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# POSSIBLE DEPLETION OF ISOMERS IN PERTURBED ATOMIC ENVIRONMENTS

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Карамян С. А., Кэрролл Дж. Дж. Возможности разрядки изомеров в возмущенном атомном окружении

Сечения кооперативных атомно-ядерных процессов могут быть выведены в благоприятных случаях из экспериментов или прямо, или посредством адаптированных теоретических оценок. Вероятность трансмутации изомера в механизмах с участием электрона должна включать как ядерную, так и атомную составляющую. Ядерные данные обширны и пополняются, в частности, в результате исследования электромагнитных ядерных процессов, таких как эмиссия гамма-квантов, реакции под действием фотонов, кулоновское возбуждение и др. В настоящей работе рассчитан и сравнивается с экспериментом темп возбуждения ядра с помощью механизма NEET. Схематически оценена также вероятность NEEC, и обсуждаются перспективы наблюдения NEEC при использовании различных экспериментальных подходов. Предложен новый метод, основанный на поиске разрядки изомеров при торможении ядер отдачи в газе.

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For atomic-nuclear cooperative processes, the cross sections can be deduced in favorable cases from experiments either directly, or through the linked theoretical estimates. Probability of isomer transmutation in electron-assisted mechanisms must contain both nuclear and atomic components. The nuclear database is abundant and extensive, in particular due to the exploration of electromagnetic nuclear processes, like  $\gamma$  emission, reactions induced by photons, Coulomb excitation and so on. In the present work, the nuclear excitation rate via NEET mechanism is calculated and compared to known experimental results. The NEEC probability is estimated schematically. Perspectives for NEEC detection within different experimental approaches are discussed. A new method is proposed to search for the isomer depletion under stopping of recoiling nuclei in gas.

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### INTRODUCTION

A problem of nuclear isomer transmutation\* is of interest in the modern literature as is expressed in a number of review articles and reports concerned various approaches (see [1-6] and references therein). In addition to the expected advances in physics, the possibilities for potential application of isomers serve for motivation of new experimental studies. As is supposed, the isomeric substance may accumulate nuclear energy for a long time, and then release upon demand due to the effect of external stimuli. Until now, it was concluded that isomer depletion induced by photons is characterized by relatively low probability because of structure hindrances and high multipolarity for  $\gamma$  transitions. The restrictions arise due to the high angular momentum and very specific configuration of isomeric states. It was realized that processes with participation of atomic electrons may supply higher abundance for isomer depletion compared to purely photon-induced nuclear reactions, just as internal electron conversion is known to dominate gamma emission in the spontaneous decay of isomers. Recently, the atomic-nuclear mechanisms for isomer transmutation were discussed in many publications describing different schemes and modifications of the processes. Among them there may be listed: nuclear excitation by electron transition, NEET, the process introduced in [7] and successfully observed [8,9] only 30 years past its introduction; nuclear excitation by electron capture, NEEC, revitalized in [10, 11]; nuclear excitation by Auger transition, NEAT, a proposed process [12]; accelerated nuclear decay due to bound electron conversion BIC [13]; and NEET through the decay of autoionization states (AS) [14]. The corresponding new proposals for experiments should also be mentioned [15–18].

#### **1. NEET PROBABILITY**

The studies with isomeric atoms in ion traps, ECRIT and EBIT, may throw some light to the problem of isomer transmutation, despite technical difficulties.

<sup>\*</sup>We use «transmutation» for isomer population and depletion, because both processes supply a transition from one nuclear species to another, even disregarding any subsequent decay.

In [15], we have discussed the rate of the  $^{242m}$ Am depletion in an ECRIT through the NEET process with atomic transitions filling an *M*-shell vacancy. The power of typical ECR systems is not enough to reach equilibrium ionization of Am atoms to the *M* shell. However, with achievable charge states of about 30<sup>+</sup>, the electron binding energy is strongly increased and the *N*-shell vacancy may become active for the nuclear excitation.

