Effective capture rates of carriers in amorphous hydrogenated silicon

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The effective capture rates of electrons and holes by the defects of undoped a-Si:H are measured using a technique based on the optical bias dependence of the modulated photocurrent. The evolution of the phase shift and modulated photocurrent spectra with the optical bias intensity is used to study the capture coefficient of the defects at different trap depths in the energy gap. The modulated photocurrent spectra are recorded using uniformly, or strongly absorbed light in order to obtain information about the trapping and recombination processes in the surface, the interface, and the bulk of the films. The calculated capture rates of carriers are studied in a series of a-Si:H films and information about the defect structure is obtained. It is found that the effective capture rates of the carriers by the defects are not constant in the studied films and may vary by up to two orders of magnitude from sample to sample. Finally, the importance of the atomic environment and the local strains of the defects of a-Si:H for the capture process of the carriers is discussed. © *1996 American Institute of Physics.* [S0021-8979(96)03016-2]

I. INTRODUCTION

It is widely accepted that the dangling bonds (dbs), which can exist in neutral positive or negative charge states (D^0, D^+, and D^- respectively), determine the optoelectronic properties of a-Si:H. Therefore, the defect density and the effective capture rate of the carriers by the defects are important for the determination of the recombination rate, which is essential in the performance analysis of the solar cells and other electronic devices based on a-Si:H. The most common methods used to characterize the electronic properties of a-Si:H cannot provide direct information about the effective capture rates of the carriers. Namely, the photothermal deflection spectroscopy[1] (PDS), the constant photocurrent method[2] (CPM), and the electron spin magnetic resonance[3] (ESR), can measure only the total or a part of the total defect density.

The charge state of the dbs of a-Si:H is believed to be an important factor for the determination of the effective capture rates of the carriers. Since the correlation energy for a-Si:H is positive ($U>0.1$ eV), it is considered that most of the dbs in undoped samples are neutral. However, theoretical studies based on thermodynamic equilibration[4] between weak bonds and dbs,[5,6] or based on potential fluctuations,[7] have shown that most of the dbs may be in a charge state. According to the theoretical predictions of these models the defect state distribution in the band gap consists of one positively charged defect band above the Fermi level, another independent negatively charged defect band below the Fermi level, and a band of neutral dbs near midgap. Several experimental studies[8–13] have been employed to measure the ratio of the charged to neutral defect density in a-Si:H and to determine the distribution of the density of states that is more appropriate to fit the experimental results. It was concluded, on one hand, that the charged defect density is higher than that of neutral defects from light induced ESR,[8] and subgap absorption measurements,[9,10] in agreement with the predictions of the defect pool model. On the other hand, it was deduced that most of the defects are in the neutral charge state from modulated photocurrent experiments.[12,13] Therefore, the charge state of the dbs dominated trapping and recombination in a-Si:H remains an open problem at present.

In this work, a technique based on the optical bias dependence of the modulated photocurrent (OBMPC technique) is used, to determine the effective capture rates of the electrons and holes[12,14–16] by the defects in undoped a-Si:H samples. In this technique, the amplitude of the modulated photocurrent (MPC) and the phase shift are measured at different optical bias light intensities as a function of the modulation frequency of the light. From these spectra, the effective capture rates of the electrons and holes by the defect states are calculated for different bias light intensities and information about the defect structure is extracted.

II. EXPERIMENTAL DETAILS

Undoped a-Si:H films 0.7–0.9 μm thick were prepared by the conventional rf glow discharge technique under various deposition conditions of the substrate temperature (T_s=250–300 °C), interelectrode distance (D=20–35 mm), and silane pressure (P=50 or 100 mTorr).[17] The films were deposited in a single high vacuum (HV) chamber or in an ultrahigh vacuum (UHV) multichamber system with base vacuum before deposition of about 5×10^{-7} and 3×10^{-8} Torr, respectively.

