^{93m}Mo ISOMER DEPLETION VIA NUCLEAR EXCITATION BY ELECTRON CAPTURE: ENERGY RELEASED FOR DIFFERENT ATOMIC SUBSHELLS AS BENCHMARKS FOR A BEAM-BASED SCENARIO APPROACH*

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For a better understanding of the nature of isomer depletion via the nuclear excitation by electron capture (NEEC) process, we have presented here, for the long-lived ($T_{1/2} \sim 6.85$ h) ^{93m}Mo isomer, the results of high-precision multiconfiguration Dirac–Fock calculations concerning the dependence of the energy released by electron capture into different atomic subshells of the N, M, and L shells on the specific charge state. These energy values can be understood as benchmarks, *i.e.*, reference parameters, which are crucial for implementations of the proposed beam-based scenario approach for different stopping targets. The authors believe that this detailed analysis of the atomic conditions for the NEEC process should be a first step to applied research, which aims to allow the controlled release of energy stored in the nuclear isomer of selected elements. The presented studies may contribute to the development of the concept of new, unconventional, and ultra-energy-dense nuclear power sources.

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1. Introduction

Long-lived nuclear isomers, *i.e.*, metastable excited states of atomic nuclei, have been proposed for extremely high-density energy storage materials and considerable research has been conducted to investigate a means to induce a release of energy on demand (see the review in [1] and references therein). One of the possible mechanisms is isomer depletion via the nuclear excitation by electron capture (NEEC) process [2–5]. In the NEEC process, which is the inverse of the well-known internal-conversion process, a free electron is captured into an unoccupied atomic subshell of an ion with the simultaneous excitation of the nucleus. The NEEC process can only occur when the kinetic energy of the free electron plus the magnitude of electron energy released once it is captured matches the energy difference between the two nuclear states. The NEEC process was predicted in 1976 [2] and has been the subject of many detailed theoretical researches (see, *e.g.*, Refs. [3–7], where isomer depletion via NEEC was proposed in Ref. [3]). For many decades, there have been unsuccessful attempts to observe the NEEC process, *e.g.*, with ^{242}Am in an electron beam ion trap [8] and with bare ^{57}Fe ions passing through an Si crystal [9].

It is worth noting that recently we have performed [10] an extensive study on the optimal conditions for the first experimental evidence of the NEEC process for the ^{93m}Mo isomer using the beam-based approach proposed in Ref. [11]. In this approach, a long-lived 6.85 h ^{93m}Mo isomer with spin parity $21/2^+$ (see Fig. 1) could be produced through a fusion–evaporation reaction in inverse kinematics, where a heavy projectile strikes a light target [10, 11]. The NEEC process allows an excitation from the ^{93m}Mo isomer (at 2425 keV) to a $17/2^+$ intermediate state (depletion level) with a half-life of 3.5 ns that lies only 4.85 keV higher in energy (see Fig. 1). The depletion level subsequently decays and releases a significant amount of stored energy (2429.8 keV). The $17/2^+$ shorter-lived isomer decays to a $13/2^+$ state via a 267.9 keV transition that gives a possibility for unique identification of NEEC because that γ ray does not exist in the natural decay of the ^{93m}Mo isomer [10].

It needs to be highlighted that very recently our results [10] have supported the first observation [12] of isomer depletion via the NEEC process for the ^{93m}Mo isomer, *i.e.*, the identification of a new physical phenomenon, using the Digital Gammasphere [13] γ-ray spectrometer installed at the linear accelerator facility (ATLAS) at Argonne National Laboratory. The ^{93m}Mo isomers were produced through the ^{90}Zr + ^{7}Li fusion–evaporation reaction according to the proposed beam-based scenario [10, 11]. Choosing fast heavy projectiles (840 MeV ^{90}Zr beam) and light ^{7}Li target results in recoiling ^{93m}Mo reaction products that move at high energies in approximately the same direction as the beam [12]. Traversal of those recoiling ^{93m}Mo ions

through a stopping medium caused them to become highly ionized. The stopping medium (^{12}C target) would also provide atomic electrons to the recoiling ^{93m}Mo ions at a high velocity, as seen in the reference frame of the ions [10–12]. Finally, the resonance condition for NEEC could be achieved during the slowing down of the recoiling ^{93m}Mo ions in the ^{12}C stopping medium. In our experimental setup [12], the confirmation for the evidence of the ^{93m}Mo isomer depletion via the NEEC process was the unique registration, in coincidence, of the characteristic γ-rays sequence 267.9 keV–684.7 keV–1477.2 keV (see Fig. 1) along with a 2475 keV transition feeding the long-lived isomer (not shown).

