

DESIGNING ATOMIC RESONANCE CONDITIONS FOR ^{93m}Mo ISOMER DEPLETION VIA NUCLEAR EXCITATION BY ELECTRON CAPTURE IN A BEAM-BASED SCENARIO FOR SELECTED TARGETS

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We present theoretical research on the optimal atomic resonance conditions for long-lived ($T_{1/2} \sim 6.85$ h) ^{93m}Mo isomer depletion via nuclear excitation by electron capture (NEEC) in a beam-based scenario. Analysis of potentially possible NEEC-resonance kinetic energy and predictions of the mean equilibrium charge state for the ^{93m}Mo recoil ion as a function of its kinetic energy in the He, C, Mg, and Al stopping media have been performed. We have found for the solid targets the high probability of observing the NEEC process for electron capture into M subshells for $q = +33$ charge state, and also significant probability for electron capture into N subshells for $q = +34$. Moreover, we have concluded that, for the studied solid targets, the occurrence of NEEC seems to be more probable than for a He gas target. The results of our study should be an important step toward the controlled release of energy stored in the nuclear isomer of some elements and may also be useful in the development of a new class of nuclear energy sources.

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1. Introduction

The properties of nucleon states in the nucleus and electron states in the atom are determined by quantum mechanics. Electromagnetic transitions between electron states and between nuclear states are governed by selection rules. As is well-known, electric-dipole transitions dominate in atoms, whereas in the case of nuclei, transitions with higher multiplicities also play a very important role. For many nuclear states, depopulating transitions with large multiplicities are highly suppressed, *i.e.*, they are characterized by the extremely low probability of occurrence. Therefore, many nuclides possess metastable states called nuclear isomers which are characterized by high spins and long, sometimes even extremely long, lifetimes in the range from nanoseconds to many years (*e.g.*, the lifetime of the ^{180m}Ta isomer exceeds 10^{16} years, which is much longer than the age of the Universe) [1, 2]. Some of these long-lived nuclear isomers can store a huge amount of energy, up to a few MeV per nucleus [3].

In 1976, Goldanskii and Namiot [4] for the first time drew attention to an important phenomenon from the point of view of fundamental science, *viz.*, the inverse of the well-known internal-conversion process. In 1989, Cue *et al.* [5] coined the term nuclear excitation by electron capture (NEEC) for this process in which a free electron is captured into a bound state of an ion with the simultaneous excitation of the nucleus. They proposed a list of isotopes which have metastable excited states suitable for the study of NEEC processes. In 1993, Yuan and Kimball predicted theoretical cross sections for the NEEC processes for highly stripped Ho, Yb, Re, and Pt ions [6], and in 2002 [7] it was proposed that NEEC could be used to induce a depletion, with energy release, from a long-lived isomer. An experiment performed at GANIL in 2004 [8], with fully stripped ^{57}Fe ions, did not observe NEEC, according to Ref. [9]. Another experimental attempt to use an electron beam incident upon ^{242}Am ions confined within an ion trap [10] was also reported as unsuccessful. In 2007, Pálffy *et al.* [9] presented the resonance strengths for NEEC processes for the isomers of a few nuclides. Moreover, in 2012, Karamian and Carroll published an article [11] in which they suggested conducting an experiment concerning the depletion of the ^{93m}Mo isomer through the NEEC process using a helium gas target in a beam-based scenario.

A fundamental step in designing the nuclear and atomic conditions for the first observation of the NEEC process for the ^{93m}Mo isomer (see Fig. 1 [12, 13]) in beam-based scenarios has been recently done [14]. This provided improved quantitative predictions to apply to the scheme of Ref. [11], where, in inverse kinematics, a fusion–evaporation reaction between a beam of heavy- Z projectiles and light- Z target nuclei would be used to produce the ^{93m}Mo isomer with an initial ion recoil kinetic energy exceeding the res-

