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Hole trapping, light soaking, and secondary photocurrent transients in amorphous silicon

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We measured and analyzed the long-time photocurrent decays in films of undoped amorphous silicon with coplanar Ohmic contacts. We interpret these as a secondary-electron current which flows as long as holes are safely trapped in states 0.3 to 0.5 eV above the valence-band edge. A *new analysis* of these transients gives the distribution of the trapped holes and implicates thermally assisted tunneling to dangling bonds as the rate-limiting step in hole recombination. Photoconductive gain is lost and the decay time shortened after small amounts of light soaking. The energetically deeper hole traps with the longer residence times are lost first and in the same number as expected for the increase in dangling bonds with a similar light soaking; this result supports a model which has hole trapping in valence-band-tail states as a precursor to light-induced dangling-bond formation. Two possible origins of these traps are discussed.

INTRODUCTION

In the Background section we present a brief history of the observation of protracted photocurrent transients in hydrogenated amorphous silicon (a-Si:H) and discuss our reasons for believing that hole trapping is essential for explaining such results. The Experimental section is followed by a Model and Analysis section which explains the recombination process and derivation of the equation used to obtain the energy distribution of these safe hole traps (SHT). The Results section shows such energy distributions derived from measured current transients on a film in the annealed and two different light soaked states. A Discussion section follows in which two sources of SHT's are described along with an explanation of how they serve as a fundamental step in the photodegradation process.

BACKGROUND

Long photocurrent decay transients have been observed on specimens of a-Si:H with coplanar contacts for some time. Hvam and Brodsky[1] noted that photoconductivity and response times increase with phosphorus content in doped films. They assumed these increases were due to a filling of deep recombination centers with electrons, thereby increasing the recombination lifetime. Their use of multiple trapping exclusively in the conduction-band tail to model the electron transport was disputed by Schiff[2] for reasons which include a low attempt-to-escape frequency. More studies of decay transients followed focusing only on multiple trapping of electrons in the conduction-band tail and recombination at dangling bonds. To explain these long photocurrent decays Pandya *et al.*[3] added unusually shallow, doubly occupied dangling bonds to the exponential conduction-band-tail distribution. In a similar study Oheda[4] took the longer than expected response times and weaker than expected temperature dependences as evidence that the conventional view of the conduction-band tail is incorrect and that a well-defined band of states exists 0.2 eV below the band edge. These authors focus on electron trapping statistics, ignoring steps to recombination occurring below the dark Fermi level, E_F.

In this study of photocurrent decays in a-Si:H, we take the point of view that, because of charge neutrality restrictions, consideration of the trapping of the minority carriers below the dark Fermi level is essential for determining steady-state and transient photoconductivities. In fact, Vanier and Griffith[5] and then Persans[6] used dual-beam photoconductivity studies to infer the presence of two types of centers: (i) recombination centers, presumably threefold cooridnated Si "dangling bonds" with normal capture-rate coefficients for electrons to trapped holes, and (ii) sensitizing centers located well below midgap, with a capture-rate coefficient three to four orders of magnitude smaller. Such centers lead to the phenomenon of electronic doping and enhanced photocurrents proposed by Rose.[7]

McMahon and Xi[8,9] have used nonequilibrium steady-state statistics[10] to model dc photoconductivity (σ_{pc}) for undoped a-Si:H when trapping and recombination occur at two types of centers. They assume that positively correlated dangling-bond (DB) states are the recombination centers and exponential band-tail states with an energy-dependent capture-rate coefficient are the sensitizing centers and calculate the temperature dependence of σ_{pc}. A similar calculation was made by Gu *et al.*[11] using a Gaussian distribution for the sensitizing center. Later, McMahon and Xi[12] noted that the deepest valence-band-tail states have the lowest capture-rate coefficient for electrons to trapped holes ($\simeq 10^{-13}$ cm^3 sec^{-1}), and named them "safe hole traps."[13] Trapped holes reside for long times in the deep valence-band-tail states, safe from recombination, causing the commonly observed low-temperature peak in σ_{pc} versus temperature (T) and associated photoconductive gain. These authors proposed that those SHT's which have the longest hole residence times and originate from the most highly strained bonds break after light soaking into two DB's; they concluded

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that SHT's were a cause of the instability in a-Si:H.

