Bound internal conversion versus nuclear excitation by electron transition: Revision of the theory of optical pumping of the ^{229m}Th isomer

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Two-photon optical pumping of the 7.6-eV nuclear isomer in the singly ionized atoms of ^{229}Th is considered. Differences between two mechanisms of the pumping, nuclear excitation in the electronic transition (NEET) and bound internal conversion (BIC), are derived and analyzed numerically. The BIC mechanism turns out to be more effective, by orders of magnitude, in accordance with previous calculations. Moreover, a numerical smallness in the NEET scheme is explicitly pointed out concerning singly and doubly charged ions. That is related to the smallness of the final vertex, responsible for conservation of energy. In the case of BIC, the calculated pumping rate of the isomer for the most effective scheme may be as high as 0.03 s^{-1}.

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I. INTRODUCTION

Combined atomic-nuclear transitions firmly occupy an essential part of modern basic research. The story goes back to the 1940s, when Wheeler suggested that the ^{238}U nucleus has a chance to be excited in a radiationless muonic transition $2s \rightarrow 1s$ [1]. Zaretsky advanced the idea and proposed that the probabilities of the radiationless electric dipole transitions $2p \rightarrow 1s$ and $3p \rightarrow 1s$ are of close value [2]. Since then, it has been shown in a number of papers [3–5] that the probabilities of many other transitions, including electric quadrupole $3d \rightarrow 1s$ and electric octupole $3d \rightarrow 2p$, are essentially of the same value.

Excited actinide atoms undergo fission, which is called prompt fission as distinct from delayed fission, occurring as a result of the muon capture. Study of prompt fission provides unique information on the fission dynamics (e.g., Refs. [6,7]). These predictions were fully confirmed in experimental research [8–10]. Furthermore, in Ref. [11] a resonance phenomena of muonic x-ray radiation from the heavy fragments of prompt fission was observed, predicted in Ref. [12]. That was the first manifestation of bound internal conversion (BIC) in the form of resonance conversion (RC) of the nuclear γ rays on the bound muon. As a result, the muon is resonantly lifted up to the $2s$ orbit, from where it re-emits the given energy in the form of the x ray.

There are many manifestations of the combined transitions in usual atoms. Letokhov noted the highly attractive potential manifestations of nuclear excitations in optical phenomena (e.g., Ref. [13]). Morita [14] proposed that a creation of a hole in the inner electron shells might result in nuclear excitation in the electronic transition (NEET). The latter process was finally discovered experimentally in gold atoms as a result of many purposeful attempts [15]. From the viewpoint of the combined transitions, of great interest are nuclides, in which there are excited states with extremely low energies, within the scale of a few eV or keV: ^{201}Hg, ^{189}Os, ^{237}Np, ^{235}U, ^{229}Th, and other nuclides. Such levels are isomeric owing to small their energies. They effectively mix up with close atomic levels, forming resonances in the optical domain [7,16]. This gives a chance of operating with the lifetimes of these isomers in a resonant field of laser radiation. Specifically, this offers a way of accelerating nuclear decay via laser-assisted resonance conversion [17,18]. Electron shells appear here as resonators. Moreover, electron shells can be used in order to master α decay rates [19]. There are indications that this can be also applied for drastic acceleration of β decay rate (e.g., [20] and references therein).

Use of the electron shell as the mediator for the purpose of mastering nuclear reactions with the help of lasers was proposed in Ref. [17]. BIC was shown to be the proper instrument for this purpose. The effective way was offered of accelerating the nuclear de-excitation rate by virtue of the nonlinear effect of merging two photons on the electron shell. One of the photons comes from the excited nucleus and the other from the externally applied laser field. If the energy of the both photons matches the atomic energy, the probability increases drastically. Namely for this reason, BIC was also called the resonance conversion. The theory of RC was summed up in Ref. [21]. Specifically, the role of energy conservation was described in detail. BIC was experimentally observed in Ref. [22].

