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Laser-assisted nuclear γ deexcitation by electronic-bridge process

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The recently observed process of nuclear γ deexcitation, called the electronic-bridge mechanism, is investigated in an intense laser field. In this process the nuclear excitation energy is transferred into an electronic excitation, which, in turn, results in the emission of an x-ray photon. The analysis of the process of deexcitation, whose energy somewhat differs from the energy difference of two bound states in the electronic shell shows that it can be drastically modified by an intense laser field if the condition of N-photon resonance is met. The transition probability per unit time of the laser-assisted process is derived for s states and the ratio η of this probability to that of the spontaneous γ decay is given for EL transitions. The expressions are also extended to the case of two laser beams of different photon energies but of a parallel state of polarization.

The first experimental observation of the "electronic-bridge mechanism" was made recently.[1] It is a third-order process that occurs in the following manner: after the nuclear deexcitation, one electron of the atom is excited and leaves the atom; at the same time an x-ray photon is also emitted. Thus the energy of the nuclear transition is converted into the energy of the outgoing electron and x-ray photon. The process is similar to internal conversion with the difference of the additional emission of an x-ray photon.

The laser-assisted internal conversion process was investigated in three recent articles,[2-4] which led to the following conclusion. The effect of laser radiation is essential in those processes in which at least one of the characteristic energies is comparable to the laser-photon energy.[5] It was shown in Refs. 3 and 4 that the internal-conversion process can be most easily modified near but below the threshold. In such a process the energy of the γ photon is less than the binding energy of the electron in the shell in question. If the magnitude of the energy defect is comparable to the laser-photon energy, then the process, which is originally energetically forbidden, can take place after absorbing the necessary number of laser photons.

In what follows we are investigating a special type of laser-assisted electronic-bridge mechanism. In the processes to be examined the energy of the γ transition is nearly resonant to one of the bound-bound electronic transitions but the energy misfit is equal to a small integer times the photon energy of the applied intense laser beam. The initial and final electronic states are the same. The three steps of the process are nuclear deexcitation, electron excitation into a dressed state, and electron transition to the initial state by x-ray emission. The intermediate electronic state, which is also a bound one, is strongly modified by the intense laser field[6] and, therefore, it is called a dressed state. We deal with resonant processes only, i.e., with those where the sum of the energy of the γ photon and that of a few laser photons is equal to one of the energies of the possible bound-bound electronic transitions.

The mechanism can be traced with the following simple model. The total Hamiltonian of the system

$$H=H_0+H_1+H_2+H_3\,, \tag{1}$$

where

$$H_0=-\frac{\hbar^2}{2\mu}\nabla^2-\frac{Ze^2}{r}+H_N\,, \tag{2}$$

$$H_1=e\mathbf{r}\cdot\mathbf{E}_L(t)\,, \tag{3}$$

$$H_2=\frac{Ze^2}{r}-\sum_{p=1}^{Z}\frac{e^2}{|\mathbf{r}-\mathbf{x}_p|}\,, \tag{4}$$

and

$$H_3=e\mathbf{r}\cdot\mathbf{E}_x(t)\,. \tag{5}$$

Here the following notation is used: μ is the rest mass of the electron, Z is the proton number of the nucleus and $\mathbf{x}_p$, $p=1,\ldots,Z$ denote their coordinates, $\mathbf{r}$ stands for the electron coordinate, H_N is the Hamiltonian of the nucleus. $\mathbf{E}_L(t)$ is the electric-field strength of the laser radiation, which is treated classically and has the form

$$\mathbf{E}_L(t)=E_0\hat{\mathbf{z}}\cos(\omega t)\,, \tag{6}$$

where E_0 and ω are the amplitude and the angular frequency of the laser radiation which is polarized linearly parallel to direction z. Finally,

$$\mathbf{E}_x(t)=i\sum_{\omega',\epsilon}\left(\frac{2\pi\hbar\omega_x}{V}\right)^{1/2}\epsilon(ae^{-i\omega_x t}-a^{\dagger}e^{i\omega_x t}) \tag{7}$$

is the operator of the quantized x-ray field. $\hbar\omega_x$ is the x-ray photon energy, ϵ determines the state of linear polarization, a and $a^{\dagger}$ are the annihilation and creation operators, and V is the volume of normalization. H_1 describes the laser-electron interaction, H_2 is responsible for internal conversion, i.e., for electron-nucleus interaction, and H_3 gives the interaction of the electron with the quantized electromagnetic field and leads to x-ray emission.

