Theory of three-step absorption of three light pulses

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Abstract. The theory of the absorption of three light pulses with an arbitrary duration in four-level systems is proposed. The pulses are in resonance with the corresponding transitions. By using the time-dependent perturbation theory the probability that the fourth level is excited at the moment t is found. The possible absorption lines and parameters on which their widths depend are analysed.

1. Introduction
Multistep absorption/excitation has been investigated in many ways and also both monochromatic light and light pulses have been used [1-7]. In [7] the possible time dependent two-step absorption spectra of two light pulses are theoretically investigated by using the simplest model for matter and in the case of an arbitrary duration of absorbed light pulses from monochromatic light to ultrashort pulses.

Thereby, we note that if at the two-step absorption in the three-level system the monochromatic light with the frequency \( \omega_1 \) is absorbed at the first step \( 0 \rightarrow 1 \), then in the spectrum where the frequency \( \omega_1 \) is fixed and the frequency \( \omega_2 \) of the maximum of the second light pulse (which is absorbed at the second step \( 1 \rightarrow 2 \)) varies, there exists one line whose width is determined by the rate of energy relaxation of the second excited level 2 and by the spectral width of the second pulse. In our opinion, this is exactly the same situation as in [6]. We note that if at the first step of the absorption the light pulse is absorbed and/or the pure phase relaxation of the first excited level exists, the second line is added to the aforementioned spectrum. The width of this line depends on the rates of the energy relaxations of both excited levels 1 and 2, the rate of the pure phase relaxation and on the spectral width of the second pulse, but not on the spectral width of the first pulse. On the other hand, the width of the initial line, in addition to the aforementioned parameters, depends on the spectral width of the first pulse. This line corresponds to the coherent contribution of the spectrum [3]. We note that the linewidths depend on the timeframes between the pulses and the time t as well.

In this paper the three-step absorption of three light pulses in a four-level system (the pulses are in resonance with the corresponding transitions) is observed to analyse which absorption lines are possible at all and which parameters their widths depend on. The interaction with the environment (phonons) is taken into account phenomenologically via the relaxation constants of the electronic levels. The presented model holds for impurity centers at low temperatures with weak electron-phonon coupling.
2. Starting formulas and model

The process starts from ground level 0. The resonance conditions are \( \omega_1 \approx \Omega_{01} \), \( \omega_2 \approx \Omega_{12} \) and \( \omega_3 \approx \Omega_{23} \), where \( \omega_1 \), \( \omega_2 \) and \( \omega_3 \) are the frequencies of the maximums of the pulses, \( \Omega_{01} \), \( \Omega_{12} \) and \( \Omega_{13} \) are the frequencies of the transitions between levels 0 \( \rightarrow \) 1, 1 \( \rightarrow \) 2 and 2 \( \rightarrow \) 3.

By using the time-dependent perturbation theory \([8, 9]\) analogically with the theory of the absorption of two light pulses in three-level systems \([7]\), the probability that at the time moment \( t \) all three pulses are absorbed and the fourth level 3 is excited is the following:

\[
W(t) = \int dt_1 \int dt_2 \int dt_3 S_1(t_1, t_3) \int dt_2' \int dt_3' S_2(t_2, t_3') \int dt_1' S_3(t_1, t_2, t_3) F(t, t_1, t_1', t_2, t_2', t_3, t_3'),
\]

(1)

where \( F \) is the correlation function of the four-level system and \( S_1, S_2 \) and \( S_3 \) are the correlation functions of the first, second and third pulses. This probability \( W(t) \) decreases to zero with the increase of time \( t \) because of energy relaxation of the fourth level.

To simplify the calculations, the light pulses are taken coherent and of a single-sided exponential shape. The corresponding correlation functions are

\[
S_1(t_1, t_3') = \theta(t_1 - t_1) \theta(t_1' - t_1) \Delta_1 \exp[i \omega_1 (t_1 - t_1') - \Delta_1 (t_1 + t_1' - 2t_1)/2],
\]

\[
S_2(t_2, t_3') = \theta(t_2 - t_2) \theta(t_2' - t_2) \Delta_2 \exp[i \omega_1 (t_2 - t_2') - \Delta_2 (t_2 + t_2' - 2t_2)/2],
\]

\[
S_3(t_1, t_1') = \theta(t_1 - t_1) \theta(t_1' - t_1) \Delta_3 \exp[i \omega_1 (t_1 - t_1') - \Delta_3 (t_1 + t_1' - 2t_1)/2], \tag{2}
\]

where \( \theta(x) \) is Heaviside step function, \( t_1, t_2 \) and \( t_3 \) are the time moments when the pulses begin to pass through the impurity centre, \( \Delta_1, \Delta_2 \) and \( \Delta_3 \) are the FWHM spectral widths of the pulses.

