Effect of atomic electrons on the 7.6-eV nuclear transition in $^{229}Th^{3+}$

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We have considered an effect of atomic electrons on the nuclear ^{229m}Th-^{229g}Th transition in $^{229}Th^{3+}$ due to the electronic bridge process. Based on a recent experimental result we assumed the energy difference between the isomeric and the ground nuclear states to be equal to 7.6 eV. We have calculated the ratios of the electronic bridge process probability ($\Gamma_{\rm EB}$) to the probability of the nuclear radiative transition (Γ_N) for the electronic $5f_{5/2} \to 6d_{3/2}, 6d_{5/2}, 7s$ and the $7s \to 7p_{1/2}, 7p_{3/2}$ transitions and found $\Gamma_{\rm EB}/\Gamma_N \sim 0.01$–$0.1$ for the former and $\Gamma_{\rm EB}/\Gamma_N \sim 20$ for the latter.

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I. INTRODUCTION

The ^{229}Th nucleus is unique in the sense that the energy splitting of the ground state doublet is only several eV. Though prediction of the existence of such a low-lying level was made more than 30 years ago [1], the definite value of energy of the isomeric state ^{229m}Th is not known so far. In 1990 Reich and Helmer [2] measured this excitation energy (ω_N) to be 3.5 ± 1.0 eV. In Ref. [3] it was obtained as 5.5 ± 1.0 eV. Finally, a recent experiment of Beck *et al.* [4] has given an even larger value (with least error) $\omega_N = 7.6 \pm 0.5$ eV.

As to the lifetime of ^{229m}Th, measurements performed by different experimental groups led to different values. The results differ from each other by several orders of magnitude, changing from a few minutes [5] to many hours [6]. Hence, new experimental and theoretical investigations are required.

Special interest in the nuclear transition from the isomeric state to the ground state is motivated by a possibility to build a superprecise nuclear clock [7] and very high sensitivity to the effects of possible temporal variation of the fundamental constants, including the fine structure constant α, strong interaction, and quark mass [8].

Laser cooling of the $^{232}Th^{3+}$ ion was recently reported by Campbell *et al.* in their article [9]. This was the first time when a multiply charged ion has been laser cooled. As a next step this experimental group plans to investigate the nuclear transition between the isomeric and the ground state in a trapped, cold $^{229}Th^{3+}$ ion. Motivated by this experimental progress we have considered the $^{229}Th^{3+}$ ion and calculated the transition probability of the ^{229}Th nucleus from its lowest-energy isomeric state ^{229m}Th to the ground state ^{229g}Th due to the electronic bridge (EB) process.

Our calculations, based on the value of $\omega_N = 7.6\,$eV, showed that if the electrons are in their ground state the ratio of the probability of the EB process, $\Gamma_{\rm EB}$, to the probability of the nuclear radiative $M1$ transition, $\Gamma_N(M1)$, is of the order of (a few) $\times 10^{-2}$. If the valence electron is in the metastable $7s$ state then $\Gamma_{\rm EB}/\Gamma_N(M1) \sim 20$.

The article is organized as follows. In Sec. II we present the general formalism describing the EB process. Section III is devoted to the method of calculation of the properties of Th^{3+}. In Sec. IV we discuss the results of calculations and Sec. V contains concluding remarks. If not stated otherwise the atomic units ($\hbar = |e| = m_e = 1$ and the speed of light $c = 137$) are used.

II. GENERAL FORMALISM

A. Configurations mixing between combined electron-nucleus states

The 7.6-eV transition in ^{229}Th is a $M1$ transition with the amplitude of a fraction of the nuclear magneton μ_N. An amplitude of an allowed electric dipole transition of the valence electron, $\sim$1 a.u., is 10^6 times larger. If there is an electron excited state close to the energy of the nuclear excitation, an energy transfer from the nuclear excited state to the electron excited state accompanied by the electron electric dipole transition to a lower state may radically decrease the lifetime of the nuclear isomeric state. Even if there is no electron state very close to the nuclear excited state, the electron bridge process produces significant effect.

The EB process can be represented by two Feynman diagrams in Fig. 1. In the following we assume that the initial i and the final f electronic states are of opposite parity and a real photon which is emitted or absorbed is the electric dipole photon. The probability of the EB process in this case is much larger than in the case when the i and the f states are of the same parity.

