Optical Bias Control of Dispersive Relaxations in a-Si:H

D. Pfost, Z. Vardeny,[a] and J. Tauc

Division of Engineering and Department of Physics, Brown University, Providence, Rhode Island 02912

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Relaxation of the photoinduced ir absorption band in a-Si:H was studied in the microsecond time domain as a function of cw bias illumination. The decays follow a power law $t^{-\alpha}$ where the dispersion parameter α increases with bias illumination. At low temperatures, α increases linearly with the steady-state carrier density while at high temperatures it saturates at high illumination levels. These results are interpreted as evidence for the bias-controlled tunneling process at low temperatures and multiple trapping process with tail states under the saturation condition at high temperatures.

Illumination of amorphous silicon (a-Si:H) with supergap photon energy produces a transient midgap absorption (PA) band whose decay has been used for studying carrier recombination.[1,2] The integrated PA band decay can often be described over several decades of time by a power law $t^{-\alpha}$ where α was identified as the dispersion parameter of carriers. At high temperatures, in samples with a low density of gap states, the dispersion process has been analyzed[1,2] in terms of the multiple-trapping (MT) model.[3,4] The dispersion parameter has been associated with the high-mobility carriers—electrons in a-Si:H[5]—whose transport determines the recombination rate. When the distribution of traps below the mobility edge is exponential, the dispersion parameter is $\alpha_0 = T/T_0$ where T_0 is the width of the distribution. This model has provided good agreement with experimental data. However, Schiff[6] has noted that this agreement cannot be interpreted as a proof of the MT model in the original version.[3,4] This model assumes a small occupancy of the trap states while it is likely that with the illumination levels used in the reported experiments saturation of the traps occurs.[6,7] In this case, the effective carrier mobility is limited by recombination rather than by trapping and thermal reexcitation. We will show that if the recombination is bimolecular, the $t^{-\alpha_0}$ law still applies at long times as in the original MT model; however, if the recombination is monomolecular, the decay is proportional to $t^{-\alpha_0/(1-\alpha_0)}$.

We measured the PA band decay in the presence of cw bias illumination. This illumination generates a large steady-state density of electrons and holes and forces the recombination to be monomolecular. We observed that at long times and high bias intensities the decay is indeed proportional to $t^{-\alpha_0/(1-\alpha_0)}$. We believe that this result is the first identification of saturation in the MT model.

At low temperatures, it was proposed that the prevailing recombination mechanism is tunneling.[8] The bias illumination method enabled us to vary the density of partners N_H available for tunneling recombination. Our data show that the change of α is proportional to N_H in agreement with theory.[9,10]

For measuring the PA decay under bias illumination we used three different light sources. The transient excess carrier population $N(t)$ was produced by a 14-Hz nitrogen-pumped dye laser with 10-ns pulses of 2.1 eV photon energy and 50 μJ energy per pulse. The steady-state carriers, of concentration N_{ss}, were produced by a cw Ar$^+$ laser with a controlled intensity up to 50 mW/cm^2. The probe source was an incandescent light with filters restricting the photon energy to between 0.8 and 1.4 eV and was used to measure the induced change in transmission ΔT associated with transient excess carriers. A fast Ge detector with an amplifier and a signal averager was used to measure $\Delta T(t)$. The steady-state induced change in transmission, ΔT_{ss}, was measured by a conventional phase detection technique. In all cases, at 10 μs, ΔT_{ss} was larger by at least an order of magnitude than ΔT. The sample was a 1-μm-thick film of glow-discharge a-Si:H on a quartz substrate kept in a cryostat with controlled temperatures between 10 and 300 K.

