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DEPENDENCE OF THE SHAPE OF A SPECTRAL HOLE ON THE RATES OF SWITCH-IN AND SWITCH-OFF AND DURATION OF THE BURNING LIGHT

(Presented by H. Keres)

The shape of a spectral hole in the function of the inhomogeneous distribution of centres over the transition frequency is calculated. The switch-in and switch-off of the burning light take place by an exponential rule and the duration of burning may be of any value, including shorter than the relaxation time of the excited electronic level. Temporal response of the spectral hole as a spectral filter on δ-pulse of light has been found.

1. Introduction

Spectral hole burning (SHB) [1,2] is widely used as a method of eliminating the inhomogeneous broadening of the spectra of electronic transitions in impurity systems. In a number of publications on SHB by a light with stationary intensity and one frequency in the time interval $(0, T)$ has been considered (see, e.g., [3]). Thereby, the SHB efficiency, $P(\Omega_{01}, \tau)$ (see (1) and (2)), determining through (1) the shape of the spectral hole in the inhomogeneous distribution of the centres at the frequency of the given transition, is considered proportional to the product of the homogeneous absorption spectrum and the irradiation dose of the burning light (see (14)). It is evident that such approach does not hold for the duration of burning, T, which is shorter or comparable with the relaxation times of the excited electronic level. In this work, SHB efficiency is calculated in a model where switch-in and switch-off of the burning light take place during a finite time by an exponential rule, and the duration of burning, T, may be of any value. The homogeneous absorption spectrum determines the spectral distribution of SHB efficiency only when the burning duration essentially exceeds the relaxation times of the excited level and the duration of swicth-in and swicth-off. On the other hand, the shape of the spectral hole coincides with the spectral distribution of SHB efficiency only at small irradiation doses. With the growth of the burning duration the irradiation dose increases, the process of saturation of the spectral hole starts [3], and the shape of the spectral hole depends on the SHB efficiency already in a complicated way.

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2. Hole in the inhomogeneous distribution function

We consider SHB in a planar, optically thin sample of a dilute solid solution of photochromic molecules at the perpencidular incidence of the burning light of the frequency ω_0 and intensity I_0 in the time interval $(0, T)$. Under certain assumption (absence of reverse processes, identical orientation of all impurities, etc. [3]) the inhomogeneous distribution function (IDF), $\varrho(\Omega_{01}, t)$, which takes into account the inhomogeneous distribution of the frequency Ω_{01} of the electronic transition $0 \rightarrow 1$ in the impurity, changes exponentially in time [3]:

$$\varrho(\Omega_{01}, \tau) = \varrho_0(\Omega_{01}) \exp[-P(\Omega_{01}, \tau)]. \qquad (1)$$

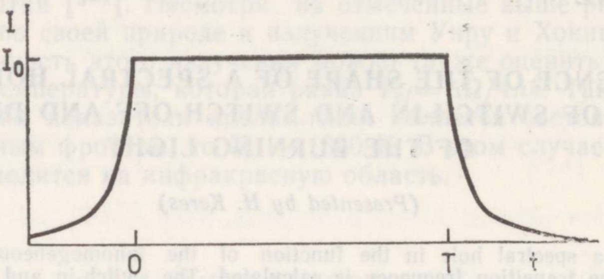

Fig. 1. The dependence of the burning light intensity $I(t) \equiv S(t, t)$ on time.

