Quasiclassical theory of ^{229m}Th excitation by laser pulses via electron bridges

A. Ya. Dzyublik*

Institute for Nuclear Research, National Academy of Sciences of Ukraine, avenue Nauki 47, 03680 Kyiv, Ukraine and National Research Nuclear University MEPhI, Kashirskoe shosse 31, 115409 Moscow, Russian Federation

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In the quasiclassical approximation it is studied excitation of the 8.2 eV isomer ^{229m}Th in the electron bridges (EB), initiated by a laser pulse. While the nucleus and atomic electron are treated quantum mechanically, the laser pulse is described by a time-dependent wave packet, formed by classic electromagnetic waves. Two types of the EB are taken into consideration: via the continuous electron spectrum and via discrete atomic levels. In calculations of the excitation probability of ^{229m}Th by a laser pulse I employ the generalized formalism of Floquet functions, which allows one to solve the time-dependent Schrödinger equation by the methods of stationary scattering theory. The task is solved for weak lasers as well as for the conventional lasers of arbitrary intensity. In the first case the results well correlate with those obtained previously when treating the laser pulse as a bunch of uncorrelated photons. If conventional lasers generate EB through an atomic level, its population suffers Rabi oscillations. Therefore application of the π pulses may strongly enhance the effect of the isomer excitation. For the same EB the excitation probability of the isomer as a function of the laser carrier frequency is shown to have two peaks associated with two resonant levels.

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I. INTRODUCTION

The nucleus ^{229}Th has uniquely small energy E_{is} of the first isomeric $3/2^+$ excited state, labeled as ^{229m}Th. The most reliable measurements of E_{is} have been done recently in the works [1,2]. By measuring the energies of the internal conversion electrons, emitted by a thorium atom, and taking the ionization energy 6.308 ± 0.003 eV [3] into account, Seiferle *et al.* [1] found that $E_{is} = 8.28 \pm 0.17$ eV. It is worth to note that the isomer ^{229m}Th in neutral atom predominantly decays through the internal conversion (IC) channel (the conversion coefficient $\alpha \approx 10^9$ [1]). On the other hand, Sikorsky *et al.* [2] with the aid of magnetic microcalorimeters measured the energies of γ transitions from higher-lying levels to the ground state and to the isomeric level. By subtracting them the authors obtained close value of the energy: $E_{is} = 8.10 \pm 0.17$ eV.

Both in these experiments and in majority of previous ones (see the review [4]) the level ^{229m}Th was pumped in the α decay of ^{233}U via the second 29.2 keV level. A more straightforward optical experiment had been done by Jeet *et al.* [5], who realized direct excitation of the isomer ^{229m}Th by synchrotron radiation and observed its spontaneous decay. Synchrotron radiation has been used also by Masuda *et al.* [6] for excitation of the second 29.2 keV level with subsequent population of the isomer. A complete theory for direct excitation of the ^{229m}Th by lasers was provided in Refs. [7,8]. Rather weak two-photon direct excitation of ^{229m}Th was carefully analyzed by Romanenko *et al.* [9]. An alternative effective method of the isomer population by electron current has been regarded by Tkalya [10]. Moreover, Wang *et al.* [11] proposed to employ for this aim a laser-driven electron recollision process, taking place in the field of superstrong lasers.

Great interest to the isomer ^{229m}Th is dictated by a realistic possibility to create new etalon of frequency and time (so-called nuclear clock) [12–15]. Whereas modern atomic optical clocks have relative frequency uncertainty $\Delta\omega/\omega$ up to 10^{-18} [16], in the nuclear clock it can be of the order of 10^{-19} [15]. In addition, the nuclear clock is more robust against external influences than the optical ones [14,15].

Such precision clocks are needed for geodesy and astronomy as well as for GPS navigation, sensors of the gravitation field potential, measurements of time variations of the Earth's gravitation [17]. Furthermore, they will enable the search for possible time variations of the fundamental constants of physics [18–24] and will be useful in solution of the dark matter puzzles [25]. Besides, features of the nuclear γ laser based on deexcitation of ^{229m}Th were analyzed in the papers [26,27].

Smallness of the ^{229m}Th energy gave rise to idea of using electron bridges (EB) for excitation of this isomer. In a number of theoretical papers [28–34] the following scheme of EB was discussed. First, an electron of the thorium ion is excited by lasers into an upper-lying atomic level. Then it passes down to any vacant level transferring its energy to the nucleus. This process represents the well-known nuclear excitation at electron transition (NEET), which was studied theoretically in Refs. [35–41] and observed for the nuclei ^{197}Au [42], ^{189}Os [43], and ^{237}Np [44]. Rather large mismatch of the nuclear and atomic transitions leads to small probability of the NEET. In best case of ^{197}Au it equals 5×10^{-8} [42]. The same difficulty with the resonance detuning exists also

*dzyublik@ukr.net

in the EB via bound electronic levels of the thorium atom. Besides, a contribution of EB into the lifetime of ^{229m}Th has been studied in Refs. [45,46].

In order to avoid the problem of mismatch of the atomic and nuclear transitions Borisyuk *et al.* [47] proposed to populate ^{229m}Th through the electron continuum. The S matrix for such EB transition was always calculated in the second or third order of the perturbation theory by using prescriptions of the quantum electrodynamics. It was introduced then the complex energy $E_{\rm is} - i\Gamma_{\rm is}/2$ for the isomeric state, where $\Gamma_{\rm is}$ is the level width. As a consequence, the corresponding wave function $\Psi_{\rm is}(\mathbf{r})e^{-i(E_{\rm is}-i\Gamma_{\rm is}/2)t/\hbar}$ diverges as $t \to -\infty$. For removal of these divergences the integration over time in the chronologically ordered series for the S matrix was only performed at $t > 0$, although by definition it should be done on the whole time axis [48]. More correct approach is ensured by the collision theory [49,50], where the transition matrix incorporates all the orders of the perturbation operator. Therefore in my previous paper [51] the excitation cross section of the isomer by lasers via EB has been calculated in the framework of the scattering theory.

In all mentioned above theories the laser radiation was regarded as a bunch of uncorrelated photons. Namely, it was first calculated the excitation cross section of ^{229m}Th by single photon and then it was averaged over the frequency distribution of the laser pulse. However, the laser pulse of correlated photons is more adequately described by the classical electromagnetic wave packet, which explicitly depends on time (see, e.g., Refs. [52,53]). Evolution of the quantum system (^{229}Th + electrons) in the field of classical wave is described by the time-dependent Shrödinger equation. At the same time, by applying the approach, developed in Refs. [54–56], one can solve the Shrödinger equation by the methods of the stationary scattering theory. Below I use this formalism in order to calculate the excitation probability of ^{229m}Th generated by laser pulses. The EB are considered via both electron continuum and discrete atomic levels.

