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Apparent stretched-exponential luminescence decay in crystalline solids

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Abstract

The relaxation of different physical systems has been found to follow the stretched-exponential law, $\exp[-(t/\tau)^\beta]$ with $0 < \beta < 1$. In particular, the photoluminescence from porous silicon, nm size silicon in SiO_2 , glassy materials and other solids have been reported in the literature to behave this way. It has been pointed out that the key role for this behavior is played by some kind of disorder in the system. The time constants τ reported were between 10^{-12} and 10^{-2} s. In the present work, it is shown using numerical simulation relevant to the case of trapping controlled luminescence, that the decay from a single crystal with a single trapping state and a single kind of recombination center yields results which agree very well with the stretched-exponential function. Taking trapping parameters in the ranges known in luminescent materials for the stimulation of the decay curves yield different values of the parameter β between 0 and 1, and different values of the time constant τ , typically in the micro- to milli-second range. Thus, the stretched-exponential function has been shown to be even more ubiquitous than thought so far, being able to describe the decay of luminescence in an ordered crystal.

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1. Introduction

Several relaxation phenomena in complex condensed-matter systems have been found to follow the stretched-exponential decay law

$$\phi(t) = \exp[-(t/\tau)^\beta] \quad 0 < \beta < 1. \quad (1)$$

The parameters β and τ depend on the material and the specific phenomenon under consideration, and can be a function of external variables such as

the temperature [1]. As early as 1863, Kohlrausch [2] described mechanical creep by this function. Williams and Watts [3] described dielectric relaxation in polymers as being a stretched-exponential function and apparently were the ones who coined the term “stretched-exponential”. The “Brinkman Report” [4], published in 1986, stated that “there seems to be a universal function that slow relaxations obey. If the system is driven (or normally fluctuates) out of equilibrium, it returns according to the function $\exp[-(t/\tau)^\beta]$. Unfortunately, this is not a mathematical expression that is frequently encountered in physics, so little idea exists of what the underlying mechanisms are”. Scher et al. [5] pointed out that there are several

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derivations of the stretched-exponential for systems in three dimensions, involving diverse concepts such as percolation, hierarchical relaxation of constraints and multi-polar interaction transitions. Obviously, for the limiting case of $\beta \rightarrow 1$, we get the “normal” exponential decay with the characteristic lifetime τ . It should be mentioned that some authors chose to describe the stretched-exponential decay with $\beta < 1$ as the result of superposition of many exponential decays [6]. In other cases, a use has been made of a time-dependent rate constant such as that given by Plonka [7]. He presented a bimolecular reaction described by a second order kinetics equation

$$-dn_A/dt = k(t)n_A n_B, \quad (2)$$

where n_A and n_B are the concentrations of two reactants and with the time-dependent rate constant $k(t) = Bt^{\beta-1}$, $0 < \beta \leq 1$ and where $n_A + c = n_B$ with c being constant. The specific rate constant function given here has been explained to result from the continuous time random walk model developed for dispersion in amorphous solids and utilized in describing the decay of excess electrons trapped in rigid glassy matrices.

A large number of papers have been published in the last decade describing a stretched-exponential decay of luminescence in different materials and in different time scales. This was preceded by a work by Even et al. [8] who presented a decay law of the form

$$P(t) = \exp[-\gamma(t/\tau)^\beta - (t/\tau)] \quad (3)$$

which is a product of an exponential and stretched-exponential functions; this function described the decay of fluorescence in porous glass doped with rhodamine B and malachite green. Chen et al. [9] studied the decay of luminescence in porous silicon and in CdSe–ZnSe superlattice, and found that the stretched-exponential behavior

$$I = I_0 \exp[-(t/\tau)^\beta] \quad (4)$$

with $0 < \beta < 1$ describes it very well. Here, I_0 is the initial luminescence intensity following the excitation. For CdSe–ZnSe at 13 K, the decay time scale was ~ 100 ns; for porous silicon at room temperature ~ 100 μ s and for porous silicon at 13 K, ~ 10 ms. For explaining this behavior which they

call “anomalous luminescence decay”, they utilize the probability distribution $\psi(t) \sim t^{-(1+\beta)}$ taken from the above-mentioned paper by Scher et al. [5]. Chen et al. [9] state that the fitting parameters β and τ were found to depend strongly on excitation conditions such as the excitation pulse width, intensity and photon energy.