The correlation function of the four-level system is \([9]\)

\[
F(t, t_1, t_1', t_2, t_2', t_3, t_3') = \left< v_{\omega_1}^* \exp \left[ i \left( \hat{H} + \frac{i}{2} \hat{\gamma} \right) (t_2 - t_3) \right] v_{\omega_1}^* \right> \left< v_{\omega_2}^* \exp \left[ i \left( \hat{H} + \frac{i}{2} \hat{\gamma} \right) (t_1 - t_2) \right] v_{\omega_2}^* \right> \times \exp \left[ -i \hat{H} \left( t_1' - t_1 \right) \right] v_{\omega_2} \exp \left[ -i \hat{H} \left( t_2' - t_1' \right) \right] v_{\omega_2} \times \exp \left[ -i \hat{H} \left( t_3' - t_3 \right) \right] v_{\omega_2} \exp \left[ -i \hat{H} \left( t_2' - t_3' \right) \right]. \tag{3}
\]

In (3) \( \hat{H} \) is Hamiltonian and \( \hat{\gamma} \) is the operator of the radiation decay of the four-level system, \( \langle \ldots \rangle \) is the symbol of averaging over the ensemble of vibrations on the initial electronic level, and the single-photon matrix elements of the interaction Hamilton \( \hat{V} \) of the field and matter are

\[
v_{\omega_1} = \left< N - 1_{\omega_1} \hat{V} \right| N \rangle, v_{\omega_2} = \left< N - 1_{\omega_1} - 1_{\omega_2} \hat{V} \right| N - 1_{\omega_2} \rangle,
\]

\[
v_{\omega_3} = \left< N - 1_{\omega_1} - 1_{\omega_2} - 1_{\omega_3} \hat{V} \right| N - 1_{\omega_3} \rangle. \tag{4}
\]

They describe the annihilation of the photons of the frequencies \( \omega_1, \omega_2 \) and \( \omega_3 \). In (4) \( |N\rangle \) is the initial state of the electromagnetic field. In our elementary model the phase relaxation and the phonon wings
are not taken into account, the relaxation processes of the excited levels 1, 2 and 3 are described by the rates of energy relaxation $\gamma_1$, $\gamma_2$, and $\gamma_3$. Thus the interaction with phonons is taken into account phenomenologically via the relaxation constants of the electronic levels.

Then the correlation function of the four-level system is

$$
F(t, t_1, t_1', t_2, t_2', t_3, t_3') = C \exp\left[-\gamma_3(2t - t_1 - t_1') / 2 - i\Omega_{01}(t_1 - t_1') - \gamma_2(t_1 + t_1' - t_2 - t_2') / 2 - i\Omega_{12}(t_2 - t_2') - \gamma_1(t_2 + t_2' - t_3 - t_3') / 2 - i\Omega_{03}(t_3 - t_3')\right],
$$

where $C$ is a constant.

In summary we have twelve parameters/variables which have influence on the spectra ($\Delta_1$, $\Delta_2$, $\Delta_3$, $\tau_2 - \tau_1$, $\tau_3 - \tau_2$, $t - \tau_3$, $\omega_1 - \Omega_{01}$, $\omega_2 - \Omega_{12}$, $\omega_3 - \Omega_{23}$, $\gamma_1$, $\gamma_2$, and $\gamma_3$).

3. Results

Subsequently, $t_1 = 0$, $T_1 = t_2 - \tau_1$, $T_2 = t_3 - \tau_2$, $x = \Omega_{01} - \omega_1$, $y = \Omega_{12} - \omega_2$, and $z = \Omega_{23} - \omega_3$. In figures 1–3 the spectral widths of light pulses $\Delta_1$, $\Delta_3$, the time intervals $T_1$, $T_2$, the time $t$ and the relaxation constants $\gamma_1$, $\gamma_2$, and $\gamma_3$ are the same.

In figure 1 the dependence of the probability $W(t)$ on the frequency of the maximum of the first pulse $\omega_1$ is calculated, the other frequencies $\omega_2$, $\omega_3$ are fixed and the quantities $\omega_2 - \Omega_{12}$, $\omega_1 - \Omega_{23}$ are chosen different from zero to separate possible lines spectroscopically.

