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The decay of OSL signals as stretched-exponential functions

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Abstract

Optically stimulated luminescence (OSL) and pulsed OSL have been utilized broadly for luminescence dosimetry as well as archaelogical and geological dating. It has been pointed out that in many cases, the decay of the OSL *during* continuous stimulating light (cw-OSL) and that of pulsed OSL *following* a pulse of stimulating light, do not generally behave according to a simple exponential function. In the present work, it is shown by the use of numerical simulation, that with the simplest model of a single trapping state and a single kind of recombination center, a decay curve significantly slower than a "normal" exponential can emerge. These results could be fitted to a stretched-exponential law, $\exp[-(t/\tau)^{\beta}]$ with $0 < \beta < 1$ with surprisingly good agreement for the decay of OSL following a stimulating light pulse. As for the decay of OSL during the exposure to stimulating light, a typical behavior found in the simulation is an initial nearly exponential decay, followed by stretched-exponential decay at longer times. In particular, the cases where β is significantly smaller than unity (e.g., $\beta \sim 0.5$) are of interest. It is to be pointed out that several relaxation phenomena in complex condensed-matter systems have been found to follow the mentioned stretched-exponential decay law. This includes some reports in the literature of stretched-exponential decay of luminescence, usually in the very short time range. It has been suggested, however, that this behavior is always associated with some kind of disorder in the sample, e.g. the disorder occurring in porous silicon. The main new points in the present work are that this kind of relaxation can be expected to occur in the two kinds of OSL mentioned above and that they result from a single crystal with only one trapping state and one kind of recombination center. The concept of half-life of the decay in these cases is considered in view of the present results.

Keywords: Optically stimulated luminescence (OSL); Stretched-exponential; Half-life

1. Introduction

The use of optically stimulated luminescence (OSL) for dating of sediments has first been suggested by Huntley et al. (1985). These authors stated that the discrimination of induced luminescence from scattered incident light can be made on the basis of wavelength or time, and opted for the former. They reported that the decay of OSL during the laser illumination of previously irradiated (during antiquity or in the laboratory) quartz grains, is non-exponential. The method of OSL has further been investigated by several researchers both experimentally and theoretically, and has been extended for dosimetry and archaeological dating. Smith and Rhodes (1994) elaborated on the decay of OSL and stated that the decrease of OSL emission with laser exposure does not follow a simple exponential (in quartz) as would be expected from first-order detrapping from a single trapping site. The authors explain this as being due to at least two sources of charge when OSL measurements are made at 17°C. They tried to simplify the results by repeating the measurements at 220°C, but still found a decay which was not a simple exponential. They explained this as being due to phototransfer into a trapping state stable at 220°C. These authors mention the role of retrapping in their samples (based on a previous work by Aitken and Smith, 1988), but suggest that this retrapping produces only a minor

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modification to the exponential OSL decrease so long as the traps are not near saturation. In the present work, we will show, using numerical simulation, that substantial retrapping can cause significant deviation from the simple exponential decay even when only a single trapping state and a single kind of recombination center are involved. This is opposed to the conclusion of Smith and Rhodes (1994) that the released charge originates necessarily from trapping sites with more than one mean life under optical stimulation. Poolton et al. (1995) studied the OSL decay of porcelain and found that the RT signal shows highly non-exponential decay characteristics which they ascribed to a range of defects with varying depth and/or optical capture cross-sections.

McKeever et al. (1997a) presented results of computer-simulated OSL (cw-OSL) during the stimulation, based on a model with three trapping states and two recombination centers which explained a number of TL and OSL phenomena occurring in the single aliquot technique. They found a decay curve, which is approximately exponential at short times, but deviate from exponential at longer times. They define the "OSL intensity" as the area under the OSL curve between 0 and 100 s. Further theoretical work by McKeever et al. (1997b) describes the behavior of OSL as a function of temperature. The authors consider retrapping by shallow traps, thermally assisted optical stimulation, thermal quenching and localized donor–acceptor recombination. They repeat the point that the decay is usually non-exponential, typically exhibiting a long "tail" to the decay at long illumination times. Bailey (2000) reported a sub-exponential decay of OSL in quartz, which could be fitted to the stretched-exponential function (Bailey, private communication).

