PHYSICAL REVIEW B VOLUME 46, NUMBER 19 15 NOVEMBER 1992-I

Decay of photocurrent from the steady state in *a*-Si:H films

J-H. Zhou and S. R. Elliott
Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge, CB2 1EW, United Kingdom

(Received 23 December 1991; revised manuscript received 11 June 1992)

Results are reported on the long-time decay of the photocurrent from the steady state in undoped *a*-Si:H films. The decay has been studied for various illumination intensities and over a wide temperature range, from 300 down to 50 K. It is found that the long-time photocurrent decay is a strong function of the steady-state illumination intensity when the intensity is low. As the light intensity is increased, the decay gradually approaches a power-law behavior $I_{\text{ph}} \sim t^{-\beta}$ and eventually becomes intensity independent. β is found to vary nonmonotonically with temperature; a peak appears between ~ 110 and ~ 150 K, depending on the history of the sample as well as on the sample itself. Some possible photocurrent decay models are examined.

I. INTRODUCTION

Transient photocurrent measurements with pulsed excitation have been widely used to study the states in the band gap as well as the properties of carrier transport and recombination in amorphous semiconductors.[1–7] A sandwich configuration is used in time-of-flight (TOF) experiments and a coplanar configuration in photoconductivity decay (PCD) measurements. Usually a power-law decay behavior is observed in both TOF and PCD experiments,

$$I_{\text{ph}} \sim t^{-\beta} . \tag{1}$$

The decay exponent β lies between zero and unity and decreases monotonically with increasing temperature. The power-law photocurrent decay observed in a TOF experiment is commonly interpreted as being due to multiple trapping (MT) of carriers in exponentially distributed band-tail states.[8,9] However, the interpretation of the power-law photocurrent decay observed in a PCD experiment has been rather controversial; while some apply directly the MT theory to their results,[2,4] others argue that hole-release-controlled recombination dominates the photoconductivity decay.[5–7] In order to study carrier relaxation in the presence of recombination, a bias-illumination approach has been introduced, in which the transient photoconductivity or infrared absorption induced by an optical pulse is monitored in the presence of a cw background illumination.[10–14]

In this paper, we present results on the excitation-intensity and temperature dependences of the decay of the photocurrent from the steady state in undoped *a*-Si:H in the time range $t > 1$ sec. We will show that, in general, the long-time photocurrent decay (LTPD) depends on excitation intensity. However, at sufficiently high excitation intensities, the decay becomes intensity independent and the limiting decay can be described by a power-law behavior as given by Eq. (1). It is found that the general features of the LTPD do not change with temperature, but the limiting power-law decay exponent β is strongly temperature dependent and can be greater than unity.

II. EXPERIMENTAL DETAILS

The samples studied in this work were standard undoped rf glow-discharge *a*-Si:H films on Corning-7059 glass, silica, or sapphire substrates prepared at the University of Dundee. The substrate temperature was 300 °C and film thicknesses were about 2 μm. The transport characteristics of similar samples can be found in the literature.[15] Coplanar Al electrodes 7 mm long were evaporated on top of the films, with the electrode spacing being 0.2, 0.38, or 0.56 mm. Ohmic contacts were used. The surfaces of some of the samples were freshly etched with 10%-HF solution just before they were put into the measurement chamber. No dependence of the photocurrent decay was found on the electrode spacing or on the surface condition (etched or unetched). As discussed in a previous paper,[16] in order to obtain the true intensity dependence of the photocurrent decay, it is vital that uniformly absorbed photons be used to generate the photocurrent in the sample. In the present study, the photon energy $h\nu = 1.8$ eV was used for this purpose. A monochromator, associated with a tungsten lamp, was used to produce monochromatic light. Variation of the light intensity was achieved by varying the voltage applied to the lamp. However, for reasons that will become clear later, a He-Ne laser with $h\nu = 1.96$ eV was used as the excitation source in the measurement of the limiting power-law decay behavior at various temperatures. The laser beam was expanded so that the area between the electrodes was uniformly illuminated. The unattenuated intensity of the laser beam was $F_0 = 3.7$ mW/cm^2. Neutral optical density filters were employed to attenuate the laser beam. The excitation intensity was sufficiently low so that no photoinduced changes[17] were introduced by the exciting light. The photocurrent is defined as the difference between the total current and the thermal-equilibrium dark current.

The effect of intense light soaking[17] on the LTPD was investigated for sample S2. The annealed state (*A* state) was attained by annealing the sample at 200 °C for 30 min and then cooling it slowly to room temperature, whereas the light-soaked state (*B* state) was achieved by exposing the sample at room temperature to intense white light (about 300 mW/cm^2) for 30 min. All measurements, including the annealing and light soaking, were carried out with the sample in the measurement chamber under a vacuum less than 4×10^{-5} Torr maintained by a diffusion pump with a liquid-N$_2$ cold trap. A detailed study has been carried out for two samples, S1 and S2. The results, however, are typical of all the samples studied. S1 has a 0.2-mm electrode spacing and an unetched natural surface, while S2 has a 0.56-mm electrode spacing and an etched surface. The applied electric field was typically 10^3 V/cm.

