Nuclear-coupled Rabi oscillations

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(Received 15 June 2001; published 1 April 2002)

The role of electronic resonances due to a laser field of appropriate frequency is investigated in laser-assisted internal conversion (IC). After the IC creates a vacancy in the electron shell, Rabi oscillations of an electron may take place in the presence of a resonant laser field. By deriving a no-go theorem, we show that a resonant process of this or, in fact, any related sort has no effect on the nuclear decay rate. This finding seems to contradict some earlier theoretical predictions in this field.

DOI: 10.1103/PhysRevA.65.045402 PACS number(s): 32.80.Wr, 23.20.Nx

In the last 15 years there has been a great deal of interest in laser-assisted internal-conversion (IC) processes [1]. In the early works nonrelativistic Volkov and Coulomb-Volkov solutions were used for the description of dressed electronic states to include those processes where IC was assisted by the absorption of many laser photons [2]. Afterwards, electron-bridge processes were also studied in the presence of laser radiation [3], including the role of resonances in case of some special isomers [4]. Then several papers were addressing the role of electronic resonances in the presence of a laser field tuned to the electronic transition in these exotic cases [5]. Moreover, in the case of processes involving at least two steps, such as, electron-nucleus and laser-electron ones, emerging resonances were treated using approaches very similar [6–8] to the approach of Breit and Wigner, which was used to describe nuclear resonances [9].

Some of these papers [7,8] predict the possibility that the rate of the laser-modified inverse IC process (the laser-assisted process of nuclear excitation in electronic transition) may be increased significantly due to its resonant character. Specifically, these papers deal with $^{229}\mathrm{Th}^{m}$, which is an exotic nuclear isomer with nuclear excitation energy of 3.5 ± 1.1 eV. The assumption was made in Refs. [6–8] that the nuclear transition may be resonant with a bound-bound electronic one. However, the difference of the nuclear and electronic transition energies is much larger than the corresponding nuclear energy width. Consequently, one cannot speak about resonance in this case at all (the term becomes very misleading) and the use of the Breit-Wigner formula cannot be justified. In the two-step process the laser helps to satisfy the requirement of energy conservation only. In a recent paper [10], we have shown that contributions of both second-order graphs to the process investigated in Refs. [6,7] as well as all intermediate states have to be taken into account in the calculation. As improper treatment of resonances has happened before, it is important to clarify which process can be considered a resonant one in the case of laser-assisted electron-nucleus combined processes.

In fact, three different kinds of resonances can occur in the case of laser-assisted IC processes. First, when two electronic states, one occupied and one empty, are coupled by a laser of appropriate frequency, Rabi oscillation of the electron arises [11]. Then IC takes effect on the oscillating electron, kicking it into a free state. If the two coupled electronic bound states are sufficiently close to each other, so that the IC rate can be considered to be equal for both, the resonant character of the process has no influence on the rate of the IC. Second, if there is resonance with some intermediate state of a higher-order nucleus-electron-laser process, then it has been shown [12] that the poles that arise can be avoided and ultimately do not give rise to any increase in the IC rate and no effect is expected due to the resonant character of the process. However, in the third case, the resonance may occur in the final state, viz., after the IC has created a vacancy, Rabi oscillations can arise due to a laser with appropriate frequency. In this paper we investigate the effect of the resonance in the final state and take into account the Rabi oscillation of the participating electron, in a general way.

To describe the electron-nucleus-laser processes, we introduce a system that consists of two electrons and the nucleus interacting with them, and a laser field which can resonantly modify the state of the participating electrons, but cannot directly affect the state of the nucleus. Let us suppose that initially both electrons are bound and they are in their ground state, but the nucleus is excited, e.g., it is in a metastable state. The deexcitation of the nucleus causes one of the bound electrons to get into a free state (in the process that is called IC) and the atom becomes ionized. The resonant laser field can give rise to Rabi oscillation of an other electron between two bound states of the ion. The whole process is treated in the second order of the perturbation theory.

