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The effect of light induced degradation on the sensitization phenomenon in a-Si:H

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We report a more detailed understanding of the thermal-quenching and sensitization processes in tetrahedrally bonded amorphous semiconductors. In particular we reveal in detail the effect of light soaking, in hydrogenated amorphous silicon (a-Si:H), on the shift of the recombination transition, from the dangling bonds to the valence band-tail states as the temperature is lowered. Our experimental observations and model simulations are shown to account for various results in the literature, explaining in detail how the charge neutrality condition determines the recombination process in a-Si:H. This, in turn, demonstrates for the first time that it is possible to deduce the three valence states of the dangling bonds only on the basis of the recombination processes implied by the phototransport observations.

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1. Introduction

The phenomena of sensitization and thermal quenching in hydrogenated amorphous silicon (a-Si:H) have been studied extensively, both theoretically and experimentally, in the last twenty years and they became the most conspicuous signature of the photoelectronic processes in this material [1-7]. While the basis of their origin, i.e. the shift of the dominant recombination channel from one type of recombination centers to another is well established, the details of these phenomena, such as the differences between the behavior of different a-Si:H materials, are not well understood to this date [5-7]. This is in spite of the importance of the basic understanding of the phototransport properties in thin film optoelectronic materials in general, and in amorphous semiconductors in particular. What is well accepted by now is that at about room temperature the recombination takes place via the dangling bonds and that at low temperatures it takes place via the valence band tail states [1-4] and/or via some impurity states [5]. However, in addition to this basic understanding there were, over the years, attempts to interpret some behaviors of the phototransport properties outside the framework of the above basic processes. These attempts usually followed the unfortunate practice [6-8] of trying to derive conclusions from a measurement of a single dependence of the photoconductivity on an externally controllable parameter. As pointed out by others [9] and by us [10] the main problem with such a practice is that since there are very many parameters involved in the description of the density of states (DOS) map and the recombination process in a-Si:H, a single dependence is "too easy" to fit by very different scenarios, which may or may not be representative of the true scenario in the material.

Our motivation to study the sensitization effect [3] in a-Si:H in more detail than done in previous studies was the report of Brandt and Stutzmann [8] on the effect of light soaking on the photoconductivity in a-Si:H, which is a conspicuous example of the above practice. In particular, a comparison of their experimental results with those of ours [11] led us to a comprehensive computer simulation study that appears to reveal the details of the sensitization process in that interesting and widely studied material [1-3]. The highlights of this study are reported in this paper.

In the work of Brandt and Stutzmann [8] the concentration of the dangling bonds, N_{db}, the photoconductivity, σ_{ph}, and the corresponding light intensity exponent, γ_e, have been measured as a function of the light soaking time, t. The quantity γ_e that is associated with the lifetime of the majority carriers (the electrons here) τ_e, is defined by $d[\log(\sigma_{ph})]/d[\log(G)]$, where G is the carrier generation rate that is proportional to the illumination intensity that is applied to the corresponding sample [1,3]. Brandt and Stutzmann found a systematic increase of N_{db} and γ_e and a systematic decrease of σ_{ph} with t. These findings were translated by them to imply a monotonic decrease of σ_{ph} with N_{db} as well as a monotonic increase of γ_e with increasing N_{db}. While the first dependence is easy to understand [3-5] as due to the decrease of the electron's lifetime τ_e ($= \sigma_{ph}/q\mu_e G$, where q is the electronic charge and μ_e is the electrons mobility) with the increase of N_{db}, the $\gamma_e(N_{db})$ behavior is not straight forward. To give their results some significance Brandt and Stutzmann "interpreted" the effect of light soaking on the recombination process as follows [8]: "γ_e approaches the monomolecular value of 1, while we observe bimolecular recombination in the annealed sample ($\gamma_e \approx 0.5$)". This, rather vague, statement does not only mean very little in the context of a-Si:H but it implies that there

is some *systematic monotonic dependence* of γ_e on N_{db} that can be accounted for.

