# Excitation of low-lying isomer level of the nucleus $^{229}$Th by optical photons

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An efficiency approaching 100% can be achieved in the pumping of a low-lying isomer state of the nucleus $^{229}$Th, with an energy $\leqslant 5$ eV, by the beam from an optical-range laser when the mechanism of an inverse electron bridge is used.

In early 1990, a group from the Idaho National Engineering Laboratory reported[1] that the energy ($E_{\rm is}$) of the first excited state of the nucleus $^{229}$Th was $1 \pm 4$ eV. The predominant decay mechanism for this level depends on the value of $E_{\rm is}$ in the neutral thorium atom. This predominant mechanism may be either direct nuclear emission, if $E_{\rm is} \leqslant 1.5$–2 eV, or an electron bridge, if $E_{\rm is}$ lies between 2 and 5 eV (Ref. 2). In the former case the lifetime of the low-lying isomer, $T^{\rm is}_{1/2}$, is a matter of days or more, while in the latter case it should be more than 10 min (except, of course, in the case of a resonant coincidence of $E_{\rm is}$ with the energy of one of the atomic M1 transitions).[2]

In the present letter we are interested in the excitation of an isomer state of the nucleus $^{229}$Th by laser light by the mechanism of an inverse electron bridge[3] (this mechanism is also called "Compton excitation" of a nucleus[4]). Figure 1a is a direct diagram of this process. For efficient excitation of this isomer level during the direct application of the laser light to the nucleus, we need to know the energy of the nuclear transition within the width of the laser line. In the scheme proposed below for exciting the nucleus $^{229}$Th by the mechanism of an inverse electron bridge, all that is required is to tune the laser light to the wavelengths of well-known atomic transition. In this case, even if there is a significant difference between the energies of the atomic and nuclear transitions, the cross section for excitation of the nucleus is large enough to allow efficient pumping of the isomer state.

The cross section for the excitation of a nucleus in an inverse electron bridge reaches a maximum when the energy of the laser photons, $\omega_L$ (we are using a system of units with $\hbar = c = 1$), is equal to $\omega_{\rm in}$, i. e., the energy of the atomic transition from the initial state to an intermediate state (Fig. 1a). The real part of one of the energy denominators in the amplitude for the direct diagram vanishes in this case. In the exchange diagram, which differs from Fig. 1a in that the two electron–photon vertices are interchanged, there is no resonance at $\omega_L = \omega_{\rm in}$. Correspondingly, the contribution of this resonance to the excitation of the nucleus is small in this case. The exchange diagram can thus be ignored. In the single-level approximation with $\omega_L = \omega_{\rm in}$, we can therefore write the cross section as follows:

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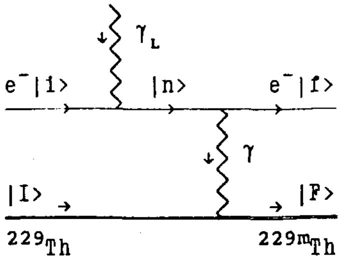


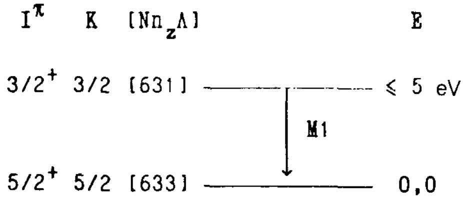


FIG. 1. a—Direct diagram of the excitation of a nucleus by laser photons by the mechanism of an inverse electron bridge; b—ground and isomer states of the nucleus $^{229}$Th (K is the rotation number, and $[Nn_z\Lambda]$ are the quantum numbers in the Nilsson model).

$$\sigma = \frac{\lambda_L^2}{\pi}\left(1+\frac{\Gamma_f}{2\Gamma_n}\right)\frac{\Gamma_A(i\to n; M(E)1)}{\Gamma_n}\frac{E_{int}^2(\omega_N; M1)}{(\epsilon_n-\epsilon_f-\omega_N)^2+\Gamma_n^2(1+\Gamma_f/2\Gamma_n)^2}. \quad (1)$$

Here $\lambda_L$ is the wavelength of the laser photons; $\omega_N$ is the energy of the nuclear transition; $\Gamma_{n,f}$ are the total widths of the corresponding atomic levels [the initial state $|i\rangle = 6d^27s^2(^3F_2)|$, with an energy[5] $\epsilon_i = -6.08$ eV, is a stationary state; the width of the nuclear level is being ignored in comparison with $\Gamma_n$ ]; $\Gamma_A(i\to n; M(E)1)$ is the probability for the $M(E)1$ atomic transition from the initial state to an intermediate state; and $E_{\rm int}^2$ is the square of the absolute value of the energy of the interaction of the electron current $j_{nf}^\mu(\vec r) = -e\bar\psi_f(\vec r)\gamma^\mu\psi_n(\vec r)$, and the nuclear current $J_{IF}^\nu(\vec R) = e\Psi_F^+(\vec R)\hat J^\nu\Psi_I(\vec R)$, averaged over the initial states and summed over the final states, in second-order perturbation theory $[\int d^3\vec r d^3\vec R j_{nf}^\mu(\vec r)D_{\mu\nu}(\omega;\vec r-\vec R)J_{IF}^\nu(\vec R)$, where $D_{\mu\nu}(\omega;\vec r-\vec R) = -g_{\mu\nu}\exp(\omega|\vec r-\vec R|)/|\vec r-\vec R|]$. If the width of the laser line, $\Delta\omega_L$ is considerably greater than the width of the radiative transition, $\Gamma_A(i\to n)$ expression (1) must also be multiplied by $\Gamma_A(i\to n)/\Delta\omega_I$. The quantity $E_{\rm int}^2$ is calculated from[6]