When the real ionization rate does not allow high-charge states in equilibrium, there is still a probability for single-ionization events creating vacancies in deeper orbits. The charge equilibration is reached only if the ionization and vacancy-decay rates are in balance. Remind that the latter rate is typically high due to short lifetime of vacancies, so for the balance the ionization rate must also be very high. But, a balance is not needed for the NEET process. It would be enough if deep ionization vacancies appear systematically as a result of regular charge-state oscillations around the equilibrium charge q. This is not similar to typical requirements for ion-beam current maximization. When ions are extracted from an ECRIS, the maximum current corresponds to charge states near the equilibrium q.

Past ionization or excitation, relaxation of the excited atomic shell happens within a short time range, and electrons are going to occupy the lowest orbits. For NEET in an ECR system, it is essential to perturb the static occupation function. A nuclear conversion may happen within the characteristic lifetime of the atomic excitation, being a branch of the relaxation processes.

Let us discuss now the time evolution of ionized-atom configurations. Spontaneous decay of vacancies in inner shells, from K to O shells is typically characterized by lifetime  $\tau \leq 1$  ps. After some individual ionization or excitation event, the electron transitions follow from upper to lower levels and are accompanied in some cases with an autoionization cascade. All these cascades may over in a short time, shorter than the typical period between two consecutive perturbations by external impact. Thus, it would be reasonable to suppose that electrons are sitting in the lowest levels corresponding to a definite charge state, and such a configuration serves as an initial target for each impact of an external electron or ion. That is true for processes in ion traps because the rate of external perturbations is lower than the decay rate.

Very short and powerful laser pulses may generate different scenarios, when decay and nuclear conversion of the atomic excitations are delayed past the impact of the short radiation burst. But in traps, a static electronic configuration exists between consecutive collisions. NEET events may happen within a short time past each perturbation. In the presence of effective electron capture in ion collisions with the ballast gas, the occupation function should not be static even during the period between fast-electron impacts. This may enhance the rate of NEET. Anyway, the probability function must dynamically characterize the occupation of different atomic levels by electrons with account of all active processes. It would be possible to introduce an electron-orbit occupation function  $P_{\nu}$  that may be mathematically approximated by the Fermi function. The distribution should be normalized applying the equation:

$$\Sigma_{\nu} n_{\nu} P_{\nu} = (Z - q), \tag{1}$$

where  $n_{\nu}$  is electron number corresponded to the complete filling of level  $\nu$ ; q is the equilibrium charge state of the ion in a trap under definite conditions. The  $\nu$ is an ordering number of levels from the deeper bound orbitals to higher orbitals. The  $P_{\nu}$  function describes the probability of an electron existing in a definite level. It is averaged over many impact events, although each individual collision will produce some particular individual configuration. The distribution  $P_{\nu}$  reflects the dynamics of electron capture and loss processes, but it may be more or less stable as a description of the overall ionic characteristics. The rate of nuclear excitations in traps should be defined by  $P_{\nu}$ , as well as by basic parameters of the atomic and nuclear transitions.

In Fig. 1, the occupation probability  $P_{\nu}$  is shown as a function of  $\nu$  with the solid curve. The dashed line corresponds to the static distribution past relaxation of atomic excitations. It is clear that the electron transition to a deeper-bound level would be possible when  $P_{\nu}$  is not zero for a higher level, and  $P_{\nu}$  is not unity for a lower level. Such a transition is shown by the curve with an arrow, representing NEET in radiationless events.

Confinement of atoms in an ion trap provides multiple opportunities for ionization of each atom, and an experiment with  $^{242m}Am$  isomeric atoms in an



Fig. 1. Occupation-probability function for atomic levels: solid curve — in dynamics due to the perturbations, and dashed line — past relaxation

ECRIT [15] remains attractive. The EBIT system may supply higher charge states, but the general production of trapped ions in an ECRIT exceeds that in an EBIT by orders of magnitude. Nevertheless, both types of traps are promising for experiments. A possibility to organize perfect resonance conditions for NEEC [17] supports the application of a tunable EBIT system as an advantageous scheme.