Following this summary of Ref. 8 and our finding of a corresponding non-monotonic behavior of the dependence of γ_e on t, i.e. on N_{db} [11], we initiated a study with a three fold purpose; to find out if indeed our experimental observation of $\gamma_e(N_{db})$ is representative, to see if it fits into the above-mentioned picture of sensitization and thermal quenching and to try and understand the effect of light soaking on the latter processes in more detail than available in the literature so far. We start then by showing that the $\gamma_e(N_{db})$ dependence reported by Brandt and Stutzmann [8] is not general and that the gross features of the representative $\gamma_e(N_{db})$ dependence can be well understood within the framework of the sensitization process. We further show that our experimental results exhibit a particular behavior that has not been discussed before, and that, by carrying out extensive simulations, this behavior leads to a much better understanding of the photoelectronic processes in a-Si:H and similar systems. The results of the simulations are found to confirm and explain the details of the experimentally observed dependencies and shed light on the evolvement of the sensitization process in a-Si:H. Since our samples [11,12] and our experimental procedure [4,5] have been described previously we mention them only briefly, in Sec. 2. In Sec. 3 we present our experimental results and the basic physics that they imply, and in Sec. 4, we present the conspicuous results of our comprehensive simulation study, the procedure of which has been given in detail in our former studies of recombination processes [5,13]. In Sec. 5 we discuss and summarize the implications of these results on the understanding of the sensitization process in a-Si:H.

2. Experimental details

The experimental results described in the present work were obtained on a sample of undoped, device quality, a-Si;H film that was deposited, using the direct current glow discharge technique [11], on a Corning 7059 glass substrate that was held at a temperature of 250C. This film had a thickness of 1.3 μm. However, similar results were obtained on samples deposited at other substrate temperatures and on various samples that were prepared by the radio frequency glow discharge and the hot wire techniques [11,12].

In our experimental work the photoconductivity measurements were carried out in the co-planar photocarrier grating configuration (0.4 mm separation between two NiCr contacts) [11,12] as a function of the sample's temperature (in the range between 80 and 300 K) and as a function of the HeNe laser illumination intensity that was applied to the sample. The highest photocarrier generation rate that was provided by this illumination was $G = 5\times10^{20}$ $cm^{-3}s^{-1}$ and it was reduced by steps down to $G = 5\times10^{18}$ $cm^{-3}s^{-1}$ in order to find the dependence of the photoconductivity on this rate. The results presented below for the mobility lifetime product $(\mu\tau)_e = (\sigma_{ph}/qG)$ were taken at a generation rate of $G = 5\times10^{19}$ $cm^{-3}s^{-1}$ while the γ_e values were obtained over the above range of generation rates by fitting the experimental data to a $\log(\sigma_{ph}) = \gamma_e[\log(G)]$ dependence.

3. Experimental results

Following the above-mentioned issues associated with the results of Brandt and Stutzmann [8] let us start by examining our own experimental results [11] regarding the effect of light soaking on the relevant two phototransport properties. Our experimental work included a systematic study of the mobility-lifetime product of the majority carriers, $(\mu\tau)_e$ (= σ_{ph}/qG), and the corresponding light intensity exponent γ_e, as a function of the sample's temperature, T. The results of our measurements, before and after 10 hours of, 100 mW/cm^2 (HeNe laser), light soaking are shown in Fig. 1. In this figure we see the typical thermal quenching effect that is manifested in semiconductors in general [3,14] and in a-Si:H in particular [1,3,4,5], by the decrease of $(\mu\tau)_e$ with increasing temperature (here above about 150 K), as well as by the corresponding typical sensitization effect of the recombination channel alteration [14], that is manifested by the $\gamma_e(T)$ peak with $\gamma_e > 1$ values.