III. RESULTS

A. General features of the LTPD

In the present study, the photocurrent decay is monitored from the steady state. It is observed that, following termination of the steady-state illumination, the photocurrent drops rapidly at first, followed by a very slow decay at long times; the excess photocurrent remains well above the thermal-equilibrium dark current and persists to times greater than 10^3 sec, depending on the temperature.[18] The long-time decay of the photocurrent is in general a function of the steady-state excitation intensity. Figure 1 shows the typical long-time decay of the photocurrent of *a*-Si:H following termination of the steady-state illumination, plotted on a double-logarithmic scale for several illumination intensities. For reasons that will be discussed shortly, we represent the intensity dependence of the LTPD by showing the dependence of the decay on the steady-state photocurrent (which, of course, increases with the illumination intensity). It can be seen that the decay of the photocurrent depends on illumination intensity when the intensity is low. As the light intensity is increased, however, the decay gradually approaches a limiting behavior, which can be well represented by Eq. (1), and then becomes intensity independent as if "saturation" has been achieved for the decay process. For the intensity dependence presented in Fig. 1, "saturation" begins to occur at a steady-state photocurrent of about 1×10^{-8} A. The limiting power-law decay shown in the figure is due to the highest steady-state photocurrent attainable in the experiment. A very similar intensity dependence was observed over the entire temperature range studied, from 300 down to 50 K. Beyond this temperature range, reliable measurements become difficult; above 300 K, the photocurrent during the decay becomes comparable with or even lower than the thermal-equilibrium dark current, while below 50 K, the photocurrent drops to the detection limit of the measurement system within about 10 sec after removal of the illumination.

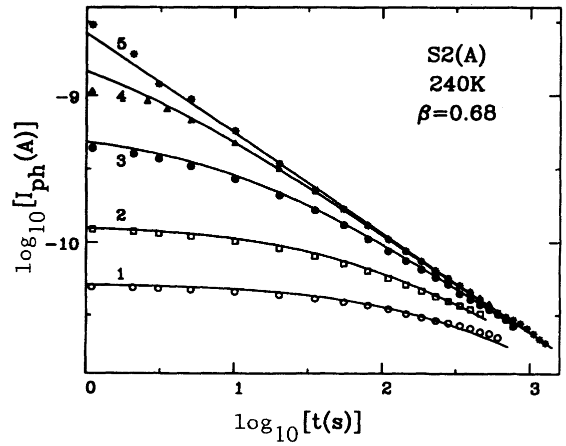

FIG. 1. Typical excitation-intensity dependence of the LTPD in *a*-Si:H, presented in terms of the steady-state photocurrent. The results were obtained for sample S2 in the annealed state with an applied voltage $V=50$ V. The steady-state photocurrent for each decay curve is as follows: 1, 5.23×10^{-11} A; 2, 1.31×10^{-10} A; 3, 5.86×10^{-10} A; 4, 3.26×10^{-9} A; 5, 9.4×10^{-7} A. Symbols: experimental data. Solid lines: calculations using Eq. (22).

It is clear from Fig. 1 that a non-power-law decay will be observed unless the illumination intensity is such that the sample is "saturated." Also clear from Fig. 1 is that, at a given temperature, the limiting power-law decay is the only decay that is determined entirely by the sample. Thus, if one is to compare the photocurrent decays for different samples, or for the same sample at different temperatures, it is the limiting behaviors that should be compared. Furthermore, the limiting power-law decay can also be obtained with non-uniformly-absorbed photons, provided that the excitation intensity is high enough to produce "saturation" in the whole sample.

The reason that we avoid linking directly the light intensity to the decay is as follows. The photocurrent decay actually measures the temporal decay of the excess photocarriers. Therefore, the meaningful variable for characterizing the initial state of the sample is the steady-state density of the photocarriers, n_{SS}, or equivalently, the steady-state photocurrent, I_{SS}, which is linearly proportional to n_{SS}. On the other hand, the illumination intensity is much less meaningful for this purpose, because n_{SS} is normally not linearly dependent on the illumination intensity, and because the same light intensity usually produces different n_{SS} at different temperatures due to the temperature dependence of the recombination rate and the quantum efficiency, neither of which is well known for *a*-Si:H.[19] However, for the results shown in Fig. 1, the highest light intensity (which generated the highest I_{SS}) was about 5×10^{-2} mW/cm^2, which, if the reflectivity is assumed to be 0.5, the absorption coefficient to be 3×10^3 cm^{-1} at $h\nu=1.8$ eV,[20] and the quantum efficiency to be unity, is equivalent to a carrier generation rate of 4×10^{18} cm^{-3}sec^{-1}. The corresponding steady-state free-carrier mobility-lifetime product is $(\mu\tau)_{\mathrm{SS}}=1.2\times10^{-5}$ cm^2/V. For the time range studied in Fig. 1, saturation begins to occur at a generation rate of about 6×10^{15} cm^{-3}sec^{-1}, and the corresponding $(\mu\tau)_{\mathrm{SS}}$ is about 8×10^{-5} cm^2/V. The lower light intensities were not measured owing to technical difficulties.

Although a very similar intensity dependence of the LTPD was observed over the entire temperature range studied, the decay rate was found to be strongly temperature dependent. In addition, the illumination intensity needed for saturating the sample increases with decreasing temperature. Figure 2 shows the limiting power-law decays at various temperatures for sample S1 in an intermediate state (I state), obtained with $h\nu = 1.96$ eV. The intermediate state was attained by exposing the sample to intense white light at room temperature for 5 min, followed by annealing it at 350 K for 30 min, and finally keeping the sample in the dark at room temperature for a few weeks. The purpose of putting the sample in the intermediate state was to ensure that the characteristics of the sample would not change during subsequent decay measurements, which took several weeks to complete. This is also one of the reasons for choosing the laser as the excitation source, as the photoemission intensity of the tungsten lamp will change when the lamp is used over a long period of time. Another reason for using the laser is to employ a high-intensity monochromatic light, which was necessary for obtaining the limiting decays at low temperatures. One can see from Fig. 2 that both the slope of the decay and the magnitude of the photocurrent are strongly temperature dependent.

In Fig. 3, the power-law-decay exponent β is shown as a function of temperature. A given symbol in Fig. 3 represents the values measured in the same run, i.e., without letting the sample return fully to thermal equilibrium between the decay measurements at different temperatures. Apparently, very consistent results were obtained no matter whether the sample fully recovers or not between the decay measurements. The average error bar in the values of β is about 0.2. We see that β exhibits a peak at about 120 K, and that the peak value $\beta_{\max}$ is greater than unity. We will show later that the peak position, $T_{\max}$, depends not only on the sample but also on its history.

