Excitation of the Isomeric ^{229m}Th Nuclear State via an Electronic Bridge Process in ^{229}Th^{+}

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We consider the excitation of the nuclear transition ^{229g}Th-^{229m}Th near 7.6 eV in singly ionized thorium via an electronic bridge process. The process relies on the excitation of the electron shell by two laser photons whose sum frequency is equal to the nuclear transition frequency. This scheme allows us to determine the nuclear transition frequency with high accuracy. Based on calculations of the electronic level structure of Th^{+} which combine the configuration-interaction method and many-body perturbation theory, we estimate that a nuclear excitation rate in the range of 10 s^{-1} can be obtained using conventional laser sources.

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The nuclear transition between the low-energy isomeric state and the ground state in the ^{229}Th nucleus is very interesting due to the possibility to build a very precise nuclear clock and its high sensitivity to a hypothetical temporal variation of the fundamental constants [1,2]. The value of the transition frequency is known from γ spectroscopy [3–5] with an uncertainty that is many orders of magnitude higher than the presumed linewidth. Presently, the most precise value of the transition energy is 7.6(5) eV [5], placing the transition in the vacuum-ultraviolet spectral range. Further investigations are therefore needed in order to allow a direct optical excitation and detection of the line and to gradually increase the spectroscopic resolution to the level adequate for an optical clock [1].

Radiative transitions in nuclei are generally impeded by the small size of the nucleus in comparison to the wavelength of the radiation, especially for higher multipole orders. Forbidden transitions in γ decay are often accompanied by internal conversion, i.e., the release of the nuclear excitation energy by ejection of an electron from the shell. The enhancement of γ-decay rates through the excitation of shell electrons to higher-lying *bound* states has been the subject of extensive theoretical studies, and the term ''electronic bridge'' (EB) was introduced in this context [6]. The inverse process is known as nuclear excitation by electron transition. It was first discussed by Morita [7] and demonstrated experimentally for the 80.9 keV nuclear transition in ^{197}Au [8].

Since the energy of the nuclear transition between the isomeric ^{229m}Th state and the ground ^{229g}Th state is in the same range as that of excited states of the outer-shell electrons, the electronic environment may have a significant influence on the nuclear transition rate in this case [9]. Decay by internal conversion is not possible for positively charged thorium ions because the ionization potential of Th^{+} and of all higher charge ions exceeds the nuclear excitation energy. This makes ^{229}Th ions an ideal test case for EB processes in various electronic environments.

In a recent article [10] we have studied the effect of atomic electrons on the nuclear transition from the isomeric ^{229m}Th state to the ground ^{229g}Th state in ^{229}Th^{+} due to an EB process. Here we consider the process of the *excitation* of the nuclear $g \to m$ transition by an EB process that is driven by a two-photon excitation of the electron shell. Taking advantage of the dense electronic level structure of Th^{+}, we suggest a new experiment to study the resonant laser excitation of the nuclear ^{229g}Th-^{229m}Th transition. Atomic units ($\hbar = |e| = m_e = 1$) are used unless noted otherwise.

Experimental scheme.—The EB process considered in this work can be represented by the Feynman diagram in Fig. 1. In the following we assume a resonant character of the process. For this reason we take into account only one Feynman diagram given by Fig. 1 which mainly contributes to the probability of the process. Other diagrams which can be obtained from this one by permutations of the photon lines will be neglected. We suppose that the initial state i is the electronic ground state and the initial and final electronic states are the same. Further we assume a two-photon excitation of high-lying states k. For the first excitation at ω_1, it seems convenient to use a continuous laser tuned to the transition from the Th^{+} ground state $(6d^2 7s)J = 3/2$ to the $(6d7s7p)J = 5/2$ state at 24 874 cm^{-1} [11]. This electric-dipole ($E1$) transition is known as the strongest

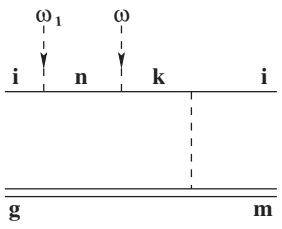

FIG. 1. Two-photon electronic bridge process. The single and double solid lines relate to the electronic and the nuclear states, respectively. The dashed lines are the photon lines.