The Hamiltonian of the unperturbed electron system can be written as

$$H_0=\hbar\begin{pmatrix}\omega_1 & 0 & 0\\ 0 & \omega_2 & 0\\ 0 & 0 & \omega_3\end{pmatrix}, \tag{1}$$

where $\omega_1=E_1/\hbar$, $\omega_2=E_2/\hbar$, and $\omega_3=E_3/\hbar$, and E_1, E_2, and E_3 are the energies of the electronic states of the system;

numeric 1 refers to the state where both electrons are bound, while 2 and 3 refer to those cases when one electron is free and the other is in one of the two bound states coupled by the laser field. To take into account the decay of these electronic states, we introduce the complex energy operator

$$i\hbar\Gamma = i\hbar \begin{pmatrix} \gamma_1 & 0 & 0 \\ 0 & \gamma_2 & 0 \\ 0 & 0 & \gamma_3 \end{pmatrix} \tag{2}$$

and γ_1, γ_2, and γ_3 describe the rate at which the states 1, 2, and 3 decay.

The interaction between the laser field and the bound electron can be represented by the matrix

$$H_1 = -2\hbar\Omega_z \cos(\omega t) \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}. \tag{3}$$

Here $\Omega_z = p_z E_z/\hbar$ is the Rabi frequency, characterizing the coupling strength between the electronic transition and the laser. Furthermore, E_z is the amplitude of the laser field (the laser is linearly polarized in the z direction), $2p_z$ is the z component of dipole moment of the transition $2 \rightarrow 3$, ω is the angular frequency of the laser, and t is the time in the frame of reference.

To describe the interaction with the nucleus, i.e., to describe the IC process, we write its Hamiltonian as

$$H_2 = Ve^{-i\omega_\gamma t} \begin{pmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{4}$$

where V is the potential created by the nuclear transition four current and felt by the electron. It contains the Coulomb potential and current-current terms in the case without retardation, and its explicit form is not needed in our calculation. $\omega_\gamma = E_\gamma/\hbar$ and E_γ is the energy of the nuclear transition transferred to one of the electrons in the first step of the process. The above asymmetric form of H_2 is a consequence of the fact that we have neglected the probability of reexcitation of the nucleus. Thus H_2 can only induce an $1 \rightarrow 2$ transition.

Now let us transform our system by the unitary operator $U_0 = e^{i(H_0 t)/\hbar}$. In this interaction picture, the Hamiltonian of the laser interaction is

$$H_1 = -\hbar\Omega_z (e^{i\omega t} + e^{-i\omega t}) \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & e^{i\omega_{23} t} \\ 0 & e^{i\omega_{32} t} & 0 \end{pmatrix}, \tag{5}$$

where $\omega_{23} = \omega_2 - \omega_3$ and $\omega_{32} = \omega_3 - \omega_2$. After employing the rotating-wave approximation, i.e., neglecting the rapidly oscillating terms, we get

$$H_1 = -\hbar\Omega_z \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & e^{i\Delta_0 t} \\ 0 & e^{-i\Delta_0 t} & 0 \end{pmatrix}, \tag{6}$$

with $\Delta_0 = \omega_{23} - \omega$. Note that $\omega_{23} + \omega \gg \Delta_0$. We also obtain for H_2 in this transformed picture

$$H_2 = Ve^{i\Delta t} \begin{pmatrix} 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{7}$$

where we have introduced the notation $\Delta = \omega_{12} - \omega_\gamma$ and $\omega_{12} = \omega_1 - \omega_2$.

Let us transform our system again [13], getting into the oscillating picture which is defined by H_1. The unitary operator U_1 of such a transformation is given by the equation $i\hbar(dU_1/dt) = H_1 U_1$, and the solution is easy to find if we take $\Delta_0 = 0$, i.e., the laser is resonant with the transition $2 \rightarrow 3$,

$$U_1 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos(\Omega_z t) & i\sin(\Omega_z t) \\ 0 & i\sin(\Omega_z t) & \cos(\Omega_z t) \end{pmatrix}. \tag{8}$$

The transformed Hamiltonian, $\hat{H}_2 = U_1^\dagger H_2 U_1$, takes the form

$$\hat{H}_2 = Ve^{i\Delta t} \begin{pmatrix} 0 & 0 & 0 \\ \cos(\Omega_z t) & 0 & 0 \\ -i\sin(\Omega_z t) & 0 & 0 \end{pmatrix}. \tag{9}$$

