JOURNAL OF EXPERIMENTAL AND THEORETICAL PHYSICS, 2024, Vol. 165, No. 2, pp. 135–141

ATOMS, MOLECULES, OPTICS

RESONANT OPTICAL PUMPING OF THE ISOMER ^{229}Th WITH ENERGY 8 eV

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Received August 13, 2023
Revised October 12, 2023
Accepted October 24, 2023

Abstract. The most likely candidate for the role of a nuclear optical standard is the isomer of the nuclear isotope ^{229m}Th with an energy of 8.338 eV. The possibility to specify the value of this energy by resonant optical pumping through an electron bridge was discussed. Proper use of the natural atomic linewidths, which are many orders of magnitude larger than the natural nuclear isomeric linewidth, is critical. Recent studies have shown that broadening due to internal conversion in neutral thorium atoms leads to a gain in scan time of nine orders of magnitude, facilitating the search for electron-nuclear resonance to a feasible level. The reverse resonance conversion method proposed in this article is applicable to ionized thorium atoms. It has the potential to improve experimental efficiency by orders of magnitude. The implementation of this method requires simultaneous excitation of the nucleus and electron shell in the final state. The causal connection between this principle and the solution to the thorium riddle is shown.

Keywords: *isomer ^{229m}Th*, *nuclear-optical clock*, *frequency standard*, *resonance conversion*

DOI: 10.31857/S00444510240201e5

1. INTRODUCTION

The use of spectral lines corresponding to the transitions of nuclear isomers promises good prospects for the creation of a frequency standard and next-generation clocks. Nuclei, being located in the center of the electron shell, are less susceptible to external interference and intracrystalline fields than atomic or molecular systems. These lines are narrow and stable. The problem is that in most nuclei the transitions have energies of tens of keV. Such transitions are problematic to manipulate with lasers. There is a unique nuclide ^{229}Th, whose excited state $3/2^+[631]$ has an energy of $\omega_n = 8.338(24)$ eV (see [1]), which is higher than that of the ground state $5/2^+[633]$.

The main obstacle to the construction of a frequency standard is to reduce the uncertainty in the value of the isomeric transition energy down to the natural width of the nuclear line, which lies at the level of 10^{-19} eV. A possible way to solve this problem is given by the method of resonant photoexcitation of the isomer (optical pumping). For this purpose, a tunable continuous-time laser with a wavelength near the resonance of $\lambda \approx 150$ nm could be used. The problem is that the natural linewidth of the isomeric transition proper is too narrow for scanning: $\Gamma_n = 0.667 \cdot 10^{-19}$ eV (10^{-5} Hz), which would take too much time.

In this connection, let us repeat that the most effective, if not the only way to accelerate scanning is to use the resonance properties of the electron shell. And it is not only the strengthening of the influence of the external field on the nucleus in the case of resonance. An even more important property in the application to scanning is the broadening of the resonance line by many orders of magnitude with respect to the widths of atomic and nuclear transitions. The interaction of the nucleus with the electron shell is realized by internal conversion (IC), which in the subthreshold region turns into discrete or resonance conversion (RC). The concept of RC was formulated as early as [2] with respect to the deexcitation of fission fragments in muon atoms. In [3], a method of laser-assisted nuclear transition enhancement using RC was first proposed using

the example of the isomer ^{235}U with energy of 76 eV. The paper noted the commonality of RC with electron bridges (EB) considered by Krutov [4]. Krutov considered the case when EB is formed in a continuum. EBs are especially effective in the case of large ICCs (IC coefficients). For example, according to calculations [5], EBs increase the probability of radiative decay of the ^{235}U nucleus isomer with energy of 76 eV by five orders of magnitude. In [6], EB was observed in the decay of the ^{93}Nb isomeric level with energy of 30.7 eV. In turn, both RC predictions were observed experimentally in [7] in muon atoms and in ^{125}Te ions [8]. Following the work of [9] and others, the term "inverse EB" was widely used to refer to the mechanisms of electron-shell-mediated photoexcitation of the ^{229}Th isomer. In further details, the concept of inverse EB has been recently reviewed in [10, 11]. It is still worth mentioning the important contribution to the RC theory construction of [12], in which the crucial role of mixing of the electron configurations of the atom's mean-field was demonstrated. In [13], it was shown that the resonance properties of the shell become stronger as electrons are removed from it, up to hydrogen-like ions, in which RC can be observed in the absence of attenuation otherwise created by other electrons. In [14], a set of RC processes was analyzed in terms of their efficiency for optical pumping of the isomer ^{229}Th, whose energy at that time was taken to be 3.5 eV.

