Trap-controlled dispersive transport and exponential band tails in amorphous silicon

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The theory of trap-controlled dispersive transport based on exponential band-tail traps is extended to include the saturation of trap occupancy at high excitation levels. The absence of saturation effects in the transient photocurrent experiment of Hvam and Brodsky is shown to contradict the band-tail hypothesis of this model.

Several recent experiments on transient photocurrents[1-3] in amorphous semiconductors offer impressive evidence that the transients following impulse photoexcitation are describable by a simple model[2-4] of trap-controlled dispersive transport which invokes two remarkable assumptions. First, transport is due entirely to the fraction of excited carriers occupying electronic states above a mobility edge. Second, the states below the mobility edge (the traps) fall off exponentially with energy in the experimentally accessible region, which lies from ~ 0.2 to ~ 0.5 eV below the mobility edge. The proximity of this exponential region to the mobility edge strongly suggests that these trap states are the disorder-induced exponential tail of the conduction or valence bands, as has in fact been proposed for plasma-deposited hydrogenated amorphous silicon (a-Si:H).[1,2]

The model which these experiments support is linear in the excitation impulse strength. One consequence of a multiple-trapping approach which invokes band tail or other defect sites as the traps is the saturation of the trap occupancy which must occur in the absence of recombination. In the present work these saturation effects, as well as recombination, are included in a multiple-trapping model for dispersive transport. I find that the nonobservation of trap saturation effects in the experiment of Hvam and Brodsky[1] on a-Si:H cannot be accounted for by the proposed band-tail model. The question remains open as to whether a more sophisticated model invoking band-tail traps can surmount this difficulty, or whether a fundamentally different approach to the origin of the trapping effects must be found.

MULTIPLE-TRAPPING MODEL

The present work is a straightforward extension of the theory proposed by Tiedje and Rose[4] and by Orenstein and Kastner.[3] Saturation effects have also been treated independently in recent work by Orenstein and Kastner.[5] There are two fundamental assumptions in this theory regarding the distribution of excited carriers. First, at any time prior to their diminution by sweepout or recombination, the carrier distribution is determined entirely by a quasi-Fermi level $E_d(t)$

$$F(t)\int_{-\infty}^{\infty} dE\, g(E)\left[1+\exp\left(\frac{E-E_d}{kT}\right)\right]^{-1}=N \ , \quad (1)$$

where $g(E)$ is the electronic density of states for the traps and $F(t)$ is an occupancy factor which acts to conserve the excitation density N. Second, E_d evolves according to the expression

$$E_d(t)=-kT\ln(\nu t) \ , \quad (2)$$

where kT is the temperature in energy units and ν is an attempt-to-escape frequency related by detailed balance to the trap-capture radius. These are strong assumptions. They require that the capture cross sections of all the traps be constant and that detailed balance hold. For an exponential density of states below the mobility edge (note that E is referenced to this edge)

$$g(E)=g_0\exp(E/kT_0) \ . \quad (3)$$

We obtain for the occupancy factor and the time-dependent drift mobility[4]

$$F(t)=(N/kT_0g_0)\,\mathrm{sinc}(\alpha\pi)(\nu t)^{\alpha} \ , \quad (4)$$

$$\alpha\equiv T/T_0 \ , \quad (5)$$

$$\mathrm{sinc}(\alpha\pi)=\sin(\alpha\pi)/\alpha\pi \ ,$$

$$\mu(t)\equiv\mu_0\frac{n(t)}{N}=\mu_0\frac{F(t)N_c\exp(E_d/kT)}{N}$$

$$=\mu_0\frac{N_c}{kT_0g_0}\,\mathrm{sinc}(\alpha\pi)(\nu t)^{-1+\alpha} \ , \quad (6)$$

where N_c is the effective density of states at the mobility edge, n is the density of mobile carriers, and μ_0 is their mobility. The definition of the drift mobility in (6) is appropriate for photocurrent transient exper-

iments and is not identical with the use of this term for time-of-flight measurements.[1,2]

The implications of these results are shown graphically in Fig. 1. The occupancy function f is defined

$$f(E,E_d,t)=F(t)(1+\exp\{[E-E_d(t)]/kT\})^{-1}$$

and has two attributes. The quasi-Fermi level E_d obeys expression (2) and falls logarithmically with time; the scaling factor $F(t)$ increases with time to conserve the total excitation density N. The evolution of the energy distribution of the excitation is illustrated in the second part of the figure; this distribution is simply the product $f(E,E_d,t)g(E)$.