As pointed out above, Huntley et al. (1985) mentioned the possibility of discrimination of the induced luminescence from the incident light on the basis of time, but did not use this option. Markey et al. (1995) introduced the pulsed OSL (later termed POSL) where the laser excitation source was pulsed with pulse widths of 10 ns. The OSL emission (of α-Al_2O_3: C) was followed as a function of time after the excitation pulse. The decay curve under these circumstances was found not to be a simple exponential, and in the analysis, was fitted to the sum of two exponentials. Akselrod et al. (1998) further studied the TL and OSL properties of Al_2O_3: C and reported a non-exponential decay of the "delayed" OSL (following the stimulating pulse), and ascribed it to the occurrence of shallow trapping states. Similar results were reported by McKeever and Akselrod (1998) who found that the decay after the light exposure shows multiple components.

Another demonstration of the non-exponential decay of OSL, both during and following the stimulation has been given by Chen and Leung (2001) and by Chen (2002). They showed that within the framework of the simplest model of a single trapping state and one kind of recombination center, and provided that retrapping is substantial, the dose dependence of the initial part of the decay curve is quadratic. On the other hand, the whole area under the decaying luminescence curve is linearly dependent on the dose. Obviously, these properties are characteristic of a non-exponential decay curve.

In the present work, we would like to demonstrate that a possible decay function of OSL *during* as well as *following* the exposure to stimulating light might be closely approximated by the quite ubiquitous stretched-exponential function $\exp[-(t/\tau)^{\beta}]$ with $0 < \beta < 1$. As early as in the 19th century, Kohlrausch (1863) described mechanical creep by this function. Williams and Watts (1970) ascribed dielectric relaxation in polymers as being a stretched-exponential function, and apparently were the ones who coined the term "stretched exponential". The 'Brinkman Report' (1986) stated that "there seems to be a universal function that slow relaxations obey. If a system is driven (or normally fluctuates) out of equilibrium, it returns according to the function $\exp[-(t/\tau)^{\beta}]\ldots$ Unfortunately, this is not a mathematical expression that is frequently encountered in physics, as little idea exists of what the underlying mechanisms are". In general, 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 temperature as shown by Klafter and Shlesinger (1986).

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. Chen et al. (1992) studied the decay of luminescence in porous silicon and in CdSe–ZnSe superlattice, and found that the stretched-exponential behavior

$$I = I_0 \exp[-(t/\tau)^{\beta}] \tag{1}$$

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 $\sim$ 100 ns; for porous silicon at room temperature it was $\sim$ 100 μs and for porous silicon at 13 K, $\sim$ 10 ms. Chen et al. reported that the fitting parameters β and τ were found to depend on excitation conditions such as the excitation pulse width, intensity and photon energy.

In the following years, many papers have been published, describing the decay of luminescence as a stretched-exponential function. Much of the work reported results observed in porous silicon, but stretched-exponential decay of luminescence has been observed in other materials as well. Pavesi and Ceschini (1993) who studied the stretched-exponential decay of luminescence suggested 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 a p-Si skeleton, (II) a random spatial arrangement of the nanocrystals and (III) the structure of the nanocrystal surfaces. In conclusion, they state that the occurrence of the stretched-exponential decay of luminescence strongly points to the role of disorder. These authors report 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. (1)) where τ is a lifetime and β, a dispersive factor. The authors suggest that this decay law, often encountered in disordered systems, can be considered a consequence of the dispersive diffusion of the photoexcited carriers. The diffusion of carriers among different spatial sites can be due to either the excitation of carriers from localized to extended states or due 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 trapping release rates. In a later work, Pavesi (1996) reiterated the importance of disorder in bringing about the stretched-exponential decay behavior. He states that values of $\beta < 1$ correspond to the existence of a broad distribution of lifetimes which describe the elementary relaxation processes, either radiative or non-radiative.

Several other papers have been published, discussing different aspects of the stretched-exponential decay of luminescence in porous silicon as well as other materials such as PbI_2 monocrystals embedded in porous silica films, nanometer-sized oxidized crystallites such as SiO_2 and Si, InGaN light-emitting diodes and multiple quantum wells. Stretched-exponential decay of luminescence has also been found in nanoporous SiGe alloys and in poly(ethylene terephtalate) films. All these works point out the importance of some disorder in the samples in question for the stretched-exponential decay.