II. ELECTRON BRIDGE

The Hamiltonian of the whole system (^{229}Th + atomic electron + quantized electromagnetic field) is written as

$$H(t) = H_0 + V_r + V_f(t), \tag{1}$$

where the unperturbed Hamiltonian H_0 is represented by a sum of the Hamiltonians for the nucleus H_n, electron H_e and the quantized electromagnetic field $H_{\rm rad}$, the perturbation $V_r = V_r^n + V_r^e$ is responsible for interaction of the nucleus and electron with the quantized electromagnetic field, while $V_f(t)$ represents the interaction of the atomic electron with the classical laser field. The operator H_e is a sum of the kinetic energy operator of the electron and the screened Coulomb field of the nucleus $V_{\rm C}(r)$. The perturbation

$$V_r^{n(e)} = -\frac{1}{c}\int \mathbf{J}_{n(e)}(\mathbf{r})A(\mathbf{r})d\mathbf{r}, \tag{2}$$

where $\mathbf{J}_n(\mathbf{r})$ and $\mathbf{J}_e(\mathbf{r})$ are the electric current density operators for the nucleus and electron, respectively, $A(\mathbf{r})$ the vector potential operator of the quantized field. The interaction with the laser field $V_f(\mathbf{r}, t)$ is determined by the same formula but with the vector potential $\mathbf{A}(\mathbf{r}, t)$ of the classical electromagnetic wave packet instead of the operator $A(\mathbf{r})$. For a linearly polarized wave

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}_0(t)\sin\left(\mathbf{k}_0\mathbf{r} - \omega_L^0 t\right). \tag{3}$$

where $k_0 = \omega_L^0/c$. Usually $\mathbf{A}(\mathbf{r}, t)$ is replaced by the electric field strength

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0(t)\cos\left(\mathbf{k}_0\mathbf{r} - \omega_L^0 t\right), \tag{4}$$

where $\mathbf{E}_0(t) = k_0\mathbf{A}_0(t)$.

I take the envelope in the form

$$E_0(t) = E_0\exp(-\Gamma_L t/2\hbar)\theta(t), \tag{5}$$

where $\theta(t)$ is the Heaviside step function:

$$\theta(x) = \begin{cases} 1, & x > 0, \\ 0, & x < 0. \end{cases} \tag{6}$$

The duration of such laser pulse will be $\tau_L = \hbar/\Gamma_L$ and the frequency distribution of its photons be determined by the Lorentzian curve centered at ω_L^0 and having the width Γ_L.

The interaction $V_f(t)$ depends on the Fourier transform of the electron current density

$$J_\lambda^e(\mathbf{k}) = \int d\mathbf{r}\mathbf{J}_e(\mathbf{r})e^{i\mathbf{k}\mathbf{r}}\cdot\mathbf{e}_\lambda, \tag{7}$$

projected on the circular polarization vector $\mathbf{e}_\lambda$. In the long-wave approximation [57]

$$\frac{1}{c}J_\lambda^e(\mathbf{k}) = -\sqrt{2\pi}\sum_{j=1}^{\infty} i^j k^j \sqrt{\frac{(j+1)(2j+1)}{j}}\frac{1}{(2j+1)!!} \times \sum_{\mu=-j}^{j} D^j_{\mu\lambda}(\alpha,\beta,0)\left[\lambda\mathfrak{M}^e_\mu(Mj) + i\mathfrak{M}^e_\mu(Ej)\right], \tag{8}$$

where $\mathfrak{M}^e_\mu(Ej)$ and $\mathfrak{M}^e_\mu(Mj)$ are, respectively, the electric and magnetic multipole operators of the electron, $D^j_{\mu\lambda}(\alpha,\beta,0)$ is the rotation matrix, depending on the spherical angles β, α of the photon wave vector $\mathbf{k}$.

The electron bridge considered here is determined by the dipole electromagnetic transitions, for which in the long-wave approximation

$$\mathfrak{M}^e_\mu(E1) = -erY_{1\mu}(\hat{\mathbf{r}}) \tag{9}$$

and

$$\mathfrak{M}^e_\mu(M1) = -\frac{k^2}{2c}\int \mathbf{J}_e(\mathbf{r})\cdot\mathbf{l}\left(h_1^{(1)}(kr)Y_{1\mu}(\hat{\mathbf{r}})\right)d\mathbf{r}, \tag{10}$$

where $\hat{\mathbf{r}}$ denotes spherical angles of the electron radius vector $\mathbf{r}$ and $h_l^{(1)}(x)$ the spherical Hankel function of the first kind.

Using the expansion (8) one can easily show that the $E1$ transitions, generated by a linearly polarized wave, are provided by the operator

$$V_f^e(t) = -\sqrt{\frac{2\pi}{3}}\sum_{\mu=-1}^{1}\left(D^1_{\mu 1}(\alpha\beta 0) + D^1_{\mu,-1}(\alpha\beta 0)\right) \times \mathfrak{M}^e_\mu(E1)E_0(t)\cos\omega_L^0 t. \tag{11}$$

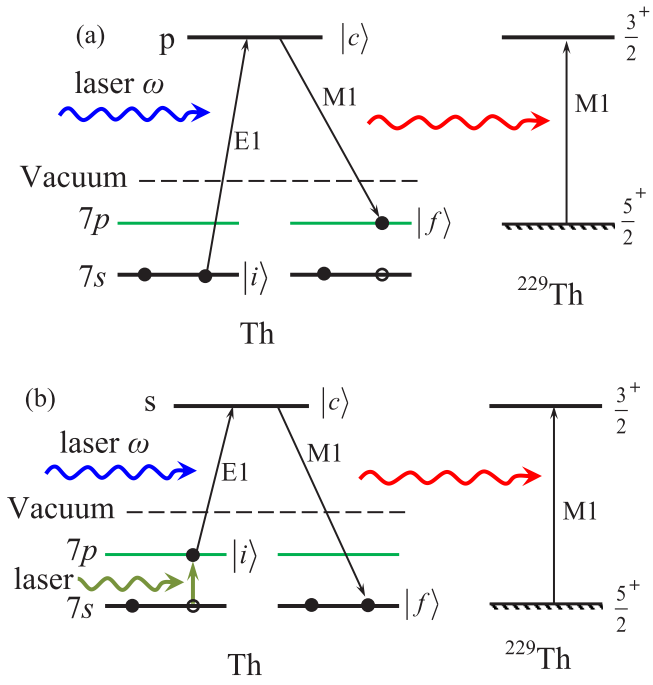

FIG. 1. Sketch of the EB via continuum, induced by laser, with excitation of the isomer ^{229m}Th. The continuum lies above the dashed line (vacuum). (a) EB begins from the upper 7s level of a thorium atom and ends in the excited 7p one. (b) EB begins from the level 7p, excited by the second laser, and ends when electron is captured by the atom into the 7s vacancy. In both cases ionization of the atom proceeds in $E1$ transitions. Resonant excitation of the nucleus happens at the reverse electron transitions of the same type and multipolarity as the nuclear $M1$ transition.

Following Refs. [47,51] I will first consider here the EB shown in Fig. 1. In case (a) the atomic electron from the upper 7s state absorbs the laser photon and performs $E1$ transition to the p state of the continuum, where it carries the orbital angular momentum $l = 1$. In the alternative case (b) the initial state is the 7p atomic level, excited by an additional laser, the second laser in $E1$ transition creates a photoelectron in the s state with $l = 0$. In both events the electron, coming back to the thorium atom, excites the isomer.