In the previous work on this theory it is implicitly assumed that $F(t)\ll 1$. Obviously this factor cannot exceed one, and thus a limit on the downward progress of $E_d(t)$ can be set from this condition

$$E_d > kT_0 \ln[N\,\mathrm{sinc}(\alpha\pi)/kT_0g_0] \quad . \tag{7}$$

The value of $E_d(t)$ predicted by (2) crosses this lower bound at a time t_1

$$\nu t_1=[N\,\mathrm{sinc}(\alpha\pi)/kT_0g_0]^{-1/\alpha} \quad , \tag{8}$$

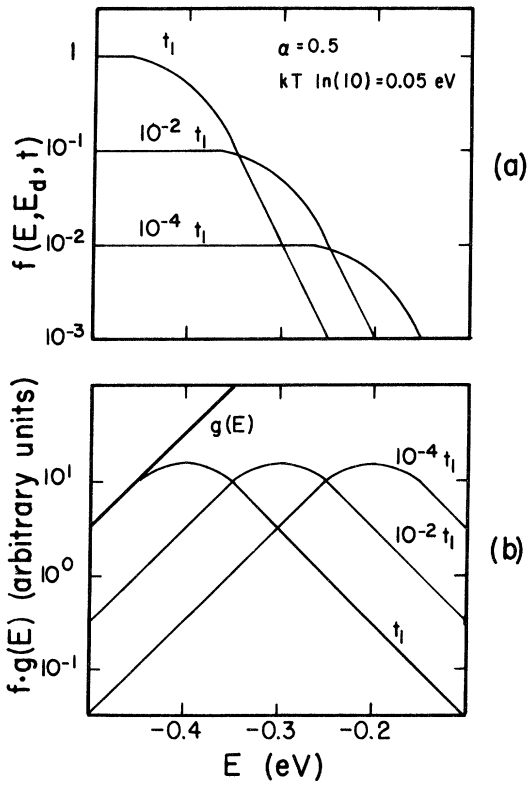

FIG. 1. (a) Occupation probability f at three times following impulse photoexcitation for an exponentially increasing density of traps $g(E)$. (b) Trap-occupation density at three times. At t_1 the traps below the quasi-Fermi level are completely filled.

t_1 is thus a measure of the thermalization time. The density of mobile carriers n after t_1 remains constant until recombination sets in

$$n=N_c(\nu t_1)^{-1}=N_c[N\,\mathrm{sinc}(\alpha\pi)/kT_0g_0]^{1/\alpha} \quad t\gg t_1 \quad . \tag{9}$$

The behavior of $n(t)$ including these saturation effects is shown in Fig. 2 for several excitation densities. It should be noted that the saturated value for n is nonlinear in the excitation density N.

Figure 2 also illustrates the behavior of $n(t)$ for long times at which "diffusion-limited bimolecular recombination" is assumed to occur.[6] This terminology means in the present context that recombination occurs by the capture of mobile carriers by a density of recombination centers equal to the total excitation density N (which is now assumed time dependent). Thus

$$N(t)=\int dE\, g(E)\left[1+\exp\left(\frac{E-E_d(t)}{kT}\right)\right]^{-1} \quad , \tag{10}$$

$$\frac{dN}{dt}=-bNn \quad . \tag{11}$$

The expression (10) assumes $F(t)=1$. Using (9) to relate n and N, the solution for n is

$$n(t)=\frac{N_c(\nu t_1)^{-1}}{1+(bN_c/\alpha\nu)(t/t_1)}, \quad t\gg t_1 \quad . \tag{12}$$

