Persistent Spectral Hole-Burning by Two Pulses of Arbitrary Duration

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Abstract: Two-step spectral hole-burning are considered in the cases of three- and four-level systems and an arbitrary duration of burning pulses. The shapes of the spectral holes (more exactly – the spectral hole-burning efficiencies that determined them) in inhomogeneous distribution functions of the centers over the transition frequencies of the first and the second steps are calculated. It is shown that with increasing the time delay between pulses the widths of the spectral holes in the inhomogeneous distribution function corresponding to the absorption of the pulses on end at the first step of this process decrease. This phenomenon does not depend on the duration of the second burning pulse and is absent at the second step of the process.

1 INTRODUCTION

The inhomogeneous broadening of the spectra of the electronic transitions in the impurity systems can be eliminated by using a method of the spectral hole-burning (SHB) (Kharlamov, Personov and Bykovskaya, 1974; Gorokhovskii, Kaarli and Rebane LA, 1976; Moerner, 1988). The first experimental works on the two-step SHB in the three- (Winnacker, Shleby and Macfarline, 1985) and the four-level (Friedrich and Haarer, 1984; Lee, Gertz, Marinero and Moerner, 1985) systems were published about 25 years ago. The spectral holes in the inhomogeneous distribution functions (IDF) are determined through SHB efficiencies (Rebane LA, Gorokhovskiy and Kikas, 1982) (e.g. exponentially or linearly). In this work we use the second and the third orders of the perturbation theory for calculating SHB efficiencies in the three- and four-level systems correspondingly; the durations of the pulses are arbitrary and computer calculations are done.

2 SHB EFFICIENCIES

The electronic levels are described by the rates of the energy relaxation γ_0, γ_1, $\gamma_{1'}$, and γ_2. The first pulse with arbitrary duration is applied between the levels 0 and 1 and the second pulse with arbitrary duration is applied between the levels 1 and 2 (for the three-level system) or the levels 1' and 2 (for the four-level system) (Figure 1). The inhomogeneous distributions of the frequencies Ω_{01} and Ω_{12} (for the three-level system) or $\Omega_{1'2}$ (for the four-level system) of the transitions $0 \rightarrow 1$ and $1 \rightarrow 2$ (for the three-level system) or $1' \rightarrow 2$ (for the four-level system) are taken into account.

SHB efficiencies $P_3(\Omega_{01}, \Omega_{12}, t)$ and $P_4(\Omega_{01}, \Omega_{1'2}, t)$ at sufficiently small excitation intensities can be considered in the second and the third orders of the perturbation theory,

$$P_3(\Omega_{01},\Omega_{12},t) = \alpha \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt_1 \int_{-\infty}^{t'} dt'_1 S_2(t_1,t'_1) \times \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t'_1} dt'_2 S_1(t_2,t'_2) F_3(t',t_1,t'_1,t_2,t'_2), \quad (1)$$

$$P_4(\Omega_{01},\Omega_{1'2},t) = \alpha \int_{-\infty}^{t} dt' \int_{-\infty}^{t'} dt_1 \int_{-\infty}^{t'} dt'_1 S_2(t_1,t'_1) \times \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t'_1} dt'_2 G(t_2,t'_2) \int_{-\infty}^{t_2} dt_3 \int_{-\infty}^{t'_2} dt'_3 S_1(t_3,t'_3) \times F_4(t',t_1,t'_1,t_2,t'_2,t_3,t'_3). \quad (2)$$

In (1) and (2) α is the photochemical quantum yield, F_3 and F_4 are the correlation functions of the three-level and the four-level systems, S_1 and S_2 are the correlation functions of the first and the second pulses. The correlation function G in (2) describes

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the relaxation processes from the level 1 to the level 1' in the four-level system.

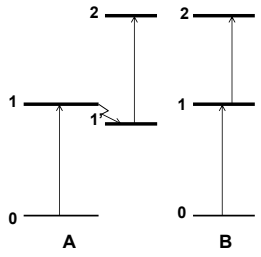

Figure 1: Scheme of SHB in the four- (A) and the three-level (B) systems by the pulses.