The calculations are made in electric-field gauge using the dipole approximation. The evolution of the many-electron system is approximated by the evolution of one electron, but the effect of the other electrons is taken into

account by using effective nuclear charges $Z_{\rm eff}(n)$ in the one-electron eigensolutions of H_0.

The motion of the electron due to the joint influence of the Coulomb and laser fields is described by the wave function Ψ, which can be approximately written for a given subshell of principal quantum number n as[6]

$$\Psi(n,n_1,n_2,m)=\Phi_{n_1n_2m}\sum_{N=-\infty}^{\infty}J_N(\lambda_{n_1n_2})\,e^{-i(E_n+N\hbar\omega)t/\hbar}, \tag{8}$$

where $\Phi_{n_1n_2m}$ is a hydrogen-type solution in parabolic coordinates and J_N denotes a Bessel function of the first kind, N is the number of absorbed or emitted laser photons, $E_n=\hbar\epsilon_n-i\hbar\gamma_n/2$ is the complex energy of the intermediate electronic state, and

$$\lambda_{n_1n_2}=\frac{\frac{3}{2}n(n_1-n_2)E_0ea_B}{Z_{\rm eff}\hbar\omega}. \tag{9}$$

Here n is the principal quantum number of the subshell, $n=n_1+n_2+|m|+1$, m is the magnetic quantum number, n_1 and n_2 are the parabolic quantum numbers,[7] and a_B is the Bohr radius. For the sake of simplicity the indices of $\lambda_{n_1n_2}$ will be omitted.

The electronic Green function of the laser-Coulomb problem can be approximately written as[6]

$$G(t_2-t_1)=-i\Theta(t_2-t_1)\sum_{n_1,n_2,m}|\Psi(n,n_1,n_2,m)\rangle\langle\Psi(n,n_1,n_2,m)|, \tag{10}$$

where Θ denotes the step function.

The S-matrix element governing the process

$$S_{fi}=(i\hbar)^{-2}\int_{t_0}^{t}dt_2\int_{t_0}^{t_2}dt_1\langle f(t_2)|H_3(t_2)|b(t_2)\rangle|0\omega_x\rangle G(t_2-t_1)\langle 0\omega_x|\langle b(t_1)|H_2|i(t_1)\rangle, \tag{11}$$

where

$$|b(t)\rangle=|b\rangle e^{-i\epsilon_b t} \tag{12}$$

and $|b\rangle$ denotes the space-dependent part of the nuclear final state (b).

$$|f(t)\rangle=\phi_0(\mathbf{r})e^{-i\epsilon_0 t}|b(t)\rangle|1\omega_x\rangle, \tag{13}$$

$$|i(t)\rangle=\phi_0(\mathbf{r})e^{-i\epsilon_0 t}|a(t)\rangle|0\omega_x\rangle, \tag{14}$$

are the final and initial states of the electron-nucleus x-ray system, respectively.

$$|a(t)\rangle=|a\rangle e^{-i\epsilon_n t} \tag{15}$$

is the initial nuclear state with $|a\rangle$ denoting its space dependent part and $|0\omega_x\rangle$ and $|1\omega_x\rangle$ are photon-number states of angular frequency ω_x. The initial and final nuclear states have energies $E_a=\hbar\epsilon_a$ and $E_b=\hbar\epsilon_b$, respectively. The space-dependent part of the initial and final electronic states, which are the same, is denoted by $\phi_0(\mathbf{r})$ and its energy is $E_0=\hbar\epsilon_0$. It is supposed that ϕ_0 describes an inner state, thus the effect of the laser on this state can be neglected because of the shielding of the outer electrons. Thus, ϕ_0 is a hydrogenic type solution in the usual spherical coordinates. The following notations are also introduced: $\omega_{ab}=\epsilon_a-\epsilon_b$, $\omega_{n0}=\epsilon_n-\epsilon_0$.

In Eq. (11) the cross term of the S matrix element is neglected as we search for a resonance-type process only. Furthermore, as in this simple model one only uses H_2 for the description of electron-nucleus interaction,[8] the γ-photon exchange between the nucleus and the electron cloud does not appear in the formalism.