In accordance with Brandt and Stutzmann [8] and the simple expectation from the increase of N_{db} (i.e. that $\tau_e \propto 1/N_{db}$ [1,3,4]) we see a decrease of $(\mu\tau)_e$ due to the light soaking. We note however, already, the weaker manifestation of the thermal quenching (i.e. the "flatter" T dependence of $(\mu\tau)_e$) that follows the light soaking. This latter effect has not been noticed before, but as we show below it is a genuine-general effect. The interesting behavior however is that of $\gamma_e(T)$. As will be shown in the next section, this behavior is not only typical, but is in accord with the simplest understanding of the DOS map and the recombination mechanisms in a-Si:H which constitute the well known "standard model" of this material [1]. First, however, let us examine the results around room temperature (300 K) where we see that just above that temperature γ_e *increases with light soaking* (i.e. with N_{db}) while just below this temperature *the monotonic behavior is reversed*. This observation suggests then that with further increase of N_{db} this transition, from a monotonic-increasing to a monotonic-decreasing behavior of $\gamma_e(N_{db})$, will shift to lower temperatures. This result shows already that the conclusion of a monotonic increase of $\gamma_e(N_{db})$ and the attempt to assume that it represents a systematic behavior, as was suggested by Brandt and Stutzmann [8], is not founded, and that it applies only to specific conditions (say, particular combination of the values of N_{db} and T) of the system.

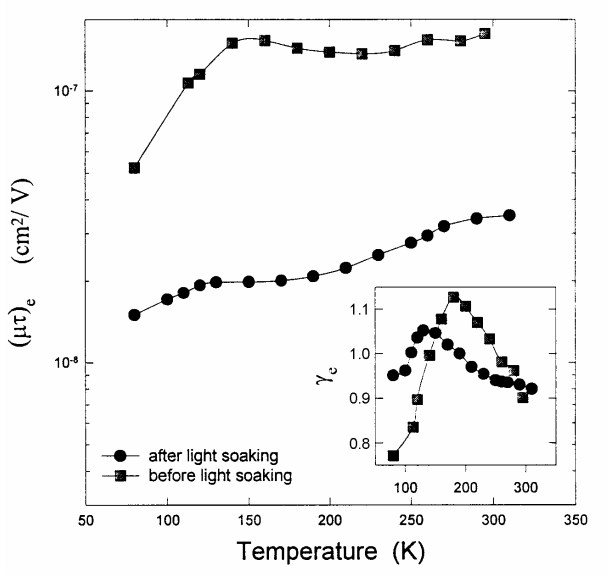

Fig. 1. Our experimentally determined temperature dependence of the electrons' mobility-lifetime product and the corresponding light intensity exponent in a-Si:H samples that were deposited by the dc glow discharge technique.

Let us try now to understand the details of the $\gamma_e(T,N_{db})$ behavior as shown in Fig. 1 within the framework of the thermal quenching and the sensitization process assumed so far for a-Si:H [1,3]. The most significant observation regarding the effect of light soaking, i.e. the shift of the $\gamma_e(T)$ peak to lower temperatures, can be well understood, in light of the numerous works on the above phenomena [1-5], in a-Si:H as follows. At high (around room) temperature the dominant recombination takes place via the dangling bonds, through their neutral, N_{db}°, and positively charged, N_{db}^{+}, centers, since these have a larger capture coefficient for electrons than the valence band-tail states. When the temperature is lowered the energy range (between the two demarcation lines [3,14]) in the DOS map, in which the latter states are active in the recombination process, broadens, and the volume concentration of these states increases, yielding the strengthening of the recombination process through them. Hence, as the temperature decreases, the dominant recombination channel shifts from the dangling bonds to the valence band tail states in spite of the smaller carrier capture coefficient of the latter. This shift is manifested [1,5] by the decrease of the lifetime (the thermal quenching effect [3,14]) upon the increase of temperature in a certain temperature range (here between 150 and 200 K) that is followed by an increase of the lifetime with further increase of the temperature. Since the increase in the carrier generation rate (illumination intensity) expands the above energy range in the DOS map (thus having the opposite role to that of the increase in temperature [3,14]), there is initially an increase in the lifetime with increasing G and then a decrease of this quantity [1,3,5]. This is manifested then by a $\gamma_e(T)$ peak with a $\gamma_e > 1$ value in the regime where the dominant recombination channel is altered. As well understood and shown below, this simple picture is confirmed by the "standard model" of a-Si:H [1].