Recently, Main *et al.*[14] pointed out that in phosphorus-doped a-Si:H, transport of electrons is essentially nondispersive and recombination of free carriers is controlled by the slow thermal release of trapped holes from valence-band-tail traps. Pickin *et al.*[15] suggested that the same recombination mechanism explains the intensity dependence of the decay time in undoped a-Si:H. In the present study, we use a similar mechanism to develop an expression based on the thermal release of holes trapped deep in the valence-band tail to analyze the secondary current transients. With this model, we can determine the energy spectrum of SHT populations giving rise to the long current transient in undoped a-Si:H. The energy depths and small values of attempt-to-escape frequency (ν) we find are consistent with the following process. Trapped holes are thermally excited out of deep, highly localized valence-band-tail states (SHT's) to shallower, more dense, less localized tail states where they tunnel[16,17] to dangling bonds and complete the recombination process. Our analysis shows that the number of SHT's lost during light soaking is of the same order as the number of light-induced DB's generated and that the deepest states in the distribution are lost first. This strengthens the arugment that SHT's are a source of the Staebler-Wronski effect.[18]

EXPERIMENTAL PROCEDURE

Current transients as a function of temperature were measured on a 2.1-μm-thick glow discharge a-Si:H film. This film is the unused portion of a sample used earlier for dc photocurrent experiments.[19] It is important to note that the analysis we develop is strictly valid only in the temperature range where "thermal quenching" dominates the behavior seen in the σ_{pc} versus $1/T$ curve. Such behavior is associated with a $\sigma_{pc}(T)$ that decreases with increasing T. For our sample this region lies between 160 and 200 K. Within this region the response time becomes exponentially longer and, therefore, σ_{pc} increasing with an observed activation energy of 0.050 eV. Outside this temperature range our analysis does not apply, since above 200 K the quasi-Fermi level has not reached the SHT's and below 160 K the emission times become so long that other recombination pathways, such as tunneling of holes to DB's (Ref. 15) and direct capture of electrons to occupied SHT's, become viable.

One hundred volts is applied across electrical contacts of painted colloidal graphite space 1 mm apart and 1 cm long. Such contacts exhibit Ohmic behavior at the photocurrents measured (≤ 200 nA). The current is amplified by a Keithley 485 picoammeter; the analogue output is sufficiently fast for the times (t) 0.005 sec $< t < 2.2$ sec used in our analysis. The picoammeter output is monitored with a PAR Model 162 boxcar averager after an established steady-state photocurrent is terminated with a shutter. Generally, 48 points were measured in a 2.2-sec interval spaced equally in $\log(t)$ for uniform energy resolution.

The analysis requires that steady-state photocurrents be established before the transient decay; we illuminated the sample through a shutter which remained open for several seconds with 10^{14} photons/cm^2 sec using a 10-nm band-pass filter centered at 630 nm. This flux yielded an adequate signal-to-noise ratio without causing undue photodegradation, even at the lowest measurement temperatures. The sample remained in a turbo molecular-pumped vacuum system during all phases of the experiment. Each light-soaking exposure was 20 min long at a flux level of 7.5×10^{15} photons/cm^2 sec at 630 nm.