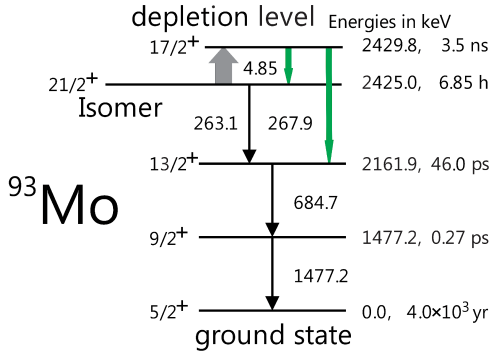

Fig. 1. (Colour on-line) Partial level scheme for the ^{93}Mo isotope. The thick grey arrow is the transition excited by NEEC and the thin black arrows show the natural decay cascade from the ^{93m}Mo isomer. The thick dark grey/green arrow transitions are only emitted following NEEC [10].

On the basis of the analysis of some preliminary results of our study [10], it has been concluded that the electron capture into M and N subshells are the most probable for observation of the NEEC process within the advanced beam-based scenario approach for the long-lived ^{93m}Mo isomer. However, our previous studies [10, 12] indicate that for comprehensive knowledge and better understanding of the nature of the NEEC process, it is necessary to carry out for ^{93m}Mo ions further detailed and extensive analysis of the atomic parameters, especially the energy released by electron capture into different atomic subshells.

2. Theoretical background

Extensive high-precision calculations have been carried out using the relativistic multiconfiguration Dirac–Fock (MCDF) method. The MCDF method was mainly developed by Grant and co-workers [14, 15] and was de-

scribed in many articles [14–19]. The Hamiltonian for an N-electron system can be expressed as

$$H = \sum_{i=1}^{N} h_{\mathrm{D}}(i) + \sum_{j>i=1}^{N} C_{ij}\,, \tag{1}$$

where $h_{\mathrm{D}}(i)$ is the Dirac operator and the terms C_{ij} account for electron–electron interactions that are a sum of the Coulomb operator and the transverse Breit operator. An atomic state function (ASF) with the total angular momentum J and parity p is assumed in the form of

$$\Psi_s(J^p) = \sum_m c_m(s)\Phi(\gamma_m J^p)\,, \tag{2}$$

where $\Phi(\gamma_m J^p)$ are configuration state functions (CSF), $c_m(s)$ are mixing coefficients for state s, and γ_m gives information uniquely defining a certain CSF. In addition, the calculations include QED corrections: self-energy and vacuum polarization. Our study has been performed using the General-purpose Relativistic Atomic Structure Package (GRASP) [15] and GRASP2K [18] packages.

3. Results

We have presented here, for the ^{93m}Mo isomer, the results of high-precision MCDF calculations concerning the dependence of a significant parameter, *i.e.*, the energy released by electron capture into different atomic subshells of the N, M, and L shells, on the specific charge state (q) and assumed electronic configuration. These energy values can be understood as benchmarks, *i.e.*, reference parameters, which are crucial for implementations of the proposed beam-based scenario approach for different stopping targets. Here, it is worth noting that the energy released by the electron capture process to the system with charge state q can be treated as the binding energy of the caught electron in the $(q-1)$ ionized system.

Table I collects the values of the energy released after electron capture into given M and N atomic subshells of the ^{93m}Mo isomer for specific charge states. The ^{93m}Mo ion is slowing down (from its recoil energy after the nuclear reaction) while traversing the stopping medium, and the NEEC process can occur when the sum of the kinetic energy of the free electron (in the reference frame of the ^{93m}Mo ion) and the magnitude of electron energy released once it is captured into a particular subshell is equal to the energy difference between the two nuclear states, *i.e.* 4.85 keV (see Fig. 1) [10, 12]. So the conditions for the NEEC process can be obtained for electron capture into particular atomic subshells and for appropriate q of ^{93m}Mo. Our

previous analyses [10, 12] have shown that, in the case of electron capture into the M and N atomic subshells, the lowest charge state for which the NEEC process occurs is $q = +32$, *i.e.*, when the ^{93m}Mo isomer kinetic energy is still high enough [10]. Therefore, charge states of a ^{93m}Mo isomer from $q = +32$ [10] to $q = +42$ (*i.e.*, bare ion) have been considered in Table I. The dependence of energy released by electron capture into M and N subshells of the ^{93m}Mo isomer on q is also presented in Fig. 2.