We note that Shimakawa and co-workers also measured the decay of the photocurrent from the steady state in a-Si:H near room temperature.[21,22] They found that the photocurrent persisted to $t > 100$ sec and that the decay could be described by a power-law behavior [Eq. (1)] for $254 < T < 298$ K, but with a temperature-independent β. It seems clear from the present study that the reason that Shimakawa and co-workers did not observe a temperature-dependent β is that their temperature range was too small to reveal the temperature dependence of β. It is also apparent that they were dealing with the case of "saturation."

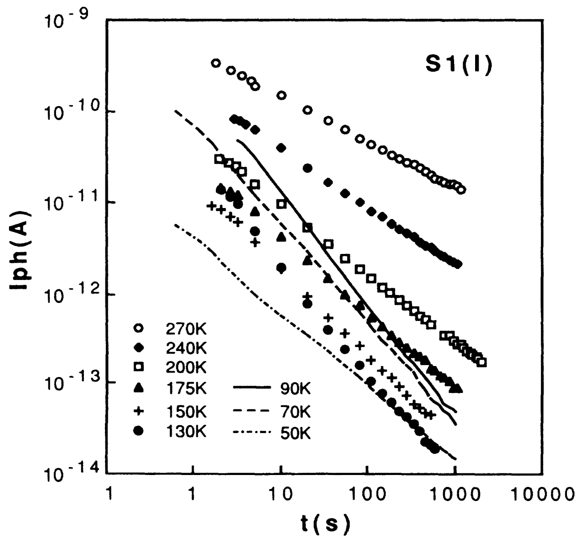

FIG. 2. Limiting power-law decays of photocurrent at various temperatures for sample S1 in an intermediate Staebler-Wronski state (I state).

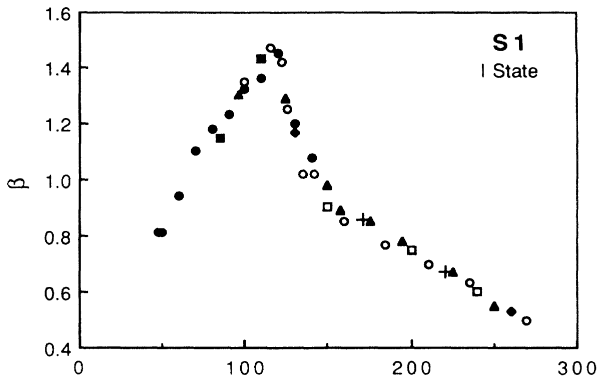

FIG. 3. Temperature dependence of the power-law-decay exponent β for sample S1 in an intermediate Staebler-Wronski state (I state). A given symbol represents the values measured in the same run, i.e., without letting the sample return fully to thermal equilibrium between the decay measurements at different temperatures.

As for the temperature dependence of the photocurrent, the first impression from Fig. 2 is probably that it is very complicated. However, if we plot the photocurrent at a given delay time as a function of temperature, an interesting feature is revealed. This is done in Fig. 4, where the photocurrents at delay times $t = 10$ and 110 sec are shown. It is interesting to see that the temperature dependence of the photocurrent is actually

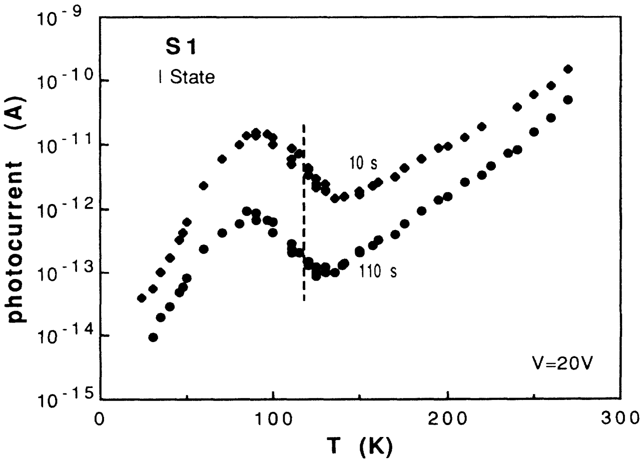

FIG. 4. Temperature dependence of the photocurrents remaining at 10 and 110 sec after termination of the steady-state illumination for sample S1 in an intermediate Staebler-Wronski state (I state). The dashed line indicates the temperature at which the peak in the temperature dependence of β appears.

structured; a peak and a minimum appear. The temperature dependence of the photocurrent can be divided into three regimes: the two regimes of increasing photocurrent at low and at high temperatures, and the regime of decreasing photocurrent between the peak and the minimum. The peak in the temperature dependence of β (see Fig. 3), at $T_{\max}$, lies in the temperature region corresponding to the regime of decreasing photocurrent. The dashed line in Fig. 4 indicates the temperature $T_{\max}$. There is also a shift of both the peak and the minimum to a lower temperature as the delay time is increased.

B. Effect of light soaking on the LTPD

The effect of intense light soaking on the LTPD has been studied for sample S2 over the temperature range between 80 and 300 K. The overall behaviors of the decays in both the *A* and *B* states are qualitatively similar to that described above for sample S1 in the *I* state. However, the details of the decays in the two states are quantitatively different. Figure 5 shows the temperature dependence of the exponent β of the limiting power-law decay for sample S2 in both the *A* and *B* states. Values of β between ~100 and ~130 K are missing in the *B* state because the photocurrent decreases so fast in this temperature region on removal of the steady-state illumination that it becomes too low to be measured in about 20 sec, which makes it impossible for a reasonably reliable value of β to be calculated from the decay data. As will be argued below, the peak in the temperature dependence of β in the *B* state probably lies at about 110 K. Thus the peak in the temperature dependence of β is shifted from about 150 K in the *A* state to about 110 K in the *B* state. In addition, light soaking has opposite effects on β in the high- and low-temperature regions; in the high-temperature region, β is decreased, while in the low-temperature region, β is increased. However, at the highest temperatures, β is little changed.