Taking into account the current energy value of the isomer, the dominant channel of its decay in neutral atoms becomes IC with ICC

$$\alpha(M1) = 0.987 \cdot 10^{9}.$$

The observed lifetime of the isomer in neutral atoms was found to be10 μs [15]. Taking into account the IC leads to an increase in the natural width of the isomeric line to the value of

$$\Gamma_a = 0.7 \cdot 10^{-10}\,\text{eV}(10\text{kHz}).$$

In practice, the application of the scanning method encounters great difficulties. Lasers in the vacuum violet range with a wavelength of about 150 nm are practically absent. To overcome this problem, the two-photon absorption method was used at the PTB – the Physikalisch-Technische Bundesanstalt, the National Metrology Institute of Germany (Braunschweig) [16]. A unique laser was used to pump the isomer at the University of California Los Angeles (UCLA) in the group of Prof. E. Hudson. Several years of unsuccessful attempts, however, encourage the search for new ways. The project [17] based on the use of the IC channel in neutral isomeric atoms seems to be the most promising at present. Similar developments are underway at the Petersburg Nuclear Physics Institute (PNPI) of the National Research Center "Kurchatov Institute" [18]. Let us consider this project in more detail in order to outline ways to further improve the efficiency of the research based on it as an example of the current state of the experiment.

2. ESTIMATION OF THE REQUIRED SCANNING TIME BY THE IC MECHANISM IN NEUTRAL ATOMS ^{229}Th

For resonance excitation of some system, it is natural to use a light beam with a spectral width approximately equal to the resonance width. Then the cross section of photoexcitation of a level with energy ω[1] can be estimated by the formula [14]

$$\sigma_\gamma(0 \to \omega) = \Gamma_\gamma(\omega \to 0)\frac{2I_\omega + 1}{2I_0 + 1}\left(\frac{\pi}{\omega}\right)^2 S_\omega, \qquad (1)$$

where S_ω is the spectral density of the beam, I_0, I_ω are the spins of the system in the initial and excited states, respectively, $\Gamma_\gamma(\omega \to 0)$ is the radiative width of the back transition. Putting $S_\omega \approx 1/\Gamma_\omega$, where Γ_ω is the total state width ω, and considering $\Gamma_\gamma(\omega \to 0) \approx \Gamma_\omega$, we obtain a universal formula for the estimations

$$\sigma_\gamma(0 \to \omega) = \frac{2I_\omega + 1}{2I_0 + 1}\left(\frac{\pi}{\omega}\right)^2, \qquad (2)$$

applicable equally to both nuclear and atomic systems. This generality is extremely important when considering the phenomena of electron-nuclear resonance.

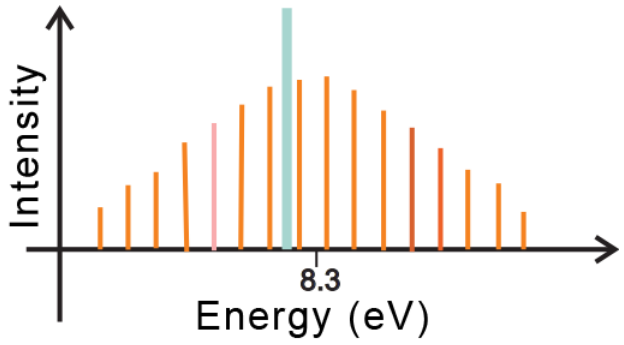

Fig. 1. Frequency comb proposed for isomer energy determination [15]. It consists of $1.2 \cdot 10^5$ comb-like modes (spikes) with variable frequency, centered around 8.3 eV. The width of each mode is 490 Hz, and the power is 10 nW in the center region of the comb. The position of the nuclear line is conventionally shown by a wide vertical line. During the scanning process of the comb teeth frequency, a resonance search is performed.

[1] We use the relativistic system of units $\hbar = c = m_e = 1$.