Similarly the transformation of Γ yields

$$\hat{\Gamma} = U_1^\dagger \Gamma U_1 = \begin{pmatrix} \gamma_1 & 0 & 0 \\ 0 & \gamma_2 \cos^2(\Omega_z t) + \gamma_3 \sin^2(\Omega_z t) & i(\gamma_2 - \gamma_3)\sin(2\Omega_z t)/2 \\ 0 & -i(\gamma_2 - \gamma_3)\sin(2\Omega_z t)/2 & \gamma_2 \sin^2(\Omega_z t) + \gamma_3 \cos^2(\Omega_z t) \end{pmatrix}, \tag{10}$$

which becomes diagonal if we assume $\gamma_2=\gamma_3=\gamma$,

$$\hat{\Gamma}=\begin{pmatrix}\gamma_1 & 0 & 0\\ 0 & \gamma & 0\\ 0 & 0 & \gamma\end{pmatrix}. \tag{11}$$

In the following, we introduce the wave function of the three-level system as $|\psi(t)\rangle=c_1(t)|1\rangle+c_2(t)|2\rangle+c_3(3)|3\rangle$, where $|1\rangle$ is the vector representing state 1, and, respectively, for the others. The time dependence of the system is contained in the expansion coefficients and the Schrödinger equation in the second interaction picture can be written as

$$i\hbar\frac{dc_n}{dt}=(\hat{H}_2-i\hbar/2\hat{\Gamma})_{nm}c_m, \tag{12}$$

for $n,m=1,2,3$ and summation over m is assumed. From here we obtain the following explicit equations for the expansion coefficients:

$$\frac{dc_1}{dt}=-\frac{\gamma_1}{2}c_1, \tag{13}$$

$$\frac{dc_2}{dt}=-i\frac{V}{\hbar}e^{i\Delta t}\cos(\Omega_z t)c_1-\frac{\gamma}{2}c_2, \tag{14}$$

$$\frac{dc_3}{dt}=-\frac{V}{\hbar}e^{i\Delta t}\sin(\Omega_z t)c_1-\frac{\gamma}{2}c_3. \tag{15}$$

Here, due to the series of transformations, c_2 and c_3 are already decoupled. Inserting the solution of Eq. (13), $c_1=c_1(0)\exp(-\gamma_1 t/2)$, into the equations for c_2 and c_3, and making the reasonable assumption $\gamma_1\ll\gamma$ we can integrate these equations. Note that $\gamma_1=\ln 2/T_1$, where T_1 is the lifetime of state 1. Since $T_1>1s$ typically, as it is governed by the lifetime of a nucleus in the metastable state, and $\gamma=\ln 2/T$ where T is the lifetime of the vacancy created by the IC, and it is of the order of nanoseconds or less, we feel that the above approximation is well justified. In fact, in the following we set $\gamma_1=0$.

We chose the initial conditions $c_2(0)=c_3(0)=0$ and $c_1(0)=1$, corresponding to the situation when the excitation of the isomeric state of the nucleus coincides with the beginning of the measurement. For the time-dependent solution we then obtain

$$c_2=-i\frac{V}{\hbar}\frac{[(i\Delta+\gamma/2)\cos(\Omega_z t)+\Omega_z\sin(\Omega_z t)]e^{i\Delta t}-(i\Delta+\gamma/2)e^{-\gamma t/2}}{(i\Delta+\gamma/2)^2+\Omega_z^2} \tag{16}$$

and

$$c_3=\frac{V}{\hbar}\frac{[\Omega_z\cos(\Omega_z t)-(i\Delta+\gamma/2)\sin(\Omega_z t)]e^{i\Delta t}-\Omega_z e^{-\gamma t/2}}{(i\Delta+\gamma/2)^2+\Omega_z^2}. \tag{17}$$

To obtain the probability of the laser-assisted IC process, we integrate the quantity $|c_2|^2+|c_3|^2$ over the entire phase space, i.e.,

$$\int_{\Omega_k}\int_0^\infty(|c_2|^2+|c_3|^2)\frac{L^3}{(2\pi)^3}k^2dkd\Omega_k. \tag{18}$$

Here k is the absolute value of the wave number vector of the free electron with energy E_2, and so $k=\sqrt{2mE_2}/\hbar$. Ω_k is the solid angle in the k space; m is the rest mass of the electron, L is the volume of the normalization. Using $kdk=(m/\hbar)d\Delta$ we can carry out the integration over Δ. As the quantity $|c_2|^2+|c_3|^2$ gives significant contribution around $\Delta=0$, we can substitute $k=\sqrt{2m(E_1+E_\gamma)}/\hbar$ (its value for $\Delta=0$) into the above integral. Moreover, we have expanded the interval of integration from $-\infty$ to ∞, because $|c_2|^2+|c_3|^2$ vanishes with increasing $|\Delta|$.