In figure 2 the dependence of the probability $W(t)$ on the frequency of the maximum of the second pulse $\omega_2$ is presented, the other frequencies $\omega_1$, $\omega_3$ are fixed.

![Figure 1](image1.png) ![Figure 2](image2.png)

Figure 1. Probability $W(t)$ for different values of $\Delta_2$, $x = \Omega_{01} - \omega_1$ is variable, $y = -10\gamma_3$, $z = -15\gamma_3$. Curve 1 $- \Delta_2 = 0$, curve 2 $- \Delta_2 = 3\gamma_3$, curve 3 $- \Delta_2 = 10\gamma_3$, $\Delta_1 = 0.1\gamma_3$, $\Delta_3 = \gamma_3$, $\gamma_1 = \gamma_2 = 5\gamma_3$, $T_1 = T_2 = \gamma_3^{-1}$, $t = 5.1\gamma_3^{-1}$. Curves are normalized.

Figure 2. Probability $W(t)$ for different values of $\Delta_2$, $y = \Omega_{12} - \omega_2$ is variable. $x = -15\gamma_3$, $z = -10\gamma_3$. Curve 1 $- \Delta_2 = 0$, curve 2 $- \Delta_2 = 3\gamma_3$, curve 3 $- \Delta_2 = 10\gamma_3$. The other parameters are the same as in figure 1. Curves are normalized to 1.
In figure 3 the dependence of the probability $W(t)$ on the frequency of the maximum of the third pulse $\omega_3$ is calculated, the other frequencies $\omega_1$, $\omega_2$ are fixed. At different values of the spectral width of the second pulse $\Delta_2$ different lines exist. When the frequency $\omega_3$ is variable, then the possible maximums of the lines are at $x = 0$, $x = -y$ (in figure 1 $x = 10\gamma_3$) and $x = -y - z$ (in figure 1 $x = 25\gamma_3$). When the frequency $\omega_2$ is variable, the possible maximums of the lines are at $y = 0$, $y = -x$ (in figure 2 $y = 15\gamma_3$), $y = -z$ and $y = -x - z$ (in figure 2 $y = 25\gamma_3$). Finally, when the frequency $\omega_3$ is variable, the possible maximums of the lines are at $z = 0$, $z = -y$ and $z = -x - y$ (in figure 3 $z = 25\gamma_3$).

**Figure 3.** Probability $W(t)$ for different values of $\Delta_2$, $z \equiv \Omega_{23} - \omega_3$ is variable. $x = -15\gamma_3$, $y = -10\gamma_3$. Curve 1 – $\Delta_2 = 0$, curve 2 – $\Delta_2 = 3\gamma_3$, curve 3 – $\Delta_2 = 10\gamma_3$. The other parameters are the same as in figure 1. Curves are normalized to 1.

Figure 4 shows the case where $y \equiv \Omega_{12} - \omega_2$ varies, in which all four possible lines are visible.
The widths of the different lines depend on different parameters (see table). It can be seen from the Table that if \( z \) varies, the width of the line with the maximum at \( z = 0 \) depends on the parameters \( \gamma_1, \gamma_3 \) and \( \Delta_3 \); the width of the line with the maximum at \( z = -y \) depends on the parameters \( \gamma_1, \gamma_3, \Delta_2 \) and \( \Delta_3 \); the width of the line with the maximum at \( z = -x - y \) depends on the parameters \( \gamma_3, \Delta_1, \Delta_2 \) and \( \Delta_3 \).

In the case, where the monochromatic light (\( \Delta_1 = 0 \)) is used at the first step of the absorption (0 → 1) in the spectrum where \( x \) varies, all three lines remain, in the spectrum where \( y \) varies the lines with the maximums at \( y = -x \) and \( y = -x - z \) remain, and in the spectrum where \( z \) varies the lines with the maximums at \( z = 0 \) and \( z = -x - y \) remain.

If, additionally, the monochromatic light is used at the second step of the absorption (1 → 2) in the spectrum where \( z \) is a variable, the line with the maximum at \( z = 0 \) disappears as well.

Note, that the lines for which the equation \( x + y + z = 0 \) applies are the coherent parts of the spectrums. Furthermore, the widths of all lines depend on the timespans \( T_1, T_2 \) and the time \( t \). In general, with the increase of \( T_1, T_2 \) and \( t \) the lines become narrower.

