Deexcitation of $^{229}Th^m$: Direct γ decay and electronic-bridge process

Péter Kálmán and Tamás Bükki
Budapest University of Technology and Economics, Department of Experimental Physics, Budafoki út 8 F.I.I.10, H-1521 Budapest, Hungary

(Received 9 December 1999; revised manuscript received 24 March 2000; published 23 January 2001)

Direct γ and electronic-bridge (EB) decay modes of the low-energy isomeric level $^{229}Th^m$ are compared theoretically and it is found that the decay rates for the two channels are approximately equal. Our findings disagree with those of a recent theoretical paper, where it was found that deexcitation happens mainly through the EB process. Recent experimental results also mandate a reinterpretation of previous data and suggest, in particular, that no unambigous evidence of the isomeric decay has been seen in previous experiments since luminescence due to α decay may have concealed the effect. Therefore an optical method for the demonstration of existence and the determination of energy separation of these nearby-lying nuclear states is proposed again.

DOI: 10.1103/PhysRevC.63.027601 PACS number(s): 23.20.Lv, 23.20.Nx, 27.90.+b

It has been found in high precision γ-ray measurements that the ^{229}Th nucleus has a metastable state ($I_A=\frac{3}{2}^+$, [631] Nilsson state) with 3.5 ± 1 eV above the ground level (assigned $I_B=\frac{5}{2}^+$, [633] Nilsson state) [1]. In the last few years many laboratories have attempted to demonstrate experimentally the existence of this low-lying level. The work of Irwin and Kim [2] was a precursor for several studies [3–5] that were intended to observe both the direct γ and the electronic-bridge (EB) channels of the decay of $^{229}Th^m$. In the case of the EB process, the excitation energy of the nucleus is transferred to an atomic electron in a bound-bound transition and a photon is also emitted. Its energy corresponds to the difference between the excitation energy of the nucleus and the energy of the electronic transition, to ensure energy conservation. In Ref. [2] the optical spectrum of different samples (the $^{229}Th^m$ was populated by alpha decay of ^{233}U) was examined and it was presumed that, in addition to photons with an energy of about 3.5 eV from the direct $M1$ transition of the nucleus, also photons with an energy of 2.4 eV originating from the EB process were observed. It was supposed in Ref. [2] that in the EB process the thorium atom was excited from the $6d_{3/2}$ ground state to the $7p_{1/2}$ state (see Fig. 1, where the relative energies of the electron shells are taken from Ref. [6]). Richardson *et al.* [3], using an ^{232}U control source in order to compare and filter their spectra, confirmed the observations of Ref. [2]. They found, however, several well separable lines around 3.5 eV because of their better resolution. They stated that the observed lines can refer to the decay of the thorium isomer, but they did not see unambiguous evidence of it. In contrast, according to Utter *et al.* [4] the optical emission of the ^{233}U sample containing $^{229}Th^m$ can be attributed to the luminescence of nitrogen in the air induced by α particles emitted by the sample. The detailed study conducted by Shaw *et al.* [5], in which the optical spectrum of the ^{233}U sample was compared to a N_2 discharge spectrum, showed that the multiplet in the spectrum at 350 nm (around 3.5 eV photon energy) was mainly due to N_2 luminescence. However, the broad peak at 500 nm (about 2.4 eV [2,3]) was definitely not consistent with the N_2 discharge spectrum [5]. Still, the authors are reluctant to state that it may arise from $^{229}Th^m$ and they have found the footprints of this exotic nuclear level, because there are several other effects, e.g., uranyl luminescence [7], from which the extra observations may originate.

Along with the experimental investigations, Karpeshin *et al.* [8] provided a theoretical study of the subject, commenting on Ref. [2]. They have calculated the ratio $R^*=W_{\mathrm{EB}}/W_\gamma$, where W_{EB} is the transition probability per unit time of the EB process and W_γ is that of the direct γ decay. They report a value of $R^*\sim782$ at the nuclear transition energy of 3.5 eV. They have also stated that in the decay of $^{229}Th^m$ through EB process, instead of a single peak, one should have found a doublet associated with the two final electronic states, $7p_{1/2}$ and $7p_{3/2}$. Thus, from Ref. [8], one can conclude that the signal of the direct γ transition at about 3.5 eV is so weak that it is undetectable and the EB process is the prevailing channel.

