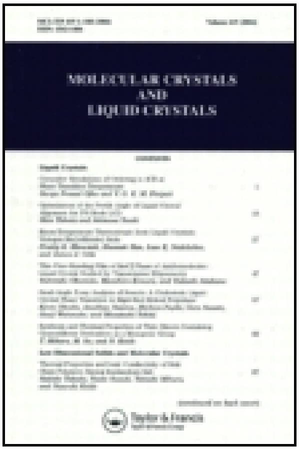

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THEORY OF ONE- AND TWO-STEP PULSED SPECTRAL HOLE BURNING

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Abstract A theory of spectral hole burning (SHB) by light pulses of an arbitrary shape and duration is proposed. One- and two-step (photon-gated) SHB processes induced by laser pulses in two-, three- and four-level systems are theoretically considered. It is shown that for two-step SHB there exists a possibility of obtaining spectral holes which are narrower than in the one-step SHB with monochromatic light. In case of one- and two-step SHB the dependence of the shape of the hole on the duration of the burning pulse is considered (on two-step SHB the second pulse is δ-pulse).

Spectral hole burning (SHB) has perspective applications in optical information storage. Different storage methods (frequency-and-time-domain) and SHB mechanisms (one- and two-step) lay emphasis on a general theoretical consideration.

One-step SHB process induced by laser pulse is considered in the first order of the perturbation theory. A theoretical consideration of the one-step pulse SHB has shown that if the distribution of frequencies and the total irradiation doses are the same, the burning with a single pulse and under stationary conditions form identical holes.[1]

Two-step SHB in three- and four-level systems is considered from the viewpoint of the theory of the transient (time-dependent) spectra of secondary emission.[2] The holes burnt in two-step processes differ essentially in the cases of pulsed and stationary SHB.[3,4]

THE MINIMUM WIDTH OF SPECTRAL HOLES

For one-step ($0 \rightarrow 1$) SHB a minimal width of the hole in the inhomogeneous distribution function (IDF) of the transition frequency is obtained by photoburning with monochromatic light and is determined by the homogeneous linewidth $\sigma_0 = (2\pi c T_1^*)^{-1} + (\pi c T_2^*)^{-1} = \gamma_1 + \Gamma_1$ [5], where T_1^* and T_2^* are the energy and phase relaxation (pure dephasing) times of the excited level, γ_1 and Γ_2 are the energy and phase relaxation constants, respectively.

For two-step SHB in three-level systems ($0 \to 1 \to 2$) there exists a possibility of obtaining spectral holes which are narrower than σ_0. This possibility is based on the introduction of a time delay T between the selective (first) and fixing (second) pulses. If the selective pulse is coherent and single-sided exponential then, in the case of an extremely short fixing pulse (δ-pulse), on the increase of the time delay T, a monotonic narrowing of the spectral hole in the IDF of the frequency Ω_{01} of the electronic transition $0 \to 1$ takes place up to the limit width $\Gamma_1 + |\gamma_1 - \Delta|$ (where Δ is the FWHM spectral width of the selective pulse)[3] (see Figure 1). The minimal width of the hole is determined by the phase relaxation time of the first excited level. Thus the energy relaxation constant γ_1 of the first excited level is eliminated through compensation by the spectral width of the selective pulse. Moreover, this spectral hole can be narrowed further, i.e. the width, which is caused by phase relaxation, may also be eliminated if the interference of the selective pulse with an additional δ-pulse at the first burning step is introduced (see Figure 2). Then the spectral hole narrows up to $|\gamma_1 + \Gamma_1 - \Delta|$ at $\tau_1 \to -\infty$, or $|\gamma_1 - \Gamma_1 - \Delta|$ at $\tau_1 > 0$, and $T \to \infty$ (where τ_1 is the time delay between the additional δ-pulse and the selective pulse). Even if the most suitable pulses are chosen the uncertainty principle limited width $\sim T^{-1}$ remains.

On two-step SHB in a four-level system[4] level 1 is selectively excited first by the selective pulse (transition $0 \to 1$), then the system relaxes to intermediate level 2. The following absorption of the second short laser pulse (fixing pulse) in the transition $2 \to 3$ introduces effective photoburning of the spectral hole. An analysis of the formulae shows that the hole in IDF of the frequency Ω_{01} is monotonically narrowed with the increasing time delay T between the burning pulses. If $\Delta < \gamma_1, \gamma_2$ or $\gamma_1 < \Delta, \gamma_2$ (γ_2 is the energy relaxation constant of level 2), the limiting holewidth $\sigma = \Gamma_1 + |\gamma_1 - \Delta|$ (at $T \to \infty$). This result coincides with the one obtained for a three-level system. Unfortunately, in all the known cases of the two-step SHB, intermediate level 2 has a long lifetime, i.e. $\gamma_2 \ll \gamma_1$. The limit holewidth in the case of $\gamma_2 < \Delta$ is $\sigma = \Gamma_1 + \gamma_1 + \Delta - 2\gamma_2$. In order that the holewidth σ be less than σ_0 the condition $\Delta < 2\gamma_2$ must be fulfilled. However, because of the smallness of γ_2, the holewidth σ differs little from σ_0 and the manifestation of the width compensation effect is weak.