In the following years, a large number of papers have been published, describing the decay of luminescence as a stretched-exponential function. Much of this work reported results observed in porous silicon but stretched-exponential decay of luminescence has been observed in other materials as well. The point made by most of the authors of these papers has been that some disorder in the materials at hand is of prime importance in producing the stretched-exponential decay of luminescence. Pavesi and Ceschini [10] studied the stretched-exponential decay of luminescence in porous silicon. They suggest that a key role is played by disorder in the form of (i) a wide distribution of the size of the Si nanocrystals which form the p-Si skeleton, (ii) a random spatial arrangement of the nanocrystals and (iii) the structure of the nanocrystal surfaces. They further sum up that all the previously published recombination models make use of the notion of disorder: the quantum recombination model assumes that the luminescence is due to the recombination of localized excitons in undulating quantum wires or dots; the surface-state emission model postulates the formation of localized states due to the random termination of the Si nanocrystals; the amorphous model assumes the formation of an amorphous Si layer on top of the Si nanocrystals where the carriers recombine, and in the chemical models the luminescence is due to some chemical entity (SiH_x, polysilane or siloxene groups), the random distribution of which causes disorder. Pavesi and Ceschini [10] maintain that the occurrence of the stretched-exponential decay of luminescence strongly points to the role of disorder. They mention the two components in the decay of luminescence in porous silicon. The fast in the time range of 10^{-9} s and the slow in the time range of 10^{-6} – 10^{-2} s, which can be modeled by a stretched-exponential (Eq. (4)) where τ is a lifetime and β , a dispersion factor. According to

these authors, this decay law often encountered in disordered systems is considered a consequence of the dispersive diffusion of the photoexcited carriers. Carriers' diffusion among different spatial sites can be due to the excitation of carriers from localized to extended states or to hopping between localized states. In the former, the localized states act as traps and the disorder causes a distribution of release rates and of trap energies. The diffusion arises from a multiple trapping–detrapping (MTD) mechanism, where the parameter β is associated with the density of trap states and trap release rates. Pavese and Ceschini [10] also investigated the dependence of β and τ on temperature and found that τ decreases with increasing temperature whereas β increases from ~ 0.6 to 0.9 in the temperature range of 15 – 200 K. In a later work, Pavese [11] reiterated the importance of disorder in bringing about the stretched-exponential decay behavior. More specifically, he states that values of $\beta < 1$ correspond to the existence of a broad distribution of lifetimes which describes the elementary relaxation processes, either radiative or nonradiative. Pavese [11] suggests that β represents a measure of the departure from the isolated quantum dot (QD) picture and says that for isolated QDs, one expects $\beta = 1$, with smaller values of β indicating the presence of exciton dispersive motion. Several other papers have been published [12–18], discussing different aspects of the stretched-exponential decay of luminescence in porous silicon, and stressing the role of disorder in producing this kind of decay.

Stretched-exponential decay of luminescence in other materials has also been reported in recent years. Dag and Lifshitz [19] discuss the dynamics of recombination processes in PbI_2 monocrystals embedded in porous silica films. They show that in the nanoscale regime, the results can be nicely fitted to a stretched-exponential function. They distinguish between a shallow trapped state band-L, related to internal defects in the particle volume and a deeper trapped states band-G related to surface defects. They suggest that the recombination of both L and G are strongly multi-exponential due to a repopulation process, which creates a distribution in the decay times.

Kamenitsu [20] describes the PL decay of nanometer-sized oxidized crystallites and explains the stretched-exponential decay observed as being closely associated with the disorder resulting from a distribution of the crystalline size and shape, and fluctuations of the surface structure and surface stoichiometry. Linnros et al. [21,22] also discuss the photoluminescence decay in nm-sized SiO_2 and Si crystallites in SiO_2 and suggest that the stretched-exponential behavior indicates the migration of excitons through nearby crystallites concurrent with trapping at localized states. They maintain that the stretched-exponential decay suggest a partially interconnected system of nanocrystals where excitons may migrate and trap in large crystals.