There are however two new observations in Fig. 1 that are significant for the more detailed understanding of the sensitization process in general and in a-Si:H in particular. The peak in $\gamma_e(T)$ is not only shifted but it is, *both, broadened and lowered* with the increase of N_{db}. These new observations are not trivially understood from the gross features of the sensitization process that we described above, or from the many previous works in the literature [1-8]. In the next section we explain these observations in detail and show that they are related to the nature of the dangling bonds as having three charging states.

4. Results of the simulation study

In order to find out whether the experimental results shown in the previous section are very general and not particular to our samples, on the one hand, and whether they are representative of the previous understanding of a-Si:H [1-5], on the other hand, we have performed a detailed simulation study of the standard model [1] of this material. In particular we conducted runs of Tran's version of the standard model with a few values of the parameter N_{db}, which is assumed here, and in general [2,3,8], to be the only parameter that is affected by the light soaking. In other words, except for the various values of N_{db} all the equations and parameters used in our simulations are those given in Ref. 1 for the standard model of a-Si:H that is denoted there as the B1 model. For simplicity, we have used however discrete states for the dangling bonds rather than Gaussian peaks [1], finding that within the parameters used here the results for both descriptions are the same. Our results of the runs of this model [1] (which was suggested prior to our experimental study [11] and thus was not biased by them) for the computation of the $(\mu\tau)_e$ and γ_e dependencies on T, are shown in Fig. 2. Considering the effect of light soaking (i.e. the increase of N_{db}) we see first, that as in the experimental results that we have shown Fig. 1, the increase of N_{db} suppresses the variations in $(\mu\tau)_e(T)$ and shifts the $\gamma_e(T)$ peak to lower temperatures. It is also conspicuous, as in Fig. 1, that the $\gamma_e(T)$ peak is *broadened and lowered* with the increase of N_{db}. In fact, it appears from Fig. 2 that this effect results in a variety of $\gamma_e(N_{db})$ behaviors. In particular, there are temperature ranges at which there is a monotonic increase of $\gamma_e(N_{db})$, temperatures ranges at which there is a monotonic decrease of $\gamma_e(N_{db})$, and temperature ranges at which there is a "*crossover*" in the direction of the variation of $\gamma_e(N_{db})$. We conclude then that both, our experimental results and model simulations, disprove the "single direction" significance that Brandt and Stutzmann [8] tried to associate with the $\gamma_e(N_{db})$ dependence that they observed at room temperature.