MODEL AND ANALYSIS

Our analysis uses the recombination paths shown in Fig. 1, which have as steps, processes common to other recombination models (see, for example, Refs. 14–17, 20 and 21); our addition is to recognize that the step that limits the recombination rate at long times is the thermal excitation of holes $[H_{vb}(t)]$ trapped in deep, isolated states to more delocalized, shallower valence-band-tail states at E_{tnl}. We assume that the principle recombination center is the positively correlated DB. The D^+/D^0 level represents the energy of the first electron to occupy a neutral DB and the D^0/D^- level is the energy location of the doubly occupied DB. Electrons cannot recombine at DB's until holes trapped deep in the valence-band tail are thermally excited to shallower tail states at E_{tnl} through which they tunnel to the deeper, midgap DB-recombination centers. It is implicit in the analysis that SHT's do not act as recombination centers as do the midgap DB's: reasons for the small capture rate constant of SHT's are left until the Discussion section. This model allows us to determine the energy distribution of the SHT's from the time dependence of their emission to E_{tnl}.

We assume first that the steady-state photocurrent and subsequent current transient after the light is shuttered are due to a time-dependent free-electron population $[n(t)]$ and a *time-independent* extended-state mobility (μ_0) and write

$$I(t)=n(t)e\mu_0FA \ , \tag{1}$$

where e is the electronic charge, F the applied voltage divided by the distance between the coplanar (0.5 mm), and A the absorption length of 630 nm light times the contact length $(0.5\times10^{-4}\ \text{cm}\times1\ \text{cm})$. Since in our experiment, steady-state quasiequilibrium is established prior to shuttering the light at $t=0$, the electron quasi-Fermi level $[E_{Fn}(t)]$ can be determined from[7]

$$n(t)=N_0e\frac{E_{Fn}(t)-E_c}{kT} \ , \tag{2}$$

where N_0, the effective density of states at the conduction-band mobility edge (E_c), is taken to be $N_ckT=7\times10^{19}\ \text{cm}^{-3}$ at 163 K. N_c, the density of states at E_c, is taken to be $5\times10^{21}\ \text{cm}^{-3}\,\text{eV}^{-1}$.[22] Further, we assumed that electrons remain in equilibrium in order that Eq. (2) apply for all $t\geq0$. The number of electrons trapped in the conduction-band tail and in thermal equilibrium with the free electrons is

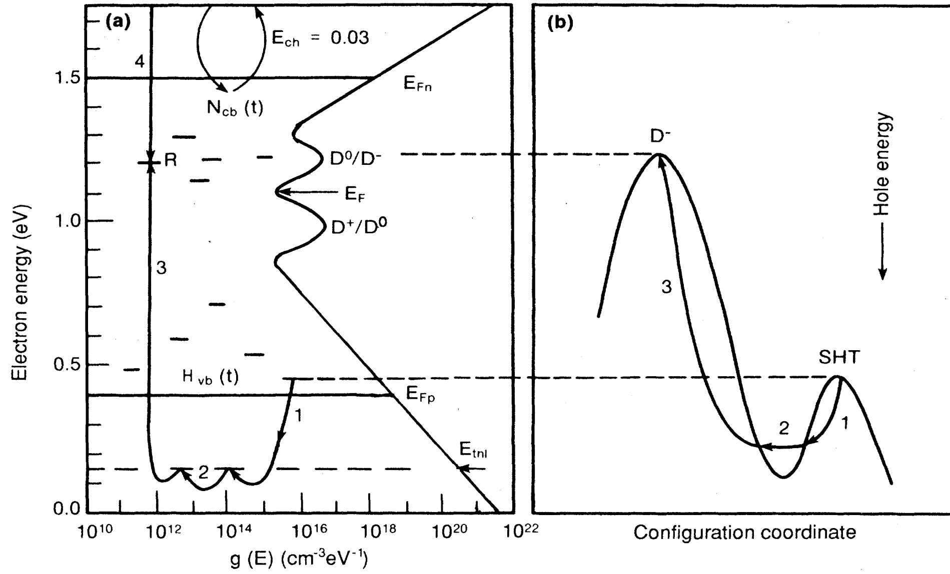


FIG. 1. (a) Density-of-states diagram showing the defects used in the recombination model. The recombination pathway includes step 1, the thermal emission of a hole out of a SHT to the shallower valence-band-tail states. Step 2 is the tunneling process at $E_{\rm tnl}$ by which the hole diffuses to the D^- state. Steps 3 and 4 show the recombination R of an electron and hole at a DB. (b) depicts the potential-energy-configuration coordinate diagram. Steps are the same as for (a).

