Journal of Non-Crystalline Solids 66 (1984) 151-156
North-Holland, Amsterdam

INFRARED QUENCHING OF PHOTOLUMINESCENCE AND PHOTOCONDUCTIVITY IN a-Si:H

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We have studied quenching and enhancement effects of photoconductivity (PC) and photoluminescence (PL) by IR-excitation from the non-equilibrium trap populations in glow discharge deposited a-Si:H films. In a dual beam arrangement ($h\nu_1$ = 1.92 eV, $h\nu_2$ < 0.7 eV) we have observed quenching of the PL in the temperature range 5 K < T < 100 K. It is suggested that this effect originates from photoionization of electron-hole pairs which have high energy and large separation. At low temperature after trap filling, IR-excitation is shown to effectively quench the residual LESR-signal and to create transients of PL and PC. It is suggested that these IR-induced effects originate from the same non-equilibrium carrier distributions in the band tails.

1. INTRODUCTION

The recombination of excess carriers in a-Si:H has intensively been studied in particular by photoluminescence and photoconductivity. It is widely accepted that at low temperature carriers are trapped in band tail states from where they either recombine radiatively by tunneling to carriers in the other band tail or non-radiatively by tunneling to defect states. The relevant defect centers have been shown to be Si-dangling bonds[1]. It is still a matter of debate, whether the PL-kinetics has to be described in a geminate or distant pair model[2]. At higher temperature detrapping and diffusion to defects enhances the non-radiative recombination rate. There are many experimental results which indicate that at low temperature non-equilibrium distributions of trapped electrons and holes excist in a-Si:H[2,3,4]. In this paper we report on the effect of IR-excitation from the trapped carrier distributions on the photoconductivity (PC), photoluminescence (PL) and light-induced ESR (LESR) of glow discharge deposited a-Si:H.

2. EXPERIMENTS AND DISCUSSION

In a dual beam arrangement the photoconductivity of undoped a-Si:H (n-type) of high quality is effectively quenched by IR-light[5,6,7]. This effect is explained by the enhancement of the recombination due to optical excitation of minority carriers from deep traps. In undoped a-Si:H the relevant hole traps

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* Supported by the Bundesministerium für Forschung und Technologie (BMFT)

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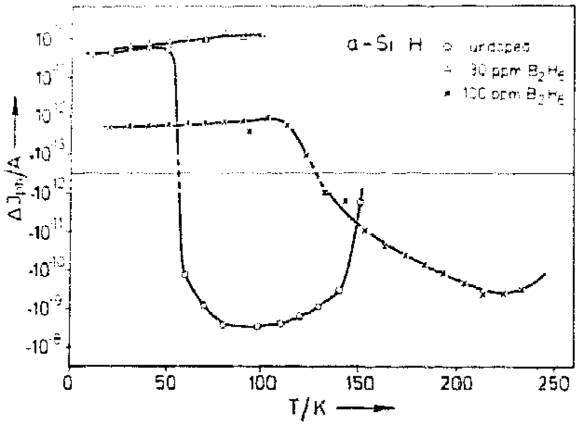

FIGURE 1
Change of the photocurrent, ΔI_{ph}, by additional IR-illumination as a function of temperature
($\Delta I_{ph} = I_{ex+IR} - I_{ex} - I_{IR}$, $\Delta I_{ph} < 0$ quenching, $\Delta I_{ph} > 0$ enhancement)

are band tail states and the recombination centers have been identified as dangling bonds[8]. A measure for the trap depth can be obtained from the low energy cut off in the spectral dependence which we found near 0.6 eV[7]. We observed optical quenching also in boron-doped samples, which clearly have p-type photoconduction, with a cut off energy also near 0.6 eV. This result is surprising because for band tail electrons one would expect a considerably lower trap depth. There are pronounced differences in the temperature dependence of the optical quenching effect of n- and p-type samples (Fig. 1). In the p-type film quenching ($\Delta I_{ph} < 0$) is observed above 125 K and ΔI_{ph} increases with rising temperature. In the undoped n-type film ΔI_{ph} decreases continuously above 100 K, when with rising temperature the diffusion of the trapped holes to defect centers is enhanced or bypassed by thermal activation and vanishes near 150 K. For the p-type sample thermal detrapping of the electron traps sets in at considerably higher temperature ($T > 220$ K) and optical quenching can still be observed at 300 K.

At low temperatures ($T < 50$ K) the PC-quenching vanishes and in all kinds of a-Si:H films the photocurrent is enhanced ($\Delta I_{ph} > 0$). In this temperature range the quasi Fermi levels are very near to the mobility edges. Then the photoconductivity is determined by the time for localization of excited carriers near the mobility edges, the $\eta\mu\tau$-product amounts to 10^{-11} cm^2/V irrespective of the defect density in the film and is independent of temperature[4]. The onset of PC-enhancement is closely related to the transition in the photoconduction from a multiple trapping dominated to a temperature independent behaviour as the temperature is lowered. Thus it is reasonable to assume that the enhancement effect results from electrons and holes which are excited by IR-light from the deep trapping levels to the conducting states and contribute to conduction until they are localized or recombine at defect states.

