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PHYSICS

Time-dependent theory of three-step absorption of three light pulses

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Abstract. The time-dependent theory of the three-step absorption of three different light pulses in the electronic four-level systems is considered using the time-dependent perturbation theory. The first, the second, and the third pulse are in resonance with transitions from level 0 to level 1, from level 1 to level 2, and from level 2 to level 3, respectively. The spectral and temporal behaviour of the probability that the fourth level is excited at the moment of time t depends on the energy relaxation constants of the excited electronic levels, the frequencies of the maxima, the spectral widths (and so the durations) of the light pulses, and the time delays between the pulses. To conclude, there are 12 parameters that affect the spectra in the simple model used. We calculate and analyse the case where the frequencies of the maxima of the first and the second pulse are fixed and the frequency of the maximum of the third pulse varies. On the whole, three lines may exist, one of which corresponds to the coherent contribution and the others do not.

Key words: theory of three-step absorption, time-dependent perturbation theory, three different light pulses.

1. INTRODUCTION

Two-step absorption of light by matter has been researched for decades [1–6]. In this paper the time-dependent theory of three-step absorption in the electronic four-level model is presented. We used time-dependent perturbation theory. Using the simplest model in calculations, we analyse the temporal and spectral behaviour of the spectrum. The probability that the fourth level is excited at the moment t depends on the energy relaxation constants of the excited electronic levels, the frequencies of the maxima, the spectral widths (and so the durations) of the light pulses, and the time delays between the pulses. The phase relaxation and the phonon wings are not taken into account. The absorbed pulses are different and may have arbitrary spectral widths and arbitrary durations.

Different limit cases are studied analytically. The spectra with fixed frequencies of the maxima of the first and second pulses and with varied frequency of the maximum of the third pulse are calculated and analysed. On the whole, three lines may exist; one of these corresponds to the coherent contribution [4] and the others do not. The three-step absorption allows deeper understanding of which lines, coherent or non-coherent, appear in the absorption spectrum and of which parameters their widths depend on. The present consideration applies for impurity centres at low temperatures with weak electron–phonon coupling.

2. PROBABILITY OF THREE-STEP TRANSITION

The process started from ground level 0. Resonance conditions are $\omega_1 \approx \Omega_{01}$, $\omega_2 \approx \Omega_{12}$, and $\omega_3 \approx \Omega_{23}$, where Ω_{01}, Ω_{12}, and Ω_{23} are the frequencies of the transitions 0→1, 1→2, and 2→3, and ω_1, ω_2, and ω_3 are the frequencies of the maxima of the pulses.

Let us find the probability that at time t the system is in the final state by applying time-dependent perturbation theory. First we find the amplitude of the probability. For general consideration we need formulas in which the initial state of the system consists of the electromagnetic field and matter is given at initial time $t_0 = -\infty$. In this case we can use any shapes of excitation pulses of light.

The system is described by the Hamiltonian

$$\hat{H} = \hat{H}_{CR} + \hat{V} = \hat{H}_C + \hat{H}_R + \hat{V}. \tag{1}$$

At initial time t_0 the system is in the state

$$|\psi(t_0)\rangle = |\psi_C(t_0)\rangle |\psi_R(t_0)\rangle. \tag{2}$$

In Eq. (1) $\hat{H}_C$ is the Hamiltonian of matter, $\hat{H}_R$ is the Hamiltonian of the electromagnetic field, $\hat{V}$ is the Hamiltonian of interaction. In Eq. (2) $|\psi_C(t_0)\rangle$ and $|\psi_R(t_0)\rangle$ are the initial states of matter and of the electromagnetic field.

The characteristic states and eigenvalues of these Hamiltonians are the following:

$$\hat{H}_{CR}|j\rangle = E_j|j\rangle, \quad \hat{H}_C|i\rangle = E_i|i\rangle, \quad \hat{H}_R|\omega\rangle = \omega|\omega\rangle. \tag{3}$$

The initial state of matter

$$|\psi_C(t_0)\rangle = \sum_{i'} b_{i'}|i'\rangle \exp(-iE_{i'}t_0) \tag{4}$$

and the initial state of the electromagnetic field (three light pulses)

$$|\psi_R(t_0)\rangle = \int_{-\infty}^{\infty} d\omega B_1(\omega-\omega_1)\exp(-i\omega t_0) \int_{-\infty}^{\infty} d\omega' B_2(\omega'-\omega_2)\exp(-i\omega' t_0)$$
$$\times \int_{-\infty}^{\infty} d\omega'' B_3(\omega''-\omega_3)\exp(-i\omega'' t_0)|\omega,\omega',\omega''\rangle, \tag{5}$$

where

$$\int_{-\infty}^{\infty} d\omega |B_1(\omega)|^2 = 1, \quad \int_{-\infty}^{\infty} d\omega' |B_2(\omega')|^2 = 1, \quad \int_{-\infty}^{\infty} d\omega'' |B_3(\omega'')|^2 = 1. \tag{6}$$

Equations (6) describe normalization conditions of three single photon wave packages with maxima at frequencies ω_1, ω_2, and ω_3, respectively.