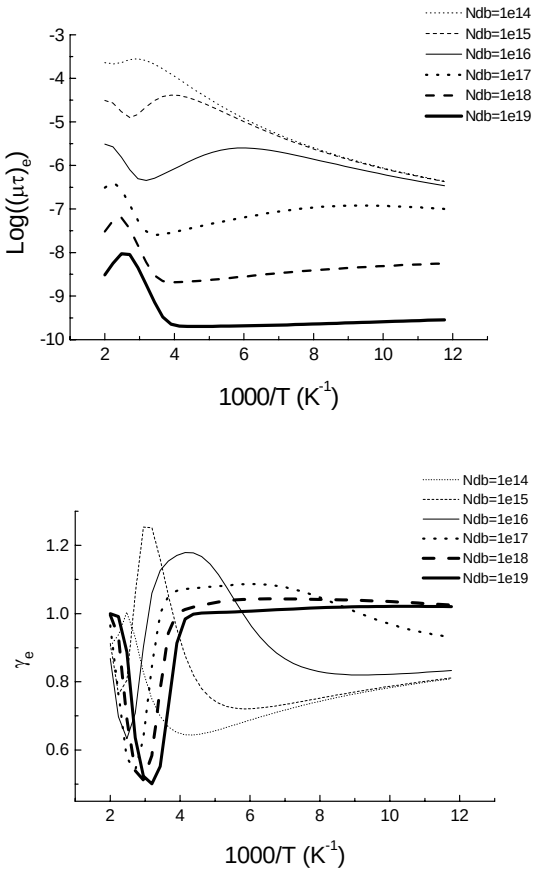

Fig. 2. Our computed temperature dependence of the electrons' mobility-lifetime product and the corresponding light intensity exponent for six concentrations of the dangling bonds. The model and parameters used were those of Tran's B1 version of the standard model of a-Si:H.

The more interesting challenge is however to understand the above-mentioned, previously unnoticed, dependence of the γ_e(T) peak on N_{db}. A hint of the processes that take place is already given in the behavior of $(\mu\tau)_e$ at low temperatures. We see that for $10^{14} \leq N_{db} \leq 10^{16}$ cm^{-3}, the values of $(\mu\tau)_e$ are almost independent of N_{db}, indicating that for the corresponding combinations of N_{db} and T the recombination takes place not via the dangling bonds but via other states that are assumed to be the valence band tail states. For higher N_{db} values the $(\mu\tau)_e$ value does depend on N_{db} even at low temperatures suggesting that the dangling bonds are the dominant recombination centers even then. Qualitatively then, the transition between the two possible recombination channels is not only shifted but is also "weakened" with the increase of N_{db}, as is manifested by the "smearing" of the γ_e(T) peak.

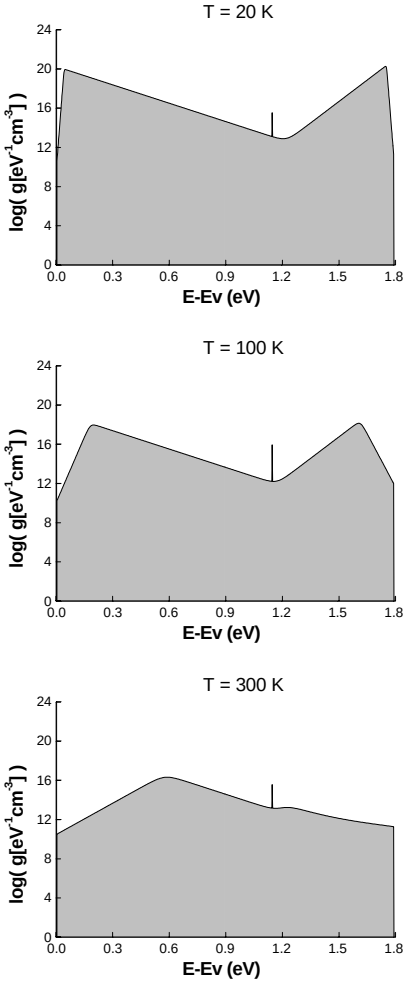

Fig. 3. The variation of the recombination-active band tails states (gray areas) with temperature, for Tran's B1 model. Also indicated are the position and the total volume concentration of the dangling bonds (discrete peaks) in the case of $N_{db} = 10^{16}$ cm^{-3}. The T = 20 K result represents essentially the "common" DOS map of the "standard model" of a-Si:H.

To appreciate the above behavior let us follow first the temperature dependence of the band tails states occupations in the standard model. This is done in Fig. 3 where we show the DOS map of the band-tail states that participate in the recombination (gray areas) as well as the position and the concentration of the dangling bonds (the discrete peak). The map is presented here for the band gap energies, E, with respect to the mobility edge of the valance band, Ev, for three temperatures. As is clearly seen the volume concentration of the band tail states, that are available for recombination, decreases with increasing temperature thus making the corresponding role of the concentration of the dangling bonds, N_{db} ,more dominant.