At low temperature optical excitation from the non-equilibrium distribution of trapped carriers results in addition in quenching of the photoluminescence. This is achieved in a dual beam arrangement when the sample is simultaneously excited by unmodulated beams of photon energies $h\nu_1 = 1.92$ eV and $h\nu_2 < 0.7$ eV. The IR-light was obtained by passing the light of a tungsten lamp through a

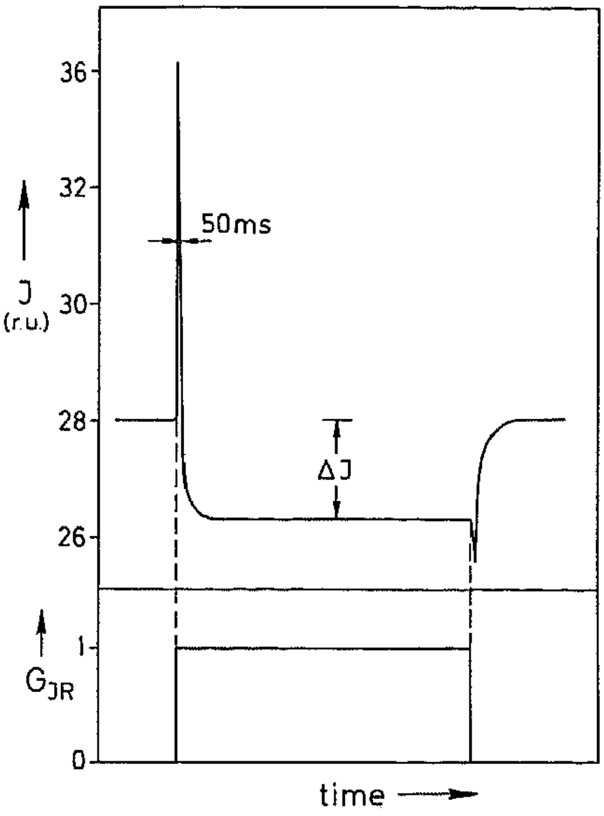

FIGURE 2
Transient PL-signal when the sample is exposed to additional IR-light of intensity G_{IR}

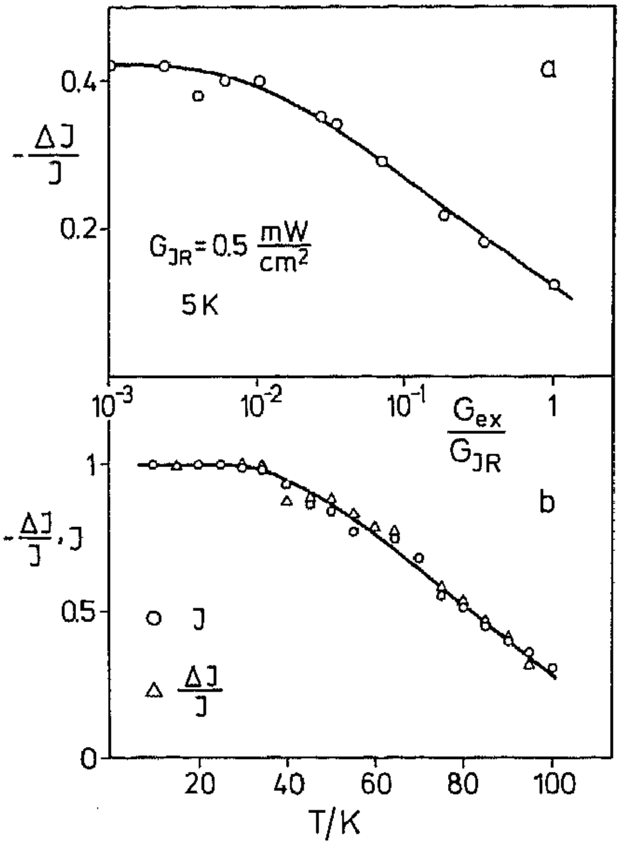

FIGURE 3
Dependence of $\Delta I/I$ on the excitation intensity G_{ex} (a). Temperature dependence of I and $\Delta I/I$, both normalized to their values at 15 K (b).