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emission line in the Th^+ spectrum and it can be excited efficiently in collisionally cooled ion clouds in a radio frequency trap [12]. As a result, only the $(6d7s7p)J = 5/2$ state needs to be accounted for in the sum over intermediate states n in Fig. 1. We also note that the monitoring of the fluorescence resulting from excitation of the $(6d^27s)J = 3/2 \rightarrow (6d7s7p)J = 5/2$ transition allows us to detect the excitation of $^{229}Th^+$ ions from the nuclear ground state to the isomeric state, making use of the differences between the hyperfine structure characteristics of electronic transitions in $^{229m}Th^+$ and in $^{229g}Th^+$ [1].

The second excitation step with frequency ω gives rise to the excitation of higher-lying states k. Their decay to the ground state is accompanied by the nuclear $g \rightarrow m$ transition and leads to the appearance of isomeric Th^+ ions in the electronic ground state. For this excitation scheme, energy conservation implies that $\omega_1 + \omega = \omega_N$, where ω_1 and ω are the frequencies of the incident photons, and $\omega_N = E_m - E_g$ is the nuclear transition frequency as determined by the difference between the energies of the isomeric state E_m and the ground state E_g.

This excitation scheme offers the prospect to determine the nuclear transition frequency ω_N with the accuracy afforded by high-resolution laser spectroscopy. In an experiment based on this scheme, one would use a widely tunable laser source to produce narrow-bandwidth radiation with variable frequency ω. If the laser frequency is scanned over the nuclear resonance, the probability of excitation to the isomeric state exhibits a resonance peak. In an ion-trap experiment with collisionally cooled $^{229}Th^+$ ions, the width of the resonance is determined essentially by Doppler broadening and by the combined linewidth of the employed laser sources. If one assumes saturated excitation to the state n and a fixed detuning of ω relative to the electronic transitions $n \rightarrow k$, the probability of the EB process shown in Fig. 1 is proportional to the spectral intensity of the laser field at ω.

Based on the result of Ref. [5] we assume that the most interesting range of the electronic excitations is around 7.6 eV $\approx 61\,300$ cm^{-1}. Following this assumption, we expect that in the sum over the intermediate states k the atomic energy levels lying between 60 000 and 64 000 cm^{-1} will give the predominant contribution to the probability of the EB process. Unfortunately, these energy levels are not yet identified experimentally. Therefore all the following results are based on *ab initio* calculations. As we noted in [10], the achieved accuracy of calculations of the high-lying states of Th^+ is at the level of several percent. This is not sufficient to reliably predict the resonance enhancement occurring in the EB process. Hence, the experimental identification of the energy levels should be considered as the next step towards realizing the method considered here.

Because at present very accurate calculations are not needed, we will make one more assumption simplifying the calculations. We assume that only one intermediate state n, the state at 24 874 cm^{-1}, contributes to the probability of the EB process and that 100% population of this state can be achieved. As a result, the frequency ω_1 is assumed to be fixed at 24 874 cm^{-1} and the process which we discuss in the following can be described by the diagram represented by Fig. 2, where the state at 24 874 cm^{-1} is denoted as t and is considered as the initial state.

Calculation.—Figure 2 shows an EB process that relies on the absorption of an incident photon. As we mentioned in Ref. [13], an EB process of this type can be effectively treated as a ''generalized'' electric-dipole transition from the initial to the final state. If the incident radiation with spectral intensity I_ω is isotropic and unpolarized, the relation between the probabilities W_{ab} of a spontaneous transition $a \rightarrow b$ and $W^{\rm in}_{ba}$ of the corresponding induced transition $b \rightarrow a$ is given by [14]

$$W^{\rm in}_{ba} = W_{ab} \frac{4\pi^3 c^2}{\omega^3} I_\omega. \tag{1}$$

The spontaneous EB process can be formally described by the mirror image of Fig. 2 with an outgoing photon arrow. The general formalism was discussed in detail in two recent articles [10,13]. Assuming the resonance character, the expression for the probability of the spontaneous EB process $\Gamma_{\rm EB}$ can be written as [10]

$$\Gamma_{\rm EB} \approx \frac{4}{9}\left(\frac{\omega}{c}\right)^3 \frac{|\langle I_g||\mathcal{M}_1||I_m\rangle|^2}{(2I_m+1)(2J_t+1)} G_2, \tag{2}$$