Carrying out the time integrations in Eq. (11), the following result is obtained:

$$S_{fi}=(i\hbar)^{-2}\sum_{\substack{n_1,n_2,m,\\N,K}}H_{3,0n}H_{2,n0,ba}2\pi J_N(\lambda)J_K(\lambda)\times\frac{\delta(\omega_x+N\omega-\omega_{ab}-K\omega)}{\omega_{ab}-\omega_{n0}-N\omega+i\gamma_n/2}. \tag{16}$$

Here the following notation is used:

$$H_{3,0n}=\sum_{l=0}^{n-1}\langle lm|\mu_1\mu_2\rangle H_{3,0nlm} \tag{17}$$

with

$$H_{3,0nlm}=-\int e[\phi_0(\mathbf{r})]^*i(2\pi\hbar\omega_x/V)^{1/2}\boldsymbol{\epsilon}\cdot\mathbf{r}\Phi_{nlm}d^3r \tag{18}$$

and

$$H_{2,n0,ba}=\frac{4\pi}{2L+1}\sum_M Q_{LM}(a,b)I_{LM,n0}. \tag{19}$$

Here $Q_{LM}(a,b)$ is the matrix element of the multipole moment of order L,M between nuclear states a and b, which, applying the Wigner-Eckart theorem[9] can be written through its reduced matrix element $\langle b\|Q_L\|a\rangle$ as

$$Q_{LM}(a,b)=(-1)^{j_b-m_b}\langle b\|Q_L\|a\rangle\begin{pmatrix} j_b & L & j_a\\ -m_b & M & m_a\end{pmatrix} \tag{20}$$

and

$$I_{LM,n0}=\sum_{l=0}^{n-1}\langle 1m|\mu_1\mu_2\rangle I_{LM,nlm0} \tag{21}$$

with

$$I_{LM,nlm0}=e\int\Phi^*_{nlm}Y_{LM}\phi_0(\mathbf{r})r^{1-L}drd\Omega. \tag{22}$$

Y_{LM} is a spherical harmonics, Ω denotes solid angle, $\langle lm|\mu_1\mu_2\rangle$ is a Clebsch-Gordan coefficient, $\mu_1=(m+n_1-n_2)/2$, and $\mu_2=(m-n_1+n_2)/2$.[10]

Now we restrict ourselves to an initial (and also final) state of ks type, which has the form $\phi_0(\mathbf{r})=R_{k0}(r)Y_{00}$, where $R_{k0}(r)$ is the radial part of a hydrogen-type solution of principal quantum number k and orbital angular momentum quantum number $l=0$. Next we use the identity

$$\boldsymbol{\epsilon}\cdot\mathbf{r}=(4\pi/3)r\sum_s Y^*_{1s}(\hat{\mathbf{r}})Y_{1s}(\hat{\boldsymbol{\epsilon}}),$$

the formula

$$W_{fi}=|S_{fi}|^2\frac{V}{(2\pi)^3}\omega_x^2c^{-3}d\omega_x d\Omega_\epsilon ,$$

and the condition of orthogonality of the $3j$ symbols[9] and the spherical harmonics. As a result, we obtain for the transition probability per unit time of the resonance-type process

$$W_{fi}^{\rm las}=\frac{16\pi|\langle b\|Q_L\|a\rangle|^2}{9(2L+1)^3(2j_a+1)}\frac{e^4}{\hbar c^3}\times\frac{|J_{nk}|^2|I_{L,nk}|^2}{\Delta^2+(\hbar\gamma_n/2)^2}\sum_K\omega_{xK}^3\mathcal{F}_K , \quad (23)$$

where $\Delta=\hbar(\omega_{ab}-N\omega-\omega_{n0})$ is the detuning,

$$J_{nk}=\int R_{k0}(Z_{\rm eff}(k),r)R_{n1}(Z_{\rm eff}(n),r)r^3dr , \quad (24)$$

$$I_{L,nk}=\int R_{k0}(Z_{\rm eff}(k),r)R_{nL}(Z_{\rm eff}(n),r)r^{1-L}dr , \quad (25)$$

and

$$\mathcal{F}_K=\sum_M\left[\sum_{\mu_1,\mu_2}\langle 1M|\mu_1\mu_2\rangle\langle LM|\mu_1\mu_2\rangle J_K(\lambda_{n_1n_2})J_N(\lambda_{n_1n_2})\right]^2 . \quad (26)$$

The energy of the x-ray photon is $\hbar\omega_{xK}=\hbar(\omega_{ab}-N\omega+K\omega)$. For the sake of clarity we have explicitly denoted the different effective charges of the states of different principal quantum numbers in Eqs. (24) and (25).