A sinusoidal modulated light (probe light) and a dc bias light (pump light) provided by two light-emitting diodes (LEDs) of band-gap red light (660 nm) (Refs. 12 and 14–16) are used to illuminate the films in the OBMPC experiments. The probe light intensity was adjusted by neutral density filters to be in the range of 0.5%–25% of the dc bias light intensity. The amplitude of the MPC and the phase shift between the modulated light and the resulting MPC, were measured by a Lock-in amplifier at room temperature as a function of the modulation frequency (ν) at different bias light intensities in the region of 1×10^{10}–5×10^{13} cm^{-2} s^{-1}. In order to study the influence of the surface or interface states to the trapping and the recombination process, LEDs

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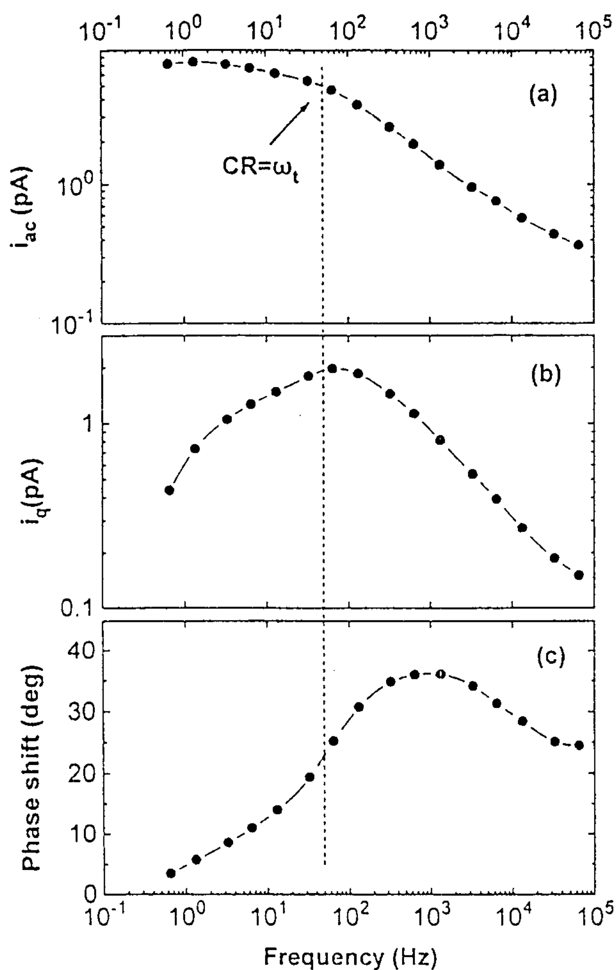

FIG. 1. Experimental $\Phi(\nu)$, $i_{ac}(\nu)$, and $i_q(\nu)$ spectra of an a-Si:H film deposited on the UHV multichamber at a substrate temperature of 250 °C measured with a bias light intensity of 1.1×10^{12} cm^{-2} s^{-1}. The dashed line shows the frequency $\omega_t=2\pi\nu_t$ that corresponds to the demarcation level of the trapped carriers.

of strongly absorbed blue (470 nm), green light (563 nm), or uniformly absorbed infrared light (880 nm) were also used. The films were illuminated from the free surface or through the substrate.

The photocurrent measurements were performed in samples with Al ohmic contacts in coplanar geometry. Eventual space charge regions of the contacts may be a problem in the MPC measurements because the modulated light may generate a modulated photovoltage that might affect significantly the MPC measurements. In this case, one expects that the phase-shift values may be voltage dependent. The results presented in this work referred to films with ohmic contacts where the phase-shift values are independent of the applied voltage, while the amplitude of the MPC was found to be zero with a zero applied voltage.

III. RESULTS

A. The MPC and phase shift spectra

The amplitude of the MPC $i_{ac}(\nu)$, the amplitude of the out of phase MPC $i_q(\nu)$, and the phase shift $\Phi(\nu)$ spectra of a film deposited at 250 °C are presented in Fig. 1(a), 1(b), and 1(c), respectively. The defect density for this sample measured by CPM was 1.3×10^{16} cm^{-3}. The $\Phi(\nu)$ spectra exhibit characteristic broad peaks that are typical for a-Si:H samples.[11,12,14–17] Similar broad spectra have been obtained by simulations assuming that the carriers interact with dbs.[15,16]

There are two regimes in the spectra of Fig. 1 that are characterized by different behavior. The two regimes are separated by the dashed line at the frequency ω_t defined by the break point of $i_{ac}(\nu)$, the maximum of $i_q(\nu)$, and the inflection point of $\Phi(\nu)$ spectra as shown in Fig. 1.

