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Evidence for an energy dependent localization length of gap states in *a*-Si:H?

A new interpretation of time-resolved photoluminescence spectra

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In honor of Emanuel Mooser's 60th birthday

Abstract. We present an extended multiple-trapping model for the description of the relaxation of photoexcited carriers in *a*-Si:H which in particular takes into account transitions between the localized tail states. Time- and energy-resolved photoluminescence spectra are interpreted in terms of this model. We show that the experimentally observed features of the photoluminescence spectra indicate a strong energy dependence of the localization length of the tail states near the mobility edges.

I. Introduction

It is nowadays well accepted that the electronic states in amorphous semiconductors can be divided in two distinct classes: *Extended states* above the mobility edges [1] and *localized states* within the gap. In the past many experiments have been devised in order to obtain some information about the character of the localized states within the gap and their distribution in energy. In the case of *a*-Si:H, optical absorption [2], photoinduced absorption [3] and photoemission yield experiments [4] suggest that the density of states decreases exponentially with the distance in energy from the mobility edges. It appears to be difficult to obtain conclusive experimental information about the wave function of the localized states in the exponential tails. The localization lengths for states near the Fermi level can be estimated from hopping transport data [5]. The energy dependence of the localization length is however unknown from the experimental point of view. Theoretically one expects a localization length which decreases as the distance in energy from the mobility edges increases [6].

In the present paper we show that time- and energy-resolved photoluminescence (PL) experiments may give useful information on the localized states near the mobility edges. PL experiments on *a*-Si have been extensively discussed in the literature [7]. The time decay of the PL intensity following a pulsed excitation follows a power law $t^{-\beta}$ over two to three decades in time and is similar to the decay found in transient photocurrent experiments. The latter experiments have been successfully interpreted in terms of the multiple-trapping model (MTM) [8, 9]. The similarity between the results of PL and photoconductivity experiments

suggests that in both cases the physical relaxation mechanism is the same and that it should be possible to explain both experiments within the same model [10, 11, 12]. A tentative explanation in this spirit of the PL decay has been proposed recently [11]. However, several problems still persist:

i) The temperature dependence of the exponent β deduced from the MTM is much too large compared with experimental PL results.

ii) The distribution in photon energy of the PL remains unexplained.

iii) The red shift of the luminescence spectra with time is not understood.

In the present work we propose an extended version of the MTM. In contrast with the conventional MTM our extended model

i) allows for transitions between localized states.

ii) allows for radiative recombinations which originate from the localized levels.

iii) allows the investigation of the effect of an energy dependent localization length.

We compare the obtained theoretical results with experiment. It is found that the problems mentioned above are resolved by the new approach. We present evidence for a strongly energy dependent localization length of the tail states.

II. The extended multiple-trapping model

(a) Basic assumptions

In the following we group the energy levels in the tail region on one side of the gap in intervals of width ΔE and centered at energies E_i . The index i numbers the intervals starting with $i = 0$ at the mobility edge. If $g(E)$ describes the density of tail states, we obtain for the total number of levels within that interval $N_i = g(E_i) \Delta E$. The transitions considered are presented in Fig. 1. The model includes:

i) transitions between the extended states in one band and localized tail

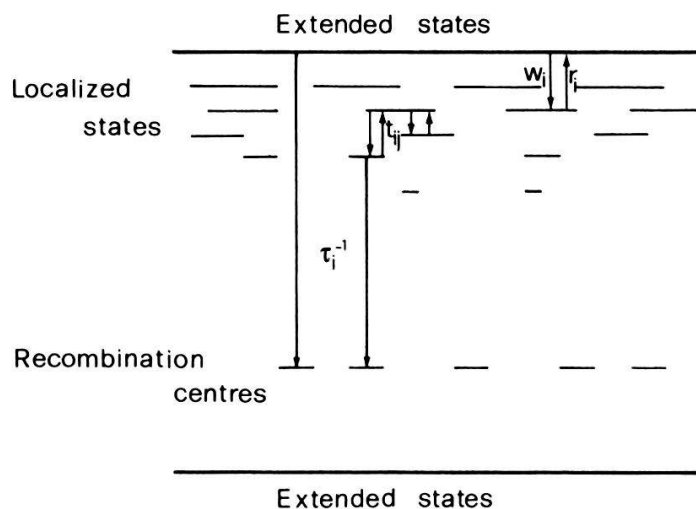


Figure 1
Transitions taken into account in the extended multiple-trapping model.