Unfortunately, however, the calculation of Ref. [8] has some serious flaws and therefore the above, seemingly clarified, picture cannot hold. We report here the results of our calculation on the $^{229}Th^m$ EB decay process leading to a conclusion that contradicts Ref. [8]. As a preliminary to the discussion we note the following. The two graphs that describe the EB process are depicted in Fig. 2. Karpeshin *et al.* have carried out the calculations [8] on the basis of their

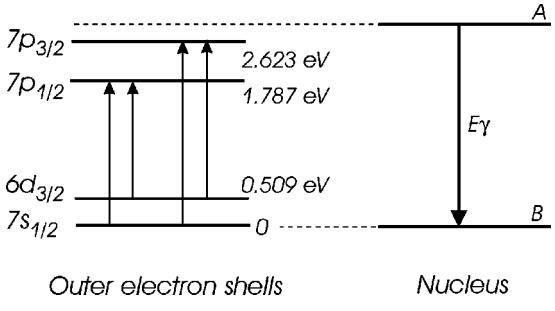

FIG. 1. Energy level scheme of the two nuclear states and the outermost electron shells of ^{229}Th. The figure shows those electronic states which may participate in an electronic-bridge process. In the ground state the $7s$ subshell is filled, there are two electrons in the $6d_{3/2}$ state, and the $7p_{1/2}$ and $7p_{3/2}$ states are empty.

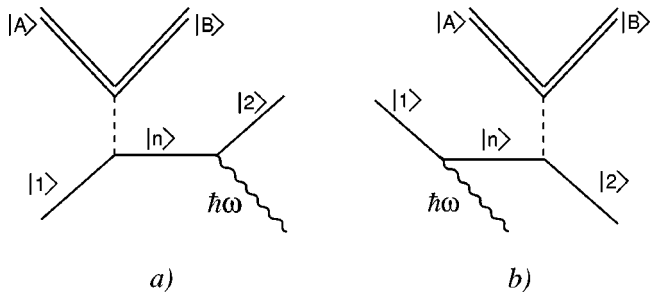

FIG. 2. Feynman diagrams of the electron-bridge process: (a) is the direct; (b) is the crossed graph of the process. Double lines indicate the nucleus (deexciting from metastable state $|A\rangle$ to ground state $|B\rangle$), solid lines indicate an atomic electron (time passes from left to right), dashed lines indicate the electromagnetic interaction between the bound electron and the nucleus, and photon emission is depicted by wavy lines.

earlier work [9] and they have considered some contributions only from the direct graph [Fig. 2(a)] of the EB process while unjustifiably neglecting others. Furthermore, even in the direct graphs, they have not summed over all intermediate states mandated by theory. In our calculation we have taken into account the effect of both the direct and the crossed graphs of Fig. 2 and applied a method that is equivalent to summing over all intermediate states.

We have two aims in this paper. We claim, contrary to Karpeshin *et al.* [8], that the direct γ decay channel is about as probable as the electronic-bridge decay mode. Furthermore, an experimental method for the determination of the energy separation of these nearby-lying nuclear states based on the application of the inverse electronic-bridge process is proposed again.