In the high frequencies regime ($\omega\gg\omega_t$), the phase shift takes relatively higher values, while the amplitude of the MPC $i_{ac}(\nu)$ depends on the frequency. This is a characteristic behavior observed at higher frequencies and/or when low optical bias intensities are used indicating that trapping and thermal reemission dominates.[14–19] In general, in this regime the empty gap states at trap depth E_ω, which are characterized by thermal emission rates equal to the modulation frequency, have the predominant contribution to the phase shift due to trapping and detrapping.[18] Assuming that the electrons dominate in the MPC measurements, then different empty gap states at trap depths E_ω below the conduction band can be probed by changing the modulation frequency according to: $\omega=2\pi\nu=e_n(E_\omega)$ or $\omega=c_nN_c\exp(-E_\omega/kT)$ where $e_n(E_\omega)$ is the thermal emission rate of electrons from E_ω, c_n is the capture coefficient of the electrons, and N_C is the effective density of the states in the conduction band.

In the low frequencies regime ($\omega\ll\omega_t$), the amplitude of the MPC $i_{ac}(\nu)$ shows a plateau and the phase shift $\Phi(\nu)$ decreases to zero. This is a characteristic behavior observed at low frequencies and/or when a relatively high bias light intensity is applied.[14–19] This is an indication that recombination dominates and the gap states are almost completely occupied and do not induce a phase shift.

The transition region between the low frequencies regime and the higher frequencies regime, defined by the dashed line at the frequency ω_t in Fig. 1, can be used to define the position of the demarcation level in the frequency domain, which allows one to distinguish the gap states that act as traps from those that play the role of recombination centers. The concept of demarcation levels as defined by Simmons and Taylor[20] that are called "quasi-Fermi-level of the trapped electrons E_{tn}, or holes E_{tp}," can be used as the most appropriate distinction of the shallow traps and recombination centers. According to the Simmons and Taylor statistics, the capture rate of the electrons, nc_n and holes, pc_p at the quasi-Fermi-level of trapped electrons E_{tn} is equal to the thermal emission rate of electrons from E_{tn} $e_n(E_{tn})=nc_n+pc_p$, where c_p is the capture coefficient of holes, and n and p are the density of free electrons and holes respectively. Calling $\mathrm{CR}=nc_n+pc_p$, the capture rate of electrons and holes, then at the demarcation level it is expected that $\mathrm{CR}=\omega_t$ and, therefore, ω_t can be used to measure the effective capture rate (CR) of the carriers by the defects that dominate trapping and recombination at the E_{tn} level.

In order to test the validity of the assignment of the frequency ω_t to the demarcation level of trapped carriers MPC measurements were made as a function of the optical bias intensity (F). If electrons dominate in the photoconductivity, by increasing F the demarcation level defined by the