$$N_{\rm cb}(t)=N_c f\int_0^{E_{Fn}(t)} e^{+[(E-E_c)/E_{\rm ch}]}dE$$
$$=N_c f E_{\rm ch} e^{+\{[(E_{Fn}(t)-E_c)/E_{\rm ch}]\}} , \quad (3)$$

where $E_{\rm ch}$ is the characteristic energy of the exponential conduction-band tail distribution, and f, the value of the modulated Fermi function,[10] is one due to the small occupied-to-unoccupied capture-rate constant ratio.

The generation rate, $G \cong 10^{18}$ cm^{-3}sec^{-1}, is large enough so that at the temperatures investigated, conduction-band and valence-band-tail populations are greater than populations trapped at midgap dangling-bond states. The free charge is small compared with the amount of charge in any of the traps, so that the condition of charge neutrality is simply

$$H_{\rm vb}(t)=N_{\rm cb}(t)+N_{\rm DB} , \quad (4)$$

where $N_{\rm DB}$, the net negative charge trapped at dangling bonds, is independent of t when compared to the relatively fast changes in $N_{\rm cb}(t)$ or $H_{\rm vb}(t)$.

Combining Eqs. (2)–(4) gives us an expression for the free-electron population in terms of the total number of holes trapped in the valence-band-tail states identically equal to $H_{\rm vb}(t)$

$$n(t)=N_0\left[\frac{H_{\rm vb}(t)-N_{\rm DB}}{fE_{\rm ch}N_c}\right]^{1/\alpha} , \quad (5)$$

where the dispersion parameter α is $kT/E_{\rm ch}$. Using Eq. (1), the secondary photocurrent transient becomes

$$I(t)=e\mu_0 AFN_0\left[\frac{H_{\rm vb}(t)-N_{\rm DB}}{fE_{\rm ch}N_c}\right]^{1/\alpha} . \quad (6)$$

Equation (6) describes a secondary photocurrent transient in which electrons flow as long as holes are trapped and unable to recombine. Figure 1 depicts the recombination process. The holes remain trapped until they are thermally emitted to more dense, less localized states at $E_{\rm tnl}$ (step 1). These provide a "conduction path" by which the holes tunnel to doubly occupied midgap DB's (step 2) and complete the recombination process (R, steps 3 and 4).

Since recombination at the DB is so much faster than the emission of the deeply trapped holes, $H_{\rm vb}(t)$ is controlled only by thermal emission from traps which are within a tunneling distance of a doubly occupied DB. Transitions of holes among SHT's serves only to establish quasiequilibrium of the $H_{\rm vb}(t)$'s. Therefore, using first-order rate equations, we can write

$$H_{\rm vb}(t)=\int P_{\rm vb}(E)e^{-e_p t}dE , \quad (7)$$

where $P_{\rm vb}(E)$ is the starting distribution for holes in SHT's and e_p is the emission coefficient of the holes to $E_{\rm tnl}$ given by

$$e_p=\nu e^{(E_{\rm tnl}-E)/kT} . \quad (8)$$

Raising both sides of Eq. (6) to the αth power and combining with Eq. (7) we find

$$[I(t)/e\mu_0 AFN_0]^\alpha$$
$$=\frac{1}{fE_{\rm ch}N_c}\left[\int P_{\rm vb}(E)e^{-e_p t}dE-N_{\rm DB}\right] . \quad (9)$$