The wave function of the initial state

$$|a\rangle = |I_g M_g\rangle \phi_{j_i m_i}(\mathbf{r}) \tag{12}$$

describes the nucleus in the ground state $|I_g M_g\rangle$ with spin I_g and its projection on the quantization axis M_g, as well as the electron with the total angular momentum j_i and its projection m_i. The corresponding energy E_a equals the electron energy ϵ_i in the initial state $|j_i m_i\rangle$. Hereafter the vacuum wave function $|0\rangle$ of the quantized electromagnetic field is omitted for brevity.

Having absorbed a laser photon, the electron flies away from the atom with the wave vector $\boldsymbol{\kappa}$ and spin projection ν. Then the first intermediate state of the whole system is described by

$$|c_1\rangle = |I_g M_g\rangle \psi^+_{\boldsymbol{\kappa}\nu}(\mathbf{r}). \tag{13}$$

Due to rotational invariance of the Hamiltonian H it is convenient to write down $\psi^+_{\boldsymbol{\kappa}\nu}(\mathbf{r})$ as an expansion [51]

$$\psi^+_{\boldsymbol{\kappa}\nu}(\mathbf{r}) = \sum_{l=0}^{\infty} \sum_{j_c m_c} \mathfrak{Y}^{m_c^*}_{j_c}(lsv;\hat{\boldsymbol{\kappa}}) \phi_{j_c l m_c}(\mathbf{r}) \tag{14}$$

in terms of the functions $\phi_{j_c l m_c}(\mathbf{r})$, which describe free electron with the orbital momentum l, total angular momentum j_c and its projection m_c.

The expansion coefficients $\mathfrak{Y}^{m_c^*}_{j_c}(lsv;\hat{\boldsymbol{\kappa}})$ depend on the spherical angles $\hat{\boldsymbol{\kappa}}$ of the wave vector $\boldsymbol{\kappa}$. They satisfy the following orthogonality relations [51]:

$$\sum_{\nu} \int d\Omega_{\boldsymbol{\kappa}} \mathfrak{Y}^{m_c' *}_{j_c'}(l'sv;\hat{\boldsymbol{\kappa}}) \mathfrak{Y}^{m_c}_{j_c}(lsv;\hat{\boldsymbol{\kappa}}) = \delta_{ll'} \delta_{j_c j_c'} \delta_{m_c m_c'}. \tag{15}$$

There is some probability that the photoelectron comes back from the continuous spectrum to the atomic empty level $|j_f m_f\rangle$ and transfers its energy by a virtual $M1$ photon to the nucleus. As a result, the system occurs in the second intermediate state

$$|c_2\rangle = |I_e M_e\rangle |j_f m_f\rangle, \tag{16}$$

where $|I_e M_e\rangle$ is the wave function of the isomer ^{229m}Th. Such a transition from $|c_1\rangle$ to $|c_2\rangle$ is nothing but the nuclear excitation by electron capture (NEEC) from continuous spectrum to an atomic vacancy (see, e.g., Ref. [58] and references therein).

The corresponding eigenvalues of H_0, associated with these intermediate states, are

$$E_{c_1} = \epsilon_c(\kappa), \quad E_{c_2} = E_{\rm is} + \epsilon_f, \tag{17}$$

where $\epsilon_c(\kappa) = \hbar^2\kappa^2/2m$ is the energy of the photoelectron, ϵ_f the final energy of this electron, being captured by the vacancy specified by quantum numbers $j_f m_f$.

The isomer decays mainly through the conversion channel (the conversion coefficient $\alpha = 10^9$). Then the final state $|b\rangle$ of the whole system will have the energy $E_b = \epsilon_f + \epsilon'(\kappa') + \epsilon_v$, where $\epsilon'(\kappa')$ stands for the energy of the emitted conversion electron and ϵ_v is the energy of the vacancy left in the atomic shell. Note that $\epsilon_v > 0$, whereas the energy of an electron to occupy this vacancy equals $-\epsilon_v$.

III. FORMALISM OF FLOQUET FUNCTIONS

The wave function of any quantum system is determined by the time-dependent Schrödinger equation

$$i\hbar \frac{\partial \Psi(\mathbf{r},t)}{\partial t} = H(t)\Psi(\mathbf{r},t), \tag{18}$$

where $\mathbf{r}$ denotes a set of coordinates of the system. If the Hamiltonian is a periodic function of time $H(t) = H(t+T)$, then Eq. (18) has the well-known Floquet solutions

$$\Psi_b(\mathbf{r},t) = \psi_{b,n}(\mathbf{r},t) e^{-i\mathscr{E}_{b,n}t/\hbar} \tag{19}$$

with the periodical functions $\psi_{b,n}(\mathbf{r},t) = \psi_{b,n}(\mathbf{r},t+T)$ and quasienergies $\mathscr{E}_{b,n}$. They form an equidistant spectrum:

$$\mathscr{E}_{b,n} = \mathscr{E}_b + n\hbar\omega, \quad n = \pm 1, \ \pm 2, \ldots, \tag{20}$$

where $\omega = 2\pi/T$ is the circular frequency, while the corresponding factors $\psi_{bn}(\mathbf{r},t)$ are

$$\psi_{b,n}(\mathbf{r},t) = \psi_b(\mathbf{r},t)e^{in\omega t}. \tag{21}$$

It is convenient to deal with such quasienergetic states in the composite Hilbert space of square-integrable periodical functions $\psi(\mathbf{r},t) = \psi(\mathbf{r},t+T)$, depending on both the spatial coordinates $\mathbf{r}$ and time t. In this space the scalar product of the functions $u(\mathbf{r},t)$ and $v(\mathbf{r},t)$ is determined by [54]

$$\langle\langle u(\mathbf{r},t)|v(\mathbf{r},t)\rangle\rangle = \int_{-T/2}^{T/2} dt \int d\mathbf{r} u^*(\mathbf{r},t)v(\mathbf{r},t). \tag{22}$$

In addition, Sambe [54] proposed to use the Floquet operators

$$\mathscr{H}_0(t) = H_0 - i\hbar\frac{\partial}{\partial t}, \quad \mathscr{H}(t) = H(t) - i\hbar\frac{\partial}{\partial t} \tag{23}$$

instead of the Hamiltonians H_0 and $H(t)$.

As a result, Eq. (18) takes the form of the eigenvalue equation

$$\mathscr{H}(t)u_{b,n}(\mathbf{r},t) = \mathscr{E}_{b,n}u_{b,n}(\mathbf{r},t). \tag{24}$$

It resembles the stationary Schrödinger equation, which enables one to employ formally all the methods of the stationary perturbation theory [54] as well as of the scattering theory [55,56].