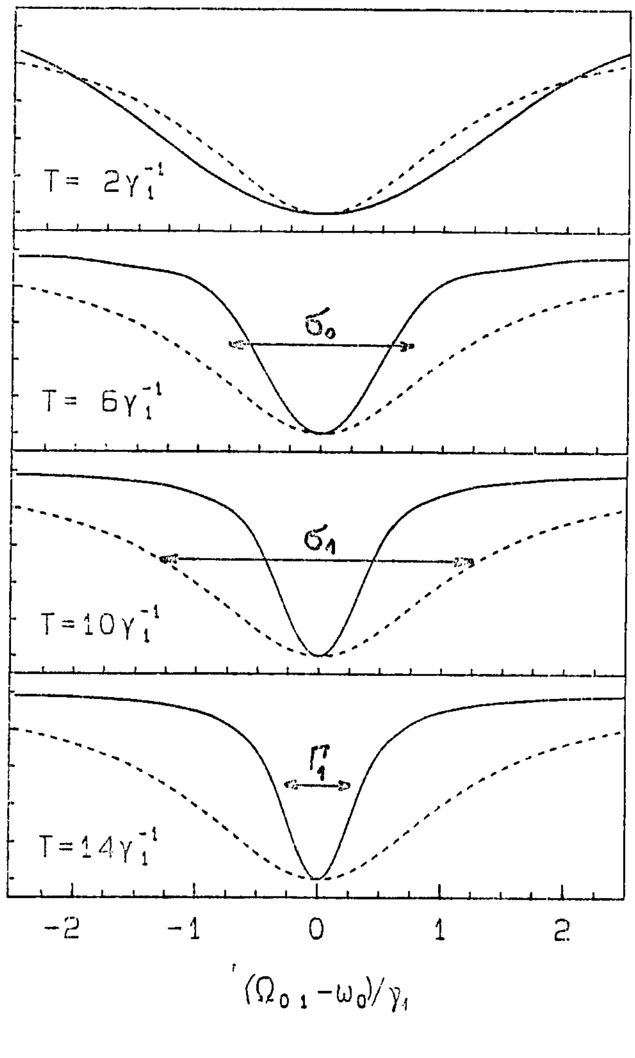

FIGURE 1 The hole in the IDF $\rho(\Omega_{01})$ for the cases of two-step (full curve) and one-step (broken curve) SHB. Parameters: Γ_1=0.5γ_1, Δ = 0.99γ_1 (σ_0 = 1.5γ_1). ω_0 is the frequency of the maximum of the first pulse.

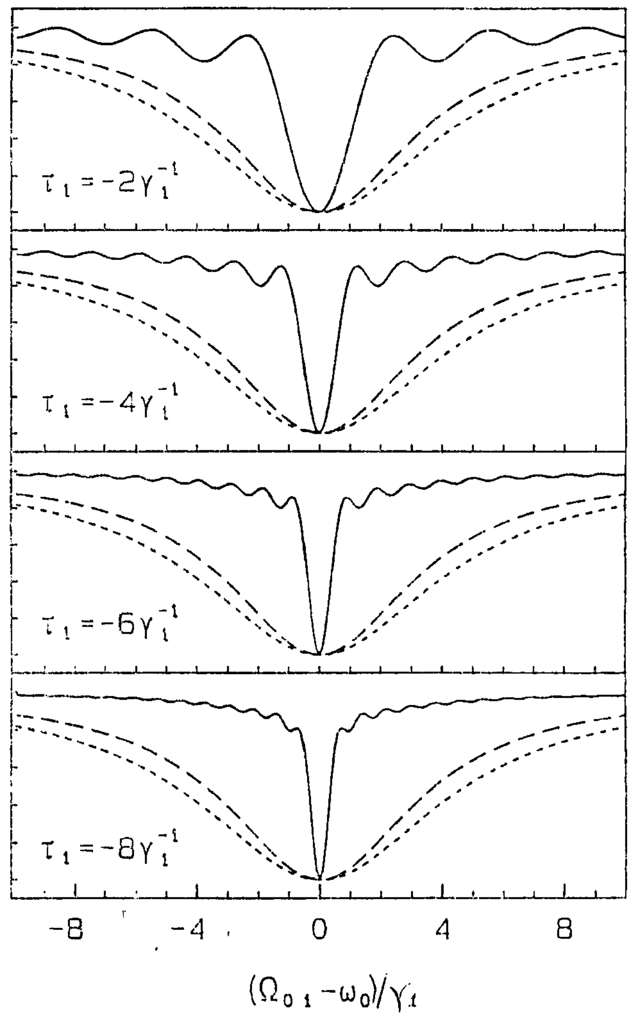

FIGURE 2 The hole in the IDF $\rho(\Omega_{01})$ in the cases of one-step (broken curve) and two-step (long broken curve) SHB by the selective pulse, and also two-step SHB by the sum of the selective pulse and the additional δ-pulse (full curve). Parameters: $\Gamma_1 = 4\gamma_1$, $\Delta = 4.5\gamma_1$, $\tau_2 = \gamma_1^{-1}$ $(\sigma_0 = 5\gamma_1)$.