We use the following correlation functions for exciting pulses:

$$S_1(t_3,t'_3)=\theta(t_3-\tau_1)\theta(t'_3-\tau_1)\Delta_1\exp[i\omega_1(t_3-t'_3 -\Delta_1(t_3+t'_3-2\tau_1)/2],$$
$$S_2(t_1,t'_1)=\theta(t_1-\tau_2)\theta(t'_1-\tau_2)\Delta_2\exp[i\omega_2(t_1-t'_1 -\Delta_2(t_1+t'_1-2\tau_2)/2] \quad (3)$$

(the pulses are coherent and of a single-sided exponential shape) where ω_1 and ω_2 are the frequencies of the maximums, and Δ_1 and Δ_2 are the fwhm spectral widths of the pulses, τ_1 and τ_2 are the time moments when the pulses begin to pass through the impurity centre.

The correlation functions of the three- and the four-level systems are the following:

$$F_3(t',t_1,t'_1,t_2,t'_2)=C_3\exp[-\gamma_2(2t'-t_1-t'_1)/2 -i\Omega_{12}(t_1-t'_1)-\gamma_1(t_1+t'_1-t_2-t'_2)/2 -i\Omega_{01}(t_2-t'_2)-\gamma_0(t_2+t'_2)/2], \quad (4)$$

$$F_4(t',t_1,t'_1,t_2,t'_2,t_3,t'_3)=C_4 \times\exp[-\gamma_2(2t'-t_1-t'_1)/2-i\Omega_{1'2}(t_1-t'_1) -\gamma_{1'}(t_1+t'_1-t_2-t'_2)/2-i\Omega_{11'}(t_2-t'_2) -\gamma_1(t_2+t'_2-t_3-t'_3)/2-i\Omega_{01}(t_3-t'_3) -\gamma_0(t_3+t'_3)/2], \quad (5)$$

where C_3 and C_4 are constants. In the correlation function

$$G(t_2,t'_2)=\theta(t_1-\tau_1)\theta(t'_1-\tau_1)g \times\exp[-g(t_1+t'_1-2\tau_1)/2] \quad (6)$$

the parameter g describes the rate of the energy transfer from the level 1 to the level 1'.

IDF $\rho_3(\Omega_{01}, \Omega_{12}, t)$ (or $\rho_4(\Omega_{01}, \Omega_{1'2}, t)$) takes into account the inhomogeneous distribution of the frequencies and under certain assumptions changes exponentially with time (Rebane LA, Gorokhovskiy and Kikas, 1982),

$$\rho_3(\Omega_{01},\Omega_{12},t)=\rho_{30}(\Omega_{01},\Omega_{12})\exp[-P_3(\Omega_{01},\Omega_{12},t)], \quad (7)$$

$$\rho_4(\Omega_{01},\Omega_{1'2},t)=\rho_{40}(\Omega_{01},\Omega_{1'2})\exp[-P_4(\Omega_{01},\Omega_{1'2},t)]. \quad (8)$$

Here ρ_{30} (ρ_{40}) is the inhomogeneous distribution function before the beginning of SHB, whereas the regions of the frequencies Ω_{01} and Ω_{12} ($\Omega_{1'2}$) do not overlap. Under small irradiation doses

$$\rho_3(\Omega_{01},\Omega_{12},t)\approx\rho_{30}(\Omega_{01},\Omega_{12})[1-P_3(\Omega_{01},\Omega_{12},t)], \quad (9)$$

$$\rho_4(\Omega_{01},\Omega_{1'2},t)\approx\rho_{40}(\Omega_{01},\Omega_{1'2})[1-P_4(\Omega_{01},\Omega_{1'2},t)]. \quad (10)$$

The final spectral holes we get at $t\to\infty$:

$$P_3(\Omega_{01},\Omega_{12})\equiv\lim_{t\to\infty}P_3(\Omega_{01},\Omega_{12},t) \quad (11)$$

and

$$P_4(\Omega_{01},\Omega_{1'2})\equiv\lim_{t\to\infty}P_4(\Omega_{01},\Omega_{1'2},t). \quad (12)$$