Our model is a generalization of an earlier theoretical work [10], which has also dealt with the decay of the $^{229}\mathrm{Th}^{m}$ isomer in the EB channel. We work in a one-electron model and use a hydrogenlike Hamiltonian, $H_e=-(\hbar^2/2m)\nabla^2-Z_{\mathrm{eff}}e^2/r$, for the electron, where e is the elementary charge, m is the rest mass of the electron, Z_{eff} is the effective proton number of the nucleus, and r stands for the electron coordinate (the origin of the system of reference is fixed to the center of mass of the nucleus). The electron interacts with the quantized electromagnetic field through the operator

$$H_1=H_1^{\dagger}e^{i\omega t}+H_1^{-}e^{-i\omega t}, \tag{1}$$

where $H_1^{\dagger}=e\vec{r}\cdot\vec{E}_q^{\dagger}$, $H_1^{-}=e\vec{r}\cdot\vec{E}_q$, and

$$\vec{E}_q=i\sum_{\vec{k},\nu}\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2}a_{\vec{k},\nu}\vec{\varepsilon}_{\nu}. \tag{2}$$

Here ω is the angular frequency of the emitted/absorbed photon, V is the volume of normalization, $a^{\dagger}_{\vec{k},\nu}$ is the photon creation, $a_{\vec{k},\nu}$ is the photon annihilation operator, $\vec{k}$ and ν stand for the wave number vector and the state of polarization of the photon, and $\vec{\varepsilon}_{\nu}$ denotes the transverse polarization vector of the quantized electromagnetic field. Finally, H_N is the Hamiltonian of the nucleus and $H_R=\Sigma_{\vec{k},\nu}\hbar\omega(a^{\dagger}_{\vec{k},\nu}a_{\vec{k},\nu}+1/2)$ is the Hamiltonian of the free electromagnetic field. Thus the total Hamiltonian can be written as $H=H_0+H_1+H_2$, where $H_0=H_e+H_N+H_R$.

The H_2 term of the total Hamiltonian describes the electromagnetic interaction between the nucleus and the electron. It involves an instantaneous Coulomb interaction term between the charge densities of the nucleus and the electron cloud and a magnetic interaction term. Retardation effects are negligible since the energy of the nuclear transition is small and comparable to the transition energies of the outermost electronic shell. Thus, in this case, the interaction operator H_2 of the electron and the nucleus is simplified to the sum

$$H_2=V_M+V_E, \tag{3}$$

where V_M is the magnetic-magnetic dipole interaction energy that causes hyperfine splitting of the electronic states and V_E is the well-known static Coulomb-interaction energy of two extended charge clouds. We mention here that our calculation is basically the same as the one in Ref. [10]. The main difference is that besides V_E, we also have a magnetic-magnetic interaction term V_M in H_2. The magnetic interaction consists of three terms, $V_M=V_{Ma}+V_{Mb}+V_{Mc}$, with

$$V_{Ma}=2\mu_0\frac{8\pi}{3}\vec{s}\cdot\vec{\mu}\,\delta^{(3)}(\vec{r}), \tag{4}$$

$$V_{Mb}=-2\mu_0(\vec{s}\cdot\vec{\mu}-3s_r\mu_r)/r^3, \tag{5}$$

and

$$V_{Mc}=2\mu_0(\vec{L}\cdot\vec{\mu})/(\hbar r^3), \tag{6}$$

where $\mu_0=e\hbar/2mc$ is the Bohr magneton, $\vec{s}=\vec{\sigma}/2$ is the Pauli spin operator for the electron ($\vec{\sigma}$ is a vector whose components are the Pauli spin matrices), $\vec{\mu}$ represents the nuclear magnetic moment, s_r and μ_r are the components of $\vec{s}$ and $\vec{\mu}$, respectively, in the direction of $\vec{r}$, and $\vec{L}$ stands for the orbital angular momentum operator of the electron [11]. Furthermore,

$$V_E=-\sum_{\lambda_2>0,\mu_2}\frac{4\pi e}{(2\lambda_2+1)}\mathcal{M}_{\mathrm{nuc}}(E\lambda_2,\mu_2)\frac{Y_{\lambda_2,\mu_2}}{r^{\lambda_2+1}}, \tag{7}$$

where $\mathcal{M}_{\mathrm{nuc}}(E\lambda_2,\mu_2)$ denotes electric multipole operator of multipolarity λ_2 of the nucleus, $Y_{\lambda,\mu}$ are spherical harmonics depending on the electronic coordinates.