Figure 6 shows the photocurrent at a delay time $t = 10$ sec as a function of temperature in both the *A* and *B* states. Once again, the results in both states are broadly similar and possess all the features appearing in the results of Fig. 4 for sample S1 in the *I* state. From Fig. 6 one can see that the photocurrent is significantly decreased by light soaking over the entire temperature range studied. It is also clear that the peak in the temperature dependence of the photocurrent in the *B* state is more pronounced than that in the *A* state. By inspection of Figs. 5 and 6, one can see that, in the *A* state, the peak in the temperature dependence of β lies in the temperature region corresponding to the regime of decreasing photocurrent in the temperature dependence of the photocurrent, similar to the results for sample S1 in the *I* state. If the same is also true in the *B* state, then the temperature dependence of the photocurrent suggests that the peak in the temperature dependence of β in the *B* state lies between 90 and 130 K, probably at about 110 K. The low-temperature shift of the peak and the minimum in the photocurrent is consistent with a low-temperature shift of the peak in the temperature dependence of β.

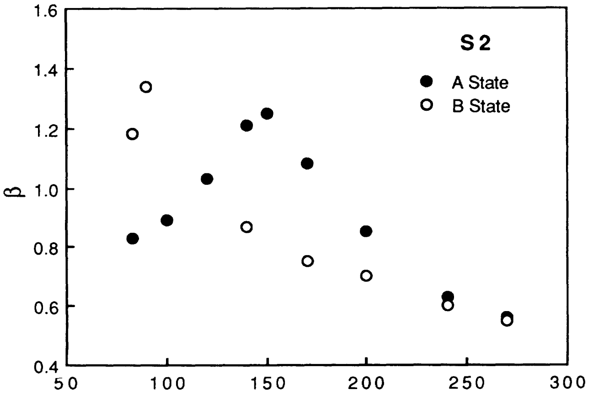

FIG. 5. Effect of light soaking on the decay exponent β for sample S2. Values of β between ~100 and ~150 K are missing in the *B* state (see the text for details). *A*: annealed state; *B*: light-soaked state.

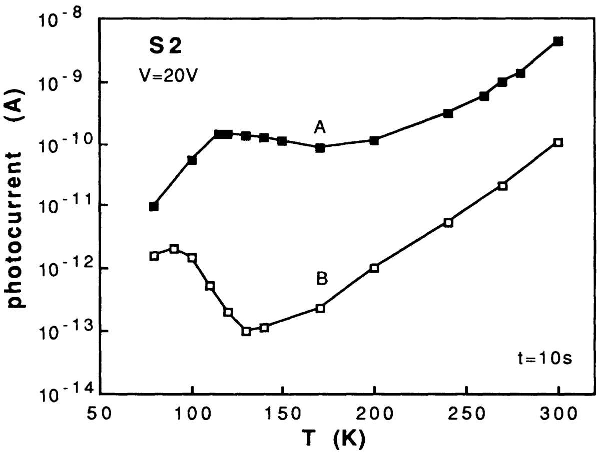

FIG. 6. Effect of light soaking on the photocurrent remaining in sample S2 10 sec after termination of the steady-state illumination. *A*: annealed state; *B*: light-soaked state.

C. Some other features of the LTPD

Despite the nonmonotonic temperature dependence of β, the long-time carrier-relaxation process in *a*-Si:H appears to be thermally activated. It was found that, for a given photoexcitation condition, the sample is further away from thermal equilibrium after photoexcitation at a lower temperature. As a matter of fact, in actual practice, one always has to raise the temperature of the sample to room temperature or above in order for the sample to return to thermal equilibrium in a reasonable time. The thermally activated nature of the long-time carrier relaxation can be clearly demonstrated by performing the following experiment. The sample is first brought into saturation by illumination at a given temperature, and then the illumination is removed. After a delay period t_d, the sample is cooled to a lower temperature and then heated up to above the illumination temperature. The photocurrent is monitored throughout this experiment. Figure 7 shows the results for illuminations at 220 and 260 K for sample S2 in the *B* state, with a delay time of 10 min, and a cooling and heating rate of 3 K/min. One can see that during the cooling and heating below the il-

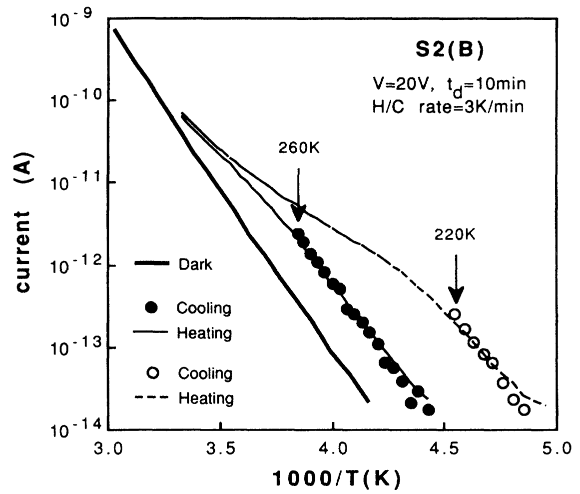

FIG. 7. Total current of sample S2 in the light-soaked state during cooling and heating after the sample was illuminated at the temperatures indicated and after a 10-min delay period. The cooling and heating rate was 3 K/min.

lumination temperature, the logarithm of the total current (dark current plus photocurrent) $I_{\rm total}$ varies linearly with $1/T$. This is a clear indication that the electrons, which are the dominant conduction carriers, are in quasithermal equilibrium, and that there is virtually no further recovery taking place below the illumination temperature. The activation energy for the linear portion of the $\log_{10}(I_{\rm total})$ vs $1/T$ curve increases with illumination temperature. Recovery is resumed when the sample is heated above the illumination temperature. It can also be seen that when the sample is heated from 220 to 260 K, after the aforementioned illumination and delay, the current is higher than that after the same illumination and delay at 260 K, indicating that for the same illumination and delay, the sample is further away from thermal equilbrium at a lower temperature.

