Tuned electron-nucleus resonance as a tool of producing the ^{229m}Th isomer

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Abstract: - Possible ways of resonant optical pumping of the 8.36-eV ^{229}Th nuclear isomer – the most likely candidate for the role of a nuclear frequency standard – are discussed. The two-photon laser-assisted mechanism is generalized to optical pumping of the isomer through the electronic bridge transition in externally applied field of three radiative sources. Attention is focused on the role of the final atomic state width. Its proper variation allows one to make use of the resonating properties of the electron shell to have a gain in the pumping rate of orders of magnitude.

Key-Words: - ^{229}Th isomer, nuclear clocks, nuclear isomers, optical pumpimg, resonance conversion

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1 Introduction

Much attention is being paid to the problem of creating nuclear optical clocks and, accordingly, the next generation frequency standard. Record samples of atomic clocks demonstrate a relative error within several units of 10^{-18} [1], while in order to solve challenging fundamental and applied problems it is necessary to further reduce the errors by another order of magnitude. The development of heavy-ion clocks [2] has good prospects. A further reduction in the error would allow to resolve the long-standing question about the possible drift of the fundamental constants [3]. The most challenging task of modern physics is the search for dark matter and energy. Here the fundamental idea is to detect wave oscillations of particles of ultralight matter in its interaction with ordinary matter [4]. The strengths of nuclear clocks are indicated in many reviews and original works (for example, [5] and references therein). Some of them seem self-evident, such as protection from external fields provided by the electron shell. And their use to search for the drift of fundamental constants has irreplaceable features, since the contribution from the nuclear component, compared to the Coulomb component, to the transition frequency is much stronger than in optical ones. Some projects are based on the joint use of atomic and nuclear clocks, using the specified features of the latters [2,6].

The number one candidate for the creation of nuclear clocks is the unique nuclide of ^{229}Th, whose excited state $3/2^+[631]$ lies at a height of only $\omega_n \approx$ 8.355740(3) eV above the ground state $5/2^+[633]$ [7,8]. For estimates, let us set its lifetime in neutral atoms to be within 10 μs [9,10]. The proper lifetime of the nucleus is much longer due to internal conversion (IC), which greatly enhanced the decay rate of the isomer: ICC (internal conversion coefficient) $\alpha(M1) = 0.987\times10^9$. Thus, impact of IC consists in an increase in the natural width of the isomeric line from $\Gamma_n = 0.667\times10^{-19}$ eV (10^{-5} Hz) to $\Gamma_a = 0.7\times10^{-10}$ eV (10 kHz). An indication has been obtained that in a crystalline environment it can be several times shorter]7,11], not changing the qualitative conclusions, though.

The recent breakthrough [7] was achieved using a highly doped CaF_2 crystal and a four-wave mixing laser. As a result, the thorium isomer transition was optically excited for the first time. Next successful excitation was performed in Ref. [8] by means of frequency comb. In the latter paper, uncertainty in the isomer energy was further diminished by 6 digits, the energy having become 2020.4073843350(21) THz. In this concern, we remind that another effective way of searching for the nuclear optical resonance is offered by exploiting the resonance properties of the electron shell. The interaction of the nucleus with the electron shell is carried out through internal conversion (IC), which in the subthreshold region

turns into discrete, or resonance conversion (RC). The RC concept was formulated yet in relation to deexcitation of fission fragments in muonic atoms [12]. RC was confirmed experimentally in Ref. [13]. A method for laser-induced radiative decay of the 76-eV isomer ^{235}U by means of RC was proposed in Ref. [14]. The method is based on two-photon resonance absorption by the atomic electrons: one photon – virtual – is emitted by the nucleus, the other comes from the external radiation of a tunable laser, which is used in order to tune the resonance. In this way, the question of scanning the laser frequency range at the isomer energy was first posed. Common features of RC with the electronic bridges (EB), introduced by Krutov [15], were noted. It is worth noting that EB was observed experimentally in [16] in the decay of the 30.7-keV isomeric level of ^{93}Nb. RC is actually the cross-invariant continuation of EB into the subthreshold field. In the form of bound internal conversion (BIC) it was discovered in the 35-keV transition in highly charged ^{125}Te ions [12]. Note also Ref. [17] where special features of the two-photon excitation was considered in a general form.