Pophristic et al. [23,24] discussed the time-resolved PL of InGaN light-emitting diodes and multiple quantum wells. They report a value of β increasing from 0.75 to 0.85 with increasing indium phase segregation, and emphasize that the stretched-exponential kinetics is consistent with the presence of disorder.

Lebib et al. [25] studied the red emission in nanoporous SiGe alloys, found a stretched-exponential decay of the PL and showed that τ decreases with temperature and that both τ and β vary with the emission energy.

Teysse re et al. [26] report on the results of PL decay in poly (ethylene terephthalate) films and show that it is a stretched-exponential function. They also state that this behavior reflects disorder of some kind and suggest that in this case, since phosphorescence is associated with the ground state dimers, the relative orientation of nearby chromophores is a likely source of disorder. These authors show that τ and β increase with temperature in a certain range, and decrease at higher temperature.

Further work on the PL decay in Si nanocrystals has been reported [27,28] to result in stretched-exponential decay function. These authors stated that $\beta < 1$ corresponds to a distribution of single exponentials, each of which characterized by a different τ value. The occurrence of the stretched-exponential behavior is associated with migration of excitons from one nanocrystallite to another.

Kushovsky et al. [29] report on the decay dynamics of disordered systems, in particular heavily doped semiconductors such as amorphous silicon, glasses or ceramics. They associate the stretched-exponential decay of luminescence with recombination in donor–acceptor pairs, provided that there is an alternate, activated decay path.

Huber [30] and García-Adeva and Huber [31] give a two-state model for the stretched-exponential decay-law, assuming a modification of the Arrhenius law for the trapping process. They make the point that in contrast with approaches which assume a hopping mechanism as being the origin of the stretched-exponentials, it is enough to consider the simpler model of the detrapping process localized in the vicinity of the fluorescent center. The multiphonon absorption can lead to a β , which is a decreasing function of temperature as sometimes found experimentally.

In the present work it will be shown that stretched-exponential decay of luminescence can be expected from a model of a single crystal with one trapping state and one kind of recombination center, when transition of carriers takes place through the conduction band with no inherent necessity to have a disorder in the sample, provided retrapping is not negligible. No analytical proof is given to this behavior due to the inability of solving analytically the relevant sets of nonlinear simultaneous equations, however, numerical solution of the equations for plausible choices of sets of trapping parameters demonstrates this result of stretched-exponential decay of luminescence. It is to be noted that Van de Walle [32] has given a somewhat similar (though with some important differences) model for the case of the relaxation process in α -Si:H. He considers the trapping and retrapping of H, and the occurrence of deeper energy trap reservoir. This resembles only to a limited extent the hole recombination center considered here (see below). The observed quantity in this work is associated with the total concentration of H in traps whereas in luminescence it has to do with the rate of electron–hole recombination. The relevant equations are similar but not identical. Also, Van de Walle [32] resorts to a set of simplifying assumptions whereas here we prefer to perform a numerical solution of the

coupled rate equations with no simplifying assumptions.

2. The model

The energy level diagram depicted in Fig. 1 is usually employed for the explanation of the occurrence of the thermoluminescence (TL) and phosphorescence phenomena [33]. The underlying assumptions here are that excitation takes place by an electron–hole production in the conduction and valence bands by the excitation photon, that direct band-to-band recombination is negligible and that at least one trapping state N and one kind of recombination center M are involved. Due to the trapping–detrapping process involved, this model is utilized mainly for long-life phosphorescence, however, with the right choice of the parameters, short time luminescence (which is indistinguishable experimentally from fluorescence) can also be explained. In fact, in the present work we limit the cases under consideration to the relatively simple situation in which no detrapping is allowed; this is the situation when the temperature is low enough so that neither electrons are released thermally from traps nor holes from the centers. In Fig. 1, $N(\text{m}^{-3})$ and $M(\text{m}^{-3})$ denote, respectively, the concentrations of the traps and centers in question whereas $n(\text{m}^{-3})$ and $m(\text{m}^{-3})$ denote their respective instantaneous occupancies. $x(\text{m}^{-3}\text{s}^{-1})$ is the rate of production of electrons and holes by the