When building the perturbation theory, Sambe [54] used the eigenfunctions and eigenvalues of the unperturbed Hamiltonian H_0, not depending on time,

$$H_0\varphi_b(\mathbf{r}) = E_b\varphi_b(\mathbf{r}). \tag{25}$$

Respectively, the products

$$u_{bn}^{(0)}(\mathbf{r},t) = \varphi_b(\mathbf{r})\frac{1}{\sqrt{T}}e^{in\omega t} \tag{26}$$

are the eigenfunctions of the Floquet operator $\mathscr{H}_0$,

$$\mathscr{H}_0 u_{bn}^{(0)}(\mathbf{r},t) = \mathscr{E}_{bn}^{(0)}u_{bn}^{(0)}(\mathbf{r},t), \tag{27}$$

with unperturbed quasienergies

$$\mathscr{E}_{bn}^{(0)} = E_b + n\hbar\omega. \tag{28}$$

The functions $u_{bn}^{(0)}$ form a complete orthonormal basis set in the composite Hilbert space:

$$\left\langle\left\langle u_{b'n'}^{(0)}\middle|u_{bn}^{(0)}\right\rangle\right\rangle = \delta_{b'b}\delta_{n'n}. \tag{29}$$

However, all this is valid for the periodical perturbation, but not for short laser pulses. Therefore I apply here the results of the paper [56], where the formalism of Sambe was generalized to the case of arbitrary perturbations by imposing periodic boundary conditions $\Psi(\mathbf{r},t) = \Psi(\mathbf{r},t+T)$. The scalar product is determined now by the same equation (22) but with the period $T \to \infty$. In this limiting case the spectrum of quasienergies becomes continuous with $n\omega$ replaced by ω:

$$\mathscr{E}_{b,\omega}^{(0)} = E_b + \hbar\omega. \tag{30}$$

The corresponding quasienergetic functions take the form [56]

$$u_{b,\omega}^{(0)}(\mathbf{r},t) = \varphi_b(\mathbf{r})\frac{1}{\sqrt{2\pi}}e^{i\omega t}. \tag{31}$$

This procedure is completely equivalent to transition from a Fourier series to a Fourier integral.

In the composite Hilbert space any transition is determined by the operator [56]

$$\mathscr{T}(t) = V + V\mathscr{G}^+(\mathscr{E}_a)V, \tag{32}$$

where $V = V(t)$ represents a perturbation, $\mathscr{G}^+(z)$ is the time-dependent Green's operator:

$$\mathscr{G}^+(z) = [z + i\eta - \mathscr{H}(t)]^{-1}, \qquad \eta \to +0. \tag{33}$$

Then the transition matrix between the states $u_{a,0}(\mathbf{r},t)$ and $u_{b,\omega}(\mathbf{r},t)$ will be written as [56]

$$\mathscr{T}_{ba}(\omega) = V_{ba}(\omega) + \sum_{c',c}\int_{-\infty}^{\infty} d\omega'' \times \int_{-\infty}^{\infty} d\omega' V_{bc'}(\omega-\omega'')\mathscr{G}^+_{c',\omega'';c,\omega'}(\mathscr{E}_a)V_{ca}(\omega'), \tag{34}$$

where the following notations are used:

$$\mathscr{G}^+_{c',\omega'';c,\omega'} = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt e^{-i\omega''t}\langle c'|\mathscr{G}^+|c\rangle e^{i\omega' t}, \tag{35}$$

$$\mathscr{T}_{ba}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dt e^{-i\omega t}\langle b|\mathscr{T}(t)|a\rangle, \tag{36}$$

and $V_{bb'}(\omega)$ are analogous Fourier transforms of the functions $V_{bb'}(t)$.

The Green's matrix $\mathscr{G}^+_{c',\omega'';c,\omega'}(\mathscr{E}_a)$ is determined by the system of equations [56]

$$(\mathscr{E}_a - \mathscr{E}_{c',\omega'} + i\Gamma_{c'}/2)\mathscr{G}^+_{c',\omega';c,\omega}(\mathscr{E}_a) = \delta_{c'c}\delta(\omega'-\omega) + \sum_{c''}\int_{-\infty}^{\infty} d\omega'' V^f_{c'c''}(\omega'-\omega'')\mathscr{G}^+_{c'',\omega'';c,\omega}(\mathscr{E}_a). \tag{37}$$

The S matrix is related to transition matrix by [56]

$$S_{ba} = \delta_{ba} - \frac{2\pi i}{\hbar}\mathscr{T}_{ba}(\omega_{ab}), \tag{38}$$

where the frequency $\omega_{ab} = (E_a - E_b)/\hbar$. This S matrix determines a probability that system at the moment $t \to \infty$ occurs in the final state $|b\rangle$:

$$P_{ba} = |S_{ba}|^2. \tag{39}$$

IV. EXCITATION PROBABILITY OF ^{229m}Th

Here I will consider the case of weak laser pulses triggering the excitation process. Then the interaction $V_f(t)$ is only significant at the initial stage of the EB and can be omitted in the Green's operator:

$$\mathscr{G}^+(\mathscr{E}_a) \approx (\mathscr{E}_a + i\eta - \mathscr{H}_0(t) - V_r)^{-1}. \tag{40}$$

Then the quasienergies of the intermediate and final states for EB are

$$\mathscr{E}_{c_1,\omega} = E_{c_1} + \hbar\omega, \quad \mathscr{E}_{c_2,\omega} = E_{c_2} + \hbar\omega, \quad \mathscr{E}_{b,\omega} = E_b + \hbar\omega \tag{41}$$

and the corresponding Green's function takes the form

$$\mathscr{G}^{+}_{c_2,\omega';c_1,\omega}(\mathscr{E}_a) = G^{+}_{c_2c_1}(\mathscr{E}_a - \hbar\omega)\delta(\omega' - \omega). \tag{42}$$

Here $G^{+}_{c_2c_1}(z)$ is the matrix for the time-independent Green's operator, taking place in the conventional collision theory [49,50]:

$$G^{+}(z) = (z + i\eta - H_0 - V_r)^{-1}. \tag{43}$$

In this case the matrix of the transition operator (32) takes the form

$$\mathscr{T}_{ba}(\omega) = \sum_{c_2,c_1} V'_{bc_2} G^{+}_{c_2c_1}(\mathscr{E}_a - \hbar\omega) V^{f}_{c_1a}(\omega), \tag{44}$$

where V' determines decay of the intermediate level $|c_2\rangle$ (decay of the isomer ^{229m}Th as well as possible decay of the atomic state $|j_f m_f\rangle$).

A. EB via continuum

In the case of EB via continuum the summation over c_1 in Eq. (44) includes integration over the wave vector of the photoelectron $\boldsymbol{\kappa}$ at infinity as well as summation over its spin projections $\nu = \pm 1/2$. The summation c_2 is carried over the magnetic quantum numbers of the nucleus M_e and electron m_f.

There are two overlapping resonant quasilevels $|c_1, \omega\rangle\rangle$ and $|c_2, \omega\rangle\rangle$, having the widths Γ_1 and Γ_2. The first width

$$\Gamma_1 = \Gamma_{\mathrm{RR}} + \Gamma_{\mathrm{DR}} + \Gamma_{\mathrm{EB}}, \tag{45}$$

where Γ_{RR} and Γ_{DR} are standard widths for the radiative and dielectronic recombinations, respectively. In the case of EB there appears an additional width Γ_{EB}, responsible for the electron capture from continuous spectrum to any vacant atomic level, followed by emission of the virtual photon to be absorbed by the nucleus (NEEC).

The second width is a sum of the widths for decay of the final electron level Γ_f and the isomer Γ_{is},

$$\Gamma_2 = \Gamma_f + \Gamma_{\mathrm{is}}. \tag{46}$$

When the atomic state $|f\rangle$ is stable $\Gamma_2 = \Gamma_{\mathrm{is}}$, otherwise $\Gamma_2 \approx \Gamma_f$.

