Optical-bias-enhanced transient photocurrent in amorphous silicon

Xing Chen and Chen-Yu Tai

University of Toledo, Toledo, Ohio 43606 (Received 20 March 1989; revised manuscript received 5 June 1989)

Transient photocurrent in *a*-Si:H film is increased by a factor of more than 10 due to optical-bias illumination. The transient photocurrent is found to decay exponentially shortly after the light pulse is turned off. The exponential-decay time constant of the transient photocurrent, which is interpreted as the lifetime of electrons trapped at the quasi-Fermi-level, is found to be proportional to $F^{-0.38}$ (*F* is the optical-bias level) at room temperature. Enhancement in transient photocurrent response may be understood as a result of partial saturation of the trap states in the band tail by the optical-bias illumination. Because of the partial saturation of the gap states and the use of a weak probe pulse, the system studied by this technique is in a quasi-steady-state with small perturbation where experimental results can be interpreted with little ambiguity. This effect is applied to study the density of the band-tail states of the sample as well as the process of charge recombination.

I. INTRODUCTION

Transient-photocurrent response in *a*-Si:H films with coplanar electrodes has been widely applied to study the transport of photoinduced excess carriers in amorphous semiconductors.¹⁻⁴ Dispersive transport in amorphous semiconductors is generally interpreted in terms of the multiple-trapping model.⁵⁻⁸ Although this model appears to account for many experimental data,⁹⁻¹² some difficulties arise.^{2,3,13-15} Since conventional experiments on transient photocurrent are not studied in a steady state and the system studied is extremely complicated, extensive work is needed to derive information such as the nature and density of states (DOS) in the band tail from the decay curve of the photocurrent. It is therefore desirable to develop a technique that can obtain information on the DOS as well as the process of charge transport in a quasi-steady-state where experimental results can be interpreted with little ambiguity.

In this work we report the observation of optical-bias enhancement of transient photocurrent by a factor of more than 10 and the application of this effect to measure the DOS. The process of recombination of charge carriers is also studied in this work. The observed enhancement in transient-photocurrent (TPC) response may be understood as a result of partial saturation of the trap states in the band gap by the optical-bias illumination. Because of the partial saturation of the gap states, the system studied by this technique is in a quasi-steady-state with small perturbation. Observation of optical-biasenhanced transient photocurrent also implies that the time required to "thermalize" a charge carrier is much shorter than the time of recombination. This result contradicted with the conclusion of some earlier works.^{16,17}

It is also discovered in this work that transient photocurrent decreases exponentially with time several microseconds after the probe light pulse is turned off. The decay rate of the transient photocurrent is found to be sensitive to the bias illumination level as well as the temperature. The exponential-decay time constant observed in this work is interpreted as the lifetime of the electrons trapped in the vicinity of the quasi-Fermi-level E_{ft} , which is determined by the rate of emission of electrons trapped in the band-tail states and the rate of recombination of charge carriers in the conduction band. This discovery is important in understanding the mechanism of the process of charge recombination.

Transient photocurrent (with the Fourier-transform method)² and transient photoinduced optical absorption with optical bias^{10,15} have been studied. Moderate enhancement (approximately 20% enhancement with both the method of Fourier transform and the method of using a 15-ns laser impulse) was reported by Pandya and Schiff.^{2,17} A theory based on a saturated band-tail model was proposed by Zeldov and Weiser¹⁴ to explain the influence of optical biasing on the transient-response experiments of Pandya and Schiff as well as the decay of photoinduced absorption in a-Si:H observed by Pfost, Vardeny, and Tauc.¹⁵ Exponential decay was also predicted by Zeldov and Weiser with the saturated band-tail model. However, the saturated band-tail model for undoped a-Si:H was later disputed by Comrad, Pandya, and Schiff¹⁶ and by Stoddart, Tauc, and Vardeny¹⁸ because of disagreement with their data. Nevertheless, exponential decay of small-signal photocurrent on an a-Si:H sample was reported by Ritter, Zeldov, and Weiser¹⁹ with the method of modulating 5% of an illumination beam by a square wave. The decay time constant of 1.8 μ s observed in that work was interpreted as the carrier lifetime. However, since the observation time span in that work was only 5 μ s, it is not clear that the system studied has reached a steady state and the data can be fitted by only an exponential curve. It is therefore important to perform a more complete study of optical bias enhancement of the transient photoconductivity in a-Si:H.

In this work, optical-bias enhancement in transient photocurrent is observed in a quasi-steady-state with small perturbation. As will be discussed later, in most of the reported work on transient photoconductivity, charge carrier density created by the light pulse at its maximum is not negligibly small compared to that generated by the bias illumination. Furthermore, in most of the earlier works the charges injected by the probe pulse were continuously immobilizing into deeper states and thus the system studied were not in a steady state. The probe pulse used in the current experiment (1 nJ/pulse or 10 nJ/cm²) is approximately four orders weaker than what was used in a typical transient experiment in *a*-Si:H (~50 μ J/pulse).¹⁵

The apparatus setup and experimental results are presented in Sec. II. A theory based on a multipletrapping (MT) model is presented in Sec. III to explain the enhancement in transient photocurrent as well as the rate of its decay.