Over recent years, there has been some progress in determining the NEET probability in experiments [8,9,19] and by theoretical simulations [20]. A low probability for NEET at a level of  $10^{-8}$ – $10^{-9}$  per one vacancy in the K shells of <sup>197</sup>Au [8] and <sup>193</sup>Ir [9] atoms provokes a question: Is the probability diminished as a consequence of the weak atomic-nuclear coupling due to the wave functions localized in different volumes, or by other factors? It is known that internal conversion manifests itself in many cases as a high-probability process. Thus, in general the atomic-nuclear coupling should not be very weak, so that the explanation for the low NEET probability may be found in the mismatch of the widths of atomic and nuclear resonances. This was discussed in the review article [6] and earlier in [15].

In analogy to radio physics, the connected systems interact strongly if their resonance parameters are matched, both in frequency and in the quality Q factor. For the atomic-nuclear coupling, the frequencies just correspond to the energies of the transitions in both subsystems and the Q factors are defined by the resonance widths. Atomic levels are typically much wider than nuclear excited states. But, in the favorable case of the strong atomic-nuclear coupling, the hybridization of the atomic and nuclear states cannot be excluded [15, 21]. A detailed description of this hybridization has not yet been obtained, and it would be good to find a simplified procedure by which to produce a theoretical estimate of the probability  $P_{\text{NEET}}$  in order to deduce some conclusions about the coupling strength by comparison to experiments.

The NEET probability must be given by an overlap integral between the atomic and nuclear resonances, multiplied by the ratio of their widths. The nuclear resonance width  $\Gamma_n$  is negligible as compared to the atomic-state ionization width  $\Gamma_a$ . The expression for  $P_{\text{NEET}}$  becomes:

$$P_{\text{NEET}} = \frac{\Gamma_n' 2 \sqrt{\Gamma_a^2 + (\Gamma_a')^2}}{\Gamma_a' \sqrt{\pi} \Gamma_a \Gamma_a'} \int \exp(-4(\ln 2) \left[ \left(\frac{E}{\Gamma_a}\right)^2 + \left(\frac{E - \Delta E}{\Gamma_a'}\right)^2 \right] dE.$$
<sup>(2)</sup>

A Gaussian shape of the atomic resonance is assumed, and the  $E_n$ ,  $E_a$  values correspond to the peak positions of the nuclear and atomic resonances, respectively. The detuning between the resonances is given by  $\Delta E = (E_n - E_a)$ . Equation (2) contains the ratio of nuclear-to-atomic transition strengths, with a simple widths ratio and a folding integral of the resonances. After a NEET

event, the hybridized system remains with a vacancy at the M1 shell and its width  $\Gamma'_a$  defines the NEET resonance. The atom-ionization resonance is due to the total width  $\Gamma_a$  of the K vacancy [8]. The overlap between resonances is illustrated in Fig.2. Equation (1) should define the NEET probability per one event of ionization, unless other factors like atomic-nuclear decoupling are taken into account.



Fig. 2. Overlap of the atomic and nuclear resonances responsible for NEET

Consider the observed NEET process due to atomic transition of an M1 electron to a K vacancy in <sup>197</sup>Au [8]. Equation (2) is used for the nuclear level at 77.339 keV. The width of the K vacancy in the gold atom is taken to be  $\Gamma_a = 54$  eV, following [22], and a detuning of  $\Delta E = 41$  eV according to the experiment [8].

The nuclear data [23] are used to define the nuclear resonance width. In <sup>197</sup>Au, the first excited level with  $1/2^+$  decays to the  $3/2^+$  ground state with a lifetime of 2.75 ns by an electromagnetic transition mainly of M1 multipolarity. The transition is converted with the coefficient  $\alpha = 4.36$  due to emission of electrons from L shell to the continuum. One may suppose that only the radiative part of the nuclear resonance strength must be active for NEET since the mechanism may be understood in terms of a virtual photon exchange between the atom and the nucleus. Indeed, the nuclear decay with emission of electrons from L shell cannot be considered as an inverse process to the nuclear excitation by the electron transition from M1 shell to K vacancy. Therefore, the nuclear excitation width can be expressed as

$$\Gamma'_n = \Gamma^{\gamma}_n = \hbar g / (1 + \alpha) \tau, \tag{3}$$

where  $\Gamma_n^{\gamma}$  corresponds to the partial gamma width for the nuclear excitation, deduced from the experimentally-measured lifetime of  $\tau = 2.75$  ns and excluding the partial decay by electron conversion by introducing the standard factor of  $(1 + \alpha)^{-1} = 0.187$ . The statistical spin factor is  $g = (2I + 1)/(2I_0 + 1) =$ 

0.5 which arises because of different phase volumes in the spin space for the excitation instead of the decay of the nuclear level. The numerical value of  $\Gamma_n^{\gamma} = 2.24 \cdot 10^{-8}$  eV is obtained from (3).