Reverse BIC can be applied as an effective tool of nuclear excitation by laser [23–25]. At first sight, seemingly it works like NEET. Morita noted that NEET may be called the inverse conversion process [14]. However, there is a critical feature with respect to which both mechanisms of nuclear excitation can be distinguished experimentally. It is energy conservation that remained out of scope explicitly in Ref. [14], and the latter circumstance caused an intense discussion [15,23,26]. Moreover, this makes strong consequences for experiments, as we will see below. Morita considered the two processes of creating the hole and the succeeding nuclear excitation as independent of one another. That means that a regular hole is produced, with its eigenenergy, that is on the mass shell, with its proper state width. Instead of a hole, with respect to the ^{229m}Th isomer, one can also resonantly excite an external electron to a discrete level and search for the nuclear excitation which may be expected in the next electron

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transition [27]. With some assumptions, such a mechanism can be conditionally named as a kind of NEET. The fundamental difference is that in the classical NEET as proposed by Morita the initial electron brings away an energy (and momentum, e.g., Ref. [28] and references cited therein). In the case of absorption of laser photon, its full energy remains in the atom. In contrast with NEET, BIC is based on the interplay of the on-shell and off-shell processes from the very beginning. The RC mechanism turns out to be much more effective for laser pumping the few-eV isomer of ^{229}Th [23,24]. Thus, it was shown in these papers that the most effective is the reverse RC mechanism with the chain excitation $7s$-$8p_{1/2}$-$8s$-$7s$, with the nuclear excitation on the last step. It is essential that the resonant $8s$ state is formed not with its eigenenergy, as it would be in the case of NEET, but with the nuclear isomeric energy that is off the mass shell. Conservation of energy is restored when this energy is transferred to the nucleus. In contrast, the NEET mechanism turns out not to be efficient at all. This question was considered in finer detail in Ref. [24]. Moreover, employment of the NEET scheme bares other problems, noted in Refs. [29], namely, three-photon ionization of the Th^+ ions and oscillating population of the electronic levels. These processes grow with increasing laser power. This limits the allowable power of the lasers. We will return to this question in Sec. IV. All these may comprise the reason for the lack of experimental success (why the isomer has not been obtained yet). The main hindering effect, however, in single and double ions may be due to the specific smallness of the concluding relaxing electronic transition to the ground state, Γ_4, in Fig. 1, as shown below in Sec. IV. Open scientific discussion around this topic aimed at a search for most efficient ways, similar to Ref. [30], will be certainly fruitful and looks necessary at this stage of investigations.

Note that, unlike the atomic spectra, the nuclear lines are considered to be much more stable against influence of external fields and environment. They possess narrower

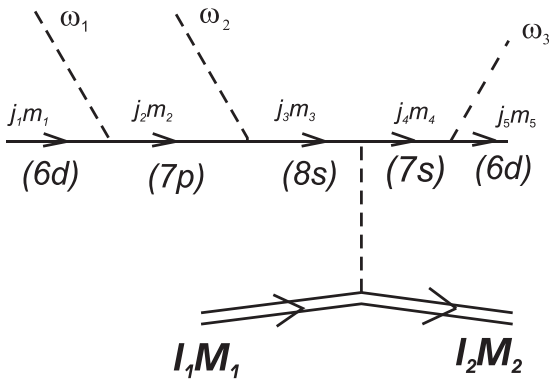

FIG. 1. Feynman graph of the two-photon pumping of the ^{229m}Th nuclear isomer. A resonance photon ω_1 is absorbed, transferring the atom from the ground state j_1m_1 into the first intermediate state j_2m_2. Absorption of the second photon ω_2 transfers the atom to the second intermediate state j_3m_3. Then the atom transfers part of the absorbed energy to the nucleus, inducing its transition from the ground state $|I_1M_1\rangle$ to the isomeric state $|I_2M_2\rangle$, and jumps to the $7s$ state $|j_4m_4\rangle$. Conservation of energy is restored (if necessary) through emission of the photon ω_3, the atom coming back to the ground state.

widths. These advantages do make their use attractive in many aspects, including creation of reference points of frequency in the optical domain and nuclear clock. This gives basis for development of the new nuclear technologies, founded on application of lasers. From such a standpoint, one of the most promising appears to be ^{229}Th, in which the splitting of the basic and excited levels in the nucleus is minimal and makes less than 10 eV (e.g., Refs. [31,32] and references therein). We will use the value of 7.6 eV, following the last data [33]. In the past, many papers were written under the assumption that this energy is 3.5 eV. Knowledge of the precise value is extremely important for applications, but also extremely difficult to measured. A recent publication [31] on the direct observation of the isomer radically changed the situation and also has created a practical footing for the problem of creating a nuclear clock. The unique properties of the nuclide allow construction of a nuclear frequency standard and nuclear clock, with the minimum error at the level of 10^{-19} and less. The latter just meets the contemporary requirements of modern technology and the fundamental physics that require minimal relative error no larger than 10^{-18} to 10^{-19}. This greatly exceeds the capabilities of the currently known devices of the fountain type, which may not significantly reduce the error below 10^{-16}. So far, only with recording devices available in Japan, USA, and elsewhere, is it possible to reduce the error to a few units of the eighteenth decimal [34].