states with energy E_i in its band tail. The transition rates are given by the trapping coefficients w_i and the release coefficients r_i .

ii) radiative transitions from the same band and its tail states at energy E_i , into the recombination centres on the other side of the gap. For sake of simplicity all recombination centres are assumed to have the same energy. The transition rates are given by τ_i^{-1} .

iii) transitions between the localized levels at energies E_i and E_j . The transition rates for transitions from states at E_i (E_j) to states at E_j (E_i) are given by t_{ij} (t_{ji}).

We shall assume in the following that the number of occupied levels $n_i(t)$ in an interval i is negligible compared with the total number of states N_i in that interval for all times considered. The same condition is applied to the recombination centres. Thus we are explicitly excluding any effects of saturation.

(b) Transition matrix elements

For the calculation of the transition rates t_{ij} for an electron in interval i to one of the levels in interval j we assume that

i) states $i_1 \cdots i_{N_i}$ with energy E_i have hydrogen-type envelope functions $(\alpha(E_i)^3 \pi^{-1})^{1/2} \exp(-\alpha(E_i)r)$, where $\alpha(E_i)$ represents the reciprocal localization length;

ii) the reciprocal localization length α increases with the distance in energy from the mobility edge;

iii) in analogy with the assumptions made in the hopping theory [13] the transition matrix elements are proportional to the spatial overlap between the envelope functions corresponding to the localized functions in question.

We consider first the transition rate P_{i_s, j_t} from state i_s to state j_t . The two states belonging to the energies E_i and E_j are centered at \mathbf{r}_s and \mathbf{r}_t , respectively. With

$$\mathbf{r} = \mathbf{r}_s - \mathbf{r}_t$$

we obtain

$$\begin{aligned} P_{i_s, j_t} &= P_{i, j}(|\mathbf{r}_s - \mathbf{r}_t|) \\ &= cf_{i, j}(T)(\alpha(E_i)\alpha(E_j))^{3/2}\pi^{-1} \\ &\quad \times \int \exp(-\alpha(E_i)r) \exp[-\alpha(E_j)|\mathbf{r} - (\mathbf{r}_s - \mathbf{r}_t)|] d^3r \end{aligned} \tag{1}$$

with

$$f_{ij}(T) = \begin{cases} \exp(-(E_j - E_i)/kT) & \text{for } E_i < E_j \\ 1 & \text{otherwise.} \end{cases}$$

The total transition rate from interval i to interval j is then obtained from

$$\begin{aligned} t_{ij} &= \sum_t P_{i_s, j_t} \\ &= \frac{N_j}{V} \int P_{i, j}(|\mathbf{r}_s - \mathbf{r}_t|) d^3r_t \end{aligned} \tag{2}$$

where V is the volume of the sample.

It is evident that this replacement of the sum by an integral is only justified if enough states overlapping with the state i_s exist in the interval j , such that the actual spatial distribution of levels j_i can be replaced by their concentration N_j/V . We note that this condition is generally not satisfied for low temperatures and long times. In this case practically all electrons are confined in strongly localized levels and the spatial distribution of neighboring localized levels and recombination centres must be taken into account explicitly [14].

The integrals in equations 1, 2 can be solved analytically and one obtains

$$t_{ij} = b_c N_j f_{ij}(T) (\alpha(E_i) \alpha(E_j))^{-3/2}. \quad (3)$$

As the wave functions of the localized states are not true exponential functions, the prefactor b_c will in principle be energy dependent. In the following this will be neglected, i.e. we assume that the energy dependence of t_{ij} is dominated by the energy dependence of the spatial overlap. Similarly, we neglect any temperature dependence of b_c . The expressions for the reciprocal life times τ_i^{-1} and for the trapping and release rates w_i and r_i are obtained from equation (3):