Differentiating with respect to time yields

$$\gamma \frac{d[I(t)]^\alpha}{dt} = -\int P_{vb}(E) e_p e^{-e_p t} dE \;, \qquad (10)$$

where $\gamma \equiv f E_{ch} N_c / (e\mu_0 AFN_0)^\alpha$. It has been shown[23] that unless $P_{vb}(E)$ is a δ function in energy, the integral of Eq. (10) can be approximated by $(kT/t)P_{vb}(E - E_{tnl})$ where $E - E_{tnl}$ is related to t by

$$E - E_{tnl} = kT \ln(\nu t) \;. \qquad (11)$$

This leads to an expression for the SHT population which can be compared with experiment:

$$P_{vb}(E - E_{tnl}) = -\frac{t}{kT} \gamma \frac{d}{dt} [I(t)]^\alpha \;. \qquad (12)$$

RESULTS

Measured current transitions at four different temperatures are shown in Fig. 2. We apply Eq. (12) to these photocurrent transients to determine $P_{vb}(t)$, which gives the time evolution of charge out of SHT's, thus limiting the rate of recombination. $P_{vb}(t)$, shown in Fig. 3 as a function of time, exhibits a fairly well-defined peak at all but the lowest T. At short times it approaches zero since the shallower states are not occupied at the level of illumination used to establish a steady state at these temperatures. The inset to Fig. 3 shows an Arrhenius plot of the natural logarithm of the peak position (t_p) as a function $10^3/T$. The best fit leads to a value of $\nu = 5 \times 10^8$ sec^{-1} and an activation energy of 0.25 eV. The attempt-to-escape frequency is reduced from the more usual phonon frequency of $\cong 10^{+12}$ sec^{-1} by the tunneling process (step 2).[24] Equation (11) can now be used to determine the energy distribution.[25]

The energy distributions of $P_{vb}(E - E_{tnl})$ measuring at 163 K using a photon flux of 10^{14} photons/cm^2 sec at 630 nm are shown in Fig. 4. We focus our attention first on the distribution for the annealed film. To locate this distribution in the band gap, i.e., to find E_{tnl}, we use results from measurements of the wavelength dependence of

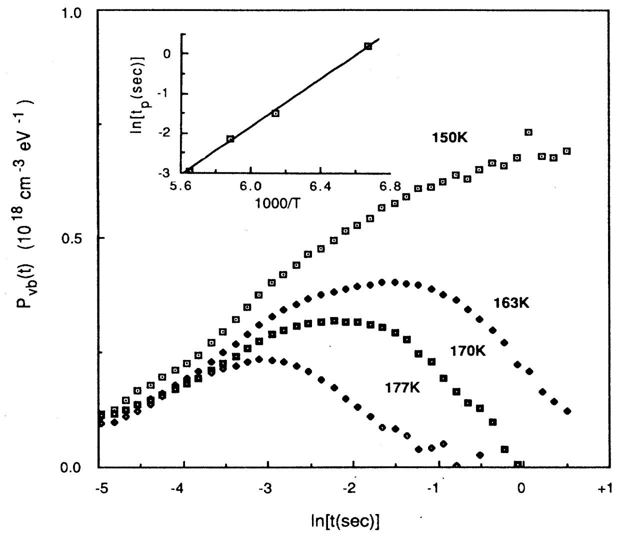


FIG. 3. SHT population $P_{vb}(t)$ as a function of time calculated from the secondary current transients shown in Fig. 2. Inset shows the logarithm of the time (t_p) of peak values of $P_{vb}(t)$ vs $1000/T$. The slope gives $E - E_{tnl}$ and the intercept ν; the best fit is $\ln(t_p) = 3050/T - 20$.