Therefore, the EB process can be effectively treated as the electric dipole $i \to f$ transition of the electron accompanied by the nuclear transition from its isomeric state to the ground state. Denoting by $\mathbf{D}_{\rm EB}$ the amplitude of this "generalized" electric dipole transition and assuming that the initial and the final states are fixed, we obtain

$$\mathbf{D}_{\rm EB} = \sum_n \frac{\langle f|\mathbf{D}|n\rangle\langle g,n|H_{\rm int}|m,i\rangle}{\varepsilon_i + E_m - \varepsilon_n - E_g + i\Gamma_n/2} + \sum_k \frac{\langle g,f|H_{\rm int}|m,k\rangle\langle k|\mathbf{D}|i\rangle}{\varepsilon_f + E_g - \varepsilon_k - E_m + i\Gamma_k/2}, \quad (1)$$

where the indices i, (n,k), and f denote initial, intermediate, and final electronic states, correspondingly; and the indices g and m denote the ground state and the isomeric state of the nucleus. ε_l are the atomic energies, $E_{m,(g)}$ are the nuclear energies of the isomeric (ground) states, and Γ_l are the widths of the intermediate states which may be neglected in the case of ^{229}Th. The operator $\mathbf{D} = -\mathbf{r}$ is the electron electric dipole moment operator and $H_{\rm int}$ is the hyperfine coupling Hamiltonian, which may be represented as a sum over multipole nuclear moments $\mathcal{M}^{\lambda}_K$ of rank K combined

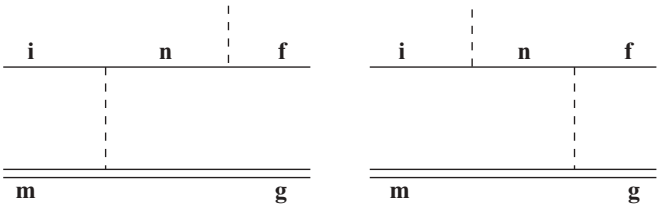

FIG. 1. The single and double solid lines relate to the electronic and the nuclear transitions, correspondingly. The dashed line is the photon line.

with the even-parity electronic coupling operators $\mathcal{T}_{K\lambda}$ of the same rank as

$$H_{\rm int} = \sum_{K\lambda} \mathcal{M}^{\lambda}_{K} \mathcal{T}_{K\lambda}. \tag{2}$$

Neglecting the hyperfine splitting of levels, we can represent the total wave function as a product of the nuclear wave function and the electronic wave function. For instance, $|g,n\rangle = |g\rangle|n\rangle \equiv |I_g M_g\rangle|\gamma_n J_n m_n\rangle$, where I_g is the nuclear spin, M_g is the projection of the nuclear spin, J_n is the electron total angular momentum, m_n is its projection, and γ_n encapsulates all other electronic quantum numbers. Taking into account Eq. (2) we can rewrite Eq. (1) as

$$\mathbf{D}_{\rm EB} = \sum_{K\lambda} \left[\sum_{n} \frac{\langle f|\mathbf{D}|n\rangle\langle n|\mathcal{T}_{K\lambda}|i\rangle}{\omega_{in}+\omega_N} + \sum_{k} \frac{\langle f|\mathcal{T}_{K\lambda}|k\rangle\langle k|\mathbf{D}|i\rangle}{\omega_{fk}-\omega_N} \right] \langle g|\mathcal{M}^{\lambda}_{K}|m\rangle, \tag{3}$$

where $\omega_{ab} \equiv \varepsilon_a - \varepsilon_b$ and $\omega_N = E_m - E_g$.

Thus, we need to carry out the atomic calculation which is similar to that for a forbidden $E1$ transition opened by the hyperfine interaction (see, e.g., Refs. [10–14]). The only difference is that the matrix element (ME) of the nuclear moment $\langle g|\mathcal{M}^{\lambda}_{K}|m\rangle$ here is nondiagonal (there is also a few percentages correction due to variation of the electron magnetic field inside the nucleus). Note that the conventionally defined nuclear moments are related to the tensors $\mathcal{M}^{\lambda}_{K}$ as $\mu \equiv \langle IM_I = I|\mathcal{M}^{0}_{1}|IM_I{=}I\rangle$ and $Q \equiv 2\langle IM_I{=}I|\mathcal{M}^{0}_{2}|IM_I{=}I\rangle$.

The probability $\Gamma_{\rm EB}$ of the electric dipole transition determined by its amplitude $\mathbf{D}_{\rm EB}$ is given by a simple formula (see, e.g., Ref. [15])

$$\Gamma_{\rm EB} = \frac{4}{3}\left(\frac{\omega}{c}\right)^3 |\mathbf{D}_{\rm EB}|^2, \tag{4}$$

where ω is the real photon frequency determined from the low of conservation of energy as $\omega = \varepsilon_i - \varepsilon_f + \omega_N$.

If we average over the initial projections of the electronic and the nuclear total angular momenta m_i and M_m and summing over the final projections m_f and M_g, Eq. (4) is transformed to

$$\Gamma_{\rm EB} = \frac{4}{3}\left(\frac{\omega}{c}\right)^3 \frac{1}{(2I_m+1)(2J_i+1)} \sum_{M_m M_g, m_i m_f} |\mathbf{D}_{\rm EB}|^2 . \tag{5}$$