For the current practical applications, first the isomer energy must be measured precisely. To this end, we modify the two-photon excitation of the nuclear isomer through reverse RC in the electron shell to the $8s$-$7s$ electronic transition proposed in Refs. [23–25]. Then knowing the energies of the photons and atomic energies will allow one to know precisely the nuclear isomer energy. This is an example of possible advances given by the new nuclear-optics technologies. In the present paper, we present a comparative prospect of application of BIC or NEET for the two-photon pumping of the isomer. The BIC mechanism is confirmed to be much more effective. Calculations show that with commercially available lasers, the time of pumping may typically be of approximately half a minute in the case of ThII.

The present article is structured as follows. In the next section, we describe the two-photon scheme in general form, taking into account both mechanisms of RC and NEET. The limiting cases of the formulas for the RC and NEET cases are derived in Sec. III. Their qualitative analysis for the case of optical pumping ThII is performed in Sec. IV, and the dominating strength of the reverse RC mechanism is demonstrated. Experimental research of the single ions of ^{229}Th was performed in Refs. [35,36]. Section V is devoted to the numerical calculations of the rate of the optical pumping for the reverse RC mechanism. In Sec. VI the results obtained are summed up and the prospects are outlined.

II. GENERAL FORMULAS FOR THE TWO-PHOTON PUMPING

A general Feynman graph of the two-photon pumping, involving both the RC and NEET mechanisms, is presented in Fig. 1. We mark electron levels with simple single-electron

labels for the sake of clarity. We calculate the diagram in the Furry's representation, making use of the resonance approximation [7,16,37]. Neutral thorium atoms have the ground-state configuration $(7s)^2(6d_{3/2})^2$. Both single-electron states, $7s$ and $6d_{3/2}$, are nearly degenerate. Transformation to the singly charged ions occurs through removing a $7s$ electron. A strong absorption line corresponding to the transition from the ground state $7s(6d_{3/2})^2$ to the state $(7s7p6d)\ J = 5/2$ with the energy of 24874 $\mathrm{cm}^{-1} = 3.084$ eV was observed in Ref. [35]. We consider the latter state as the first intermediate one. The second intermediate state must be one of the states where the $8s$ single-electron component dominates. Nuclear isomer excitation occurs in the $8s$-$7s$ transition. For this transition, coupling of the electrons to the nucleus is maximal. For comparison, in the case of, e.g., the $8s$-$6d$ $M1$ transition, the discrete α_d values turn out to be smaller by six orders of magnitude. Another circumstance which is of interest for experiments is discussed in Sec. IV. The $7s$ state should not necessarily be the ground one. We consider level $(7s^26d)\ J = 5/2$ with the energy of 4113 cm^{-1} to be a probable candidate.

In the case of the RC mechanism, graph in Fig. 1 is the same as that considered in Ref. [24], Fig. 5(a), with the replacement of the initial electron state $7s$ by $6d$ according to the concrete choice of the electron configurations, as explained above. Turning to the numerical analysis, first, we separate out the angular variables, using the Wigner-Eckart theorem (see Appendixes A and B for the definition of the reduced matrix elements), and integrate over them, as described, e.g., in Ref. [22]. This allows one to break the whole expression for the rate of the pumping M into a product of probabilities of constituent elementary processes: consecutive absorption of laser photons ω_1, ω_2, transfer of the absorbed energy to the nucleus, and removal of the residual excitation energy of the atom by emission of photon ω_3.[1]

$$M = \frac{\langle j_2||H'_\gamma||j_1\rangle\langle j_3||H'_\gamma||j_2\rangle\langle I_2 j_4||H'_c||I_1 j_3\rangle}{\left(\omega_1 - \epsilon_2 + i\Gamma^a_2/2\right)\left(\omega_1+\omega_2-\epsilon_3+i\Gamma^a_3/2\right)} \times \frac{\langle j_5||H'_\gamma||j_4\rangle}{\left(\omega_1+\omega_2-\omega_n-\epsilon_4+i\Gamma^a_4/2\right)} D_M \equiv \mathcal{M} D_M, \quad (1)$$

with

$$D_M = \sum_{m_1\mu_1\mu_2\mu_3 M_1 M_2} \frac{C(j_1 m_1 \lambda_1\mu_1|j_2 m_2)}{\sqrt{2j_2+1}} \frac{C(j_2 m_2\lambda_2\mu_2|j_3 m_3)}{\sqrt{2j_3+1}} \times \frac{C(I_2 M_2\lambda_4\mu_4|I_1 M_1) C(j_3 m_3\lambda_4\mu_4|j_4 m_4)}{\sqrt{(2I_1+1)(2j_4+1)}} \times \frac{C(j_4 m_4\lambda_3\mu_3|j_5 m_5)}{\sqrt{2j_5+1}}. \quad (2)$$