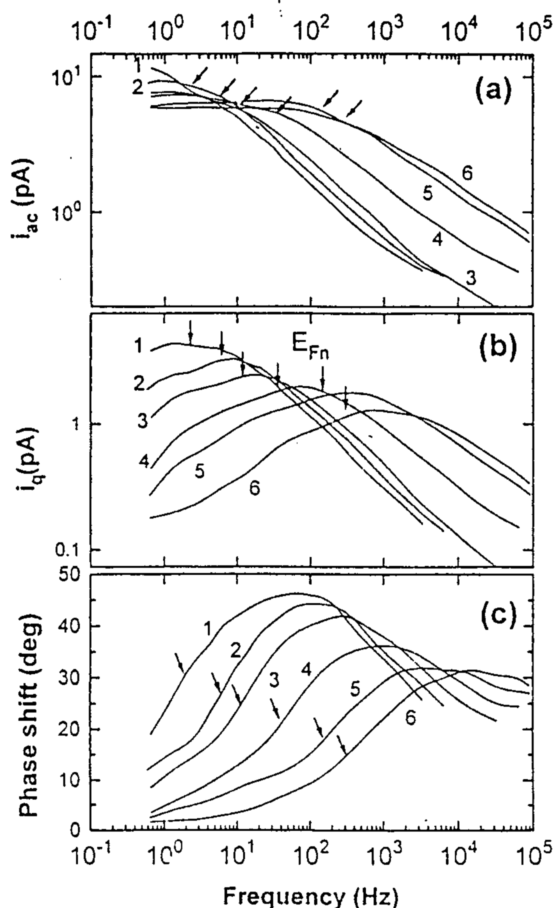

FIG. 2. Experimental $i_{ac}(\nu)$, $i_{ac}(\nu)$, and $\Phi(\nu)$ spectra in (a), (b), and (c), respectively, of a UHV a-Si:H film ($T_s=250$ °C) at various bias light intensities, F, in $cm^{-2} s^{-1}$. (1) 4×10^{10}, (2) 1.1×10^{11}, (3) 2.7×10^{11}, (4) 1.1×10^{12}, (5) 2.5×10^{12}, and (6) 6.2×10^{12}. The arrows represent the position (ν_{Fn}) of the quasi-Fermi-level E_{Fn} in the frequency domain as calculated from the dc photoconductivity.

frequency ω_t should shift to higher frequencies following the shift of the quasi-Fermi-level E_{Fn} of free electrons towards shallower states. Typical results showing the effect of the optical bias intensity F on the amplitude of the MPC, the out of phase MPC, and the phase-shift spectra are presented in Fig. 2(a), 2(b), and 2(c), respectively. These spectra are measured by keeping the amplitude of the modulated light constant while increasing F. The arrows of Fig. 2 indicate the position of the frequency ν_{Fn} that corresponds to the respective quasi-Fermi-level E_{Fn} in the frequency domain. This frequency was calculated from the dc photoconductivity (σ_{ph}) according to the equation[15] $2\pi\nu_{Fn}=c_n\sigma_{ph}/(\mu e)$, by assuming a mobility $\mu=10$ $cm^2 V^{-1} s^{-1}$ and $c_n=1.2\times10^{-8}$ $cm^{-3} s^{-1}$. Thus, in the spectra measured with increasing F, the shift of the demarcation level defined by the frequency ω_t that corresponds to the break point of $i_{ac}(\nu)$, the maximum of the $i_q(\nu)$, and the inflection point of the $\Phi(\nu)$, follows the shift of E_{Fn} to higher frequencies and to shallower states. The behavior of Fig. 2 was observed in all the studied a-Si:H films[11,12] and it can be taken as a conclusive demonstration of the assignment of frequency ω_t to the position of the demarcation level of the trapped electrons in the frequency domain. In Fig. 3 the ω_t is plotted as a function of the $\omega_{Fn}=2\pi\nu_{Fn}$. By assuming a constant effective capture co-

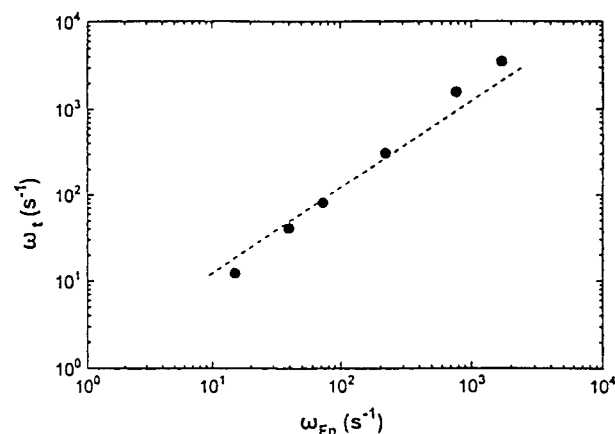

FIG. 3. The demarcation level of trapped electrons ω_t from the MPC spectra of Fig. 2 as a function of the frequency ω_{Fn} that corresponds to the quasi-Fermi E_{Fn} as calculated from the dc photoconductivity. The dashed line is the linear dependence of $\omega_t \propto \omega_{Fn}$.

efficient for the probed states it is observed in Fig. 3 that the relation between ω_t and ω_{Fn} do not significantly deviate from the linear relation (dashed line), indicating that ω_t is roughly proportional to ω_{Fn} ($\omega_t \propto \omega_{Fn}$). Therefore, it can be concluded that the effective capture coefficient of the gap states probed by MPC does not have a significant dependence on the trap depth.