The off-diagonal Green's function is given by [56]

$$G^{+}_{c_2,\omega;c_1,\omega} = \frac{R^{+}_{c_2c_1}}{\left(E_a - \mathscr{E}_{c_1} + i\frac{\Gamma_1}{2}\right)\left(E_a - \mathscr{E}_{c_2} + i\frac{\Gamma_2}{2}\right)}, \tag{47}$$

where the matrix

$$R^{+}_{c_2c_1} = \sum_{b\neq c_2,c_1} \frac{\langle c_2|V_r|b\rangle\langle b|V_r|c_1\rangle}{E_a + i\eta - E_b} \tag{48}$$

defines the NEEC transition.

Substitution of (41) and (47) into (44) results in

$$\mathscr{T}_{ba}(\omega) = \sum_{c_2c_1} \frac{V'_{bc_2} R^{+}_{c_2c_1} V^{f}_{c_1a}(\omega)}{\left(\epsilon_i - \epsilon_c(\kappa) - \hbar\omega + i\frac{\Gamma_1}{2}\right)\left(\epsilon_i - E_{\mathrm{is}} - \epsilon_f - \hbar\omega + i\frac{\Gamma_2}{2}\right)}. \tag{49}$$

Replacing here ω by

$$\omega_{ab} = (\epsilon_i - \epsilon_f - \epsilon'(\kappa') - \epsilon_v)/\hbar, \tag{50}$$

one gets the transition matrix for EB shown in Fig. 1:

$$\mathscr{T}_{ba}(\omega_{ab}) = -\sum_{c_2c_1} \frac{V'_{bc_2} R^{+}_{c_2c_1} V^{f}_{c_1a}(\omega_{ab})}{\left(\epsilon_c(\kappa) - \epsilon_0(\kappa') - i\frac{\Gamma_1}{2}\right)\left(\epsilon'(\kappa') - \epsilon'_0 + i\frac{\Gamma_2}{2}\right)}, \tag{51}$$

where

$$\epsilon_0(\kappa') = \epsilon_f + \epsilon'(\kappa') + \epsilon_v \tag{52}$$

and

$$\epsilon'_0 = E_{\mathrm{is}} - \epsilon_v \tag{53}$$

are the resonance energies of the photoelectron and conversion electron, respectively. Notice that summation over the states c_1 includes integration over the wave vector of photoelectron $\boldsymbol{\kappa}$ and summation over projections of its spin ν.

The EB probability generated by single laser pulse P_{EB} is obtained by summing the transition probability P_{ba} over all final states $|b\rangle$ and averaging over the initial states $|a\rangle$:

$$P_{\mathrm{EB}} = \frac{1}{2I_g + 1} \sum_{M_g M'_g} \frac{1}{2j_i + 1} \sum_{m_f m_i} \sum_{\nu'} \int d\boldsymbol{\kappa}' P_{ba}, \tag{54}$$

where ν' means spin projection of the conversion electron. Besides, the probability P_{ba} with $b \neq a$ according to (38) and (39) is given by

$$P_{ba} = \left(\frac{2\pi}{\hbar}\right)^2 |\mathscr{T}_{ba}(\omega_{ab})|^2. \tag{55}$$

Having inserted here $\mathscr{T}$ matrix (51), I integrate first over the wave vector of the photoelectron $\boldsymbol{\kappa}$ by taking into account the orthogonality relation (15) and approximating the factor $(\Gamma_1/2\pi)[(\epsilon_c(\kappa) - \epsilon_0(\kappa'))^2 + (\Gamma_1/2)^2]^{-1}$ by the delta function $\delta(\epsilon_c(\kappa) - \epsilon_0(\kappa'))$.

Integrating further over $\boldsymbol{\kappa}'$ and repeating calculations of Ref. [51], I look for the EB probability $P_{\mathrm{EB}}(\delta)$ as a function of the detuning

$$\delta = \omega_L^{\mathrm{res}} - \omega_L^0 \tag{56}$$

of the central laser frequency ω_L^0 from the resonance value

$$\omega_L^{\mathrm{res}} = (E_{\mathrm{is}} + \epsilon_f - \epsilon_i)/\hbar. \tag{57}$$

The result is

$$P_{\mathrm{EB}}(\delta) = \sum_{j_c} P^{j_c}_{\mathrm{EB}}(\delta), \tag{58}$$

where $j_c = 1/2$ and $3/2$ for the case (a), while $j_c = 1/2$ for (b) (see Fig. 1) and the partial excitation probability

$$P^{j_c}_{\mathrm{EB}}(\delta) = g_{j_c} \frac{\Gamma^{\mathrm{IC}}_{\mathrm{is}}(f)^{j_c}}{\Gamma} \mathcal{L}(\delta) P^{j_c}_{\mathrm{ion}}(\omega_L^{\mathrm{res}}) \tag{59}$$

Here $\Gamma = \Gamma_L + \Gamma_2$, the spin factor

$$g_{j_c} = \left(\frac{2I_e + 1}{2I_g + 1}\right)\left(\frac{2j_f + 1}{2j_c + 1}\right), \tag{60}$$

$\mathcal{L}(\delta)$ denotes the Lorentzian function:

$$\mathcal{L}(\delta) = \frac{(\Gamma/2)^2}{\hbar^2\delta^2 + (\Gamma/2)^2}, \tag{61}$$

$\Gamma_{\rm is}^{\rm IC}(f)^{j_c}$ is the internal conversion (IC) width when the electron is emitted from the level j_f, carrying away the energy $E_{\rm is} - |\epsilon_f|$ and the total angular momentum j_c. In (59) it is taken into account that NEEC and IC are inverse processes.

Besides, $P_{\rm ion}^{j_c}(\omega_L^{\rm res})$ means the ionization probability by a laser pulse. For electric dipole transitions it is expressed in terms of the reduced transition probability $B(E1; j_i \to j_c)$ by

$$P_{\rm ion}^{j_c}(\omega_L^{\rm res}) = \frac{2\pi^2}{9}\frac{E_0^2}{\Gamma_L} B(E1; j_i \to j_c)\rho(\epsilon_{\rm res}), \tag{62}$$

where

$$\rho(\epsilon_{\rm res}) = \frac{mk_{\rm res}}{\hbar^2} \tag{63}$$

is the density of the electron states of continuum in vicinity of the resonant energy

$$\epsilon_{\rm res} = \frac{\hbar^2 k_{\rm res}^2}{2m} = \epsilon_i + \hbar\omega_L^{\rm res}. \tag{64}$$

Returning to Eq. (51), let me remark that the integration of $\mathcal{T}_{ba}(\omega_{ab})$ over $\epsilon(\kappa)$ leads to appearance of the smooth factor $\rho(\epsilon_0(\kappa'))$, still depending on κ'. But after integration of $|\mathcal{T}_{ba}(\omega_{ab})|^2$ over $\epsilon(\kappa')$ it is replaced by $\rho(\epsilon_{\rm res})$. This can be done because $|\mathcal{T}_{ba}(\omega_{ab})|^2$ contains a sharp peak like the δ function $\delta(\epsilon'(\kappa') - \epsilon_0')$.