With substitution of all these quantities in Eq. (2), one obtains

$$P_{\rm NEET} = 5 \cdot 10^{-10},\tag{4}$$

that is by a factor of 0.01 lower than the experimentally determined value in [8]. The origin of this discrepancy is not yet clear: What could be wrong in the simple estimates above? As an assumption for the discussion, we propose the following: Remind that the nuclear excitation rate was deduced from the  $\gamma$  decay of the same nuclear level applying the reversibility rule, so that  $\Gamma'_n = \Gamma^{\gamma}_n$  was used. This may be incorrect. Really, the inverse process for NEET in this case would be the BIC transition with electron excitation from K to M1 orbital, so that  $\Gamma'_n = \Gamma^{\text{BIC}}_n$  should be used. The BIC rate for this transition was never measured, but now we can estimate it as removing the discrepancy between the experimental [8] and presently-calculated  $P_{\text{NEET}}$  values. Taking  $\Gamma^{\text{BIC}}_n = 100 \cdot \Gamma^{\gamma}_n$  one finds rate

$$r_{\rm BIC} = \tau^{-1} = 3 \cdot 10^9 \,\,{\rm s}^{-1}.\tag{5}$$

It is difficult to say if this is realistic until a detailed theoretical simulation or direct experiments are carried out for BIC. To compare, recall the electron conversion rate  $r = \alpha/\tau(1 + \alpha) = 2.96 \cdot 10^8 \text{ s}^{-1}$ , for the decay of first excited nuclear level in <sup>197</sup>Au. This conversion is known to result primarily from *L*-shell electrons. From the described analysis, the conclusion follows as well that the decoupling of atomic and nuclear modes is not clearly manifested because the real NEET probability even exceeds the value obtained in this simple model accounting only for the resonances overlap. Hence, there is no need to introduce an additional suppression due to the decoupling.

The NEET probability for  $^{242m}$ Am could also be estimated provided that the atomic transition energy is tuned exactly to the nuclear transition by the choice of a specific charge state. This depends on the environment for atoms in an ECRIT. At some favorable conditions in the trap,  $P_{\text{NEET}} \sim 10^{-13} - 10^{-14}$  can be obtained. For NEET, the ECRIT system will be more productive than an EBIT due to the higher number of ions in the active volume of the trap, and also because of the existence of modesty rather than deep ionization which allows the presence of electrons at higher levels and, therefore, transitions to a vacancy in a deeper-bound orbital. The latter possibility is illustrated in Fig. 1; it does not look promising for NEEC, but is acceptable for NEET. At the same time, advantages of an EBIT for NEEC are as follows: the tunable energy of the electron beam, high density of beam current, and higher charge states achievable as compared to an ECRIT.

In [15], the depletion rate for  $^{242m}$ Am in an ECRIT was estimated to be about  $10^4 \text{ s}^{-1}$ , assuming  $P_{\text{NEET}} = 10^{-14}$ . This estimate was probably too optimistic. In reality, the rate may be smaller due to both lower  $P_{\text{NEET}}$  and lower productivity of the system for the generation of the needed electronic vacancy. But at the moment it is impossible to exclude even the opposite possibility that more abundant depletion of isomers happens in an ECRIT or in an EBIT as compared to the estimates of [15, 17]. Such traps may serve as a sort of boiler for isomers to produce an energy release, provided that a sufficiently high rate of isomer depletion can be reached.