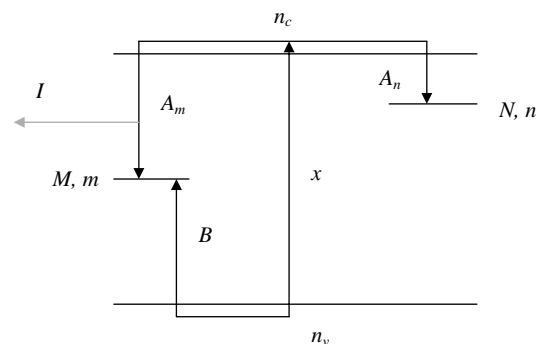


Fig. 1. Energy level diagram of a crystalline solid with concentrations of N traps and M recombination centers. The meaning of the other magnitudes shown is given in the text.

excitation irradiation, and is proportional to the excitation intensity of the sample which is assumed to be constant along the irradiation (this may be a short pulse of excitation or a longer period one). n_c and n_v (m^{-3}) are the instantaneous concentrations of electrons in the conduction band and holes in the valence band, respectively. A_n ($\text{m}^3 \text{s}^{-1}$) and A_m ($\text{m}^3 \text{s}^{-1}$) are the retrapping and recombination coefficients, respectively, and B ($\text{m}^3 \text{s}^{-1}$) is the trapping coefficient of free holes during the excitation. The equations governing the process during the excitation are

$$dn_v/dt = x - B(M - m)n_v, \quad (5)$$

$$dm/dt = -A_m m n_c + B(M - m)n_v, \quad (6)$$

$$dn/dt = A_n(N - n)n_c, \quad (7)$$

$$dn_c/dt = dm/dt + dn_v/dt - dn/dt. \quad (8)$$

These equations are solved numerically for a certain period of excitation time. Once the excita-

tion is terminated, we set $x=0$ and continue the numerical solution of the equations for a further period of time. The luminescence emission intensity during this time is assumed to be associated with the rate of electron-hole recombination

$$I = -dm/dt = A_m m n_c. \quad (9)$$

3. Numerical results

In this section, some numerical results of luminescence decay curves will be shown, along with their best fit to the stretched-exponential function (4). The “best fit” procedure chosen consisted of minimizing the sum of squares of the differences between the simulated experimental points (I_i) (from Eq. (9)) and the relevant points on the stretched-exponential function