B. The calculated DOS

The density of states (DOS) as a function of frequency calculated from the data of Fig. 2 according to the MPC method[11] is presented in Fig. 4. The top axis represents the energy range that is calculated from $E_\omega=kT\ \ln(c_n N_C/\omega)$ assuming that the interaction of electrons with the defect states above midgap dominates in the MPC. Thus, it was taken $N_C=2.5\times10^{19}$ cm^{-3} and $c_n=1.2\times10^{-8}$ cm^{-3}/s. As the bias light intensity F increases, the calculated DOS, especially at lower frequencies, decreases. This is a characteristic behavior with increasing F because the MPC method measures the unoccupied states that are available for trapping and thermal release. As the quasi-Fermi-level shifts towards shallower

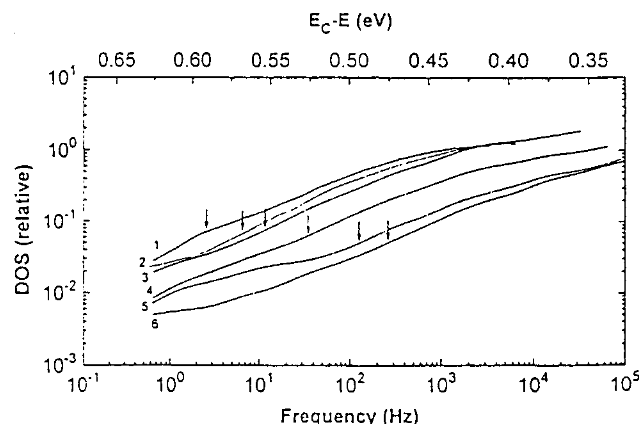

FIG. 4. The DOS calculated from the data of Fig. 2. The arrows show the frequency ν_{Fn} and the numbers are as in Fig. 2.

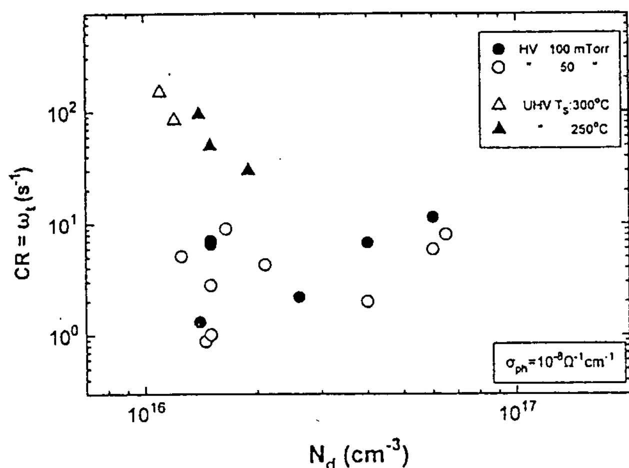

FIG. 5. The effective capture rates of carriers (CR) of undoped a-Si:H samples deposited in the HV or UHV calculated from the MPC spectra that recorded under a constant dc photoconductivity of $\sigma_{ph}=10^{-8}\ \Omega^{-1}\ cm^{-1}$.

states in the band gap, the deeper gap states become populated and in these states the calculated DOS is underestimated. The calculated DOS of Fig. 4 is typical for a-Si:H films measured by the MPC method[11,14,15] indicating that a defect band at 0.35–0.65 eV below E_C dominates electron trapping and recombination.

C. The effective capture rates

In order to study the variations of the calculated effective capture rates (CR) of the carriers by the defects, which are probed by MPC in different undoped a-Si:H samples, the MPC spectra were recorded under a constant dc photoconductivity σ_{ph}. In this way, the quasi-Fermi-level is expected to be fixed at about the same energy level in the band gap.