Averaging over the initial and summing over the intermediate and final magnetic quantum numbers, we arrive at the following expression for the transition rate:

$$w = \int |\mathcal{M}|^2 \Sigma_D W_1 S_1(\omega_1) d\omega_1\ W_2 S_2(\omega_2) d\omega_2. \quad (3)$$

[1]We use units of $\hbar = c = 1$.

For convenience, we explicitly introduce in (3) the two parameters for each laser, which determine the rates of the resonance processes: intensity W_i and spectral density of the radiation $S_i(\omega_i)$ of the ith laser, normalized at unity. The latter may be approximately taken as the inverse spectral width. I_1, M_1 and I_2, M_2 stand for the nuclear spins and their projections on the quantization axis in the ground and isomeric states, respectively. j_i, m_i are the electronic quantum numbers, λ_i, μ_i designate the multipole orders and magnetic numbers of the external photons, λ_n, μ_n—the quantum numbers of the intermediate photon.

Performing the summation in (2), we arrive at the following expression:

$$\Sigma_D = \frac{1}{(2I_1+1)(2j_1+1)} \sum_{m_2 m_3 \mu_n m_4} |D_M|^2 = 1/[(2j_2+1)(2j_3+1)(2I_2+1)(2j_4+1)(2\lambda_4+1)]. \quad (4)$$

Taking into account the definition (B2), we can put down expression (4) in the form of

$$|\langle I_2 j_2||H'_c||I_1 j_1\rangle|^2 = |\langle I_1 j_1||H'_c||I_2 j_2\rangle|^2 = (2I_2+1)(2\lambda+1)\Gamma_c/2\pi = (2I_2+1)(2\lambda+1)\alpha_\vartheta(7s\to 8s)\Gamma^{(n)}_\gamma/2\pi. \quad (5)$$

In the same way, the two-bar radiative matrix element reads as follows [cf. (A3)]:

$$|\langle j_5||H'_\gamma||j_4\rangle|^2 = \frac{2j_4+1}{2\pi}\Gamma^{(4\to5)}_\gamma. \quad (6)$$

Let us designate

$$t = \frac{\pi^4}{\omega_1^3\omega_2^3} W_1 W_2. \quad (7)$$

Then the general formula for the two-photon pumping becomes

$$w = \frac{t(2I_{ex}+1)}{9(2j_1+1)(2I_g+1)} \alpha_d \Gamma^{(n)}_\gamma |BW|^2. \quad (8)$$

The resonance effects are now expressed in terms of the double integral of the three Breit–Wigner resonance factors:

$$|B_W|^2 = \int b_1 b_2 b_3 S_1(\omega_1) S_2(\omega_2)\, d\omega_1\, d\omega_2, \quad (9)$$

where

$$b_1 = \frac{\Gamma^{(a)}_\gamma(2\to1)/2\pi}{(\epsilon_1+\omega_1-\epsilon_2)^2+(\Gamma_2/2)^2}, \quad (10)$$

$$b_2 = \frac{\Gamma^{(a)}_\gamma(3\to2)/2\pi}{(\epsilon_1+\omega_1+\omega_2-\epsilon_3)^2+(\Gamma_3/2)^2}, \quad (11)$$

$$b_3 = \frac{\Gamma_4/2\pi}{(\epsilon_1+\omega_1+\omega_2-\omega_n-\epsilon_4)^2+(\Gamma_4/2)^2}, \quad (12)$$

and Γ_j stands for the total width of decay of the jth atomic state. $\Gamma^{(a)}_\gamma(i\to j)$ means the radiative atomic transition from the state i to the state j.

III. RESONANCE EFFECTS ARISING IN THE RC AND NEET MECHANISMS

Factor b_1 (10) has a sharp maximum at $\omega_1 = \epsilon_2 - \epsilon_1$. This is condition for the resonance absorption of the first quantum ω_1. Introducing a new variable $x_1 = \epsilon_1 + \omega_1 - \epsilon_2$, one can see that b_1 is essentially reduced to a smeared δ function of this variable x_1.