$$\Delta = \sum_{i=1}^K \{I_i - I_0 \exp[-(t_i/\tau)^\beta]\}^2, \quad (10)$$

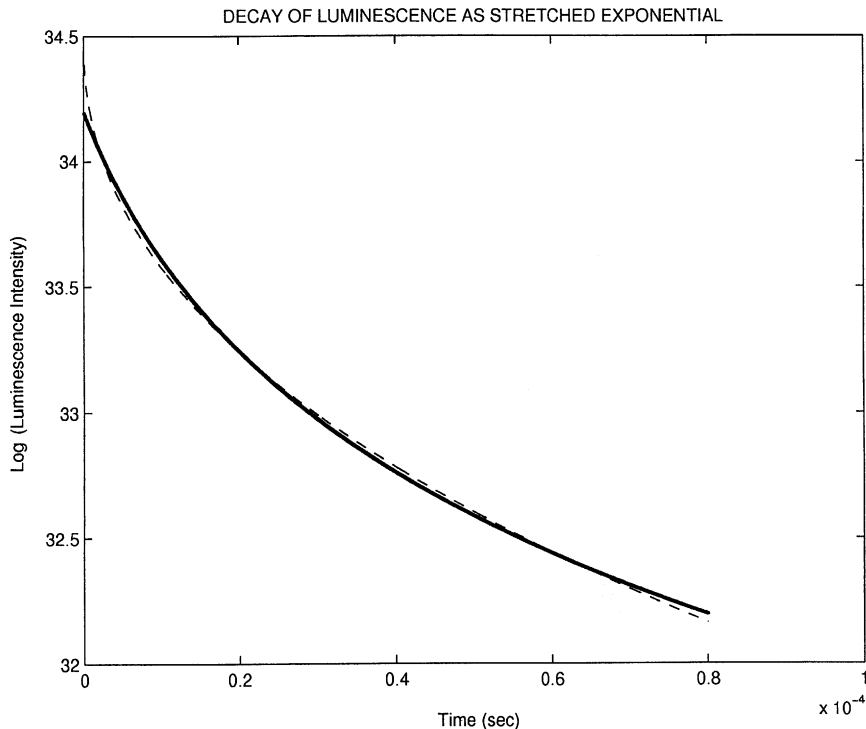


Fig. 2. Decay of luminescence as calculated for a given set of parameters (given in the text) in the range of 0–80 μs and its stretched-exponential best fit with $\beta = 0.45$ and $\tau = 11.8 \mu\text{s}$ and $\text{FOM} = 6.7 \times 10^{-4}$.

using the *fmins* minimization program in the Matlab package, with the three variables, I_0 , τ and β . K is the number of points at which the luminescence intensities were evaluated. As a “figure of merit” for the goodness of the fit we have taken

$$\text{FOM} = (\Delta/K)^{1/2}/I_0. \quad (11)$$

Here, Δ is the value of the expression in Eq. (10) at its minimum. The division by the number of points, K , is in order to be able to compare the goodness of fit between cases with different number of points, and the square root is taken so that the numerator has dimensions of intensity. Thus, FOM as defined in Eq. (11) is dimensionless.

Fig. 2 depicts, on a semilog scale an example simulated with the following parameters: $x = 10^{19} \text{ m}^{-3} \text{ s}^{-1}$; $A_m = 10^{-17} \text{ m}^3 \text{ s}^{-1}$; $A_n = 10^{-9} \text{ m}^3 \text{ s}^{-1}$; $B = 10^{-17} \text{ m}^3 \text{ s}^{-1}$; $N = 10^{18} \text{ m}^{-3}$; $M = 10^{19} \text{ m}^{-3}$. Luminescence intensities at 800 points along the decaying curve have been calculated following a

simulated excitation for 0.1 s. The thick line consists of the 800 computed points whereas the dashed line is the best fitted stretched-exponential (Eq. (4)). The best-fit parameters are $\beta = 0.45$ and $\tau = 11.8 \mu\text{s}$. The fit does not look very good, and the value of FOM (Eq. (11)) is 6.7×10^{-4} . However, the main feature of the stretched-exponential function is seen, namely, that on the semilog scale it is concave, having a negative first derivative and a positive second derivative. Furthermore, had we simulated a moderate noise, the agreement with the stretched-exponential curve would have been quite good. However, a closer look at the results indicates that the behavior at the very beginning of the curve is different than the rest of it. This brings to mind the idea that, perhaps, a better agreement will be reached if different best-fit procedures are performed for the first short period of time and for the rest of the curve. This idea seems to be in accord with the work mentioned in the Introduction by Pavesi and Ceschini [10] who described a

