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PHOTOLUMINESCENCE QUENCHING BY ELECTRIC FIELDS IN HYDROGENATED AMORPHOUS SILICON

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Abstract

We have measured the decrease of photoluminescence intensity at low temperature as a function of external fields (up to $3x10^5$ V/cm) in hydrogenated amorphous silicon (a-Si:H). The results are discussed within the framework of recombination of geminate pairs together with basic physical phenomena like carrier separation during thermalization, trapping of photoexcited carriers in band tails and possible subsequent reemission or tunneling out of traps.

Introduction

Measurements of steady-state photoluminescence (PL) give an overall quality criterium for a-Si:H. It is well known that the PL intensitity drops rapidly as the defect concentration exceeds 10^{17} cm^{-3}. There are still many open questions in the interpretation of more subtle effects like time-resolved PL and thermal and electric field quenching. Reported field quenching data have been interpreted in terms of Poole-Frenkel effect /1/ and exciton recombination, i.e. thermal emission of carriers over the Coulomb barrier, which is lowered by external fields. But also the possibility of carrier separation of geminate pairs during thermalization in extended states was taken into account /2/. To date a unique theory is not yet developped.

Recently the anticorrelation between field dependent photoconductivity (PC) and PL for fields up to $6x10^4$ V/cm has been discussed by Jahn *et al.*. This anticorrelation is based on the idea that the quantum efficiency of radiative plus non-radiative recombination plus PC must be equal to one /3/. Mainly from the temperature dependence it is concluded that Poole-Frenkel effect is not important.

Our measurements were done on samples with sandwich geometry, thereby fields up to $3x10^5$ V/cm could be obtained. For the interpretation we will consider (1) carrier separation during thermalization, (2) reemission and (3) tunneling out of traps. From the magnitude of field quenching we suggest that the average electron mobility during thermalization can be estimated.

Photoluminescence set-up and results

For the PL excitation the green line (514.5 nm) of an Ar ion laser was used. A bundle of optical fibres connected the laser source, the sample in helium gas and the monochromator. We used a liquid N_2 cooled Ge detector. For more detail see ref. /4/. The sample structure was Cr/n^+/i-a-Si:H/Au. The light was passed through the semitransparent Au contact. During the PL measurements we monitored the I-V characteristic to ensure that the sample was not heated due to photocurrents.

Fig. 1 shows three PL spectra taken at 50 K as a function of external field F. Fig. 2 gives the field dependence of the spectrally integrated photoluminescence for a 0.17 μm thick sample. Results from ref. /1/ are also shown.

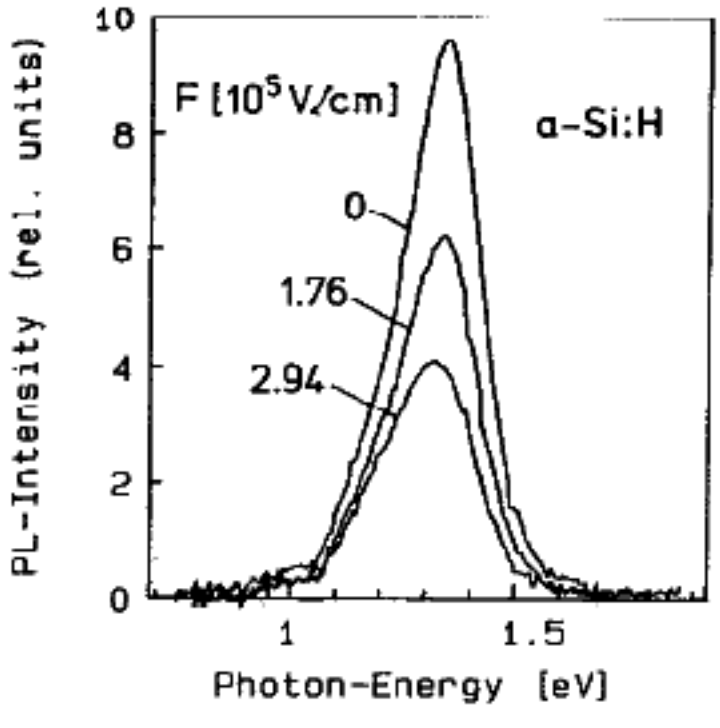

Fig. 1: PL spectra at 50 K of a-Si:H at the given external fields F.