Substituting Eq. (3) to Eq. (5), applying the Wigner-Eckart theorem and performing the summation over all magnetic quantum numbers of the initial, intermediate and final states, we can reduce Eq. (5) to the form $\Gamma_{\rm EB} = \sum_K \Gamma^{(K)}_{\rm EB}$, where $\Gamma^{(K)}_{\rm EB}$ can be represented by

$$\Gamma^{(K)}_{\rm EB} = \frac{4}{3}\left(\frac{\omega}{c}\right)^3 \frac{|\langle I_g||\mathcal{M}_K||I_m\rangle|^2}{(2K+1)(2I_m+1)(2J_i+1)} \times \left(G^{(K)}_1 + G^{(K)}_{12} + G^{(K)}_2\right), \tag{6}$$

where

$$G^{(K)}_1 \equiv \sum_{J_n} \frac{1}{2J_n+1} \times \left| \sum_{\gamma_k} \frac{\langle \gamma_f J_f||D||\gamma_k J_n\rangle\langle \gamma_k J_n||\mathcal{T}_K||\gamma_i J_i\rangle}{\omega_{ik}+\omega_N} \right|^2, \tag{7}$$

$$G^{(K)}_{12} \equiv 2\sum_{J_t J_n} (-1)^{J_t+J_n} \begin{Bmatrix} J_i & J_t & 1 \\ J_f & J_n & K \end{Bmatrix} \times \sum_{\gamma_k} \frac{\langle \gamma_f J_f||D||\gamma_k J_n\rangle\langle \gamma_k J_n||\mathcal{T}_K||\gamma_i J_i\rangle}{\omega_{ik}+\omega_N} \times \sum_{\gamma_s} \frac{\langle \gamma_f J_f||\mathcal{T}_K||\gamma_s J_t\rangle\langle \gamma_s J_t||D||\gamma_i J_i\rangle}{\omega_{fs}-\omega_N}, \tag{8}$$

and

$$G^{(K)}_2 \equiv \sum_{J_n} \frac{1}{2J_n+1} \times \left| \sum_{\gamma_k} \frac{\langle \gamma_f J_f||\mathcal{T}_K||\gamma_k J_n\rangle\langle \gamma_k J_n||D||\gamma_i J_i\rangle}{\omega_{fk}-\omega_N} \right|^2. \tag{9}$$

The terms $G^{(K)}_1$ and $G^{(K)}_2$ characterize the contributions of the first and second diagrams in Fig. 1 while the "interference" of two these diagrams is given by $G^{(K)}_{12}$.

It is worth noting that Eq. (6) is valid in a general case because deriving it we did not make any approximations. In particular, we did not suppose that there is an electronic transition whose frequency is close to the nuclear transition frequency ω_N. In systems where such a "resonance" transition exists, the expression for $\Gamma_{\rm EB}$ can be significantly simplified.

B. Derivation of the coefficients β_{M1} and β_{E2}

Since the frequency of the nuclear transition from the isomeric state to the ground state of ^{229}Th is very small, in the following we will take into consideration only first two terms in Eq. (6), involving the nuclear magnetic-dipole ($K = 1$) and electric-quadrupole ($K = 2$) moments. Another consequence of the smallness of the nuclear transition frequency is that the probability of the $m \xrightarrow{E2} g$ transition $\Gamma_N(E2)$ is strongly suppressed in comparison to the probability of $m \xrightarrow{M1} g$ transition $\Gamma_N(M1)$.

The probability $\Gamma_N(\tau K, m \to g)$ of the τK transition (where τ denotes M or E) in the ^{229}Th nucleus can be written in a form used in the nuclear physics as (see, e.g., Ref. [16])

$$\Gamma_N(\tau K, m \to g) = 8\pi \frac{k_N^{2K+1}}{[(2K+1)!!]^2} \frac{K+1}{K} B(\tau K, m \to g). \tag{10}$$

Here $k_N \equiv \omega_N/c$ and the reduced probability of the nuclear $m \to g$ transition $B(\tau K, m \to g)$, expressed in terms of the operator $\mathcal{M}_K$, reads as

$$B(\tau K, m \to g) = \frac{1}{2I_m + 1} \frac{2K+1}{4\pi} |\langle I_g || \mathcal{M}_K || I_m \rangle|^2. \quad (11)$$

Using Eq. (10) we find for this transition

$$\frac{\Gamma_N(M1)}{\Gamma_N(E2)} = \frac{100}{3} \frac{1}{k_N^2} \frac{B(M1, m \to g)}{B(E2, m \to g)}. \quad (12)$$

The theoretical value of $B(M1, m \to g)$ was obtained in [17]

$$B(M1, m \to g) \approx 0.086 \mu_N^2. \quad (13)$$

To the best of our knowledge the accurate value of $B(E2, m \to g)$ is unknown. An estimate of this quantity is found in Ref. [18], where Strizhov and Tkalya, referring to Ref. [19], cite the value of several Weisskopf units (W.u.) for $B(E2, m \to g)$.

The definition of 1 W.u. for the $E2$ transition from a nuclear excited state to the ground state (in usual units) is

$$E2\text{: 1 W.u.} = 5.940 \times 10^{-6} A^{4/3} e\mathrm{b}^2, \quad (14)$$

where e is the electron charge and A is the number of nucleons in the nucleus.