TABLE I

The energy released (in eV) by electron capture into different M and N atomic subshells of the ^{93m}Mo isomer for specific charge states q.

Charge state, q	Initial config.	$3s$ $4s$	$3p_{1/2}$ $4p_{1/2}$	$3p_{3/2}$ $4p_{3/2}$	$3d_{3/2}$ $4d_{3/2}$	$3d_{5/2}$ $4d_{5/2}$
+32	$1s^2 2s^2 2p^6$	1790	1720	1693	1601	1595
		968	940	929	894	892
+33	$1s^2 2s^2 2p^5$	1877	1808	1784	1701	1694
		1020	992	982	951	947
+34	$1s^2 2s^2 2p^4$	1963	1896	1875	1803	1794
		1071	1044	1034	1008	1003
+35	$1s^2 2s^2 2p^3$	2052	1995	1964	1911	1900
		1124	1100	1088	1065	1060
+36	$1s^2 2s^2 2p^2$	2141	2094	2054	2007	1990
		1177	1157	1141	1123	1117
+37	$1s^2 2s^2 2p^1$	2233	2189	2152	2125	2112
		1232	1214	1199	1183	1178
+38	$1s^2 2s^2$	2326	2285	2250	2218	2208
		1288	1271	1257	1244	1240
+39	$1s^2 2s^1$	2416	2383	2347	2338	2327
		1348	1330	1315	1307	1302
+40	$1s^2$	2506	2482	2445	2437	2425
		1409	1389	1373	1370	1365
+41	$1s^1$	2619	2607	2566	2575	2562
		1469	1460	1442	1441	1435
+42	—	2732	2732	2688	2688	2674
		1530	1530	1511	1511	1505

As can be seen from Table I and Fig. 2, with the increase of charge state, the energy released by electron capture into given subshells increases significantly, but grows faster for M subshells than for N subshells. It is worth underlining that the energies released by electron capture for M subshells are much higher than for N subshells. This is due to the fact that the electrons in the M subshells are more tightly bound with the nucleus than electrons in the N subshells.

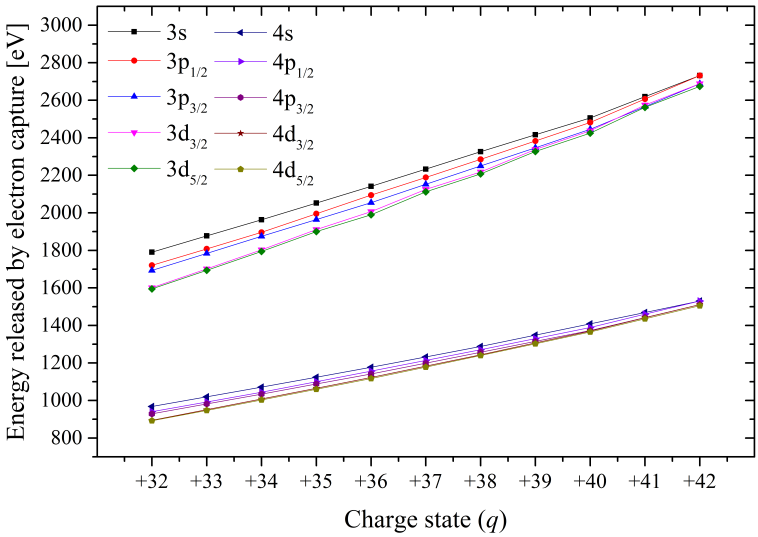

Fig. 2. Dependence of energy released by electron capture into the M and N subshells of the ^{93}Mo isomer on the specific charge state q.

TABLE II

The energy released by electron capture into different L subshells of a ^{93m}Mo isomer for specific charge states q. The excited initial configurations are those of the final configurations with a single electron absent from the $2s$, $2p_{1/2}$, or $2p_{3/2}$ subshell.