B. EB via discrete atomic level

In addition, I analyze also the case of EB through a discrete intermediate atomic levels $|j_1 m_1\rangle$, having the energy ϵ_1 and width Γ_1. It is convenient to write down the corresponding transition matrix as

$$\mathcal{T}_{ba}(\omega_{ab}) = -\sum_{M_e m_1} \frac{V'_{bc_2} R^+_{c_2 c_1} V^f_{c_1 a}(\omega_{ab})}{\left(\epsilon'(\kappa') - \epsilon_0 + i\frac{\Gamma_1}{2}\right)\left(\epsilon'(\kappa') - \epsilon_0' + i\frac{\Gamma_2}{2}\right)}, \tag{65}$$

where

$$\epsilon_0 = \epsilon_1 - \epsilon_f - \epsilon_v \tag{66}$$

$$\epsilon_0' = E_{\rm is} - \epsilon_v. \tag{67}$$

The atomic excitation probability is determined by the expression

$$\mathcal{P}_1(\epsilon') = \frac{1}{2j_i+1}\sum_{m_1, m_i}\left(\frac{2\pi}{\hbar}\right)^2 \left|V^f_{c_1 a}(\omega_{ab})\right|^2. \tag{68}$$

It still depends on ϵ' because

$$\hbar\left(\omega_L^0 + \omega_{ab}\right) = -(\epsilon' - \tilde{\epsilon}_0) \tag{69}$$

with

$$\tilde{\epsilon}_0 = \hbar\omega_L^0 + \epsilon_i - \epsilon_f - \epsilon_v. \tag{70}$$

Employing the Wigner-Eckart theorem

$$\langle j_f m_f|\mathfrak{M}_\mu(\lambda l)|j_i m_i\rangle = \frac{(j_i l m_i \mu | j_f m_f)}{\sqrt{2j_f+1}}\langle j_f||\mathfrak{M}(\lambda l)||j_i\rangle, \tag{71}$$

where $\langle j_f||\mathfrak{M}(\lambda l)||j_i\rangle$ is the reduced matrix element, as well as the orthogonality rule for the Clebsh-Gordan coefficients

$$\sum_{m_1 m}(j_1 j_2 m_1 m_2|jm)(j_1 j_2' m_1 m_2'|jm) = \frac{2j+1}{2j_2+1}\delta_{j_2 j_2'}\delta_{m_2 m_2'}, \tag{72}$$

unitarity of the Wigner functions

$$\sum_{\mu=-j}^{j} D^{j*}_{\mu\nu}(\alpha\beta\gamma)D^{j}_{\mu\nu'}(\alpha\beta\gamma) = \delta_{\nu\nu'}, \tag{73}$$

and the well-known expression for the reduced probability

$$B(\lambda l; i \to f) = (2j_i+1)^{-1}|\langle j_f||\mathfrak{M}(\lambda l)||j_i\rangle|^2, \tag{74}$$

one arrives at

$$\mathcal{P}_1(\epsilon' - \tilde{\epsilon}_0) = \frac{\pi}{9}\frac{B(E1; j_i \to j_1)E_0^2}{(\epsilon' - \tilde{\epsilon}_0)^2 + (\Gamma_L/2)^2}. \tag{75}$$

Combining Eqs. (54), (55), (65) with (75) one has the integral representation for the EB probability:

$$P_{\rm EB} = \frac{\pi}{9}E_0^2\int_{-\infty}^{\infty} d\epsilon' \frac{\Gamma_2(\epsilon')}{\left[(\epsilon' - \epsilon_0)^2 + \left(\frac{\Gamma_1}{2}\right)^2\right]} \times \frac{|E_{\rm int}|^2 B_e(E1; j_i \to j_1)}{\left[\left(\epsilon' - \epsilon_0'\right)^2 + \left(\frac{\Gamma_2}{2}\right)^2\right]\left[(\epsilon' - \tilde{\epsilon}_0)^2 + \left(\frac{\Gamma_L}{2}\right)^2\right]}. \tag{76}$$

Here I used the following notation:

$$|E_{\rm int}|^2 = \sum_{M_e m_f}\left|R^+_{c_2 c_1}\right|^2, \tag{77}$$

which denotes the NEET coupling parameter, calculated in Refs. [36,37,41]. In particular, for magnetic dipole transitions it is defined by

$$|E_{\rm int}|^2 = \frac{4\pi}{9}e^2 k_\gamma^4\left(j_1 1\frac{1}{2}0|j_f\frac{1}{2}\right)^2 \times |\mathcal{M}(M1)|^2 B(M1; I_g \to I_f), \tag{78}$$

where $B(M1; I_g \to I_e)$ is the reduced probability of the nuclear transition from the ground state to the excited one, $\mathcal{M}(M1)$ denotes the matrix element for the $M1$ electronic transition (see Refs. [36,37]), and the wave vector of the virtual photon $k_\gamma = E_{\rm is}/\hbar c$.

The integrant in (76) contains apart of the function $\Gamma_2(\epsilon')$ yet the factors like the δ functions. Therefore after integration over ϵ' it transforms to usual width Γ_2.

By introducing new integration variable $x = \epsilon' - \tilde{\epsilon}_0$ and applying the relationship

$$\frac{1}{x^2 + (\Gamma_L/2)^2} = \frac{2}{\Gamma_L}{\rm Re}\int_0^\infty d\mu e^{ix\mu - \Gamma_L\mu/2}, \tag{79}$$

calculation of $P_{\rm EB}$ reduces to estimation of the integral

$$\mathcal{I} = \mathrm{Re}\frac{1}{2\pi}\int_0^\infty d\mu \times \int_{-\infty}^{\infty} \frac{e^{ix\mu-\Gamma_L\mu/2}dx}{\left[(x-\hbar\delta_a)^2+\left(\frac{\Gamma_1}{2}\right)^2\right]\left[(x-\hbar\delta)^2+\left(\frac{\Gamma_2}{2}\right)^2\right]}, \tag{80}$$

where the detunings of the atomic and nuclear-atomic resonances are

$$\delta_a = \omega_a - \omega_L^0, \quad \delta = \omega_{\rm res} - \omega_L^0. \tag{81}$$

with $\omega_a = \epsilon_1 - \epsilon_i$. The integral over x is calculated by means of the contour integration. I introduce now one of the main NEET parameters, the mismatch of the atomic and nuclear transitions:

$$\Delta = \hbar(\delta_a - \delta) = \epsilon_1 - \epsilon_f - E_{\rm is}. \tag{82}$$

Then in the approximation $|\Delta| \gg \Gamma_{1(2)}$ one gets

$$\mathcal{I} = \frac{4}{\Gamma_L}\frac{1}{\Delta^2}\left[\frac{1}{\Gamma_1\Gamma_a}\mathcal{L}_a(\delta_a) + \frac{1}{\Gamma_2\Gamma}\mathcal{L}(\delta)\right], \tag{83}$$

where $\Gamma_a = \Gamma_L + \Gamma_1$ and $\Gamma = \Gamma_L + \Gamma_2$ as well as

$$\mathcal{L}_a(\delta_a) = \frac{(\Gamma_a/2)^2}{\hbar^2\delta_a^2 + (\Gamma_a/2)^2}, \quad \mathcal{L}(\delta) = \frac{(\Gamma/2)^2}{\hbar^2\delta^2 + (\Gamma/2)^2}. \tag{84}$$