The project [17] proposes to use the seventh harmonic of the reference beam of a fiber laser with a wavelength of 1070 nm. In the path of the thus obtained continuously generated beam with a power of 1.2 mW, a rotating flap with holes is placed, which produces its cyclic opening and interruption at regular time intervals 100 μs. As a result, the Fourier spectrum of the transformed beam takes the form of a frequency comb located near the target energy and consisting of $1.2 \cdot 10^5$ equidistant teeth. The target is a thin circle with a diameter of 0.3 mm. On its surface, $1.6 \cdot 10^{13}$ atoms ^{229}Th are applied by sputtering or otherwise. The light beam thus obtained is focused onto this target in the form of a frequency comb. The fact of resonance absorption is established by recording the conversion electrons arising from the deexcitation of the isomer. The equipment for recording conversion electrons is developed on the basis of detectors previously used by the authors to detect the decay of the isomer [15] and the first direct measurement of its energy [19].

The irradiation power in each tooth of the comb is 10 nW, half-width 490 Hz ($2 \cdot 10^{-12}$ eV), distance between the teeth is 77 MHz ($3 \cdot 10^{-8}$ eV). From formula (2), it follows that if the irradiation frequency falls in resonance with the nuclear transition, approximately 60 atoms will transition to the isomeric state during each exposure time of 100 μs. The scanning frequency is changed every second. The step of frequency change is equal to the width of the desired line, i.e., 10^{-10} eV. Then it would take 5000 steps to scan the interval between the teeth of the comb, which would take 5000 seconds. To more accurately determine the energy of the isomer, the interval between the tines must be changed. Therefore, several more scanning cycles will be required to refine the isomer energy.

3. RESONANT PUMPING OF THE ISOMER THROUGH THE ELECTRON SHELL

We have dwelt in such detail on the description of the project with Th I to give the best idea of the level of development of the modern experiment, its problems and aims. The main purpose of the present work is to draw attention once again to the fact that one can significantly reduce the time of the experiment if one takes advantage of the resonance properties of the electron shell to enhance the effect of the external field on the nucleus. The example discussed above already utilizes the enhancement of the isomeric transition by the IC channel by 9 orders of magnitude. However, this mechanism can be attributed to the factors of kinematic amplification that passively utilize the broadening of the spectral line of the isomer due to the IC. Resonance with the electron shell as such is not exploited. However, already in single ions, the electron detachment energy becomes larger than the isomer energy, which turns off the IC channel. At the same time, most projects assume the use of exactly ions ^{229}Th.

A suitable dynamic broadening mechanism for ions was presented in [14]. Let us consider its application on the example of single ions ^{229}Th.

In the initial state of the atom, both the nucleus and the valence electron are in the ground state. The shell configuration is $7s6d^2$ with angular momentum $j = 3/2$ [20]. In the virtual discrete conversion process $7s$-electron transfers to the nucleus the energy ω_n required for isomer formation. In the spectral decomposition of the Green's function (Fig. 2), we can, to a first approximation, restrict ourselves to the same $7s$-state, since $8s$- and other levels are much farther away in energy. The Warsaw effect is that it opens up the possibility of mixing by means of RC of the ground and isomeric nuclear states, despite the fact that they have different spins, while preserving the total angular momentum of the F atom [21]. This leads to accelerated decay of the isomer by hundreds of times in the case of hydrogen-like ions ^{229m}Th. If hydrogen-like ions are considered instead of single ions in the diagram in Fig. 2, the full moments $F = 2$ and 3, with the level with $F = 2$ being the ground state, would be possible in the initial state. Similarly, the isomer could form states with the electron with $F = 1$ and 2. A virtual transition to an isomeric state is possible with $F = 2$, but with an initial state with $F = 3$ a process like the one shown in Fig. 2 would be impossible. The IC selection rules automatically take these combinations into account in the formulas for calculating discrete ICCs $\alpha_d(M1)$. In the considered case of single ions, due to the even larger electron shell momentum, $j = 3/2$, a number of combinations are all the more possible for the total momentum F in both initial and final states. All of them are still accounted for by the selection rules for $\alpha_d(M1;7s-7s)$.