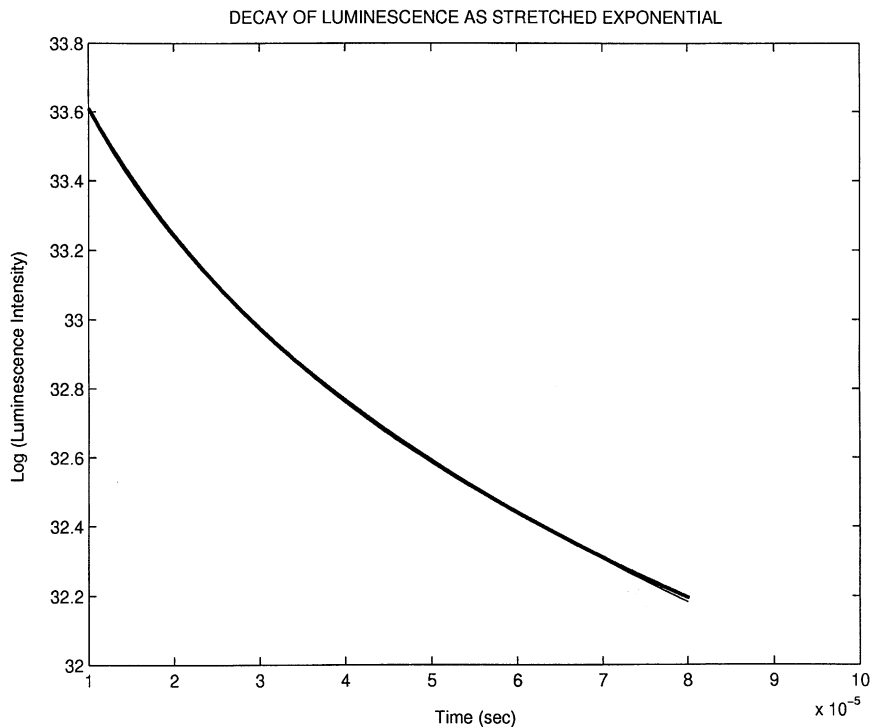


Fig. 3. Fit of the same simulated luminescence points as in Fig. 2 in the range of 10–80 μs , to a stretched-exponential curve. The best fit yields $\beta = 0.33$ and $\tau = 3.3 \mu\text{s}$ and $\text{FOM} = 1.7 \times 10^{-4}$.

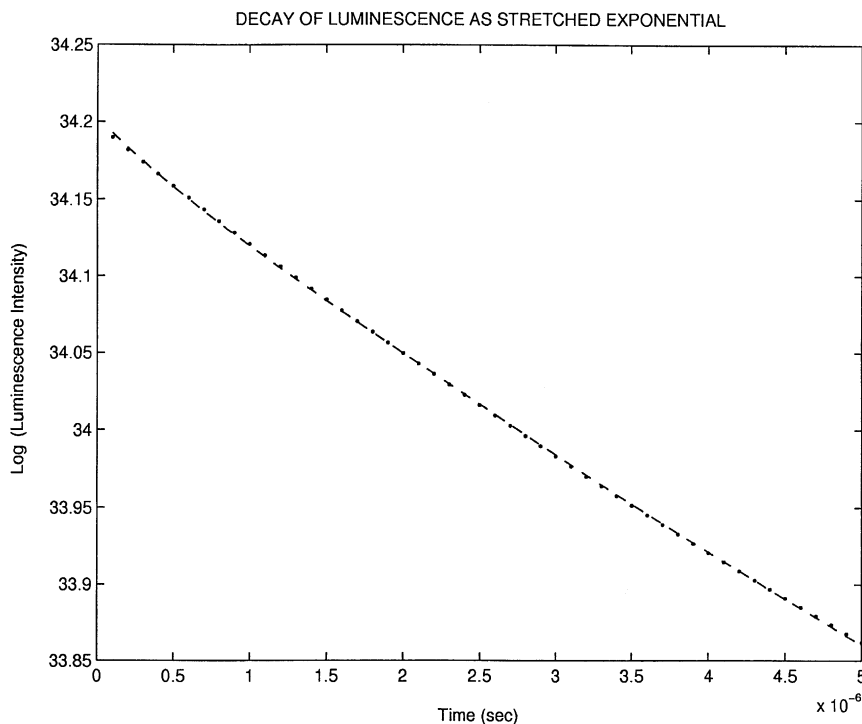


Fig. 4. Fit of the simulated luminescence points as in Fig. 2 in the range of 0–5 μs . The best fit yields $\beta = 0.875$, $\tau = 17 \mu\text{s}$ and $\text{FOM} = 2.65 \times 10^{-5}$.

fast, nearly exponential range of decay in the luminescence of porous silicon, followed by a relatively slow, stretched-exponential decay.

Fig. 3 shows the fit of the same results as in Fig. 2 to a stretched-exponential when the first 10 μs are excluded; thus the time range is 10–80 μs . The values of the relevant parameters here are $\beta = 0.33$ and $\tau = 3.3 \mu\text{s}$. As compared to other simulated curves (not reported here in detail), this is a very low value of β , probably associated with the fact that the retrapping coefficient A_n is eight orders of magnitude larger than the recombination coefficient A_m . The agreement between the simulated and best-fitted curves is significantly better, which is demonstrated by the value of $\text{FOM} = 1.7 \times 10^{-4}$, about 4 times smaller than with the results of Fig. 2. A slight deviation is seen at the longer time end.

