Pis'ma v ZhETF, vol. 92, iss. 3, pp. 152 – 156

Triggering of nuclear isomers by x-ray laser

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Submitted 25 May 2010

We studied the decay of nuclear isomeric states in the field of the x-ray laser. The laser pulses are described by the Gaussian wave packet of linearly polarized electromagnetic waves. At first stage the laser short pulse generates nuclear transition in the intermediate excited state, which afterwards decays into the final state with emission of γ quantum. Simple formulas are derived for the induced transition probability, which well correlate with known results, obtained previously for the incoherent x-ray radiation.

1. 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 – 5] 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 following experiments with more strong radiation sources (see the reviews [6, 7]). Theoretical estimations [7 – 10] were also negative, predicting too little probability of the hafnium triggering for the case, when the nucleus directly interacts with the radiation. Nevertheless, the question with role of the electronic shells remains still open.

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 [11, 12]. Collins et al. [5] assumed the NEET to be a realistic reason of the hafnium triggering observed by them. In more detail this question was discussed in [13]. Trying to solve the Hf problem, Izosimov [14] considered triggering of isomers via decay of autoionization states, which can arise in electronic shells during irradiation by x-rays, while Karpeshin et al. [15] studied the NEET accompanied by emission of photon.

It was also discussed similar process, called NEEC (Nuclear Excitation by Electronic Capture)(see, e.g., [16, 17]), when free electrons of the continuous spectrum are captured into unoccupied atomic orbits, giving off their energy to the nucleus. Possible realization of NEEC in hot plasma has been investigated in [18 – 20], and the triggering of isomers by means of NEEC in [21]. Alternative process was analyzed by Gosselin et al. [22], 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 [23 – 25], while coupling of the electronic shells to the laser field has been discussed earlier by Berger et al. [26]. 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 λL is proportional to $(kR)^{2L+1}$, where k is the wave vector of the photon and R is the radius of the nucleus [27]. Hence, the gain in the transition probability for the x-ray photon with the energy $E \sim 1$keV compared to the optical photons with $E \sim 1$eV becomes very large.

Bürvenich et al. [28] 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. [29] 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 deexcitation 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.

2. 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

$$\mathbf{A}(\mathbf{r},t) = \mathbf{A}_0(t)\cos(\mathbf{kr} - \omega(k)t). \tag{1}$$

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

$$A_0(t) = A_0 \exp\left[-t^2/2\tau^2\right]. \tag{2}$$

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 $\boldsymbol{\kappa}$ and circular polarization $\boldsymbol{\epsilon}_p$. 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.,[27]):

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

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

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

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

where

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

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

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

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

The laser pulses are repeated with the period T, so that the field interaction is a periodic function of time $V_f(t) = V_f(t+T)$ with $T \gg \tau$. This enables us to use here the results of the paper [30], obtained for quantum systems subject to periodic classical fields.

The projections of $\hat{\mathbf{j}}(\mathbf{k})$ along the circular polarization vectors $\boldsymbol{\epsilon}_{p=\pm 1}$ are determined by the following expansion in multipoles:

$$\frac{1}{c}\hat{j}_p(\mathbf{k}) = \sqrt{2\pi}\sum_{L=1}^{\infty} i^L k^L \sqrt{\frac{(L+1)(2L+1)}{L}}\frac{1}{(2L+1)!!},$$
$$\sum_{m=-L}^{L} D^L_{pm}(\alpha,\beta,0)^*\left[\hat{\mathcal{M}}_m(EL) - ip\hat{\mathcal{M}}_m(ML)\right], \tag{10}$$

where $\hat{\mathcal{M}}_m(\lambda L)$ are the electric ($\lambda = E$) and magnetic ($\lambda = M$) multipole operators (see, e.g., [31]), $D^L_{pm}(\alpha,\beta,0) = e^{ip\alpha}d^L_{pm}(\beta)$ 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

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

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 $|I_f M_f\rangle$, 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 photon energy $\varepsilon(k) = \hbar\omega(k)$ is close to the transition energy $E_0 = W_e - W_i$ and introduce the detuning parameter

$$\delta = E_0/\hbar - \omega(k). \tag{14}$$

3. Transition probability. The probability of finding the system at the moment $T/2$ in the final state $|b\rangle$ is given by [30]

$$P_b = \left|\delta_{ba} - \frac{2\pi}{\hbar}\mathcal{T}_{ba}(\omega_{ab})\right|^2, \tag{15}$$

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

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

The two-step transition matrix is [30]

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

where Γ_e is the width of the intermediate (excited) level, and $\tilde{V}_{ei}^f(\omega_{ab})$ 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