Following the train of thought of Ref. [10] it can be shown that the leading part of the total S matrix element (S_{fi}) of the EB process [$S_{fi}=S^a_{fi}+S^b_{fi}$, where S^a_{fi} and S^b_{fi} are the second order matrix elements corresponding to the two graphs of Figs. 2(a) and 2(b), respectively], can be written as

$$S_{fi}=(i\hbar)^{-1}\langle f|H_{\mathrm{EB}}|i\rangle 2\pi\,\delta(\omega_{BA}+\omega-\omega_{12}), \tag{8}$$

where

TABLE I. Computed values of magnetic dipole and electric quadrupole EB coefficients, α, for different electron transitions at nuclear transition energies $E_\gamma = 3.5$ eV and $E_\gamma = 2.4$ eV. [$\alpha(M1) = W_{EB}(M1)/W_\gamma(M1)$; $\alpha(E2) = W_{EB}(E2)/W_\gamma(E2)$, where W_{EB} is the rate of the EB process, W_γ is the rate of the direct γ decay; $\eta = W_\gamma(E2)/W_\gamma(M1)$.] R is the ratio of the EB rate to the direct γ decay rate, $R = W_{EB}^{tot}/W_\gamma^{tot}$. The energy, $\hbar\omega$, of the photon, emitted in the EB process, is also tabulated for each electron transition. The $7s \rightarrow 7p_{3/2}$ transition at $E_\gamma = 2.4$ eV is energetically forbidden. The R^* values are taken from Ref. [8] and should be compared to the $\alpha(M1)$ values of this table.

$E\gamma$ (eV)	Electron transition	$\alpha(M1)$	$\eta\alpha(E2)$	R	R^*	$\hbar\omega$ (eV)
	$7s_{1/2} \rightarrow 7p_{1/2}$	0.28			783	1.713
	$7s_{1/2} \rightarrow 7p_{3/2}$	0.59			782	0.877
3.5				0.87		
	$6d_{3/2} \rightarrow 7p_{1/2}$	5.58×10^{-4}	7.97×10^{-5}			2.222
	$6d_{3/2} \rightarrow 7p_{3/2}$	5.91×10^{-4}	4.73×10^{-5}			1.386
	$7s_{1/2} \rightarrow 7p_{1/2}$	2.41			8.35	0.613
	$7s_{1/2} \rightarrow 7p_{3/2}$				0.07	
2.4				2.41		
	$6d_{3/2} \rightarrow 7p_{1/2}$	3.43×10^{-3}	1.90×10^{-3}			1.122
	$6d_{3/2} \rightarrow 7p_{3/2}$	8.92×10^{-3}	1.51×10^{-3}			0.286

$$H_{EB} = \vec{\alpha}^\dagger \cdot \vec{\nabla}_r H_2, \tag{9}$$

and

$$\vec{\alpha}^\dagger = \frac{e}{m} i \sum_{\vec{k},\nu} \left(\frac{2\pi\hbar\omega}{V} \right)^{1/2} \frac{1}{\omega^2} a^\dagger_{\vec{k},\nu} \vec{\varepsilon}_\nu. \tag{10}$$

Here $\hbar\omega_{BA} = E_A - E_B$ and $\hbar\omega_{12} = E_1 - E_2$ are the nuclear and electronic transition energies, respectively, E_A, E_B and E_1, E_2 are the energy eigenvalues of the corresponding nuclear and electronic states. The above form of the total S matrix element takes into account both graphs and all intermediate states. With its help the transition rate for the process under consideration can be calculated in the standard manner. (For the initial, intermediate, and final states applied and for more details see Ref. [10].)

On the outer electron shells of the thorium atom there are $7s$ and $6d$ electrons (see Fig. 1). We have carried out calculation with those initial ($7s$,$6d$) and final ($7p$) electronic states that were partly included and partly mentioned in Ref. [8]. We have employed Pauli states [11] for the description of the spinning electron. In our case, besides the magnetic coupling ($M1$ transitions in the electronic shell and in the nucleus, as well), electric coupling can also occur as a consequence of the possibility of quadrupole ($E2$) transition in the nucleus. We have considered both possibilities.