where $\mathcal{M}_1$ is the magnetic-dipole nuclear moment and $|I_g\rangle = 5/2^+$ [633] and $|I_m\rangle = 3/2^+$ [631] are the ground and the isomeric nuclear state, respectively, given in their Nilsson classification. J_i is the electron total angular momentum of the initial state and $\omega = \omega_N - \omega_1$ is the frequency of an absorbed photon. The explicit expression for the coefficient G_2 was derived in [13] and reads as

$$G_2 = \sum_{J_k} \frac{1}{[J_k]} \left| \sum_{\gamma_s} \frac{\langle \gamma_i J_i \| \mathcal{T}_1 \| \gamma_s J_k\rangle \langle \gamma_s J_k \| D \| \gamma_t J_t\rangle}{\omega_{si} - \omega_N + i\Gamma_s/2} \right|^2, \tag{3}$$

with $\omega_{si} \equiv \varepsilon_s - \varepsilon_i$. Here $[J_k] \equiv 2J_k + 1$, ε_l denotes the energy of level l, $\mathcal{T}_1$ is the electronic magnetic-dipole hyperfine coupling operator, D is the electric-dipole

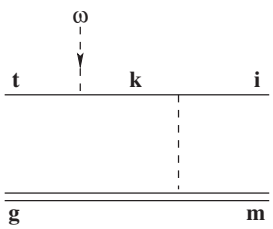

FIG. 2. Simplified Feynman diagram of the considered electronic bridge process. Here it is assumed that the first excitation step leads to efficient population of the state $(6d7s7p)J = 5/2$ at 24 874 cm^{-1}, which is denoted as t.

moment operator, and γ_s encapsulates all other electronic quantum numbers. The explicit expressions for the matrix elements of the operators $\mathcal{T}_1$ and D are given in Ref. [13]. The possible values of the total angular momentum J_k are determined by the selection rules of the operators $\mathcal{T}_1$ and D. In our case $J_i = 3/2$ and $J_t = 5/2$ and, correspondingly, $J_k = 3/2$ and $5/2$.

In the following we will use the dimensionless quantity β_{M1} introduced in Ref. [13] and defined as the ratio of the probability $\Gamma_{\rm EB}$ to the probability of the spontaneous $M1$ radiative nuclear $m \to g$ transition Γ_N:

$$\beta_{M1} = \frac{\Gamma_{\rm EB}}{\Gamma_N} \approx \left(1 - \frac{\omega_1}{\omega_N}\right)^3 \frac{G_2}{3(2J_t + 1)}. \tag{4}$$

We use the method of calculation described in detail in Ref. [10]. We consider Th^+ as an ion with three valence electrons above the closed-shell core $[1s^2, \ldots, 6p^6]$ and employ the CI + MBPT approach combining the configuration-interaction (CI) method in the valence space with many-body perturbation theory (MBPT) for core-polarization effects [15]. At the first stage we solved Dirac-Hartree-Fock (DHF) equations [16] in V^{N-3} approximation and then we determined the $5f$, $6d$, $7p$, $7s$, and $8s$ orbitals from the frozen-core DHF equations. The virtual orbitals were found with the help of a recurrent procedure [17]. The one-electron basis set included $1s$–$18s$, $2p$–$17p$, $3d$–$16d$, and $4f$–$15f$ orbitals on the CI stage.

We formed the configuration spaces allowing all possible (up to triple) excitations from the $6d^27s$ configuration (for the even states) and from the $5f7s^2$ configuration (for the odd states) to the $7s$–$13s$, $7p$–$12p$, $6d$–$11d$, and $5f$–$10f$ shells. It allows us to take into account most completely the configuration interaction for all considered states.

The energies and the wave functions are determined from the eigenvalue equation in the model space of the valence electrons $H_{\rm eff}(E_p)|\Phi_p\rangle = E_p|\Phi_p\rangle$. The effective Hamiltonian is defined as $H_{\rm eff}(E) = H_{\rm FC} + \Sigma(E)$, where $H_{\rm FC}$ is the relativistic three-electron Hamiltonian in the frozen-core approximation and $\Sigma(E)$ is the energy-dependent core-polarization correction. We also introduce an effective electric-dipole operator $D_{\rm eff}$ and an operator $(\mathcal{T}_1)_{\rm eff}$ acting in the model space of valence electrons. These operators were obtained within the relativistic random-phase approximation (RPA) [18,19].