The amplitude of the probability of finding the system at time $t \geq t_0$ in the state $|j\rangle$ according to the Hamilton equation is

$$c_j(t) = \langle j|\exp\left[-i(t-t_0)\hat{H}\right]|\psi(t_0)\rangle$$
$$= \sum_{i'} \int_{-\infty}^{\infty} d\omega \int_{-\infty}^{\infty} d\omega' \int_{-\infty}^{\infty} d\omega'' \langle j|\exp\left[-i(t-t_0)\hat{H}\right]|i'\rangle|\omega,\omega',\omega''\rangle b_{i'} \exp(-iE_{i'}t_0)$$
$$\times B_1(\omega-\omega_1)B_2(\omega'-\omega_2)B_3(\omega''-\omega_3)\exp\left[-i(\omega+\omega'+\omega'')t_0\right]$$
$$= \sum_{j'} \langle j|\exp\left[-i(t-t_0)\hat{H}\right]|j'\rangle b_{j'} \exp(-iE_{j'}t_0). \tag{7}$$

Let us take into account that at y ≥ 0

$$\exp\left(-iy\hat{H}\right)=\exp\left(-iy\hat{H}_{CR}\right)T\exp\left[-i\int_0^y ds\hat{V}(s)\right], \tag{8}$$

where $\hat{V}(s)=\exp\left(is\hat{H}_{CR}\right)\hat{V}\exp\left(-is\hat{H}_{CR}\right)$, T is the operator of chronological arranging, which puts the operators from right to left in the order of increasing s. To go to limit $t_0 \to -\infty$, the decay of the characteristic states of the Hamiltonian $\hat{H}_{CR}$ has to be taken into account. Then the amplitude $c_j(t)$ can be approximately presented as follows:

$$c_j(t)\approx\sum_{n=1}^{\infty}(-i)^n\sum_{j_1,j_2,\dots,j_n}{}' \int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\dots\int_{t_0}^{t_{n-1}}dt_n\exp\left[-i\left(E_j-i\gamma_j\right)(t-t_1)\right]\langle j|\hat{V}|j_1\rangle$$
$$\times\exp\left[-i\left(E_{j_1}-i\gamma_{j_1}\right)(t_1-t_2)\right]\langle j_1|\hat{V}|j_2\rangle\dots\langle j_{n-1}|\hat{V}|j_n\rangle b_{j_n}\exp\left(-iE_{j_n}t_n\right), \tag{9}$$

where γ_j is the decay constant of the state $|j\rangle$ and the apostrophe means that the terms with the coinciding numbers of states are omitted. This formula can be used for all values of t_0 including $-\infty$ only if the interaction is small enough.

Here we assume that the final state does not coincide the with the initial state ($b_j = 0$), the integration variables $t_1, t_2, \dots, t_n$ are the times of the transitions of the amplitude of the probability from one characteristic state of Hamiltonian $\hat{H}$ into the other, and the differences $t - t_1$, $t_1 - t_2$, … $t_{n-1} - t_n$, and $t_n - t_0$ determine time intervals during which the amplitude of the probability is in state $|j\rangle$, $|j_1\rangle$, ..., $|j_{n-1}\rangle$, and $|j_n\rangle$, respectively.

To describe the process with the three photons the term of the third order of expansion is needed:

$$c_j(t)\approx -i\sum_{j_1,j_2,j_3}{}'\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_2}dt_3\exp\left[-i\left(E_j-i\gamma_j\right)(t-t_1)\right]\langle j|\hat{V}|j_1\rangle\exp\left[-i\left(E_{j_1}-i\gamma_{j_1}\right)(t_1-t_2)\right]$$
$$\times\langle j_1|\hat{V}|j_2\rangle\exp\left[-i\left(E_{j_2}-i\gamma_{j_2}\right)(t_2-t_3)\right]\langle j_2|\hat{V}|j_3\rangle b_{j_3}\exp\left(-iE_{j_3}t_3\right). \tag{10}$$

Since

$$\begin{gathered}E_j-i\gamma_j=E_i-i\gamma_i,\quad E_{j_1}-i\gamma_{j_1}=E_{i_1}-i\gamma_{i_1}+\omega'',\\ E_{j_2}-i\gamma_{j_2}=E_{i_2}-i\gamma_{i_2}+\omega'+\omega'',\ E_{j_3}=E_{i_3}+\omega+\omega'+\omega'',\end{gathered} \tag{11}$$

then

$$c_j(t)\approx -i\sum_{i_1,i_2,i_3}{}'\int_{-\infty}^{\infty}d\omega\int_{-\infty}^{\infty}d\omega'\int_{-\infty}^{\infty}d\omega''\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\int_{t_0}^{t_2}dt_3\exp\left[-i\left(E_i-i\gamma_i\right)(t-t_1)\right]$$
$$\times\langle i|\langle 0|\hat{V}|\omega''\rangle|i_1\rangle\exp\left[-i\left(E_{i_1}-i\gamma_{i_1}+\omega''\right)(t_1-t_2)\right]\langle i_1|\langle\omega''|\hat{V}|\omega',\omega''\rangle|i_2\rangle$$
$$\times\exp\left[-i\left(E_{i_2}-i\gamma_{i_2}+\omega'+\omega''\right)(t_2-t_3)\right]\langle i_2|\langle\omega',\omega''|\hat{V}|\omega,\omega',\omega''\rangle|i_3\rangle$$
$$\times b_{i_3}\exp\left[-i\left(E_{i_3}+\omega+\omega'+\omega''\right)t_3\right]B_1(\omega-\omega_1)B_2(\omega'-\omega_2)B_3(\omega''-\omega_3). \tag{12}$$