The energy balance is restored by absorption of an external photon with frequency (and energy) ω. Having absorbed the energy of the photon, the electron passes to the final $7p$-state. The frequency of the beam is determined from the energy conservation condition:

$$\omega = \omega_n + \varepsilon_{7p}, \tag{3}$$

where ε_{7p} is the energy of $7p$-level. At this frequency, the amplitude passes through a resonance with a width $\Gamma_a = \Gamma_{7p}$, equal to the width of $7p$-level. Therefore,

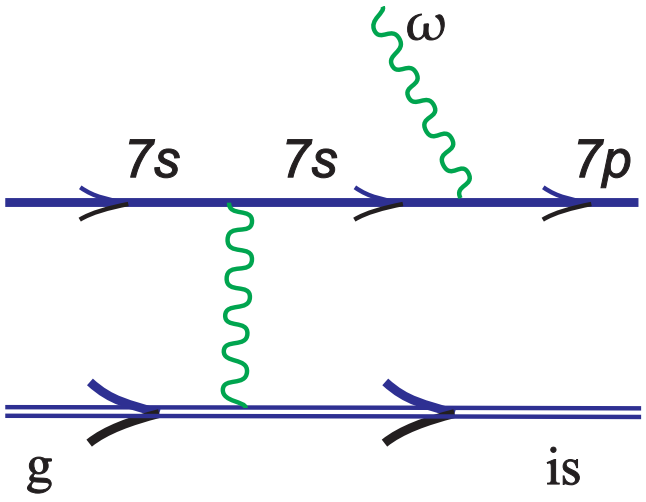

Fig. 2. Feynman plot of the resonant optical pumping of an isomer based on the principle of oscillation of the ground and isomeric states.

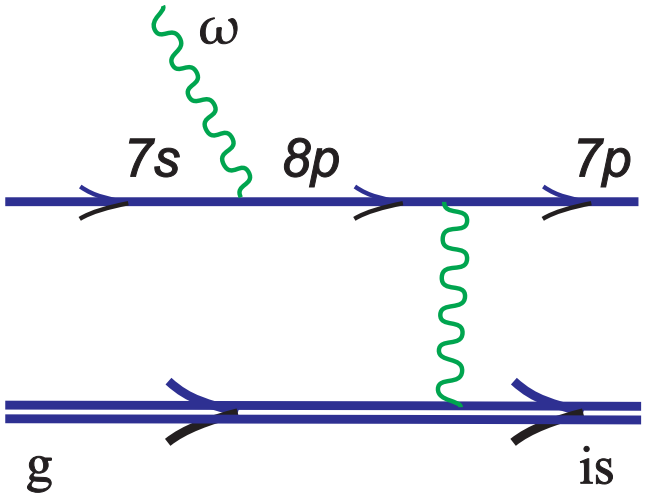

Fig. 3. Feynman plot of the resonant optical pumping of the isomer by the direct mechanism.

for scanning purposes it is reasonable to use a beam with resonance frequency ω and with spectral width equal to the resonance width Γ_a.

Conventionally, let us call this method the first one and compare it with another method, which uses a beam with the same flux but with frequency ω_n and with spectral width equal to the half-width of the isomeric line Γ_n, and which can be used to pump a bare nucleus. For the ratio of the average cross sections of such two processes – the acceleration factor R, the following formula was obtained in [14]

$$R = \frac{\alpha_d(M1;7s-7s)\Gamma_\gamma^{(a)}(\omega;7s-7p)}{2\pi\omega_n^2}. \qquad (4)$$

where $\alpha_d(M1;7s-7s)$ is the dimensional analog of the ICC for $M1$-transition with energy ω_n, $\Gamma_\gamma^{(a)}(\omega;7s-7p)$ is the radiation width of the transition $7s-7p$ with frequency ω (outside the mass surface). It is related to the usual width by the detailed balance relation

$$\Gamma_\gamma^{(a)}(\omega;1-2) = \frac{2j_2+1}{2j_1+1}\Gamma_\gamma^{(a)}(\omega;2-1), \qquad (5)$$

where the numbers $1-2$ and $2-1$ denote the direction of the process in the general case, and $2j_i+1$ is the statistical weight of the i-state. We emphasize that the acceleration factor (4) is achieved on a wider beam but with the same photon flux. However, the resonance energy, according to (4), will be somewhat higher. Correspondingly, the beam power will be also higher – in proportion to the ratio ω / ω_n.