Fig. 4 depicts the results of the best fit for the 50 points in the first 5 μs . Here, $\beta = 0.875$, $\tau = 17 \mu\text{s}$ and $\text{FOM} = 2.65 \times 10^{-5}$, more than 6 times smaller

than in Fig. 3. The large value of β , rather close to unity, means that the behavior here is close to be a simple exponential decay, in agreement with the mentioned results by Pavesi and Ceschini [10] and the work by Even et al. [8] (see Eq. (3)).

Several other runs of the solution of the sets of differential equations with different sets of trapping parameters have been performed. Typically, like in the results shown here, the initial decay was close to exponential which later turned into stretched-exponential with the values of β and τ depending on the set of trapping parameters. Although here, the value of τ in the initial, nearly exponential range has been found to be larger than that at the stretched-exponential region, with other sets of parameters, the initial decay had τ values smaller than in the stretched-exponential range. Obviously, when the parameters were such that recombination was significantly stronger than retrapping, the resulting decay curve was a simple exponential.

4. Conclusion

In this work it has been demonstrated that the stretched-exponential decay of luminescence can be the result of carriers transport in single crystals with at least one kind of recombination center and one trapping state. The well-known rate equations utilized for the study of TL and phosphorescence [33] have been solved numerically for the two stages of excitation by a light pulse and the decay of light emission following it, and the results have been best-fitted to the stretched-exponential (4). Typically, the decay curve has the features of the stretched-exponential function provided the re-trapping is stronger than the recombination. The main feature here is that on a semilog scale, the decaying curve is concave, namely has a positive second derivative. The fact that the fit of the whole decaying curve was not perfect could be masked by adding a moderate simulated noise, which indeed is usually seen in the measurements. As demonstrated here, however, a significantly better fit is usually reached if the fit is performed separately for the initial time range where a larger value of β is found, closer to unity, and for the longer time range where smaller values of β are found, indicating “real” stretched-exponential behavior. It should also be mentioned that perfect agreement could not be expected since the number of free parameters entering the model (Eqs. (5)–(8)) exceeds the number of parameters in the stretched-exponential expression (I_0 , β and τ in Eq. (4)). The main point made here is that there is a possibility of getting the quite common stretched-exponential decay of luminescence from such a system with no obvious source of disorder, which is usually suggested to be the reason for this kind of decay. It appears that the main reason for the occurrence of the stretched-exponential decay has to do with the possibility of re-trapping of free electrons from the conduction band into the electron traps. The role of re-trapping in bringing about stretched-exponential decay has been suggested before [10], but always in the context of disordered systems.

Variations of the resulting β and τ with the experimental parameters such as the intensity of the excitation light and the pulse length have been observed, however more work is required to

describe in a physical manner the effect of these experimental factors on the stretched-exponential parameters. Also, the dependence on temperature not dealt with here, appears to be of importance. Previous works [10,11,14,16,18–20,25,26] have reported temperature dependence of β and τ in different systems with either an increase or decrease of these parameters observed with increasing temperature. In our preliminary results similar changes were observed, but more work is required to understand the physical significance of these changes.

Finally, it might be suggested that the disorder, so common in stretched-exponential yielding systems is manifested in the present case of single crystals by the random distribution of traps and centers in the sample. This argument is somewhat weakened by the fact that when recombination dominates, the centers are still randomly distributed but the luminescence decay is a regular exponential. It is to be noted that in the present system, there is no straight-forward way of describing the stretched-exponential decay as a sum (integral) over a distribution of exponential decays as explained to be the source of the stretched-exponential function in several other systems [6]. It may be argued that the fact that the decay is not a simple exponential function has to do with the nonlinear terms $A_m m n_c$, $B(M - m)n_v$ and $A_n(N - n)n_c$, appearing in Eqs. (5)–(7).

It should be stressed that the present work does not contradict any of the known ways of explaining the stretched-exponential decay of luminescence. It merely suggests one more possibility of explaining the phenomenon. This should be considered, for example, when a sample made of small crystallites emits luminescence. The possibility that the stretched-exponential decay results from each of the crystallites rather than the whole sample should not be ruled out.

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