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PHOTOCONDUCTIVITY AND PHOTOLUMINESCENCE OF a-Si:H AT LOW TEMPERATURE

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We have studied the temperature dependence of the photoluminescence steady state and transient photoconductivity of a-Si:H down to 4 K. Below 50 K we observe photoconductivity with $\eta\mu\tau = 10^{-11}$ cm^2/V, which is independent of temperature and varies only little with the defect density in the films. We propose that this conduction arises predominantly from the drift of the photoexcited electrons and holes during thermalization in the extended states prior to the localization in states below the mobility edge. An important implication of the present data is that, even at helium temperatures, a considerable part of the carriers recombines in a non-geminate process.

1. Introduction

From quite a number of photoluminescence studies [1] a model for recombination in hydrogenated amorphous silicon has been developed: The photogenerated electrons and holes thermalize quickly down to the mobility edge in times of about 10^{-12} s. The subsequent thermalization within the localized states occurs by tunneling at a rate which decreases as the density of states in the band tail decreases. At low temperatures ($T < 50$ K) the quantum efficiency for radiative recombination of the localized electrons and holes is of the order of 1 in low defect material but appreciably smaller in less perfect material due to non-radiative recombination via tunneling to defects. It is still a matter of debate whether radiative recombination is predominantly geminate or whether the distant-pair model is more appropriate [2,3]. At higher temperatures, ionization of localized electron–hole pairs is believed to be responsible for the temperature dependent quenching mechanism of the photoluminescence. This process leads to mobile carriers which give rise to the photoconductivity. According to these ideas one would expect the photoconductivity to decrease strongly with decreasing temperature due to the decreasing probability of thermal pair ionization until, at low temperature, the photoconductivity vanishes if only geminate recombination occurs. There are many investigations of photoconductivity and photoluminescence above liquid nitrogen temperature which support this view [4] but no investigations down to helium tempera-

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ture, although particularly in this temperature range such studies could help to decide whether geminate – or distant pair recombination is dominant. In this paper we report on a combined investigation of photoluminescence (PL) and steady state as well as transient photoconductivity (PC) in a-Si : H in the temperature range 4–500 K.

2. Experimental

The present study was carried out on a series of a-Si : H films deposited by the glow discharge technique or by rf-sputtering onto fused quartz substrates. The preparation conditions are given in table 1. In the glow discharge depositions the pressure was between 0.3 and 0.5 mbar and the deposition rates were 1.5–3 Å/s.

The room temperature conductivity of these undoped and doped films varied from $10^{-12}\ \Omega^{-1}\ \mathrm{cm}^{-1}$ to $4\times10^{-3}\ \Omega^{-1}\ \mathrm{cm}^{-1}$. After the film deposition Cr-contacts were evaporated which provided fairly ohmic contacts for field strengths up to 2×10^{4} V/cm. In order to improve the geometry grid-like contacts were used which had a total length of 15 cm and an electrode spacing of 400 μm. In the transient experiments the light of a He–Ne-laser (25 mW) was cut off by a pockels cell. The decay of the photocurrent from the steady state value was recorded by means of a digital storage oscilloscope (Gould OS4040) and a fast preamplifier. The time resolution of this equipment was 1 μs. In order to determine the $\eta\mu\tau$-product, absorption measurements were carried out on all our samples. The luminescence measurements were performed in a conventional arrangement using a-Si : H films deposited in the same run onto ground substrates.

3. Experimental results and discussion

When the temperature is lowered the intensity of the photoluminescence increases and attains below 50 K, for samples of low defect density, a nearly constant value (fig. 1). The absolute quantum efficiency, η_{PL}, in this tempera-

Table 1

Sample no.	System	Gas mixture	T_s (°C)	Flow (sccm)	Doping
C1	capacitively	5% SiH_4 in He	280	80	0
C2	coupled g.d.	5% SiH_4 in He	280	80	33ppm PH_3
C3		100% SiH_4	280	3	0
C4		100% SiH_4	280	3	1000ppm B_2H_6
I1	inductively coupled g.d.	100% SiH_4	300	6	100ppm PH_3
S1	rf sputter	40% H_2 in Ar $p_{tot} = 2\times10^{-2}$ mbar	200		0