Let us introduce the single photon matrix elements

$$v_{\omega_1} = \langle \omega', \omega'' | \hat{V} | \omega, \omega', \omega'' \rangle, \\ v_{\omega_2} = \langle \omega'' | \hat{V} | \omega', \omega'' \rangle, \quad v_{\omega_3} = \langle 0 | \hat{V} | \omega'' \rangle, \tag{13}$$

and the functions $B_i(t)$, which determine time profiles of the pulses:

$$B_i(\omega) = \int_{-\infty}^{\infty} dt \exp(i\omega t) B_i(t). \tag{14}$$

In the end we get

$$c_j(t) \approx -i \sum_{i_1,i_2,i_3}' \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t_2} dt_3 \exp\left[-i(E_i - i\gamma_i)(t - t_1)\right] \langle i | v_{\omega_3} | i_1 \rangle \exp\left[-i\left(E_{i_1} - i\gamma_{i_1}\right)(t_1 - t_2)\right] \\ \times \langle i_1 | v_{\omega_2} | i_2 \rangle \exp\left[-i\left(E_{i_2} - i\gamma_{i_2}\right)(t_2 - t_3)\right] \langle i_2 | v_{\omega} | i_3 \rangle b_{i_3} \exp\left(-iE_{i_3} t_3\right) \varepsilon_1(t_3) \varepsilon_2(t_2) \varepsilon_3(t_1), \tag{15}$$

where

$$\varepsilon_i(t) = \exp(-i\omega_i t) B_i(t). \tag{16}$$

In Eq. (16) it is assumed that the pulses are quasi-monochromatic, i.e. $B_i(t)$ are slowly changing functions in comparison with $\exp(-i\omega_i t)$.

To get the probability it is necessary to average the quantity $|c_i(t)|^2$ over the initial states and to summarize over the final states. In the conditions of thermal equilibrium

$$\left\langle b^*_{i_3} b_{i'_3} \right\rangle_c = n_{i_3} \delta_{i_3 i'_3}, \quad n_{i_3} = \frac{1}{Z} \exp\left(-E_{i_3}/kT\right), \tag{17}$$

where Z is the statistic sum, T is temperature, and $\langle \ldots \rangle$ is the mark of averaging over ensemble.

Taking the preceding into account, we get

$$W(t) \approx \sum_{i,i_3} n_{i_3} \sum_{i_1,i_2,i'_1,i'_2} \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t} dt'_1 S_3(t_1, t'_1) \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t'_1} dt'_2 S_2(t_2, t'_2) \\ \times \int_{-\infty}^{t_2} dt_3 \int_{-\infty}^{t'_2} dt'_3 \langle i_3 | v^+_{\omega_1} | i'_2 \rangle \exp\left[i\left(E_{i'_2} - i\gamma_{i'_2}\right)(t'_2 - t'_3)\right] \langle i'_2 | v^+_{\omega_2} | i'_1 \rangle \\ \times \exp\left[i\left(E_{i'_1} - i\gamma_{i'_1}\right)(t'_1 - t'_2)\right] \langle i'_1 | v^+_{\omega_3} | i \rangle \exp\left[-iE_i(t'_1 - t_1) - \gamma_i(2t - t_1 - t'_1)\right] \\ \times \langle i | v_{\omega_3} | i_1 \rangle \exp\left[-i\left(E_{i_1} - i\gamma_{i_1}\right)(t_1 - t_2)\right] \langle i_1 | v_{\omega_2} | i_2 \rangle \\ \times \exp\left[-i\left(E_{i_2} - i\gamma_{i_2}\right)(t_2 - t_3)\right] \langle i_2 | v_{\omega_1} | i_3 \rangle \exp\left[-iE_{i_3}(t_3 - t'_3)\right] S_1(t_3, t'_3). \tag{18}$$

Here

$$S_i(t_j, t'_j) = \left\langle \varepsilon_i{}^*(t_j) \varepsilon_i(t'_j) \right\rangle_R \tag{19}$$

are the correlation functions of the absorbed pulses, $\langle \ldots \rangle_R$ denotes averaging over states of pulses. The functions $S_i(t_j,t'_j)$ are considerably different from zero in the region $|t_j|$, $|t'_j| \le \Delta_i^{-1}$, where Δ_i is the spectral width of pulse i. If the pulse is coherent, its duration is determined by the time Δ_i^{-1}.

Taking into account that $E_i |i\rangle = \hat{H}_C |i\rangle$ and introducing the operator of damping $\gamma_i |i\rangle = \hat{\gamma} |i\rangle$, Eq. (18) can be rewritten in the form:

$$W(t)=\int_{-\infty}^{t}dt_1\int_{-\infty}^{t}dt'_1S_3(t_1,t'_1)\int_{-\infty}^{t_1}dt_2\int_{-\infty}^{t'_1}dt'_2S_2(t_2,t'_2)\int_{-\infty}^{t_2}dt_3\int_{-\infty}^{t'_2}dt'_3F(t,t_1,t'_1,t_2,t'_2,t_3,t'_3)S_1(t_3,t'_3), \quad (20)$$

where

$$F(t,t_1,t'_1,t_2,t'_2,t_3,t'_3)=\langle v^+_{\omega_1}\exp\left[i\left(\hat{H}_C+i\gamma\right)(t'_2-t'_3)\right]v^+_{\omega_2}\exp\left[i\left(\hat{H}_C+i\gamma\right)(t'_1-t'_2)\right]$$
$$\times v^+_{\omega_3}\exp\left[-i\hat{H}_C(t'_1-t_1)-\gamma(2t-t_1-t'_1)\right]v_{\omega_3}\exp\left[-i\left(\hat{H}_C-i\gamma\right)(t_1-t_2)\right]v_{\omega_2}$$
$$\times\exp\left[-i\left(\hat{H}_C-i\gamma\right)(t_2-t_3)\right]v_{\omega_1}\exp\left[-i\hat{H}_C(t_3-t'_3)\right]\rangle_C. \quad (21)$$

At the end this probability $W(t)$ decreases to zero with the increase of time t.