If $\Omega_z\to 0$ then $|c_3|^2=0$, and we receive the quantity

$$|c_2|^2=\frac{|V|^2}{\hbar^2}\frac{1+e^{-\gamma t}-2e^{-\gamma t/2}\cos(\Delta t)}{\Delta^2+(\gamma/2)^2}. \tag{19}$$

Integrating it over the phase space Δ we obtain

$$N_0(t)=\mathcal{K}\int_{-\infty}^{\infty}|c_2|^2d\Delta=\frac{2\pi|V|^2\mathcal{K}}{\hbar^2\gamma}(1-e^{-\gamma t}), \tag{20}$$

where $\mathcal{K}=L^3/\sqrt{2(mc^2)^3(E_1+E_\gamma)}/(2\pi^2/\hbar^2c^3)$ is the phase-space constant and c is the velocity of light. The transition probability per unit time of the laser-free IC process can be given as $W_0=N_0\gamma$.

In the case when a resonant laser field is present, from Eqs. (16) and (17) we obtain the expression

$$\begin{aligned}|c_2|^2+|c_3|^2=&\frac{|V|^2}{\hbar^2}\frac{1}{[\Delta^2+\Omega_z^2+(\gamma/2)^2]^2-4\Omega_z^2\Delta^2}\\ &\times\{[\Delta^2+\Omega_z^2+(\gamma/2)^2][1+e^{-\gamma t}\\ &-2e^{-\gamma t/2}\cos(\Omega_z t)\cos(\Delta t)]\\ &-4e^{-\gamma t/2}\Delta\Omega_z\sin(\Omega_z t)\sin(\Delta t)\}.\end{aligned} \tag{21}$$

We can now integrate the above expression over the entire phase space, i.e., over Δ, and receive

$$N(t)=\frac{2\pi|V|^2\mathcal{K}}{\hbar^2\gamma}[1+e^{-\gamma t}-2e^{-\gamma t}\cos^2(\Omega_z t) - 2e^{-\gamma t}\sin^2(\Omega_z t)]=\frac{2\pi|V|^2\mathcal{K}}{\hbar^2\gamma}(1-e^{-\gamma t}). \tag{22}$$

The transition probability per unit time of the laser-assisted IC process is given by $W=N\gamma$.

Note that $N_0(t)=N(t)$ and, therefore,

$$W=W_0, \tag{23}$$

which is our most important result, stating that the rates of the laser-free and laser-assisted IC processes are equal.

In conclusion, we have shown that the presence of a laser field has no influence on the IC process, in spite of the resonant character of the interaction with the final state. Nor do the other kind of the resonant processes exert any influence on the rate of the IC, as we have discussed the state of the theoretical art in the Introduction. If one is interested in a feasible experiment, a possible realization of the above situation is offered by, e.g., the ^{90}Zr isomer which has a metastable state with a lifetime of 61 ns, much longer than the lifetime of the vacancy in an inner (e.g., K) electron shell [14]. The isomeric state is populated through a cascade from another metastable state of the same nucleus with much longer lifetime (809 ms). This long-lived metastable state is, in turn, populated by the β-decay of ^{90}Nb [15]. When the state with the lifetime of 61 ns is populated, a γ pulse with energy of 425.5 keV and duration of 93 fs is emitted, marking the event and triggering the measurement. Then this isomeric nuclear state decays via an $E0$ transition into the ground state of the nucleus through IC only. The emitted energetic IC electron should also be registered. This way, one can obtain our $N_0(t)$ or $N(t)$ quantities. Since their explicit expressions, Eqs. (20) and (22), are identical, the spectrum must be independent of whether or not the resonant laser field is present. Therefore, if the rate of the IC process is found to be altered by the presence of lasers in future experiments, the change should not be attributed to the resonant interaction with the laser field of appropriate frequency.

This work was partly supported by the Hungarian National Science Research Foundation (OTKA) under Grant No. T031825. The research of J.B. was partially supported by the Office of Naval Research (Grant No. N00014-92J-1233).

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