Laser-driven inverse electronic bridge process: An optical tool for determining low-energy separation of nearby nuclear states

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(Received 25 February 1992; revised manuscript received 1 June 1992)

An optical method to determine the low-energy separation (1 ± 4 eV) of nearby nuclear states of ^{229}Th is presented. It is shown that the Coulomb field of a deexciting electron of the ^{229}Th atom can cause transition between the near-lying nuclear states if the atom emits a photon with an energy equal to the difference between the energies of the electron and nuclear transitions. It is also predicted that the process can be enhanced through induced emission by applying a laser of resonant photon energy. The transition probabilities per unit time of the spontaneous and the laser induced processes are given.

PACS number(s): 23.20.Nx, 23.20.Lv, 32.30.Jc, 27.90.+b

The energy separation D of two intrinsic states at the ground state of ^{229}Th nucleus takes on the value $D = 1 \pm 4$ eV (but in any event it is certainly less than 10 eV) [1]. In the present paper we deal with the possibility of modification of the transition between these states by an inverse electronic bridge process. (The electronic-bridge process was observed a few years ago [2].) In contrast to the common belief that lasers of optical or UV frequencies that are available nowadays are not appropriate to modify nuclear transitions [3–4], it will be shown below that, not questioning this statement, due to the smallness of its transition energy there is a chance to modify the above-mentioned nuclear process [5,6].

In the case of the doublet nuclear states of ^{229}Th we suggest studying a laser-driven inverse electronic-bridge process. It is supposed that a laser (or lasers) excites (or excite) the atomic electron to a state of angular momentum defined by the selection rules in the next deexcitation phase. During the electronic deexcitation the excitation of the nucleus and the emission of a photon take place simultaneously. The energy of the photon is then equal to the difference of the electronic and nuclear transition energies (Fig. 1). The nuclear quadrupole transition with the photon emission in the dipole regime impose the condition $j_1 + j_2 \geq 3$ on the sum of initial and final electronic angular momenta j_1 and j_2. The electron excitation mechanism is not discussed here. It is supposed that the electron has already reached the initial electronic state. Since the electronic deexcitation initiating a nuclear excitation is accompanied by a photon emission of definite energy, a new line appears in the optical spectrum of the laser excited ^{229}Th sample. By measuring the energy (i.e., the angular frequency ω_0) of this new line the nuclear transition energy $\hbar\omega_{ba}$ can be determined as $\hbar\omega_{ba} = \hbar\omega_{12} - \hbar\omega_0$, where $\hbar\omega_{12}$ is the electron transition energy and $\hbar\omega_0$ is the photon energy of the new line observed.

Once the value of ω_0 is known the rate of the process can be increased by applying a laser of frequency $\omega_L = \omega_0$ as the laser field of frequency resonant with the spontaneously outcoming optical photon induces the emission of the photon.

In recent papers [7,8] similar processes were studied with the important difference that no outcoming photon was considered in them. In Refs. [7,8] it is the excitation process of the nuclear isomeric state which is mainly traced and which can come off owing to the finite width of the final electronic state. In the above papers the calculations were carried out near resonances which allows for retaining only a few terms from the infinite series constituting the S-matrix element.

In this paper we try to show the consequences of the emission of a resonant photon and handle the infinite series of the S matrix in a different way—without restricting the calculations to one or two resonant terms—resulting in an estimation of a compact form. We treat the process in the simplest way. We use a one-electron, hydrogenlike Hamiltonian

$$H_e = -\tfrac{1}{2}\hbar^2\nabla^2/\kappa - Ze^2/r$$

for the electron, where κ is the rest mass of the electron, e is the elementary charge, and Z is the proton number of the nucleus.