3. MODEL

In our model the pulses are coherent and of a single-sided exponential shape. The corresponding correlation functions are

$$\begin{aligned}S_1(t_3,t'_3)&=\theta(t_3-\tau_1)\theta(t'_3-\tau_1)\exp[i\omega_1(t_3-t'_3)-\Delta_1(t_3+t'_3-2\tau_1)/2],\\S_2(t_2,t'_2)&=\theta(t_2-\tau_2)\theta(t'_2-\tau_2)\exp[i\omega_2(t_2-t'_2)-\Delta_2(t_2+t'_2-2\tau_2)/2],\\S_3(t_1,t'_1)&=\theta(t_1-\tau_3)\theta(t'_1-\tau_3)\exp[i\omega_3(t_1-t'_1)-\Delta_3(t_1+t'_1-2\tau_3)/2],\end{aligned} \quad (22)$$

where

$$\theta(x)=\begin{cases}0, & if\ x<0,\\1, & if\ x\geq 0,\end{cases}$$

τ_1, τ_2, and τ_3 are the time moments when the pulses begin to pass through the impurity centre, Δ_1, Δ_2, and Δ_3 are the spectral full widths at half maximum (FWHM) of the pulses.

In the model used the rates of energy relaxation γ_1, γ_2, and γ_3 describe the relaxation processes of the excited levels 1, 2, and 3. In this model the phase relaxation and the phonon wings are not taken into account. Then the correlation function of the four-level system is

$$F(t,t_1,t'_1,t_2,t'_2,t_3,t'_3)=C\ \exp[-\gamma_3(2t-t_1-t'_1)/2-i\Omega_{23}(t_1-t'_1)-\gamma_2(t_1+t'_1-t_2-t'_2)/2-i\Omega_{12}(t_2-t'_2)$$
$$-\gamma_1(t_2+t'_2-t_3-t'_3)/2-i\Omega_{01}(t_3-t'_3), \quad (23)$$

where C is a constant.

Even in this elementary model there are 12 parameters that have an influence on the spectra.

4. MONOCHROMATIC LIGHT

If monochromatic light is used at all three steps of the absorption process, i.e. in Eq. (22), the FWHM spectral widths of the pulses $\Delta_1=\Delta_2=\Delta_3=0$ (stationary case). Then

$$W\approx\frac{1}{\left[(\Omega_{01}-\omega_1)^2+\gamma_1^2/4\right]}\frac{1}{\left[(\Omega_{01}+\Omega_{12}-\omega_1-\omega_2)^2+\gamma_2^2/4\right]}\frac{1}{\left[(\Omega_{01}+\Omega_{12}+\Omega_{23}-\omega_1-\omega_2-\omega_3)^2+\gamma_3^2/4\right]}. \quad (24)$$

In Eq. (24) the first term describes the absorption of light with frequency ω_1 between the levels $0\to1$, the second term describes the absorption of light with frequency $\omega_1 + \omega_2$ between the levels $0 \to 2$, and the third term describes the absorption of light with frequency $\omega_1 + \omega_2 + \omega_3$ between the levels $0 \to 3$. In the case where the frequencies ω_1 and ω_2 are fixed and the frequency ω_3 varies, only the line between the levels $0 \to 3$ exists, which corresponds to the coherent contribution. The width of this line depends only on the rate of energy relaxation γ_3 of the final state 3. It is so only in this model. If we take into account the phase relaxation of levels 1 and 2, other two lines, which correspond to the non-coherent contribution, will appear. If the light is not monochromatic, i.e. spectral widths of the coherent pulses $\Delta_1 \neq 0$, $\Delta_2 \neq 0$, and $\Delta_3 \neq 0$, these other two lines will appear even though the phase relaxation is not taken into account (see Chapter 6). In [7] it is shown that the ratio of non-coherent and coherent parts, i.e. the ratio of luminescence and scattering of resonant secondary radiation (the process between levels $0\to1\to0$, the second order of perturbation theory), is determined by the ratio of the rates of the phase relaxation and the rate of the energy relaxation of the excited level 1.