Ge-Filter. Fig. 2 displays a typical transient PL-signal. When the IR-light is turned on, enhancement of PL occurs which decays within some 10 ms. In the steady state the PL is quenched, $\Delta I < 0$. Finally, when the IR is turned off a transient decrease is observed before the PL-intensity attains its original value. Such behaviour is found in a large variety of doped and undoped gd-a-Si:H-samples. $\Delta I/I$ depends sensitively on the intensities of the two light beams. When the IR-intensity, G_{IR}, is kept constant, $\Delta I/I$ increases with decreasing intensity of the excitation beam, G_{ex}, and tends to saturate at low excitation levels (Fig. 3a). At high levels of G_{ex}, e.g. 0.5 mW/cm^2, $\Delta I/I$ is roughly proportional $G_{IR}^{1/2}$. At low excitation intensities this dependence is considerably weaker.

IR-quenching can be explained in the geminate pair model as well as in the distant pair model by optical ionization of electron-hole pairs which enhances the non-radiative recombination rate, when the carriers diffuse to defects. This process thus is similar to that one which leads to thermal quenching of the PL-intensity. If one assumes that the rate of photoinduced pair ionization is proportional to G_{IR} and to the total density of electron-hole-pairs, n_p, the

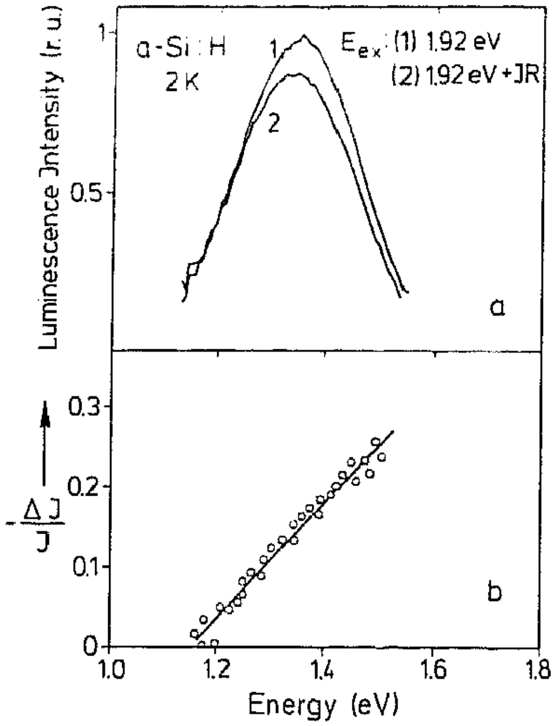

FIGURE 4
Influence of IR-illumination on the PL spectra (a) and spectral dependence of the quenching effect ΔI/I (b). $G_{ex} \approx G_{IR}$ = 0.5 mW/cm^2

PL-intensity I and ΔI/I are given by

$$I = \frac{P_R\, G_{ex}}{P_R + P_{NR}} \qquad \frac{\Delta I}{I} = -\frac{c\, G_{IR}}{P_R + P_{NR}} \qquad (1)$$

In these expressions P_R and P_{NR} denote the probabilities for radiative and non-radiative recombination. Since the temperature dependence of I is caused by that of the non-radiative path, $P_{NR}(T)$, relations (1) predict the same temperature dependence for both quantities. Fig. 3b shows that this is indeed the case. If I and ΔI/I are normalized to their values at 15 K, the points nicely fall on one line.

However, the influence of the IR-light on the spectral dependence of the PL suggests that the rate of optical pair ionization is not simply proportional to the total pair concentration. With IR-excitation the spectrum (Fig. 4) becomes unsymmetric and shifts slightly to lower energy. ΔI/I decreases with decreasing photon energy and there is no IR-quenching on the low energy side of the PL-band. Hence IR photons preferably quench the luminescence from high-energy-pairs. In addition, one should expect that optical excitation of pairs does not necessarily lead to ionization. If the excitation occurs well inside the Onsager radius, diffusion will not quench the radiative recombination but may even enhance it by transforming pairs of large separation to those of short separation. This may be the origin of the transient enhancement effect in Fig. 2. We thus conclude that the photo-induced quenching will be caused primarily by high-energy-pairs which have a large separation.

In a geminate pair model, where each electron recombines with its geminate hole, the distribution of the pair separations does not depend on the exciting light intensity G_{ex}. Therefore the fraction of pairs with high energy and large separation should be independent of G_{ex} in accordance with the behaviour in Fig. 3a, where ΔI/I attains a constant value at low excitation levels. At higher intensities this model breaks down when there is spatial overlap of the pairs. Then one expects that with increasing G_{ex} the fraction of pairs with large separation strongly decreases. This might be the origin of the de-