Finally the EB probability splits into two terms:

$$P_{\rm EB}(\omega_L^0) = P_{\rm EB}^{(1)}(\delta_a) + P_{\rm EB}^{(2)}(\delta), \tag{85}$$

corresponding to two coupled NEET resonances $|c_1\rangle$ and $|c_2\rangle$ and having the form

$$P_{\rm EB}^{(1)}(\delta_a) = \frac{\Gamma_2}{\Gamma_1}\frac{\Gamma_L}{\Gamma_a}\frac{|E_{\rm int}|^2}{\Delta^2}\mathcal{P}_1^{\rm res}\mathcal{L}_a(\delta_a) \tag{86}$$

with

$$P_{\rm EB}^{(2)}(\delta) = \frac{\Gamma_L}{\Gamma}\frac{|E_{\rm int}|^2}{\Delta^2}\mathcal{P}_1^{\rm res}\mathcal{L}(\delta). \tag{87}$$

Here

$$\mathcal{P}_1^{\rm res} = \frac{4\pi}{9}B(E1; j_i \to j_1)E_0^2/\Gamma_L^2 \tag{88}$$

stands for the resonant value of the electron transition probability to the first excited state if $\omega_L^0 = \omega_a$.

V. EXCITATION BY CONVENTIONAL LASERS

In this section I will consider the role of conventional lasers, which can in principle realize the Rabi oscillations in two-level atom. Let the laser pulse be short compared to the lifetimes τ_e of all the electron states associated with the EB, that is $\tau \ll \tau_e$. Hence at first the electron is excited with a probability $\mathcal{P}_1$ to the level $j_1 m_1$, degenerated in m_1, and only then the NEET starts up.

In order to describe multiphoton excitation of the isomer one rewrites the resonant part of the operator $\mathcal{T}(t)$ in the form[1]

$$\mathcal{T}(t) = V'\mathcal{G}_r^+(\mathcal{E}_a)\mathcal{T}_f(t), \tag{89}$$

where the reduced Green's operator $\mathcal{G}_r^+(\mathcal{E}_a)$ does not include the interaction with the laser field V_f:

$$\mathcal{G}_r^+(\mathcal{E}_a) = (\mathcal{E}_a + i\eta - \mathcal{H}_0(t) - V_r)^{-1}, \tag{90}$$

while new transition operator depends on it:

$$\mathcal{T}_f(t) = V_f(t) + V_f(t)\mathcal{G}^+(\mathcal{E}_a)V_f(t), \tag{91}$$

and describes the ionization process in all the orders of the perturbation theory.

Then the EB via continuum is described by the following $\mathcal{T}$ matrix:

$$\mathcal{T}_{ba}(\omega_{ab}) = -\sum_{c_2c_1}\frac{V'_{bc_2}R^+_{c_2c_1}\mathcal{T}^f_{c_1a}(\omega_{ab})}{\left(\epsilon_c(\kappa) - \epsilon_0(\kappa') - i\frac{\Gamma_1}{2}\right)\left(\epsilon'(\kappa') - \epsilon'_0 + i\frac{\Gamma_2}{2}\right)}, \tag{92}$$

which coincides with (51) if the laser field is so weak that it is sufficient to keep in $\mathcal{T}_f(t)$ only the first term $V_f(t)$. With the aid of (92), one again arrives at the expressions (58), (59) for the EB probability triggered by laser pulse. However, now the ionization probability incorporates contribution of all orders of the interaction $V_f(t)$.

A much more interesting situation arises in the case of the EB via discrete atomic levels. Again excitation of the isomer is determined by Eqs. (85)–(87), but with the probability $\mathcal{P}_1$, which demonstrates Rabi oscillations. As it is usually done, degeneration of the atomic levels is ignored and the atom is considered with two levels $|i\rangle$ and $|1\rangle$, to be coupled by a short laser pulse. In adiabatic approximation it is introduced the instantaneous Rabi frequency $\Omega(t) = -\mathbf{d}_{1i}\mathbf{E}_0(t)/\hbar$, where $\mathbf{d} = -e\mathbf{r}$ is the dipole moment of the electron. In the case of exact resonance $\omega_0 = \omega_a$, if duration of the pulse $\tau \ll \tau_1$ the probabilities of finding electron in the states $|i\rangle$ and $|1\rangle$ are [53]

$$p_i(t) = \cos^2\frac{\vartheta(t)}{2}, \quad p_1(t) = \sin^2\frac{\vartheta(t)}{2}, \tag{93}$$

where

$$\vartheta(t) = \int_{-\infty}^{t}\Omega(t')dt'. \tag{94}$$

Then the transition probability is determined by the formula

$$\mathcal{P}_1 = \sin^2\frac{\Theta}{2}, \tag{95}$$

depending on the so-called pulse square $\Theta = \vartheta(\infty)$. The pulses with $\Theta = \pi$, called π pulses, ensure inverse population of the levels, that is $\mathcal{P}_1 = 1$.

[1] See the appropriate operator transformations in Ref. [49], p. 487.

VI. COMPARISON WITH OTHER MODELS

The power flux density per unit square (intensity) $I_L(t)$ of the laser radiation, averaged over quick oscillations with frequency ω_L^0, is related with the pulse envelope by

$$I_L(t) = \frac{c}{8\pi} E_0^2(t). \tag{96}$$

The number of photons transmitting through unit square per unit time is determined by

$$j(t) = I_L(t)/\hbar\omega_L^0. \tag{97}$$

The excitation rate of the isomer reads

$$w(t) = j(t)\langle\sigma_{\rm EB}(\omega_L)\rangle, \tag{98}$$

where $\langle\sigma_{\rm EB}(\omega_L)\rangle$ stands for the averaged cross section of EB, generated by the laser pulses. When deriving it in Ref. [51] I missed the factor 2. The correct expression in the vicinity of the resonance should be

$$\langle\sigma_{\rm EB}(\omega_L)\rangle = \sum_{j_c} g_{j_c} \frac{\Gamma_{is}^{\rm IC}(f)^{j_c}}{\Gamma} \mathcal{L}(\delta)\sigma_{\rm ion}^{j_c}\left(\omega_L^{\rm res}\right), \tag{99}$$

where $\sigma_{\rm ion}^{j_c}(\omega_L^{\rm res})$ is the partial ionization cross section by photons having the resonance frequency, when a photoelectron attributes the angular momentum j_c.

Integrating (98) one obtains the EB probability in the laser field:

$$P_{\rm EB} = j_{\rm tot}\langle\sigma_{\rm EB}(\omega_L)\rangle, \tag{100}$$

where the total flux of photons

$$j_{\rm tot} = \frac{W}{S\hbar\omega_L^0} = \frac{1}{8\pi}\frac{E_0^2}{\Gamma_L k_0}. \tag{101}$$

Here W is the energy of the laser pulse and S is a square of the beam focusing spot.