In the present work, it will be shown that stretched-exponential decay of OSL can be expected from a model of a single crystal with one trapping state and one kind of recombination center, when transition of the carriers is through the conduction band, with no inherent necessity to have a disorder in the system. This is the case for both OSL *during* the application of the stimulating light or *following* it. The differences in the nature of the decay in these two cases are indicated. No analytical proof is given to this behavior since it is not possible to solve analytically the relevant sets of non-linear simultaneous equations. However, numerical solution of the equations for plausible choices of sets of trapping parameters demonstrates this result of stretched-exponential decay of OSL.

2. The model

As pointed out above, we follow the electronic transitions in the simplest possible relevant model, namely, a model with a single trapping state and a single kind of recombination center. The process to be followed first is the radiation-induced excitation of the sample by transferring electrons from the valence into the conduction band. The forbidden gap, the relevant levels and the transitions taking place are shown in Fig. 1. N (m^{-3}) and n (m^{-3}) denote,

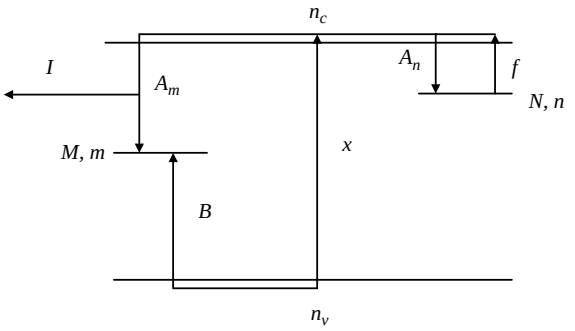

Fig. 1. Energy level diagram of a solid with one trapping state and one kind of recombination center. N and M are the concentrations of the trap and center, respectively and n, m their instantaneous occupancies. n_c and n_v are the free electron and hole concentrations, respectively. A_n and A_m are the retrapping and recombination coefficients, respectively. x is the rate of production of electrons and holes by the excitation irradiation, and f is the optical stimulation rate.

respectively, the concentration of the trapping state and its instantaneous occupancy. M (m^{-3}) and m (m^{-3}) are the concentration of the center and its instantaneous occupancy, respectively. A_n ($m^3\ s^{-1}$) and A_m ($m^3\ s^{-1}$) are the retrapping and recombination coefficients, respectively and B ($m^3\ s^{-1}$) is the trapping coefficient of the free holes during the excitation. n_c (m^{-3}) and n_v (m^{-3}) are the instantaneous concentrations of electrons in the conduction band and holes in the valence band, respectively. x ($m^{-3}\ s^{-1}$) is the rate of production of electrons and holes by the excitation irradiation, and is proportional to the excitation intensity of the sample which is assumed to be constant along the irradiation. It is also assumed that the excitation is homogeneous across the sample, which is typical of γ irradiation, X-rays as well as β particles, provided the sample is relatively thin. We assume here that during the irradiation, the temperature is low enough so that there is no thermal release of trapped electrons (or, in other words, that the trap is deep enough not to allow the thermal release of electrons during the irradiation). The equations governing the process during the excitation are:

$$dn_v/dt = x - B(M - m)n_v, \tag{2}$$

$$dm/dt = -A_m m n_c + B(M - m)n_v, \tag{3}$$

$$dn/dt = A_n(N - n)n_c, \tag{4}$$

$$dn_c/dt = dm/dt + dn_v/dt - dn/dt. \tag{5}$$

This set of equations can be numerically solved using a standard Matlab solver for given sets of the parameters. At the end of the excitation, electrons and holes may remain in the conduction and valence bands, respectively. In order to simulate the experimental conditions properly, we set $x = 0$ and continue the numerical solution for a further period of

relaxation time so that the concentrations n_c and n_v decay to negligible values.

