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Monte-Carlo method for determining the quenching function from variable heating rate measurements

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ABSTRACT

In thermoluminescence (TL) measurements radiative recombination takes place at various temperatures. Typically, the quantum efficiency of luminescence decreases with increasing temperature. We call this mechanism the thermal quenching. There is no simple method to 'restore' unquenched TL data. This paper presents an algorithm, based on the Monte-Carlo method, for calculating the quenching function and restoring the unquenched TL curve. For this purpose we use a series of TL glow curves measured at the same initial conditions with variable heating rates. The method is quite general and no particular kinetic model of TL need to be assumed. The reliability of the method is tested using computer generated TL glow curves obeying the simple trap model (STM) kinetics.

1. Introduction

During thermoluminescence (TL) measurements various processes take place simultaneously. Prior to the measurement, a sample under study has to be excited by a high-energy (e.g. UV, X-ray or gamma) radiation. A portion of the energy is stored in traps in the solid. When the sample is heated the energy is released in the form of light. Trapped charge carriers are thermally released to a transport band undergoing radiative recombination in recombination centers (RCs). A series of peaks appearing in a TL glow curve is usually attributed to trap levels characterized by different activation energies.

Radiative recombination takes place at various temperatures. Typically, the quantum efficiency of luminescence decreases at higher temperatures. We call this mechanism the thermal quenching. In TL and optically stimulated luminescence (OSL) measurements it may be an important factor influencing the experimental data.

The effect of thermal quenching may be observed while performing a series of TL measurements with different heating rates. Typically, with increasing heating rate, the maximum of a TL glow peak shifts to higher temperatures. At a higher temperature, the luminescence is quenched more intensely so that the whole area under TL peak decreases. An illustration of this phenomenon is shown in Fig. 1. It is assumed that the quenching function (QF) has the form:

$$\eta(C,W,T) = \left[1 + C\exp\left(\frac{-W}{kT}\right)\right]^{-1} \quad (1)$$

where C and W are 'quenching parameters'. T is the sample temperature and k is the Boltzmann constant (Akselrod et al., 1998). Therefore, instead of the normal TL intensity $J_{TL}(T)$, we measure the function disturbed by quenching:

$$J(T) = J_{TL}(T)\eta(C,W,T). \quad (2)$$

There is no simple way to determine the quenching parameters as well as the 'unquenched' intensity $J_{TL}(T)$. However, some ideas for approximate determination of QF, assuming e.g. its constant value throughout the peak or a special order of kinetics were presented by Pagonis et al. (2006), Dallas et al. (2008) and others.

This paper presents a different method for calculating the quenching parameters from a series of variable heating rates TL measurements. The method is quite general and does not assume any particular type of TL kinetics.

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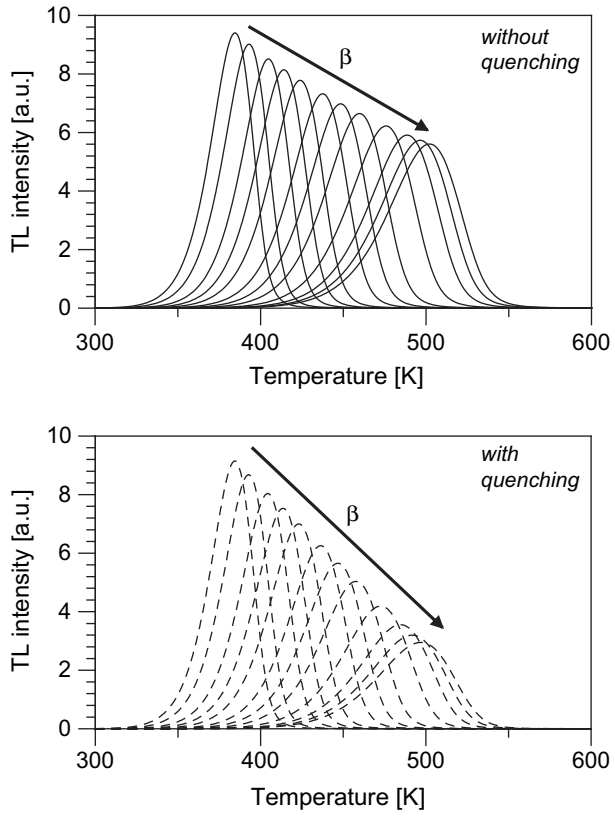

Fig. 1. Set of TL glow curves calculated with different heating rates from 0.01 to 20 K/s without (solid lines) and with (dotted lines) thermal quenching. The TL is given in photon-counts/K.

2. The algorithm for calculating quenching function parameters

The difficulty in estimating the quenching parameters is related to the fact, that during the measurements we are dealing with luminescence intensities already distorted by quenching. Quenching changes the TL peak shapes and the positions of the maxima, which are important i.e. for determining basic parameters of traps and recombination centers. For these reasons we cannot use directly the quenched data for calculations. To solve the problem we formulate an 'area-invariant' hypothesis and then we will propose a numerical algorithm to determine QF.