Let us follow now, beyond the results presented in Ref. 1, the temperature dependencies of the concentrations and the recombination rates for various values of N_{db}. This is done in Fig. 4 as follows. The total concentration of dangling bonds, N_{db}, consists of the concentration of the

neutral dangling bonds, $N_{db}°$, the positively charged dangling bonds, N_{db}^{+}, and the negatively charged dangling bonds N_{db}^{-}, while the band tails volume contributions are given by the energy-integrated concentrations of the recombination available, acceptor-like, conduction band tail states, Q_n, and, recombination available, donor-like, valence band tail states, Q_p. Also shown are the steady state concentrations of the electrons, n, and the holes, p. The corresponding recombination rates through the various dangling bonds are $G_{db}°$, G_{db}^{+} and G_{db}^{-}, and through the valence and conduction band tails states they (the integrated values of the rates [1]) are G_{vt} and G_{ct}. These results, as those presented previously by Tran [1] for the relatively low N_{db} values show that at low temperatures the charge neutrality is maintained by the equality of Q_p and Q_n, while the crossing of G_{vt} with G_{db}^{+} and $G_{db}°$ represents the sharp transition of the dominant recombination channel, from that of the valence band tail states to that of the dangling bonds states upon the increase of temperature.

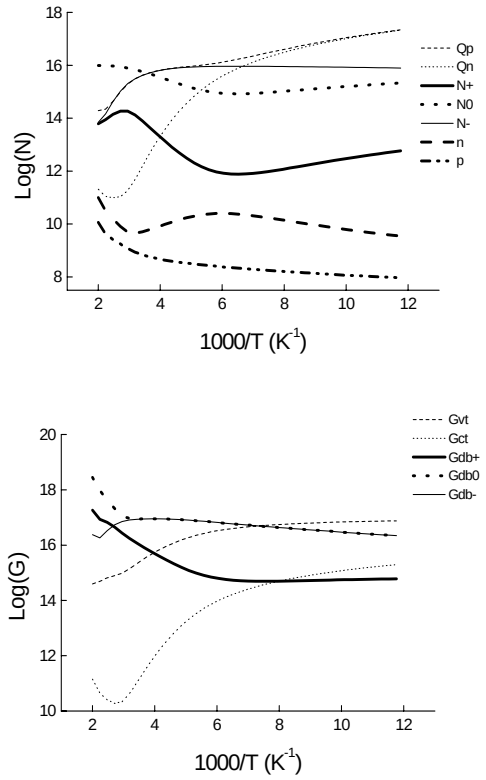

Fig. 4. The temperature dependence of the concentrations (N) and the recombination rates (G) via the different types of centers in the mobility gap of a-Si:H, for a relatively low ($N_{db} = 10^{16}$ cm^{-3}) concentration of dangling bonds.

In contrast with the above well-known and well understood case [1,5] we show in Fig. 5 the behavior of the other extreme, i.e. that of the temperature dependencies of the N's and the G's for very high values of N_{db}. Here, we see that even at low temperatures the values of Q_n and Q_p are smaller than the concentrations of the various dangling bonds and thus, throughout the entire temperature range, the basic well-known [3,14] condition for the sensitization, i.e. that the concentration of the sensitizing centers is significantly larger than the concentration of the sensitized centers, is not fulfilled. In our case, of course, the neutral and the positively charged dangling bonds (that have a relatively larger electron capture coefficients) are the latter, and the band tail states are the former [1]. Correspondingly, sensitization and thermal quenching phenomena are not observed in this case. It is further important to note that in contrast with the case shown in Fig. 4, the charge neutrality in the case of Fig. 5 is maintained by the dangling bonds-only charge neutrality condition ($N_{db}^{-} \approx N_{db}^{+}$) and the recombination rate through their charged states is as significant as the recombination rates through the much higher concentration of neutral states.