$$E_{int}^2(\omega_N; M1) = \frac{1}{4}\Gamma_A(\omega_N; n\to f; M1)\,\Gamma_N^r(\omega_N; I\to F; M1)\left(1+\frac{1}{\delta^2}\right), \quad (2)$$

where $\Gamma_N^r$ is the probability for the radiative nuclear transition, and $\delta = \mathrm{Re}[\mathcal{M}_{M1}(\omega_N)]/\mathrm{Im}[\mathcal{M}_{M1}(\omega_N)]$ is an analog of the ratio (well known in the theory of internal conversion) of the real part and imaginary part of the atomic matrix element

$$\mathcal{M}_{M1}(\omega) = (\kappa_n+\kappa_f)\int_0^\infty dr\, r^2 h_1^{(1)}(\omega r)[g_n(r)f_f(r)+f_n(r)g_f(r)]. \quad (3)$$

The latter quantity was calculated numerically. The mean field and the electron wave functions were determined in a relativistic version of the Hartree–Fock–Slater method by the program of Ref. 7. The quantities $g(r)$ and $f(r)$ in (3) are the major and minor components of the radial wave functions of the electron; $h_1^{(1)}(\omega r)$ are the Hankel function of index 1; and $\kappa = (l - j)(2j + 1)$, where $l$ and $j$ are the orbital and total angular momenta of the electron. In the calculation of $\Gamma_N^r$ we used the most probable value of the reduced probability for the nuclear $M1$ transition illustrated in Fig. 1b: $B(M1; I \to F) \sim 10^{-2}$ Weisskopf units.[2,8] As a result, the typical value of $E_{\rm int}^2$ turns out to be about $10^{-9}$–$10^{-10}$ eV$^2$. One can determine $E_{\rm int}$ experimentally and compare the result with theoretical predictions. This is an important point, since the parameter $E_{\rm int}^2/\Delta^2$ which appears in (1) is a basic characteristic of the NEAT process (nuclear excitaiton during atomic transitions). Opinion in the literature is divided on the relative probability for this process (see the analysis and citations in Ref. 6).

The cross section for the excitation of the isomer calculated from Eqs. (1)–(3) for $\omega_L = 0.3$–$5$ eV turns out to be $10^{-19}$–$10^{-20}$ cm$^2$ for a difference $\Delta = 1$ eV between the energies of the atomic transition $\omega_{nf} = \epsilon_n - \epsilon_f$ and the nuclear transition $\omega_N$. Two cases are important. (1) First, there is the case of the transition $i \to n$–$M1$, with the final state of the electron being the same as the initial state (an "elastic" inverse electron bridge). The quantity $\Gamma_A(i \to n; M1)$ can be one or two orders of magnitude smaller than $\Gamma_n$. This situation is realized in the case of, for example, $|n\rangle = 6d^3 7s(^5F_3)$ with $\epsilon_n = -5.15$ eV (Ref. 5). (2) Second, there is the transition $i \to n$–$E1$, with the final state of the electron being different from the initial state. The ratio $\Gamma_A(i \to n; E1)/\Gamma_n$ is on the order of one in this case. One possible realization is $|n\rangle = 6d\,7s^2 7p(^3F_2)$ with $\epsilon_n = -4.74$ eV and $|f\rangle = 5f\,6d\,7s^2(^3F_2)$ with $\epsilon_f = -5.06$ eV (Ref. 5). By moving "up" in terms of the energies of the excited atomic states, we can easily find[9] suitable atomic levels which should be stimulated if $E_{is}$ turns out to be larger than 1 eV. The value found for $\sigma$ is a lower estimate. Because of the relatively high density of excited atomic levels,[5,9] the actual value of $\Delta$ may be considerably less than 1 eV.

By definition, the excitation efficiency $\zeta$ is equal to the ratio of the number of $^{229m}$Th isomer nuclei which are formed to the number of thorium atoms exposed to the light. This efficiency is found from the expression ($\zeta \simeq \phi_L \sigma \tau$, where $\phi_L$ is the flux density of laser photons, and $\tau$ is the exposure time. A 0.1-W laser with $\omega_L$ up to 5 eV, with 1 $\mu g$ of pure $^{229}$Th over an area of 1 cm$^2$ on a substrate, in a layer $\simeq 10$ Å thick, can put essentially all the nuclei in the sample in the excited state over $\tau \simeq 10^2$–$10^3$ s. As a result, the activity of the isomer is $10^9$ Bq, even at $T_{1/2}^{\rm is} \simeq 10$ days. As a result, a "laser" experiment to pump, and to determine the lifetime and energy of, an anomalously low-lying isomer level is realistic; in fact, it would be rather simple in comparison with other possible measurement schemes.

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