Results and discussion.—In the following, we assume that the value of ω_N is between 60 000 and 64 000 $\rm cm^{-1}$, which corresponds to the range 7.4–7.9 eV, and suppose that the main contribution to G_2[see Eq. (3)] comes from intermediate states lying in this range. In [13] we discussed that the largest value of β_{M1} is expected if the initial and intermediate states k (see Fig. 2) are connected by an $E1$ transition. Then the states t and k are of opposite parity and we have to consider the transition $(6d7s7p, J = 5/2) \xrightarrow{E1} k \xrightarrow{\mathcal{T}_1} (6d^27s, J = 3/2)$.

Using Eq. (3) we obtain

$$G_2 \approx \sum_{J_k=3/2}^{5/2} \frac{1}{[J_k]} \sum_{\gamma_n} \frac{R_{n,J_k}}{(\omega_{ni} - \omega_N + i\Gamma_n/2)^2}, \tag{5}$$

where the quantity R_{n,J_k} is determined as

$$R_{n,J_k} \equiv |\langle 6d^27s, J = 3/2\|\mathcal{T}_1\|\gamma_n J_k\rangle\langle\gamma_n J_k\|D\|6d7s7p, J = 5/2\rangle|^2. \tag{6}$$

As follows from Eq. (4), the equation for β_{M1} reads as

$$\beta_{M1} \approx \frac{1}{18}\left(1 - \frac{\omega_1}{\omega_N}\right)^3 G_2 \approx 0.012 \times G_2, \tag{7}$$

where we took into account that the quantity $\omega_1/\omega_N \simeq 0.4$ if ω_N is between 60 000 and 64 000 $\rm cm^{-1}$.

High-lying energy levels of Th^+ in the range from 60 000 to 64 000 $\rm cm^{-1}$ were determined theoretically in [10]. In Table I we list the values of the coefficients R_{n,J_k} found for the even-parity states in the frame of the CI + MBPT + RPA approximation. The good correspondence between experimental and theoretical levels at lower energies makes us confident that the level structure is complete for the considered electron configurations. We expect that there will be more levels from other configurations but that the coefficients R for configurations with multiple electron excitations are smaller than the dominant ones calculated here.

Using Eq. (5), the tabulated coefficients R_{n,J_k}, and energy values we can find G_2 and β_{M1} for a given value of ω_N. The values of G_2 and β_{M1} depend critically on the position of ω_N relative to the electronic levels. This is indicated in Fig. 3, which shows the variation of G_2 with ω_N in the considered wave number range. As seen from this figure, $G_2 \geq 800$ if ω_N lies within the range of the electronic energy levels listed in Table I. For $\omega_N = 7.6$ eV [5],

TABLE I. Calculated energy levels (see text) in the range from 60 000 to 64 000 $\rm cm^{-1}$ and coefficients $R_{n,J}$. Δ_n is the difference between the energies of the excited state and the ground state. The notation $y[x]$ means $y \times 10^x$.

n	J	Δ_n[a] ($\rm cm^{-1}$)	$R_{n,J}$ (a.u.)
$5f6d7p$	3/2	60 287	3[−4]
$6d7s8s$	5/2	60 416	3[−2]
$5f6d7p$	5/2	60 462	2[−1]
$6d7s8s$	3/2	61 763	2[−3]
$5f^26d$	5/2	61 996	5[−4]
$6d^28s$	5/2	62 345	1[−5]
$6d7s8s$	3/2	62 927	9[−4]
$6d^27d$	5/2	63 308	2[−2]
$6d^27d$	3/2	63 381	4[−3]
$6d^27d$	3/2	63 729	3[−4]
$6d^28s + 6d7s8s$	5/2	63 955	3[−5]

[a]Reference [10].