In Fig. 1 the transient relative change $-\Delta T(t)/T$ is plotted versus the delay time following the 10-ns pump pulse, on logarithmic scales, at various temperatures and bias illuminations. In agreement with previous work[1,2,5] we observed power-law decays $t^{-\alpha}$ in the time interval 10^{-5} to 10^{-3} s. The data for the no-bias case are shown in Fig. 1(a) and in the inset. The dispersion parameter α_0 increases slowly with temperature from a nonzero value at $T = 0$ K. This has been
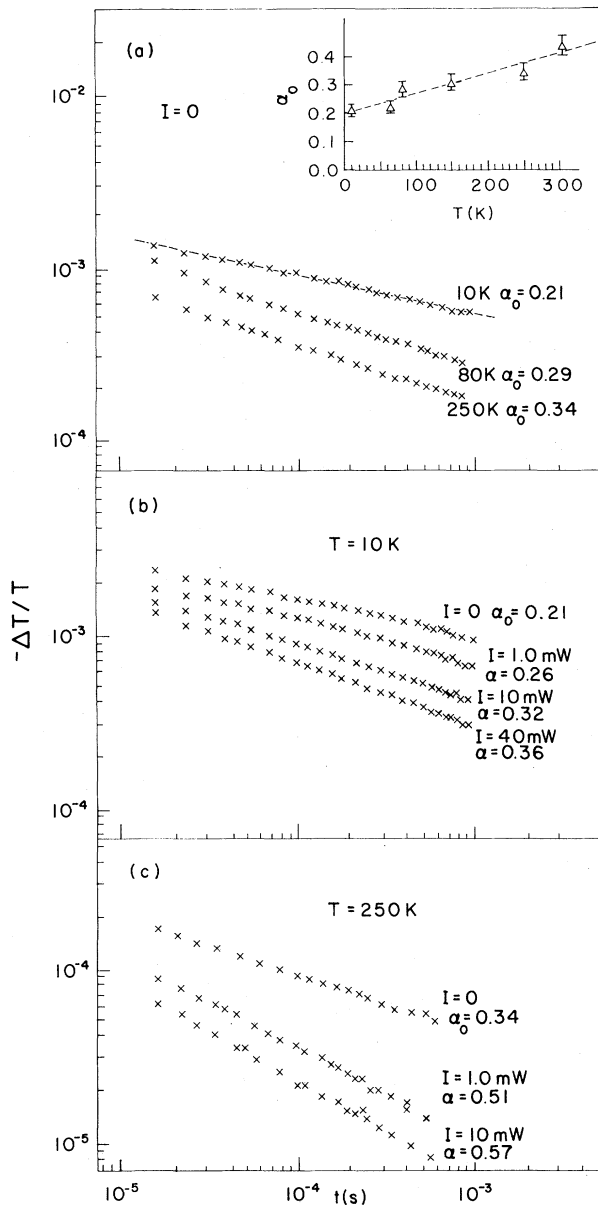
FIG. 1. Decays of induced absorption band in a-Si:H at different temperatures and different optical biasing levels I.

FIG. 2. $\Delta\alpha = \alpha - \alpha_0$ as a function of the strength of the PA band produced by optical biasing.

interpreted as recombination by tunneling at low temperature with the MT recombination channel becoming progressively more important at high temperatures.[8]

PA decays at various biasing illuminations (BI) are presented in Fig. 1(b) for 10 K and Fig. 1(c) for 250 K. At both temperatures α increases with the intensity I of the cw laser. At 250 K the increase is more pronounced than at 10 K: α increases from 0.2 to 0.3 at 10 K under BI of 10 mW/cm² and under the same bias α increases
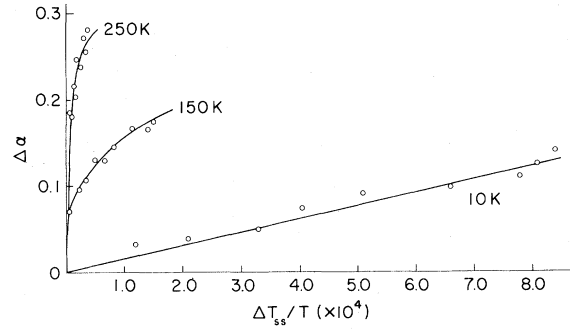
from 0.37 to 0.58 at 250 K. The intensity I is not the directly relevant variable for studying the dependence of α on biasing. A more meaningful variable is the steady-state excess carrier concentration N_{ss} produced by the BI. This is because N_{ss} depends sublinearly[11] on I and also because the same I produces different N_{ss} at different temperatures as a result of the temperature-dependent recombination rate.[11] The steady-state change in transmission produced by BI, ΔT_{ss}, is proportional[2] to N_{ss}. For these reasons, we plot in Fig. 2 $\Delta\alpha = \alpha - \alpha_0$ versus $\Delta T_{ss}/T$. We note a significant difference in the low- and high-temperature forms of this function. At 10 K, $\Delta\alpha$ is proportional to $\Delta T_{ss}/T$ (therefore N_{ss}); at high temperatures, a trend toward saturation is seen starting at low values of $\Delta T_{ss}/T$. We will show that these functional dependencies are in agreement with the tunneling model (applicable at low T) and MT model (at high T).