In analogy with (100) one can write

$$P_{\rm ion}(\omega_L^{\rm res}) = j_{\rm tot}\sigma_{\rm ion}\left(\omega_L^{\rm res}\right). \tag{102}$$

Then multiplying both sides of (99) by $j_{\rm tot}$ one arrives at Eq. (59). Thus, both approaches lead to the same result.

Karpeshin and Trzhaskovskaya [33] using the perturbation theory analyzed the EB, which leads to excitation of ^{229m}Th via bound atomic levels. They found two peaks in its pumping rate as a function of the photon frequency. Using the above technique one can show that their results well correlate with my equations. In particular, from Eqs. (86), (58) it is seen that the intensities of the peaks in the EB probability are related by

$$\frac{P_{\rm EB}^{(1)}(0)}{P_{\rm EB}^{(2)}(0)} = \frac{\Gamma_2}{\Gamma_1} \tag{103}$$

if the laser has wide frequency distribution. The same result has been previously obtained in Ref. [33], where it was outlined that in the case when $\Gamma_2 \ll \Gamma_1$ it is preferable to tune the laser into resonance with the second atomic-nuclear transition.

However, I cannot share the opinion of Karpeshin and Trzhaskovskaya [33] about the nature of the peaks in the function $P_{\rm EB}(\omega_L^0)$. They stated that the first peak results from the NEET process, while the second results from the inverse bound internal conversion. On the contrary, both peaks are due to two resonant levels of the unified atomic-nuclear system, whose coupling is described by the formulas of the NEET theory.

VII. CONCLUSION

Interaction of the laser radiation with atoms is usually described either in the quasiclassical approximation or with the aid of the Bloch equations, which govern the population probabilities of the atomic levels (see, e.g., Refs. [52,53]). Here I preferred the first approach and approximate a laser pulse by the classical electromagnetic wave packet with the Lorentzian frequency distribution. The interaction of the atomic electrons with such a laser field $V_f(t)$ explicitly depends on time. Therefore one must solve rather cumbersome time-dependent Schrödinger equations in order to find the excitation probability of ^{229m}Th by the laser pulse via EB. However, the task isconsiderably simplified by applying the formalism of generalized Floquet functions with their period $T \to \infty$ [55]. Then the time-dependent problem is reduced to calculations of the transition matrix, using the technique of the stationary scattering theory.

At first I considered the EB through the continuing electron spectrum (see Fig. 1). In this case the atomic electron under the influence of the laser pulse passes to the continuum with the wave vector $\boldsymbol{\kappa}_c$ and polarization ν. Then the whole system (nucleus+electrons) occurs in one of the first resonance states $|c_1\rangle$, which differ by κ. After that the electron returns to the atom, filling the vacancy $|j_f m_f\rangle$ and transferring its energy to the nucleus ^{229}Th by the emitted virtual photon. This transition is called NEEC [57]. In the approximation of weak laser pulses I derived simple formulas (58), (59) for the EB probability $P_{\rm EB}$ and compared them with previous calculations [51] of the average excitation cross section. When the ionization probability of the atom is calculated in the first order of $V_f(t)$ (weak laser), it is proven that both approaches provide the same result.

Moreover, the EB via a bound atomic level is also regarded. It is interesting that in this case the $P_{\rm EB}(\omega_L^0)$, determined by Eqs. (85)–(87) as a function of the laser carrier frequency ω_L^0, represents two peaks associated with the resonant levels of the atomic-nuclear system $|c_1\rangle$ and $|c_2\rangle$. The first peak is located at the frequency ω_a of direct electron transition from the initial state $|j_i m_i\rangle$ to the excited one $|j_1 m_1\rangle$. The second peak at the frequency $\omega_{\rm res}$ is provided by the two-step atomic-nuclear transition to the state $|c_2\rangle$ through the intermediate state $|c_1\rangle$. Coupling of these states is realized by the virtual $M1$ photons, bringing the electron energy to the nucleus. The same peculiarities of the NEET spectra, generated by x rays near K photoabsorption threshold, have been discussed in Refs. [39–41]. At the same time, in the case of electronic transitions via continuum there survives only the second peak of $P_{\rm EB}(\omega_L^0)$. It is caused by the destructive interference of transitions into the continuum of states differing by the kinetic energy of the photoelectron.

Consider now in more detail the case (b) of Fig. 1, when the EB via continuum is excited by means of two lasers. Then Γ_2 in Eq. (59) coincides with the isomer width $\Gamma_{\rm is}$. In neutral

Th atom decay of the isomeric nucleus occurs mainly in the conversion channel, so that $\Gamma_{is} \approx \Gamma_{is}^{\rm IC}(7s)$ and the expression (59) for the EB probability considerably simplifies. Its resonant value can be written as

$$P_{\rm EB}(0) = \frac{2}{3}\frac{1}{1+\tau_{is}/\tau_L}P_{\rm ion}, \tag{104}$$

where the isomer lifetime $\tau_{is} = \hbar/\Gamma_{is}$. The experimental value of the half-life for ^{299m}Th in neutral atom is $T_{is}^{1/2} = 7 \pm 1\mu$s [59]. Respectively, its lifetime $\tau_{is} \approx 10\ \mu$s.

In possible experiment the neutral thorium atoms with nuclei ^{229}Th in the ground state should be embedded in the surface of any target. Perhaps, it can be done in the same manner as in the skilful experiment of Seiferle *et al.* [1]. Next after irradiation of the target by two lasers one must detect the IC electrons as a benchmark of the isomer ^{229m}Th population. However, the energies of both the photoelectrons and the IC electrons are approximately equal to $E_{is} + \epsilon_f$, because $\epsilon_v = -\epsilon_f$. In order to separate IC electrons from the background of photoelectrons the duration of the laser pulse τ_L should be much less than the isomer lifetime τ_{is}. Then applying a time-delay scheme one can safely detect the conversion electrons. For $\tau_L = 1\mu$s if $P_{\rm ion} = 0.5$ one finds from (104) that the relative number of realized EB equals $P_{\rm EB}(0) = 3\%$.

Combining Eqs. (101)–(102) one gets the following formula for estimation of the pulse energy:

$$W = \hbar\omega_L^{\rm res}\left(S/\sigma_{\rm ion}\left(\omega_L^{\rm res}\right)\right)P_{\rm ion}\left(\omega_L^{\rm res}\right), \tag{105}$$

where $\omega_L^{\rm res} = E_{\rm is} + \epsilon_f - \epsilon_i$. These energies are $E_{\rm is} = 8.28$ eV [1] and $\epsilon_i - \epsilon_f = 1.793$ eV [60]. It is also used the ionization cross section $\sigma_{\rm ion}(\omega_L^{\rm res}) \approx 5.2 \times 10^{-18}$ cm^2 [47] of the initial atomic state $6d_2 7s^1 7p^1$ by photons with frequency $\omega_L^{\rm res}$. In addition, a square of the beam spot $S = 1$ mm^2 and $P_{\rm ion} = 0.5$ are taken. Then the EB can be realized by a laser with the pulse energy $W \sim 1$mJ in 3% of nuclei ^{229}Th when the atom is in the state $6d_2 7s^1 7p^1$, excited by the first laser. The estimations of Ref. [33] show that standard lasers ensure significant populations of this atomic level.

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