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DECAY ACCELERATION OF ISOMERS BY X-RAY LASER

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We analyzed the possibility of the two-step triggering of nuclear isomers by short X-ray laser pulses via an excited intermediate level. The estimations of the decay acceleration are performed for the 6^- isomer of ^{84}Rb in the field of future European X-ray laser.

Keywords: X-ray laser, nuclear isomer, triggering, energy release, decay accelaration.

Introduction

A few dozens nuclei have excited metastable (isomeric) states with long lifetimes. Different attempts were undertaken in recent years to release great energy of these isomers, affecting them by any external fields. Collins et al. [1] irradiated the 16^+ isomer of ^{178}Hf (whose half-life is 31 y and energy 2.446 MeV) by X-rays, and found acceleration of its decay by few per cent. These authors described the acceleration as a two-step process, when the nucleus, absorbing X-ray photon, goes first to the higher level $|e\rangle$ and then decays through the chain of levels to the ground state. However, the nature of such intermediate state $|e\rangle$ was not identified so far. These results have not been confirmed in the recent experiments with more strong radiation sources (see the review [2]).

The Coulomb interaction of the nucleus with surrounding electrons can significantly change the nuclear decay rate. Therefore intensive studies have been devoted to NEET (Nuclear Excitation by Electronic Transition) - reciprocal process with respect to the bound electron conversion. Trying to solve the Hf problem, Karpeshin et al. [3] studied the NEET accompanied by emission of photon.

It was also discussed similar process, called NEEC (Nuclear Excitation by Electronic Capture) when free electrons of the continuous spectrum are captured into unoccupied atomic orbits, giving off their energy to the nucleus [4]. Possible triggering of isomers by means of NEEC was discussed in [5]. Alternative process was analyzed by Gosselin et al. [6], who regarded the Coulomb excitation of nuclei in the process of inelastic scattering of free electrons in hot plasma.

The decay of bare isomeric nuclei via an excited intermediate level induced by optical lasers has been studied in [7]. More realistic chance for the isomeric triggering is associated with the appearance of X-ray lasers on free electrons (XFEL). Advantages compared to optical lasers in this case are obvious. First, one can match the radiation frequency with the nuclear transition frequency. Moreover, the probability for the transition of multipolarity L is proportional to $(kR)^{2L+1}$, where k is the wave vector of the photon and R is the radius of the nucleus [8]. Hence, the gain in the transition probability for the X-ray photon with the energy $E \propto 1$ keV compared to the optical photons with $E \propto 1$ eV becomes very large.

Bürvenich et al. [9] analyzed the electric dipole transitions between the ground and excited nuclear states, induced by the X-ray laser pulse. For this aim the muster equations for the level populations have been solved numerically. These equations contained the Rabi frequency, explicitly depending on time. The next paper of Pálffy et al. [10] dealt with similar calculations for laser-induced transitions of arbitrary multipole order.

The problems of the coherent nuclear optics and triggering of isomers are very intriguing. Therefore it is important to derive analytic formulas for the de-excitation probability of nuclei exposed to X-ray laser field, which would explicitly show the role of all relevant physical parameters. Our paper is devoted to this aim.

Semiclassical approach

In semi-classical approach the nucleus together with the field of γ quanta are described quantum-mechanically, while the laser radiation is treated as a classical electromagnetic wave packet. Let the wave be linearly polarized and its vector potential

$$\mathbf{A}(r,t) = \mathbf{A}_0(t)\cos(\mathbf{kr} - \omega_k t). \quad (1)$$

We approximate the time-dependent envelope $\mathbf{A}_0(t)$ by the Gaussian function

$$\mathbf{A}_0(t) = \mathbf{A}_0 \exp\left[-t^2 / 2\tau^2\right], \quad (2)$$

where parameter τ characterizes the pulse duration.