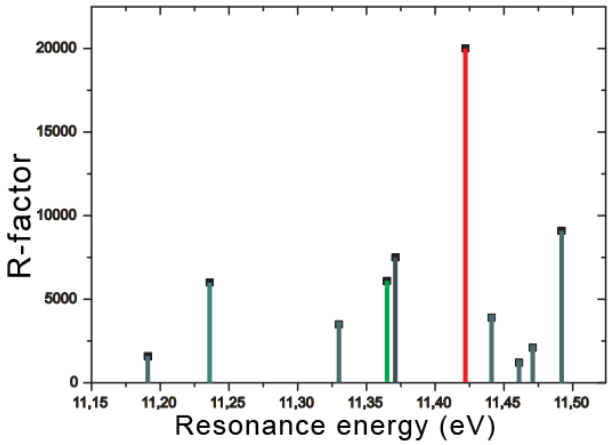

Fig. 4. Resonance picture taking into account fragmentation of 7 p-levels according to [20]; R-factor is given in relative intensity units.

The diagram in Fig. 2 interferes with the diagram in Fig. 3a from [14], in which the order of interaction of the $7s$-electron with the nucleus and the beam is reversed and the intermediate state $7s$ is replaced by $8p$. The Feynman diagram thus obtained is shown in Fig. 3. Qualitatively, it carries the same physical meaning [22]. A laser photon with frequency ω is absorbed by $7s$-electron, which goes into a virtual state. Near the resonance, the main contribution is made by $8p$-electron. It transfers a part of the received energy to the nucleus, transferring it to the isomeric state. The electron remains in the excited state $7p$ with energy ε_{7p}. The resonance condition is still given by formula (3). The acceleration factor can be calculated by the formula [14], similar to (4):

$$R = \frac{\alpha_d(M1; 8p-7p)\Gamma_\gamma^{(a)}(\omega; 7s-8p)}{2\pi(\omega-\varepsilon_{8p})^2}. \qquad (6)$$

Summarizing this section, we recap that the gain in cross section can be used to reduce the scanning time of the first technique by R times. Spectral beam broadening can also reduce the scanning time by Γ_a/Γ_n times compared to the second scheme.

4. CALCULATION RESULTS

At the time of publication of [14], another value of the isomer energy was considered the most probable: 3.5 eV. Therefore, the calculations were performed with this value. The present calculations were performed within the framework of the Dirac-Fock method using the RAINE utility suite for the calculation of atomic structures [23]. As a result, the following values were obtained:

$$\alpha_d(\omega_n; 7s-7s) = 1.95\cdot 10^{10}\,\text{eV},$$

The ICC for $M1$-transition with energy $\omega_n = 8.338$ eV in Th I is equal to $\alpha(M1) = 0.987\cdot 10^9$. If we put the lifetime in neutral atoms 10 μs [15], then the intrinsic width of the isomer follows from here

$$\Gamma_n = 0.667\cdot 10^{-19}\,\text{eV}.$$

Within the framework of the method, the resonance cross section consists of two components corresponding to the final levels $7p_{1/2}$ and $7p_{3/2}$, with the intensity of the second component twice as high as the first. For the energy of the level $7p_{3/2}$ the value 3.52 eV is obtained, hence $\omega = 11.86$ eV. The energy of the level $7p_{1/2}$ is obtained 2.49 eV, hence $\omega = 10.83$ eV for this level. Further, the radiation width of the atomic transition

$$\Gamma_\gamma^{(a)}(\omega; 7s-7p) = 3.30\cdot 10^{-6}\,\text{eV},$$

which is thirteen orders of magnitude larger than the natural width of the isomeric line Γ_n. Substituting these values into formula (4), we find the value of the acceleration factor $R = 147$.

The contribution shown in the diagram in Fig. 3 in [14] was 20 times smaller. It becomes relatively even smaller with the present value of the isomer energy. Therefore, we can limit ourselves to considering the diagram in Fig. 2.