The next stage is that of optical stimulation and we assume that the applied light releases electrons from the trapping state at a rate of fn (m^{-3} s^{-1}) where f (s^{-1}) is the optical stimulation rate (see Fig. 1). We assume here homogeneous illumination of the sample. fn is analogous to x (m^{-3} s^{-1}) in the excitation stage, except that x is constant whereas fn varies with n. We can take now the final values of the functions n, m, n_c and n_v (the latter two having very small values) at the end of the relaxation as initial values for the optical stimulation. Three simultaneous differential equations are to be considered:

$$-\mathrm{d}m/\mathrm{d}t = A_m m n_c, \tag{6}$$

$$\mathrm{d}n/\mathrm{d}t = -fn + A_n(N-n)n_c, \tag{7}$$

$$\mathrm{d}n_c/\mathrm{d}t = \mathrm{d}m/\mathrm{d}t - \mathrm{d}n/\mathrm{d}t. \tag{8}$$

These are also solved using the Matlab package. Since we associate the intensity of the OSL signal with the recombination rate, we can write the OSL intensity I as

$$I = -\mathrm{d}m/\mathrm{d}t. \tag{9}$$

Here, a proportionality factor should have preceded the right-hand side of Eq. (9); however, omitting it merely means that we measure the emission intensity in some different units.

While considering the OSL decay *during* stimulation, the solution of these Eqs. (6)–(9) should yield the decaying curve. As for the OSL *following* stimulation, we should run Eqs. (6)–(8) for the length of time of the simulated pulse, and continue the run setting $f = 0$. Here, the decay time is taken starting from the end of the light pulse, and the emitted light intensity is given again by Eq. (9).

3. Numerical results

Several runs of the mentioned sets of differential equations have been performed with different sets of the relevant parameters. As mentioned above, an important condition for getting a decay which is far from being a simple exponential is the occurrence of a relatively high retrapping coefficient. However, the conditions for getting different values of β depended on all the relevant trapping parameters. Anyway, in practically all the cases checked, a very good agreement with the stretched-exponential function took place with values of β typically ranging from 0.5 to unity. Also, it has been pointed out that the agreement with the stretched-exponential is usually better in the pulsed OSL where the emitted light is monitored following the stimulating light pulse than in the case of decay of OSL during the exposure to the stimulating light.

The "best fit" procedure chosen consisted of minimizing the sum of squares of the differences between the simulated experimental points (I_i) and the relevant points on the stretched-exponential function

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

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

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

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.

Before going to some numerical examples, it should be mentioned that within the limited framework discussed here of one trapping state and one kind of recombination center, performing the numerical solution of Eqs. (2)–(5) is not needed. The reason is that here at the end of the stages of excitation and relaxation, and if the latter is long enough, we end up with equal final occupancies of the trap and center. These values are the initial occupancies for the next stage of optical stimulation, and therefore we can just choose some values of $n_0 = m_0$ and proceed. Eqs. (2)–(5) were mentioned since they are required when one is interested in the decay of luminescence following the initial excitation. Also, the extension of this set of equations is of interest if the system in question has more than only one trapping state and one kind of recombination center, and in this case, the solution of the relevant set of coupled equations during the excitation stage is essential.

The relevant parameters to be chosen here are A_m and A_n, the recombination and retrapping coefficients, N the total concentration of trapping states, $n_0 = m_0$ the initial filling of traps and centers following the excitation and relaxation, and f, the optical stimulation rate. In the case of OSL *following* the light pulse, the length of the pulse t_f is also of interest.

Fig. 2 shows the results of a simulation of cw-OSL with the chosen parameters: $A_m = 10^{-12}$ m^3 s^{-1}; $A_n = 10^{-13}$ m^3 s^{-1}; $N = 10^{17}$ m^{-3}; $n_0 = m_0 = 10^{16}$ m^{-3}; $f = 1$ s^{-1}. The results are shown on a semilog scale. The solid line consists of 1000 simulated points at intervals of 0.01 s. The dashed line is the best-fitted curve to the logarithm of Eq. (1), namely, $\ln I_0 - (t/\tau)^{\beta}$. The simulated decay curve is seen to be sub-exponential; however, it is visually observed that the agreement is not very good. The effective fitting parameters found are $\tau = 0.79$ s and $\beta = 0.57$. The value of the FOM defined in Eq. (11) here is $\mathrm{FOM} = 4.9 \times 10^{-3}$.

