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ON THE CLASSIFICATION THEORY OF TIME-DEPENDENT RESONANCE SECONDARY EMISSION

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Инна РЕБАНЕ. К ТЕОРИИ КЛАССИФИКАЦИИ ЗАВИСЯЩЕГО ОТ ВРЕМЕНИ РЕЗОНАНСНОГО ВТОРИЧНОГО СВЕЧЕНИЯ

(Presented by V. Hizhnyakov)

In this paper, the time decay of the resonance secondary emission (RSE) spectrum of a two-level system on an instantaneous disruption of monochromatic excitation is calculated. The effect of the phase relaxation of the excited electronic state on this spectrum is calculated. The theoretical scheme used to describe the measurement recording of the transient RSE spectrum contains a dispersion system and a time-gate-supplied point photon detector behind it*. In this case, the transient spectrum $I(\Omega,\eta,t)$ determined as the photon counting rate at $t+\tau_0$ (τ_0 is the flight time of photons from the light source to the counter) on tuning the dispersing system to the frequency Ω with the spectral resolution η, is described by the following formula [1]

$$I(\Omega,\eta,t)=\iint\limits_{-\infty}^{t} dt_1\,dt'_1 \exp[i\Omega(t'_1-t_1)]\,C(t-t_1,\ t-t'_1)\times$$

$$\times\int\limits_{-\infty}^{t_1} dt_2 \int\limits_{-\infty}^{t'_1} dt'_2\, a(t'_1-t_1,\ t'_1-t'_2,\ t_1-t_2)\,S(t_2,t'_2). \tag{1}$$

Here $a(t'_1-t_1,\ t'_1-t'_2,\ t_1-t_2)$ is the three-time correlation function of the system, $S(t_2,t'_2)$ is the correlation function of the exciting light, and $C(t-t_1,\ t-t'_1)$, the correlation function of the recording.

Let the monochromatic excitation of the frequency ω_0 be disrupted at the moment $t=0$. In this case the correlation function of excitation is of the form

$$S(t_2,t'_2)=\begin{cases}\exp[i\omega_0(t_2-t'_2)], & t_2,t'_2\leqslant 0,\\ 0, & t_2 \text{ and (or) } t'_2>0.\end{cases} \tag{2}$$

We take the correlation function of spectral transmission in an exponential form

$$C(t-t_1,\ t-t'_1)=\exp[-\eta(t-t_1)]\exp[-\eta(t-t'_1)]. \tag{3}$$

Let us use the Condon approximation. The relaxation processes in the excited electronic state will be described by means of energetic (longitudinal) and phase (transversal) relaxation constants, γ and Γ, respectively. For describing the effects of transversal relaxation in this state the results of [3] can be used, according to which the correlation function of a two-level system with consideration for the effects mentioned equals

* In [2] the effect of the measuring scheme upon the transient spectrum is considered.

$$a(t'_1-t_1,\ t'_1-t'_2,\ t_1-t_2)=C\exp[i\Omega_{01}(t_1-t_2+t'_2-t'_1)-$$
$$-\gamma(t_1-t_2+t'_1-t'_2)-\Gamma(|t'_1-t_1|+t'_1-t'_2+t_1-t_2+$$
$$+|t'_2-t_2|-|t'_2-t_1|-|t'_1-t_2|)]. \qquad (4)$$

Here Ω_{01} is the frequency of the electronic transition and C is a constant.

After substituting formulae (2)—(4) into (1) and integrating, we get the following formula

$$I(\Omega,\eta,t)=C[(\omega_0-\Omega_{01})^2+(\gamma+\Gamma)^2]^{-1}\times$$
$$\times\{\exp(-2\eta t)[|\alpha_1|^{-2}-(\alpha_1^{-1}(\alpha_2^*-\Gamma)^{-1}+\text{c. c.})+$$
$$+(1+\Gamma/\eta)(1-\Gamma/(\eta-\gamma))|\alpha_2-\Gamma|^{-2}]+ \qquad (5)$$
$$+\exp(-2\gamma t)(1+\Gamma/\gamma)[1+\Gamma/(\eta-\gamma)]|\alpha_2+\Gamma|^{-2}+$$
$$+\exp[-(\eta+\gamma+\Gamma)t][(\alpha_2^*-\Gamma)^{-1}(\alpha_1^{-1}-(1+\Gamma/\gamma)(\alpha_2+\Gamma)^{-1}+$$
$$+(\Gamma/\gamma)(\alpha_2+2\gamma+\Gamma)^{-1})\exp(i(\Omega-\Omega_{01})t)+\text{c. c.}]\},$$

where

$$\alpha_1=i(\omega_0-\Omega)+\eta,$$
$$\alpha_2=i(\Omega_{01}-\Omega)+\eta-\gamma.$$

In formula (5), the term proportional to $|\alpha_1|^{-2}$ describes scattering, the terms proportional to $|\alpha_2\pm\Gamma|^{-2}$, $[(\alpha_2^*-\Gamma)(\alpha_2+2\gamma+\Gamma)]^{-1}\times\exp[i(\Omega-\Omega_{01})t]$, $[(\alpha_2^*-\Gamma)(\alpha_2+\Gamma)]^{-1}\exp[i(\Omega-\Omega_{01})t]$ describe luminescence (the two last ones describe oscillation on the wings of the luminescence line [1,4]), and the terms proportional to $[\alpha_1(\alpha_2^*-\Gamma)]^{-1}$, $[\alpha_1(\alpha_2^*-\Gamma)]^{-1}\exp[i(\Omega-\Omega_{01})t]$ describe the interference between scattering and luminescence. For the luminescence line an effect of width compensation takes place [4] — the registration width (spectral resolution η) and the decay width γ, i. e. on the increase of t the width of the luminescence line $\sigma(t)$ tends to the limit

$$\lim_{t\to\infty}\sigma(t)=\Gamma+|\eta-\gamma|. \qquad (6)$$

The constant of transverse relaxation Γ contributes to the width of the luminescence spectrum and to the decay constant of oscillating terms. It also determines the intensity distribution between the luminescence and scattering lines. For example, in the case of $\Gamma\gg\gamma\cong\eta$ at $t\gg\Gamma^{-1}$ $I_{\text{lum}}/I_{\text{scatt}}\sim\Gamma$.

Note that the decay of the term corresponding to scattering is determined only by the spectral resolution η and is independent of the phase relaxation rate Γ (one of the two interference terms and the luminescence term describing its oscillations depend on that rate). This result contradicts the conclusion of the author of [5] that the decay of the scattering component in RSE is determined by the phase relaxation rate.

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