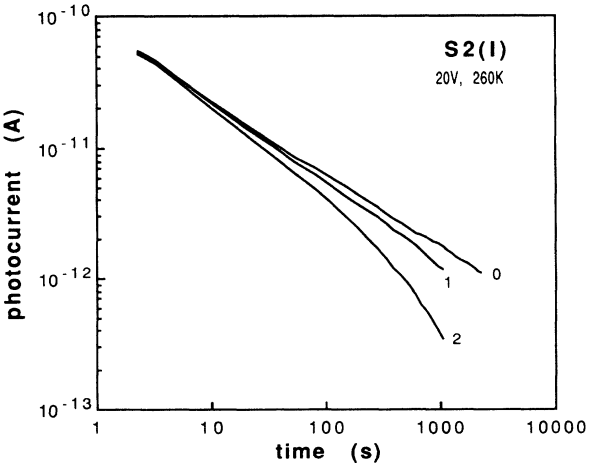

FIG. 8. Effect of bias illumination (BI) on the LTPD in *a*-Si:H. The results were obtained for sample S2 in an intermediate Staebler-Wronski state (*I* state). Curve 0: without BI; curves 1 and 2: with BI. See the text for details.

We also studied the effect of bias illumination[14] on the LTPD. Figure 8 shows the limiting long-time decays in sample S2 in an intermediate state (light soaked for 5 min) at 260 K for three bias-illumination levels. The thermal-equilibrium dark current I_d was 4.3×10^{-13} A. Curve 0 was obtained without bias illumination, and curves 1 and 2 with bias illumination. The bias illuminations were done with white light. Again, the intensities of the bias illuminations were not measured; however, the steady-state background currents for curves 1 and 2 were 1.17×10^{-12} and 2.97×10^{-12} A, respectively. One can see that the limiting decay with bias illumination no longer exhibits a power-law behavior and becomes faster with increasing level of bias illumination.

IV. POSSIBLE MODELS

A. Conventional considerations

Under steady-state illumination, a large number of nonequilibrium carriers are trapped in the gap states. To a good approximation, the occupancy of the states can be described by the quasi-Fermi levels.[23] Following termination of the steady-state excitation, unlike in the case of pulsed excitation, recombination of nonequilibrium carriers, rather than capture of free excess carriers by the traps, is the predominant event. This recombination determines the decay of the photocurrent. The long-time decay of the photocurrent indicates the long recombination lifetimes of the nonequilibrium carriers. In undoped *a*-Si:H the photoconductivity is dominated by electrons due to their much larger mobility.[15] If the temperature is not too low, it is likely that after removal of the excitation, the trapped electrons are thermally released to extended states at the conduction-band mobility edge E_c before they can recombine with holes. Whether retrapping is important depends on the relative rates of thermal emission and recombination. Thus, we will consider two cases: strong recombination and weak recombination.

Case 1. Strong recombination. When recombination is strong, retrapping is negligible and the decay of the photoconductivity is limited by thermal emission of trapped electrons. A similar case has been considered by Fritzsche and Ibaraki.[24] Assuming that there exists quasiequilibrium between thermal emission and recombination, they show that the photoconductivity at time t can be approximated by

$$\sigma_{\rm ph}(t)=ekTN_t(E_{dn})f_0(E_{dn})\mu_n\tau_n/t\ , \tag{2}$$

where e is the electronic charge, k is the Boltzmann constant, $N_t(E)$ is the density of gap states, $f_0(E)$ is the occupation function in the steady state, μ_n is the extended-state conductivity mobility, and τ_n, the free electron recombination lifetime. E_{dn} is the demarcation energy for electrons and is given by[24,25]

$$E_{dn}=E_c-kT\ln(\nu_0 t)\ , \tag{3}$$

where ν_0 is the attempt-to-escape frequency. For the limiting decay, $f_0(E)\approx1$.

For monomolecular recombination, τ_n is constant.

Equation (2) then leads to, for a uniform distribution of traps, $\sigma_{\text{ph}} \sim t^{-1}$. For an exponential distribution of traps

$$N_t(E) \sim \exp[\mp(E_c - E)/kT_0] , \tag{4}$$

we have

$$\sigma_{\text{ph}} \sim t^{-(1 \pm T/T_0)} . \tag{5}$$

When the density of the traps decreases as the energy moves towards midgap, bimolecular recombination can occur. Assuming an exponential form for the trap distribution, i.e.,

$$N_t(E) = N_0 \exp[-(E_c - E)/kT_c] , \tag{6}$$

we have for the density of trapped electrons

$$n_t \sim kT_c N_c \exp[-(E_c - E_{dn})/kT_c] . \tag{7}$$

It follows from Eqs. (3) and (7) that

$$n_t \sim t^{-\alpha_c} , \tag{8}$$

where $\alpha_c = T/T_c$. As a result

$$\tau_n = 1/b_r n_t \sim t^{\alpha_c} , \tag{9}$$

where b_r is the recombination coefficient. Substitution of Eqs. (3), (6), and (9) into (2) leads to $\sigma_{\text{ph}} \sim t^{-1}$.