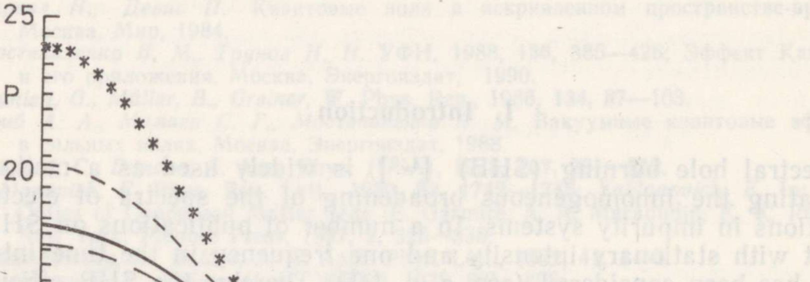

Fig. 2. SHB efficiency $P(x)$ on different switch-in durations: $\Delta_i = \gamma$ (dotted line); $\Delta_i = 2\gamma$ (dashed line, long dashes); $\Delta_i = 4\gamma$ (solid line); $\Delta_i = 6\gamma$ (dashed line, short dashes). Parameters: $\Delta_0 = 10^6\gamma$, $T = 6\gamma^{-1}$, $\beta = 1$.

Here $\varrho_0(\Omega_{01})$ is the initial IDF and $P(\Omega_{01}, \tau)$ is SHB efficiency at the moment τ. To consider the times shorter than or comparable to the relaxation time of level 1 we can use a general formula for SHB efficiency, which describes SHB by any light with a sufficiently small intensity, including the pulses with any shape and duration [4]. Thus, SHB efficiency in the first order of the perturbation theory is the following:

$$P(\Omega_{01}, \tau) = \alpha \int_{-\infty}^{\tau} dt' \iint_{-\infty}^{t'} dt_1\, dt'_1\, S(t_1, t'_1) F(t', t_1, t'_1), \tag{2}$$

where F is the correlation function (CF) of the impurity centre [4], S is the CF of the burning light and α is the quantum yield of the burning process. When the hole-burning process is finished $(t \gg T)$, we get the final SHB efficiency, $P(\Omega_{01})$, after which the final spectral hole in IDF is determined through (1).

To describe the impurity centre we use a model where the relaxation processes of excited level 1 are described by the rate of energy relaxation, γ. The corresponding CF is the following:

$$F(t', t_1, t'_1) = \frac{\sigma\gamma}{2\pi} \exp\left[i\Omega_{01}(t'_1 - t_1) - \frac{\gamma}{2}(2t' - t_1 - t'_1) \right], \tag{3}$$

where σ is the integral absorption cross-section. If we use this CF, we get the following absorption spectrum

$$\varkappa(x) = \frac{\sigma\gamma}{2\pi} \frac{1}{x^2 + \gamma^2/4}, \tag{4}$$

where $x = \omega_0 - \Omega_{01}$.

Let the switch-in and switch-off of the burning light take place by an exponential rule (Fig. 1), then the CF of the burning light is the following:

$$S(t_1, t'_1) = I_0 \varepsilon^*(t_1)\varepsilon(t'_1) = I_0 \exp[i\omega_0(t_1 - t'_1)]\, |\varepsilon(t_1)|\, |\varepsilon(t'_1)|, \tag{5}$$

where $|\varepsilon(t)| = \exp(\Delta_i t/2)$, if $t \leqslant 0$, $|\varepsilon(t)| = 1$, if $0 < t < T$ and $\varepsilon|(t)| = \exp[-\Delta_0(t - T)/2]$, if $T \leqslant t$ and the constants Δ_i and Δ_0 define respectively the rate of the switch-in and switch-off of the burning light. Substituting (3) and (5) into (2) and integrating over (2), we obtain the following SHB efficiency:

$$P(x) = P_\infty(x) + P_i(x) + P_0(x), \tag{6a}$$

where

$$P_\infty(x) = \frac{\beta}{\zeta_1}\left\{ \gamma T + \left(2 - \frac{\gamma^2}{\zeta_1}\right) + \frac{2}{\zeta_1} \exp\left(-\frac{\gamma T}{2}\right) \times \right.$$
$$\left. \times [\zeta_2 \cos(xT) - \gamma x \sin(xT)] \right\}, \tag{6b}$$