$$\begin{aligned} \frac{1}{\tau_0} &= b_v \frac{N_{rc}}{(\alpha(0) \alpha_{rc})^{3/2}} =: \frac{\tilde{b}_v}{\alpha(0)^{3/2}} \\ \frac{1}{\tau_i} &= b_v \frac{N_{rc}}{(\alpha(E_i) \alpha_{rc})^{3/2}} =: \frac{\tilde{b}_v}{(\alpha(E_i))^{3/2}} \\ w_i &= \frac{b_c N_i}{(\alpha(0) \alpha(E_i))^{3/2}} \\ r_i &= w_i \exp(-E_i/kT) g(0) kT/N_i \end{aligned} \quad (4)$$

α_{rc}^{-1} is the localization length of the recombination centres, N_{rc} is the number of recombination centres. $\alpha(0)^{-1}$ is the 'localization length' of the extended states, which is taken equal to the sample dimension. $g(0)$ is the density of states at the mobility edge.

(c) Rate equations and solution

With the transition rates calculated above we can write the rate equations for the number of electrons $n_i(t)$ in the intervals i

$$\begin{aligned} \frac{dn_0}{dt} &= \sum_i n_i r_i - n_0 \sum_i w_i - \frac{n_0}{\tau_0} \\ \frac{dn_i}{dt} &= -n_i r_i + n_0 w_i - n_i \sum_j' t_{ij} + \sum_j' n_j t_{ji} - \frac{n_i}{\tau_i} \end{aligned} \quad (5)$$

After a Laplace transform

$$n_i(t) = \int_0^\infty \exp(-st) n_i(s) ds$$

we obtain a system of linear equations

$$\begin{aligned}
 -sn_0(s) &= \sum_i n_i(s)r_i - n_0(s) \sum_i w_i - \frac{n_0(s)}{\tau_0} \\
 -sn_i(s) &= -n_i(s)r_i + n_0(s)w_i - n_i(s) \sum_j' t_{ij} + \sum_j' n_j(s)t_{ji} - \frac{n_i(s)}{\tau_i}
 \end{aligned}
 \tag{6}$$

The eigenvalues s_i and eigenvectors \mathbf{u}_i of the system can be found easily by standard numerical techniques. With

$$\mathbf{u}_i = \begin{pmatrix} n_0(s_i) \\ \vdots \\ n_N(s_i) \end{pmatrix}
 \tag{7}$$

where N is the number of intervals considered, we obtain

$$\begin{pmatrix} n_0(t) \\ \vdots \\ n_N(t) \end{pmatrix} = \sum c_i \mathbf{u}_i \exp(-s_i t)
 \tag{8}$$

The coefficients c_i are defined by the initial conditions at $t = 0$, which are taken as

$$\begin{aligned}
 n_0(t = 0) &= \tilde{n}_0 \\
 n_i(t = 0) &= 0 \quad \text{for } i = 1 \cdots N.
 \end{aligned}
 \tag{9}$$

III. Application to the case of a -Si:H

In order to describe the luminescence properties of a -Si:H we must still specify the energy dependence of the density of tail states $g(E)$ and of the localization length $\alpha^{-1}(E)$ near the conduction mobility edge. We write

$$\begin{aligned}
 g(E) &= \frac{N_L}{kT_0} \exp(-E/kT_0) \\
 \alpha(E) &= A \exp(E/kT_1)
 \end{aligned}
 \tag{10}$$

where N_L is the total number of tail states. The exponential energy dependence assumed for $\alpha(E)$ may appear surprising at first sight. Generally, for reasons of simplicity a constant localization length independent of the energy is assumed. The parameters of the models are then those in equations (4) and (10): A , N_L , kT_0 , kT_1 , b_c , \tilde{b}_v . We note that we have assumed that all the transitions involving the recombination centres are radiative, i.e. the contribution of electrons in interval i to the PL is equal to n_i/τ_i . In general one should allow for non-radiative transitions [15] and write

$$\frac{1}{\tau_i} = \frac{1}{\tau_{ir}} + \frac{1}{\tau_{inr}}$$

where τ_{ir} , (τ_{inr}) is the radiative (non-radiative) life time. However, this would increase the number of parameters. The successful fit to the experimental curves presented in Section IV, using only the limited set of parameters above show, that the non-radiative transitions do not significantly perturb the recombination statistics, except perhaps for high excitation intensity and short times ($<0.1 \mu\text{s}$).