Apparently, various values of β can be accounted for individually by a consideration of strong recombination. It should be noted, however, that the relationship between β and T found for the LTPD is in general nonlinear, while the relationship predicted by a consideration of strong recombination is linear. On the other hand, in the I and B states, β appears to decrease linearly with T at high temperatures (Figs. 3 and 5), and can be expressed empirically as

$$\beta = \beta_0 - T/T_0 , \tag{10}$$

where β_0 and T_0 are constants. However, β_0 is found to be greater than unity, which does not agree with the theoretical prediction. From the above discussion, it can be concluded that the photocurrent decay is unlikely to be limited by thermal emission of trapped electrons. In other words, retrapping is not negligible and recombination is weak. That recombination is weak is supported by the fact that the recombination mobility-lifetime product is much greater than the deep-trapping mobility-lifetime product.[26,27]

Case 2. Weak recombination. If recombination is weak, an electron will on the average experience release and retrapping events many times before it is captured by a recombination center and recombines. In this case, the electrons are in quasithermal equilibrium, and the occupancy of the electron traps and the states at E_c can be described by a quasi-Fermi level E_{fn}. Assuming again that the trap distribution has the exponential form of Eq. (6), we can write for the densities of free and trapped electrons

$$n = kTN_c \exp[-(E_c - E_{fn})/kT] \tag{11}$$

and

$$n_t \approx kT_c N_0 \exp[-(E_c - E_{fn})/kT_c] . \tag{12}$$

Equations (11) and (12) lead to

$$n_t \sim n^{\alpha_c} . \tag{13}$$

It can be shown very easily that, for monomolecular recombination, $n \sim t^{1/(1-\alpha_c)}$, and that, for bimolecular recombination, $n \sim t^{-1}$. The same results have also been obtained by Zeldov and Weiser for photoconductivity decay with pulsed excitation for the case in which the band-tail states are fully occupied by the injected carriers.[13] Thus, weak recombination gives rise to limiting power-law decays with $\beta \geq 1$ only, in disagreement with our experimental observation (Figs. 3 and 5).

B. Hole-emission-limited recombination

If trapped holes also have to be released to extended states before they can recombine with electrons, thermal emission of trapped holes can be the limiting step for recombination. In fact, this recombination mechanism has been invoked by some authors to explain their experimental results on the photocurrent decay in *a*-Si:H.[6,7,28]

Assume an exponential distribution for both the electron traps (ET's) and the hole traps (HT's),

$$N_t(E) = N_0 \exp[-(E_c - E)/kT_c] \quad (\text{ET}) , \tag{14}$$

$$P_t(E) = P_0 \exp[-(E - E_v)/kT_v], \quad (\text{HT}) . \tag{15}$$

We also assume that all the trap states concerned are occupied by the appropriate carriers at the beginning of the decay, and that $T < T_c, T_v$. Hole-emission-controlled recombination is another case of weak recombination (for electrons). Thus, the electrons are in quasithermal equilibrium. Following the discussions given in Sec. IV A for the case of quasithermal equilibrium, we obtain the relationship between the densities of free and trapped electrons

$$n \sim n_t^{1/\alpha_c} \tag{16}$$

with $\alpha_c = T/T_c$.

The occupancy of the hole traps is determined by thermal emission of holes. In the present case, retrapping of released holes is implied to be negligible. By analogy with the arguments presented in Sec. IV A for the case of electron-emission-limited recombination, we have for the density of trapped holes

$$p_t \approx kT_v P_0 \exp[-(E_{dp} - E_v)/kT_v] , \tag{17}$$

where E_{dp} is the hole demarcation energy and is given by

$$E_{dp} = E_v + kT \ln(\nu_0 t) . \tag{18}$$

Substitution of Eq. (18) into (17) yields

$$p_t \approx kT_v P_0 (\nu_0 t)^{-\alpha_v} , \tag{19}$$

where $\alpha_v = T/T_v$.

Charge neutrality requires $n_t = p_t$. Thus

$$n \sim n_t^{1/\alpha_c} = p_t^{1/\alpha_c} \sim t^{-T_c/T_v} . \tag{20}$$

Equation (20) shows that the hole-emission-limited recombination gives rise to a power-law-decay exponent that is independent of temperature. This is obviously in disagreement with the experimental results.

It should be noted, however, that the above arguments are valid even if the distributions of the traps are not exponential, but can be approximated by an exponential form over a small energy range. In this case, T_c and T_v are each a function of energy. Since at different temperatures the experiment is sensitive to different energy regions, the ratio T_c/T_v (i.e., β) will in general vary with temperature. Apparently, such a possibility cannot be ruled out. Further work is needed to find out how realistic this possibility is.

A special case of the hole-release-controlled recombination is $T_c < T < T_v$. In this case, the maximum in the energy distribution of the excess electrons in the steady state is at E_c, i.e., $n \gg n_t$. Consequently, the condition for charge neutrality changes to $n = p_t \sim t^{-T/T_v}$. This is the case considered by Werner and Kunst for their pulsed-transient decay data.[6] However, given the fact that the temperatures studied here are below 300 K, which is probably the lower limit for T_c, we do not think the condition $T_c < T < T_v$ is satisfied even at the highest temperatures studied. Moreover, the predicted β for $T_c < T < T_v$ can only be less than unity and has the opposite temperature dependence to that of the decay observed in our samples at high temperatures (Figs. 3 and 5).