At low temperatures, in the time range of our measurements, the photogenerated carriers are immobile and their recombination is governed by one-event tunneling.[12,13] The dispersive character of the recombination is due to a distribution of tunneling times as produced, for example, by the distribution of tunneling distances. Since $N(t) \ll N_{ss}$ the recombination kinetics is monomolecular. Under this condition α extracted from the decay curves can be directly related to the dispersion parameter, avoiding the complication of the bimolecular recombination.[14,15] Therefore the increase in α with I shows that the recombination becomes less dispersive when I is increased. According to the theory[9,10] the dispersion parameter should be proportional to the density of N_{ss} as observed (Fig. 2).

We note that the concentration of tunneling partners can be increased by doping as reported

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in Ref. 8. However, by doping one introduces additional states in the gap that have a strong influence on the recombination process. It is therefore no surprise that in this previously reported case α is not proportional to the carrier concentrations introduced by doping. However, the biasing experiment avoids the complication of new states being introduced in the gap by doping.

At high temperatures recombination by tunneling is unlikely because carriers leave their sites very quickly. The results have been interpreted[1] in terms of the MT model. The recombination rate is limited by electron transport which depends on the trapping in tail states below the mobility edge of the conduction band E_c, assumed to have the distribution

$$g(E) = (N_t/kT_0)\exp[-(E_c - E)/kT_0]$$

where N_t is the total density of states in the tail. The occupancy of these states by electrons is determined by the Fermi distribution with a demarcation energy E_d which separates states whose probability of thermal reexcitation is smaller or larger than $\frac{1}{2}$. The occupancy of the states below E_d depends on the competition between the trapping and recombination processes. If recombination is neglected, trap saturation occurs[6] at

$$t_s = \nu^{-1}(N_0 \sin\alpha_0\pi/N_t\alpha_0\pi)^{-1/\alpha_0},$$

where ν is the attempt-to-escape frequency, $\alpha_0 = T/T_0$, and N_0 is the total electron density. Saturation can occur if the recombination time τ is longer than t_s. We present an analysis which indicates that in our experiments saturation has occured before the time of measurement. We assume that the states below E_d are completely filled and that E_d sinks deeper with time by recombination rather than by thermalization as in the MT model. The process is described by the following equations:

$$dN/dt = -b_r N_r n, \qquad (1)$$

$$N = \int^{E_d} g(E)dE = N_t \exp[-(E_c - E_d)/kT_0], \qquad (2)$$

$$n = [g(E_c)/g(E_d)]\exp[-(E_c - E_d)/kT], \qquad (3)$$

where N is the time-dependent total electron density in the tail, n, is the electron density in the conduction band $(n \ll N)$, b_r is the recombination coefficient, N_r is the density of recombination partners, and $g(E_c)$ and $g(E_d)$ are the densities of electron states at the bottom of the conduction

band E_c and at E_d, respectively.