Using Eqs. (12), (13), and (14) we arrive at the estimate

$$\frac{\Gamma_N(M1)}{\Gamma_N(E2)} \sim 10^{11}. \quad (15)$$

An accurate calculation of the probability of the nuclear $E2$ transition is beyond the topic of this work. In the following we rely on the estimate given by Eq. (15) and concentrate our efforts on the computation of the ratios

$$\beta_{M1} = \Gamma_{\mathrm{EB}}^{(1)}/\Gamma_N(M1) \quad \text{and} \quad \beta_{E2} = \Gamma_{\mathrm{EB}}^{(2)}/\Gamma_N(E2),$$

where $\Gamma_{\mathrm{EB}}^{(1,2)}$ are given by Eq. (6) and $\Gamma_N(M1)$ and $\Gamma_N(E2)$ can be found from Eq. (10).

Using these equations we obtain

$$\beta_{M1} = \left(\frac{\omega}{\omega_N}\right)^3 \frac{1}{3(2J_i+1)} \left(G_1^{(1)} + G_{12}^{(1)} + G_2^{(1)}\right) \quad (16)$$

and

$$\beta_{E2} = \left(\frac{\omega}{\omega_N}\right)^3 \frac{1}{k_N^2} \frac{4}{(2J_i+1)} \left(G_1^{(2)} + G_{12}^{(2)} + G_2^{(2)}\right). \quad (17)$$

As follows from the estimate Eq. (15), the probability of the nuclear radiative $E2$ transition from the isomeric state to the ground state in ^{229}Th is completely negligible in comparison with the probability of the $M1$ transition. Based on this estimate one can expect that the electronic part of the EB process mainly contributing to Γ_{EB} can be represented as $i \xrightarrow{\mathcal{T}_1} n \xrightarrow{E1} f$, while the channel $i \xrightarrow{\mathcal{T}_2} n \xrightarrow{E1} f$ can be neglected.

As we will demonstrate below this assumption is valid for $^{229}\mathrm{Th}^{3+}$ in spite of that β_{E2} is many orders of magnitude larger than β_{M1}. The physical meaning of this is as follows. It is known that a neutral atom is not affected by an external electric field. It means that an effective electric field acting on the nucleus is equal to zero because the electrons completely screen the external electric field. Respectively, gradient of electrostatic potential created by the electrons at the nucleus is very large. For a static case (in our consideration it corresponds to $\omega_N = 0$) a similar phenomenon was investigated in Refs. [20,21] where magnetic-dipole shielding factors and electric-quadrupole antishielding factors were calculated for a number of atoms and ions. For instance, for such a heavy atom as Hg, the latter was shown to be four orders of magnitude larger than the former.

Note also that the probability of so-called elastic process (when the final state is the same as the initial state) is much smaller, since instead of $E1$ transitions we have to consider $M1$ (or $E2$) transitions. But the probability of an allowed $M1$ transition is four orders of magnitude smaller than the probability of an allowed $E1$ transition.

The triply ionized thorium $^{229}\mathrm{Th}^{3+}$ is an univalent ion. Respectively, the total electronic angular momentum as well as other quantum numbers coincide with the quantum numbers of the valence electron. The expressions for the single-electron operators T_1 and T_2 and for the MEs of the operators D, T_1, and T_2 are presented in the Appendix.

III. METHOD OF CALCULATION

At the first stage we have solved Dirac-Hartree-Fock (DHF) equations [22] in V^{N-1} approximation. It means that the DHF equations were solved self-consistently for the core electrons. After that we determined valence orbitals for several low-lying states from the frozen-core DHF equations. The virtual orbitals were determined with the help of a recurrent procedure [23]. One-electron basis set of the following size was constructed: 1–20s, 2–20p, 3–20d, 4–25f, 5–18g.

To find wave functions needed for calculation of β_{M1} and β_{E2} we applied a relativistic many-body method initially suggested in Refs. [24,25] and subsequently developed in Refs. [26,27]. In this method one determines wave functions from solution of the effective many-body Schrödinger equation

$$H_{\mathrm{eff}}(E_n)|\Psi_n\rangle = E_n|\Psi_n\rangle, \quad (18)$$

with the effective Hamiltonian defined as

$$H_{\mathrm{eff}}(E) = H_{\mathrm{FC}} + \Sigma(E). \quad (19)$$

Here H_{FC} is the frozen-core DHF Hamiltonian and self-energy operator Σ is the energy-dependent correction, involving core excitations, which recovers second order of perturbation theory in residual Coulomb interaction and additionally accounts for certain classes of many-body diagrams in all orders of perturbation theory. We will refer to this approach as the DHF + Σ formalism.

Together with the effective Hamiltonian H_{eff} we introduce effective ("dressed") electric-dipole operator D_{eff} and operators $(\mathcal{T}_K)_{\mathrm{eff}}$ acting in the model space of valence electrons. These operators were obtained within the relativistic random-phase approximation (RPA) [21,26] which describes a shielding of the externally applied electric field by the core electrons. The RPA sequence of diagrams was summed to all orders of the perturbation theory.