II. EXPERIMENT

The experimental setup is shown schematically in Fig. 1. The bias illumination was obtained from a 10-mW helium-neon laser. Intensity of the bias illumination was varied by using neutral-density (ND) filters. Pulses of duration 0.3 μ s and a repetition rate of 300 Hz were obtained by reflecting a 5-mW helium-neon laser beam from a mirror mounted on a rotor driven by compressed air. The laser beams had a diameter of approximately 3 mm and the energy density per pulse at the sample was approximately 10 nJ/cm^2 . As will be shown later, the probe pulse was sufficiently weak that it caused only a small perturbation to the steady state created in the sample by the bias illumination. The transient photocurrent was detected using $\sim 2 \times 10^3$ V/cm bias, a fast preamplifier, and a boxcar integrator (Princeton Applied Research 162-163). The gate width of the boxcar integrator was 50 ns. The trigger signal was provided by a fast photodiode as shown in the diagram. dc current caused by the bias illumination was measured by a Hewlett Packard 4140B pA meter/dc-voltage source. The sample was mounted in vacuum (inside a Janis 8DT optical cryostat). Data was recorded with a computer.

The measurements were performed on an undoped a-Si:H film. The sample, approximately 1 μ m thick, was prepared by plasma-assisted chemical-vapor deposition



FIG. 1. Schematic diagram of the experimental setup. $0.3-\mu s$ pulses are produced by reflecting a He-Ne laser beam from a rotating mirror. Transient photocurrent is detected by a boxcar integrator. The bias illumination is provided by another He-Ne laser also shown in this diagram. Neutral density (ND) filters are used to adjust the bias level.

(PACVD) on a glass substrate (Corning 7059) at a temperature of 260°C, pressure of 0.4 torr, a flow rate of 3 sc cm for SiH₄, and 30 sc cm for argon. The electrodes, evaporated aluminum film, were separated by approximately 1.5 mm.

The transient photocurrent with and without bias illumination are shown in Fig. 2. As shown in this diagram, a bias illumination may increase the transient current by a factor of more than 10 and will also change the decay rate of the transient current. Note that significant enhancement in the transient current occurs at a surprisingly low-bias illumination level of approximately 2 mW/cm².

In Fig. 3 we plotted the photocurrent on a logrithmic scale at several levels of bias illumination versus time. As shown in the diagram straight lines fit the decay curves well except at small t. Shortly after the pulse is turned off, carriers left at levels higher than the effective Fermi level E_{ft} are still going through the process of immobilization to the quasi-steady-state. The decay rate of the current immediately after the probe pulse is turned off is therefore expected to be faster than that rate at a later time. Figure 3 also indicates that the rate of decay of the transient current is sensitive to the level of bias illumination. With the level of optical-bias illumination increased from 1 mW/cm² (curve d) to 200 mW/cm² (curve a), the decay rate increases by a factor of 10. Note that the scale in transient photocurrent (y axis) in Fig. 3 is not normalized for different curves.

It is interesting to compare our data to that reported by Ritter, Zeldov, and Weiser.¹⁹ The data shown in Fig. 3 can also be fitted by a straight line with a time constant of approximately 1.6 μ s (at a bias level of 30 mW/cm², curve b) for a time span of 5 μ s immediately after the probe beam is turned off. This is in good agreement with their value of 1.8 μ s.¹⁹ It should be pointed out that the decay time constant studied in this work is measured



FIG. 2. Transient photocurrents at bias illumination levels: a, 100 mW/cm²; b, 15 mW/cm²; c, 2 mW/cm²; c with no bias illumination.

FIG. 3. Intensity of the transient photocurrent measured as a function of time at the optical-bias illumination equal to (a) 200 mW/cm², (b) 30 mW/cm², (c) 7 mW/cm², and (d) 1 mW/cm².

several μ s after the probe pulse is turned off. It is not the "free-electron lifetime" measured by Ritter, Zeldov, and Weiser.

III. DISCUSSION OF RESULTS

Optical-bias enhancement of the transient photocurrent may be understood with the multiple-trapping (MT) model. According to the multiple-trapping model for carrier kinetics,^{7,8,11} free carriers generated by a light pulse rapidly condense into the traps with a distribution that runs parallel to the distribution of traps. Trapped electrons can be thermally released with an immobilization time given by $t = v^{-1} \exp[(E_c - E)/kT]$, where v $(\sim 10^{12} \text{ Hz})$ (Ref. 7) is the attempt-to-escape rate, $E_c - E$ is the depth of electron traps below the conduction-band mobility edge E_c , and kT is the thermal energy. At time t_0 ($\approx 0.6 \ \mu s$) which is the experimentally observed rise time for the transient signal (shown in Fig. 2), the bulk of the trapped electrons produced by the probe pulse is concentrated in the vicinity of the demarcation level,^{8,9} E_d , when the photocurrent is observed to have its maximum value. Here

$$E_c - E_d = kT \ln(vt_0) . \tag{1}$$

As shown in Fig. 4 the bias illumination will raise the "effective Fermi level" for trapped electrons E_{ft} from the dark Fermi level E_{ft0} . If the bias illumination is sufficiently intense to raise E_{ft} to a level higher than E_d , as shown in Fig. 4(b), then the bulk of the charge carriers due to the probe pulse must now be concentrated in the vicinity of E_{ft} . With the electron emission rate from the trap given by²⁰

$$e_n(E) = v \exp[-(E_c - E)/kT],$$
 (2)

the number of released electrons from the trap increased as E was raised from E_d to E_{fi} . As a result, the "drift mobility" of the electrons responsible for the transient photocurrent is increased and a corresponding enhancement in the transient photocurrent is observed. In this work enhancement of the transient photocurrent as well as the decay rate of the transient current are studied with the MT model.