dual beam photoconductivity measurements[4] and photo-induced optical absorption[26] that placed hole trap sensitization centers 0.40 eV above the valence-band edge. The states involved in tunneling at E_{tnl} are 0.25 eV shallower than the SHT distribution edge or 0.15 eV above E_v. At this position we expect a valence-band-tail state spacing of $[0.060 N_c \exp(-0.15/0.06)]^{-1/3} = 50$ Å. Tunneling is viable over such distances, since potential barrier heights must be less than 0.15 eV. Since the distribution of SHT's falls short of matching the number of states in the conventional valence-band tail with a characteristic energy of 0.060 eV (see Fig. 1), it is clear that these states do

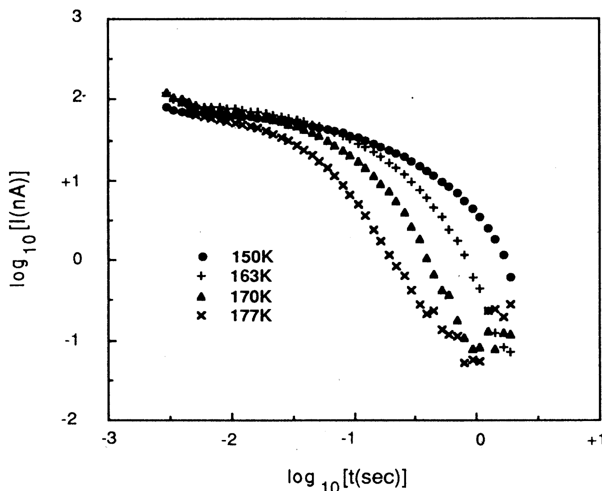


FIG. 2. Current transients for the annealed a-Si:H film with coplanar contacts at the four temperatures noted.

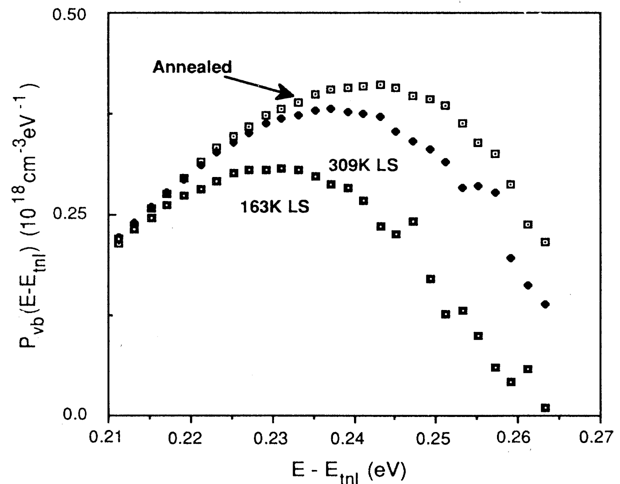


FIG. 4. $P_{vb}(E - E_{tnl})$ vs energy depth from E_{tnl} for the annealed, 309- and 163-K light-soaking exposures. $E_{tnl} - E_v = 0.15$ eV.

not make up the entire tail.

The lower two curves show the SHT energy distributions remaining after 20 min of light soaking with a photon flux of 7.5×10^{15} photons/cm^2 sec at each of two temperatures. At either temperature, the energetically deeper states are removed first. Light soaking at 309 K destroys about 5×10^{15} states/cm^3, a value similar to the number of light-induced DB's produced for a similar exposure of an annealed sample grown in the same system.[27] We have also observed that shallower states anneal back faster than the deeper states with a complete anneal requiring 20 h at 403 K.

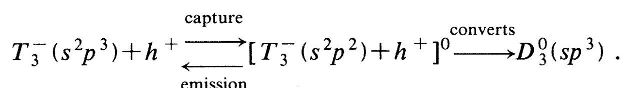
The same light-soaking exposure at 163 K removes about three times as many SHT's, which again are the deeper SHT's with the longer hole emission times. More states are lost for two reasons: (i) at lower T the effects of back anneal during light soaking are smaller[19,28] and (ii) the hole residence times are longer. This temperature dependence is also observed in photoluminescence studies.[29,30] Unlike the states lost after the 309-K light soaking, these states will almost completely anneal after 15 h at 303 K and require only 2 h at 365 K for a full anneal. The deepest states lost in the SHT distribution take the longest time to anneal.