Possible physical phenomena underlying PL field quenching

a) Carrier separation during thermalization

Radiative recombination is strongly dependent on the distance r of electron-hole separation. In the geminate pair picture the overlap factor of localized wave functions is given by $\exp(-2r/r_0)$, where r_0 denotes the spatial extent of the wave function of the less localized carrier, i.e. electron in the case of a-Si:H. Values deduced are in the range of 10 Å/5/. Considering drift we can assume a simple relation between the distance s of separated carriers and the external field F, $s = \mu_0 t F$, where μ_0 is the extended states mobility and t is the thermalization time in extended states. If one neglects non-radiative processes at low temperatures, the luminescence intensity I_{PL} is proportional to $\exp(-2s/r_0)$,

$$I_{PL} \sim \exp(-\gamma F), \tag{1}$$

with

$$\gamma = \frac{2\mu_0 t}{r_0}. \tag{2}$$

The decay of PL intensity shown in Fig. 1 is clearly exponential, with $\gamma = 3.2\text{x}10^{-6}$ cm/V. With the value of $t=10^{-12}$s, taken from the literature /6/, an extended states electron mobility of 0.16 cm^2/Vs is deduced. (Slightly higher values for t up to 10 ps are obtained for relaxation into flat tail states /7/). We assume at the same time that the hole mobility is much smaller than the electron mobility. This value is roughly two orders of magnitude lower than those obtained by extrapolation to high temperatures in TOF measurements /8/. However, one should consider that thermalization occurs very rapidly above the mobility edges of valence and conduction bands and is slowed down when the band tails are reached. Then the measured mobility has to be interpreted not purely due to transport in high-mobility extended states but also involving transport in low-mobility flat tail states.

Another possible explanation why μ_0 is smaller than the usually quoted free carrier mobility is the observation that due to inhomogenities during the initial film groth the average quality of a 0.17 μm film is poorer than the one of thicker films. This was found through thickness dependent PDS and PC measurements /9/.

In the following we will denote our measured mobility the average mobility $\overline{\mu_0}$ during thermalization. We suppose that $\overline{\mu_0}$ should show a slight temperature dependence, according to temperature dependent PL quenching measurements at low fields done by Jahn *et al.* /3/.

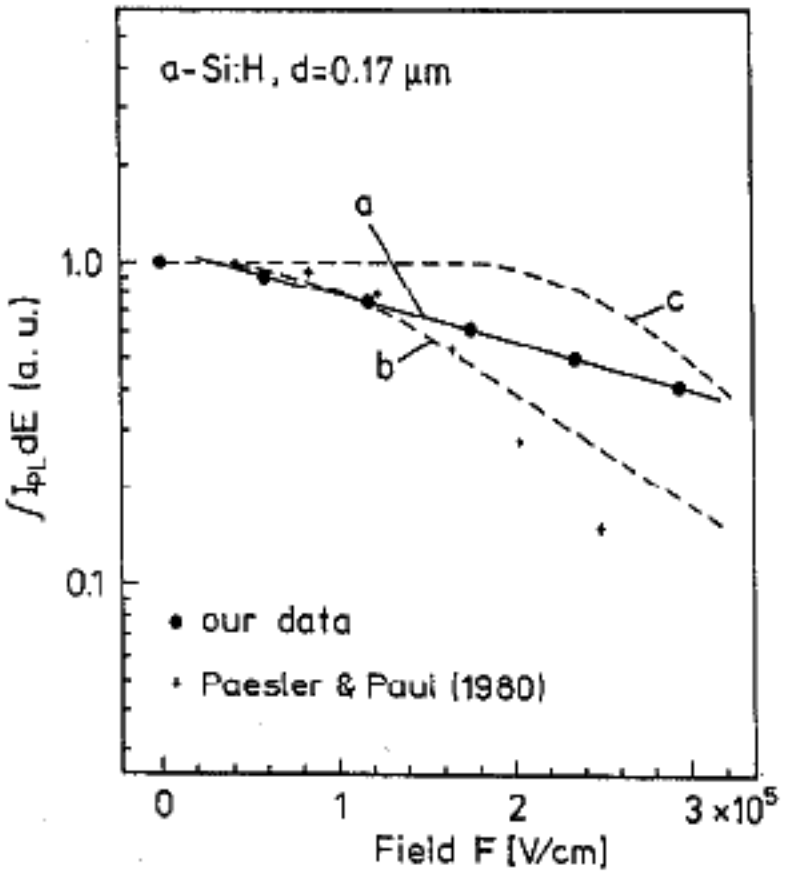

Fig. 2: Field dependence of the spectrally integrated photoluminescence intensity (a). Results from ref. /1/ are also shown. Curves (b) and (c) denote calculations to Poole-Frenkel effect (eq. (4)) and tunneling out of traps (eq. (6)).