A. Enhancement of the transient photocurrent

In order to explain the enhancement in transient photocurrent let us first consider the situation with no bias illumination. In this case the multiple-trapping model predicts that the bulk of electrons produced by an extremely short pulse concentrates in the vicinity of E_d

FIG. 4. Schematic diagram of the process of optical-biasenhanced transient photoconductivity in *a*-Si:H. (a) After being illuminated by a shot pulse at t=0, the multiple-trapping theory predicts that the bulk of the charge in *a*-Si:H is concentrated in the vicinity of $E_d(t)$ at time *t* if there is no optical-bias illumination. (b) With optical-bias illumination the quasi-Fermi-level for the trapped electron will be raised to E_{ft} from the dark Fermi level. If E_{ft} is above E_d , charge due to the light pulse must concentrate in the vicinity of E_{ft} . Since the probability of electron emission from the trap increases as the energy of these electrons is increased from E_d to E_{ft} , the transient photocurrent is enhanced.

 $\equiv E_d(t_0) = E_c - kT \ln(vt_0)$ at $t = t_0$. Since the most probable number of release events for states above E_d is greater than 1 at t_0 it is reasonable to make the approximation that, at $t = t_0$, electrons above E_d are thermalized and have a Boltzmann distribution. Similarly, states deeper than E_d are unlikely to thermally release an electron at $t = t_0$, and electron distribution in these states remains frozen into the initial form that parallels the den-

sity of traps.⁸ If we assume the DOS in the tail states is of the form

$$g_t(E) = (N_L / kT_c) \exp[-(E_c - E) / kT_c],$$
 (3)

here N_L is the total conduction-band-tail state density and kT_c is the conduction-band-tail slope, the resulting distribution of electrons is thus given by

$$n_{t}(E) = \begin{cases} \beta(N_{L}/kT_{c}) \exp[-(E_{c}-E_{d})/kT] \exp[(E_{c}-E)(1-\alpha)/kT] & (E_{d} < E < E_{c}) \\ \beta(N_{L}/kT_{c}) \exp[-(E_{c}-E)/kT_{c}] & (E < E_{d}) \end{cases}.$$
(4)

Here $\alpha = T/T_c$. The constant β is determined by the normalization condition that the total number density of the trapped electrons

$$N_t = \int_{E_{ft}}^{E_c} n_t(E) dE \tag{5}$$

is a constant. For a pulse of finite duration, distribution of the generated charge carrier trapped in the band-gap states can be obtained from Eqs. (4) and (5) by integrating the charge distribution function over the duration of the pulse.

Optical-bias illumination will raise the value of the effective Fermi level for the trapped electron E_{ft} . For simplicity, we can assume that all of the levels below E_{ft} are occupied and are not accessible to electrons injected by the probe pulse. Since the capture cross section for states above E_{ft} is independent of E_{ft} , we can also assume that the distribution of charge carriers $n_t(E)$ (for an extremely short pulse) above E_{ft} has the form given above. Two cases should be considered here.

Case I. $E_{ft} < E_d$ [shown in Fig. 4(a)]. In this case

$$n_{t}(E) = \begin{cases} \beta'(N_{L}/kT_{c})\exp[(E_{c}-E)(1-\alpha)/kT]\exp[-(E_{c}-E_{d})/kT] & (E_{d} < E < E_{c}) \\ \beta'(N_{L}/kT_{c})\exp[-(E_{c}-E)/kT_{c}] & (E_{ft} < E < E_{d}) \\ 0 & (E < E_{ft}). \end{cases}$$
(6)

With the normalization condition given in Eq. (5) the normalization constant β' is found to be

$$\beta' = \frac{N_t}{N_L} \left[e^{-(E_c - E_d)/kT_c} - e^{-(E_c - E_f)/kT_c} - \frac{\alpha}{1 - \alpha} e^{-(E_c - E_d)/kT} (1 - e^{(1 - \alpha)(E_c - E_d)/kT}) \right]^{-1}.$$
(7)

Case II. $E_{ft} > E_d$ [shown in Fig. 4(b)]. In this case

$$n_t(E) = \begin{cases} \beta''(N_L/kT_c) \exp[(E_c - E)(1 - \alpha)/kT] \exp[-(E_c - E_d)/kT] & (E_{ft} < E < E_c) \\ 0 & (E < E_{ft}). \end{cases}$$
(8)

