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Data Processing of Time-Resolved Pico-Second Photoluminescence Spectra of Quasi Bi-Dimensional Systems

Dobri Atanassov Batovski[1], Philippe Roussignol[2], Emanuelle Deleporte[2], and Arianna Filoramo[2]
Faculty of Science and Technology, Assumption University

Abstract

This paper considers data processing of time-resolved photoluminescence (PL) spectra obtained with two-dimensional streak camera. Effective numerical algorithms are implemented in an applied software product (program LumFit).

Keywords: *Pico-Second Photoluminescence, Data processing.*

Introduction

The relaxation process of the optical spontaneous emission (for example photoluminescence) of semiconductor heterostructures can be described theoretically by means of a series of phenomenological parameters, the time-constants of the recombination processes (Bastard 1992). The radiative channel (after laser pumping) is in competition with the non-radiative relaxation processes (photon emission, capture by deep centers, Auger effect, etc.) which send the excited carriers to lower states from which they can emit photons or relax non-radiatively, etc. (Fig.1).

The theoretical and numerical description of the relaxation pico-second PL lineshapes in the presence of a series of discrete recombination levels is the subject of the present work. The peculiarities of data processing of experimental noisy spectra are considered in terms of

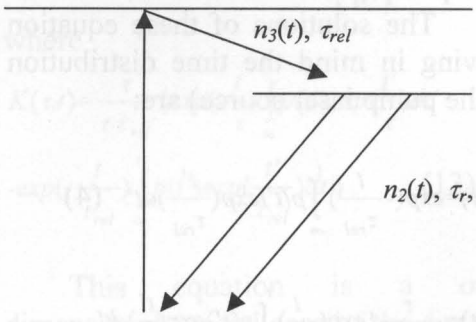

Fig.1. Schematic diagram of the generation-recombination process of three-energy level optical system.

1 E-mail: dobri@science.s-t.au.ac.th
2Ecole Normale Superieure, Departement de Physique, Laboratoire de Physique de la Matiere Condensée, 24, rue *Lhomond, 75231 Paris Cedex 05, FRANCE*

multivariable constrained optimization theory. The case of continuous distribution of levels is described in terms of the ill-posed problems theory.

Basic Theoretical Relations

The time-constants of the recombination processes can be described using the rate equations for the non-radiative relaxation process n_3 (t), and the time process $n_2(t)$ from the lower state (deep level)[1]

$$\frac{dn_3}{dt} = p(t) - \frac{n_3}{\tau_{rel}} \qquad (1)$$

$$\frac{dn_2}{dt} = \frac{n_3}{\tau_{rel}} - \frac{n_2}{\tau_r} - \frac{n_2}{\tau_{nr}} = \frac{n_3}{\tau_{rel}} - \frac{n_2}{\tau} \qquad (2)$$

$$\frac{1}{\tau} = \frac{1}{\tau_r} + \frac{1}{\tau_{nr}} \qquad (3)$$

where τ_{rel}, τ_r, and τ_{nr} are correspondingly the relaxation, radiative and non-radiative time-constants; $p(t)$ is the line- shape of the laser pump pulse.

The solutions of these equation (having in mind the time distribution of the pump laser source) are:

$$n_3(t) = exp(-\frac{t}{\tau_{rel}}) \int_{-\infty}^{t} p(t')exp(\frac{t'}{\tau_{rel}})dt' \qquad (4)$$

$$n_2(t) = \frac{\tau}{\tau-\tau_{rel}}\{exp(-\frac{t}{\tau}) \int_{-\infty}^{t} p(t')exp(\frac{t'}{\tau})dt'$$

$$-exp(-\frac{t}{\tau_{rel}}) \int_{-\infty}^{t} p(t')exp(\frac{t'}{\tau_{rel}})dt'\} \qquad (5)$$

In the case when the pump laser pulse can be presented via time δ-function

$$p(t) = n_0\delta(t) \qquad (6)$$

we obtain

$$n_3(t) = n_0 Y(t) exp(-\frac{t}{\tau_{rel}}) \qquad (7)$$

$$n_2(t) = n_0 Y(t) \frac{\tau}{\tau-\tau_{rel}}$$

$$\times\{exp(-\frac{t}{\tau}) - exp(-\frac{t}{\tau_{rel}})\} \qquad (8)$$

The experimental pump laser pulse can be presented if the first approximation via Gaussian distribution with time dispersion γ (≈2 ps, for example) and middle time point t_0

$$p(t) = \frac{1}{\gamma\sqrt{2\pi}} exp(-\frac{1}{2}(\frac{t-t_0}{\gamma})^2) \qquad (9)$$

However, it is more convenient to measure the lineshape of the pump laser pulse with the same experimental set-up. Then the experimental spectrum consists of two parts:
- the lineshape of the pump laser pulse;
- the pico-second photoluminescence spectrum of the sample.