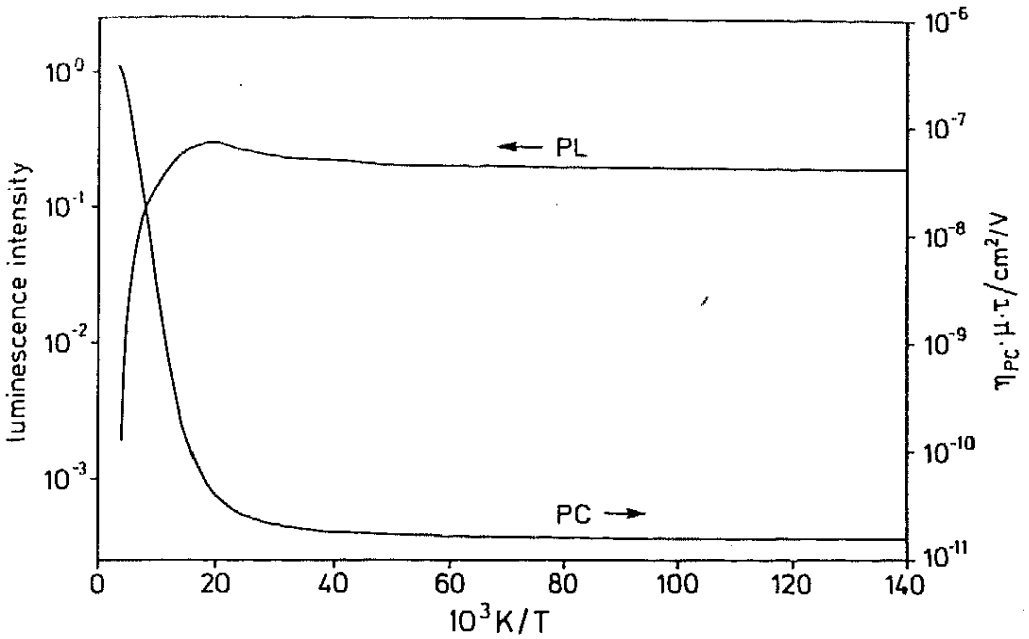

Fig. 1. Temperature dependence of the photoluminescence intensity (PL) and the $\eta_{PC}\mu\tau$-product (PC) of sample C3.

ture range amounts to about 30%. This value is in good agreement with other reports on films of low defect density [5]. In less perfect films, i.e. highly doped or sputtered a-Si : H, η_{PL} is considerably smaller. If the temperature dependent quenching mechanism of the photoluminescence is assigned to the ionization of electron–hole pairs, the photoconductivity is expected to decrease strongly with decreasing temperature. This behaviour, indeed, is found above 50 K (fig. 1). Below this temperature, however, the photoconductivity becomes independent of temperature, the $\eta_{PC}\mu\tau$-product amounting to 2×10^{-11} cm^2/V. It should be emphasized that this quantity in the case of a linear dependence of the photoconductivity on the light intensity has a simple meaning, it is connected with the schubweg of the photoexcited carriers. This result demonstrates that, even at the lowest temperatures, mobile electrons and/ or holes exist. Hence, only part of the excited carriers, if at all, recombine in a geminate process.

It is remarkable that the low temperature photoconductivity is observed with very similar $\eta_{PC}\mu\tau$-values in quite different samples. Fig. 2a shows results for undoped and doped glow discharge films as well as hydrogenated sputtered material. The different positions of the Fermi level and the different defect densities give rise to a wide spread of $\eta_{PC}\mu\tau$ and to quite different temperature dependences above 50 K. Below this temperature, however, the $\eta_{PC}\mu\tau$-product of these samples becomes independent of temperature and differs by not more than a factor of 4 although the defect densities in these films differ appreciably, as can be seen from the different quantum efficiencies of the photoluminescence: $\eta_{PL}(C4) < 3\%$, $\eta_{PL}(S1) < 0.5\%$. It is thus obvious that there is no close relation between photoconductivity and photoluminescence. In this temperature range the photoconductivity is only weakly influenced by the density