b) Poole-Frenkel effect

The Poole-Frenkel effect describes thermal emission of carriers over the Coulomb barrier, which builds up in the presence of an external field. The height of the barrier E_{max} is determined by the field strength F,

$$E_{max} = \sqrt{\frac{e^3 F}{\pi \varepsilon \varepsilon_0}}. \tag{3}$$

The reemission rate τ_e^{-1} for trapped carriers into extended states is given by

$$\tau_e^{-1} = \nu_0 \exp(-\frac{E - E_{max}}{kT}), \tag{4}$$

where $\nu_0 = 10^{12}s^{-1}$ and the energy E of the carriers is taken to be positive in mid-gap direction. Depending on the energetic distribution of carriers in the tail states after exposure to light, carriers at different energy levels will have different emission probabilities. We calculated the carrier densities using a model due to Dunstan and Boulitrop /10/. Assuming that reemission is the only non-radiative process, i.e. $\tau_{nr}^{-1} = \tau_e^{-1}$, modified electron and hole densities were calculated, using the rate equation between radiative and non-radiative recombination /4/

$$n_r(E,F) = \frac{\tau_r^{-1}}{\tau_r^{-1} + \tau_{nr}^{-1}(E,F)} n(E,0). \tag{5}$$

n_r denotes the fraction of electrons contributing to radiative recombination, $\tau_r = 10^{-8}$s the radiative lifetime at

the maximum of the lifetime distribution /11/. The luminescence intensitiy is then calculated by convolution of electron and hole distributions. The result is shown in Fig. 2 (curve b) for T=50 K.

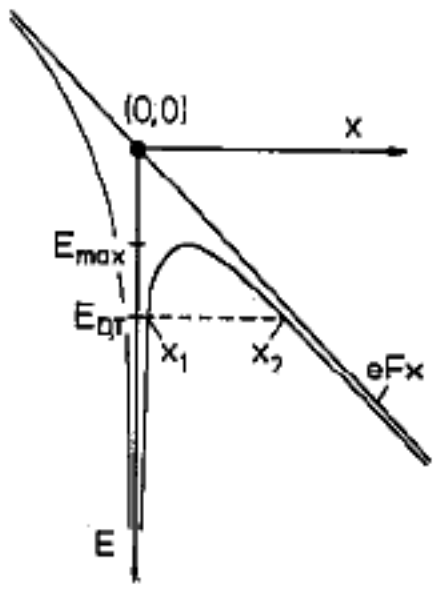

Fig. 3: One dimensional sketch of the Coulomb potential in the presence of an external field.

c) Tunneling out of traps

Besides from being thermally emitted over the barrier, trapped carriers have the possibility to tunnel through the barrier. In analogy to eq. (5) one can calculate the fraction of electrons contributing to radiative recombination, now with the tunneling probability τ_t^{-1} as the non-radiative rate

$$\tau_{nr}^{-1} = \nu_0 \exp\left(-2\int_{x_1}^{x_2} dx \sqrt{\frac{2m}{\hbar^2}\left(E - \left(\frac{e^2}{4\pi\epsilon\epsilon_0 x} + eFx\right)\right)}\right), \tag{6}$$

where m is the free electron mass and ϵ=12. The calculation of the field dependent luminescence intensity is analogous to the case of Poole-Frenkel effect. The result is shown in Fig. 2 (curve c). In the measured field range the quenching predicted by the tunneling model is too weak. This may partly be due to oversimplifications in the Dunstan model. Furthermore, not all of the carriers that have tunneled through the barrier may be lost for radiative recombination. This will strongly depend on the estimate of the effective Bohr radius r_0.

The above two processes can be illustrated by defining demarcation energies $E_{D,PF}$ and $E_{D,T}$, at which the radiative rate equals the non-radiative one, i.e. $\tau_r^{-1} = \tau_t^{-1}$ and $\tau_r^{-1} = \tau_t^{-1}$, respectively (see also Fig. 3). The field dependence of these energies is shown in Fig. 4 together with the electron distribution derived from the Dunstan model.