In Fig. 5 the capture rates of carriers calculated from the MPC spectra according to the discussion in Sec. III A and equation $CR=\omega_t$, are presented as a function of the defect density N_d measured by CPM. The MPC spectra were recorded under a constant photoconductivity $\sigma_{ph}=10^{-8}\ \Omega^{-1}\ cm^{-1}$, on a series of samples deposited in the HV chamber (circles) and on a series of samples deposited in the UHV multichamber (triangles). The series of films from the HV chamber were deposited at 250 °C and at various interelectrode separation distances $D=20$–35 mm and silane pressures of 50 or 100 mTorr. The films from the UHV multichamber system were deposited at 250 or 300 °C, at 100 mTorr of silane pressure and $D=25$ mm. The deposition rate, the defect density, and the photoelectric properties of the films are affected by changing the interelectrode separation and/or the silane pressure.[17] From the series of samples of Fig. 5, it is observed that the calculated CR values do not exhibit a systematic dependence on the defect density. Even in samples with comparable defect density the CR may vary by 1–2 orders of magnitude from sample to sample. In the films deposited in different deposition runs but in the same preparation conditions, the defect density is about the same and the calculated CR may vary by up to a factor of 3.

Since the CR are calculated under a constant dc photoconductivity, $\sigma_{ph}=ne\mu$, the density of free carriers n is kept constant if the mobility μ of carriers is not significantly varied in the studied films. In this case, the fluctuations of th[e] calculated CR in Fig. 5 may originate from variations of th[e] effective capture cross section of the defects. However, it [is] possible that some samples may have a poor mobility μ. [A] lower μ is compensated by an inversely proportional in[crease] of the density n that might result in a enhanced capture rate to be calculated that is not related to a higher capture cross section. On the other hand, a poor μ results in [a] lower diffusive velocity of carriers to the defects,[21,22] whic[h] counteracts the effect of the increase of the capture rate du[e] to the increase of n. Therefore, the variations of the calculated capture rates of Fig. 5 from sample to sample are expected to be practically less dependent on variations of μ an[d] to mainly depend on changes of the capture cross section [o]f the defects.

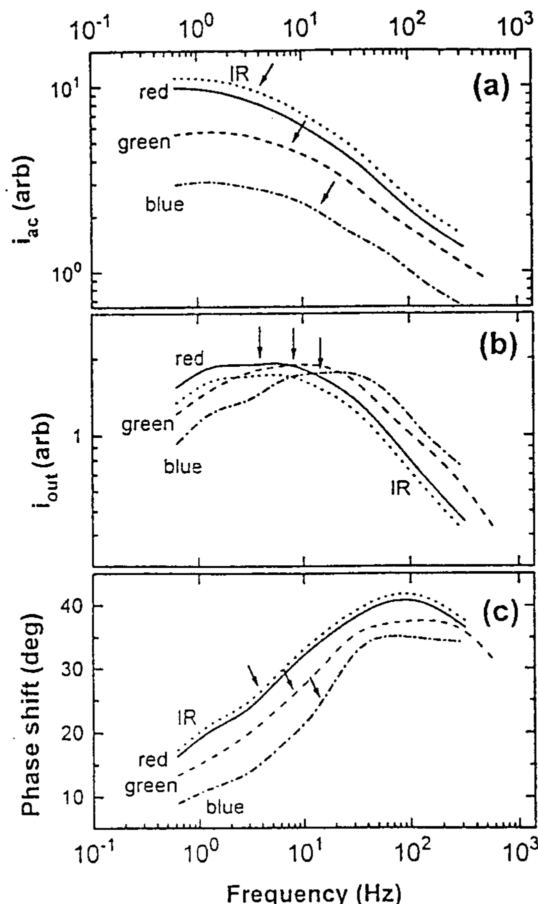

FIG. 6. Experimental $\Phi(\nu)$, $i_{ac}(\nu)$, and $i_q(\nu)$ spectra measured under a co[n]stant photoconductivity and with a blue (470 nm), green (563 nm), red (66[?] nm), and infrared (880 nm) light, respectively, of a UHV sample deposit[ed] at 250 °C. The arrows show the demarcation level of trapped carriers at ω

D. Influence of the surface or interface

The amplitude of the MPC, the out of phase MPC, an[d] the phase shift spectra for a UHV sample deposited at 250 °[C] measured under a constant dc photocurrent are presented i[n] Figs. 6(a), 6(b), and 6(c). The sample is illuminated from th[e] front side using blue (470 nm), green (563 nm), red (66[?] nm), or infrared (880 nm) light. The same results were ob[tained] when the illumination is from the substrate side (no[t] shown here). The arrows indicate the frequency ω_t corre[sponding] to the demarcation level of the trapped carrier[s]

The behavior observed in Fig. 6 is typical for undoped a-Si:H films and it was observed in films with relatively high or low CR.