With the normalization condition $\int_{E_t}^{E_c} n_t(E) dE = N_t$, the normalization constant β'' is found to be

$$\beta^{\prime\prime} = \left[\frac{1-\alpha}{\alpha}\right] \left[\frac{N_t}{N_L}\right] (e^{(1-\alpha)(E_c - E_{ft})/kT} - 1)^{-1} .$$
(9)

Transient photocurrent at $t = t_0$ can be derived from the relation

$$I_p \sim \int_{E_{fl}}^{E_c} n_l(E) e(E) dE \quad .$$

Here e(E) is given by Eq. (2). Write $I_p = \Gamma \int_{E_{ft}}^{E_c} n_t(E) e(E) dE$ where the proportional constant is Γ . Using Eqs. (6)–(9), one has for case I $(E_{ft} < E_d)$

$$I_{p} = \beta' \frac{N_{L}}{kT_{c}} \nu \Gamma \left[e^{-(E_{c} - E_{d})/kT} \int_{E_{d}}^{E_{c}} e^{-(E_{c} - E)/kT_{c}} dE + \int_{E_{fl}}^{E_{d}} e^{-(1+\alpha)(E_{c} - E)/kT} dE \right]$$

$$= \Gamma \nu N_{t} \frac{e^{-(E_{c} - E_{d})/kT} (1 - e^{-(E_{c} - E_{d})/kT}) + \frac{\alpha}{1+\alpha} (e^{-(1+\alpha)(E_{c} - E_{d})/kT} - e^{-(1+\alpha)(E_{c} - E_{fl})/kT})}{e^{-(E_{c} - E_{d})/kT_{c}} - e^{-(E_{c} - E_{fl})/kT_{c}} - \frac{\alpha}{1-\alpha} e^{-(E_{c} - E_{d})/kT} (1 - e^{(1-\alpha)(E_{c} - E_{d})/kT})}, \qquad (10)$$

for case II $(E_{ft} > E_d)$

$$I_{p} = \beta'' \frac{N_{L}}{kT_{c}} \Gamma e^{-(E_{c} - E_{d})/kT} \int_{E_{ft}}^{E_{c}} e^{-\alpha(E_{c} - E)/kT} dE$$
$$= \left(\frac{1 - \alpha}{\alpha}\right) \Gamma \nu N_{t} \frac{1 - e^{-(E_{c} - E_{ft})/kT_{c}}}{e^{(1 - \alpha)(E_{c} - E_{ft})/kT} - 1} .$$
(11)

The above results show that I_p at $t = t_0$ will first increase slowly as E_{ft} is increased until its value is increased by a factor of approximately $1/\alpha$ when $E_{ft} = E_d$. The transient photocurrent I_p will then increase faster (approximately exponentially with a rate of $\exp[-(E_c - E_{ft})(1-\alpha)/kT]$) as E_{ft} is further increased.

For a pulse of duration T_0 , the transient current generated I_p can be calculated by integrating Eqs. (10) and (11) over the pulse duration T_0 . In our experiment the pulse can be approximated by a square wave. The integrated results for I_p plotted as a function of E_{fi} with the value $\alpha = 0.77, 0.72, 0.65, 0.57, and 0.52$ are shown in Fig. 5. Experimental data with the value of the peak of the transient current measured as a function of $kT \ln(i_b/i_{b0})$ (i_b is the photocurrent due to the bias illumination, i_{b0} is an adjustable parameter) are also shown in this diagram. Because the quasi-Fermi level for free electrons in the conduction band E_{fn} is defined by

$$n = N_c \exp[(E_{fn} - E_c)/kT],$$
 (12)

and the quasi-Fermi-level for the trapped electron is defined by

$$n + Rp = N_c \exp[(E_{ft} - E_c)/kt],$$
 (13)

(here R is the ratio of the electron and hole capture cross

sections and p is the hole number density), the quasi-Fermi-level for trapped electrons is always positioned above (but may be close to) the quasi-Fermi-level for free electrons. The value of $kT \ln(i_b/i_{b0})$ is therefore a rough measurement of E_{ft} . For the sample used, i_b is found to be proportional to $F_b^{0.9}$ (F_b is the optical bias level). The experimental data plotted with E_{ft} given by $0.9kT \ln(F_b/F_{b0})$ is shown on Fig. 5 and is compared with the theoretical calculation to give a rough estimate of the value of α . The data fits very well to the curve calculated with $\alpha = 0.65$.

The value of α can also be determined from a measurement of the dependence of the exponential decay rate of the transient photocurrent τ on the bias illumination level F_b . As shown in Fig. 6 τ varies as $F_b^{-0.38}$ over the whole range of the bias illumination level studied. When the optical-bias level is changed by a small amount ΔF_b the change in the number density of the trapped electron ΔN_t can be written as

$$\Delta N_t \propto \Delta (F_b \tau) \propto \Delta (F_b^{0.62}) \propto 0.62 F_b^{-0.38} \Delta F_b \quad . \tag{14}$$

The corresponding change in E_{ft} is determined from the enhancement in the transient photocurrent I_p . With $I_p \sim \exp[-(E_c - E_{ft})(1-\alpha)/kT]$, one has

$$\Delta E_{ft} = \frac{kT}{1-\alpha} \Delta (\ln I_p) = \frac{kT}{1-\alpha} \frac{\Delta I_p}{I_p} .$$
 (15)

At a high level of bias illumination I_p varies as F_b^m (*m* is a constant). As a result, one has

FIG. 5. Calculated transient photocurrent is plotted as a function of the optical-bias illumination level with the value of α equal to (a) 0.52, (b) 0.57, (c) 0.65, (d) 0.72, and (e) 0.77. The experimentally measured value of the transient photocurrent is also shown in this diagram.