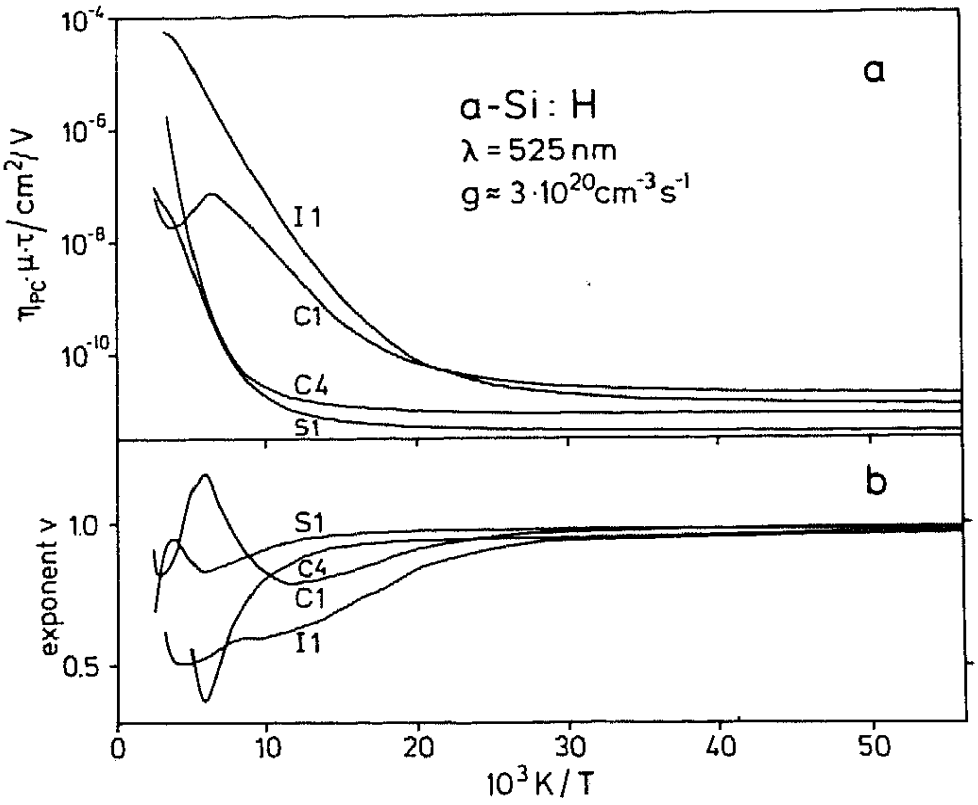

Fig. 2. Temperature dependence of the $\eta_{PC}\mu\tau$-product (a) and the exponent ν of the dependence of the photoconductivity on light intensity (b) of different a-Si : H samples.

of localized gap states. This conclusion is supported by the observation that light induced defects, which are known to lead to pronounced differences of $\sigma_{ph}(T)$ in the high temperature range [6,7], do not affect σ_{ph} below 50 K. After prolonged illumination with white light of 70 mW/cm^2 at room temperature, there is no observable difference between the photocurrents in the light soaked and annealed state of the film at temperatures below 50 K.

The dependence of the photoconductivity on the light intensity, in the whole temperature range investigated, obeys a power law: $\sigma_{ph} \sim g^{\nu}$. Above 50 K the exponent ν (fig. 2b) varies between 0.4 and 1.2 depending on temperature and the kind of sample. Below 50 K, in all films investigated, we found a linear dependence, i.e. $\nu = 1$, over more than 3 orders of magnitude in intensity. This contribution to the photocurrent varies linearly with the applied electric field and exhibits the same spectral dependence as the high temperature photocurrent.

It is usually assumed that the transport of the photoexcited carriers occurs in the extended states above the mobility edge E_c with a mobility of 1 to 10 cm^2/Vs and is dominated by trapping and thermal release from localized tail states [8]. It was proposed that, below 250 K, the transport path changes and that hopping in tail states predominates [8]. However, combined measurements of the steady state and transient photoconductivity [6] as a function of temperature have indicated, that near this temperature it is not the current path but the recombination mechanism which changes. Presumably, the initial

state for the recombination transitions changes from extended to localized tail states. The predominance of the latter process has recently been established by Dersch et al. [9] from the study of spin dependent photoconductivity. With decreasing temperature multiple trapping will become increasingly less important, since most of the tail states are converted to recombination centers. Once a carrier is trapped in a tail state the probability for reemission to the conducting states is much smaller than that for further thermalization in the tail states until recombination occurs, e.g. via a defect state. Therefore, the thermalization time rather than the recombination or trapping time determines the value of the photoconductivity. We propose that, immediately after the photogeneration, the carriers are in high mobility states and contribute to the conduction during their thermalization until they are localized in tail states. The "lifetime" of the carriers in extended states thus is determined by the localization i.e. capture in localized states near the mobility edge. Since the thermalization time is independent of the carrier density this leads to a linear dependence of the photocurrent on the light intensity. Furthermore, this characteristic time does not depend on the density of defects in the film but is an intrinsic property of the amorphous silicon network, being determined by the density of states above and near the mobility edge. Further indication for this type of transport in a-Si : H has recently been obtained by Johnson et al. [10] from picosecond transient photocurrents in the high temperature range. For a-As_2Se_3 similar conclusions were drawn from concomitant transient measurements of photoinduced absorption and photoconductivity [11].

One implication of this interpretation is, that the response time should be very fast in the low temperature range since trapping processes are unim-