Based on Ref. [18], application of inverse EB was proposed in Ref. [19]. Morita suggested that creation of holes in inner shells can induce a non-radiative electron transition accompanied with excitation of the nucleus (known as NEET). Actually guided by paper by I. S. Batkin [20], Tkalya suggested that lifting up a valence electron to an excited level may induce excitation of the nucleus. For the nucleus to be effectively excited, however, the intermediate electron state should have exactly the same energy as the nuclear transition energy. In fact, Batkin never told that such a resonance can be realized in nature indeed, having considered energy transfer through a virtual electron level. In view of the improbability of such a coincidence, as well as the uncertainty of the isomer energy, a "non-resonance" method was proposed in Ref. [21]. According to the method, resonating electronic levels are populated in a spontaneous transition from a higher shell. Moreover, it was shown that in fact, there is a number of the levels which *form* resonances with the isomer transition. As a result, *all* these resonance intermediate states are populated automatically, contrary to the method of Ref. [19]. An experiment aimed at testing this mechanism was carried out in Ref. [22]. The problem, however, was that the most probable isomer energy at that time was considered to be 3.5±1 eV. The value of 7.6±1 eV [23], close to the present one, appeared much later.

When it became clear that the energy of the isomer lies in the region of 7–8 eV, this induced appearance of projects based on the absorption of two or three laser photons ([24-29] and Refs. cited therein). Whatever project we take, however, we will definitely be faced with the issue of optimizing the scanning step by a proper choice of the bandwidth of the pumping beam. Thus, in Ref. [28] it was noted that the effective width of the isomer line is 40 nuclear widths, which, however, is still too small to make it as a scanning step. In project [8], the line broadening due to the IC reaches 9 orders of magnitude, but exactly the same amount is lost in the excitation cross section [30]. In Ref. [7] the laser beam spectral width was deliberately modulated to an

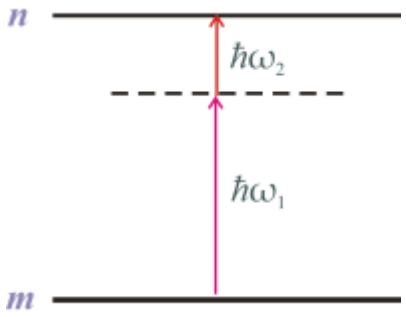

Figure 1. Resonance two-photon absorption of light by atoms. The sum of the energies of the two photons $\hbar\omega_1$ and $\hbar\omega_2$ is exactly equal to the energy of the atomic transition E_{mn}.

effective bandwidth of 10–20 GHz to for better coverage of the search region. As a consequence, the resonance excitation cross section of the isomer, again, decreases by orders of magnitude. Different physics lies in the idea of excitation of the nucleus through electron-nuclear resonance. For example, the natural width of the $7p$ level is eleven orders of magnitude greater than the nuclear one, whilst its excitation cross section does not suffer at all, but on the contrary, gains many orders of magnitude in comparison to the nuclear cross section. The combination of these factors creates attractive prospects. Let us consider some peculiarities of pumping the nuclei through the resonating shell.

2 Remind of the method

Fig. 1 shows a diagram of the resonance absorption of two photons with the energies $\hbar\omega_1$ and $\hbar\omega_2$ by an atomic electron, transferring it from state m to state n. Let the transition energy be E_{mn}. Resonance occurs under the condition that

$$\hbar\omega_1 + \hbar\omega_2 = E_{mn} . \quad (1)$$

The process can be virtual, when condition (1) is violated. It is characterized with a resonance defect Δ:

$$\Delta = \hbar\omega_1 + \hbar\omega_2 - E_{mn} . \quad (2)$$

As Δ increases, the absorption probability rapidly vanishes at Δ > Γ, where Γ is the resonance width equal to the sum of the widths of the levels *m* and *n*. Now imagine that photon $\hbar\omega_1$ is emitted by the nucleus. Then the process goes over the inverse RC in the laser field, and the scanning of the frequency $\hbar\omega_2$ goes over the resonance tuning. In the case of exact resonance, it becomes possible to obtain an acceleration of the nuclear transition by a factor of

$$G \approx (\Delta/\Gamma)^2 , \quad (3)$$

which can comprise more than ten orders of magnitude.

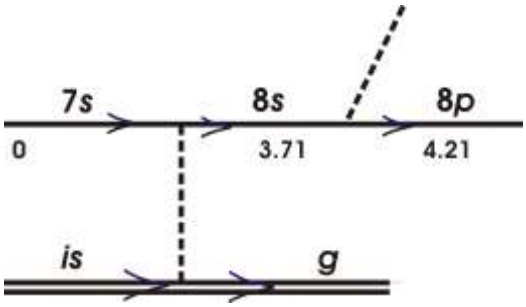

Figure 2. Feynman graph of laser induced resonance decay of the ^{229m}Th isomer. Double line specifies nuclear transition from the isomeric to ground states. Digitals below the electronic line indicate the energies of the electronic levels in eV.

