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RECOMBINATION OF PHOTOGENERATED CARRIERS IN DOPED HYDROGENATED AMORPHOUS SILICON

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Abstract. - Bulk recombination of photogenerated carriers is studied by transient photocurrent measurements in phosphorus doped a-Si:H. With light doping, the monomolecular recombination lifetime exceeds 10 ms and only a slow bimolecular recombination from extended states is observed. For very high initial carrier densities, a fast decay is suggested to be direct recombination from shallow band tail states.

Introduction. - The relaxation and recombination mechanisms of excess carriers in amorphous semiconductors have been investigated by a number of authors (1-7) relating to different experiments such as photocurrent (1,3,7), photoluminescence (2,6) and photoinduced absorption (3,4). In a recent Letter (7), hereafter referred to as HB1, we have shown that transient photocurrent (TPC) measurements with a short-pulse excitation can yield valuable information about these mechanisms and that, under certain circumstances, a bulk recombination lifetime can be extracted from TPC decays, even in the situation of strongly dispersive transport (8).

The present work addresses further the interpretation of the TPC decay under similar conditions. In particular, we investigate the fast initial decay and the form of the final recombination decay, i.e. those parts of the decay that are not easily described by a time-dependent drift mobility reflecting multiple trapping in a distribution of band tail states (7,8).

Experimental Details. - The experiments were performed on phosphorus doped hydrogenated amorphous silicon (a-Si:H) films supplied with Mo contacts in the form of stripes 1 mm apart (see figure 1). The films were prepared by glow discharge as previously described (7) but with a substrate temperature T_s=300°C during deposition.

Fig. 1. Sample geometry and focusing arrangement.
The excitation source was a N_2-laser pumped dye laser with pulse energy, pulse duration and repetition rate of 5 μJ, 4 ns and 20 Hz, respectively. The laser light was focused on the sample by a cylindrical lens as shown in figure 1. Thereby, we could illuminate homogeneously the region between the contacts to obtain maximum signal for a given excitation density. On the other hand, the light could be focused further to a line, less than 50 μm wide, to increase the excitation density keeping the total excitation power $P_{exc}$ constant. $P_{exc}$ was changed by inserting neutral density filters. The a-Si:H films were about 1 μm thick and they were excited in their full thickness by the laser light of photon energy=1.9 eV. Of the incident light about 23% was transmitted (as measured) and about 28% was reflected. Hence, only about 50% was absorbed in the film.

The TPC was investigated in a low drift field ($<10^3$ V/cm$^{-1}$) parallel to the film. For times $t<$500 μs, the decay was recorded via a sampling oscilloscope yielding a system time resolution of about 20 ns. For longer times (up to 100 ms), we used a PAR waveform eductor with a modified time resolution of 100 μs. In both cases, logarithmic converters were used to obtain directly a log-log plot of the TPC decay (7,8).

Experimental Results. - Our previous results showed that phosphorus doping strongly increased the effective recombination lifetime of photoexcited carriers in a-Si:H (7). Further investigations have revealed that this effect is even more pronounced when the films are prepared at a slightly higher substrate temperature $T_s$. This is illustrated in figure 2. The solid curves are results from the present work; TPC decays for different levels of phosphorus doping in samples prepared at $T_s=300^\circ$C. The dashed curves are from HB1 with $T_s=250^\circ$C.

It is obvious from figure 2 that for a given doping level, as specified by the amount (in volume percent) of PH_3 added to SiH_4 during deposition, the effective recombination lifetime is increased by about an order of magnitude in the present set of samples as compared to the set in HB1. It is not clear from the present experiments whether this is caused by a more efficient doping at the higher substrate temperature or by structural differences in the a-Si:H network.

Another difference between the present and the previous TPC decays is in the actual form of the decay. The fast initial decay observed in HB1 is absent in the present TPC decays as observed in figure 2. Furthermore, the dispersive transport, caused by multiple trapping in band tail states (7,8), does not show up as a simple power law decay over several orders of magnitude in time as in HB1, but rather displays a gradual change in slope of the log-log plot. This indicates that instead of the initial decay, a slow bimolecular recombination competes with the relaxation through band tail states which is conveniently described by a time dependent drift mobility.

![Fig. 2. TPC decays from the present experiment (solid curves) compared with previous results (dashed curves) from HB1.](attachment:image.png)
For the lowest doping level (X=0.001%), the final decay in figure 2 (solid curve) is faster than \( t^{-1} \) due to a final monomolecular recombination as observed in HB1. For higher doping levels this situation is never reached indicating that in these films, the monomolecular recombination lifetime exceeds 10 ms.

Due to the focusing arrangement with the cylindrical lens, the present TPC decays of figure 2 correspond to a lower initial carrier density than in HB1 although the total number of photoexcited carriers is about the same in both cases. The results of figure 2 therefore suggest a connection between the initial carrier density and the form of the initial TPC decay. Such a connection is further established in figure 3.