The unperturbed Hamiltonian of the system nucleus in the laser field + quantized electromagnetic field is

$$\hat{H}_0 = \hat{H}_n + \hat{H}_{rad}, \tag{3}$$

where $\hat{H}_n$ and $\hat{H}_{rad}$ define respectively the Hamiltonian of the nucleus and the Hamiltonian of the quantized electromagnetic field. The latter in the Coulomb gauge is given by

$$\hat{H}_{rad} = \sum_{\boldsymbol{\kappa}} \sum_{p=\pm 1} \hat{a}^{+}_{\boldsymbol{\kappa}p} \hat{a}_{\boldsymbol{\kappa}p}, \tag{4}$$

where $\hat{a}^{+}_{\boldsymbol{\kappa}p}$ and $\hat{a}_{\boldsymbol{\kappa}p}$ are the creation and annihilation operators of the photon with the wave vector $\vec{\kappa}$ and circular polarization $\boldsymbol{\varepsilon}_p$. For brevity, we do not talk about the conversion electrons.

The total Hamiltonian

$$\hat{H} = \hat{H}_0 + \hat{V}_r + \hat{V}_f(t), \tag{5}$$

where $\hat{V}_r$ is the interaction operator of the nucleus with the quantized electromagnetic field and $\hat{V}_f(t)$ is the interaction operator of the nucleus with the laser field.

The first interaction is defined by well-known expression (see, e.g., [8]):

$$\hat{V}_r = -\frac{1}{c}\int d\mathbf{r}\mathbf{j}(\mathbf{r})\mathbf{A}(\mathbf{r}), \tag{6}$$

where $\mathbf{j}(\mathbf{r})$ is the flux density operator of the nucleus, $\mathbf{A}(\mathbf{r})$ is the vector potential operator of the quantized electromagnetic field.

Similarly, the interaction operator of the nucleus with the laser wave $\hat{V}_f(t)$ is defined by the same expression (6) with the operator $\mathbf{A}$ replaced by the classical potential (1). Using Eq. (1), we represent the operator $\hat{V}_f(t)$ as

$$\hat{V}_f(t) = -\frac{1}{2c}\left[\hat{j}_{\|}(k)e^{-i\omega(k)t} + \hat{j}_{\|}(-k)e^{i\omega(k)t}\right]A_0(t), \tag{7}$$

where

$$\hat{j}_{\|}(\mathbf{k}) = \mathbf{j}(\mathbf{k})\mathbf{e}, \tag{8}$$

is the product of the Fourier-transform of the nuclear flux density operator

$$\mathbf{j}(\mathbf{k}) = \int d\mathbf{r}\mathbf{j}(\mathbf{r})e^{i\mathbf{k}\mathbf{r}} \tag{9}$$

and the polarization vector $\mathbf{e} = \mathbf{A}_0(t)/A_0(t)$.

The projections of $\mathbf{j}(\mathbf{k})$ along the circular polarization vectors $\boldsymbol{\varepsilon}_{p=\pm1}$ are determined by the following expansion in multipoles:

$$\frac{1}{c}\hat{j}_p(\vec{k}) = \sqrt{2\pi}\sum_{L=1}^{\infty} i^L k^L \sqrt{\frac{(L+1)(2L+1)}{L}}\frac{1}{(2L+1)!!}\times$$
$$\times\sum_{m=-L}^{L} D^{L}_{pm}(\alpha,\beta,0)^{*}\left[M_m(EL) - ipM_m(ML)\right], \tag{10}$$

where $M_m(\lambda L)$ are the electric $(\lambda = E)$ and magnetic $(\lambda = M)$ multipole operators [11], $D^L_{pm}(\alpha,\beta,0) = e^{ip\alpha}d^L_{pm}(\beta)$ are the rotation matrices, depending on spherical angles β,α of the wave vector $\mathbf{k}$ of the laser wave with respect to the nuclear center-of-mass coordinate frame x, y, z.