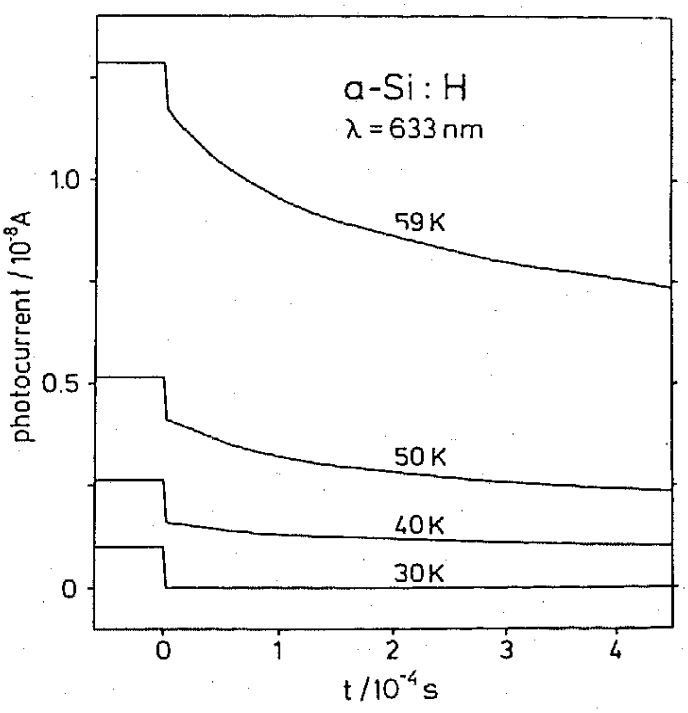

Fig. 3. Decay of the photocurrent of sample C2 after illumination for different temperatures. The initial fast drop amounts to 10^{-9}A independent of temperature.

portant. We, therefore, studied the decay of the photocurrent from the steady state as a function of temperature. According to fig. 3 the decay is clearly nonexponential. From the transient a response time can be defined as the time constant at the onset of the decay

$$\tau_R = -\left(\frac{1}{I}\frac{dI}{dt}\right)^{-1}_{t=0}.$$

Above 50 K this quantity varies between 10^{-4} and 10^{-2} s and depends only weakly on temperature [6,12]. The interesting result shown in fig. 3 is that, at low temperature, in addition to the slow non-exponential decay there is an initial fast decay. The time constant of this component, obviously, is much shorter than the time resolution of the experimental arrangement, namely 1 μs. For the slow component we find, as for high temperatures, $\tau_R \approx 4 \times 10^{-4}$s practically independent of temperature. This slow decay is attributed to the multiple trapping mechanism involving flat tail states [6,12]. With decreasing temperature this slow contribution decreases and finally vanishes below about 30 K. The fast contribution to the photocurrent, however, is independent of temperature and amounts to $\Delta I_{ph} = 10^{-9}$A in the present example, corresponding to an $\eta_{PC}\mu\tau$-product of 6×10^{-12} cm^2/V.

For the interpretation it is important to realize that, at intermediate temperatures, the fast decay is superimposed on the decay determined by multiple trapping and that, below 50 K, there is not detectable temperature dependence of the $\eta_{PC}\mu\tau$-product. These results support the conclusion that the photoresponse at very low temperature does not include thermal reemission from deeper states but is due to photoexcited carriers thermalizing above the mobility edge. An additional contribution to the low temperature photoconductivity in principle may also occur during the slower thermalization in the localized tail states. From time resolved measurements of the photoluminescence one expects that the thermalization is much enhanced with rising temperature [13]. We expect therefore that this process would lead to a temperature dependent photoconductivity which however is not observed. We therefore believe that predominantly fast thermalization near the mobility edge leads to photoconduction with $\eta_{PC}\mu\tau \approx 10^{-11}$ cm^2/V. Inserting an extended states mobility of 10 cm^2/Vs and $\tau_{th} = 10^{-12}$s for the thermalization time in the extended states [1], one obtains for the quantum efficiency for the generation of mobile carriers: $\eta_{PC} \approx 1$. Since only carriers contribute to the photoconduction which recombine in a non-geminate process, this rough estimate leads to the conclusion that even at low temperature non-geminate recombination plays a major role. This result seems to be in conflict with interpretations of photoluminescence studies [1]. However, recent measurements of electroluminescence in pin-diodes [2] as well as recent investigations of photoluminescence [3] have thrown some doubt on these earlier interpretations at least in the case of experiments with continuous excitation. If, on the other hand, one assumes $\eta_{PC} \ll 1$ one would need a correspondingly larger value for the mobility in the extended states in order to account for the observed $\eta_{PC}\mu\tau$-

product. At first sight this sounds rather unreasonable. However, large values for the mobility in excess of 100 cm^2/Vs recently have been deduced by Silver et al. [14] from junction recovery studies at pin-diodes.

4. Conclusion

We have observed a low temperature steady state photoconductivity with $\eta_{PC}\mu\tau \approx 10^{-11}$ cm^2/V which is completely independent of temperature and only slightly sensitive to the density of deep gap states. It is argued that this photoconduction occurs predominantly during the thermalization of the photo-excited electrons and holes in the extended states before the carriers are localized in states below the mobility edges. An implication of this interpretation is that even at low temperature non-geminate recombination is important. As a consequence, experiments at low temperatures ($T < 50$ K) allow the direct study of electronic transport near the mobility edge since there is no interaction with traps. Such investigations may yield information on the nature of the involved states and the mobility edge.

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