5. ULTRASHORT PULSES

1. The first pulse is much shorter than the relaxation time of the second level γ_1. Then

$$S_1(t_3,t'_3) \approx \delta(t_3-\tau_1)\delta(t'_3-\tau_1)\exp[i\omega_1(t_3-t'_3)], \tag{25}$$

$$W(t) = \int_{\tau_1}^{t} dt_1 \int_{\tau_1}^{t} dt'_1 S_3(t_1,t'_1) \int_{\tau_1}^{t_1} dt_2 \int_{\tau_1}^{t'_1} dt'_2 S_2(t_2,t'_2) F(t,t_1,t'_1,t_2,t'_2,\tau_1,\tau_1). \tag{26}$$

Equation (26) describes two-step absorption between levels $1\to2\to3$. The initial level 1 is excited at the time moment τ_1. In our model with correlation functions from Eqs (22) and (23) the probability is the following:

$$\begin{aligned}
W(t) \approx{}& \frac{1}{\left[y^2+(\Delta_2+\gamma_1-\gamma_2)^2/4\right]}\left\{\frac{\exp\left[-\gamma_1(\tau_2-\tau_1)\right]}{\left[z^2+(\Delta_3+\gamma_2-\gamma_3)^2/4\right]}\right.\\
&\times\Big[\exp\big(-\gamma_2(\tau_3-\tau_2)-\gamma_3(t-\tau_3)\big)+\exp\big(-\Delta_3(t-\tau_3)-\gamma_2(t-\tau_2)\big)\\
&-2\cos\big(z(t-\tau_3)\big)\exp\big(-(\Delta_3+\gamma_3)(t-\tau_3)/2-\gamma_2(t+\tau_3-2\tau_2)/2\big)\Big]\\
&+\frac{1}{\left[(y+z)^2+(\Delta_2+\Delta_3+\gamma_1-\gamma_3)^2/4\right]}\\
&\times\Big[\exp\big(-\Delta_2(\tau_3-\tau_2)-\gamma_1(\tau_3-\tau_1)-\gamma_3(t-\tau_3)\big)+\exp\big(-\Delta_2(t-\tau_2)-\Delta_3(t-\tau_3)-\gamma_1(t-\tau_1)\big)\\
&-2\cos\big((y+z)(t-\tau_3)\big)\exp\big(-\Delta_2(t+\tau_3-2\tau_2)/2-\Delta_3(t-\tau_3)/2-\gamma_1(t+\tau_3-2\tau_1)/2-\gamma_3(t-\tau_3)/2\big)\Big]\\
&-\Bigg[\frac{\exp\big(iy(\tau_3-\tau_2)\big)}{\big(iz+(\Delta_3+\gamma_2-\gamma_3)/2\big)\big(-i(y+z)+(\Delta_2+\Delta_3+\gamma_1-\gamma_3)/2\big)}\\
&\times\Big(\exp\big(-\gamma_2(\tau_3-\tau_2)/2-\gamma_3(t-\tau_3)/2\big)-\exp\big(-iz(t-\tau_3)-\Delta_3(t-\tau_3)/2-\gamma_2(t-\tau_2)/2\big)\Big)\\
&\times\Big(\exp\big(-\Delta_2(\tau_3-\tau_2)/2-\gamma_1(\tau_3+\tau_2-2\tau_1)/2-\gamma_3(t-\tau_3)/2\big)\\
&\left.-\exp\big(i(y+z)(t-\tau_3)-\Delta_2(t-\tau_2)/2-\Delta_3(t-\tau_3)/2-\gamma_1(t+\tau_2-2\tau_1)/2\big)\Big)+c.c.\Bigg]\right\}.
\end{aligned} \tag{27}$$

Here $y \equiv \Omega_{12} - \omega_2$ and $z \equiv \Omega_{23} - \omega_3$.

2. When the second pulse is ultrashort

$$S_2(t_2,t'_2) \approx \delta(t_2-\tau_2)\delta(t'_2-\tau_2)\exp[i\omega_2(t_2-t'_2)], \tag{28}$$

then

$$W(t)=\int_{\tau_2}^{t} dt_1 \int_{\tau_2}^{t} dt'_1 S_3(t_1,t'_1) \int_{-\infty}^{\tau_2} dt_3 \int_{-\infty}^{\tau_2} dt'_3 F(t,t_1,t'_1,\tau_2,\tau_2,t_3,t'_3) S_1(t_3,t'_3). \tag{29}$$

Here we have multiplication of two one-step processes: 0→1 and 2→3, in the latter case level 2 is excited at moment τ_2. In our model the probability is the following:

$$\begin{aligned} W(t) \approx & \frac{1}{\left[x^2+(\Delta_1-\gamma_1)^2/4\right]}\left\{\exp\left[-\Delta_1(\tau_2-\tau_1)\right]+\exp\left[-\gamma_1(\tau_2-\tau_1)\right]\right. \\ & \left.-2\cos\left[x(\tau_2-\tau_1)\right]\exp\left[-(\Delta_1+\gamma_1)(\tau_2-\tau_1)/2\right]\right\} \\ & \times\frac{1}{\left[z^2+(\gamma_2+\Delta_3-\gamma_3)^2/4\right]}\left\{\exp\left[-\gamma_2(t-\tau_2)-\Delta_3(t-\tau_3)\right]+\exp\left[-\gamma_2(\tau_3-\tau_2)-\gamma_3(t-\tau_3)\right]\right. \\ & \left.-2\cos\left[z(t-\tau_3)\right]\exp\left[-\gamma_2(t+\tau_3-2\tau_2)/2-(\Delta_3+\gamma_3)(t-\tau_3)/2\right]\right\}. \end{aligned} \tag{30}$$

Here $x \equiv \Omega_{01}-\omega_1$ and $z \equiv \Omega_{23}-\omega_3$.