Furthermore, the integrand in expression (9) has two distinctive maxima with respect to the energy of the second photon ω_2. Each of the maxima corresponds to that or another mechanism, NEET or reverse RC, respectively. Resonance absorption of the second photon corresponds to the NEET mechanism, in accordance with what is said in the introduction. In this case, it is convenient to introduce a new variable, $x_2 = \epsilon_2 + \omega_2 - \epsilon_3$. The resonance condition for NEET is $x_2 = 0$, that is,

$$\omega_2 = \epsilon_3 - \epsilon_2. \tag{13}$$

Note that (13) is independent of the nuclear energy. Energy conservation is restored when the photon ω_3 is emitted, i.e., the secondary hole undergoes decay. The latter decay occurs from the state which is out of the mass shell. Therefore, Γ_4 in the denominator of b_3 in (12) may be omitted. In turn, $b_1 b_2$ is essentially reduced to the product of the two smeared δ functions

$$b_1 b_2 \sim \delta(x_1)\delta(x_1 + x_2). \tag{14}$$

Integration over ω_1 and ω_2 results in

$$|B_W|^2_{\rm NEET} = S_1 S_2 \frac{\Gamma_\gamma^{(a)}(2 \to 1)}{\Gamma_2} \frac{\Gamma_\gamma^{(a)}(3 \to 2)}{\Gamma_3} \frac{\Gamma_4/(2\pi)}{\Delta^2}, \tag{15}$$

where $\Delta = \epsilon_3 - \epsilon_4 - \omega_n$ is the difference of the atomic and nuclear transition energies (the defect of resonance). Note that this probability vanishes at $\Gamma_4 \to 0$, i.e., if state 4 is stable. This has a clear physical sense, in that if violated in NEET the balance of energy cannot be restored.

The other choice was suggested in Refs. [23–26]. This choice is that the state 4 is on the mass shell:

$$\omega_2 = \omega_n + \epsilon_4 - \epsilon_2. \tag{16}$$

This choice can be put in correspondence with the reverse RC mechanism, as explained in the introduction. In this case, it is convenient to introduce the variable $x_3 = \epsilon_2 + \omega_2 - \omega_n - \epsilon_4$. It is now a product of $b_1 b_3$ which essentially reduces to the fold of two smeared δ functions, similar to (14):

$$b_1 b_3 \sim \delta(x_1)\delta(x_1 + x_3). \tag{17}$$

Now the main contribution comes from $x_1 \approx 0$, $x_3 \approx 0$. With the account of (17), integration in (9) yields in

$$|B_W|^2_{\rm RC} = S_1 S_2 \frac{\Gamma_\gamma^{(a)}(2 \to 1)}{\Gamma_2} \frac{\Gamma_\gamma^{(a)}(3 \to 2; \omega_2)/2\pi}{\Delta^2}. \tag{18}$$

where $\Delta = \epsilon_3 - \epsilon_4 - \omega_n$ is again the defect of resonance. Thus, state 4 is on the mass shell. In comparison with (15), notation $\Gamma_\gamma^{(a)}(3 \to 2; \omega_2)$ highlights the fact that the radiative width of the $3 \to 2$ transition is calculated at the energy $\omega_2 \neq \epsilon_3 - \epsilon_2$. State 4 undergoes natural decay to state 5 with the probability of unity and its own lifetime [26]. Integration over ω_3, in view of (17), yields in this unity. Consequently, this vertex can be omitted. The resulting diagram is presented in Fig. 2. In turn, the latter goes over the leading diagram of Ref. [24]. From this consideration, one already can intuitively conclude that the RC mechanism should be stronger, as the NEET mechanism (Fig. 1) contains an additional radiative vertex, and therefore, it looks to be of a higher order in the perturbation series.

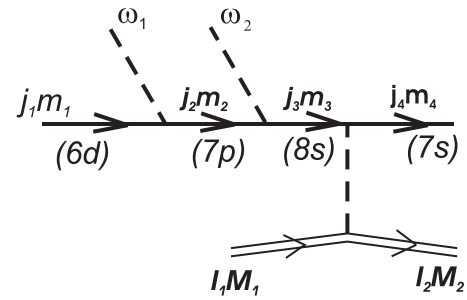

FIG. 2. Feynman graph of the two-photon pumping the ^{229m}Th nuclear isomer within the framework of the inverse resonance conversion mechanism.