The initial and final electronic states are

$$|1(t)\rangle = \varphi_1(\mathbf{r})e^{-1\epsilon_1 t}$$

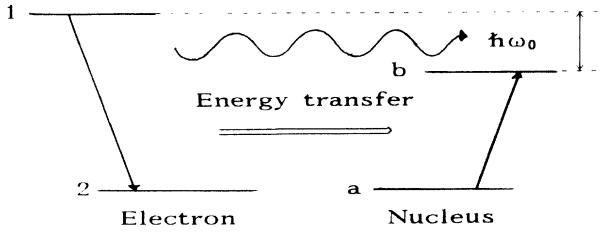

FIG. 1. Energy scheme of the inverse electronic-bridge process of ^{229}Th.

and

$$|2(t)\rangle = \varphi_2(\mathbf{r})e^{-i\varepsilon_2 t},$$

where $\varphi_1 = R_{n_1 l_1} Y_{l_1 m_1}$ and $\varphi_2 = R_{n_2 l_2} Y_{l_2 m_2}$ with R_{nl} and Y_{lm} denoting the radial and angular parts of the electronic states of hydrogenic type in spherical coordinates. $E_1 = \hbar\varepsilon_1$ and $E_2 = \hbar\varepsilon_2$ are the energy eigenvalues of the states, respectively.

The nuclear Hamiltonian is denoted by H_N whose detailed form is not needed in the present study. We assume that $|a\rangle$ and $|b\rangle$ are two eigenstates of H_N with the usual time evolution $|a(t)\rangle = |a\rangle e^{-i\varepsilon_a t}$ and $|b(t)\rangle = |b\rangle e^{-i\varepsilon_b t}$ with $E_a = \hbar\varepsilon_a$ and $E_b = \hbar\varepsilon_b$ being the corresponding energy eigenvalues.

The interaction of the electron and the quantized electromagnetic field is represented by

$$H_1 = e\mathbf{r}\cdot\mathbf{E}_q(t). \tag{1}$$

$\mathbf{r}$ stands for the electron coordinate, $\mathbf{E}_q(t)$ is the electric field strength of the quantized radiation field, which in dipole approximation can be expressed in the usual way in terms of the photon annihilation and creation operators $a_{\mathbf{k}\nu}$ and $a^\dagger_{\mathbf{k}\nu}$ as

$$\mathbf{E}_q(t) = i\sum_{\mathbf{k},\nu}\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2}\varepsilon_\nu(a_{\mathbf{k}\nu}e^{-i\omega t} - a^\dagger_{\mathbf{k}\nu}e^{i\omega t}). \tag{2}$$

ε_ν determines the state of polarization, $\hbar\omega$ and $\mathbf{k}$ are the energy and wave number of the photon, and V is the volume of normalization.

For the description of the electron-nucleus interaction a simple Hamiltonian

$$H_2 = \frac{Ze^2}{r} - \sum_{p=1}^{Z}\frac{e^2}{|\mathbf{r}-\mathbf{x}_p|} \tag{3}$$

is used [9], where $\mathbf{x}_p$ for $p = 1,\ldots,Z$ are proton coordinates. Thus the total Hamiltonian of the system is $H = H_e + H_1 + H_2 + H_N$.

The rate of the process can be calculated in the above model by ordinary perturbation calculation to the second order (Fig. 2). The corresponding S-matrix elements are

$$S^A_{fi} = \sum_n \frac{\langle f|H_1^\dagger|n,0\rangle\langle n,0|H_2|i\rangle}{-i\hbar^2(\omega_{ba}+\omega_{n1})}2\pi\delta(\omega_{ba}+\omega-\omega_{12}) \tag{4}$$

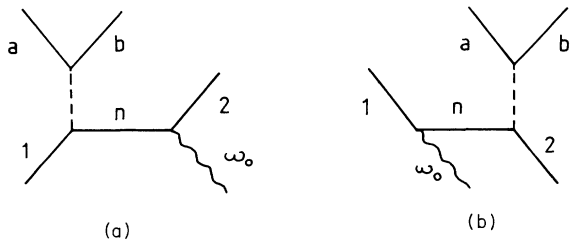

FIG. 2. The two graphs of the inverse electronic-bridge process. The dashed line represents the Coulomb interaction between the electron cloud and the nucleus.