The method was applied to ^{229}Th in [21]. Below we use relativistic units $\hbar = c = m_e = 1$. The idea of that work is illustrated by Feynman diagram in Fig. 2, which reproduces the level scheme of a singly ionized atom. It exploits the same idea as in Fig. 1, but treats it somewhat in a different way. Moreover, in that time ω_n = 3.5 eV was adopted as the most probable isomer energy. In sufficiently strong fields, a satellite appears at the *8p* level with the energy $\varepsilon_{8p} - \omega$. Its amplitude can approach unity in sufficiently strong fields. As a result, the wavefunction of the 8*p* state in the two-level approximation can be put down as follows:

$$\Psi_{8p}(\boldsymbol{r},t) = (|8p> + \beta|8s> e^{-i\omega t})\, e^{-i\varepsilon_{8p}t}. \quad (4)$$

The satellite amplitude in Eq. (4) is determined by the envelope of the electric-field amplitude $\mathcal{E}$ at the location of the atom and detuning from the resonance $\Delta = \varepsilon_{8p} - \varepsilon_{8s} - \omega$ as follows:

$$\beta = \langle 8s|H_L|8p\rangle , \quad (5)$$

where H_L is the Hamiltonian of the interaction of the laser field with the electron:

$$H_L = -e\mathcal{E}z . \quad (6)$$

The resulting expression for the laser-induced deexcitation width of the isomer reads [21]

$$\Gamma_{\rm ind} = |\beta|^2 \frac{\alpha_d(M1)\Gamma_\gamma^{(n)}\Gamma/2\pi}{(\varepsilon_{7s}+\omega_n+\omega-\varepsilon_{8s})^2+(\Gamma/2)^2} . \quad (7)$$

Here $\alpha_d(M1)$ – discrete ICC for the 7*s*-8*s* transition, $\Gamma_\gamma^{(n)}$ – the radiative isomer width, Γ – the total width of the intermediate 8*s* state, ε_i – the eigenvalue of the *i*th atomic state. Out of resonance, $\Gamma_{\rm ind} \sim \Gamma/\Delta^2$, whereas at the resonance $\Gamma_{\rm ind} \sim 1/\Gamma$. In this way, one gets the above resonance enhancement factor of $(\Delta/\Gamma)^2$ by scanning ω, thus tuning the satellite level exactly to resonance with the energy of the isomeric transition ω_n.

3 Application of the method to the thricely ionized thorium atoms

The modern trend in constructing the nuclear clocks is based on the use of three-fold ions of ^{229}Th. The effect of electron bridges is suppressed in them, whereas in neutral atoms it would lead to a 100-fold broadening of the reference line of the 7*s*-8*s* transition. Accordingly, let us consider the 3-photon sequence shown in Figure 3. That was proposed in Ref. [28], with no account of the above physics and related recent results. As a consequence, that article is well-founded experimentally, but the level of presentation leaves much to be desired. Let us consider this matter in finer detail.

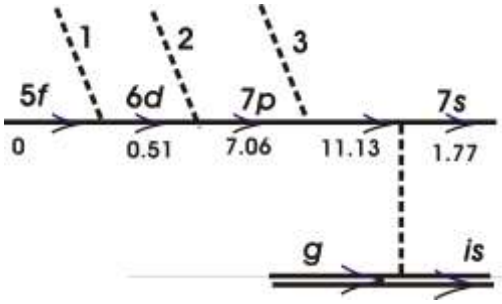

Figure 3. Feynman graph of the laser-assisted excitation of the ^{229}Th isomer via three photon absorption. The photons are numbered for convenience. Digitals below the electronic line indicate the energies of the electronic levels in eV.

In accordance with what is said above, the decisive link in the chain is the isomer excitation following the transition $7p + \omega_3 \rightarrow {}^{229m}$Th + 7*s*. In fact, the intermediate electron state after the photon absorption is formed far from resonance. As a result, a small value of the order of Γ/Δ arises in the probability. Isomer excitation needs strong enough fields with the electrical strength ~10^6 V/cm, when $|\beta|^2$ becomes closer to unity. Assuming that $\Gamma \sim 10^{-8}$ eV, Δ ~ 1 eV, we get that at resonance, the cross section would increase by 16 orders of magnitude. But resonance can be tuned. The case of ^{229m}Th is just considered in Ref. [15]. The recipe is illustrated by the Feynman graph in Figure 3. In sufficiently strong fields, a satellite occurs by the 7*p* level, with an

energy of $\varepsilon_{7p} - \omega$ with an amplitude of the order of unity. By scanning ω, this satellite can be coincided with the isomer energy, which will lead to a desired gain (3).