First of all, let us note that when the quenching is absent, the whole area under the TL peak should be strictly conserved (at least within the framework of basic TL models: STM – simple trap model and LT – localized transitions). Moreover, it seems that the peak height and peak width are (approximately) inversely proportional to each other when it is shifted towards higher temperatures with increasing heating rate β. Now, let us consider the following integral:

$$S_{\varepsilon,\beta} = \int_{t_\varepsilon^{(1)}}^{t_\varepsilon^{(2)}} J(\beta,t)\mathrm{d}t = \frac{1}{\beta}\int_{T_\varepsilon^{(1)}}^{T_\varepsilon^{(2)}} J(\beta,T)\mathrm{d}T, \tag{3}$$

where: $0 < \varepsilon < 1$ and $t_\varepsilon^{(1)}$, $t_\varepsilon^{(2)}$, $T_\varepsilon^{(1)}$, $T_\varepsilon^{(2)}$ denote positions on horizontal axis, on both sides of the TL peak where the intensity falls down to ε-th part of the maximum intensity, i.e.:

$$J\left(\beta,t_\varepsilon^{(1)}\right) = J\left(\beta,t_\varepsilon^{(2)}\right) = \varepsilon J_{\max}, \tag{4}$$

$$J\left(\beta,T_\varepsilon^{(1)}\right) = J\left(\beta,T_\varepsilon^{(2)}\right) = \varepsilon J_{\max}. \tag{5}$$

Therefore, we postulate the following hypothesis: *the integral $S_{\varepsilon,\beta}$ (3) does not depend on the heating rate β.*

Its validity will be discussed in the next section. The hypothesis allows us to formulate the following Monte-Carlo algorithm:

1. Set the peak range parameter ε (e.g. $\varepsilon = 0.2$)
2. Generate $W \in [W^{(\min)}, W^{(\max)}]$ and $\ln\_C \in [\ln C^{(\min)}, \ln C^{(\max)}]$
3. Calculate 'unquenched' data $J_{\mathrm{TL}}(\beta_i,T) = J(\beta_i,T)/\eta(C,W,T)$ where β_i is the i-th heating rate
4. For each peak $J_{\mathrm{TL}}(\beta_i,T)$ find peak position parameters $T_{\mathrm{TL},\beta_i}^{(\max)}$ and $J_{\mathrm{TL},\beta_i}^{(\max)} = J_{\mathrm{TL}}(\beta_i, T_{\mathrm{TL},\beta_i}^{(\max)})$
5. For each peak $J_{\mathrm{TL}}(\beta_i,T)$ calculate peak range parameters $T_{\mathrm{TL},\beta_i}^{(1)}$ and $T_{\mathrm{TL},\beta_i}^{(2)}$ defined as follows: $J_{\mathrm{TL}}(\beta_i, T_{\mathrm{TL},\beta_i}^{(1)}) = \varepsilon J_{\mathrm{TL},\beta_i}^{(\max)} = J_{\mathrm{TL}}(\beta_i, T_{\mathrm{TL},\beta_i}^{(2)})$ with $T_{\mathrm{TL},\beta_i}^{(1)} < T_{\mathrm{TL},\beta_i}^{(\max)} < T_{\mathrm{TL},\beta_i}^{(2)}$
6. Calculate 'unquenched peak areas' $S_i = \int_{T_{\mathrm{TL},\beta_i}^{(1)}}^{T_{\mathrm{TL},\beta_i}^{(2)}} J_{\mathrm{TL}}(\beta_i,T)\mathrm{d}T$ for each peak
7. Calculate the mean area $\bar{S} = (1/N)\sum_{i=1}^{N} S_i$ and the relative deviation

$$\sigma_{R\bar{S}} = \frac{1}{\bar{S}}\sqrt{\frac{1}{N(N-1)}\sum_{i=1}^{N}(S_i-\bar{S})^2}$$

8. If σ_{RS} belongs to the set of 100 best-fit values (with lowest σ_{RS}) then store the data
9. Go to step 2; repeat N_{loop} times
10. Estimate the final values as an average of the 10 best-fit results.

The idea of this method is simple. The algorithm tries to find appropriate values of the quenching parameters, by guessing a pair (C,W), where $C = \exp(\ln\_C)$ (in step 2), then calculating 'unquenched' data (steps 3–5) and finally comparing the areas under 'unquenched' TL peaks measured for various heating rates (in step 7). For appropriately guessed parameters, the areas for different heating rates should be the same. Hence, the relative area deviation σ_{RS} is expected to be close to zero. The accuracy of this method increases with increasing the number of trials N_{loop}, i.e. the number of loops (steps 2–9) in which the random parameters are generated, and narrowing the range of acceptable parameters $[W^{(\min)}, W^{(\max)}]$ and $[\ln C^{(\min)}, \ln C^{(\max)}]$. In practical applications, the calculations should be repeated several times with decreasing range of quenching parameters. Typically, we start with the most physically acceptable ranges $[W^{(\min)}, W^{(\max)}]$ and $[\ln C^{(\min)}, \ln C^{(\max)}]$. In our calculations N_{loop} varied from 10^4 to 10^7, depending on the assumed range of parameters.

3. Testing the hypothesis

The algorithm presented above is based on the hypothesis that the integral $S_{\varepsilon,\beta}$ (3) does not depend on the heating rate β. This assumption is not obvious. Therefore we have to check its validity numerically. For this purpose, let us consider the most popular simple trap model (STM) (see Chen and McKeever, 1997):

$$\dot{n} = n\nu\exp\left(\frac{-E}{kT}\right) - n_c A(N-n), \tag{6a}$$

$$-\dot{m} = Bmn_c, \tag{6b}$$

$$m = n + n_c + M, \tag{6c}$$

where N, n, and m denote the concentrations of trap states, electrons trapped in 'active' traps and holes trapped in recombination centers, respectively. M stands for the number of electrons in the thermally disconnected traps (deep traps). A and B stand for the