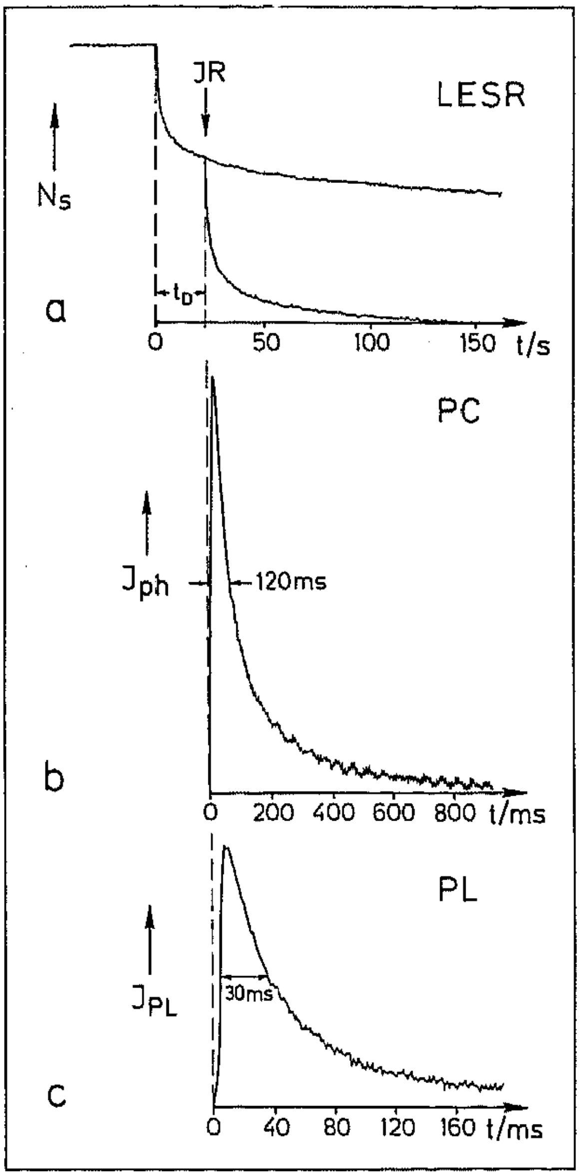

FIGURE 5
Transients of LESR (a), PC (b) and PL (c), when after a dark period t_D the films is exposed to IR-light. The same values of G_{IR} is used for (b) and (c), for (a) G_{IR} was much lower. T = 15 K.

creasing quenching effect with G_{ex}. Thus it seems that the geminate pair model qualitatively accounts for these observations. However, these preliminary results do not allow to decide between the conflicting models for recombination. More detailed information is needed, in particular on the influence of IR-illumination on the lifetime spectra and on the dependence of this effect on the IR-photon energy.

Recently Bhat et al.[9] also reported on photo-induced quenching of PL in a two-beam measurement by subbandgap excitation using $h\nu_2 > 1.55$ eV. The spectral dependence as well as the dependence on the light intensities are completely different from the results reported here indicating that the quenching mechanism is substantially different.

When the exciting light is turned off, the non-equilibrium distribution of trapped band tail carriers persists with long lifetimes at low temperature. The long time decay of the residual light-induced ESR signal (LESR) has been used to study the recombination of these metastable populations[3]. The non-equilibrium carriers can be excited by IR-light and detected in transients of PL and PC. Fig. 5 displays such transients which are generated, when the traps have been populated by bandgap excitation and when after a dark period t_D, IR-light ($h\nu < 0.7$ eV) is turned on. The IR-excitation effectively quenches the residual LESR-signal (Fig. 5a). Connected with the removal of the metastable trap population are the transients of PC and PL (Fig. 5b,c). The halfwidth of the PL-signal amounts to some 10 ms and thus is smaller by an order of magnitude than that of the PC-transient (note different time scales). This IR-induced PL-transient has some similarity with the transient enhancement effect in the two-beam experiments (Fig. 2) and can be explained in the same way by a redistribution of lifetimes due to IR-excitation. Obviously part of the excitation processes result in electron-hole pairs of shorter separation, when the carriers are retrapped, leading to radiative recombination. It is not possible to decide, whether these pairs are still geminate. If they were, they would not contribute to photoconduction. On the other hand, the PC-transient can originate only from those electrons and/or holes, which finally recombine radiatively or non-radiatively following distant pair kinetics. It is difficult to explain the different halfwidths of the transients. The halfwidth of the PL-signal is determined by the time dependent generation rate of radiative pairs and their resulting lifetime distribution. Similarly, the form of the PC-transient is expected to be mainly given by the excitation rate from the non-equilibrium distribution. At such low temperatures, namely, thermal detrapping effects have been shown to be unimportant and the photoconduction is determined by the time for localization near the mobility edges, which is extremely short[4]. Hence the larger halfwidth of the PC-signal does not arise from the transport but from the generation process. Possibly retrapping and reexcitation of carriers can lead to this behaviour.

The observation that IR-excitation quenches the LESR-signal and thereby generates transients of PL and PC, strongly indicates, that the IR-induced effects, reported here, originate from the same non-equilibrium carrier distributions in the band tails. A detailed study of these transients as a function of temperature and t_D, is being carried out. This may lead to valuable information about recombination of these excess carriers in a-Si:H films.

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