IV. Comparison with experiment

The theoretical curves presented in Figs. 2 to 5 are obtained with the following parameters:

$$\begin{aligned} N_L &= 10^{21} & kT_0 &= 60 \text{ meV} \\ kT_1 &= 15 \text{ meV} & A &= 1 \text{ cm}^{-1} \\ b_c &= 1.2 \times 10^{-8} \text{ s}^{-1} \text{ cm}^{-3} & \tilde{b}_v &= 2.5 \times 10^7 \text{ s}^{-1} \text{ cm}^{-3/2} \end{aligned}$$

The calculated total PL intensity for 77 K

$$I_{\text{tot}}(t) = \sum_i \frac{n_i}{\tau_i}$$

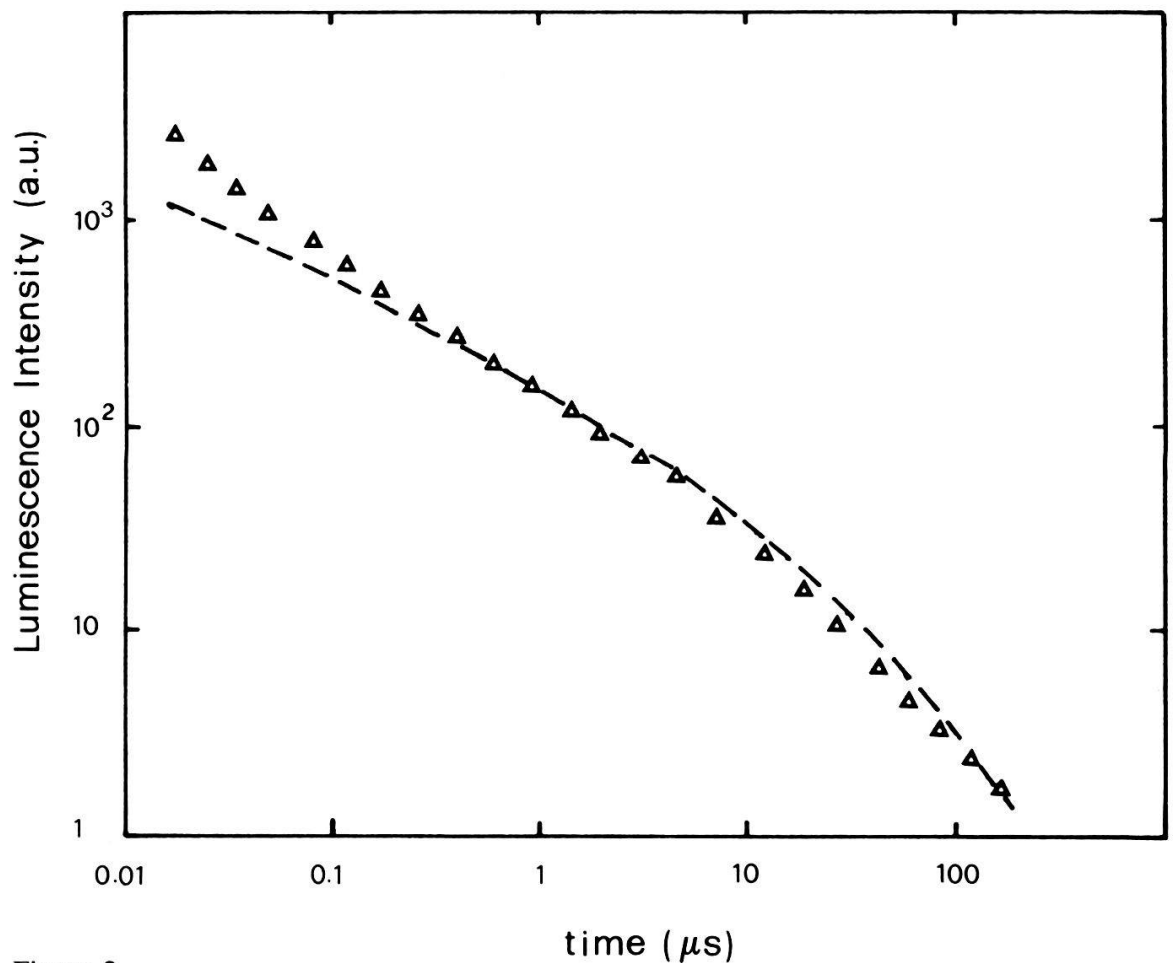


Figure 2