3 RESULTS

In Figure 2, SHB efficiency $P_3(\Omega_{01})$ for the fixed value of $\Omega_{12}-\omega_2$ is calculated for different values of the time delay $T=\tau_2-\tau_1$ between the pulses. With increasing the time delay T, the spectral line with the maximum at $\Omega_{01}=\omega_1$ monotonously narrows down. The spectral line with the maximum at $\Omega_{01}-\omega_1=15\gamma_1$ corresponds to absorption of both the pulses at the same time. This spectral line appears only when the pulses overlap, i.e. at $T=0.5\gamma_1^{-1}$, and also little bit at $T=2.5\gamma_1^{-1}$.

For the four-level system with the same parameters as in Figure 2, SHB efficiency $P_4(\Omega_{01})$ is calculated in Figure 3. In the case of $\Omega_{11'}=-10\gamma_1$ the third spectral line appears with the maximum at $\Omega_{01}-\omega_1=-10\gamma_1$. With increasing the time delay T between the pulses the same monotonous narrowing of both the spectral lines takes place. In Figure 4 the parameter $g=2\gamma_1$, unlike Figure 3 where $g=0$, which leads to result that in Figure 4 the spectral line with the maximum at $\Omega_{01}-\omega_1=-10\gamma_1$ is larger than in Figure 3. Differently from Figure 3, in Figure 4 the spectral line with maximum at $\Omega_{01}=\omega_1$ is already small in the case of the time delay $T=2.5\gamma_1^{-1}$ and is absent at $T=4.5\gamma_1^{-1}$. The spectral lines monotonously narrow down with increasing the time delay but at $T=4.5\gamma_1^{-1}$ the width of the spectral line with the maximum at $\Omega_{01}-\omega_1=-10\gamma_1$ is larger than in Figure 3.

In Figure 5, SHB efficiency $P_3(\Omega_{12})$ for the fixed value of $\Omega_{01}-\omega_1$ is calculated. The shape of the curve of this SHB efficiency changes a little at different values of the time delay T, only the

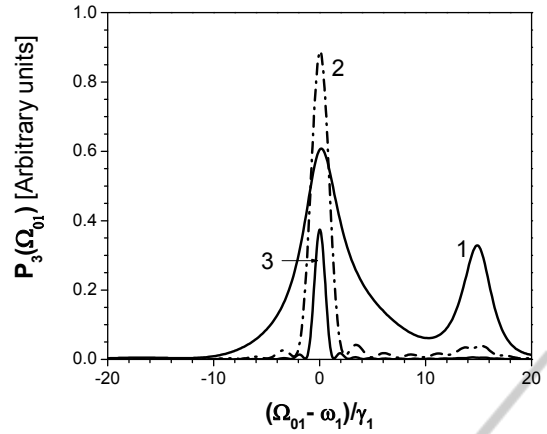

Figure 2: SHB efficiency in the three-level system $P_3(\Omega_{01})$ for the fixed value of $\Omega_{12} - \omega_2 = -15\gamma_1$ at the different time delays T between the pulses: 1 – $T = 0.5\gamma_1^{-1}$, 2 – $T = 2.5\gamma_1^{-1}$, 3 – $T = 4.5\gamma_1^{-1}$. $\gamma_0 = 0.01\gamma_1$, $\gamma_2 = 1.5\gamma_1$, $\Delta_1 = \Delta_2 = \gamma_1$.