FIG. 6. The exponential-decay time constant of the transient photocurrent is plotted as a function of the level of the opticalbias illumination level. In a logarithmic scale, the data can be fitted with a straight line of slope -0.38, indicating that monomolecular recombination is the main recombination process over the whole bias illumination range studied.

$$\Delta E_{ft} = \frac{mkT}{1-\alpha} \frac{\Delta F_b}{F_b} . \tag{16}$$

With Eqs. (14) and (16), one has

$$g(E_{ft}) = dN_t(E_{ft})/dE$$

$$\approx F_b^{0.62}(1-\alpha)$$

$$= \text{const} \exp[-0.69(E_s - E_{ft})/kT]. \quad (17)$$

In Eq. (17) we have used the relation that $E_{ft} = \text{const} + 0.9kT \ln F$. The value of α , 0.69, agrees reasonably well to the estimated value of 0.65 derived by the curve fitting of the data obtained in the I_t versus $E_{ft} = \text{const} + 0.9kT \ln F_b$ diagram shown in Fig. 5. The value of $\alpha = 0.69$ is somewhat lower than the value reported of approximately $0.8^{11,21}$ However, since the value measured in this work is the value of α at E in the vicinity of 0.25 eV, which is shallower than the region measured in the reported work, there is no contradiction.

With $\alpha = 0.69$ the DOS can be derived from the enhancement curve shown in Fig. 5. The number density of electrons trapped in the tail states for various bias illumination F_b is given by

$$N_b = f_b \tau \tag{18}$$

and

$$f_b = \Phi F_b (1-R) [1-\exp(-ad)]/d$$
 (19)

Here f_b is the electron-hole generation rate (cm⁻³s⁻¹). Φ is the quantum efficiency of electron-hole pair generation by the incident photon which can be approximated by unity. F_b is the incident photon flux of the bias illumination, R is the sample surface reflectivity, a = 7000 cm^{-1} at wavelength 6328 Å (Ref. 22) is the opticalabsorption coefficient and d is the film thickness. The "trapped-electron lifetime" τ is determined from the decay rate of the transient current shown in Fig. 3. With the corresponding change in the quasi-Fermi-level determined from the enhancement curve shown in Fig. 5, the density of state $g(E) = dN_b/dE_{ft}$ can be determined directly. The value of the DOS, g(E), plotted in Fig. 7, seems to be consistently lower than the value reported by Spear,²³ Lang,²⁴ and Archibald.²⁵ However, this calculation is based on the assumption that $v=10^{12}$ s⁻¹. An increase in the value of v by a factor of 10 will cause a corresponding change of approximately 0.06 eV in the value of *E*.

The number density of charge carrier produced by the probe pulse is given by

$$n_p = f_p T_0 \tag{20}$$

and

$$f_p = \Phi F_p (1 - R) [1 - \exp(ad)] / d .$$
 (21)

Here F_p is the probe beam photon flux, and T_0 , the pulse duration, is approximately 0.3 μ s. With the probe beam intensity of 30 mW/cm² and the bias beam intensity in the range of 0.2-200 mW/cm², n_p is $10^{-1}-10^{-2}$ of n_b . Note that in previous works on transient photocurrent or

FIG. 7. The value of the density of state determined in this work with the assumption that $v=10^{12}$ /sec is shown in curve d. As a comparison, estimates for N(E) from Spear et al. (shown as line a), Lang et al. (line b), and Archibald et al. (line c) are also shown in this diagram. This diagram seems to indicate that the correct value of v should be approximately 10^{13} /s.

photoinduced absorption experiment with bias illumination the number density of charge carriers created by each pulse (typically 50 μ J/pulse),¹⁵ n_p , is approximately 10^2n_b , and the system studied is not in a quasi-steadystate.

B. Physical interpretation of the decay rate of the transient photocurrent

The system studied in this work is in a quasi-steadystate with small perturbation. The bulk of the charges injected by the pulse is concentrated in the vicinity of the effective Fermi level E_{ft} shortly after the pulse was turned off and cannot immobilize into deeper levels. As a result, the transient current must decrease only through recombination with a constant rate determined mainly by the bias illumination level, the temperature, and the properties of the material. In this work a theory based on the assumption of a Fermi-Dirac function distribution of the trapped electrons²⁰ under a bias illumination is applied to interpret the experimental data.