3. In the case where the third pulse is ultrashort,

$$S_3(t_1,t'_1) \approx \delta(t_1-\tau_3)\delta(t'_1-\tau_3)\exp[i\omega_3(t_1-t'_1)] \tag{31}$$

and

$$W(t)=\int_{-\infty}^{\tau_3} dt_2 \int_{-\infty}^{\tau_3} dt'_2 S_2(t_2,t'_2) \int_{-\infty}^{t_2} dt_3 \int_{-\infty}^{t'_2} dt'_3 F(t,\tau_3,\tau_3,t_2,t'_2,t_3,t'_3) S_1(t_3,t'_3). \tag{32}$$

Here two-step absorption 0→1→2 takes place. In our model the probability is the following:

$$\begin{aligned} W(t) \approx & \frac{\exp\left[-\gamma_3(t-\tau_3)\right]}{\left[x^2+(\Delta_1-\gamma_1)^2/4\right]}\left\{\frac{1}{\left[y^2+(\Delta_2+\gamma_1-\gamma_2)^2/4\right]}\right. \\ & \times\left[\exp\left(-\gamma_1(\tau_2-\tau_1)-\gamma_2(\tau_3-\tau_2)\right)+\exp\left(-\Delta_2(\tau_3-\tau_2)-\gamma_1(\tau_3-\tau_1)\right)\right. \\ & \left.-2\cos\left(y(\tau_3-\tau_2)\right)\exp\left(-(\Delta_2+\gamma_2)(\tau_3-\tau_2)/2-\gamma_1(\tau_2+\tau_3-2\tau_1)/2\right)\right] \\ & +\frac{1}{\left[(x+y)^2+(\Delta_1+\Delta_2-\gamma_2)^2/4\right]} \\ & \times\left[\exp\left(-\Delta_1(\tau_2-\tau_1)-\gamma_2(\tau_3-\tau_2)\right)+\exp\left(-\Delta_1(\tau_3-\tau_1)-\Delta_2(\tau_3-\tau_2)\right)\right. \\ & \left.-2\cos\left((x+y)(\tau_3-\tau_2)\right)\exp\left(-\Delta_1(\tau_2+\tau_3-2\tau_1)/2-\Delta_2(\tau_3-\tau_2)/2-\gamma_2(\tau_3-\tau_2)/2\right)\right] \\ & -\left[\frac{\exp\left(\left(ix-(\Delta_1+\gamma_1)/2\right)(\tau_2-\tau_1)\right)}{\left(iy+(\Delta_2+\gamma_1-\gamma_2)/2\right)\left(-i(x+y)+(\Delta_1+\Delta_2-\gamma_2)/2\right)}\right. \\ & \times\left(\exp\left(-\gamma_2(\tau_3-\tau_2)/2\right)-\exp\left(\left(-iy-(\Delta_2+\gamma_1)/2\right)(\tau_3-\tau_2)\right)\right) \\ & \left.\left.\times\left(\exp\left(-\gamma_2(\tau_3-\tau_2)/2\right)-\exp\left(\left(i(x+y)-(\Delta_1+\Delta_2)/2\right)(\tau_3-\tau_2)\right)\right)+c.c.\right]\right\}. \end{aligned} \tag{33}$$

Here $x \equiv \Omega_{01}-\omega_1$ and $y \equiv \Omega_{12}-\omega_2$.

6. RESULTS OF CALCULATIONS

Due to an abundance of parameters, only some characteristic spectra are presented. Hereafter $\tau_1 = 0$, $T_1 \equiv \tau_2 - \tau_1$, and $T_2 \equiv \tau_3 - \tau_2$. Here we analysed the case where $z \equiv \Omega_{23} - \omega_3$ is variable (the frequency of the maximum of the third pulse ω_3 varies) and $x \equiv \Omega_{01} - \omega_1$ and $y \equiv \Omega_{12} - \omega_2$ are fixed (the frequencies of the maxima of the first and second pulses ω_1 and ω_2 are fixed) and chosen different from zero to separate spectroscopically possible lines.

The dependence of the spectrum on the parameter Δ_2 at a fixed time t is presented in Fig. 1. We get three lines with maxima: at $z = -(x + y)$, $z = -y$, and $z = 0$. The location of the maximum of the first line from the right at $z = -(x + y)$ ($\omega_3 = \Omega_{01} + \Omega_{12} + \Omega_{23} - \omega_1 - \omega_2$) shows that the three pulses are absorbed together so that $x + y + z = 0$ ($\omega_1 + \omega_2 + \omega_3 = \Omega_{01} + \Omega_{12} + \Omega_{23}$) without excitation of levels 1 and 2 and therefore this is the coherent contribution. The locations of the maxima of the other two lines, at $z = -y$ ($\omega_3 = \Omega_{12} + \Omega_{23} - \omega_2$) and $z = 0$ ($\omega_3 = \Omega_{23}$), show that absorption takes place correspondingly $0 \to 1 \to 3$ and $0 \to 1 \to 2 \to 3$ or $0 \to 2 \to 3$ and therefore these are not coherent contributions. In the first case the second and the third pulse are absorbed together from level 1. With the increase of the spectral width of the second pulse Δ_2 the intensity of the line at $z = 0$ increases and the intensities of the other lines decrease.

In Fig. 2 time t is much longer compared with Fig. 1. In this case the width of the line with the maximum at $z = -(x + y)$ (corresponds to the coherent contribution) diminishes, the limit width of this line is determined by the spectral widths of the pulses Δ_1, Δ_2, and Δ_3 and with the energy relaxation constant γ_3 of the excited electronic level 3. From a certain time t with the increase of time t the intensity and the width of the lines decrease. In Fig. 2 curve 1 ($\Delta_2 = 0$) is 3.2 times as large as the corresponding curve 1 in Fig. 1.