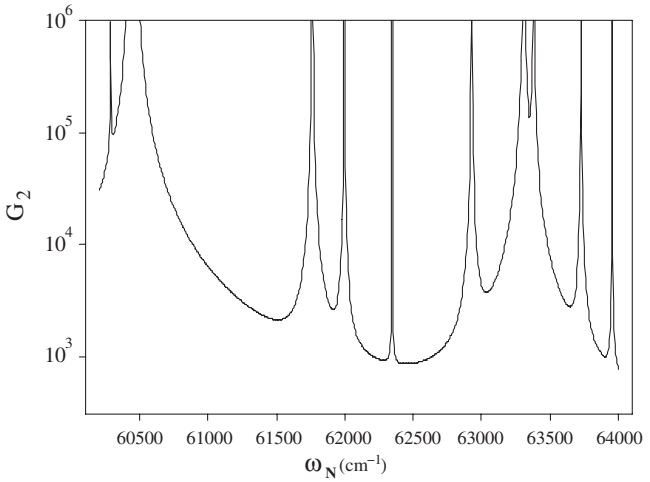

FIG. 3. Dependence of the coefficient G_2 [see Eq. (5)] on the nuclear transition frequency ω_N as calculated on the basis of the data listed in Table I.

the $(5f6d7p)J = 5/2$ state at the calculated value 60 462 cm^{-1} yields the main contribution to the EB process and one obtains $G_2 \sim 2700$ and $\beta_{M1} \sim 30$. When G_2 can be calculated on the basis of spectroscopically determined energy levels, it is likely that the dependence of G_2 on ω_N will look somewhat different from Fig. 3. But we expect that the EB excitation rate depends on a small number of dominant channels or even only a single one and that the typical minimum value of the coefficient G_2 will remain at the same order of magnitude ($\sim 10^3$).

The excitation probability to the isomeric state due to resonant laser excitation with $\omega_1 + \omega = \omega_N$ can be estimated as follows. We assume that radiation at ω is produced by a laser source which emits pulses with 10 mJ energy and with a spectral width of $\Delta\omega = 2\pi \times 3$ GHz at a repetition rate of 30 Hz. These characteristics can be achieved, for example, with commercially available frequency-doubled dye lasers as they are used in many laboratory applications. With focusing to a beam cross section of 0.1×0.1 mm^2, the resulting time-averaged spectral intensity is $I \approx 1.5 \times 10^{-3}$ (W/m^2) s. Taking into account Eqs. (1) and (4) and assuming $\beta_{M1} \approx 30$ and the value $\Gamma_N \simeq 6.6 \times 10^{-4}$ s^{-1} [13], we find that a ^{229}Th$^+$ ion is excited to the isomeric state with a probability in the range of $W_{\mathrm{EB}}^{\mathrm{in}} \approx 10$ s^{-1}. A significantly larger excitation rate can be expected if a suitable electronic transition frequency happens to be very close to ω_N. An interaction time of $(W_{\mathrm{EB}}^{\mathrm{in}})^{-1} \approx 0.1$ s would be sufficient to transfer a substantial part of the ensemble of trapped ^{229}Th$^+$ ions to the isomeric state and to detect this by the change of the fluorescence photon flux resulting from the excitation at ω_1 (see above). If the effective resonance width for the excitation to the isomeric state is dominated by the spectral width of the radiation at ω as assumed above, for one fixed setting of ω, a frequency interval $\Delta\omega = 2\pi \times 3$ GHz is probed in a time of 0.1 s. The uncertainty range of ± 0.5 eV [5] in ω_N corresponds to a frequency interval $\Omega = 2\pi \times 242$ THz so that a number of $\Omega/\Delta\omega \sim 10^5$ 3-GHz intervals must be probed. Thus, we estimate the scan time as $\Omega/(\Delta\omega W_{\mathrm{EB}}^{\mathrm{in}}) \sim 10^4$ s.

Conclusion.—We have suggested a new experimental scheme to excite the nuclear ^{229g}Th-^{229m}Th transition in Th$^+$ ions and to accurately determine its frequency. The scheme relies on an electronic bridge process that is driven by two incident laser photons whose sum frequency is resonant with the nuclear transition frequency. Using our previous calculations of the electronic energy level structure of Th$^+$ in the experimentally relevant energy range [10], we have estimated the probability of the investigated two-photon electron bridge process. Assuming that the nuclear transition energy is close to 7.6 eV, we find that a nuclear excitation probability in the range of 10 s^{-1} can be obtained with only moderate laser power and bandwidth requirements.

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