We define R, the ratio of the total probability per unit time of the EB process (including $M1$ and $E2$ transitions) to the total decay rate of the direct γ decay process [$W_\gamma^{tot} = W_\gamma(M1) + W_\gamma(E2)$], as $R = W_{EB}^{tot}/W_\gamma^{tot}$, and

$$R = \sum_p \frac{\alpha(M1,sp) + \alpha(M1,dp) + \alpha(E2,dp)\,\eta}{1+\eta}. \tag{11}$$

Here $\eta = W_\gamma(E2)/W_\gamma(M1)$ is the ratio of the rates of the direct γ transitions of multipolarities $E2$ and $M1$. $\alpha(M1) = W_{EB}(M1)/W_\gamma(M1)$ and $\alpha(E2) = W_{EB}(E2)/W_\gamma(E2)$ are magnetic dipole and electric quadrupole EB coefficients, and the letters sp or dp in the bracket refer to $7s_{1/2} \rightarrow 7p$ and $6d_{3/2} \rightarrow 7p$ electronic transitions, respectively. For the final $7p$ electronic state we have two possibilities: $7p_{1/2}$ and $7p_{3/2}$. We note here that in the case of $7s \rightarrow 7p$ transitions the EB process with electric coupling of the nucleus is forbidden. We used Weisskopf approximation in calculating η and obtained $\eta = 6.6\times10^{-14}$ (with $E_\gamma = 3.5$ eV) and $\eta = 3.1\times10^{-14}$ (with $E_\gamma = 2.4$ eV). The ratio of the reduced nuclear matrix elements of electric quadrupole and magnetic dipole processes, $B(E2)/B(M1)$, increases with increasing nucleon number [12], so it seems reasonable that the realistic values of η may be increased by two orders of magnitude compared to their Weisskopf-approximated ones. Our numerical results are collected in Table I. We deal with two possibilities for the energy separation of the nuclear levels: $\Delta E = 3.5$ eV or $\Delta E = 2.4$ eV. In the case of $\Delta E = 3.5$ eV and nuclear $M1$ transition, the γ decay is more probable than the EB process. Moreover, the contribution of the $E2$ channel to the direct decay is negligible. In the case of $\Delta E = 2.4$ eV the $M1$ channel will dominate again, but now the rate of the EB process is about twice the rate of the direct γ channel. In this case, IR photons, which were not registered in the experiments [2–5] as yet, are emitted in the EB process.

In the following we return to the experimental studies [2–5,7]. It has turned out that the lines around 3.5 eV in the observed optical spectra are mostly due to the N_2 luminescence caused by energetic α particles [5]. The detected signal around 2.4 eV was at first presumed as being due to the decay of the thorium isomer through the EB channel [3]. Indeed, regarding the experimental status quo and the misleading theoretical conclusions of Karpeshin *et al.* [8], one could conclude that the 2.4 eV signal might originate from the EB decay of $^{229}Th^m$. However, it was proved convincingly [7] that the observed broad signal about 2.4 eV [3,5] is

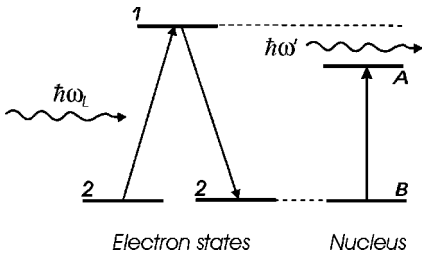

FIG. 3. Energy scheme of the laser-driven inverse electronic-bridge process of ^{229}Th. The electron shell is in the ground state (state 2) initially. A laser of photon energy $\hbar\omega_L$ induces a resonant electron transition to state 1. State 1 can decay by the inverse electronic bridge process, exciting the nucleus and emitting a photon of energy $\hbar\omega'$ in the process.

likely due to uranyl luminescence. As a conclusion, we can say that no traces of the isomeric decay were found in the experiments.