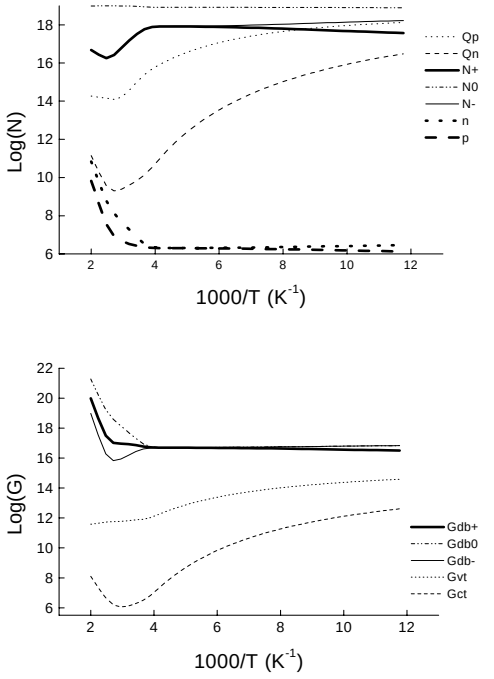

Fig. 5. The temperature dependence of the concentration of, and the recombination rates at, the different types of centers in the mobility gap of a-Si:H, for a high ($N_{db} = 10^{19}$ cm^{-3}) concentration of dangling bonds.

Following these two extreme behaviors we conclude that the experimental behavior shown in Fig. 1 for the as-prepared material, corresponds to the above low- N_{db} case (Fig. 4) while the behavior found after light soaking can not be accounted for by this or by the large N_{db} case (Fig. 5). Hence, we conclude that our experimental results, for the light soaked case shown in Fig. 1, correspond to an intermediate-N_{db} case, such as the one given by $N_{db} = 10^{17}$ cm^{-3}, in Fig. 2. To confirm this suggestion we show in Fig. 6 the corresponding temperature dependencies of the N's and G's. Here, while the increase

of N_{db} shifts the transition to lower temperatures, the concentration of the recombination available valence band-tail states Q_p is not enough to yield a recombination channel that will clearly dominate the dangling bonds channel and in turn yield a sharp transition that is manifested by a narrow $\gamma_e(T)$ peak, as found for annealed (small N_{db} value). We have then that the concentration Q_p increases with decreasing temperature but due to the light soaking induced increase of N_{db} (in comparison with the case of Fig. 4), the former offsets the latter only at low temperatures. The low temperature range in which the competition between the two recombination channels is sustained causes, in this case, the broadening and "suppression" of the sensitization transition. The latter accounts very well then for the experimentally observed "smearing" of the $(\mu\tau)_e$ and γ_e peaks, that we saw in Fig. 1.

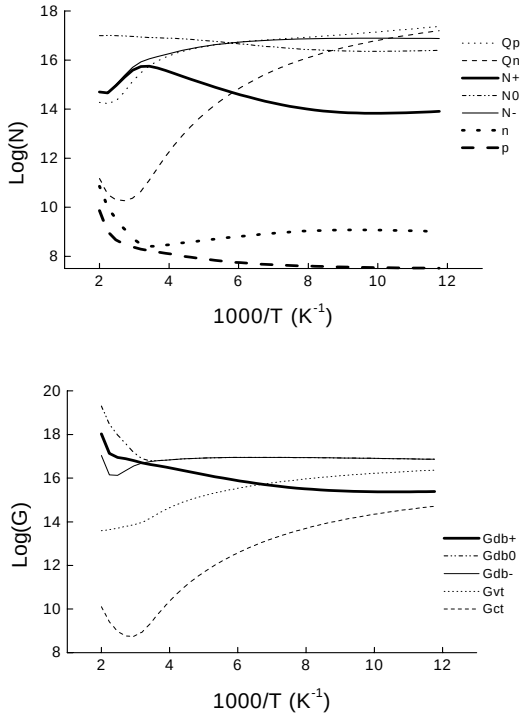

Fig. 6. The temperature dependence of the concentration of, and the recombination rates at, the different types of centers in the mobility gap of a-Si:H, for an intermediate ($N_{db} = 10^{17} cm^{-3}$) concentration of dangling bonds.