$$P_i(x) = \beta \left\{ \frac{2\gamma}{\zeta_1 \Delta_i} - \frac{1}{\zeta_i}\left(1 + \frac{\gamma}{\Delta_i}\right) + 2\exp\left(-\frac{\gamma T}{2}\right) \times \right.$$
$$\times \left[\frac{1}{\zeta_1 \Delta_i}\Big(-\gamma\cos(xT) + 2x\sin(xT)\Big) + \frac{1}{\zeta_i(1 - \Delta_i/\Delta_0)} \times \right.$$
$$\left.\left. \times \left(\left(1 + \frac{\gamma}{\Delta_i}\right)\cos(xT) - \frac{2x}{\Delta_i}\sin(xT) \right) \right] \right\} \tag{6c}$$

and

$$P_0(x)=\beta\left\{\frac{2\gamma}{\zeta_1\Delta_0}-\frac{1}{\zeta_0}\left(1+\frac{\gamma}{\Delta_0}\right)+\right.$$

$$+2\exp\left(-\frac{\gamma T}{2}\right)\left[\frac{1}{\zeta_1\Delta_0}\left(-\gamma\cos(xT)+2x\sin(xT)\right)+\right.$$

$$\left.\left.+\frac{1}{\zeta_0(1-\Delta_0/\Delta_i)}\left(\left(1+\frac{\gamma}{\Delta_0}\right)\cos(xT)-\frac{2x}{\Delta_0}\sin(xT)\right)\right]\right\}. \qquad (6d)$$

Here $\zeta_1=x^2+\gamma^2/4$, $\zeta_i=x^2+(\gamma+\Delta_i)^2/4$, $\zeta_0=x^2+(\gamma+\Delta_0)^2/4$ and $\beta=\alpha I_0\sigma/2\pi$ (Fig. 2).

3. The light pulse passing through the spectral filter

The spectral holes are widely used as spectral filters (see, e.g. [5]). The temporal response $\overline{R}(t)$ of a spectral filter to the δ-pulse of the light falling on the filter at the moment $t=0$ is given by the Fourier transform of its complex transmission ($t\geqslant 0$):

$$\overline{R}(t)=\frac{1}{2\pi}\int_{-\infty}^{\infty}d\omega\exp(-i\omega t)\,r(\omega) \qquad (7)$$