Regarding the disturbing effects, arising mainly from luminescence caused by α decay of ^{233}U, we suggest a different method for the observation of the energy separation of the nuclear doublet [10]. At the first step of the measurement a laser of photon energy $\hbar\omega_L$ resonantly excites an atomic electron of ^{229}Th with the nucleus in the ground state. (As we need Th in the ground state instead of the metastable one, the target Th nuclei, populated in ^{233}U decay, can be enriched enough and can be separated chemically from uranium.) Then in an inverse EB process, the excited electron returns to its ground state while the nucleus becomes excited and a photon of energy $\hbar\omega' = \hbar\omega_L - \hbar\omega_{AB}$, corresponding to the energy difference between the electronic and nuclear transitions, is emitted (see Fig. 3). It may happen, however, that the electron being in excited state 1 decays to an other electronic state (let us call it level 3 of energy E_3 and suppose that $E_3 - E_2 > \hbar\omega_{AB}$, E_2 is the energy eigenvalue of the electronic ground state). Then the electron can excite the nucleus starting the inverse electronic-bridge process from level 3, as well. In such a process a photon of $\hbar\omega' = E_3 - E_2 - \hbar\omega_{AB}$ is emitted. Measuring the optical signal ($\hbar\omega'$) in coincidence with the tuned laser pulse, it is expected that one would catch the traces of ^{229}Th^m better. Moreover, parallel laser irradiation of a control Th sample prepared from other isotopes of Th can help to find the signal of the above inverse EB processes. In general, the thorium exists as part of a molecular compound the energy eigenvalues of which are unknown. It is also an advantage of the above proposed inverse EB method that it is applicable in any case in which the Th compound has discrete electronic energy levels. The use of the control sample makes unnecessary the precise knowledge of the energy level scheme, as the extra lines, that appear in the case of the ^{229}Th compound sample due to the inverse EB process, are all shifted with $\hbar\omega_{AB}$.

This work was partly supported by the Hungarian National Science Research Found (OTKA) under number T031825.

[1] C.W. Reich and R.G. Helmer, Phys. Rev. Lett. **64**, 271 (1990); R.G. Helmer and C.W. Reich, Phys. Rev. C **49**, 1845 (1994).
[2] G.M. Irwin and K.H. Kim, Phys. Rev. Lett. **79**, 990 (1997).
[3] D.S. Richardson, D.M. Benton, D.E. Evans, J.A.R. Griffith, and G. Tungate, Phys. Rev. Lett. **80**, 3206 (1998).
[4] S.B. Utter, P. Beiersdorfer, A. Barnes, R.W. Lougheed, J.R. Crespo López-Urrutia, J.A. Becker, and M.S. Weiss, Phys. Rev. Lett. **82**, 505 (1999).
[5] R.W. Shaw, J.P. Young, S.P. Cooper, and O.F. Webb, Phys. Rev. Lett. **82**, 1109 (1999).
[6] F.F. Karpeshin, I.M. Band, M.B. Trzhaskowskaya, and B.A. Zon, Phys. Lett. B **282**, 267 (1992).
[7] J.P. Young, R.W. Shaw, and O.F. Webb, Inorg. Chem. **38**, 5192 (1999).
[8] F.F. Karpeshin, I.M. Band, M.B. Trzhaskowskaya, and A. Pastor, Phys. Rev. Lett. **83**, 1072 (1999).
[9] F.F. Karpeshin, I.M. Band, M.B. Trzhaskowskaya, and M.A. Listengarten, Phys. Lett. B **372**, 1 (1996).
[10] P. Kálmán and T. Keszthelyi, Phys. Rev. C **49**, 324 (1994).
[11] H.A. Bethe and E.E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms*, Handbuch der Physik, Vol. XXXV, Atoms 1 (Springer, Berlin, 1957).
[12] J.M. Eisenberg and W. Greiner, *Nuclear Theory* (North-Holland, Amsterdam, London, 1970), Vol. 1, Chap. 1.2.2.; Vol. 2, pp. 102 and 105.