Now we introduce the quantity η, which is defined as $\eta=W_{fi}^{\rm las}/W_{fi}^{\rm spont}$, where $W_{fi}^{\rm spont}$ is the transition probability per unit time of the spontaneous γ-ray emission[8]

$$\eta=\frac{2[(2L-1)!!]^2e^2}{9(L+1)(2L+1)}\left[\frac{c}{\omega_{ab}}\right]^{2L-2}\times\frac{|J_{nk}|^2|I_{L,nk}|^2}{\Delta^2+(\hbar\gamma_n/2)^2}\sum_K\left[\frac{\omega_{xK}}{\omega_{ab}}\right]^3\mathcal{F}_K . \quad (27)$$

The ratio η characterizes the yield of the laser-assisted, resonant electronic-bridge mechanism related to the spontaneous γ decay. In the presence of an intense laser beam of appropriate angular frequency that meets the condition of resonance, the transfer of the nuclear excitation energy through the electron bridge may become very effective.

Although in these cases, which are expected to be modified by intense laser field,[3,4] the energy defect is comparable to the laser-photon energies available today, the start of the process needs the absorption of more than one laser photon. Thus from an experimental point of view the use of two lasers of different frequencies seems to be more advantageous: a laser of fixed photon energy $\hbar\omega$, which is applied to inject the larger part of the energy defect, and a tunable laser with photon energy $\hbar\omega'$ to fulfill the resonance condition. If the lasers have linear and parallel states of polarization then Eqs. (6), (8), and (26) are modified as

$$\mathbf{E}_L(t)=E_0\hat{\mathbf{z}}\cos(\omega t)+E_0'\hat{\mathbf{z}}\cos(\omega' t) , \quad (28)$$

$$\Psi(n,n_1,n_2,m)=\Phi_{n_1n_2m}\sum_{N,N'=-\infty}^{\infty}J_N(\lambda_{n_1n_2})J_{N'}(\lambda'_{n_1n_2})\exp[-i(E_n/\hbar+N\omega+N'\omega')t] , \quad (29)$$

and

$$\mathcal{F}_{K,K'}=\sum_M\left[\sum_{\mu_1,\mu_2}\langle 1M|\mu_1\mu_2\rangle\langle LM|\mu_1\mu_2\rangle J_K(\lambda)J_N(\lambda)J_{K'}(\lambda')J_{N'}(\lambda')\right]^2 , \quad (30)$$

where the prime refers to the parameter of the second, tunable laser. The resonance condition determines N and N' ($N'=-1$ as the second laser is weak) thus the detuning is

$$\Delta=\hbar(\omega_{ab}-N\omega-\omega'-\omega_{n0}) \quad (31)$$

and $\hbar\omega_{xKK'}=\hbar(\omega_{ab}-N\omega-\omega'+K\omega+K'\omega')$ is the energy of the x-ray photon. In this case Eqs. (23) and (27) remain valid if $\hbar\omega_{xK}$ and $\mathcal{F}_K$ are replaced with $\hbar\omega_{xKK'}$ and $\mathcal{F}_{K,K'}$.

To find feasible candidate materials for the resonant laser-assisted electronic-bridge mechanism three points following from Eq. (27) may be considered as a guideline. The nuclear transition must have small energy, high multipolarity, and the energy defect must be small. The ^{235m}U isomer, which has a γ photon energy 73.5 eV and multipolarity $L=3$, seems to be a most promising material. The electron has 71-eV binding energy in the $P1(6s_{1/2})$ shell in the U atom.[11] For an intermediate state of binding energy 2.14 eV, the energy misfit equals 4.64 eV, which is resonant to the emission of two laser photons of energy 2.32 eV. Thus, at a laser intensity $I=10^{12}$ W/cm^2, Eq. (27) results $\eta=6.7\times10^9$ which seems to be observable. The laser plays a dual role in producing this large enhancement. In this way not only the resonance condition can be fulfilled but the originally $E3$ γ transition can be converted into the emission of an $E1$ x-ray photon. The presence of the laser beam makes it also possible to break the selection rules of x-ray transitions which are valid in the laser free case.

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