We direct the axis x along the wave vector $\mathbf{k}$ and the axis z along the polarization vector $\mathbf{e}$. Then the orientation of the frame x', y', z' with the axis z' parallel to $\mathbf{k}$ is defined by the Eulerian angles $\alpha = 0$, $\beta = \pi/2$, $\gamma = 0$, while

$$\hat{j}_{\|}(\pm k) = (\hat{j}_1(\pm k) - \hat{j}_{-1}(\pm k))/\sqrt{2}. \tag{11}$$

The eigenfunctions and eigenvalues of the unperturbed Hamiltonian $\hat{H}_0$ obey the equation

$$\hat{H}_0\chi_b = E_b\chi_b. \tag{12}$$

Let at the initial moment $t = -T/2$ the system (nucleus + quantized electromagnetic field) be described by the wave function

$$\chi_a = |I_iM_i\rangle|0\rangle, \tag{13}$$

where the function $|I_iM_i\rangle$ describes the isomeric state of the nucleus and $|0\rangle$ the vacuum of the quantized field. The corresponding E_a equals the initial energy of the nucleus W_i.

The nucleus, absorbing X-ray photon, performs transition from $|i\rangle$ to the intermediate excited state $|e\rangle = |I_eM_e\rangle$ with the energy W_e (the corresponding wave function of the system will be $\chi_c = |e\rangle|0\rangle$). Then the nucleus, emitting γ quantum with the energy $E_\gamma = \hbar\omega_\gamma$, goes into the final state $|I_fM_f\rangle$, having the energy W_f. So the energy of such a final state $|b\rangle$ of the whole system equals $E_b = W_f + E_\gamma$.

We assume that the X-ray photon energy is close to the transition energy $E_0 = W_e - W_i$ and introduce the detuning parameter

$$\delta = E_0/\hbar - \omega_k. \tag{14}$$

Transition probability

The probability of finding the system at the moment $T/2$ in the final state $|b\rangle$ is given by [12]

$$P_b = \left|\delta_{ba} - \frac{2\pi}{\hbar} T_{ba}(\omega_{ab})\right|^2, \qquad (15)$$

where the transition frequency $\omega_{ab} = (E_a - E_b)/\hbar$, or with the above definitions

$$\omega_{ab} = (W_i - W_f)/\hbar - \omega_\gamma. \qquad (16)$$

The matrix for the two-step transition is [12]

$$T_{ba}(\omega_{ab}) = \sum_{M_e} \frac{\langle b|\hat{V}_r|c\rangle \tilde{V}_{ei}^f(\omega_{ab})}{W_i - W_e - \hbar\omega_{ab} + i\Gamma_e/2}, \qquad (17)$$

where Γ_e is the width of the intermediate (excited) level; $\tilde{V}_{ei}^f(\omega_{ab})$ denotes the Fourier-transform of the matrix element $\langle I_e M_e|\hat{V}_f(t)|I_i M_i\rangle$, which for the Gaussian pulse will be

$$\tilde{V}_{ei}^f(\omega) = -\frac{1}{2c} j_\parallel(\vec{k})_{ei} \frac{A_0}{2\sqrt{2\pi}} \tau e^{-(\omega+\omega_k)^2\tau^2/2}. \qquad (18)$$

After substitution of Eqs. (17) and (18) into Eq. (15) we have to average yet the probability P_b over the initial states and sum over all possible final ones $|b\rangle$. Using Eqs. (10) and (11) as well as standard relations

$$\sum_{M_i M_e} (I_i L M_i m | I_e M_e)(I_i L M_i m' | I_e M_e) =$$

$$= \left(\frac{2I_e + 1}{2L + 1}\right) \delta_{mm'} \qquad (19)$$

and

$$\sum_{m=-L}^{L} d_{mp}^L(\beta) d_{mp'}^L(\beta) = \delta_{pp'}, \qquad (20)$$

we find that

$$\frac{1}{2I_i + 1} \sum_{M_i, M_e} |c^{-1} j_\parallel(\mathbf{k})_{ei}|^2 = \left(\frac{2I_e + 1}{2I_i + 1}\right) \frac{\Gamma_{ei}^\gamma}{4k}, \qquad (21)$$

where Γ_{ei}^γ is the partial width of the radiative transition of arbitrary multipolatity from e to i.