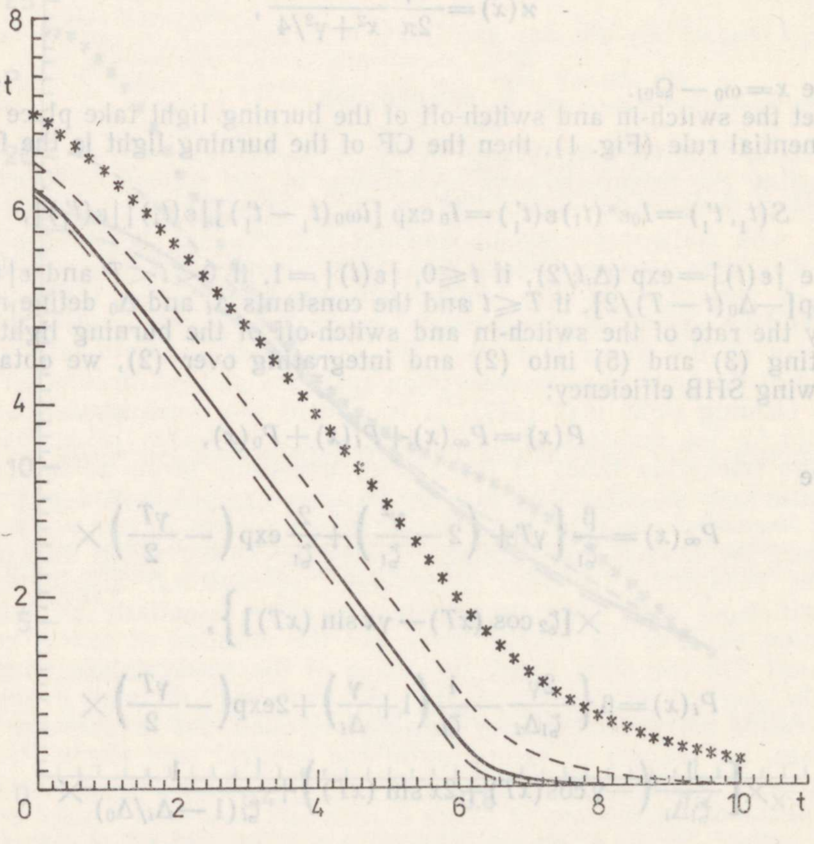

Fig. 3. The characteristic part of temporal response $Rt(t)$ on different switch-in durations: $\Delta_i=\gamma$ (dotted line); $\Delta_i=2\gamma$ (long dashes); $\Delta_i=4\gamma$ (solid line); $\Delta_i=6\gamma$ (short dashes). Parameters: $\Delta_0=10^6\gamma$, $T=6\gamma^{-1}$, $\beta=1$. The time t is given in γ^{-1}.

and the observable intensity of light is $I(t) = |\overline{R}(t)|^2$. The complex transmission $r(\omega)$ is related to the transmission $T(\omega)$ by

$$r(\omega) = \exp\{(1+i\hat{H})[\ln T(\omega)]/2\}, \tag{8}$$

where $\hat{H}$ denotes the Hilbert transmission [6]. After SHB ($\tau\to\infty$) the transmission spectrum of an optically thin sample of the thickness δL is given by

$$T(\omega) = 1 - \delta L \int_{-\infty}^{\infty} d\Omega_{01} \varkappa(\omega - \Omega_{01}) \varrho(\Omega_{01}). \tag{9}$$

Substituting (1) and (4) into (9), (9) into (8) and then (8) into (7), we obtain the following temporal response ($t \geqslant 0$):

$$\overline{R}(t) = \delta(t) - \frac{\delta L \sigma \varrho_0}{2\pi} \exp\left(-\frac{\gamma t}{2}\right) \int_{-\infty}^{\infty} d\Omega_{01} \exp\left[-i\Omega_{01} t - P(\Omega_{01})\right]. \tag{10}$$

(ϱ_0 is assumed to be constant). Direct integration over (10) is possible when $P(\Omega_{01}) \ll 1$. Then

$$\exp\left[-P(\Omega_{01})\right] = 1 - P(\Omega_{01}) + P^2(\Omega_{01})/2 - \ldots \tag{11}$$

and ($t \geqslant 0$)

$$\overline{R}(t) = \delta(t)(1 - \delta L \sigma \varrho_0) + R(t), \tag{12}$$

where in the function $R(t)$ $t > 0$.

Let us take into account the first two terms in (11). Integrating over (10), we obtain (Fig. 3)

$$R(t) = A\beta \exp(-i\omega_0 t - \gamma t) Rt(t) = \\ = A\beta \exp(-i\omega_0 t - \gamma t)\left[Rt_\infty(t) + Rt_i(t) + Rt_0(t)\right], \tag{13a}$$

where at $0 < t \leqslant T$

$$Rt_\infty(t) = (T - t), \tag{13b}$$

$$Rt_i(t) = \left[\frac{2}{\Delta_i} - \frac{1}{\Delta_i}\exp\left(-\frac{\Delta_i t}{2}\right)\right] \tag{13c}$$

and

$$Rt_0(t) = \left[\frac{2}{\Delta_0} - \frac{1}{\Delta_0}\exp\left(-\frac{\Delta_0 t}{2}\right)\right] \tag{13d}$$

and at $T < t$

$$Rt_\infty(t) = 0, \tag{13e}$$

$$Rt_i(t) = \left[\frac{2}{\Delta_i(1 - \Delta_i/\Delta_0)}\exp\left(-\frac{\Delta_i}{2}(t - T)\right) - \right. \\ \left. - \frac{1}{\Delta_i}\exp\left(-\frac{\Delta_i t}{2}\right)\right], \tag{13f}$$

and

$$Rt_0(t) = \left[\frac{2}{\Delta_0(1 - \Delta_0/\Delta_i)}\exp\left(-\frac{\Delta_0}{2}(t - T)\right) - \right. \\ \left. - \frac{1}{\Delta_0}\exp\left(-\frac{\Delta_0 t}{2}\right)\right]. \tag{13g}$$