DISCUSSION

Estimates of the drift length and photoconductive gain can be used to judge whether our model and analysis are reasonable. First we note that the low-temperature peak of the σ_{pc} versus $1/T$ occurs where the gain is maximum; at higher T the thermal emission times are shortened and at lower T the electron drift mobility decreases further and other recombination pathways become important. For this sample the peak occurs at $\cong160$ K.[19] Figure 3 shows a response time of $\cong0.2$ sec at 163 K. This number, together with an applied field of 10^3 V/cm and an electron drift mobility of 2×10^{-3} cm^2 V^{-1} sec^{-1},[31] gives an electron drift length of 0.4 cm. For a contact spacing of 0.1 cm, the peak gain is 4.

As mentioned above, we consider two microscopic defects for the SHT's: One is the strained sp^3 bonding states displaced upwards into the gap from the valence-band edge. Stutzmann *et al.*[27] argued that when these weak bonds trap a hole, the back bonds distort, stabilizing the hole. An electron localized in the antibonding state recombines nonradiatively with this hole and transfers enough energy to the weak bond so that local reconfiguration can stabilize two new DB's. Our data show that the energetically deeper SHT's disappear first upon light soaking. This agrees with Stutzmann's model because deep SHT's (i) provide the long hole residence times, making conversion more likely, and (ii) they represent the weakest bonds in the distribution. The SHT's do not act as strong recombination centers compared to DB's because the electron must lose much more energy through a multiphonon loss process in order to recombine. Since the resulting recombination rate has an exponential energy dependence, this transition is 10^{-3} less likely than a transition to a DB.

The other defect considered as the source of the SHT is T_3^- proposed by Branz and Silver.[32] This defect is an s^2p^3-hybridized DB lying below E_F and is stable because of either a negative correlation energy[33] or microscopic inhomogeneities.[34] The T_3^- center would be a good hole trap because of its negative charge. It would act as a safe hole trap because it is neutral when occupied by a hole. In addition, the phonon emission argument given above would make electron capture less likely. The process of converting a T_3^- to a DB with an sp^3 bonding configuration is given by



$$T_3^-(s^2p^3)+h^+ \underset{\text{emission}}{\overset{\text{capture}}{\rightleftarrows}} [T_3^-(s^2p^2)+h^+]^0 \xrightarrow{\text{converts}} D_3^0(sp^3) \ .$$

During light soaking, the intermediate defect state, a hole bound at a T_3^-, can do one of the following: (i) The hole can be emitted and the normal recombination process will occur as depicted in Fig. 1, or (ii) the defect can convert to a positively correlated, sp^3-configured DB. The energetically deeper states are more likely to complete this conversion process because of their longer hole residence times. This time is exponentially lengthened by the energy required to reach E_{tnl}.

CONCLUSION

We have provided a recombination model and a new analysis technique which lead to the energy distribution of SHT's in undoped *a*-Si:H and account for the extended photocurrent transients. In our model, transients, occurring after steady state is established, decay only after safely trapped holes are released thermally to shallower valence-band-tail states at E_{tnl}. There they diffuse via more dense, less localized valence-band-tail states through which they tunnel to occupied dangling-bond states completing the recombination process. Thermally assisted tunneling is indicated because of the low attempt-to-escape frequency of 5×10^8 sec^{-1} found for these trapped hole distributions which are distributed 0.21–0.26 eV from E_{tnl}. Light soaking destroys the deeper SHT's because of the longer hole residence times. Furthermore, the number destroyed is nearly equal to the number of DB's produced. Thus we conclude that hole trapping in these states is an intermediate step in bond breaking or defect conversion. Two possible sources of SHT's were discussed.

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