IV. QUALITATIVE ANALYSIS

Comparing two expressions (15) and (18), one can estimate the two mechanisms for their efficiency. Actually, comparison of the resonance factors, given by (15) and (18), is quite enough for this purpose. Taking the ratio of the two equations (15) and (18), we arrive at the required relation

$$\zeta = \frac{W_{\rm RC}}{W_{\rm NEET}} = \frac{\Gamma_3}{\Gamma_4} \gg 1. \tag{19}$$

In general, the excitation energy in the state 3 is much higher than that in state 4, the same referring to their widths. That is why the ratio in (19) is much greater than unity [26]. For this reason, state 4 cannot be the ground one. That is, in the NEET mechanism, after absorbing two photons and transferring them to the nucleus, the electron cannot return to the ground state, as was assumed in Ref. [38]. In this case, by definition, $\Gamma_4 = 0$. Consequently, the NEET amplitude vanishes. Correspondingly, ratio (19) $\zeta \to \infty$ at $\Gamma_4 \to 0$. In the case of single ions, our level $(7s^2 6d)J = 5/2$ undergoes radiative decay into the ground state $(6d^2 7s)J = 3/2$ by the electrical quadrupole transition (or two-photon one), with a correspondingly narrow width Γ_4 suppressed by orders of magnitude. There is, however, still another physical reason, which drastically enhances advantage of RC in comparison with the NEET mechanism.

It is known from experiment [15] and calculations, e.g., Refs. [23,39], that the NEET probability turns out to be extremely low, at the level of 8–10 negative orders of magnitude. This is because the NEET probability drastically depends on the energy of the initial electronic hole [26]. It would be maximal at the resonance, if the energy of the hole matched the nuclear energy. It is, however, shifted off the mass shell by the value of Δ. One might think that power of modern lasers could be opposed to this smallness. This is only true within certain limits, however. Apart from NEET, the electron in state 3 may alternatively either absorb the third

photon and leave the atom in the doubly charged state, or undergo induced back transition to the state 2 [29]. These processes grow with increasing laser power, hindering one to realize NEET. As a result, the population probabilities of the upper levels reach saturation at some critical laser power. Both hindering processes, however, should become weaker as the second laser frequency goes off the resonance, as in the case of the RC mechanism.

Along with the graph of Fig. 2, which has the counterpart in paper [24], it is worthy to classify other graphs considered in Ref. [24] in accordance with the above scheme. Those in Fig. 4 should be referred to as due to the NEET mechanism, and graphs in Figs. 2(a) and 3(a) as due to the RC mechanism. Also, Ref. [38] follows the reverse RC mechanism.

V. RESULTS FOR THE TWO-PHOTON PUMPING VIA RC

In the case of the RC mechanism, expression (8) with (18) for the two-photon pumping reads as

$$w_{RC} = \frac{(2j_4+1)}{9(2j_1+1)} W_1 W_2 S_1(\omega_1) S_2(\omega_2) \Gamma_\gamma(3\to 2) \frac{\Gamma_\gamma^{(a)}(2\to 1)}{\Gamma_2} W_n. \tag{20}$$

In expression (20) W_n is the probability of nuclear excitation in the radiationless electron transition. With the account of the occupation numbers, it reads (cf. Ref. [26])

$$W_n = \frac{1}{(2j_3+1)(2j_4+1)} \frac{\alpha_d \Gamma_\gamma^{(n)}}{2\pi \Delta^2}. \tag{21}$$

Calculations were performed for ThII atoms of the pumping rate by means of formulae (20). The related radiative widths and energies were calculated within the framework of the Dirac-Fock method, by use of the package of computer codes RAINE [40]. The following values were obtained: $\alpha_d = 5.34 \times 10^9$ eV, $\Gamma_{7p} \geqslant \Gamma_\gamma^{(7p)} \approx 10^{-7}$ eV. The latter value consists of $\Gamma_\gamma^{(a)}(7p \to 7s) = 3.2 \times 10^{-8}$ eV at the transition energy of $\omega_a = \omega_1 - \epsilon_4$, and $\Gamma_\gamma^{(a)}(7p_{1/2} \to 6d_{3/2}) = 6.89 \times 10^{-8}$ eV at $\omega_a = \omega_1$. Furthermore, $\Gamma_\gamma^{(a)}(8s \to 7p; \omega_2) = 1.96 \times 10^{-8}$ eV. Here the energies ω_1=3.08 eV, $\omega_2 \approx 5$ eV. It follows from Eq. (21) that $W_n = 2.12 \times 10^{-12}$.