Total photoluminescence intensity vs. time. Δ : Experimental decay following a pulsed excitation. Undoped *a*-Si:H, excitation energy 2.31 eV, excitation density $6 \times 10^{-7} \text{ J cm}^{-2}$. Temperature 77 K. ---: Theoretical decay as predicted by our model.

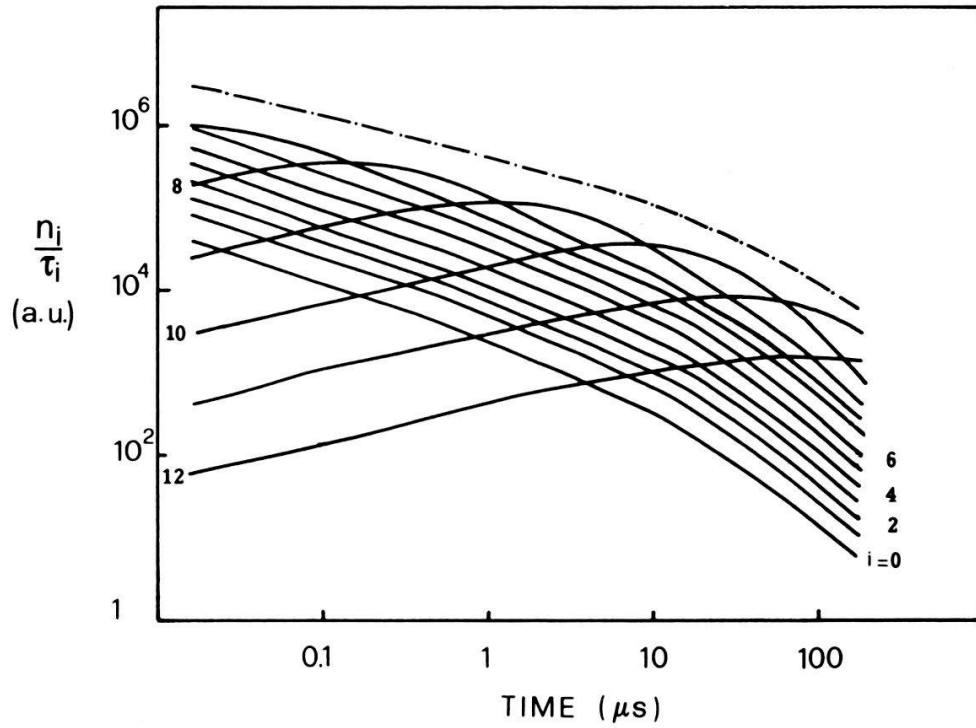


Figure 3
 Contributions to the photoluminescence emission from the different energy intervals with energies $E_i = i \cdot 0.012$ eV below the mobility edge. $i = 0$ corresponds to contributions from the mobility edge with $E_0 = 0$. — · —: total photoluminescence intensity.