### 2. POSSIBILITIES FOR NEEC DETECTION

Another atomic-nuclear process, NEEC, has recently attracted attention [10, 11, 17]. The process of nucleus excitation due to electron capture by an atomic vacancy (NEEC) was specified in [17] for the  $^{242m}$ Am isomeric nuclei confined in an EBIT and exposed to low-energy electrons. The scheme of the isomer depletion includes the excitation of the nucleus from an initial isomeric state with  $I^{\pi} = 5^-$  and 48.6 keV to the 3<sup>-</sup> potential activation level located at 4.1 keV above the isomer. Being strongly stripped to the charge state of  $42^+$ (*I*-like), the Am ion could capture an electron to the 5p3/2 atomic level, and the resonance condition for nuclear excitation corresponds to an electron beam energy of 2.658 keV that is available using a tunable EBIT system [17].

Atomic and nuclear transitions could be equalized in energy by tuning the beam–electron velocity, while multipolarity matching is not required for the transition of an electron from the continuum, unlike the conditions for NEET. The resonance matching is yet imperfect due to very different widths of atomic and nuclear modes. The atomic-level full width is smaller than that in the *K*-vacancy case, but still remains at a level of eV. The potential nuclear activation level at 52.7 keV in <sup>242</sup>Am is characterized by a width of about  $1.6 \cdot 10^{-7}$  eV due to its decay mostly to the ground state with a very small branch linked to the isomer. But, the latter branch is needed for isomer excitation to the potential activation level with subsequent depletion of the isomer population. Remind that we are looking for a process that is efficient for isomeric energy release.

For ions in an EBIT, the nuclear decay from the potential activation level back to the isomer ( $\Delta E = 4.1 \text{ keV}$ ) may happen mainly via  $\gamma$  emission because the internal conversion is suppressed [11] in strongly ionized atomic species. Therefore, only the radiative nuclear width must be considered and its value was calculated using the standard nuclear electromagnetic transition model. The reduced probability B(E2) has not yet been measured for the transition from the activation level with 3<sup>-</sup> to the isomeric 5<sup>-</sup> level. A theoretical estimate of the Weisskopf strength for this E2 transition resulted in a value of about  $b = 10^{-9}$ for decay branch to the isomer. In the case of NEEC, the effective nuclear width may be expressed similarly as above for NEET in Eq. (3):

$$\Gamma'_n = \hbar bg / [(1+\alpha)\tau], \tag{6}$$

where  $\Gamma'_n$  is a partial gamma width of the nuclear activation level for decay to the isomer. The *b* is the branching ratio for the transition from activation level back to the isomer and *g* factor is inserted again to distinguish the inverse processes of excitation and decay. The factor *b* was not included in Eq. (3) because, in definition, b = 1 for the decay of the first excited nuclear level. The situation is different for an intermediate state of the isomer depletion.

For the NEEC probability estimate, we assume that the electron beam energy can be arranged exactly at a value needed to match the nuclear transition. Therefore, the detuning factor is omitted here, unlike in Eq. (2). The atomic level width,  $\Gamma_a$ , in the discussed case is defined mainly by its radiative part. Not only electron conversion of nuclear decay, but also Auger and Coster–Kronig processes are diminished in highly stripped ions [11]. In any case, the  $\Gamma_a$  width makes not much significance for NEEC because the electron transition from the continuous spectrum is influenced more by the electron beam energy resolution  $\varepsilon$ . This inhomogeneous broadening exceeds both nuclear and atomic widths. Finally, the rate of events can be defined by the expression:

$$r_{\rm NEEC} = \sigma_{\rm REC} \, N_{\rm at} \, I \, \Gamma'_n / \varepsilon, \tag{7}$$

where  $\sigma_{\rm REC}$  is the radiative electron-capture cross section by the ion (see comments below),  $N_{\rm at}$  is the number of ions in the active volume of the trap, and I is the electron current density within the trap expressed in cm<sup>-2</sup>. s<sup>-1</sup>. The factor of  $\Gamma'_n/\varepsilon \ll 1$  just defines the rate of NEEC. Longitudinal Doppler broadening remains negligible, as compared to  $\varepsilon \approx 50$  eV typical in an EBIT.

The radiative electron capture (REC) process may be considered as the basic step initiating NEEC. Its high cross section was found to be near  $10^{-15}$  cm<sup>2</sup>/atom in atomic-collision experiments. In principle, this is a summed cross section for REC to all vacant levels in collision of an ion with the target atom. Unlike that, capture of the free electron to a definite atomic orbit in the ion is of importance for NEEC and the corresponding partial cross section should be by two orders of magnitude lower at  $\approx 10^{-17}$  cm<sup>2</sup>/electron.