It follows from (20) that the excitation rate of the two-photon pumping essentially depends on the spectral widths of both laser beams. At fixed power of the pulses, it is desirable to diminish the widths down to approximately the inverse atomic width. In order to get a realistic estimate, let us use typical parameters of the commercially available lasers: 0.3 mJ per pulse, with the repetition rate of 30 Hz, and the focusing spot of the beam at 0.1×0.1 mm^2, the spectral width being 1.25×10^{-5} eV. Inserting these values into (20), we arrive at the pumping rate of $M = 0.0281$ s^{-1}.

For comparison, in the case of NEET the expression reads

$$w_{\rm NEET} = \frac{2j_4+1}{9(2j_1+1)} W_1 W_2 S_1(\omega_1) S_2(\omega_2) \frac{\Gamma_\gamma^{(2\to1)}}{\Gamma_2} \frac{\Gamma_\gamma^{(3\to2)}}{\Gamma_3} \Gamma_4 W_n \tag{22}$$

with W_n given by (21). The relative value of (22) can be estimated by means of (19). Γ_4 in that equation is actually the probability of the $E2$ radiative transition from the state 4 to the ground one. This transition has a very small width, due to its quadrupole multipolarity. According to our calculation, $\Gamma_4 = 1.69 \times 10^{-18}$ eV. Two-photon decay is more probable. Anyway, the NEET mechanism would have a probability by orders of magnitude as small as the reverse RC one, in good agreement with what was said previously.

VI. DISCUSSION

Previously we performed comparative analysis of the two mechanisms, reverse BIC and NEET, as the instruments for the two-photon optical pumping of the ^{229}Th nuclear isomer. The calculation shows that the expected time of the pumping may be about 30 s with the commercially available lasers, if the reverse RC scheme is used. For this purpose, the second photon energy must satisfy Eq. (16): $\omega_2 = \epsilon_4 + \omega_n - \epsilon_1 - \omega_1$. From the same expression, one can find the isomer energy after its detection is established: $\hbar\omega_n = \hbar\omega_1 + \hbar\omega_2 - \epsilon_{7s}$.

NEET looks easier to exploit, while the energy of the isomer is not known with enough precision. In contrast, the calculations confirm the higher efficiency of the NEET mechanism expressed in a much higher transition rate. This especially concerns singly and doubly charged ions, where a specific smallness arises due to the necessary $6s \to 6d$ transition at the final step of the NEET chain. Another experimental advantage is that hindering NEET processes of the three-photon ionization and electron-induced oscillations between the levels should be suppressed in the RC mechanism. RC is thus shown to be a robust tool, increasing our capacity with the power of the applied lasers. Moreover, the specific smallness of the NEET effect is explicitly pointed out, which arises due to the necessity to restore conservation of energy via emission of the photon ω_3 in Fig. 1. This lowers the probability by orders of magnitude.

Hopefully, analysis of the results will help to find the optimal scheme, which will lead the experiment to its final success. In turn, that will allow production of the isomer as needed for a practical scheme to create the nuclear frequency standard and the nuclear clock. Moreover, this will open wide prospects for developing new optical-nuclear technologies.

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APPENDIX A: SPONTANEOUS AND INDUCED RADIATIVE TRANSITIONS

Let a system absorb a photon with energy ω and transfer from the ground state 1 to an excited state 2. Photoabsorption cross section is related to the radiative width of the reverse process of γ emission $\Gamma_\gamma(2 \to 1)$ as follows [7,24]:

$$\sigma_{\rm abs}(1 \to 2) = \frac{2I_2+1}{2I_1+1} \left(\frac{\pi}{\omega}\right)^2 \Gamma_\gamma(2 \to 1), \tag{A1}$$

where ω is the transition energy. By making use of the Wigner-Eckart theorem, one can separate the angular variables and dependence on the magnetic quantum numbers. As a result, the radiative amplitude $F_\gamma^{2\to1}$ can be put down as follows:

$$F_\gamma^{2\to1} = \sum_\mu \frac{C(j_1 m_1 \lambda\mu | j_2 m_2)}{\sqrt{2j_2+1}} \langle j_1 || H'_\gamma || j_2 \rangle. \tag{A2}$$