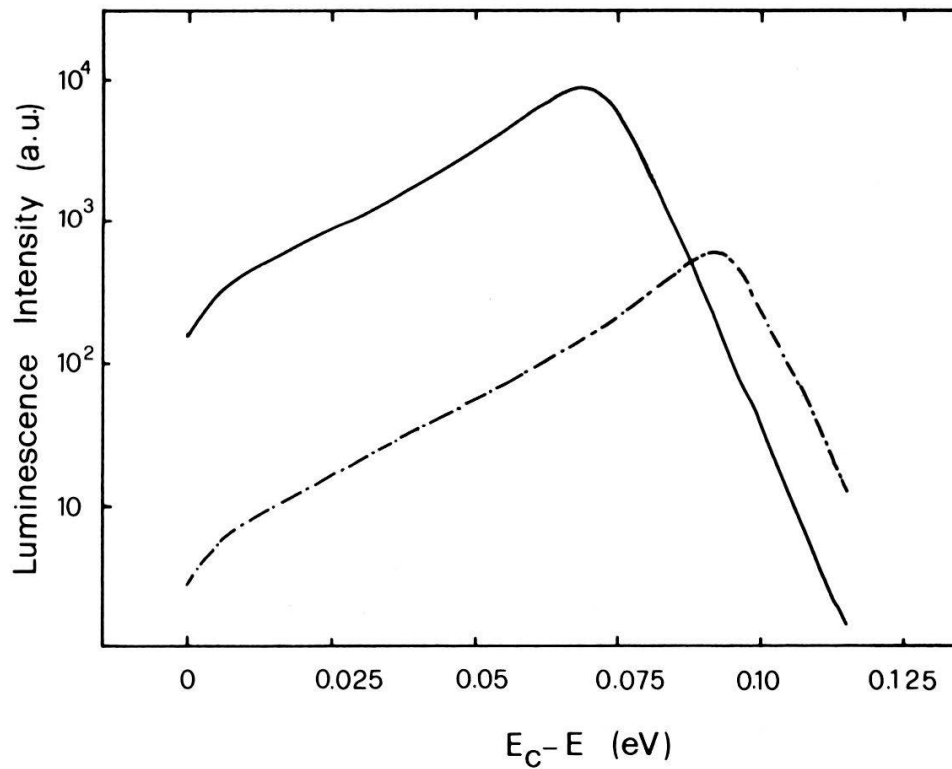


Figure 4
 Photoluminescence intensity as a function of energy for —: $t_1 = 0.1 \mu s$, — · —: $t_2 = 10 \mu s$.