Taking the numerical values of [17] for the parameters of  $N_{\rm at}$ , I and  $\varepsilon$  in an EBIT, one can deduce a NEEC rate of about  $10^{-7}$  s<sup>-1</sup> for the  $^{242m}$ Am isomer depletion. This is by orders of magnitude lower than the magnitude given in [17]. A reason for the discrepancy is obviously due to the very low branching ratio  $b \approx 10^{-9}$  obtained herein. Let us remind the use of the Weisskopf estimate for the single-particle strength of the 5<sup>-</sup> to 3<sup>-</sup> nuclear transition in  $^{242}$ Am from the isomer to the potential activation level. But it would be impossible to expect a

higher probability for this low-energy E2 transition. On the contrary, it could be even lower taking into account of a structure prohibition, for instance, due to the 3-fold K quantum number hindrance.

In principle, the estimates above were not expected to meet the requirements of high accuracy because of the many simplifications. The described scheme of calculations may underestimate the NEEC and NEET probabilities for excitation of the <sup>242</sup>Am and <sup>197</sup>Au nuclei, respectively. Additional theoretical and especially experimental studies are among challenges for the modern subatomic physics.

Many authors have already indicated [10, 17, 24] their interest to the discovery of NEEC. Experiments can be arranged at different facilities over the world using ion traps and storage rings. The methods and restrictions are also under discussion. Significant difficulties appear in all cases and they are manifested mostly in the form of backgrounds masking the NEEC signal. Intense radiation is generated in REC and Coulomb excitation processes and it will compete with NEEC detection at in-beam experiments. In Table 1, the restrictions visible for different NEEC experiments are briefly indicated. The application of a new method for NEEC detection under recoil stopping after nuclear reaction is characterized in the following section.

Table 1. Effect-to-background conditions in future experiments for NEEC

Method	Restrictions				
a) Srorage ring + gas target	Velocity spread	Physical background			
	of bound electrons	in $\gamma$ detector from			
b) Storage ring + electron	Low yield	<b>REC</b> and Coulumb			
beam in cooler		excitation			
c) EBIT	Low yield Electron beam resolution				
d) NEEC at recoil stopping	Advantage: Relative test of activities obtained in				
	direct and inverse kinematics				

In future experiments, it would be advantageous to study NEEC for nuclear levels located within a few keV above the initial state, whether ground state or isomer. Such cases can be found over the nuclear chart. Promising nuclides according to the Nuclear Data Sheets are listed in Table 2. One can see that transitions to higher levels for 12 isomeric and 11 ground states are characterized by an energy < 10 keV, and they meet the requirements essential for successful NEEC experiments. Some of the states can be useful also for NEET and some were already discussed in the literature as objects for experiments on nuclear excitation by synchrotron or laser radiation. It would not be easy to predict which nucleus will finally provide the discovery of NEEC. Even some new candidates may appear because the collection of nuclear data becomes more developed and abundant over each decade.

a) I fom isomers						
Nuclide	$^{73m}$ Se	$^{84m}$ Rb	$^{93m}$ Mo	<sup>96m</sup> Tc	$^{110m}\mathrm{Ag}$	$^{133m}$ Ba
$E^*$ , keV	0.59	3.48	4.8	1.12	1.13	2.93
Nuclide	$^{150m}$ Eu	$^{152m}$ Eu	$^{179m2}\mathrm{Hf}$	$^{189m}$ Os	$^{192m}$ Ir	$^{242m}\mathrm{Am}$
$E^*$ , keV	0.9	0.9	0.07	5.37	5.06	4.1

Table 2. Excited levels-candidates for nuclear excitation by electron capture, NEEC

b)	From	ground	states
<b>D</b> )	I I VIII	ground	states

a) From isomers

b) From ground states						
Nuclide	<sup>84</sup> Kr	<sup>151</sup> Sm	<sup>169</sup> Tm	<sup>171</sup> Tm	<sup>181</sup> Ta	$^{187}$ Os
$E^*$ , keV	9.405	4.862	8.413	5.036	6.238	9.746
Nuclide	<sup>193</sup> Pt	<sup>201</sup> Hg	<sup>205</sup> Pb	<sup>229</sup> Th	<sup>235</sup> U	
$E^*$ , keV	1.642	1.556	2.239	0.0076	0.0765	