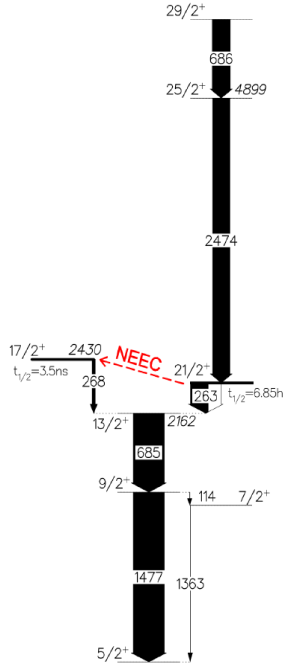


Fig. 1. (Color online) Partial level scheme of ^{93}Mo , adapted from Refs. [12] and [13]. The dashed gray/red arrow illustrates the transition to higher excitation energy via the NEEC process. The separation between the $21/2^+$ isomer and $17/2^+$ intermediate state is exaggerated for clarity.

onance energies for the NEEC process. These high-energy ^{93m}Mo recoils will subsequently slow down from high speed in the stopping medium. During the slowing down process, highly ionized isomeric atoms can reach the appropriate energy and atomic state to match, at some point along their paths, the NEEC resonance conditions (see Fig. 2). It should be underlined that the results of the analysis in [14] and a unique experimental configuration (for which the crucial elements were: using the Digital Gammasphere

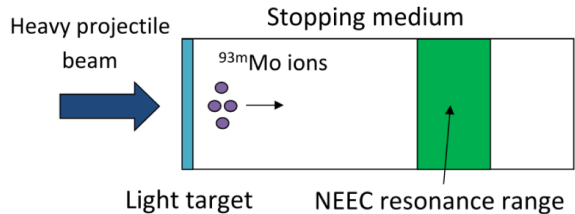


Fig. 2. NEEC process experimental observation scheme [15].

spectrometer at Argonne National Laboratory (ANL), optimal construction of the target, and the appropriate beam energy of ^{90}Zr ions) have made possible the first observation of the NEEC process for any isomer, using ^{93m}Mo , *i.e.*, the identification of a new physical phenomenon [15]. Evidence of ^{93m}Mo isomer depletion via the NEEC process was the registration of the distinctive γ -ray sequence 2474 keV–268 keV–685 keV–1477 keV (see Fig. 1).

2. Results

In previous papers, the most promising nuclear reactions $^{91}\text{Zr} + ^4\text{He}$ [14] and $^{90}\text{Zr} + ^7\text{Li}$ [15] were analyzed, where production of the ^{93}Mo isotope is significant, in particular the essential population of the desired ^{93m}Mo isomer. The cross sections for these reactions have been calculated using the Monte Carlo fusion–evaporation code PACE4 [16, 17] and their relative values verified experimentally [15]. In these reactions, a heavy projectile (^{91}Zr [14], ^{90}Zr [15]) strikes a light target (^4He [14], ^7Li [15]) and the produced ^{93m}Mo isomer has high recoil energy (7.7 MeV/nucleon [14]) because it is taking from the projectile most of the kinetic energy. Therefore, in this paper, we have taken this ^{93m}Mo isomer recoil energy (which must exceed the energy needed for the NEEC resonances) as a starting point to determine the dependence of the mean equilibrium charge state (q_{mean}) for ^{93}Mo ions on their kinetic energy during their passage through different gas and solid targets. We have used formulas proposed by Schiwietz and Grande [18], which reproduce the experimental data with the accuracy not worse than $\Delta q_{\text{mean}}/Z_p \sim 2.5\%$ [18]. In addition, the values of the discrete energy released by electron capture into different atomic subshells of the N and M shells as a function of the charge state q for ^{93m}Mo ions have been obtained using the relativistic multiconfiguration Dirac–Fock (MCDHF) method [19–22]. This method enables precise calculations, taking into account the Breit interaction and two quantum electrodynamics corrections (self energy and vacuum polarization).