4. The limit cases

1. On a quick switch-in ($\Delta_i \gg \gamma, T^{-1}$), $P_i(x) \ll P_\infty(x)$ and switch-off ($\Delta_0 \gg \gamma, T^{-1}$), $P_0(x) \ll P_\infty(x)$, i.e. in (6a) the $P_\infty(x)$ corresponds to infinite quick swicth-in and switch-off, $P_i(x)$ takes into account the finite duration of swicth-in and $P_0(x)$, the finite duration of swicth-off. Note that from $P_i(x)$ we can get $P_0(x)$ if in (6c) Δ_i is substituted by Δ_0.

2. If the condition $T \gg 2\gamma^{-1}$ is fulfilled, then the second, the constant term in (6b) for $P_\infty(x)$ is much smaller than the linearly growing first term and the third term in (6b) is already relaxed. Then we get

$$P_\infty(x) = \beta T \frac{\gamma}{x^2+\gamma^2/4} = \alpha I_0 T \varkappa(x) \equiv P_L(x). \tag{14}$$

(14) is the usual formula for SHB efficiency. However, (14) is suitable only for the times essentially longer than the relaxation time of the excited level 1 and the duration of switch-in and switch-off (Fig. 4).

3. With $P(\Omega_{01}) \equiv P_L(\Omega_{01})$ (see (14); note that the hole in IDF is then Lorentzian), we obtain

$$R_L(t) = A\beta \exp(-i\omega_0 t - \gamma t) T. \tag{15}$$

In this case temporal response decays purely exponentially. With the growing of the burning duration, T, (and $\Delta_i, \Delta_0 \gg T^{-1}$) the limit of the response $R(t)$ (see (13)) tends to $R_L(t)$.