and

$$S^B_{fi} = \sum_n \frac{\langle f|H_2|n,1\rangle\langle n,1|H_1^\dagger|i\rangle}{-i\hbar^2(\omega+\omega_{n1})}2\pi\delta(\omega_{ba}+\omega-\omega_{12}), \tag{5}$$

where the superscripts refer to the graphs of Figs. 2(a) and 2(b). Here $H_1^\dagger = e\mathbf{r}\cdot\mathbf{E}_q^\dagger$ with

$$\mathbf{E}_q^\dagger = -i\sum_{\mathbf{k},\nu}\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2}\varepsilon_\nu a^\dagger_{\mathbf{k}\nu}$$

and $|f\rangle = |2\rangle|b\rangle|1\omega,\mathbf{k},\nu\rangle$, $|i\rangle = |1\rangle|a\rangle|0\omega,\mathbf{k},\nu\rangle$, $|n,0\rangle = |n\rangle|b\rangle|0\omega,\mathbf{k},\nu\rangle$, and $|n,1\rangle = |n\rangle|a\rangle|1\omega,\mathbf{k},\nu\rangle$ are the final, initial, and intermediate states of the electron-nucleus-photon system. $|2\rangle \equiv \varphi_2(\mathbf{r}), |1\rangle \equiv \varphi_1(\mathbf{r}), |n\rangle \equiv \varphi_n(\mathbf{r})$ are electronic eigenstates and $|1\omega,\mathbf{k},\nu\rangle$ and $|0\omega,\mathbf{k},\nu\rangle$ are photon number states of angular frequency ω, wave vector $\mathbf{k}$, and polarization ν.

Now the following changes are made in the total S-matrix element $S_{fi} = S^A_{fi} + S^B_{fi}$. As $\omega_{ba}+\omega_{n1} = \omega_{ba} - \omega_{12} + \omega_{n2} = \omega_{n2} - \omega_0$ the argument of the Dirac delta allows the $\omega_{n2} - \omega_0 = \omega_{n2} - \omega$ substitution in Eq. (4). Using the identities

$$(\omega+\omega_{n1})^{-1} = \omega^{-1} - \omega^{-1}\omega_{n1}(\omega+\omega_{n1})^{-1} \tag{6}$$

and

$$(\omega_{n2}-\omega)^{-1} = -\omega^{-1} + \omega^{-1}\omega_{n2}(\omega_{n2}-\omega)^{-1} \tag{7}$$

and applying the formula of completeness

$$\sum|n\rangle\langle n| = \delta(\mathbf{r}-\mathbf{r}\prime) \tag{8}$$

in S_{fi} the terms originated from ω^{-1} and $-\omega^{-1}$ cancel. Using $\omega_{\beta a}\mathbf{r}_{\alpha\beta} = i\mathbf{p}_{\alpha\beta}/\kappa$ [10] in the two remainders

$$S_{fi} = \left(\sum_n \frac{\langle f|ie\mathbf{p}\mathbf{E}_q^\dagger/\kappa|n,0\rangle\langle n,0|H_2|i\rangle}{-i\hbar^2(\omega_{n2}-\omega)\omega} + \sum_n \frac{\langle f|H_2|n,1\rangle\langle n,1|ie\mathbf{p}\mathbf{E}_q^\dagger/\kappa|i\rangle}{-i\hbar^2(\omega+\omega_{n1})\omega}\right) \times 2\pi\delta(\omega_{ba}+\omega-\omega_{12}). \tag{9}$$

Here $\mathbf{p} = -i\hbar\nabla$. Applying again Eqs. (6) and (7) in Eq. (9) and using Eq. (8) in the terms that contain ω^{-1} and $-\omega^{-1}$, these two terms give a part of the S-matrix element as

$$S_{fi} = (i\hbar)^{-1}\langle f|H_\alpha^\dagger|i\rangle 2\pi\delta(\omega_{ba}+\omega-\omega_{12}), \tag{10}$$

where

$$H_\alpha^\dagger = -\boldsymbol{\alpha}^\dagger\cdot\boldsymbol{\nabla}\sum_{p=1}^{Z}\frac{e^2}{|\mathbf{r}-\mathbf{x}_p|} \tag{11}$$

with

$$\boldsymbol{\alpha}^\dagger = \frac{e}{\kappa}i\sum_{\mathbf{k},\nu}\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2}\frac{1}{\omega^2}a^\dagger_{\mathbf{k}\nu}\boldsymbol{\varepsilon}_\nu, \tag{12}$$

which can be considered at least as a good underestimation of the whole S-matrix element for the following reasons.