Bearing in mind the possibility mentioned by McKeever et al. (1997a) that the decay curve may consist of two components, at short and long times, the numerical OSL values were fitted separately for the first second and for the period of time from $t = 2$ to 10 s. Fig. 3 shows the latter. The fit

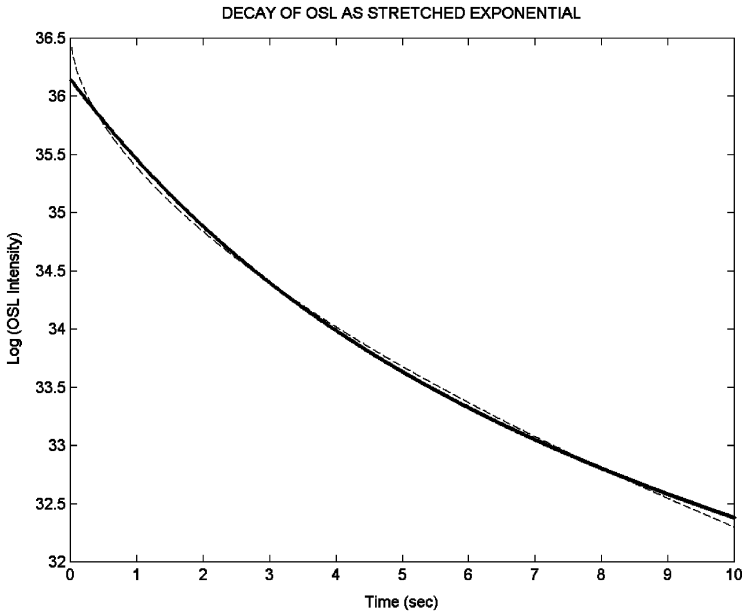

Fig. 2. Simulated decay curve (solid line) during optical stimulation (cw-OSL), and the best-fitted stretched-exponential function (dashed line) for a given set of parameters specified in the text and for a time range of 0–10 s, on a semilog scale.

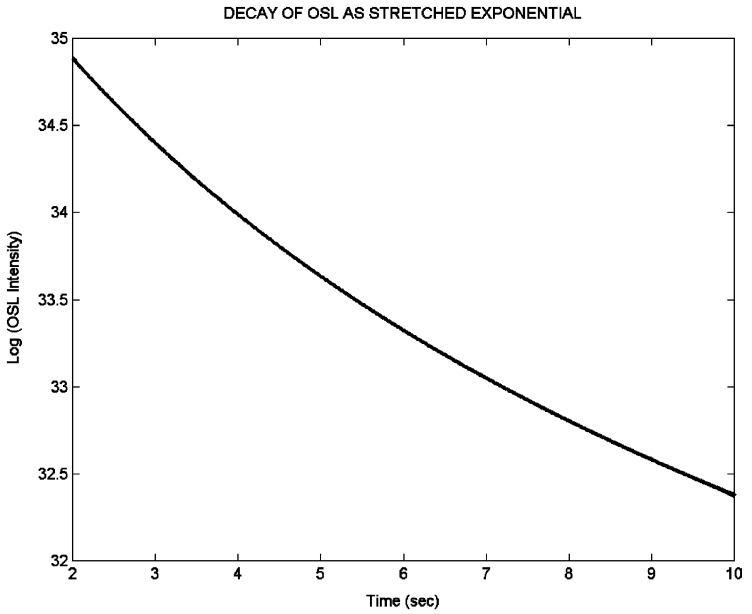

Fig. 3. Simulated decay curve and best fitted stretched exponential on a semilog scale. The results are the same as in Fig. 2, but only in the time range of 2–10 s.

looks significantly better with the fitting parameters $\beta = 0.35$ and $\tau = 0.057$ s with FOM $= 1.7 \times 10^{-4}$, more than an order of magnitude better than for the whole curve. Fig. 4 shows the best fit for the first second. Here, we get $\beta = 0.94$ and $\tau = 1.47$ s. The high value of β means that the initial decay is nearly exponential. Here we get FOM $= 3.3 \times 10^{-5}$, 5 times better than in Fig. 3.