Figure 3 illustrates the dependence of the spectrum on the parameter γ_2 at different values of time t.

Analysis of these three figures and the other spectra with different parameters shows that the limit width of the line with maximum at $z = -y$ is determined by the spectral widths of the pulses Δ_2 and Δ_3 and by the energy relaxation constants γ_1 and γ_3. The limit width of the line with maximum at $z = 0$ is determined by the spectral width of the pulse Δ_3 and by the energy relaxation constants γ_2 and γ_3.

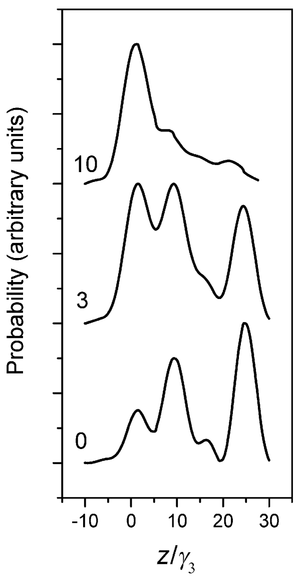

Fig. 1. Dependence of the probability $W(t)$ on $z \equiv \Omega_{23} - \omega_3$ at the fixed value of $x \equiv \Omega_{01} - \omega_1 = -15\gamma_3$, $y \equiv \Omega_{12} - \omega_2 = -10\gamma_3$ for different values of Δ_2 (in γ_3). $\Delta_1 = 0.1\gamma_3$, $\Delta_3 = \gamma_3$, $\gamma_1 = 5\gamma_3$, $\gamma_2 = 5\gamma_3$, $T_1 = 10^{-7}\gamma_3^{-1}$, $T_2 = 0.1\gamma_3^{-1}$, $t = 1.1\gamma_3^{-1}$. All curves are normalized to 1.

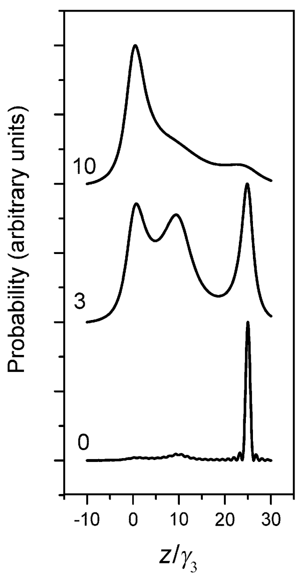

Fig. 2. Dependence of the probability $W(t)$ on $z \equiv \Omega_{23} - \omega_3$ at the fixed value of $x \equiv \Omega_{01} - \omega_1 = -15\gamma_3$, $y \equiv \Omega_{12} - \omega_2 = -10\gamma_3$ for different values of Δ_2 (in γ_3). $\Delta_1 = 0.1\gamma_3$, $\Delta_3 = \gamma_3$, $\gamma_1 = 5\gamma_3$, $\gamma_2 = 5\gamma_3$, $T_1 = 10^{-7}\gamma_3^{-1}$, $T_2 = 0.1\gamma_3^{-1}$, $t = 5.1\gamma_3^{-1}$. All curves are normalized to 1.

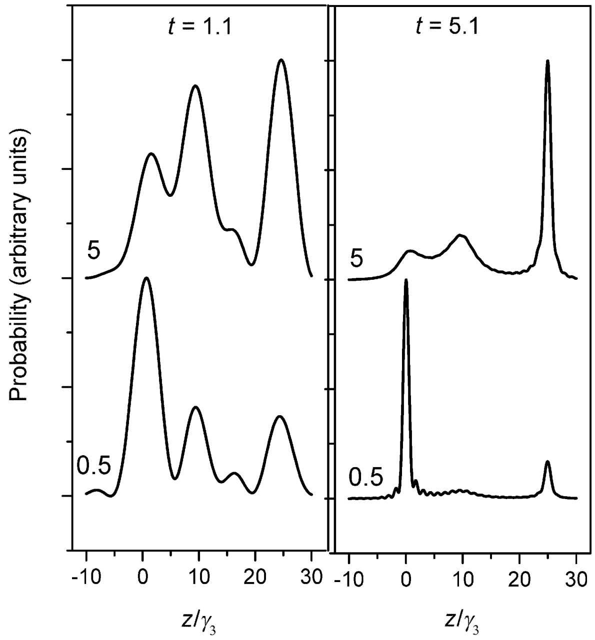

Fig. 3. Dependence of the probability $W(t)$ on $z \equiv \Omega_{23} - \omega_3$ at the fixed value of $x \equiv \Omega_{01} - \omega_1 = -15\gamma_3$, $y \equiv \Omega_{12} - \omega_2 = -10\gamma_3$ for different values of γ_2 (in γ_3) and t (in γ_3^{-1}). $\Delta_1 = 0.1\gamma_3$, $\Delta_2 = \gamma_3$, $\Delta_3 = \gamma_3$, $\gamma_1 = 5\gamma_3$, $\gamma_2 = 5\gamma_3$, $T_1 = 10^{-7}\gamma_3^{-1}$, $T_2 = 0.1\gamma_3^{-1}$. All curves are normalized to 1.