Theoretical calculations of the exponential-decay time constant have been presented by Zeldov and Weiser¹⁴ and by Pandya and Schiff.²⁶ In this work we will use a somewhat different approach to calculate the decay time constant. This approach will enable us to interpret the time constant τ observed in our work as the lifetime of the trapped electrons in the vicinity of E_{ft} . The theory presented will show that τ is determined mainly by the rate of emission of electrons trapped in the vicinity of E_{ft} times the rate of recombination of charge carriers in the conduction band.

With the standard multiple-trapping theory the rate equations for free- and trapped-electron densities can be written as follows:

$$\frac{dn}{dt} = \sum_{i} [n_i e_i - n(N_i - n_i)\sigma_i] + G - R ,$$

$$\frac{dn_i}{dt} = n(N_i - n_i)\sigma_i - n_i e_i ,$$
(22)

where σ_i is the mobile-carrier capture coefficient of the *i*th trap, $e_i = e(E_i)$ the rate for thermal release of a mobile carrier from the *i*th trap, and N_i and n_i the total and occupied density of the *i*th trap. The photogeneration and recombination rates are denoted by G and R and the free-electron density is n.

Simmons and Taylor²⁰ solved the rate multipletrapping equations by using the detailed balance relations. The occupation probability for levels above the dark Fermi level is given by

$$f(E) = \frac{n}{n+Rp} (1+e^{(E-E_{ft})/kT})^{-1} .$$
 (23)

Here R is the ratio of the traps capture coefficient for the hole to that of the electron. Equation (23) describes the steady-state solution for continuous bias illumination. The response due to the small transient generation rate δG impulse is given by differenting Eq. (22) (Ref. 26),

$$\frac{d}{dt}(\delta n) = \delta G - \delta R$$
$$-\sum_{i} \left\{ \left[\delta n (N_{i} - n_{i})\sigma_{i} \right] - \delta n_{i} (e_{i} + n\sigma_{i}) \right\}, \quad (24)$$

$$\frac{d}{dt}(\delta n_i) = \delta n(N_i - n_i)\sigma_i - \delta n_i(n\sigma_i + e_i) , \qquad (25)$$

where δn and δn_i denote the photoexcited densities of the free and trapped electrons due to $\delta G(t)$, and δR is the corresponding change in the recombination rate. Rewriting the Fourier transforms of the above equations, one obtains

$$\delta n \left[i\omega + \sum_{i} (N_{i} - n_{i})\sigma_{i} \right]$$

= $\delta G - \delta R + \sum_{i} \delta n_{i} (e_{i} + n\sigma_{i}) , \quad (26)$

$$\delta n_i [i\omega + n\sigma_i + e_i] = \delta n (N_i - n_i)\sigma_i . \qquad (27)$$

For simplicity the same symbols are used for the timedependent functions and their Fourier transforms in the above formulas. After eliminating δn_i from Eqs. (26) and (27) one finally has

$$\delta n \left[i\omega + \int_{-\infty}^{E_c} dE g(E) [1 - f(E, E_{f,n})] \times \frac{i\omega\sigma(E)}{i\omega + n\sigma(E) + e(E)} \right]$$
$$= \delta G - \delta R \quad . \tag{28}$$

Here it is assumed that all traps at energy E are identical and the traps are continuously distributed. To obtain the long-time behavior of the transient photocurrent, we make use of the Fourier integral theorem.^{27,28} This theorem states that if a function is analytic in a strip of the complex plane parallel to and including the real axis, then the Fourier-transformed function will show an exponential decay at a long time with a time constant determined by the bounds of the analytic strip. To obtain the exponential-decay time constant we define $\tau_0^{-1} = \delta R / \delta n$ and write

$$\delta n / \delta G = \left[i\omega + \sigma_t \int_{-\infty}^{E_c} dE g(E) [1 - f(E, E_{ft})] \times \frac{i\omega}{i\omega + n\sigma_t + e(E)} + \tau_0^{-1} \right]^{-1}.$$
(29)

Let $\omega = i\eta$ be the first zero of the bracketed expression in Eq. (29). $1/\eta$ ($=\tau$) is then the exponential-decay constant of $\delta n / \delta G$. Assume the trap's mobile-carrier capture coefficient $\sigma(E) = \sigma_t$ is energy independent and with g(E) given in Eq. (3), it is necessary to evaluate the integration

$$\int_{-\infty}^{E_{c}} dE \frac{e^{(E-E_{f_{l}})/kT}}{1+e^{(E-E_{f_{l}})/kT}} \frac{e^{(E-E_{c})/kT_{c}}}{i\omega + n\sigma_{l} + \nu e^{(E-E_{c})/kT}} .$$
 (30)

Equation (29) is obtained by Pandya and Schiff.²⁶ In the following we will demonstrate that contribution to the integration comes mainly from electrons trapped in the vicinity of the quasi-Fermi-level E_{ft} . This result may then be used in the interpretation of the time constant measured in this work.