Besides, note that time-dependence of the flux density of the Gaussian pulse is defined by the function

$$S(t) = S_0 e^{-t^2/\tau^2}, \qquad (22)$$

where the peak power

$$S_0 = \frac{ck^2}{8\pi} A_0^2. \qquad (23)$$

All this allows us to write down the probability of the isomeric triggering with emission of γ quantum in the form

$$P_f^\gamma = \left(\frac{2I_e + 1}{2I_i + 1}\right) \left(\frac{\pi\tau}{\hbar k}\right)^2 \frac{S_0}{\omega_k} \left(\frac{\Gamma_{ef}^\gamma \Gamma_{ei}^\gamma}{\Gamma_e}\right) I(\delta), \qquad (24)$$

where

$$I(\delta) = \frac{\Gamma_e}{2\pi} \int_{-\infty}^{\infty} dE_\gamma \frac{e^{-(\delta+\omega_\gamma-\omega_\gamma^{(0)})^2\tau^2}}{(E_\gamma - E_0')^2 + (\Gamma_e/2)^2}, \qquad (25)$$

and $E_0' = \hbar\omega_\gamma^{(0)}$ is the energy of transition from the excited level to the final one:

$$E_0' = W_e - W_f. \qquad (26)$$

The pulse duration τ is by several orders less than the typical lifetime of the excited level $\tau_n^e = \hbar/\Gamma_e$. Therefore the integrand in Eq. (25) is a product of the sharp peak due to the denominator and the exponent, smoothly dependent on ω_γ. Standard estimations give then

$$I(\delta) \approx e^{-\delta^2\tau^2}. \qquad (27)$$

Treating emission of the conversion electrons in the same manner, one obtains the complete triggering probability $P_f = (1 + \alpha_{ef}) P_f^\gamma$, where α_{ef} is the conversion coefficient for the transition $e \to f$. The induced decay occurs for a short time $\propto \tau$. Nevertheless, it is convenient to introduce the average triggering rate during the repetition period T:

$$\overline{w}_{ind}(i \to f) = P_f / T. \qquad (28)$$

Discussion

The physical meaning of the Eq. (24) becomes more clear, when we treat the laser pulse as a bunch of X-ray photons. Let us denote the number of all photons in this pulse, passing through unit square with the energies from $E = \hbar\omega$ to $E + \Delta E$, by $N(E)\Delta E$. Their energy distribution $N(E)$ is characterized by the squared Fourier-transform of the vector potential (1), i.e.,

$$N(E) \propto \exp\left[-(\omega - \omega_k)^2 \tau^2\right]. \qquad (29)$$

The total energy of the pulse per unit square amounts

$$E_{tot} = \sqrt{\pi} S_0 \tau. \qquad (30)$$

Integrating $N(E)$ and equating the result to the full number of photons $E_{tot} / \hbar\omega(k)$, we easily find the normalization factor in Eq. (29).

Let us introduce the average flux rate of photons, transmitting through unit square per unit energy, during the repetition period T :

$$\dot{N}(E) = N(E) / T. \qquad (31)$$

The resonant value of such flux rate at $E = E_0$ will be

$$\dot{N}(E_0) = \frac{S_0}{\hbar\omega(k)\Gamma_G}\left(\frac{\tau}{T}\right)e^{-\delta^2\tau^2}, \qquad (32)$$