The solid curves in figure 3 are TPC decays in lightly doped (0.01% PH\(_3\)) a-Si:H with unfocused laser excitation as in figure 2 - the incident light illuminating the entire region between the contacts (1x12 mm\(^2\)). The dashed curves are TPC decays with focused laser excitation, implying that the same excitation power \( P_{\text{exc}} \) is focused to a narrow line, increasing the initial carrier density by a factor >20. With maximum excitation power \( (P_{\text{exc}}=1) \), unfocused and focused excitation yield an initial carrier density of \( \sim 5 \times 10^{17} \) cm\(^{-3}\) and \( >10^{19} \) cm\(^{-3}\), respectively. TPC decays are also shown with \( P_{\text{exc}} \) reduced by one and two order of magnitude.

We observe that when the initial carrier density is low \((<10^{18} \) cm\(^{-3}\)) the TPC decay is governed by dispersive transport with a time dependent drift mobility combined with a slow bimolecular recombination as indicated by the final decay approaching a \( t^{-1} \) behaviour (4). When the initial carrier density, on the other hand, is high \((>10^{18} \) cm\(^{-3}\)) this slow decay is preceded by a fast decay with a characteristic decay time \( \tau \approx 5 \mu s \).

Recombination Model. - The observed TPC decays can be qualitatively understood by assuming two different recombination channels both of which are essentially band to band transitions, i.e. bimolecular in nature. It is assumed that the photoexcited holes are immediately trapped and immobilized in localized valence band tail states (1). Therefore the main recombination channel for photoexcited electrons of low density is via extended states with a recombination rate \( R_e \) given by

\[
R_e = b_e(t) \cdot n^2
\]

where \( n \) is the total density of photoexcited carriers (in the present experiment \( n \approx n_e \), the equilibrium density). The time dependence of the recombination coefficient \( b_e(t) \) in equation (1) accounts for the fact that the (small) fraction of electrons, that are in extended states, diminishes with time (4).
A competing recombination channel, which we suggest to be important at very high carrier densities, is via localized electron states below the mobility edge. The recombination rate $R_L$ from these states will depend on the degree of localization which again depends on the energy $\epsilon$ which separates the localized state from the mobility edge at $E_C$. Hence, we may write:

$$R_L(\epsilon) = b_L(\epsilon) \cdot n_L(\epsilon) \cdot n$$  \hspace{1cm} (2)

where $n_L(\epsilon)$ is the density of electrons with energy $E=E_C-\epsilon$. One may expect shallow localized states to extend over several atomic distances. The recombination in equation (2) will dominate if the probability of finding a trapped hole within a recombination radius $r_o$ of the localized electron is substantial, or equivalently:

$$\frac{4\pi}{3} r_o^3 \cdot n \geq 1$$  \hspace{1cm} (3)

Discussion and Conclusions. - We suggest equation (3) to be met initially with intensely focused laser excitation, and therefore recombination from shallow localized states (equation (2)) to be responsible for the initial decay observed in figures 2 and 3 (dashed curves).

In our experiment $n=10^{18}$ cm$^{-3}$ seems to be a limiting case for observing an initial decay which from equation (3) requires $r_o \geq 60$ Å. Only very shallow states can be expected to have such a large recombination radius $(4,6)$. On the other hand, recombination from shallow localized states may explain why only traps beyond a certain trap depth $\epsilon_t$ seem to be effective in the trap controlled dispersive transport observed on a longer time scale $(7)$. The band tail states with $0<\epsilon<\epsilon_t$ are sufficiently delocalized to act as recombination centres (at high initial carrier density) rather than as shallow traps with fast thermal release to conducting states.

In HB1 we observed $\epsilon_t=0.1$ eV with an initial carrier density $n=2 \times 10^{18}$ cm$^{-3}$. For the lower initial carrier densities in the present work (figures 2 and 3, solid curves) only more shallow states should be preempted by recombination. One would therefore expect a lower activation energy for the initial drift mobility in this case and indeed a much weaker temperature dependence is observed when the initial carrier density is kept low ($<10^{18}$ cm$^{-3}$).

In the low density case, we also expect the initial photocurrent $I_o$ to be proportional to the total number of incident photons: $I_o=K \eta P_{exc}$, where $K$ is a constant and the quantum efficiency $\eta$ includes any initial geminate recombination of photoexcited carriers. With unfocused laser excitation we do find an almost linear dependence with a quantum efficiency only slightly dependent on $P_{exc}$. However, with focused laser excitation the initial photocurrent is sublinear in excitation power which is consistent with a fast initial bimolecular recombination from band tail states.

In conclusion, monomolecular recombination in deep recombination centres seems to be almost eliminated by phosphorus doping of glow-discharge a-Si:H films deposited at a substrate temperature $T_s=300^\circ$ C. A slow bimolecular recombination from extended electron states dominates the final recombination of those photoexcited electrons that survive any fast initial ($t\leq0.1$ μs) recombination - geminate or non-geminate.

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References.