**Table.** The dependence of the widths of different lines on different parameters

| Locations of the maximums of the lines | \( \Delta_1 \neq 0, \Delta_2 \neq 0, \Delta_3 \neq 0 \) | \( \Delta_1 = 0, \Delta_2 \neq 0, \Delta_3 \neq 0 \) | \( \Delta_1 = \Delta_2 = 0, \Delta_3 \neq 0 \) | \( \Delta_1 = \Delta_2 = \Delta_3 = 0 \) |
|-------------------------------|---------------------------------|---------------------------------|---------------------------------|---------------------------------|
| \( x \) varies                 | \( y_1, \Delta_1 \)             | \( y_1 \)                        | \( y_1 \)                        | \( y_1 \)                        |
| \( x = 0 \)                     | \( y_2, \Delta_1 \)             | \( y_2 \)                        | -                               | -                               |
| \( x = -y \)                    | \( y_1, \Delta_1, \Delta_2 \)   | \( y_1, \Delta_2 \)             | \( y_2 \)                        | \( y_2 \)                        |
| \( x = -y - z \)                | \( y_3, \Delta_1, \Delta_2, \Delta_3 \) | \( y_3, \Delta_2, \Delta_3 \)   | \( y_3 \)                        | \( y_3 \)                        |
| \( y \) varies                 | \( y_1, y_2, \Delta_2 \)        | -                               | -                               | -                               |
| \( y = 0 \)                     | \( y_2, \Delta_1 \)             | \( y_2, \Delta_2 \)             | \( y_2 \)                        | \( y_2 \)                        |
| \( y = -x \)                    | \( y_1, y_3, \Delta_2, \Delta_3 \) | -                               | -                               | -                               |
| \( y = -z \)                    | \( y_3, \Delta_1, \Delta_2, \Delta_3 \) | \( y_3 \)                        | \( y_3 \)                        | \( y_3 \)                        |
| \( y = -x - z \)                | \( y_1, y_2, \Delta_3 \)        | \( y_2, \Delta_3 \)             | \( y_3 \)                        | \( y_3 \)                        |
| \( z \) varies                 | \( y_2, y_3, \Delta_3 \)        | \( y_2, y_3, \Delta_3 \)        | -                               | -                               |

**Figure 4.** Probability \( W(t) \) in the case where all four lines exist, \( y = \Omega_1 - \omega_2 \) is variable. \( x = -30\gamma_3, z = -20\gamma_3, \Delta_1 = \Delta_2 = \Delta_3 = \gamma_1 = \gamma_2 = \gamma_3, T_1 = 2\gamma_3^{-1}, T_2 = \gamma_3^{-1} \), \( t = 10.1\gamma_3^{-1} \). Curve is normalized to 1.
4. Conclusions

The time-dependence theory of three-step absorption of three different light pulses with an 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 to be depending on the time delays between the pulses $T_1$ and $T_2$, the spectral widths of the pulses $\Delta_1$, $\Delta_2$ and $\Delta_3$ and the energy relaxation constants $\gamma_1$, $\gamma_2$ and $\gamma_3$ of the excited electronic levels 1, 2 and 3. The time dependent perturbation theory is applied.

In the calculations the pulses are taken as coherent and of a single-sided exponential shape, $\omega_1$, $\omega_2$ and $\omega_3$ are the frequencies of the maximums, $0$, $T_1$ 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 $\Omega_{01}$, $\Omega_{12}$ and $\Omega_{13}$ are the frequencies of the transitions $0 \rightarrow 1$, $1 \rightarrow 2$ and $2 \rightarrow 3$.

In the general case, where the spectral widths of the pulses $\Delta_1$, $\Delta_2$ and $\Delta_3$ are comparable with the energy relaxation constants $\gamma_1$, $\gamma_2$ and $\gamma_3$, three lines may exist in the spectra in the cases where $\omega_1$ or $\omega_3$ are variable (corresponding $\omega_2$ and $\omega_3$ or $\omega_1$ and $\omega_2$ are fixed, figures 1 and 3). In the case where $\omega_2$ is variable there may exist four lines ($\omega_1$ and $\omega_3$ are fixed, figure 4). An analysis shows that the widths of the possible lines depend on different parameters (Table).

This work was supported by the European Union through the European Regional Development Fund (Centre of Excellence "Mesosystems: Theory and Applications", TK114) and by institutional research funding (IUT2-27,“Nonlinear theory of solids and fundamental fields“) of the Estonian Ministry of Education and Research.

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