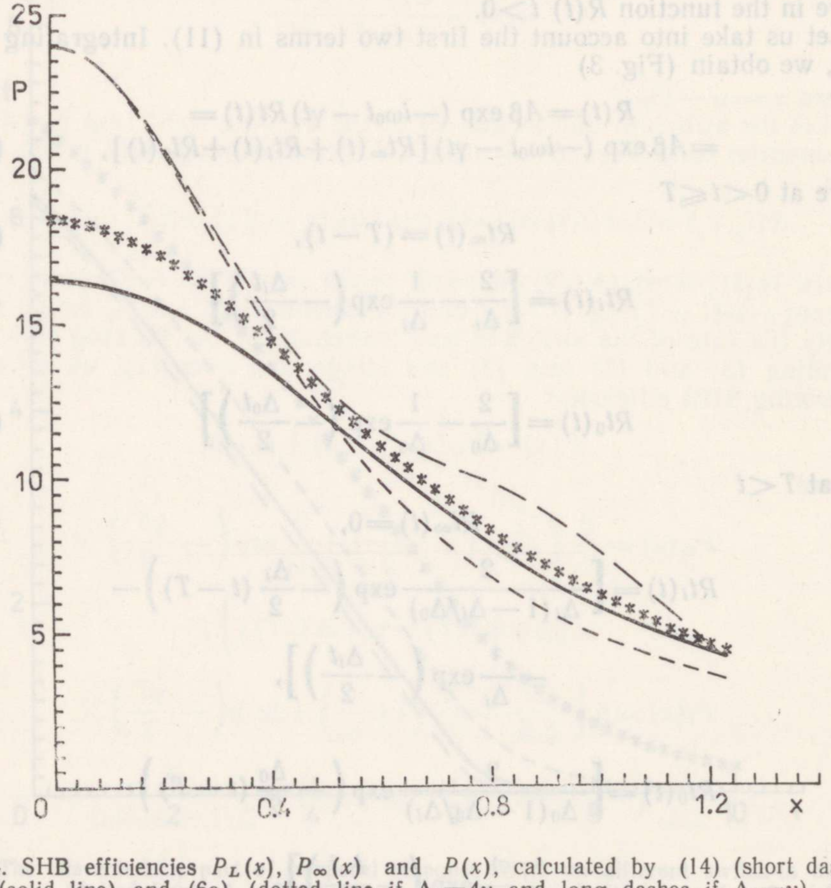

Fig. 4. SHB efficiencies $P_L(x)$, $P_\infty(x)$ and $P(x)$, calculated by (14) (short dashes), (6b) (solid line) and (6a) (dotted line if $\Delta_i = 4\gamma$ and long dashes if $\Delta_i = \gamma$). Parameters: $\Delta_0 = 10^6\gamma$, $T = 6\gamma^{-1}$, $\beta = 1$.

Thus, on common assumption that the homogeneous absorption spectrum is of a Lorentzian shape, to get a hole of the same shape the burning duration must be long enough as compared to the relaxation time of the excited level, and it must be short enough to ensure the absence of the saturation process of the hole.

REFERENCES

1. *Kharlamov, B. M., Personov, R. I., and Bykovskaya, L. A.* Opt. Commun., 1974, **12**, 191—193.
2. *Gorokhovskiy, A. A., Kaarli, R. K., and Rebane, L. A.* Opt. Commun., 1976, **16**, 282—284.
3. *Rebane, L. A., Gorokhovskii, A. A., and Kikas, J. V.* Appl. Phys., 1982, **B29**, 4, 235—250.
4. *Rebane, I. K.* J. Phys., 1989, **B22**, 2411—2421.
5. *Sõnajalg, H., Gorokhovskii, A., Kaarli. R., Palm, V., Rätsep, M., and Saari, P.* Opt. Commun., 1989, **71**, 6, 377—380.
6. *Ross, G., Fiddy, M. A., and Nieto-Vesperinas, M.* In: Inverse Scattering Problems in Optics (ed. H. P. Baltes). Berlin, Springer, 1980, 15.

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SPEKTRAALSÄLGU KUJU SÕLTUVUS PÕLETAVA VALGUSE SISSE- JA VÄLJALÜLITAMISE KIIRUSEST NING KESTUSEST

On arvutatud spektraalsälgu kuju lisanditsentrite ülemineku sageduse mittehomogeenses jaotusfunktsioonis. Põletava valguse sisse- ja väljalülitamine toimub eksponentsiaalseaduse järgi ja põletava valguse kestus võib olla suvaline, kaasa arvatud lühem kui ergastatud elektronnivoo relaksatsiooniaeg. On leitud spektraalsälgu kui spektraalfiltri ajaline koste valguse δ-impulsile.

Инна РЕБАНЕ

ЗАВИСИМОСТЬ ФОРМЫ СПЕКТРАЛЬНОГО ПРОВАЛА ОТ СКОРОСТЕЙ ВКЛЮЧЕНИЯ И ВЫКЛЮЧЕНИЯ, А ТАКЖЕ ОТ ПРОДОЛЖИТЕЛЬНОСТИ ВЫЖИГАЮЩЕГО СВЕТА

Рассчитана форма спектрального провала в функции неоднородного распределения частоты перехода примесных центров. Включение и выключение выжигающего света происходят по экспоненциальному закону. Продолжительность выжигания может быть любой, включая времена, которые короче времени релаксации возбужденного электронного уровня. Найден временной отклик спектрального провала как фильтра на δ-импульс света.