Applying the formula $\omega_{\beta a}\mathbf{p}_{\alpha\beta} = -i(\nabla Ze^2/r)_{\beta a}$ [10] in the two remainders and using Eqs. (6)–(8) in them the terms containing ω^{-1} and $-\omega^{-1}$ cancel. It can be shown with the aid of the identity

$$\omega_{\alpha\beta}(\nabla Ze^2/r)_{\beta a} = [\nabla Ze^2/r, H_e]_{\beta a}$$

that the order of magnitude of the other two terms is less by $e^2/\hbar c$ than the term given by Eq. (10). Here it is worth mentioning that when computing the transition probability per unit time the ω dependence will disappear in consequence of the Dirac delta, which imposes the $\omega = \omega_0$ substitution. As $\hbar\omega_{12}$ and $\hbar\omega_0$ both are the order of magnitude of eV, Eq. (10) may be the leading term. In any case it is certainly true that when deriving Eq. (10) from Eq. (9) and using Eqs. (6) and (7) the subtraction due to the ω^{-1} and $-\omega^{-1}$ terms led to an underestimation of Eq. (9).

The application of Eqs. (6) and (7) may lead to an incorrect result if the frequency ω_0 is nearly resonant to an electronic transition or if it becomes very small, i.e., if $\omega_{12} - \omega_{ba} \sim 0$. In these cases the approximate form Eq. (10) of the S-matrix element also fails. But, practically, this situation can be avoided with choosing an appropriately different initial electronic state.

If we restrict ourselves to the point nucleus ($x_p < r$) approximation then the expressions

$$\sum_{p=1}^{Z} \frac{e^2}{|\mathbf{r} - \mathbf{x}_p|} = e^2 \sum_{L,M} \frac{4\pi}{2L+1} Y_{LM}(\hat{\mathbf{x}}_p) Y^*_{LM}(\hat{\mathbf{r}}) \frac{\sum_p x_p^L}{r^{L+1}} \tag{13}$$

and

$$\nabla(Y^*_{LM}(\hat{\mathbf{r}})r^{-(L+1)}) = \sum_{\mu=1,0,-1} \varepsilon_\mu (L+1)^{1/2}(2L+1)(-1)^L \begin{pmatrix} L+1 & 1 & L \\ -M-\mu & \mu & M \end{pmatrix} \times Y_{L+1,-M-\mu} r^{-(L+2)} \tag{14}$$

can be applied in Eq. (11). ($\mu = 1, -1, 0$ correspond to the two transverse and the longitudinal states of polarization [11].) For convenience the following notations are introduced for the different factors appearing in the S matrix:

$$\left\langle b \left| \sum_p e x_p^L Y_{LM}(\hat{\mathbf{x}}_p) \right| a \right\rangle = e r_0^L (-1)^{j_b - m_b} \langle b \parallel \hat{Q}_L \parallel a \rangle \begin{pmatrix} j_b & L & j_a \\ -m_b & M & m_a \end{pmatrix}, \tag{15}$$

where $\langle b \parallel \hat{Q}_L \parallel a \rangle$ is the reduced matrix element of the multipole moment of order L, M between the nuclear states $|a\rangle$ and $|b\rangle$ expressed in Weisskopf units, j_b, m_b, j_a, and m_a denote the total angular momentum quantum numbers and the magnetic quantum numbers of the final and initial nuclear states, respectively. L is determined by the nuclear transition, $r_0 = R_0 A_0^{1/3}$ is the nuclear radius with A_0 being the nucleon number. We also introduce

$$\hat{I}^{n_1 l_1}_{L,n_2 l_2} = a_B^{L+2} \int R_{n_1 l_1}(Z_{\text{eff}}(n_1), r) \times R_{n_2 l_2}(Z_{\text{eff}}(n_2), r) r^{-L} dr, \tag{16}$$

with a_B denoting the Bohr radius and Z_{eff} the effective nuclear charge of the state.