The continued use of a quasi-Fermi-level approach [implicit in (10)] is reasonable only because of the bi-

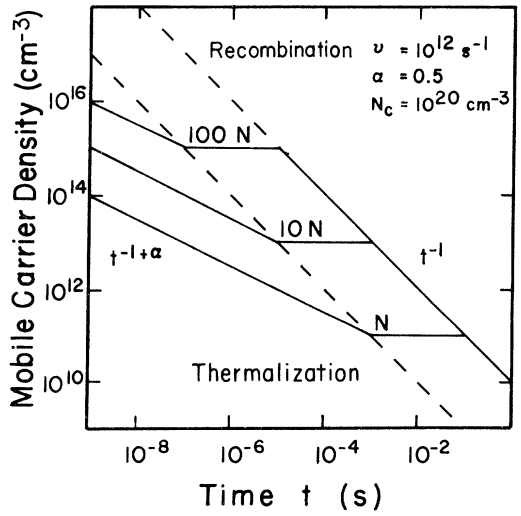

FIG. 2. Evolution of mobile carrier density following impulse photoexcitation for three excitation densities. The figure assumes trap saturation followed by diffusion-limited bimolecular recombination.

molecular assumption; if an excitation-density-independent (i.e., monomolecular) recombination rate is assumed the trap-emission-limited picture of Tiedje and Rose[4] must be used instead. However, for *a*-Si:H, the bimolecular assumption is consistent both with steady-state photoconductivity measurements[7] (which usually yield the well-known relation $n \propto F^{\gamma}$, where F is the optical flux and $0.5 < \gamma < 1$) as well as with photoinduced absorption measurements.[6] The assumption that thermalization occurs prior to recombination requires

$$bN_c/\alpha\nu << 1 \ .$$

Employing for b the estimate[5,6] 10^{-10} cm^3 s^{-1}, $N_c \sim 10^{20}$ cm^{-3}, and[2] $\nu \sim 10^{12}$ s^{-1} we obtain $bN_c/\alpha\nu \sim 10^{-2}$, which suggests that in *a*-Si:H saturation of the band-tail traps should occur prior to diffusion-limited bimolecular recombination. In *a*-As_2Se_3, on the other hand, recombination apparently sets in prior to saturation.[5]

DISCUSSION

In the experiment of Hvam and Brodsky absorbed photon densities as large as 10^{18} cm^{-3} were employed. For the most extensively studied specimen (doped 0.01% with PH_3 in the gas phase) the room-temperature transient follows the expected $t^{-1+\alpha}$ decay out to nearly 10^{-3} s; the slopes of the transients for this specimen have the dependence on temperature predicted by the multiple-trapping model.

These authors also studied the dependence of the transients on the excitation density; although these studies were only sketchily reported, they were apparently consistent with the linear dependence of the transient currents on excitation density required by the multiple-trapping model for the transient. A linear dependence on excitation density also demonstrates that fast intensity-dependent recombination effects, which might tend to reduce the effective excitation density N below the absorbed photon density, were negligible up to 10^{18} cm^{-3}. Subsequent work by the same authors[8] has shown that for excitation densities greater than 10^{18} cm^{-3} such a process may in fact occur.

These experiments indicate that this specimen has the properties expected from a multiple-trapping model for the transient, and that the effects of trap saturation were not observed out to 10^{-3} s, after which the onset of recombination would prevent their observation. This value of 10^{-3} s can be used as follows to estimate the prefactor g_0 of the exponential trap density. First, we neglect the effects of any nonunity quantum efficiency (QE) for carrier generation and thus equate the absorbed photon flux with the excitation density N. One of the lowest measured values of the QE in *a*-Si:H is 0.4 at room temperature,[9] which would not influence the present discussion. Second, we employ for ν the value of 10^{12} s^{-1} obtained by Tiedje *et al.*[2] in their time-of-flight (TOF) experiments on *a*-Si:H. This choice will be more fully discussed later. Using the experimental value at room temperature for this specimen of $\alpha = 0.6$ (and thus $kT_0 = 40$ meV) we obtain [cf. (8)]

$$g_0 = (N/kT_0)\,\mathrm{sinc}(\alpha\pi)\,(\nu t_1)^{\alpha} > 3 \times 10^{24}\ \mathrm{cm}^{-3}\mathrm{eV}^{-1} \ .$$