Correspondingly, for the radiative width one arrives at the following expression:

$$\begin{aligned}\Gamma_\gamma(2\to1) &= \frac{2\pi}{2j_2+1} \sum_{m_1\mu m_2} \frac{C^2(j_1 m_1 \lambda\mu | j_2 m_2)}{2j_2+1} \\ &\quad \times |\langle j_2 || H'_\gamma || j_1 \rangle|^2 \\ &= \frac{2\pi}{2j_2+1} |\langle j_1 || H'_\gamma || j_2 \rangle|^2. \end{aligned} \tag{A3}$$

Equations (A2) and (A3) define the reduced matrix element $\langle j_1 || H'_\gamma || j_2 \rangle$. The definition is independent of any concrete form of the width. It is equally valid for the nuclear radiative width, for which we use estimated value, and for the atomic radiative widths, which are calculated by means of package of the computer codes RAINE in the length gauge [24]. Thus defined reduced matrix elements are in compliance with those previously defined in Refs. [22,41].

Photoabsorption cross section $\sigma_{\rm abs}(1\to2)$ is related to the radiative width of the reverse process $\Gamma_\gamma(2\to1)$ by means of [24]

$$\sigma_{\rm abs}(1\to2) = \frac{(2j_2+1)}{(2j_1+1)} \left(\frac{\pi}{\omega}\right)^2 \Gamma_\gamma(2\to1). \tag{A4}$$

In terms of the reduced matrix elements of the radiative $2\to1$ transition (A3) and (A4) may be put down as follows:

$$\sigma_{\rm abs}(1\to2) = \frac{(2\pi)}{(2j_1+1)} \left(\frac{\pi}{\omega}\right)^2 |\langle j_2 || H'_\gamma(\lambda) || j_1 \rangle|^2. \tag{A5}$$

The probability $P(\omega)$ of a photon absorption per unit time in an externally applied laser field is then obtained from the cross section (A5) by multiplying it by the flux of the incoming photons $j = W/\omega$ [42,43], where W is the intensity of the laser radiation and ω is the photon energy. Furthermore, let us average the cross sections over the spectral width of the laser radiation, assuming the latter are of the order of the width of the corresponding atomic line:

$$P(\omega) = \frac{(2\pi)W}{(2j_1+1)} \frac{\pi^2}{\omega^3} |\langle j_2 || H'_\gamma(\lambda) || j_1 \rangle|^2 S(\omega). \tag{A6}$$

Thus defined $S(\omega)$ in (A6) is the spectral intensity of the laser radiation, normalized at unity. It approximately equals the inverse spectral width.

APPENDIX B: INTERNAL CONVERSION TRANSITIONS

Analogously, let us define the reduced conversion amplitude [22,41,44]. Let the nucleus make a transition from an excited state $I_1 M_1$ to a lower state $I_2 M_2$, transferring the energy to an atomic electron which is in the state $j_1 m_1$. The conversion electron is emitted into the state with quantum numbers $j_2 m_2$ in the continuum:

$$\begin{aligned}&\langle I_2 M_2 j_2 m_2 | H'_c | I_1 M_1 j_1 m_1 \rangle \\ &= \sum_\mu \frac{C(I_2 M_2 \lambda\mu | I_1 M_1) C(j_1 m_1 \lambda\mu | j_2 m_2)}{\sqrt{2I_1+1}\sqrt{2j_2+1}} \langle I_2 j_2 || H'_c || I_1 j_1 \rangle \end{aligned} \tag{B1}$$

Correspondingly, conversion width of the nuclear transition reads as follows:

$$\begin{aligned}\Gamma_c^{2\to1} &= \frac{2\pi}{2I_2+1} \\ &\quad \times \sum_{m_1 m_2 \mu M_1 M_2} \frac{C^2(I_1 M_1 \lambda\mu | I_2 M_2) C^2(j_2 m_2 \lambda\mu | j_1 m_1)}{(2I_2+1)(2j_2+1)} \\ &\quad \times |\langle I_1 j_1 || H'_c || I_2 j_2 \rangle|^2 \\ &= \frac{2\pi}{(2I_2+1)(2\lambda+1)} |\langle I_1 j_1 || H'_c || I_2 j_2 \rangle|^2 \equiv \alpha(\tau, L) \Gamma_\gamma^{(n)}. \end{aligned} \tag{B2}$$

Here α is ICC, and $\Gamma_\gamma^{(n)}$ is the radiative nuclear width. Traditionally, we define the conversion width for the primary electronic configuration of the closed shell in the initial state and open shell in the final state.

In the case of BIC, α is merely replaced by α_d, which indicates that fact that $j_2 m_2$ is a virtual state below the continuum.

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