Figures 3–6 show the q_{mean} of the ^{93m}Mo recoil ions as a function of its kinetic energy in gas (He) and solid (C, Mg, and Al) targets, in the range of ion energies from 4.4 to 7.7 MeV/nucleon. The absolute uncertainties Δq_{mean} are 0.48 for He gas and 0.54 for C, Mg, and Al solid targets [18], and they are shown in Figs. 3–6 by the dashed green lines close to the solid black ones. In addition, we have used the widths of the q distribution for a He gas target equal to ~ 1.7 [23] and for solid targets equal to ~ 1.5 [24, 25]. They are shown in Figs. 3–6 as the short-dashed red lines described as: $q_{\text{mean}} - \text{FWHM}/2$ and $q_{\text{mean}} + \text{FWHM}/2$. The potential NEEC-resonance kinetic energy of the free electron (in the ^{93m}Mo ion reference frame) has been obtained as the difference between the energy required for NEEC to occur (4.85 keV) and the energy released by electron capture into

the N or M subshells. To obtain the corresponding NEEC-resonance kinetic energy of the ^{93m}Mo ion (presented in Figs. 3–6), we have multiplied the above data by the nucleon-to-electron mass ratio. The vertical bars in Figs. 3–6 show the potential positions of the ^{93m}Mo ion NEEC-resonance kinetic energies for electron capture to the N and M subshells for $q = +32$ to $q = +38$, ending with a filled circle when the bars tops are in the q range (*i.e.*, between $q_{\text{mean}} - \text{FWHM}/2$ and $q_{\text{mean}} + \text{FWHM}/2$) and open circles when they are not in the q range. Each group of five bars illustrates, from left to right, electron capture into the $d_{5/2}$, $d_{3/2}$, $p_{3/2}$, $p_{1/2}$, and $s_{1/2}$ subshells.

In the case of a He ($Z = 2$) gas target (see Fig. 3), the potential positions of the ^{93m}Mo -ion NEEC-resonance kinetic energies may mainly be achieved for electron capture into the N subshells. It may be seen when the bar tops of the $4d_{5/2}$, $4d_{3/2}$, $4p_{3/2}$, $4p_{1/2}$, and $4s_{1/2}$ subshells (for $q = +33$) reach the line describing the ^{93m}Mo -ion q_{mean} , with the energy for the $4d_{5/2}$ subshell falling closest to the line. Moreover, after electron capture to the M subshells, the NEEC process observation seems to be also probable.

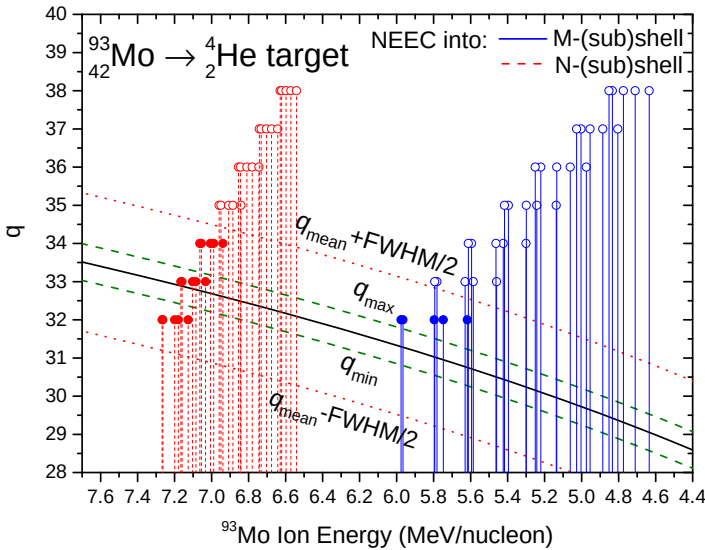


Fig. 3. (Color online) Dependence of q_{mean} (black solid line) on ^{93m}Mo -ion kinetic energy for a He gas target. The vertical bars show the potential positions of the ^{93m}Mo ion NEEC-resonance kinetic energies, that may appear from $q = +32$ to $q = +38$ if the bar tops are in the q range.