Charge state q	Final configuration	Energy released for L subshells [eV]		
		$2s$	$2p_{1/2}$	$2p_{3/2}$
+21	$1s^2 2s^2 2p^6 3s^2 3p^6 3d^4$	3614	3378	3262
+22	$1s^2 2s^2 2p^6 3s^2 3p^6 3d^3$	3693	3456	3343
+23	$1s^2 2s^2 2p^6 3s^2 3p^6 3d^2$	3774	3538	3425
+24	$1s^2 2s^2 2p^6 3s^2 3p^6 3d^1$	3855	3619	3506
+25	$1s^2 2s^2 2p^6 3s^2 3p^6$	3934	3695	3588
+26	$1s^2 2s^2 2p^6 3s^2 3p^5$	4016	3782	3670
+27	$1s^2 2s^2 2p^6 3s^2 3p^4$	4096	3863	3751
+28	$1s^2 2s^2 2p^6 3s^2 3p^3$	4177	3945	3833
+29	$1s^2 2s^2 2p^6 3s^2 3p^2$	4253	4025	3909
+30	$1s^2 2s^2 2p^6 3s^2 3p^1$	4338	4108	3997
+31	$1s^2 2s^2 2p^6 3s^2$	4423	4193	4084

Moreover, for the ^{93m}Mo isomer, it is also possible that the electron can be captured into different L subshells, in the case of excited configurations having an incompletely filled L shell. Table II presents the values of energy released after electron capture into given L atomic subshells of a ^{93m}Mo isomer for charge states from $q = +21$ to $q = +31$. For the considered q, the excited initial configurations are obtained from the final configuration by removing an electron from the $2s$, $2p_{1/2}$, or $2p_{3/2}$ subshell. As can be seen from Table II, the values of energies released by electron capture into the $2s$, $2p_{1/2}$, or $2p_{3/2}$ subshell are very high, much higher than for M and N subshells. It should be noted that the kinetic energy of the free electron for NEEC to occur (in the reference frame of the ^{93m}Mo ion), equal to the difference between the nuclear energy required for the occurrence of the NEEC process (4850 eV) and the energy released by electron capture into L subshells (see Table II), is very small. This requires that the kinetic energy of the ^{93m}Mo ion in the laboratory frame needed for NEEC resonance would also be very small, giving relatively low probability for holes existing in the L subshells, especially for higher charge state. Therefore, the possibility for the NEEC process occurring by electron capture into the L shell seems to be much lower than in the cases of the M and N shells.

On the other hand, for higher q, the existence of excited configurations with holes in the L shell would significantly broaden the resonance window widths [10] for the occurrence of the NEEC process for electron capture into the M and N shells. In particular, for the excited configurations with holes in the $2s$ subshell, the NEEC resonance window width for electron capture into the $3p$ subshell could reach the value of 0.03 eV, compared to values of the order of 0.00001 eV found without considering this effect in Ref. [10].

4. Summary and conclusions

For a better understanding of the nature of the NEEC process, we have presented here in the framework of the proposed beam-based scenario approach, the ^{93m}Mo isomer benchmark values for the energies released by electron capture into different atomic subshells of the N, M, and L shells for specific charge state q and assumed electronic configurations. These energy values have been obtained using the high-precision MCDF method, implemented with the GRASP and GRASP2K packages. Analysis of these benchmark values has shown that, with the increase of charge state, the energy released by electron capture into given subshells grows significantly, but faster for M subshells than for N subshells. In addition, the energies released by electron capture for M subshells are much higher than for N subshells. Moreover, for the studied q, the energies released by electron capture into the M and N subshells give the best opportunity for observation of the NEEC process for the ^{93m}Mo isomer [10].

It has also been found that the values of energies released by electron capture into the L shell are very high, much higher than for M and N subshells. So, for NEEC resonances, the kinetic energy of the free electron (in the reference frame of the ^{93m}Mo ion) and also the kinetic energy of the ^{93m}Mo ion in the laboratory frame are very small, providing relatively low probability for existing holes in the L subshell, especially for higher charge states. Thus, the possibility of the NEEC process occurring by electron capture into the L shell seems to be low. However, the existence of excited configurations with holes in the L subshells would significantly broaden the resonance window widths for the NEEC process for electron capture into the M and N subshells.

The authors believe that this improved analysis of the atomic conditions for the NEEC process will have an impact on the development of the theories describing isomer depletion processes. In addition, it can have an effect on a better understanding of the processes occurring in the Universe, and in particular, provide knowledge concerning the survival of the nuclei of different isotopes of the elements in stellar environments [12]. It should also be a first step to applied research, which aims to allow the controlled release of energy stored in the nuclear isomer of selected elements. These studies may contribute to the development of the concept of new, unconventional, and ultra-energy-dense nuclear power sources [1, 10].

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