It is easy to see that the value of the integrand expressed above has a maximum value close to E_{ft} . Since the system studied in this work has reached a quasi-steady-state, the emission and capture rate for electrons must be equal under this circumstance [also remember $f(E_{ft})=\frac{1}{2}$], or

$$n\sigma_t = v \exp[(E_{ft} - E_c)/kT] = e(E_{ft})$$
. (31)

With e(E) increasing fast as E is increased from E_{ft} and [1-f(E)] decreasing fast as E is decreased from E_{ft} , it is clear that the integrand decreases fast (exponentially) as E deviates from a value close to E_{ft} . We may define a function F(x) by

$$F(x) = \frac{e^{(x-E_{ft})/kT}}{1+e^{(x-E_{ft})/kT}} \frac{e^{(x-E_c)/kT_c}}{i\omega + n\sigma_t + ve^{(x-E_c)/kT}} .$$
 (32)

To find the maximum value of F(x), set dF(x)/dx = 0 at $x = x_m$. If $\omega \ll n\sigma_t$ one has

$$\exp[(x_m - E_{ft})/kT] = (1+\alpha)/(1-\alpha) = A .$$
 (33)

Here $\alpha = T/T_c$.

As is expected, the maximum contribution to the integrand comes from regions close to $E = E_{ft}$. In order to give a physical interpretation of the decay time constant let us temporarily replace the integration by the maximum value of the integrand times an "effective width" ΔE . With the Fourier integral theorem and Eq. (29) we get (34)

$$\tau^{-1}g(E_m)[1-f(E_m,E_{ft})]\sigma_t\Delta E \approx \tau_0^{-1}[n\sigma_t+e(E_m)]$$

or

$$\tau^{-1}N_t \approx \tau_0^{-1}\sigma^{-1}e(E_m) = \tau_0^{-1}\sigma^{-1}v \exp[(E_{ft} - E_c)/kT].$$

 N_t in Eq. (34) is the total number of the trapped electrons. Equation (34) indicates that the exponential-decay constant τ is determined by the rate of recombination of free charges τ_0^{-1} and the rate of emission of electrons from band-tail levels in the vicinity of E_{fl} .

The integration in Eq. (29) can be calculated more accurately by expanding F(x) in a Taylor series in the vicinity of its maximum value x_m ,

$$F(x) \approx F(x_m) + \frac{1}{2} F''(x_m) (x - x_m)^2$$
 (35)

Since F(x) decreases fast as E deviates from the value x_m , we can write

$$F(x) \approx F(x_m) \exp\left[\frac{1}{2} \frac{F''(x_m)}{F(x_m)} (x - x_m)^2\right]$$
 (36)

Defining

$$B \equiv -F''(x_m)/2F(x_m)$$

= $\frac{1}{(kT)^2} [A/(1+A)^2 - \alpha/(1+A) + \alpha(1-\alpha)],$

and integrating from x = 0 to ∞ one obtains

$$\int_{-\infty}^{E_c} F(x) dx \approx \sqrt{\pi/B} F(x_m) .$$
(37)

As a result, the integration is determined mainly by $F(x_m)$, which is

$$F(x_m) = \frac{1}{1+A} \left[\frac{1}{1+A} + \alpha \right] \left[\frac{A}{v} \right]^{\alpha} (n\sigma_t)^{\alpha-1} .$$
 (38)

The exponential-decay constant η can then be determined from

$$\eta + \sigma_t \eta C (n\sigma_t)^{\alpha - 1} = \tau_0^{-1} . \tag{39}$$

Here C is a constant. With

$$n\sigma_t = e(E_m) = v \exp[(E_{ft} - E_c)/kT]$$

and

$$N_{t} = \int_{-\infty}^{E_{c}} dE \left[\frac{N_{L}}{kT_{c}} \right] \frac{e^{(E-E_{c})/kT_{c}}}{1+e^{(E-E_{ft})/kT}}$$
$$\simeq e^{-(E_{c}-E_{ft})/kT_{c}} \int_{-\infty}^{\infty} dE \frac{e^{(E-E_{ft})/kT_{c}}}{1+e^{(E-E_{ft})/kT}}$$
$$= \left[\frac{\pi}{\sin \alpha \pi} \right] (n\sigma_{t})^{\alpha} , \qquad (40)$$

it is easy to see that Eqs. (39) and (34) are equivalent (to a

constant of the order of 1) if $\eta \ (\equiv 1/\tau)$ is small compared to τ_0^{-1} . The charge recombination rate τ_0 in Eq. (39) is determined by the processes of recombination. Two cases will be treated here.

(1) Monomolecular recombination. The recombination rate depends linearly on n (electron in conduction band) but is independent of the trapped-carrier density. In this case $F_b \propto R = n/\tau_0 = \cos t\eta n^{\alpha-1}n = \cos t\eta n^{\alpha}$. The exponential decay time constant $(\tau = \eta^{-1})$ is found to vary as $F_b^{-(1-0.9\alpha)}$. With $\alpha = 0.69$, monomolecular recombination predicts τ varies as $F_b^{-0.38}$ in the sample studied in this work.

(2) Bimolecular recombination. In this case the rate of recombination is proportional to the number of trapped molecules (or $R = n\sigma_t N_t = n\sigma_t F_b \tau$, N_t is the trappedelectron density) and $\tau_0^{-1} = \sigma_t F_b \tau$. The exponentialdecay time constant τ is found to be proportional to $(F_b n^{1-\alpha})^{-1/2}$. With $\alpha = 0.69$, bimolecular recombination predicts τ varies as $F_b^{-0.68}$ in the sample studied.