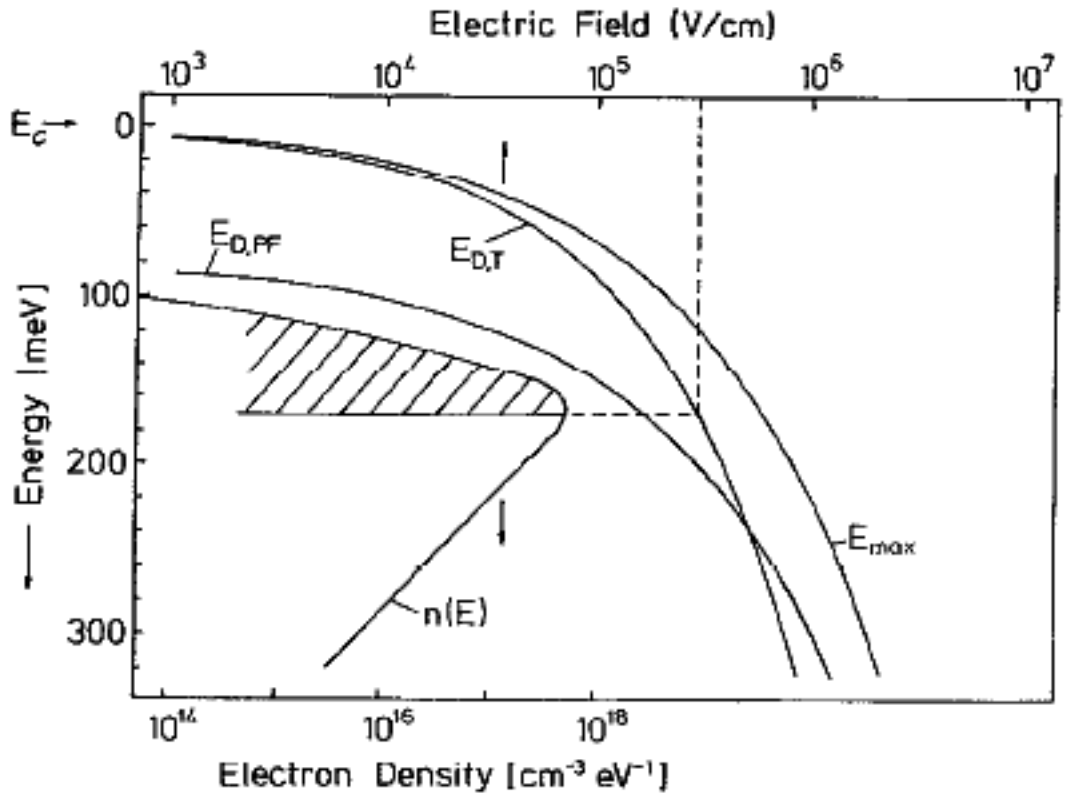

Fig. 4: Field dependence of the demarcation energies of the Poole-Frenkel effect ($E_{D,PF}$) and tunneling out of traps ($E_{D,T}$). Also shown is the electron distribution after exposure to light calculated by the Dunstan model. The shaded area schematically illustrates the portion of electrons which are lost for radiative recombination at a given field strength due to thermal emission or tunneling, respectively.

Discussion of limitations

a) Schottky barrier

Inhomogeneous field distributions inside the film may complicate the above considerations. For instance, the x-axis in Fig. 2 is based on the assumption that the internal field is linear with the applied voltage. Schottky barriers have a high field region near the surface. So at low applied voltage the simple linear relation between electric field and voltage is not correct. We have used a very thin sample in order to ensure that at large voltages this effect is no longer important.

b) Sample heating

We have observed high currents with some contacts. We suppose that due to the large voltages that were applied those contacts were partially shorted and the film below was heated. The observed PL quenching was then similar to the measurements published in ref. /1/ (see also Fig. 2). If the observed PL quenching in those cases were interpreted as being due to field effects only, then much larger values for $\overline{\mu_0}$ would be obtained. By comparison of the dissipated power in "good" and "bad" contacts and from separate temperature quenching data we have deduced that for the measurement shown in Fig. 2 the temperature effect is indeed negligible (below 5% for the highest field value).