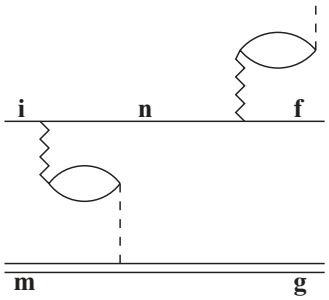

FIG. 2. The single and double solid lines relate to the electronic and the nuclear transitions, correspondingly. The dashed line is the photon line. The wavy line is the Coulomb interaction between electrons.

A representative diagram illustrating a contribution of the RPA corrections in the first order is shown in Fig. 2. As we will show below in certain cases including the RPA corrections is very important because it changes $\Gamma_{\rm EB}$ by orders of magnitude.

With the wave functions obtained from Eq. (18), the quantities $G_1^{(K)}$, $G_{12}^{(K)}$, and $G_2^{(K)}$ can be computed with the Sternheimer [28] or Dalgarno-Lewis [29] method implemented in the DHF + RPA + Σ framework.

For instance, the expression for $G_2^{(K)}$, given by Eq. (9), can be rewritten as

$$G_2^{(K)} = \sum_{J_n} \frac{1}{2J_n+1} |\langle \gamma_f J_f || \mathcal{T}_K || \delta\psi, J_n \rangle|^2, \qquad (20)$$

where an intermediate-state wave function $|\delta\psi\rangle$ can be found from the inhomogeneous equation

$$|\delta\psi\rangle = \frac{1}{\varepsilon_f - \omega_N - H_{\rm eff}} D_z |i\rangle \qquad (21)$$

and then $|\delta\psi, J_n\rangle$ is obtained by projecting the wave function $|\delta\psi\rangle$ to the state with the definite value of J_n. Similarly we can derive the expressions for $G_1^{(K)}$ and $G_{12}^{(K)}$.

Only excitations of the valence electron to higher virtual orbitals are included in the intermediate-state wave function $|\delta\psi\rangle$ due to the presence of $H_{\rm eff}$ in Eq. (21). Additional contributions to $G_1^{(K)}$, $G_{12}^{(K)}$, and $G_2^{(K)}$ come from particle-hole excitations of the core. The role of these contributions will be discussed more detailed in the next section.

Since Th^{3+} is a univalent element, the quantities $G_1^{(K)}$, $G_{12}^{(K)}$, and $G_2^{(K)}$ can be obtained by another method. We can directly sum over *all* intermediate states using the single-electron wave functions found at the stage of constructing the basis set. An accuracy of this approach is comparable to the accuracy of the more refined method of solving the inhomogeneous equation. The reason is that, despite a nonresonant character of the EB process in $^{229}Th^{3+}$ for $\omega_N = 7.6$ eV, there are only a few intermediate states in Eqs. (7), (8), and (9) (whose denominators are small) that give a dominant contribution to $\Gamma_{\rm EB}$.

We would like to stress that in the sums over the intermediate states in Eqs. (7), (8), and (9) the states $(\gamma_i J_i) = (\gamma_k J_n)$ or $(\gamma_f J_f) = (\gamma_k J_n)$ are allowed. This is due to that the ME of the nuclear moment $\langle g|\mathcal{M}_K^{\lambda}|m\rangle$ is nondiagonal and, correspondingly, the diagonal MEs of the electronic operator T_K should be included into consideration. Note that the diagonal MEs of the operators T_K are large and the inclusion of these contributions to $\Gamma_{\rm EB}$ significantly affects the final value of the latter.

IV. RESULTS AND DISCUSSION

To check the quality of the constructed wave functions we have calculated the energy levels for a number of low-lying states and compared them with the experimental data. Some details regarding the energy levels computation can be found in our recent article [30]. We present in Table I the results obtained on the stage of pure DHF approximation and in the frame of DHF + Σ formalism.

As seen from Table I on the stage of the DHF approximation the order of the low-lying levels is incorrect. For instance, the $6d_{3/2}$ state lays deeper than the $5f_{5/2}$ state. An agreement between theoretical and experimental energy levels is rather poor. The inclusion of the core-valence correlations restores the correct order of the states and significantly improves the agreement with the experimental energy levels. Nevertheless in certain cases (e.g., for the $7s$ state) the energy levels were reproduced not very accurately. For this reason in the following calculation of β_{M1} and β_{E2} we used the experimental energies for the low-lying states.

We have carried out calculations of the coefficients β_{M1} and β_{E2} for $\omega_N = 7.6$ eV considering the ground state $5f_{5/2}$ and the metastable state $7s$ as the initial state i. As follows from the discussion above the final states should be of opposite parity in comparison to the initial states. Respectively, we considered $6d_{3/2}$, $6d_{5/2}$, and $7s$ states to be the final states when the initial state was $5f_{5/2}$. The $7p_{1/2}$ and the $7p_{3/2}$ states were the final states when the initial state was $7s$.