Time-Resolved PL in case of Series of Discrete Levels

In the case of series of discrete levels (Ohnesorge 1994).

$$n_\tau(t) \approx \sum_{i=1}^{number\ of\ levels} D_i \frac{\tau_i}{\tau_i-\tau_{rel}}$$

$$\times\{exp(-\frac{t}{\tau_i}) - exp(-\frac{t}{\tau_{rel}})\} \qquad (10)$$

$$n_\tau(t) \approx \sum_{i=1}^{number\ of\ levels} D_i \frac{\tau_i}{\tau_i - \tau_{rel}}$$

$$\{exp(-\frac{t}{\tau_i}) \int_{-\infty}^{t} p(t')exp(\frac{t'}{\tau})dt'$$

$$-exp(-\frac{t}{\tau_{rel}}) \int_{-\infty}^{t} p(t')exp(\frac{t'}{\tau_{rel}})dt'\} \qquad (11)$$

where D_i are the contributions (ratios) of each level to the photoluminescence spectrum. The Eq.(10) is valid for δ-function time pump source and eq.(11) - for arbitrary $p(t)$ pump lineshape. This theoretical model represents a constrained optimization problem from numerical point of view.

We consider the case of two discrete quantum levels with different time-constants in order to obtain qualitative information from noisy experimental spectra by minimizing the least-squares quadratic functional between experimental data and theoretical model.

The simplex method (Box 1965) for constrained optimization can be used for effective data processing of such problems. The simplex method is a non-gradient iterative method for constrained optimization, which is very convenient when:

- the number of theoretical parameters is not so big. In our case, the fit parameters are 5: τ_{rel}, τ_1, τ_2, D_2 (we assume that $D_1 = 1$) and the starting time point t_{start} for the photoluminescence process;
- the calculation of the function for the theoretical model needs time;
- the first derivatives of the function for the theoretical model cannot be calculated in real time;
- the point of global minimum of the quadratic functional has to be reached neglecting the initial assumptions for the searching solution.

The simplex method forms an initial many-dimensional figure of random points (simplex). The simplex can change its form, size, and direction during the iterative process searching the point of global minimum. The method can pass through some of the points of local minimum, while the corresponding gradient methods depend strongly on the initial assumptions for the searching solution.

Time-Resolved PL in case of Continuous Distribution of Levels

In case of continuous distribution of levels the corresponding theoretical model can be represented by one-dimensional Fredholm equation of the first kind with two-dimensional integral kernel

$$n_\tau(t) \approx \int_{\tau_{min}}^{\tau_{max}} D(\tau)K(\tau,t)d\tau, \quad t_{min} \le t \le t_{max} \qquad (12)$$

where

$$K(\tau,t) = \frac{\tau}{\tau - \tau_{rel}} \{exp(-\frac{t}{\tau}) \int_{-\infty}^{t} p(t')exp(\frac{t'}{\tau})dt'$$

$$-exp(-\frac{t}{\tau_{rel}}) \int_{-\infty}^{t} p(t')exp(\frac{t'}{\tau_{rel}})dt'\} \qquad (13)$$

This equation is a one-dimensional special case of the general operator equation $\hat{A}D = S$, $D \in U$, $S \in Z$ with linear operator $\hat{A}$ and approximately known left $\hat{A}_k$ and right

parts S_δ, related to the wide class of ill-posed problems(Tihonov *et al.* 1983). The regularization methods in most cases search for a solution by minimizing the quadratic functional

$$J(D) = \frac{1}{2}\left\|\hat{A}_k D - S_\delta\right\|_Z^2, \; S_\delta \in Z$$

by projection over some *a priori* known multitude in given space $D \in U_1$ ($U_1 \in U$) below the sum of the uncertainties $h\|D\|$ and δ of the left and right parts of the operator equation, i.e.,

$$\left\|\hat{A}D - S\right\|_Z \leq \delta + h\|D\| \qquad (14)$$

where $\| \|$ denotes norm in corresponding space.

Conclusion

The discrete level approach is implemented in the program LumFit (Batovski 1995; 1998), developed for effective data processing in real time of time-resolved pico-second photo-luminescence spectra.

The assumption for continuous distribution of levels can be useful in case of data processing of both disordered and amorphous quasi bi-dimensional systems.

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