THE SHAPE OF A SPECTRAL HOLE AND ITS TEMPORAL RESPONSE AT SHORT BURNING TIMES

A number of publications has considered one-step SHB by light with stationary intensity and one truncated frequency in the time interval (0,T) (see, e.g.,[6]). Thereby, the SHB efficiency $P(\Omega_{01})$, determining through (1) (see Appendix) the shape of the spectral hole in the IDF, is considered to be proportional to the product of the homogeneous absorption spectrum (HAS) (under the usual assumption HAS is of Lorentzian shape) and the irradiation dose of the burning light ($P(\Omega_{01}) = P_L(\Omega_{01})$). It is evident that such an approach does not hold for the duration of the burning, T, which is shorter than or comparable with the relaxation time of the excited level. Here SHB efficiency $P(\Omega_{01}) = P_2(\Omega_{01})$ is calculated in a model where the duration of burning, T, may be of any value[7] (see Figure 3). The HAS determines the spectral dist-

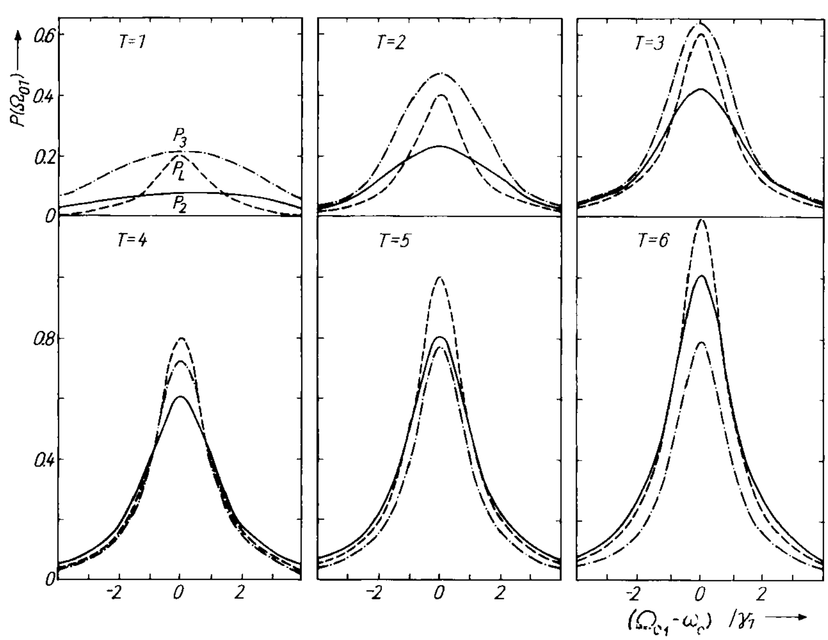

FIGURE 3 SHB efficiencies: --- $P_L(\Omega_{01})$, —— $P_2(\Omega_{01})$, and -·- $P_3(\Omega_{01})$ (in case of two-step SHB in a three-level system; here the second pulse is δ-pulse). $\Gamma_1 = \gamma_1$. Time T is given in γ_1^{-1}.

ribution of SHB efficiency only when the burning duration essentially exceeds the relaxation time of the excited level. (On the other hand, the shape of the spectral hole coincides with the spectral distribution of SHB efficiency only at small irradiation doses.)

In case of one-step SHB the dependence of the shape of the hole on the steepness of the fronts of the burning pulse is also considered.[8] Temporal responses of spectral holes as spectral filters to the δ-pulse of the light have been found[7,8].

APPENDIX

Under certain assumption the IDF, $\rho(\Omega_{01},t)$, which takes into account the inhomogeneous distribution of the frequency Ω_{01} of the electronic transition $0\rightarrow 1$ in the impurity, changes exponentially with time[6],

$$\rho(\Omega_{01},\tau) = \rho_0(\Omega_{01}) \exp[-P(\Omega_{01},\tau)]. \tag{1}$$

Here $\rho_0(\Omega_{01})$ is the initial IDF and $P(\Omega_{01},\tau)$ the SHB efficiency at the moment τ. At $t > T$ we get the final SHB efficiency, $P(\Omega_{01})$.

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