## 3. NEEC UNDER RECOIL STOPPING

The mentioned links between NEEC and REC induce an idea that some other atomic collision effects may potentially support successful detection of NEEC and NEET, in addition to those discussed above. The author of [14, 16] describes a scheme of NEET via population of autoionization states (AS) of an atom and consecutive nuclear excitation past the AS decay. The proposed method involves an ion source supplying a beam of highly-charged ions in energy range of about 100 keV for excitation of AS in a solid target. The nuclear decay radiation after NEET is to be detected in prompt or delayed time domain. The scheme of irradiation looks like a typical atomic-collision technique, and it is worthy to remind that high angular-momentum Rydberg states are populated in ion interaction with solid surface [25]. Hollow atoms are formed as a result because the outer orbits are occupied by electrons while the inner shells are empty.

The autoionization states, AS, are also among this class of configurations, with electrons at outer-bound orbits. They may not survive in transmission of projectile or recoil atoms through the solid, and the gas phase should be more safe for the exploration of AS and for other studies involving the electron states with high orbital momentum. In such sense, our proposal [15] for isomeric atom confinement in electron-cyclotron resonance ion trap, ECRIT, promises the unsuppressed rate for all processes with strongly perturbed atomic shells. The discussed above NEEC in the electron-beam ion trap EBIT [17] may also be advantageous. In general, multiple ionization, excitation and decay of atomic levels of each ion confined in a trap should be more productive for isomer depletion, as compared to the beam experiments.

However, one should not forget that for production of isomeric nuclei, beams and solid targets are normally used. Significant recoil momentum is transferred to the reaction products by the bombarding projectile. Thus, a question arises, what happens with the radioactive isomeric nuclei under stopping in the target or in a collecting absorber? It is well known that ion excitation and ionization happen with high probability during the transmission of swift ions (recoils) through a solid [26]. At high velocity of  $v \ge 0.1 c$ , the heavy-element ion can be stripped to a charge of about  $40^+$ – $50^+$ , that is, comparable to the charge states needed for NEEC in an EBIT. The electron-impact energy enough for NEEC could also be supplied by the ion velocity under stopping.

As is indicated above, the collision of a 2.658 keV electron with an *I*-like  $^{242m}$ Am<sup>42+</sup> ion corresponds to the resonance condition [17]. An electron impact of similar velocity takes place at ion energy of about 4.9 MeV/u in matter. Isomeric  $^{242m}$ Am nuclei can be produced with high cross section by bombarding a deuterium target by 5 MeV/u  $^{241}$ Pu ions through the (*d*, *n*) reaction in inverse kinematics. The produced  $^{242m}$ Am ion conserves practically the full projectile velocity after this reaction and the charge distribution is peaked at 54<sup>+</sup> when passing through a carbon degrader [26]. Lower charge states are formed at similar velocities in a gas medium, as is well known from the practice of ion charge multiplication in multi-step acceleration schemes.

A charge state of  $42^+$  must be available after recoil stopping in a gas target or degrader so that the NEEC transition in  $^{242m}$ Am ion becomes possible. The currently proposed experimental scheme is shown in Fig. 3. The recoil nucleus transmits through the gas degrader and its velocity is systematically decreasing. At some distance, the resonance conditions for NEEC might be fulfilled, and it is indicated as a defining the resonance range within the gas stopper. The resonance coordinate distribution should be wide due to the velocity spread and possible variation of the charge state for the definite NEEC transition. For instance, NEEC to the  $5p_{3/2}$  orbital in the Am ion at  $q = 42^+$  may happen at another velocity in the higher charged ions where this orbital is also vacant.

A nuclear radiation detector should be installed near the «resonance-range» location to measure prompt emission accompanying events of isomer depletion by



Fig. 3. Schematic layout of the experiment on excitation of recoil nuclei by NEEC under stopping

NEEC. Until now, absolutely no experiments have been attempted to explore the survival of isomers under stopping of the reaction products in matter. The depletion rate could be negligible, in general, but may be manifested for some special isomers. The scheme is advantageous because each ion will reach at some point in the degrader a resonance velocity required for NEEC, despite the velocity spread arising due to the straggling under stopping and by other reasons.