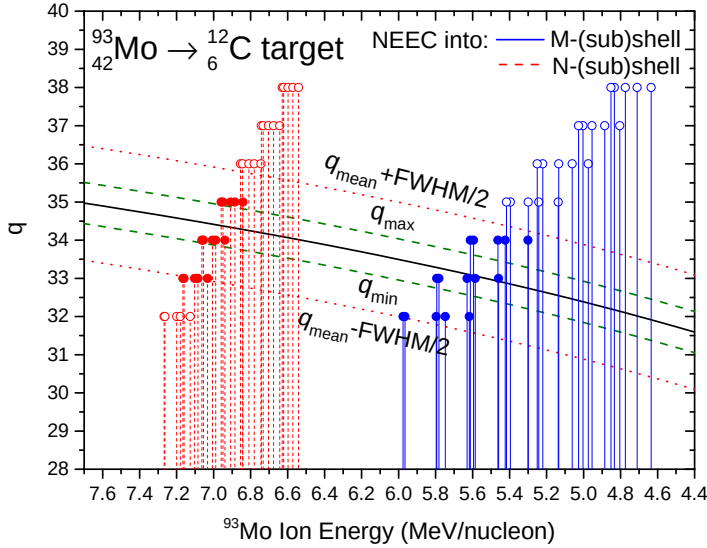


Fig. 4. (Color online) Similar to Fig. 3, but for a solid C target.

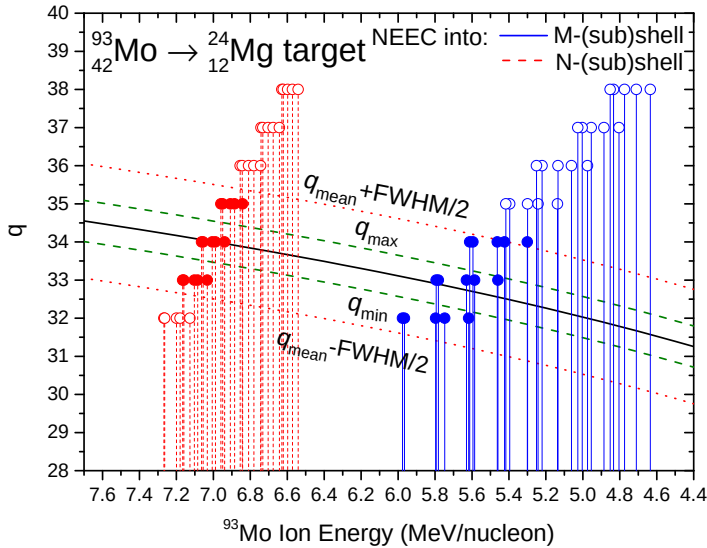


Fig. 5. (Color online) Similar to Fig. 3, but for a solid Mg target.

For a solid C ($Z = 6$) target (see Fig. 4), the potential positions of the ^{93m}Mo -ion NEEC-resonance kinetic energies may be achieved for electron capture into the M subshells (*i.e.*, $3d_{5/2}$, $3d_{3/2}$, $3p_{3/2}$, $3p_{1/2}$, and $3s_{1/2}$) for $q = +33$ because of the proximity of the line describing the ^{93m}Mo -ion q_{mean}

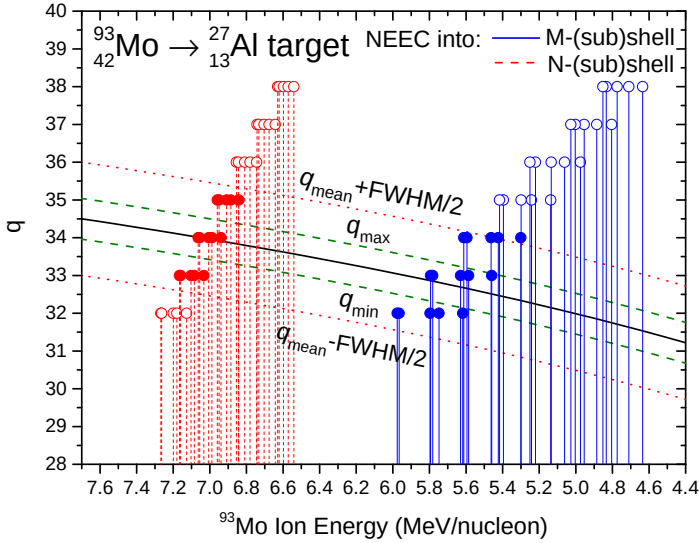


Fig. 6. (Color online) Similar to Fig. 3, but for a solid Al target.