TABLE I. The low-lying energy levels (in cm^{-1}) in the DHF and the DHF + Σ approximations are presented. The theoretical values are compared with the experimental data.

DHF		DHF + Σ		Experiment[a]
$6d_{3/2}$	–	$5f_{5/2}$[b]	–	–
$6d_{5/2}$	4225	$5f_{7/2}$	4798	4325
$5f_{5/2}$	5190	$6d_{3/2}$	9091	9193
$5f_{7/2}$	8617	$6d_{5/2}$	14835	14486
$7s_{1/2}$	11519	$7s_{1/2}$	21321	23131
$7p_{1/2}$	46702	$7p_{1/2}$	59436	60239
$7p_{3/2}$	58225	$7p_{3/2}$	72677	73056
$8s_{1/2}$	102595	$8s_{1/2}$	120085	119622
$7d_{3/2}$	103148	$7d_{3/2}$	120898	119685
$7d_{5/2}$	104763	$7d_{5/2}$	122657	121427
$6f_{5/2}$	111874	$6f_{5/2}$	128734	127262
$6f_{7/2}$	112316	$6f_{7/2}$	129227	127815
$8p_{1/2}$	117185	$8p_{1/2}$	135144	134517
$8p_{3/2}$	122194	$8p_{3/2}$	140558	139871
$9s_{1/2}$	142328	$9s_{1/2}$	161481	160728

[a]Reference [31].

[b]The removal energy of the $5f_{5/2}$ state was found to be equal to 0.9414 a.u. on the DHF stage and 1.0584 a.u. on the (DHF + Σ) stage. The experimental value is 1.0588 a.u.

TABLE II. The coefficients β_{M1} obtained for certain $i \to f$ transitions for $\omega_N = 7.6$ eV in the DHF, the DHF + RPA, and the DHF + RPA + Σ (denoted as +Σ) approximations are presented. The coefficients β_{E2} are given in the DHF + RPA approximation.

		β_{M1}			β_{E2}
i	f	DHF	DHF + RPA	+Σ	DHF + RPA
$5f_{5/2}$	$6d_{3/2}$	0.015	0.0037	0.015	2×10^8
	$6d_{5/2}$	0.0015	0.051	0.060	5×10^7
	$7s_{1/2}$	2×10^{-9}	0.032	0.037	5×10^7
$7s_{1/2}$	$7p_{1/2}$	18	19	23	7×10^8
	$7p_{3/2}$	4.1	4.4	5.3	1×10^8

In Table II we present the results obtained (i) on the stage of pure DHF approximation, (ii) in the DHF + RPA approximation, and (iii) in the frame of DHF + RPA + Σ formalism. Including the RPA corrections is formally reduced to replacement of the "bare" operators by the "dressed" operators. In particular, solving the inhomogeneous equation we have to replace the operators $\mathcal{T}_K$ in Eq. (20) and D in Eq. (21) by $(\mathcal{T}_K)_{\rm eff}$ and $D_{\rm eff}$, correspondingly.

As seen from the table in certain cases the inclusion of the RPA corrections increases the probability of the EB process by several orders of magnitude. It happens, for example, for the $5f_{5/2} \to 7s$ transition. The channel $5f_{5/2} \xrightarrow{\mathcal{T}_1} n \xrightarrow{E1} 7s$ turns out strongly enhanced because the "dressed" MEs $\langle 5f_{5/2}||(\mathcal{T}_1)_{\rm eff}||n\rangle$ are much larger in absolute value than the "bare" MEs $\langle 5f_{5/2}||\mathcal{T}_1||n\rangle$. Indeed, we have to consider the intermediate states n that admit the $E1$ transitions $n \to 7s$. But for such n the "bare" MEs $|\langle 5f_{5/2}||\mathcal{T}_1||n\rangle|$ are very small.

The coefficient β_{M1} is rather small for the $5f_{5/2} \to 6d_{3/2}$ transition and the RPA and the Σ corrections change its value significantly. The reason is that $G_1^{(1)}$, $G_{12}^{(1)}$, and $G_2^{(1)}$ are comparable in their magnitudes but $G_{12}^{(1)}$ is negative. In the DHF + RPA approximation it leads to a large cancellation between these terms.

When we consider the $7s$ state as the initial state, the main channel of the process is $7s \xrightarrow{\mathcal{T}_1} 8s \xrightarrow{E1} 7p_{1/2,3/2}$. Respectively, the first diagram in Fig. 1 (the term $G_1^{(1)}$) gives the main contribution to $\Gamma_{\rm EB}$ while $G_{12}^{(1)}$ and $G_2^{(1)}$ only slightly correct this value. As is seen $\beta_{M1}(7s \to 7p_j)$ are 2–3 orders of magnitude larger than $\beta_{M1}(5f_{5/2} \to 6d_j; 7s)$. This is due to the large value of the ME $\langle 7s||\mathcal{T}_1||8s\rangle$.