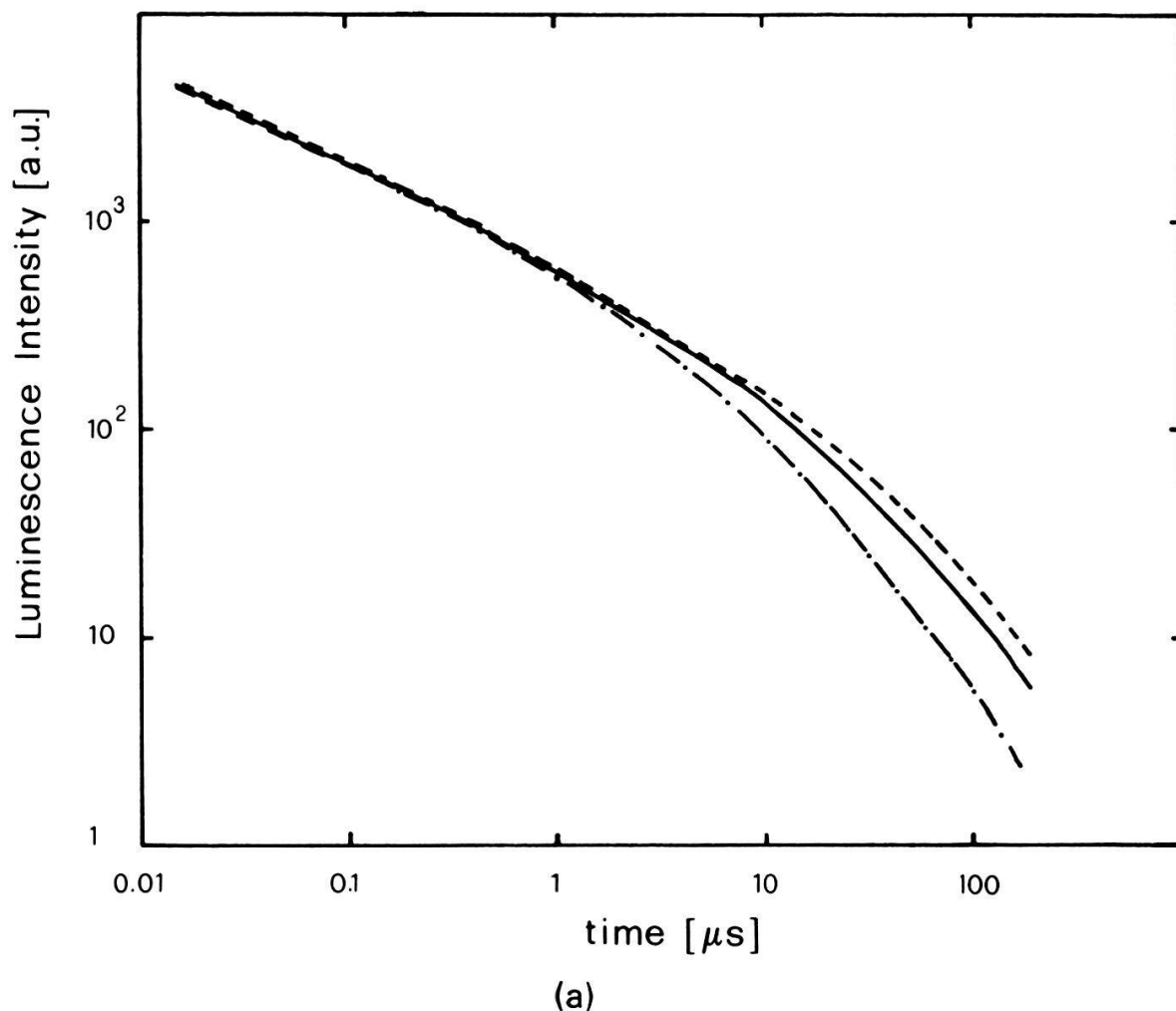


Figure 5

Total photoluminescence intensity vs. time for different temperatures.

(a) theoretical (b) experimental.

---: $kT = 3$ meV	● $kT = 3$ meV
—: $kT = 6$ meV	▲ $kT = 6$ meV
- · -: $kT = 9$ meV	■ $kT = 9$ meV

is shown for 77 K in Fig. 2. It behaves like $t^{-\beta}$ over several time decades. The comparison with the experimental curve shows that the agreement is excellent for $t > 0.1 \mu\text{s}$. The discrepancy observed at shorter times is probably due to the choice of our theoretical initial conditions.

The calculated partial contributions to the luminescence n_i/τ_i arising from the levels in the intervals i in the energy range of 120 meV are presented in Fig. 3. We note that the time dependence of the partial contributions of levels close to the mobility edge and of the total PL intensity are very similar. This is in agreement with our energy-resolved PL experiments. The maximum seen in the contributions from the deeper intervals corresponds to the filling of the levels, which are empty at $t = 0$ according to our initial conditions.

We emphasize that the power law decay for the total PL spectrum in Fig. 2 is a direct consequence of the strongly energy-dependent life times $\tau(E)$ which