This value of g_0 is much too large to permit continuity between the exponentially distributed traps and (in this case) the conduction-band density of states ($g < 3 \times 10^{22}$ cm^{-3}eV^{-1}). Interestingly, such continuity was explicitly assumed by Tiedje *et al.*[2] in order to extract μ_0 from their TOF experiments.

It may appear that the value of g_0 simply implies that the mobility edge lies above the exponential tail region, and that the direct consequences of this nonexponential region on the photocurrent transient (a decay more rapid than $t^{-1+\alpha}$) appeared at times too short to have been observed in this experiment. However, this approach can be ruled out because it requires an unrealistically high value of μ_0 to compensate for the low value of N_c relative to the tail-state density. Solving (6) for μ_0 and employing the estimates $N_c \sim 10^{20}$ cm^{-3} and $g_0 = 3 \times 10^{24}$ cm^{-3}eV^{-1} a value for $\mu_0 \sim 2.5 \times 10^4$ cm^2/V s is obtained from Hvam and Brodsky's data for $\mu(t)$ at room temperature. Such a value is much too large for an amorphous semiconductor. A second argument against this noncontinuity hypothesis is that its consequences are not observed in picosecond photocurrent transients,[10] which, however, were performed on very different specimens than Hvam and Brodsky's.

The remaining arguable estimate used in estimating g_0 is the magnitude of ν. The estimate employed here is based on time-of-flight measurements in undoped *a*-Si:H as a function of temperature,[2] which yield ν unambiguously as a fitting parameter. Moreover an estimate of $\nu > 10^{12}$ s^{-1} is consistent with picosecond photocurrent transient data on very different specimens,[10] which do not show the expected effects for smaller ν. The difficulties with a large g_0 would be significantly ameliorated only if $\nu < 3 \times 10^8$ s^{-1} for Hvam and Brodsky's specimen, which would be very surprising if all these experiments are to be interpreted in terms of band-tail traps.

In principle time-of-flight measurements (used here to estimate ν) and photocurrent transient measurements should yield similar information, and for *a*-Si:H (Refs. 1 and 2) the similar behavior of the transients with time and temperature suggests that the two techniques are in fact observing the same phenomena. Unfortunately time-of-flight measurements cannot be extended to high excitation densities without introducing known complications due to space-charge effects on the drift field. Any criticism

of the intepretation of the TOF measurements based on trap saturation effects must thus rely on the link between the TOF measurements and the direct photocurrent transient measurements. Such a link has proven elusive for chalcogenides,[3] and needs to be better established in *a*-Si:H.

In conlcusion, photocurrent transient data of Hvam and Brodsky are inconsistent with a complete interpretation based on conduction band-tail states acting as traps, despite the partial success of this model in accounting for the form, temperature dependence and intensity dependence of the transients. It seems unlikely that a band-tail model incorporating such refinements as a distribution of capture cross sections[11] would still account for the remarkably simple experimental results while avoiding the difficulties with trap saturation. A more plausible possibility would be that bimolecular recombination is occurring simultaneously with the multiple-trapping process,[5,8] but that the resulting transients nonetheless show the principal features expected from multiple trapping alone. A definitive evaluation of this possibility requires that the linearity of the current transient with excitation density be carefully checked; any significant nonlinearity of course requires modifications to a simple multiple-trapping description. If in fact the sole objection to the multiple-trapping model remains the nonobservation of trap saturation, a different speculation would be that some variation of a self-trapping picture might account for the multiple-trapping features without predicting saturation effects for defect traps.

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