C. Time-dependent bimolecular recombination

The fact that the limiting decay exhibits a power-law behavior tempts us to consider the time-dependent bimolecular-recombination (TDBR) rate equation

$$dn/dt = -bt^{-\Delta}n^2 \; , \tag{21}$$

where b and Δ are constants, and $b > 0$ and $\Delta \leq 1$. Integration of Eq. (21) gives

$$n = n_{\mathrm{SS}}/[1+(bn_{\mathrm{SS}}/\beta)t^{\beta}] \; , \tag{22}$$

where n_{SS} is the steady-state free-electron density, and $\beta = 1-\Delta$. Equation (22) predicts $n \sim t^{-\beta}$ at long times. To our surprise, Eq. (22) agrees well with the decays with $\beta < 1$, in both the high- and low-temperature regimes. An example of the fitting for $\beta < 1$ is given in Fig. 1, where the solid curves are the results of calculations made using Eq. (22). It should be noted that the decay curves in Fig. 1 were fitted as a whole and not individually, because fitting of the limiting power-law decay determines all the parameters in Eq. (22). The other fits were obtained by substituting the corresponding steady-state carrier density (which is proportional to the steady-state photocurrent) into Eq. (22). The first two points in the limiting power-law decay, which have a large uncertainty, were ignored in the fitting.

For $\beta > 1$, the result of the fitting is very unsatisfactory. However, this is not surprising. We see from Eq. (21) that $\beta > 1$ requires $\Delta < 0$. This means that the recombination would proceed with a recombination coefficient that increases with time. Obviously, such a process is physically unreasonable.

A similar TDBR rate equation has also been considered by other authors in the study of photoinduced infrared absorption.[29] In a photoinduced absorption experiment, one measures the time decay of photocarriers following a pulsed excitation. According to the theories,[3,30,31] the transport of photocarriers following a pulsed excitation is dispersive, which gives rise to the TDBR of the photocarriers. In a photoconductivity decay experiment, however, one measures the time decay of the free photocarriers only; thus it is difficult to imagine how the TDBR coefficient comes into this issue. We would therefore like to stress that Eq. (21) is purely empirical at this stage of our study; the mechanism responsible for the TDBR is yet to be uncovered. We believe, however, that such an empirical expression will help in further study of this problem.

V. DISCUSSION

We have shown that a power-law decay can be predicted by several models, which, however, predict different temperature dependences for β. Therefore, the power-law decay behavior alone is not sufficient to provide evidence for the applicability of any of the models. Although the details of the processes responsible for the LTPD in *a*-Si:H are not clear, it seems certain that the LTPD is limited by recombination which is bimolecular in nature. This view is supported by the fact that bias illumination enhances the photocurrent decay at long times (see Fig. 8). At short times, the number of photocarriers due to the terminated illumination is greater than the number of photocarriers due to the bias illumination; thus the recombination is bimolecular and the decay is not affected by the presence of bias illumination. At long times, however, the number of the photocarriers due to the terminated illumination has decayed considerably and is smaller than the number of photocarriers due to the bias illumination; thus the number of recombination centers becomes independent of time and the decay is monomolecular.

One may argue that the results presented in Fig. 7 are evidence that the LTPD is limited by thermal emission of trapped electrons, as similar results have usually been explained in terms of electron-emission-limited relaxation.[24] In fact, the results of Fig. 7 can also be understood on the basis of recombination-limited relaxation. After cessation of the excitation, excess electrons recombine with holes via extended states. At the end of the delay period, considerable recombination has already taken place, and the density of free electrons in extended states is very small, and thus recombination is slow. When the sample is cooled below the illumination temperature, the density of free electrons is further decreased, and recombination becomes even slower. Therefore, one observes virtually no further decay of the photocurrent. At the same time, the electrons are in quasithermal equilibrium, which results in a well-defined activation energy. When the temperature is raised above the illumination temperature, the density of free electrons increases significantly, resulting in an evident decay of the photocurrent.

It is not clear either what is the mechanism of the effect of intense light soaking on the LTPD, and further study is underway. However, we would like to point out here that it appears that there is a correlation between the temperature dependence of the LTPD and that of the steady-state photoconductivity.[18] This correlation is further evidence that the LTPD is limited by recombination. According to a recent study of the steady-state photoconductivity,[32] at low temperatures the recombination is predominantly via the band-tail states, while at high temperatures the deep defect states (dangling bonds) are the dominant recombination centers. Thus, the temperature $T_{\max}$ (corresponding to $\beta_{\max}$) probably marks the transition between band-tail-dominated and deep-defect-dominated recombination. Accordingly, the decrease of the photocurrent (see Fig. 6) after light soaking is caused by an increase in the number of dangling bonds.[33] The increase of the number of dangling bonds also causes the transition to occur at a lower temperature, thus giving rise to a low-temperature shift of $T_{\max}$ after light soaking.

Before ending the discussion, we would like to make a few further remarks. First, considering the work by Street and co-workers,[34,35] it is natural to ask whether the long-time photocurrent decay observed in *a*-Si:H is a true bulk property. This question has been answered in an earlier paper,[16] where we demonstrate clearly that the LTPD is indeed a bulk property.

Second, as far as the persistence of photoconductivity is concerned, the LTPD observed here is very similar to the persistent photoconductivity (PPC) observed in compensated *a*-Si:H and in various types of *a*-Si:H multilayers.[36] However, the generation of the PPC is strongly thermally activated;[36,37] for the same amount of illumination, the PPC increases with illumination temperature. By contrast, in the LTPD, the resulting excess conductivity is higher for illumination at a lower temperature than for the same illumination at a higher temperature (see Fig. 7). Furthermore, the light intensity needed for generating the PPC, which is typically 80 mW/cm^2, is many orders of magnitude higher than the light intensity needed for generating the limiting decay in the LTPD experiment. Exposure of our samples to a light of comparable intensity to that usually used for generating the PPC results in significant reduction in both the dark conductivity and the high-temperature photoconductivity (the Staebler-Wronski effect[17]). Thus, the LTPD observed here has a different origin from that of the PPC. It seems that carrier trapping by the band-gap states is the only reasonable explanation of the origin of the LTPD studied in this work.

