the atom density of the carbon target, f_q is the ion fraction in charge state q, E is the ion energy, $-dE/dx$ is the stopping power, and σ_q^α is the NEEC cross section into channel α for an initial ion with charge state q. The cross section σ_q^α can be connected to the NEEC resonance strength S_q^α [25] by

$$\sigma_q^\alpha = \sum_i \sqrt{M_p/(2E)}\, J_i(Q) S_q^\alpha, \tag{5}$$

where M_p is the mass of the ^{93m}Mo ion, $J_i(Q)$ is the Compton profile [33] of the carbon target electrons in orbital i, and Q is the momentum component of the target electron along the direction of the incident projectile. In the present Letter, we employ the same NEEC resonance strengths S_q^α for the considered 648 NEEC channels as in Ref. [24], which are based on *ab initio* NEEC cross section calculations. In order to calculate the charge-state distribution, we adopt the multiparameter least-square fit by Schiwietz and Grande [34]. Furthermore, we obtain the stopping power by the unitary-convolution-approximation stopping-power model implemented in the convolution approximation for swift particles (CasP) code by

TABLE I. Coefficients used to extract the upper limit of NEEC probability.

Coefficients	Value
$\varepsilon_{263}/\varepsilon_{268}$	1.01(1)
a_{268}	0.0356[a]
a_{263}	0.698[a]
P_E	0.91(2)
P_T	0.96(1)
P_{dp}	0.24

[a]Adopted from National Nuclear Data Center [31].

Schiwietz and Grande [35–37]. The NEEC probability is calculated to be 2.3×10^{-12} at the recoil energy of 460 MeV before entering the carbon foil and 2.8×10^{-11} at that of 840 MeV. For the latter, we assume a recoiling ^{93m}Mo ion energy of 840 MeV for the experiment in Ref. [23] to provide an upper limit. In addition to NEEC, Coulomb excitation and inelastic scattering could also lead to the isomer depletion, with probabilities on the order of 10^{-6} [23].

The theoretical results are in agreement with the nonobservation of isomer depletion in the present Letter, which sets an upper limit of 2×10^{-5} for the NEEC probability, but disagree with the previous experimental data [23] by about 9 orders of magnitude. Assuming that the observed probabilities of the isomer depletion are attributed to NEEC, the discrepancy between the two experimental results would be too large to be explained due to the differences in the experimental conditions. The theoretical calculations predict that the NEEC probability ratio at the two recoil energies is about 8%. However, the observed difference is considerably larger, suggesting that something else may play a major role. We emphasize that the present Letter measures the excitation probability under a low γ-ray background, and hence it sheds doubt on the previously reported NEEC phenomenon observed with the heavy γ-ray background. In order to confirm the ^{93m}Mo isomer depletion, it is crucial to eliminate the contaminations induced by the heavy γ-ray background at the primary reactions *in situ*.

The present Letter demonstrates the feasibility of studying isomer depletion with high sensitivity by employing isomer beams. The sensitivity is expected to be further improved by combining intense isomer beams with special stopping targets such as crystals [38] and the liquid beam electron target [39].

In summary, the isomer depletion of ^{93m}Mo has been reinvestigated under low γ-ray background employing a secondary beam line. The isomer depletion has not been observed, and an upper limit of 2×10^{-5} has been extracted for the NEEC probability, which is consistent with the theoretical expectations. The results from the present Letter highlight the necessity and feasibility of further experiments for reinvestigating the isomer depletion under low γ-ray background with higher recoil energies.

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