Interelectron interaction leads to fragmentation of atomic levels and the strength of transitions between them. Considering that the cross section of the process is proportional to the intensity of the corresponding transition line from the ground state $7s$ to the excited state $7p$ in accordance with formula (4), we can estimate the relative intensities of the corresponding components in the cross section of resonant photoexcitation. For this purpose, we use the experimental relative intensities of the atomic lines of the absorption spectrum of Th II given in [20]. According to [20], a low-fragmentation component corresponding to the transition from the ground state to the $7s7p6d$ level with $j = 5/2$ and 3.08398 eV energy is highlighted. The relative estimates for the strongest lines obtained in this way are presented in Fig. 4. As can be seen, in addition to the main component with energy 11.422 eV, there are intense satellites with energies 11.371, 11.365, 11.236 eV and others. They can be used as benchmarks, the detection of which will help to reliably identify the energies of these components in the experiment and, accordingly, to determine the isomer energy according to (3) with an accuracy corresponding to laser measurement methods.

5. DISCUSSION OF THE RESULTS

Let us note the peculiarities of the resonance excitation of the nucleus according to Fig. 2, 3 in comparison with the photon absorption by a bare nucleus. The cross section by the resonance mechanism, according to the calculation, was amplified approximately by a factor of $R \approx 150$. This shows the dynamical properties of the electron shell as a resonator.

The most important difference for the present work is the width of the resonance. If for a bare nucleus this width is given by the intrinsic half-width (in the absence of IC) of the isomeric level of the nucleus Γ_n, then by the mechanism presented in Fig. 2, 3, it is equal to the sum of all widths – electronic and nuclear. Usually, as in this case, the sum of the widths of the intermediate $8p$- and final $7p$-states of the excited atom dominates. Let us denote it as Γ_a. There is a correlation $\Gamma_a \ggg \Gamma_n$, so scanning by the mechanism shown in Fig. 2 requires Γ_a/Γ_n times less time.

For effective realization of this mechanism it is extremely important that the final atomic level, in this case $7p$, is not the ground state, but has a typical atomic width. If instead of $7p$-level the atom returns to the ground $7s$-state, the resonance would have a nuclear width Γ_n due to the absence of atomic widths as a consequence of the law of conservation of energy. The amplification factor $R = 150$ could be preserved in the case of a monochromatic laser beam of photons

with frequency $\omega = \omega_n$ and spectral width $\Gamma_\omega \approx \Gamma_n$, but the most important advantage of optical pumping of an isomer by a beam with spectral width of a typical atomic line would be lost.

In particular, this refers to the two-photon scheme of excitation of the isomer proposed in [24], in which the energy of two consecutive photons absorbed by the electron shell is completely transferred to the nucleus. At the same time, the electron shell returns to the ground state. In view of what has been said above, this scheme does not seem viable at present: the shell must remain in the excited state. Other shortcomings of [24] are discussed in [25, 26]. In due course, they led to a paradox called the "thorium enigma". The arguments given above summarize its solution.

6. CONCLUSIONS

We are on the threshold of the birth of new laser-nucleus technologies. They will be based on resonant interaction of light beams with nuclei: resonant absorption and scattering, elastic and inelastic, nonlinear effects such as generation of higher harmonics and others. To realize these manipulations with bare nuclei, it is necessary to use spectroscopically narrow almost monochromatic beams with spectral width in the range of the nuclear line width. The development of such technologies requires knowledge of the energies of nuclear transitions and isomers. It is such light beams that will form the basis of future nuclear optical clocks.

At the same time, it is possible to use the electron shell as an effective resonator that enhances the effect of light on the nucleus. The tool for constructing such a resonator is internal and resonant conversion. The above examples further demonstrate how effectively resonance can be used to optically pump an isomeric state of ^{229m}Th. Resonance can be used in two dimensions: in terms of gain and to reduce the requirements for the degree of monochromaticity of the light beam. In the above example with optical pumping of ThI, a gain of nine orders of magnitude in line broadening due to IC can be obtained. In ThII ions it is possible to realize resonance, which will give an effective gain both in the interaction strength – by a factor of 150, and in the broadening of the resonance line by two or three orders of magnitude as compared to IC in neutral atoms.

ACKNOWLEDGEMENTS

One of the authors (FFK) would like to thank L. von der Venza, L. F. Vitushkin and P. Tirolf for fruitful discussions.

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