where $\Gamma_G = \hbar / \tau$ stands for the bandwidth of the Gaussian pulse. In reality XFEL is characterized by much more wider bandwith Γ_L, so that for numerical estimations we have to replace in Eq. (32) Γ_G by Γ_L. Comparing Eq. (32) with Eqs. (25), (27) as $\tau << \tau_n^e$ we obtain the following average rate of the isomeric triggering, induced by the X-ray laser:

$$\overline{w}_{ind}(i \to f) = \left(\frac{2I_e + 1}{2I_i + 1}\right)\frac{\pi^2}{k^2}\dot{N}(E_0)F_R\Gamma_e, \qquad (33)$$

where the factor

$$F_R = \frac{(1+\alpha_{ef})R_{ei}R_{ef}}{[(1+\alpha_{ei})R_{ei} + \sum_f (1+\alpha_{ef})R_{ef}]^2} \qquad (34)$$

is expressed in terms of the branching ratios $R_{ei(f)}$ for the radiative transitions $e \to i(f)$ and the corresponding conversion coefficients $\alpha_{ei(f)}$.

The total width of the excited level Γ_e, entering into Eq. (33), is determined by its half-life:

$$\Gamma_e = \frac{\hbar \ln 2}{T_{1/2}^e}. \qquad (35)$$

Our Eq. (33) coincides with that derived in [13] for the two-step transition rate via an excited intermediate level in the case of isomeric triggering by an incoherent beam of X-rays. Note, however, that in [13] the two-step decay had been regarded as a process continuous in time, when the excitation of the intermediate level and its decay proceed simultaneously. But in our case a short laser pulse first stimulates the transition to the excited level, which then exponentially decays emitting γ-quantum. Therefore our Eq. (33) contains the averaged flux rate, while the corresponding result of [13] contains constant rate $dN(E_0)/dt$ (note that some our designations are slightly different from those of [13]).

Even small probability P_f may be associated with large acceleration of the isomeric decay during the repetition period of the pulses T. When T / τ_n^i and $\overline{w}_{ind}(i \to f)$ are much less than unity, this acceleration R is defined by the ratio of the induced decay rate to the spontaneous one $1/\tau_n^i$:

$$R = \sum_{f \neq i} \overline{w}_{ind}(i \to f)\tau_n(i). \qquad (36)$$

We estimated the triggering rate for the 6^- isomer ^{84}Rb, having the energy $W_i = 463.59$ keV and the half-life $T_{1/2}^e = 20.26$ m, assuming exact matching of the laser radiation to the nuclear transition from the isomeric state to the intermediate one $(\delta = 0)$. Using such parameters of future European XFEL as the duration of the pulse $\tau = 100$ fs, the repetition period $T = 2.5 \cdot 10^{-5}$ s, the peak power $S_0 = 6 \cdot 10^{15}$ W/cm^2 and the bandwidth 0.1% we got the induced decay probability $P_f = 2 \cdot 10^{-7}$ and the acceleration of the decay $R = 4$.

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ПРИСКОРЕННЯ РОЗПАДУ ІЗОМЕРІВ РЕНТГЕНІВСЬКИМ ЛАЗЕРОМ

О. Я. Дзюблик

Проаналізовано можливість двокрокового тригерування ядерних ізомерів короткими імпульсами рентгенівського лазера через збуджений проміжний рівень. Зроблено оцінки прискорення розпаду для 6^- ізомеру ^{84}Rb у полі майбутнього європейського рентгенівського лазера.

Ключові слова: рентгенівський лазер, виділення енергії, прискорення розпаду.

УСКОРЕНИЕ РАСПАДА ИЗОМЕРОВ РЕНТГЕНОВСКИМ ЛАЗЕРОМ

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Проанализирована возможность двухшагового тригерования ядерных изомеров короткими импульсами рентгеновского лазера через возбужденный промежуточный уровень. Сделаны оценки ускорения распада для 6^- изомера ^{84}Rb в поле будущего европейского рентгеновского лазера.

Ключевые слова: рентгеновский лазер, тригерование, выделение энергии, ускорение распада.

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