Fig. 5 depicts the numerical results for a decay *following* the exposure to the stimulating light. The parameters chosen here are: $A_m = 10^{-20}$ m^3 s^{-1}; $A_n = 10^{-16}$ m^3 s^{-1};

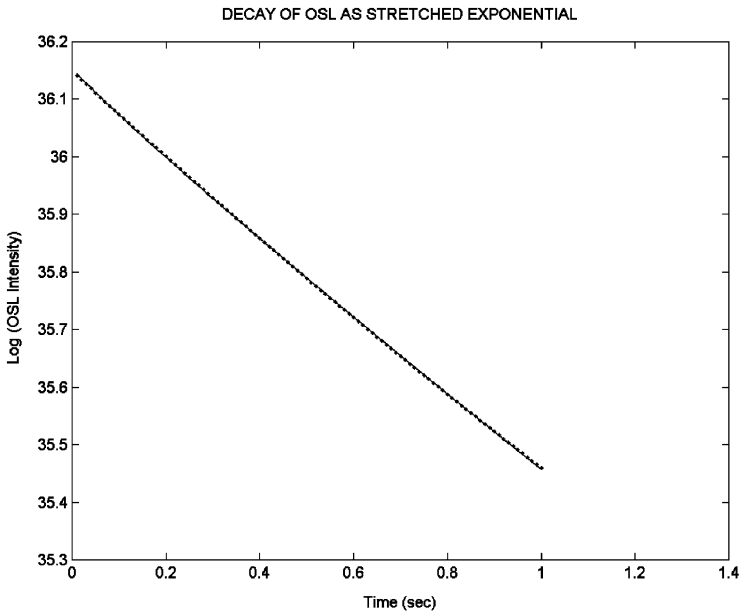

Fig. 4. The best-fitted curve for the same trapping parameters as in Figs. 2 and 3, in the time range of 0–1 s.

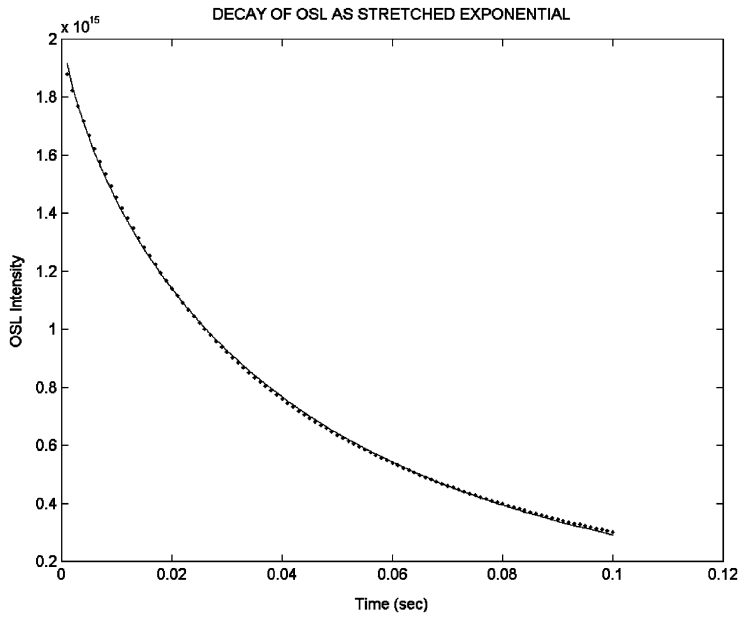

Fig. 5. Simulated decay curve (points) and best-fitted stretched-exponential function (solid line) following a light pulse. The set of chosen parameters and the resulting stretched-exponential parameters are given in the text. The results here are shown on a linear scale.

$N = 10^{18}$ m^{-3}; $n_0 = m_0 = 9 \times 10^{17}$ m^{-3}; $f = 10$ s^{-1} and the length of the stimulation pulse is $t_f = 0.1$ s. One hundred numerically simulated points are seen and the solid line indicates the best-fitted curve. The fitting parameters here are $\beta = 0.75$ and $\tau = 0.041$ s. As opposed to Figs. 2–4, the results here are on a linear (not logarithmic) scale in order to demonstrate the possibility of fitting the curve this way. Repetition of the curve fitting with the same data and on a logarithmic scale resulted in practically the same β and τ values. The FOM here is 3.1×10^{-4}.