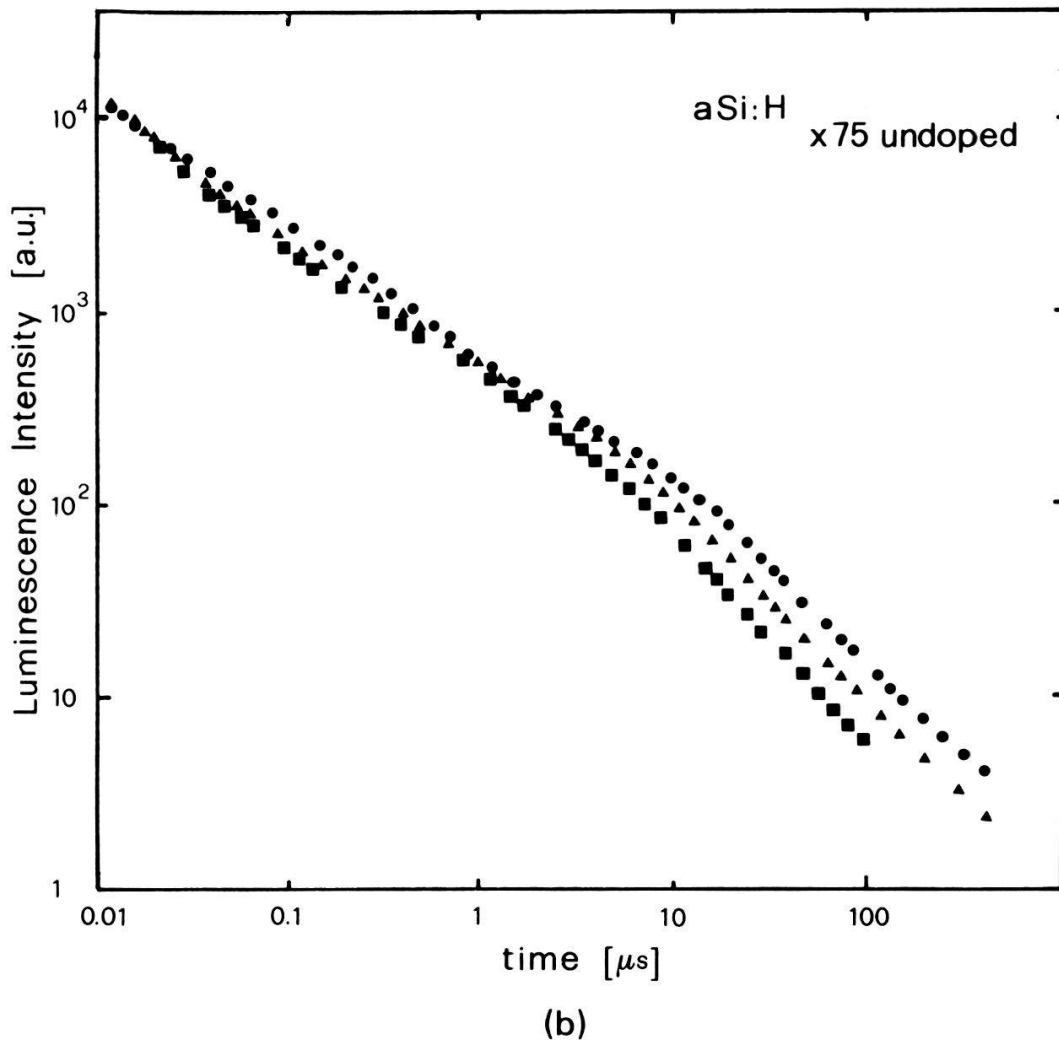


Figure 5 (Continued)

result from equation (4)

$$\begin{aligned} \frac{1}{\tau(E)} &= \frac{\tilde{b}_v}{\alpha(E)^{3/2}} \\ &= \tilde{b}_v A^{-3/2} \exp(-3E/2kT_1) \end{aligned}$$

In fact, inspection of the rate equations equation (5) shows that a constant τ would lead to an exponential decay of the total PL spectrum, since in this case

$$\frac{d\sum n_i}{dt} = -\frac{\sum n_i}{\tau}$$

Experimentally one observes a red shift of the PL spectrum with time, which is about 50 meV between $t_1 = 0.1 \mu s$ and $t_2 = 10 \mu s$. Figure 4 shows the calculated PL spectra as a function of energy for t_1 and t_2 . The relative shift is 25 meV, i.e. too small by a factor two. This result is not too bad if one considers the fact that the actual value of the shift depends strongly on the choice of t_1 , theoretically as well as experimentally.

The theoretical and experimental temperature dependences of the total PL decay are shown in Figs. 5a and 5b, respectively. It is seen that the experimental behaviour is well explained by the theory. We note that a conventional MTM cannot explain the fact that up to 3 μs the decay curves do *not* change with the temperature.

V. Conclusion

We have demonstrated that time- and energy-resolved PL data for *a*-Si:H may be successfully interpreted in terms of an extended multiple-trapping model which in particular allows for transitions between the localized tail states. Our results indicate that the localization length of the tail states decreases rather strongly with the distance in energy from the mobility edge. To our knowledge, this is the first time that it is shown that such an energy dependence – which is suggested in different theoretical approaches to amorphous systems – can lead to an explanation of significant experimental features.

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