As is seen from Table II, the inclusion of the core-valence correlations changes the values of β_{M1} at the level of 20% for all considered transitions except the $5f_{5/2} \to 6d_{3/2}$ transition. These corrections are not too large because the core orbitals lay rather deep. In particular the single-electron energy of the external core $6p_{3/2}$ orbital is -2.1 a.u.. For the same reason the contribution to $\Gamma_{\rm EB}$ from the core electrons excitations is small. It is at the level of a few percentages.

We also present in Table II the coefficients β_{E2} obtained for the $5f_{5/2} \to 6d_j; 7s$ and the $7s \to 7p_j$ transitions in the DHF + RPA approximation. We restricted ourselves by this simple approximation because these values are given mostly for reference and an order of magnitude estimate of these quantities is sufficient.

TABLE III. The probabilities $\Gamma_{\rm EB}^{(1)}$ (in s^{-1}) obtained for certain $i \to f$ transitions for $\omega_N = 7.6$ eV in the DHF + RPA + Σ approximation are presented.

i	f	$\Gamma_{\rm EB}^{(1)}$
$5f_{5/2}$	$6d_{3/2}$	9.9×10^{-6}
	$6d_{5/2}$	4.0×10^{-5}
	$7s_{1/2}$	2.4×10^{-5}
$7s_{1/2}$	$7p_{1/2}$	1.5×10^{-2}
	$7p_{3/2}$	3.5×10^{-3}

As we have already mentioned above the coefficients β_{E2} are many orders of magnitude larger than the coefficients β_{M1} found for the same transitions. In particular, for the $5f_{5/2} \to 6d_{3/2}$ transition $\beta_{E2}/\beta_{M1} \sim 10^{10}$. It is not surprising if we take into account the small value of k_N^2 in the denominator of Eq. (17) and the resonant character of the $5f_{5/2} \xrightarrow{\mathcal{T}_2} 7p_{1/2} \xrightarrow{E1} 6d_{3/2}$ transition because the frequency of the $5f_{5/2} - 7p_{1/2}$ transition $\omega_{7p_{1/2},5f_{5/2}} \approx 7.5$ eV is very close to $\omega_N = 7.6$ eV.

In spite of that the main contribution to $\Gamma_{\rm EB}$ comes from the $i \xrightarrow{\mathcal{T}_1} n \xrightarrow{E1} f$ channel. As it follows from the results listed in Table II and Eq. (15), we can neglect the contribution to $\Gamma_{\rm EB}$ coming from the $i \xrightarrow{\mathcal{T}_2} n \xrightarrow{E1} f$ channel and put $\Gamma_{\rm EB} \approx \Gamma_{\rm EB}^{(1)}$.

Using Eqs. (10) and (13) we find $\Gamma_N(M1) \approx 6.6 \times 10^{-4} s^{-1}$ at $\omega_N = 7.6$ eV and, correspondingly,

$$\Gamma_{\rm EB} \approx \Gamma_{\rm EB}^{(1)} \approx 6.6 \times 10^{-4} \beta_{M1} s^{-1}. \qquad (22)$$

The numerical results obtained for $\Gamma_{\rm EB}^{(1)}$ with use of the equation written above are listed in Table III.

Finally, we would like to return to the problem of determination of the nuclear transition frequency ω_N. As we have already mentioned the experimental data regarding this quantity are rather contradictory and its true value is unknown so far. At the same time in certain cases the coefficients β_{M1} are very sensitive to ω_N (therefore the measurements of β_{M1} can be used to find ω_N).

To illustrate it we present in Table IV the coefficients β_{M1} obtained for different values of ω_N for the most important $7s \to n \to 7p_{1/2}$ and $7s \to n \to 7p_{3/2}$ channels. Note that

TABLE IV. The coefficients β_{M1} obtained for several values of ω_N for the $7s \to 7p_{1/2}$ transition in the DHF + RPA approximation are presented.

i	f	ω_N (eV)	β_{M1}
$7s_{1/2}$	$7p_{1/2}$	5.0	0.21
		6.0	4.3
		7.0	13
		7.6	19
$7s_{1/2}$	$7p_{3/2}$	7.0	1.0
		7.6	4.4

the former is energetically allowed if $\omega_N \geqslant 4.6$ eV and the latter is energetically allowed if $\omega_N \geqslant 6.2$ eV.

V. CONCLUSION

In conclusion, we have calculated the ratios of the probabilities $\Gamma^{(1)}_{\rm EB}$ and $\Gamma^{(2)}_{\rm EB}$ to the probabilities of the nuclear radiative $M1$ and $E2$ transitions, β_{M1} and β_{E2}. We found that if the valence electron is in the ground state the coefficients β_{M1} are rather small for all considered transitions. If the valence electron is in the metastable $7s$ state the coefficients β_{M1} are 2–3 orders of magnitude larger and $\Gamma_{\rm EB}/\Gamma_N(M1) \sim 20$.

The spectrum of Th^{3+} is not too dense. As a result for the $i \xrightarrow{\mathcal{T}_1} n \xrightarrow{E1} f$ transitions considered in this work there are no electronic transitions which would be at resonance with the nuclear transition at $\omega_N = 7.6$ eV.