With the help of these formulas and notations in Eq. (11), taking $|S_{fi}|^2$ and dividing it by $2\pi\delta(0) = T$, the transition probability per unit time W_{fi} of the spontaneous process is obtained as

$$W_{fi} = W^0_{fi} |\langle b \parallel \hat{Q}_L \parallel a \rangle|^2 |\hat{I}^{n_1 l_1}_{L,n_2 l_2}|^2 \times (2l_2 + 1) \begin{pmatrix} l_1 & l_2 & L+1 \\ 0 & 0 & 0 \end{pmatrix}^2 \tag{17}$$

with

$$W^0_{fi} = \left(\frac{r_0}{a_B}\right)^{2L} \frac{e^6}{\kappa^2 c^3 a_B^4} \frac{8\pi^2(L+1)}{(2L+1)(2j_a+1)} \frac{1}{\hbar\omega_0} \times \sum_{\substack{\mu=1,-1 \\ -L \le M \le L}} \begin{pmatrix} L+1 & 1 & L \\ -M-\mu & \mu & M \end{pmatrix}^2, \tag{18}$$

where $\hbar\omega_0 = \hbar(\omega_{12} - \omega_{ba})$ is the energy of the outgoing photon and $\hbar\omega_{12} = \hbar(\varepsilon_1 - \varepsilon_2)$ and $\hbar\omega_{ba} = E_b - E_a$ are the electronic and the nuclear transition energies, respectively. In obtaining Eqs. (17) and (18), the following manipulations have also been made. The result of the angular integral of the product of three spherical harmonics has been substituted. In order to average over the initial and to sum up with respect to the final states the addition theorem of the $3j$ symbols has been used twice. The sum over $\mathbf{k}$ is changed for an integral in the phase space of the outcoming photon.

By measuring the energy of the emitted photon, the spacing of the nuclear energy levels can be determined as $\hbar\omega_{ba} = \hbar\omega_{12} - \hbar\omega_0$. At the same time, the uncertainty concerning the question of the ground state could also be resolved: in case of $\hbar\omega_{ba} < 0$ (i.e., $\hbar\omega_{12} < \hbar\omega_0$) the state of angular momentum $\frac{3}{2}^+$ is the ground state [1].

Knowing the value of ω_0 the process can be accelerated by applying a laser of frequency $\omega_L = \omega_0$. The laser of photon frequency resonant with the outcoming optical photon can induce its emission. The transition probability per unit time of this induced, inverse electronic-bridge process can be obtained repeating the calculation with the following modifications.

First, it is assumed that the electron is excited above the initial state and reaches it by spontaneous deexcitation. Under these circumstances the initial, excited

electronic state has an energy distribution

$$\rho(\varepsilon_1) = \gamma_1\{2\pi[(\varepsilon_1 - \varepsilon_{10})^2 + \gamma_1^2/4]\}^{-1},$$

where γ_1 is the width of the state and $\hbar\varepsilon_{10}$ is the central energy of the broadened level. This energy distribution has to be used as a weight factor in an integral with respect to ε_1 when we average over the initial electronic states. The integral can be calculated with the aid of the Dirac delta obtained in time integration. If the laser has an angular frequency fulfilling the condition $\varepsilon_{10} = \varepsilon_2 + \omega_{ba} + \omega_L$ then the integral results $2\gamma_1^{-1}\pi^{-1}$.

Secondly, in the case of induced emission the photon parts of the initial and final states are modified to $|N\omega_L, \mathbf{k}_0, \nu_0\rangle$ and $|(N+1)\omega_L, \mathbf{k}_0, \nu_0\rangle$, respectively, resulting in a factor $N+1 \sim N$, and since $\mathbf{k}_0$ and ν_0 are fixed the factor $\pi^{-2}V\omega_L^2c^{-3}$ brought in by the phase-space integral is not necessary.

As the laser flux $\Phi = Nc/V$ ($\Phi = I/\hbar\omega_L$, I is the laser intensity) the transition probability per unit time W_{fi}^L of the laser induced inverse electronic-bridge process can be given as

$$W_{fi}^L = (2\pi)^{-1}W_{fi}\Phi\lambda_L^2\gamma_1^{-1}, \tag{19}$$

where λ_L is the wavelength of the laser radiation.