4. Discussion

It has been demonstrated that within the framework of a model with a single trapping state and a single kind of recombination center, the decay of OSL with time can be approximated by the stretched-exponential function (Eq. (1)). For the case of the decay of OSL *following* a stimulating light pulse, the agreement has been quite good; more often than not, the decay curve was found to be nearly simple exponential. Values of β significantly smaller than unity have been found either when the initial pulse has been very large (large value of f, meaning that the system is initially significantly out of equilibrium), or when the trapping state is initially close to saturation. As for the OSL decay during simulation (cw-OSL), the characteristic behavior found has been of an initial nearly exponential decay, followed by a stretched-exponential decay. This is in good agreement with simulation based on a system with three trapping states and two kinds of recombination centers (McKeever et al., 1997a). These authors found a nearly exponential decay at short times and slower decay at longer times. Our present results indicate that this behavior may be associated with the basic process occurring in our simpler model, and not necessarily with the model by McKeever et al. which obviously includes many more parameters.

It is to be noted that even in our simpler model, the stretched exponential appears to be merely an approximation to the decay curve. We start with five parameters, namely, $A_m, A_n, N, n_0 = m_0$ and f, and in the pulsed OSL also a sixth one, namely t_f (the length of the stimulating pulse) and fit the results to a function depending on three parameters, I_0, β and τ. Obviously we cannot expect a perfect agreement. An attempt can be made in the future to try to define analytically under what circumstances (namely, what relations between the given parameters), the stretched-exponential decay should be expected. It is encouraging to note that the decay of OSL enters into a larger group of relaxation phenomena, which exhibit the stretched-exponential behavior. At the same time, the fact that with all those phenomena no analytical proof for this time dependence was found is somewhat discouraging.

A common feature of the stretched-exponential functions for different values of β is that if we define the "lifetime" as the time required to decay to e^{-1} of the initial intensity, one gets $t_{1/e} = \tau$ irrespective of β. As for the time needed to decay to half intensity, we get $t_{1/2} = \tau(\ln 2)^{1/\beta}$. Thus, for $\beta = 1$, we get the well-known relation $t_{1/2} = \tau \ln 2$, and for, say, $\beta = 0.5$, we get $t_{1/2} = \tau(\ln 2)^2$. In this respect, we mention the fact that both in the stretched and simple exponential, the expression for half-life is simple and independent of the initial intensity I_0. This is opposed to other expressions such as $A \exp[-(t/\tau_1)] + B \exp[-(t/\tau_2)\beta]$ where the expression for half-life is significantly more complicated and depends on the parameters A and B as well as on τ_1, τ_2 and β.

It might be thought that in the systems discussed here, the strong retrapping plays the role of disorder usually quoted as being the source of the stretched-exponential behavior. We tend to believe that this is not the case. Even when the retrapping is rather strong, we are talking about a rather sparse random distribution of trapping states, and the crystal under consideration basically remains unchanged. Furthermore, if we had a mean to arrange the trapping states in a perfect orderly manner, the rate equations would still be the same and therefore the consequences of having a stretched-exponential decay would remain unchanged. It has been suggested by Huber (private communication) that the real reason for the stretched-exponential behavior here has to do with the non-linearity of Eqs. (6) and (7), in which terms like $A_m m n_c$ and $A_n(N - n)n_c$ are included.

It should be noted that simulated experimental results here were ideal in the sense that, as opposed to real experimental results, no noise was included. Some preliminary calculations have been made in which different levels of simulated noise have been added. Without going into details, we report here that, as could be expected, the FOM increased with the level of the noise, and from a certain noise level up, the FOM depended nearly linearly on the noise level. In spite of this, the minimization procedure brought about nearly the same best-fitted decay curves with nearly the same values of β and τ, practically independently of the noise level.

It might be interesting in the future to check whether the fit to other decay curves such as the "general order" decay yield similar goodness of fit, as reflected by the calculated values of the FOM. In particular, it would be of interest to test whether such best-fit programs can distinguish between different decay functions in the presence of rather high noise levels.

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