With the boundary conditions $N(0) = N_0$, one obtains for the monomolecular case $(N_r = \text{const})$

$$N(t) = N_0/(1 + t/\tau_M)^{\alpha_0/(1-\alpha_0)} \qquad (4)$$

and for the bimolecular case $(N_r = N)$

$$N(t) = N_0/(1 + t/\tau_B)^{\alpha_0}, \qquad (5)$$

where the recombination times are

$$\tau_M^{-1} = b_r(1 - \alpha_0)N_r N_c N_t^{-1/\alpha_0} N_0^{(1-\alpha_0)/\alpha_0}$$

and

$$\tau_B^{-1} = b_r \alpha_0^{-2} N_c N_t^{-1/\alpha_0} N_0^{1/\alpha_0}$$

where N_c is the effective state density at the mobility edge of the conduction band $[N_c = kTg(E_c)]$.

Equation (5) shows that at long times $(t \gg \tau_B)$ the decay of PA is proportional to $t^{-\alpha_0}$, which is the same as when the traps are not saturated. Since the experiments are usually done in this time domain, the PA decay study *cannot* identify trap saturation if the recombination is bimolecular.

However, if the recombination is monomolecular, long-time decay is proportional to $t^{-\alpha_0/(1-\alpha_0)}$ [Eq. (4)] which we have actually observed by the optical biasing of PA at large BI (Fig. 3). This shows that the traps are saturated and that the biasing forces the recombination to be monomolecular.[16]

Experimental data show that ΔT_{ss} is at least ten times larger than $\Delta T(t)$ in the time range of measurement. This implies $N_r \simeq N_{ss} \gg N(t)$ and monomolecular recombination should prevail.
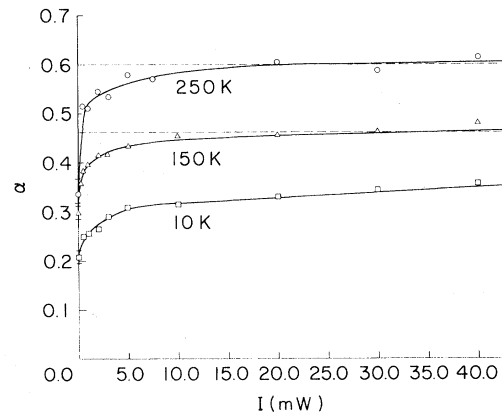

FIG. 3. Dispersion parameter α as a function of bias-light intensity. The dotted lines are asymptotes at $T = 150$ and 250 K corresponding, within the accuracy of the measurement, to $\alpha = \alpha_0/(1 - \alpha_0)$ where $\alpha_0 = \alpha(I = 0)$.

However, it is difficult to understand how this condition is realized. In our experiments, $N_0 \simeq 10^{18}$ cm^{-3} and the cw carrier generation rate is about 10^{22} cm^{-3} s^{-1} giving as a rough estimate $N_{ss} \simeq 10^{16}$ cm^{-3} (if $b_r = 10^{-10}$ cm^{-3} s^{-1}). Another difficulty is to explain why under biasing the recombination of holes is still dominated by electrons produced by the pulse rather than by the high concentration of cw electrons.

These discrepancies are removed if we assume that below the band tail of the conduction band there are deeper electron traps. Electrons in these traps do not contribute to the recombination of holes via transport in the conduction band. The presence of these states has the following consequences. All or most of the electrons produced by biasing are in the deep traps; therefore the cw hole densities can be significantly larger than the above estimate of 10^{16} cm^{-3}. The hole recombination rate and therefore $\Delta T(t)$ are determined by the transient electron density. During the thermalization process, electron trapping in the deep states competes with the MT mechanism and delays the onset of saturation[7]; therefore filling of these states by biasing facilitates saturation of the tail states. These deep electron traps are probably the dangling bonds that have been observed in many studies[17-19]; when they are doubly occupied (D^-), their energy is approximately 0.7 eV below the bottom of the conduction band.

In conclusion, we found that optical biasing of a-Si:H leads to an increase of the dispersion parameter. At low temperatures, the change of the dispersion parameter is proportional to the steady-state population of carriers in agreement with the tunneling mechanism of recombination. At high temperatures, the biasing induces monomolecular kinetics which reveals the tail-state saturation identified by the change of the dispersion parameter from the zero-bias value α_0 to the high-bias value $\alpha_0/(1 - \alpha_0)$.

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[a]Permanent address: Solid State Institute, Technion, Haifa, Israel.

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