Now we give a numerical estimation of the rate of the spontaneous process [see Eqs. (17) and (18)]. The initial and final angular momenta are $j_a = \frac{5}{2}^+$ and $j_b = \frac{3}{2}^+$, so they can be coupled by a quadrupole transition, i.e., $L = 2$. Since the thorium shell has $6d^27s^2$ configuration it seems to be an acceptable assumption that the initial state is obtained by exciting a $6d$ electron into an $8p$ state. Thus $l_1 = 1$ and $l_2 = 2$, hence $(2l_2 + 1) = 5$, $\begin{pmatrix} 1 & 2 & 3 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{3}{35}$, and

$$\sum_{\substack{\mu=1,-1 \\ -2\le M\le 2}} \begin{pmatrix} 3 & 1 & 2 \\ -M-\mu & \mu & M \end{pmatrix} = \frac{2}{3}.$$

Furthermore, Eq. (18) gives $W_{fi}^0 = 8.7 \times 10^{-5}(\hbar\omega_0)^{-1}$ s^{-1}, where $\hbar\omega_0$ is measured in electronvolts and $|\hat{I}_{2,62}^{81}|^2 = 4.49 \times 10^{-4}$ with $Z_{\text{eff}}(8) = 2.169$ ($n_1 = 8, l_1 = 1$) and $Z_{\text{eff}}(6) = 4.649$ ($n_2 = 6, l_2 = 2$). With these numerical values, Eq. (17) gives

$$W_{fi} = 1.7 \times 10^{-8}|\langle b \parallel \hat{Q}_L \parallel a\rangle|^2(\hbar\omega_0)^{-1} \text{ s}^{-1}.$$

$|\langle b \parallel \hat{Q}_L \parallel a\rangle|^2$ is expected to have a value between 10^{-2} and 10^2 [12].

The ^{229}Th has a half-life $\tau = 2.3\times10^{11}$ s [13]. A sample of N_0 ^{229}Th atoms has a radioactivity $A = 8.1\times10^{-23}N_0$ in Ci units. Thus, for a sample with $N_0 = 10^{18}$ we have $A = 81\mu$Ci. It can be expected that a few percent of the atoms (e.g., 1%) can be populated in the appropriate electronic initial state. In this case the rate of photon emission R can be estimated as

$$R = 1.7 \times 10^{8}|\langle b \parallel \hat{Q}_L \parallel a\rangle|^2(\hbar\omega_0)^{-1} \text{ s}^{-1}.$$

This is certainly larger than 10^5 s^{-1}, which seems to be an observable value. Moreover, since the state $|b\rangle$ (of angular momentum $\frac{3}{2}^+$) can have a much smaller half-life than the state $|a\rangle$, the process can result in an increase of the activity of the sample.

According to the calculations of Ref. [7] for the activity due to state $|b\rangle$ with a supposed half-life of 10 days and an irradiation time 10^2–10^3 s on $N_0 = 10^{13}$ ^{229}Th nuclei $A = 10^5$–10^7 Bq can be obtained. This activity corresponds to an induced rate 10^8–10^{10} s^{-1} of transition $a \to b$. Converting this rate to the case $N_0 = 10^{16}$, which corresponds to our choice $N_0 = 10^{18}$ with the assumption that 1% of the atoms is in the appropriate initial state, we obtain 10^{11}–10^{13} s^{-1} for their rate. Although this process is somewhat different from the one studied by us, it is comforting to see that its induced rate moves over and in the same orders of magnitude as the R obtained here.

Finally,, we can conclude that the laser-driven inverse electronic-bridge mechanism can offer a method to determine the energy difference of closely lying nuclear states of ^{229}Th and a possibility of enhancement of a radioactive decay rate.

The authors are indebted to S. Varró for the critical reading of the manuscript. This work was partly supported by the MTA-Soros Foundation (Budapest–New York) and the Hungarian National Science Research Fund (OTKA) under numbers T-4459 and E-012199.

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