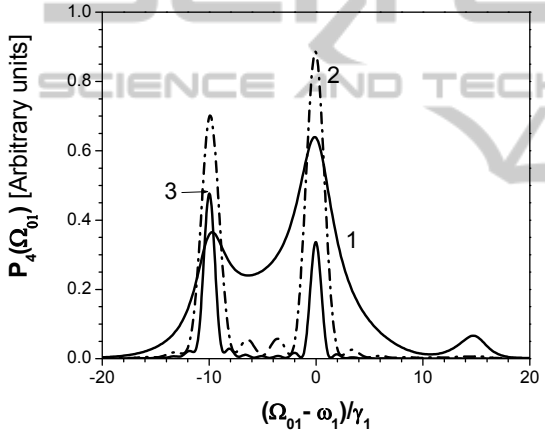

Figure 3: SHB efficiency in the four-level system $P_4(\Omega_{01})$ for the fixed value of $\Omega_{1'2} - \omega_2 = -25\gamma_1$ at the different time delays T between the pulses: 1 – $T = 0.5\gamma_1^{-1}$, 2 – $T = 2.5\gamma_1^{-1}$, 3 – $T = 4.5\gamma_1^{-1}$. $\gamma_0 = 0.01\gamma_1$, $\gamma_{1'} = 0.5\gamma_1$, $\gamma_2 = 1.5\gamma_1$, $\Delta_1 = \Delta_2 = \gamma_1$, $g = 0.001\gamma_1$, $\Omega_{11'} = -10\gamma_1$.

intensities of SHB efficiency are different at different T. In the case of the four-level system (Figure 6) the line of the absorption of two pulses together with the maximum at $\Omega_{1'2} - \omega_2 = 15\gamma_1$ appears only at the time delay $T = 0.5\gamma_1^{-1}$, the shape of the spectral line with the maximum at $\Omega_{1'2} = \omega_2$ does not change.

Thus, taking into account (9) and (10), we can say that at the first step of SHB process monotonous narrowing of the spectral hole in IDF takes place when the time delay T between the pulses increases. In this process the shape of the correlation function S_1 of the first pulse is important.

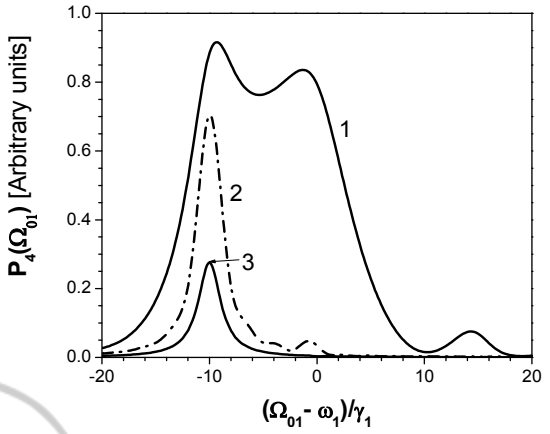

Figure 4: SHB efficiency in the four-level system $P_4(\Omega_{01})$ for the fixed value of $\Omega_{1'2} - \omega_2 = -25\gamma_1$ at the different time delays T between the pulses: 1 – $T = 0.5\gamma_1^{-1}$, 2 – $T = 2.5\gamma_1^{-1}$, 3 – $T = 4.5\gamma_1^{-1}$. $\gamma_0 = 0.01\gamma_1$, $\gamma_{1'} = 0.5\gamma_1$, $\gamma_2 = 1.5\gamma_1$, $\Delta_1 = \Delta_2 = \gamma_1$, $g = 2\gamma_1$, $\Omega_{11'} = -10\gamma_1$.

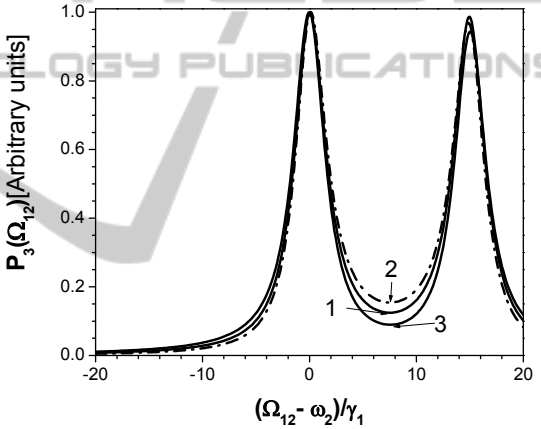

Figure 5: SHB efficiency in the three-level system $P_3(\Omega_{12})$ for the fixed value of $\Omega_{01} - \omega_1 = -15\gamma_1$ at the different time delays T between the pulses: 1 – $T = 0.5\gamma_1^{-1}$, 2 – $T = 2.5\gamma_1^{-1}$, 3 – $T = 4.5\gamma_1^{-1}$. $\gamma_0 = 0.01\gamma_1$, $\gamma_2 = 1.5\gamma_1$, $\Delta_1 = \Delta_2 = \gamma_1$. Here the curve 2 is enlarged 8.55 times and the curve 3 is enlarged 72.1 times.