Such a treating leads to an essential consequence for experiment. It is suggested in Ref. [28] that cool thorium atoms should be prepared in the 7*p* state. Then lasers 1 and 2 are switched off, and laser 3 is switched on. The above treat considers 7*p* and 7*s* levels as an entire couple arising in the field of laser 3. The couple is induced by switching on laser 2. Therefore, we conclude that it is rather the 6*d* state which could be prepared in cooled atoms. As soon as photon 3 is then absorbed, its energy together with that of photons 1 and 2 will be transferred to the nucleus. Furthermore, reasoning similarly, one can conclude that the triple ions should be prepared cooled in the ground state, then all three lasers switched on simultaneously. This is the only way how one can realize scanning with resolution better then the atomic widths of the intermediate 3*d* and 7*p* levels.

4 The role of the width of the final state in the EB chain

Another point for discussion is presented by the width of the final atomic 7*s* state in the chain in Fig. 3 [30]. It is represented by the nuclear width Γ_n, if the 7*s* level is ground, but by the electronic one if it is finite. In the latter case, the 7*s* electron leaves the state before it could be lifted up to the 7*p* state in a Raby transition. As a result, statistical equilibrium is broken, and formulas (7) and others of Ref. [28] concerning the EB probability become invalid. Contrary, the number of the isomeric nuclei increases linearly with time, so that up to inverse inverse population of the ground and isomeric nuclear states can be realized [31]. Actual gain then achieves as much as well about ten orders of magnitude, as we saw in the previous section.

Let us consider in more detail how this enhancement works in experiment. The scanning step at present during direct optical pumping of the isomer exceeds its natural width by orders of magnitude. The probability of its excitation in the laser field decreases by exactly the same number of orders of magnitude. The latter circumstance can be avoided during excitation via the resonant electron bridge \cite{PRl}. Namely, the scanning step h_s can be increased up to the width of the corresponding atomic resonance Γ_a. For $h_s < \Gamma_a$, the gain is expressed in an increased scanning step by a factor of $G = \Gamma_a / \Gamma_n >>> 1$. In the case of $h_s > \Gamma_a$, the advantage is even more transparent: the cross section through the EB is as large as that via plain pumping by a factor of *G*. In turn, due to the energy conservation law, Γ_a is determined by the width of the final 7*s* state. And to determine the isomer energy with an error equal to h_s, the spectral width of all three laser beams in Fig. 3 should not exceed h_s. This is why it is so important to have a finite width in the 7*s* level. Depending on the desired resolution, this could be the width of the *E*2 transition 7*s*-6*d*, or the induced 7*s*-7*p* transition, which can be manipulated, or even just the width of the *M*1 transition between the hyperfine components of the 7s level if the laser resolution is smaller than the hyperfine splitting.

5 Conclusion

This year was signed by an unexpected great progress in construction of the nuclear clock. The isomer was produced by direct laser excitation, by two groups independently. There is still a long way ahead of advancing the tools for laser-nuclear technology. There is still attractive way of pumping the isomer via resonance absorption of several photons from different laser sources. Up to now, there was no serious attempts of making use of resonance properties of the electron shell, apart from paper [22], in spite of substantial discussions. Summing up what is said previously, we conclude that one should exploit the resonance properties of the electron shell, either he scans the pumping laser frequency in search for resonance, or just wants to excite the isomer. This allows one to work with higher the cross-sections which exceed those for bare nuclei by orders of magnitude. In order that this advantage be realized, the electronic state in the isomeric atoms obtained must have a finite width Γ_a. The latter can be chosen in the most optimal way for the actual purposes, depending on the bandwidth of the pumping laser *r*. Then the gain will be $G \approx \Gamma_a / \Gamma_n$. Then with $\Gamma_a < r$ the EB cross-section will be as large as that for the bare nucleus by *G* times.

Three-step isomer excitation in the triple ions can be performed according to Fig. 3, using a laser cooled target of thorium atoms in the ground state. A needed width of the \$7s\$ state can be provided by the proper choice of the related method. This can be either the natural width of the *E*2- or *M*1 atomic transition 7*s*-6*d*, or that of the specially induced Rabi transition 7*s*-7*p*. One can use also the natural widths of the excited hyperfine components of the 7*s* level. All three pulses from the external sources must be applied simultaneously to a cooled target of the thorium atoms in the ground state.

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