In the case of our correlation functions of the excitation pulses the limit values of FWHM of the lines are the following (ω_1 and ω_2, i.e. x and y, are fixed):

$$\sigma_{\max \text{ at } z=0} \equiv \lim_{|t-\tau_3|\to\infty} \sigma_{z=0}(T,t) = \gamma_2 + |\Delta_3 - \gamma_3| \quad (\textit{the non-coherent contribution}),$$
$$\sigma_{\max \text{ at } z=-y} \equiv \lim_{|t-\tau_3|\to\infty} \sigma_{z=-y}(T,t) = \gamma_1 + \Delta_2 + |\Delta_3 - \gamma_3| \quad (\textit{the non-coherent contribution}), \tag{34}$$
$$\sigma_{\max \text{ at } z=-x-y} \equiv \lim_{|t-\tau_3|\to\infty} \sigma_{z=-x-y}(T,t) = \Delta_1 + \Delta_2 + |\Delta_3 - \gamma_3| \quad (\textit{the coherent contribution}).$$

The other parameters influence only the intensities of these lines.

Analysis shows that in the general case where the spectral widths of the pulses Δ_1, Δ_2, and Δ_3 are comparable with the energy relaxation constants γ_1, γ_2 , and γ_3, if the frequency of the maximum of the first pulse ω_1 (ω_2, ω_3 are fixed) is variable, three lines with the maxima at $x = 0$, $x = -y$, and $x = -y - z$ exist in the spectra. In the case where the frequency of the maximum of the second pulse ω_2 (ω_1 and ω_3 are fixed) is variable, four lines with the maxima at $y = 0$, $y = -x$, $y = -z$, and $y = - x - z$ exist in the spectra.

7. CONCLUSIONS

A time-dependent theory of three-step absorption of three different light pulses with arbitrary duration in the electronic four-level model is proposed. The probability that the fourth level is excited at the time moment t is found depending on the time delays between pulses T_1 and T_2, the spectral widths of the pulses Δ_1, Δ_2, and Δ_3, and the energy relaxation constants γ_1, γ_2, and γ_3 of the excited electronic levels 1, 2, and 3.

In calculations the pulses are taken as coherent and of a single-sided exponential shape; ω_1, ω_2, and ω_3 are the frequencies of the maxima; 0, T_1, and $T_1 + T_2$ are the time moments when the pulses begin to pass through the impurity centre. The resonance conditions are $\omega_1 \approx \Omega_{01}$, $\omega_2 \approx \Omega_{12}$, and $\omega_3 \approx \Omega_{23}$ where Ω_{01}, Ω_{12}, and Ω_{13} are the frequencies of the transitions $0 \to 1$, $1 \to 2$, and $2 \to 3$ and ω_1, ω_2, and ω_3 are the frequencies of the maxima of the pulses.

In the general case (the spectral widths of the pulses Δ_1, Δ_2, and Δ_3 are comparable with the energy relaxation constants γ_1, γ_2, and γ_3), if the frequency ω_1 or the frequency ω_3 is variable (ω_2 and ω_3 are fixed or ω_1 and ω_3 are fixed), three lines exist in the spectra. In the case where the frequency ω_2 (ω_1 and ω_3 are fixed) is variable (ω_1 and ω_3 are fixed), four lines exist in the spectra.

Analysis of the case where the frequencies ω_1 and ω_2 are fixed and the frequency of the maximum of the third pulse ω_3 varies, shows that the widths of three possible lines depend on different parameters. The limit width of the line with the maximum at $\omega_3 = \Omega_{01} + \Omega_{12} + \Omega_{23} - \omega_1 - \omega_2$ is determined by the spectral widths of the pulses Δ_1, Δ_2, and Δ_3 and by the energy relaxation constant γ_3 (the coherent contribution), the limit width of the second line with the maximum at $\omega_3 = \Omega_{12} + \Omega_{23} - \omega_2$ is determined by the spectral widths of the pulses Δ_2 and Δ_3 and by the energy relaxation constants γ_1 and γ_3, and the limit width of the third line with the maximum at $\omega_3 = \Omega_{23}$ is determined by the spectral width of the pulse Δ_3 and by the energy relaxation constants γ_2 and γ_3.

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Kolme valgusimpulsi kolmeastmelise neeldumise ajast sõltuv teooria

Inna Rebane

Kasutades ajast sõltuvat häiritusarvutust, on esitatud kolme valgusimpulsi kolmeastmelise neeldumise ajast sõltuv teooria elektroni neljanivoolistes süsteemides. Esimene, teine ja kolmas impulss on resonantsis vastavalt nivoo 0 ja nivoo 1, nivoo 1 ja nivoo 2 ning nivoo 2 ja nivoo 3 vaheliste üleminekutega. Tõenäosuse, et hetkel *t* on ergastatud nivoo 3, spektraalne ja ajaline käitumine sõltub ergastatud elektroni nivoode energia relaksatsiooni konstantidest, valgusimpulsside maksimumide sagedustest ja spektraalsetest laiustest (ja samuti kestusest) ning impulssidevahelistest ajalistest viivistest. Kokkuvõttes: kasutatud lihtsas mudelis on kaksteist parameetrit, mis mõjutavad spektreid. Arvutused on tehtud juhtumi jaoks, kus esimese kahe impulsi maksimumide sagedused on fikseeritud ja muutub kolmanda impulsi maksimumi sagedus. Üldjuhul võib spektris eksisteerida kolm spektraaljoont, üks neist vastab koherentsele panusele spektrisse ja teised kaks mittekoherentsele panusele.