to the bar tops. One can also see that in the case of $q = +33$, the bar tops for electron capture to the $3p_{3/2}$, $3p_{1/2}$, and $3s_{1/2}$ subshells are located slightly closer to the line characterizing the q_{mean} than for the $3d_{5/2}$ and $3d_{3/2}$ subshells. For electron capture into the M subshells for $q = +32$ and $q = +34$, the probability for NEEC is lower than for $q = +33$, and from $q = +36$ to $q = +38$ is rather unlikely. However, for electron capture into the N subshells, the line representing the q_{mean} is between the bar tops for $q = +34$ and $q = +35$, which results in a smaller chance for NEEC to occur than for $q = +33$ for electron capture into the M subshells. It is worth underlining that, for a C target, the q_{mean} is much higher (by about 1.6) than for a He target.

Figures 5–6 present, for solid Mg ($Z = 12$) and Al ($Z = 13$) targets, analyses that are very similar to those in Figs. 3–4. We have found that, for Mg and Al targets, the q_{mean} is lower than for a C target by about 0.4 and about 0.5, respectively. Therefore, as can be seen for the solid targets studied, with an increase of the atomic number Z , the values of q_{mean} slightly decrease. In the case of Mg and Al targets (see Figs. 5–6), the potential positions of the ^{93m}Mo -ion NEEC-resonance kinetic energies may be reached for electron capture into all of the N subshells for $q = +34$. Moreover, for $q = +33$, the NEEC probability is also high for electron capture into the M subshells, but especially for electron capture into the $3d_{5/2}$ and $3d_{3/2}$ subshells. For $q = +32$ and $q = +34$, the probability of observing the NEEC process for electron capture into the M subshells seems to be lower than for $q = +33$, but is still rather significant.

3. Summary and conclusions

The theoretical research performed here has shown the important role of complex atomic processes in establishing the optimal resonance conditions in planned experiments on ^{93m}Mo isomer depletion via the NEEC process in a beam-based scenario. The dependence of the mean equilibrium charge state for ^{93}Mo ions on their kinetic energy during passage through He gas and different solid targets (C, Mg, and Al) has been obtained using formulas proposed by Schiwietz and Grande, in a wide range of beam energies of Mo ions. In our study, we have determined the values of energy released by electron capture into different M and N subshells (established using the MCDF method) depending on the charge states, in order to evaluate possible NEEC-resonance kinetic energies of the ^{93m}Mo ion.

Analysis of possible NEEC-resonance kinetic energies and predictions of the q_{mean} for the ^{93m}Mo recoil ion (as a function of its kinetic energy in the selected stopping targets) has shown that, for all analyzed media, the favorable NEEC resonance conditions for electron capture into N and/or M subshells can be achieved. However, in view of internal-conversion-coefficient values for given subshells, one can suppose that observation of the NEEC process for electron capture into the M subshells should be more prevalent than for the N subshells. Therefore, on the basis of our analysis, one can conclude that the NEEC process seems to be more probable for the studied solid targets than for a He gas target. For the solid targets, we have found the high probability of the NEEC process for electron capture into M subshells for $q = +33$, and also significant probability for electron capture into N subshells for $q = +34$. However, it is very difficult to decide which solid target is the most promising for observing the NEEC process because the differences between them are subtle. Additional and more comprehensive analysis is planned for future investigation.

The results of this study concerning the atomic conditions for the NEEC process should be an important step toward applied research, which could allow, in the future, for the controlled release of energy accumulated in the nuclear isomer of some elements. This research may also be useful in the development of a new class nuclear energy sources.

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