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

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

$$\sum_{M_i M_e} (I_i L M_i m|I_e M_e)(I_i L M_i m'|I_e M_e) = \left(\frac{2I_e+1}{2L+1}\right)\delta_{mm'} \tag{19}$$

and

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

we find that

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

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 energy flux density of the Gaussian pulse (1), (2) is defined by the function

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

where the peak power

$$S_0 = \frac{ck^2}{8\pi}A_0^2. \tag{23}$$

All this allows us to write down the probability of the isomer de-excitation by one pulse, accompanied by emission of γ quantum:

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

where the parameter $\Gamma_G = \hbar/\tau$ determines the width of the Gaussian wave packet (1), (2) in the energy representation,

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

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

$$E_0' = W_e - W_f. \tag{26}$$

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 (25) is a product of the sharp peak due to the denominator and the exponent, smoothly dependent on ω_γ. Standard estimations give then

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

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 $e \to f$. Then the average triggering rate during the repetition period T is

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

4. Pulse as a bunch of photons. The physical meaning of the formula (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 $\varepsilon = \hbar\omega$ to $\varepsilon + \Delta\varepsilon$, by $N(\varepsilon)\Delta\varepsilon$. Their energy distribution $N(\varepsilon)$

is characterized by the squared Fourier-transform of the wave (1), (2) i.e.,

$$N(\varepsilon) \sim \exp\left[-(\varepsilon - \varepsilon(k))^2/\Gamma_G^2\right]. \quad (29)$$

Integration of (22) gives the total energy of the pulse per unit square:

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

Integrating also $N(\varepsilon)$ and equating the result to the full number of photons $E_{tot}/\varepsilon(k)$, we easily find the coefficient in (29).

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

$$\bar{\dot{N}}(\varepsilon) = N(\varepsilon)/T. \quad (31)$$

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

$$\bar{\dot{N}}(E_0) = \frac{S_0}{\varepsilon(k)\Gamma_G}\left(\frac{\tau}{T}\right) e^{-\delta^2\tau^2}. \quad (32)$$

Comparing (32) with (25), (27) as $\tau << \tau_n^e$ we obtain the following average triggering rate of the isomer, induced by the x-ray laser:

$$\bar{w}_{ind}(i \to f) = \left(\frac{2I_e + 1}{2I_i + 1}\right) \frac{\pi^2}{k^2} \bar{\dot{N}}(E_0) F_R \left(\frac{\hbar \ln 2}{T_{1/2}^e}\right), \quad (33)$$

where $T_{1/2}^e$ is the half-life of the excited level, F_R denotes the factor

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

depending on the branching ratios $R_{ei(f)}$ for the radiative transitions $e \to i(f)$ and the corresponding IC coefficients $\alpha_{ei(f)}$.

5. Discussion. Our Eq.(33) is obtained in the approximation of moderate lasers, providing small transition probability, $P_f \ll 1$. Even such small probability P_f may be associated with large acceleration of the isomeric decay, defined by the ratio of the induced and spontaneous decay rates:

$$\mathcal{R} = \sum_{f \neq i} \bar{w}_{ind}(i \to f)\tau_n^i. \quad (35)$$

Eq.(33) coincides with the expression for the two-step transition rate, derived in [32] for the isomers exposed to an incoherent beam of x-rays. The coherence of the incident radiation can ensure new effects, such as the Rabi oscillations, only in the case of super-intense lasers. Note, however, that in [32] the induced deexitation had been regarded as a process continuous in time, when the excitation of the intermediate level and its decay proceed simultaneously. As a result, the formulas of [32] contain constant photon flux rate, whereas our Eq.(33) depends on the rate $\bar{\dot{N}}(\varepsilon)$, averaged over the period T.

In reality the laser bandwidth Γ_L much exceeds Γ_G. Therefore Γ_G in Eq.(33) is to be replaced by Γ_L.

Starting from Eqs.(33), we estimated the probability P_f for triggering of the 6^- isomer of ^{84}Rb, having the energy $W_i = 463.59$ keV and the half-life $T_{1/2}^i = 20.26$ m. The x-ray pulse is assumed to be matched exactly ($\delta = 0$) with the transition to the intermediate level 5^-, characterized by $W_e = 466.64$ keV and $T_{1/2} = 9$ ns [33]. These levels are coupled by highly converted M1 transitions. We employed the following parameters of the future European XFEL: $\hbar\omega(k) \leq 12.4$ keV, $\tau = 100$ fs, $T = 2.5 \cdot 10^{-5}$ s, the peak power $S_0 = 6 \cdot 10^{15}$ W/cm^2 and $\Gamma_L = 10^{-3}\hbar\omega(k)$ [29]. In this case the transition probability $P_f \approx 2 \cdot 10^{-7}$, that corresponds to acceleration of the isomeric decay $\mathcal{R} \approx 4$.

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