VI. SUMMARY AND CONCLUSIONS

We have measured the intensity and temperature dependences of the long-time decay of the photocurrent from the steady state in undoped *a*-Si:H. The long-time photocurrent decay is found to depend on the steady-state excitation intensity when the excitation intensity is low, but to approach a limiting power-law behavior, $I_{\mathrm{ph}} \sim t^{-\beta}$, with increasing excitation intensity and eventually to become intensity independent. While a similar intensity dependence is observed over the entire temperature range studied (50–300 K), the exponent β of the limiting power-law decay is strongly temperature dependent. A peak appears between ~ 110 and ~ 150 K in the temperature dependence of β, depending on the history of the sample as well as on the sample itself. Several possible photocurrent decay mechanisms have been examined, with the conclusion that the LTPD cannot be explained in terms of the conventional considerations. Empirically, the decays with $\beta < 1$ can be described by a time-dependent bimolecular recombination; however, the mechanism responsible for the TDBR is not clear.

ACKNOWLEDGMENTS

The authors are grateful to Professor P. G. Le Comber of the University of Dundee for supplying the samples studied in this work, and to Dr. W. I. Milne of the University of Cambridge and Professor H. Fritzsche of the University of Chicago for providing other samples. The authors would also like to thank Professor Fritzsche for valuable discussions. J-H. Zhou acknowledges financial support from Trinity College, Cambridge.

---

[1] T. Tiedje, J. M. Cebulka, D. L. Morel, and B. Abeles, Phys. Rev. Lett. **46**, 1425 (1981).

[2] J. M. Hvam and M. H. Brodsky, Phys. Rev. Lett. **46**, 371 (1981).

[3] J. Orenstein, M. A. Kastner, and V. Vaninov, Philos. Mag. B **46**, 23 (1982).

[4] C-Y. Huang, S. Guha, and S. J. Hudgens, Phys. Rev. B **27**, 7460 (1983).

[5] H. Oheda, Philos. Mag. B **52**, 857 (1985).

[6] A. Werner and M. Kunst, Phys. Rev. B **36**, 7567 (1987).

[7] C. Main, R. Russell, J. Berkin, and J. M. Marshall, Philos. Mag. Lett. **55**, 189 (1987).

[8] T. Tiedje and A. Rose, Solid State Commun. **37**, 49 (1981).

[9] J. Orenstein and M. A. Kastner, Phys. Rev. Lett. **46**, 1421 (1981).

[10] R. Pandya and E. A. Schiff, J. Non-Cryst. Solids **59&60**, 297 (1983).

[11] D. Pfost, Z. Vardeny, and J. Tauc, Phys. Rev. Lett. **52**, 376 (1984).

[12] X. Chen and C.-Y. Tai, Phys. Rev. B **40**, 9652 (1989).

[13] E. Zeldov and K. Weiser, Phys. Rev. Lett. **53**, 1012 (1984).

[14] R. Pandya and E. A. Schiff, Philos. Mag. B **52**, 1075 (1985).

[15] W. E. Spear, J. Non-Cryst. Solids **59&60**, 1 (1983), and references therein.

[16] J-H. Zhou and S. R. Elliott, Phys. Rev. B **45**, 1668 (1992).

[17] D. L. Staebler and C. R. Wronski, Appl. Phys. Lett. **31**, 292 (1977); J. Appl. Phys. **51**, 3262 (1980).

[18] J-H. Zhou, Ph.D. dissertation, University of Cambridge, 1992.

[19] R. S. Crandall, in *Semiconductors and Semimetals,* edited by J. I. Pankove (Academic, New York, 1984), Vol. 21(B), p. 245.

[20] L. Ley, in *The Physics of Hydrogenated Amorphous Silicon (II),* edited by J. D. Joannopoulos and G. Lucovsky, Topics

in Applied Physics Vol. 56 (Springer-Verlag, New York, 1984), p. 61.

[21] K. Shimakawa and Y. Yano, Appl. Phys. Lett. **45**, 862 (1984).

[22] K. Shimakawa, Y. Yano, and Y. Katsuma, Philos. Mag. B **54**, 285 (1986).

[23] A. Rose, *Concepts in Photoconductivity and Allied Problems* (Interscience, New York, 1963).

[24] H. Fritzsche and N. Ibaraki, Philos. Mag. B **52**, 299 (1985).

[25] J. G. Simmons and M. C. Tam, Phys. Rev. B **7**, 3706 (1973).

[26] E. A. Schiff, Philos. Mag. Lett. **55**, 87 (1987).

[27] R. S. Crandall and I. Balberg, Appl. Phys. Lett. **58**, 508 (1991).

[28] T. J. McMahon and R. S. Crandall, Phys. Rev. B **39**, 1766 (1989); Philos. Mag. B **61**, 425 (1990).

[29] Z. Vardeny, P. O'Connor, S. Ray, and J. Tauc, Phys. Rev. Lett. **44**, 1267 (1980).

[30] H. Scher and E. W. Montroll, Phys. Rev. B **12**, 2455 (1975).

[31] T. Tiedje, in *Semiconductors and Semimetals,* edited by J. I. Pankove (Academic, New York, 1984), Vol. 21(C), p. 207.

[32] J-H. Zhou and S. R. Elliott, Philos. Mag. B (to be published).

[33] H. Dersch, J. Stuke, and J. Beichler, Appl. Phys. Lett. **38**, 456 (1981).

[34] W. B. Jackson, R. A. Street, and M. J. Thompson, J. Non-Cryst. Solids **59&60**, 497 (1983).

[35] R. A. Street, J. Non-Cryst. Solids **77&78**, 635 (1985).

[36] S. C. Agarwal and S. Guha, in *Disordered Semiconductors,* edited by M. A. Kastner, G. A. Thomas, and S. R. Ovshinsky (Plenum, New York, 1987), p. 519.

[37] A. Hamed, H. Fritzsche, X.-M. Deng, S. Kohler, and R. Gupta, J. Non-Cryst. Solids **137&138**, 287 (1991).