In another study [5] we have pointed out that the dominant recombination at low temperatures can be controlled by oxygen induced centers rather than by valence band tail states. However, since the effect of the increase of N_{db} is the same as in the case of the valence band tail states discussed above, the same considerations regarding the role of the dandling bonds apply. In another publication we will show that indeed, also in that case, the results and conclusions are very similar to those presented here.

5. Discussion and conclusions

In this work we have seen that the basics of the recombination process in a-Si:H are well understood and are well accounted for by the "standard model" of this material. The present work puts however in perspective previously reported experimental results and their interpretation. More importantly, our work sheds light on the details of the process as follows. From the simulations of the standard model we are able to explain in detail the behaviors shown in Fig. 2 and the various experimental observations that were reported by many authors [1,4,8,11]. The scenario that determines the behaviors, such as those of $\gamma_e(T)$ and $\gamma_e(N_{db})$ is concluded here to depend on the way in which the charge neutrality condition is fulfilled. While for the high N_{db} values the charge neutrality condition is determined solely by the dangling bonds ($N_{db}^- \approx N_{db}^+$) and no sensitization can take place, in the low N_{db} values it is determined by the charge neutrality condition of the band tail states ($Q_p \approx Q_n$) at low temperatures, and the sharp transition to the $Q_p \approx N_{db}^-$ condition at higher temperatures. The intermediate case, which exists in typical light soaked samples, is characterized by the prevalence of latter condition throughout a very broad temperature range. It appears then that prevalence is responsible for the conspicuous "smearing" of the sensitization transition in light soaked samples suggesting that the interplay between the above three dominant charge neutrality conditions are the reason for the richness of the behaviors of $\gamma_e(T)$ and $\gamma_e(N_{db})$.

The above picture is confirmed by our computed recombination rates. We have found indeed that for the high N_{db} values, only a small fraction of the recombination process shifts from the dangling bonds to the valence band tail states. The latter smaller contribution is superimposed on the main recombination route via the neutral and positively charged dangling bonds. In contrast then with the case of annealed samples with low N_{db} values, the increase in the concentration of, recombination available, valence band tail centers (Q_p), with decreasing temperature, is not enough to yield a sensitized recombination. On the other hand in the intermediate range of N_{db} values, the proximity to the fulfillment of the above $N_{db}^+ \approx N_{db}^-$ condition does enable only a small, superimposed, sensitization which is manifested by the broadening of the temperature range over which there is a contribution to the sensitization, bringing about the smearing of the transition in the temperature dependencies of phototransport properties $(\mu\tau)_e$ and γ_e. This replaces the sharp sensitization transition that is well known for the small values of N_{db}. Hence, the stringent constraint of the charge neutrality and its manifestation are necessary for the explanation of the results, and they yield a very convincing proof that it is the *triple valence-charge nature of the dangling bond that controls the recombination* rather than a collection of various uncorrelated deep level charged centers. This holds for a-Si:H in general and for light soaked samples in particular. While accepted for a long time, there was so far no such evidence for this triply-charged nature of the dangling bonds that relies only on

the experimentally observed phototransport properties of this material.

In conclusion, we presented the full scenario of the effect of the increase of the dangling bond concentration on the recombination processes in a-Si:H thus enabling a better understanding of the phototransport properties in light soaked a-Si:H and similar systems We have shown that the charge neutrality condition explains this scenario and that this condition can only be fulfilled by recombination centers with three charging states that have a correlated occupation.

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