The experimentally measured exponential-decay time constant τ of the transient photocurrent is shown in Fig. 6 as a function of the level of optical-bias illumination. This diagram indicates that τ varies as $F_b^{-0.38}$ over the whole range of the optical-bias illumination level studied. With no noticeable change in the slope observed in the τ versus F_b curve plotted in a logarithmic scale the experimental result seems to indicate that monomolecular recombination is the dominant recombination process in the region studied. The slope of the curve also agrees well with the prediction based on monomolecular recombination process and deviate significantly from that based on bimolecular recombination process.

IV. CONCLUSION

In conclusion we have observed optical-bias enhancement in the transient photocurrent in a-Si:H films and applied this effect to study the electric properties of the amorphous thin film. The transient photoconductivity is studied in a quasi-steady-state, and the experimental result can, therefore, be interpreted with little ambiguity. The density of the tail states g(E) is determined directly from this measurement. This analysis is also applied to study the rate of recombination of charge carriers in a quasi-steady-state as a function of the quasi-Fermi-level for the first time. It is found here that the transient photocurrent decays exponentially several microseconds after the probe pulse is turned off. The exponential-decay time constant is interpreted as the effective lifetime of electrons trapped at the quasi-Fermi-level E_{ft} . This time constant is determined by the rate of emission of electrons at E_{ft} and the free-electron lifetime τ_0 . It is also found that the exponential-decay rate of the transient current τ is sensitive to the optical-bias illumination level and may change by a factor of 14 when the optical-bias level is changed from 0.2 to 200 mW/cm². With τ varying as $F_b^{-0.38}$ over the whole bias illumination range studied, results of this experiment indicate that monomolecular recombination is the main process of recombination of charge carriers.

9660

- ¹J. M. Hvam and M. H. Brodsky, Phys. Rev. Lett. **46**, 371 (1981).
- ²R. Pandya and E. A. Schiff, J. Non-cryst. Solids **59-60**, 297 (1983).
- ³H. Oheda, Philos. Mag. B **52**, 857 (1985).
- ⁴C. Main, R. Russell, J. Berkin, and J. M. Marshall, Philos. Mag. 55, 189 (1987).
- ⁵H. Scher and E. W. Montroll, Phys. Rev. B 12, 2455 (1975).
- ⁶F. W. Schmidlin, Phys. Rev. B 16, 2362 (1977).
- ⁷T. Tiedje and A. Rose, Solid State Commun. **37**, 49 (1980).
- ⁸J. Orenstein and M. A. Kastner, Phys. Rev. Lett. **46**, 1421 (1981).
- ⁹T. Tiedje, J. M. Cebulka, D. L. Morel, and B. Abeles, Phys. Rev. Lett. 46, 1425 (1981).
- ¹⁰Z. Vardeny, P. O'Connor, S. Ray, and J. Tauc, Phys. Rev. Lett. 44, 1267 (1980).
- ¹¹J. Orenstein, M. A. Kastner, and V. Vaninov, Philos. Mag. B 46, 23 (1982).
- ¹²C. Tsang and R. A. Street, Phys. Rev. B **19**, 3027 (1979).
- ¹³E. A. Schiff, Phys. Rev. B 24, 6189 (1981).
- ¹⁴E. Zeldov and K. Weiser, Phys. Rev. Lett. 53, 1012 (1984).
- ¹⁵D. Pfost, Z. Vardeny, and J. Tauc, Phys. Rev. Lett. **52**, 376 (1984).

- ¹⁶K. A. Conrad, R. Pandya, and E. A. Schiff, Phys. Rev. Lett. 54, 247 (1985).
- ¹⁷R. Pandya, E. A. Schiff, and K. A. Conrad, J. Non-Cryst. Solids 66, 193 (1984).
- ¹⁸H. A. Stoddart, J. Tauc, and Z. Vardeny, Phys. Rev. Lett. 54, 248 (1985).
- ¹⁹D. Ritter, E. Zeldov, and K. Weiser, Phys. Rev. B 38, 8296 (1988).
- ²⁰J. G. Simmons and G. W. Taylor, Phys. Rev. B 4, 502 (1971).
- ²¹B. Gu and Z. Xu, Philos. Mag. B 55, 391 (1987).
- ²²H. Piller, Handbook of Optical Constants of Solids (Academic, New York, 1985).
- ²³W. E. Spear, and P. G. LeComber, Philos. Mag. B **33**, 935 (1976).
- ²⁴D. V. Lang, J. D. Cohen, and J. P. Harbison, Phys. Rev. B 25, 5285 (1982).
- ²⁵I. W. Archibald and R. A. Abram, Philos. Mag. B 56, 429 (1987).
- ²⁶R. Pandya and E. A. Schiff, Philos. Mag. B 52, 1075 (1985).
- ²⁷P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953).
- ²⁸E. C. Titchmarch, Introduction to the Theory of Fourier Integrals (Oxford University Press, New York, 1948).