We have found the coefficients β_{E2} to be many orders of magnitude larger than β_{M1}, but based on the estimate $\Gamma_N(M1)/\Gamma_N(E2) \sim 10^{11}$ one can state that the contribution of the $i \xrightarrow{\mathcal{T}_2} n \xrightarrow{E1} f$ channel to $\Gamma_{\rm EB}$ is negligible. It is worth noting that this statement is correct for all considered transitions in spite of the resonant character of the $5f_{5/2} \xrightarrow{\mathcal{T}_2} 7p_{1/2} \xrightarrow{E1} 6d_{3/2}$ transition.

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APPENDIX

The expressions for the single-electron operators T_1 and T_2 can be written as

$$T_{1\lambda}(\mathbf{r}) = \frac{-i\sqrt{2}\boldsymbol{\alpha}\cdot\mathbf{C}^{(0)}_{1\lambda}(\mathbf{n})}{cr^2} \tag{A1}$$

and

$$T_{2\lambda}(\mathbf{r}) = \frac{-C_{2\lambda}(\mathbf{n})}{r^3}, \tag{A2}$$

where $\mathbf{n} \equiv \mathbf{r}/r$ and $\mathbf{C}^{(0)}_{K\lambda}$ is a normalized vector spherical harmonic defined by (see, e.g., Ref. [32])

$$\mathbf{C}^{(0)}_{K\lambda}(\mathbf{n}) = \frac{\mathbf{L}}{\sqrt{K(K+1)}} C_{K\lambda}(\mathbf{n}). \tag{A3}$$

Here $\mathbf{L}$ is the orbital angular-momentum operator and $C_{K\lambda}$ is a spherical harmonic given by

$$C_{K\lambda}(\mathbf{n}) = \sqrt{\frac{4\pi}{2K+1}} Y_{K\lambda}(\mathbf{n}). \tag{A4}$$

To calculate the MEs of the operators D, T_1, and T_2 we define the one-electron wave function $|a\rangle \equiv \psi_a(\mathbf{r})$ as follows

$$\psi_a(\mathbf{r}) = \frac{1}{r}\begin{pmatrix} P_a(r)\Omega_{\kappa_a m_a}(\mathbf{n}) \\ i\,Q_a(r)\Omega_{-\kappa_a m_a}(\mathbf{n}) \end{pmatrix}, \tag{A5}$$

where $\kappa_a = (l_a - j_a)(2j_a + 1)$.

Using the ME $\langle\kappa_b||C_K||\kappa_a\rangle$:

$$\langle\kappa_b||C_K||\kappa_a\rangle = (-1)^{j_b+1/2}\sqrt{(2j_a+1)(2j_b+1)} \times \begin{pmatrix} j_b & j_a & K \\ -1/2 & 1/2 & 0 \end{pmatrix} \xi(l_b + l_a + K),$$

where

$$\xi(x) = \begin{cases} 1, & \text{if } x \text{ is even} \\ 0, & \text{if } x \text{ is odd,} \end{cases}$$

we can write the reduced ME for the electric dipole operator D in the following form

$$\langle n_b\kappa_b||D||n_a\kappa_a\rangle = \langle n_b\kappa_b||-r||n_a\kappa_a\rangle = -\langle\kappa_b||C_1||\kappa_a\rangle \int_0^\infty \{P_bQ_a + P_aQ_b\}\, r\,dr, \tag{A6}$$

where n_i is the principal quantum number.

The reduced ME for the magnetic dipole operator T_1 is represented by

$$\langle n_b\kappa_b||T_1||n_a\kappa_a\rangle = \langle -\kappa_b||C_1||\kappa_a\rangle(\kappa_b + \kappa_a) \int_0^\infty \{P_bQ_a + P_aQ_b\}\, \frac{1}{r^2}\,dr. \tag{A7}$$

Rewriting the angular part of Eq. (A7) in a more simple form we arrive at

$$\langle n_b\kappa_b||T_1||n_a\kappa_a\rangle = \xi(l_b + l_a)(-1)^{j_a+l_a-1/2}\frac{c_{j_aj_b}}{2} \int_0^\infty \{P_bQ_a + P_aQ_b\}\, \frac{1}{r^2}\,dr, \tag{A8}$$

where

$$c_{j_aj_b} \equiv \begin{cases} \sqrt{(2j_a+1)(2j_b+1)/(j_{\min}+1)}, & j_a \neq j_b \\ \sqrt{(2j_a+1)^3/(j_a(j_a+1))}, & j_a = j_b \end{cases}$$

and $j_{\min} = \min(j_a, j_b)$.

The reduced ME for the electric quadrupole operator T_2 is given by

$$\langle n_b\kappa_b||T_2||n_a\kappa_a\rangle = -\langle\kappa_b||C_2||\kappa_a\rangle \times \int_0^\infty \{P_aP_b + Q_aQ_b\}\, \frac{1}{r^3}\,dr. \tag{A9}$$

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