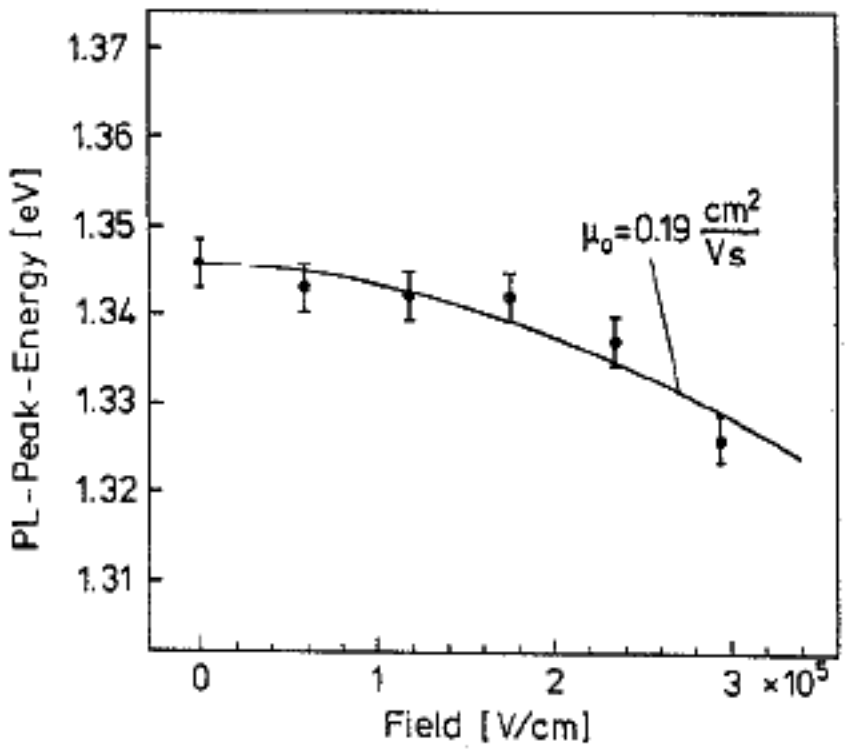

Fig. 5: Shift of PL peak energy with external field F.

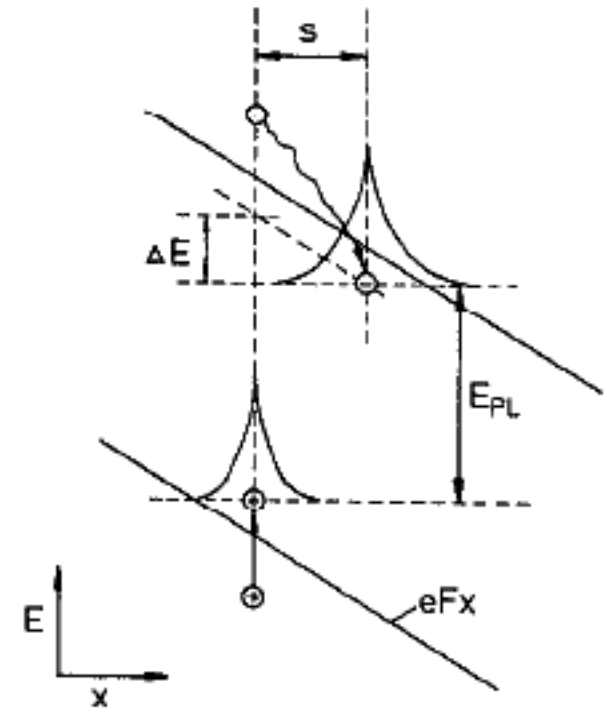

Fig. 6: Schematic illustration of the wave functions of a localized electron and hole trapped in band tail states after photoexcitation with an external field F.

Shift of PL peak with electric field

Support for the importance of process (1) is drawn from the shift of the peak position in the PL spectrum as a function of external field.

Based on the idea of carrier separation shown in Fig. 6 the shift of the PL peak energy ΔE is given by

$$\Delta E = e\overline{\mu_0}tF^2. \qquad (7)$$

The fit gives $\overline{\mu_0}$=0.19 cm^2/Vs. This is in good agreement with the result found from the PL intensity dependence under the assumption that carrier separation is the dominant process (1).

Summary

We conclude that separation of carriers during relaxation in extended and flat tail states is the dominant process for the field quenching of PL intensity. Other processes like Poole-Frenkel emission and tunneling out of traps cannot totally be ruled out. However, as we have shown through calculations based on the model by Dunstan and Boulitrop, the measured PL quenching cannot satisfactorily be accounted for in the field range up to 3x10^5 V/cm by these two processes. The basic idea of carrier separation is supported by consistent values for $\overline{\mu_0}$ from both the quenching of PL intensity with electric fields and the concomitant energy shift of the maximum in the PL spectra.

Acknowledgements

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