Variations of the electron kinetic  $E_{kin}$  and binding  $B_e$  energies in the ion rest frame are illustrated in Fig. 4, b, displaying them as a function of the stopping range. The  $E_{kin}$  and  $B_e$  values are decreased under stopping because both the ion velocity v and the equilibrium charge q(v) are regularly diminishing, as is illustrated in Fig. 4, a. The resonance takes place when a total energy of REC,  $E_e = E_{kin} + B_e$ , becomes equal to the energy  $E_n$  of the nuclear level that we want to excite. The resonance condition may even appear multiple times for single ions because of charge-state fluctuations up and down in regular charge-exchange interactions. Distribution of the resonance coordinate is shown schematically in Fig. 4, c.

In presence of the backgrounds, the sensitivity of the NEEC process detection would be a critical point in this new scheme, as in more sophisticated experiments with an EBIT or a storage ring. It was mentioned that the high cross section of



Fig. 4. a) Changes of ion velocity v and equilibrium charge q(v); together with b) kinetic, potential and total energies  $E_{kin}$ ,  $B_e$  and  $E_e$  of a free electron in the ion-rest frame, shown in a function of the stopping range L; c) NEEC resonance corresponding to the coordinate where the electron total energy  $E_e = E_{kin} + B_e$  becomes equal to nuclear level energy  $E_n$ 

REC provides a benefit for higher yield of NEEC, but it may also restrict the detection sensitivity by creating background. Recall that the equilibration of charge for swift ions in matter corresponds to a free-path length of  $\approx 1 \ \mu g/cm^2$ . Cross sections on the scale of  $10^{-15} \text{ cm}^2/\text{atom}$  can immediately be deduced, confirming high probability of electron capture and loss under stopping.

Nevertheless, it would be good to find a simple experimental scheme supplying the high sensitivity and reliability for NEEC detection. Such a variant can be realized, for instance, for the  ${}^{2}\text{H}({}^{241}\text{Pu}, n) {}^{242m}\text{Am}$  reaction. The  ${}^{242m}\text{Am}$  isomer must be produced both in the direct kinematics, when the  ${}^{241}\text{Pu}$  target is irradiated by the deuteron beam and in inverse kinematics with the  ${}^{2}\text{H}$  target and the  ${}^{241}\text{Pu}$  beam. The reaction cross section is invariant in any kinematical frame and one can compare the values obtained after two experiments in the direct and inverse kinematics. The depletion of the isomer under stopping of the 5 MeV/u  ${}^{242m}\text{Am}$  ions should reduce the detected yield of the isomer activity. Lower cross section obtained in the inverse scheme will indicate the isomer depletion due to the NEEC mechanism. The corresponding NEEC probability would be deduced from the ratio between the yields in the two experiments.

Assume that the experiment will prove a stability of isomeric configuration under stopping. Then, the general conclusion is reached that there is a low efficiency for the atomic-nuclear conversion, and some limits are to be established for the rate of processes. The in-beam detection of an isomer depletion in prompt mode may be more complicated and not straightforward in the method, but intriguing nevertheless for the future advances. The yields ratio taken at conditions of the direct versus inverse kinematics looks more attractive because activation methods are the best way to provide a high sensitivity. In addition, relative measurements are recognized as the most reliable approach.

#### CONCLUSIONS

Atomic-nuclear co-operative processes are discussed in the context of nuclear isomer transmutation. Unfortunately, few experimental studies could be quoted on this special topic. In the present work, estimates are given for the  $^{242m}$ Am isomer depletion via NEET and NEEC mechanisms using modern ion traps: ECRIT and EBIT. A simplified scheme of calculations is used, but can be verified by calibration to a measured NEET probability. The yield is found to be relatively low, and an appropriate experimental scheme is yet to be devised. The unknown probability of NEEC can be deduced from a new type of experiment studying isomer survival under stopping of reaction recoils in a gas degrader. The corresponding scheme is proposed and characterized in a semi-quantitative manner.

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