In (Rebane IK, 1988) and (Rebane I, 2012) for the three- and the four-level systems correspondingly the same effect in the case of the extremely short second pulse (δ-pulse) was calculated analytically. The widths of these holes can be narrower than the width of the hole obtainable in one-step SHB processes by monochromatic light in two-level systems (so-called compensation effect). In the case where S_1 was defined by (3) for the spectral hole width we receive

$$\lim_{T\to\infty} \sigma(T) = \gamma_0 + |\gamma_1 - \Delta_1| + \Gamma_1. \qquad (13)$$

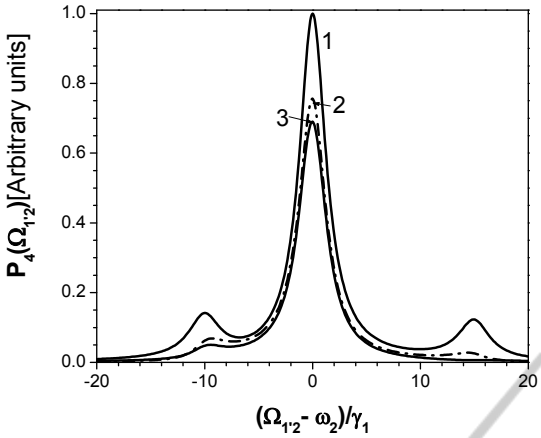

Figure 6: SHB efficiency in the four-level system $P_4(\Omega_{1'2})$ for the fixed value of $\Omega_{01} - \omega_1 = -25\gamma_1$ at the different time delays T between the pulses: 1 – $T = 0.5\gamma_1^{-1}$, 2 – $T = 2.5\gamma_1^{-1}$, 3 – $T = 4.5\gamma_1^{-1}$. $\gamma_0 = 0.01\gamma_1$, $\gamma_{1'} = 0.5\gamma_1$, $\gamma_2 = 1.5\gamma_1$, $\Delta_1 = \Delta_2 = \gamma_1$, $g = 0.001\gamma_1$, $\Omega_{11'} = -10\gamma_1$.

In (Rebane IK, 1988) the model with both rates of the relaxation of the first electronic level, the energy (γ_1) and the pure phase (Γ_1) was used. This model showed that the width conditioned by the pure phase relaxation Γ_1 adds to the spectral hole width σ without taking into account Γ_1.

In (Rebane IK, Tuul and Hizhnyakov, 1979) the analogical effect of the line narrowing was considered in the time depending resonant secondary emission (consists of the scattering and of the ordinary and the hot luminescence).

4 CONCLUSIONS

Computer calculations are carried out for the two-step SHB in the three- and four-level systems by light pulses of the arbitrary duration. In the three-level system we receive two spectral lines of SHB efficiency $P_3(\Omega_{01})$ at the first step of this process (for fixed value of $\Omega_{12} - \omega_2$) or of SHB efficiency $P_3(\Omega_{12})$ at the second step of this process (for fixed value of $\Omega_{01} - \omega_1$). In the four-level system we receive the three spectral lines of SHB efficiency $P_4(\Omega_{01})$ (for fixed value of $\Omega_{1'2} - \omega_2$) or of SHB efficiency $P_4(\Omega_{1'2})$ (for fixed value of $\Omega_{01} - \omega_1$). The calculations show that at the first step of SHB with increasing the time delay T between pulses, the spectral lines of SHB efficiencies $P_3(\Omega_{01})$ and $P_4(\Omega_{01})$ corresponding to the absorption of the pulses on end and due to these the corresponding spectral holes in IDF monotonously narrow down. This phenomenon does not depend on the duration of the second burning pulse and is absent for the spectral lines corresponding to the absorption of the two pulses together and also at the second step of the process.

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