In order to interpret the above data it must be taken into consideration the following remarks. When the films are illuminated with the strongly absorbed green or blue light, the part of the film within the respective penetration depth of the light, that is less than 200 nm, has predominant contribution to the MPC measurements. On the other hand, when the films are illuminated with the uniformly absorbed infrared light, the contributions of the surface or interface layer to the MPC are minimized. From the fact that the measured spectra with the red light (660 nm) coincide with the respective spectra measured with the uniformly absorbed infrared light (880 nm), it can be concluded that by using a red band-gap light of 660 nm the bulk states of the films are mainly probed, and the eventual contributions of the surface or interface layers are relatively small. From the fact that the demarcation level of the spectra measured with the strongly absorbed blue or green light is shifted to higher frequencies as compared to the respective demarcation level of the spectra measured with the more uniformly absorbed red or infrared light, it is concluded that the effective capture rates of carriers are higher near the surface or interface than in the bulk of the films.

The same shift with decreasing λ was also observed in the respective spectra measured under a constant dc photocurrent and higher bias light intensities than those of Fig. 6. This result excludes the possibility that the shift of the demarcation level of trapped carriers with decreasing λ can be attributed to the band bending because the later is expected to have a dependence on the bias light intensity.

IV. DISCUSSION

The fluctuation of the effective capture rates of the carriers by the defects is 1–2 orders of magnitude in the samples in Fig. 5 and does not represent any systematic dependence on the defect density. From Fig. 5 it is observed that samples with comparable defect density may have substantially different effective capture rates.

The fact that the dbs in a-Si:H may exist in a charged or a neutral state in general may explain the fluctuations of the calculated CR in Fig. 5. Therefore, probably defects in a charged state dominate trapping in films with the highest calculated CR values, while defects in a neutral state dominate trapping in films with the lower calculated CR values. Thus, the shift of the demarcation level towards higher frequencies with decreasing λ of Fig. 6 could indicate that more defects in a charged state eventually dominate trapping near the surface or interface than in the bulk of the films. In fact, if the charge state of the dbs is the main factor that determines the capture rate of the carriers, then in samples with the high calculated CR one should observe a less pronounced shift of the demarcation level to higher frequencies with decreasing λ, because both near the surface and in the bulk of the films the defects in a charged state dominate trapping. However, the strong wavelength dependence of Fig. 6 was observed in samples with high or low calculated CR. Therefore, the defect states near the surface or interface probably act as more effective capture centers for the carriers than the defect states in the bulk of the films, independent of the fact that defects in a charge state or in a neutral state dominate trapping in the bulk of the films. Based on this observation, it can be concluded that the capture process of the carriers by the defects is not only determined by the charge state of the defects but also by the structural environment of the dbs. The structural disorder is probably higher near the surface or interface than in the bulk of the a-Si:H films enhancing the capture process of the carriers.

A similar conclusion has been also derived by light soaking experiments.[15,16] Namely, it has been observed by the OBMPC technique that at the beginning of light soaking the effective capture probability of the carriers, especially that of holes, by the defects, has been found to strongly increase upon light soaking without a change of the defect density.[15,16] The light-induced increase of the capture probability was found to take place uniformly in the bulk and in the surface of the films. When the defect density increases the capture probability of the carriers by the defects increases with a slower rate. Upon thermal annealing at temperatures lower than 150 °C the defect density remains constant while the capture probability of the carriers decreases. Based on these results, it was concluded that the density of strained bonds near the defects probably increases upon light soaking because of light-induced structural modifications that increase the structural disorder. As a result, the capture process by the defects, especially that of holes, which is considered to be the rate limiting step for recombination in a-Si:H, is enhanced upon light degradation. Therefore, the capture rate of carriers may vary even in the same sample and depend on the light soaking and annealing history.

Assuming that the dbs dominate in the trapping and recombination process, the recombination rate (R) is proportional to the dbs density (N_d): $R = C \times N_d$. The proportionality factor (C) is a complicated function[23] of the capture rate of electrons (nc_n) and the capture rate of holes (pc_p). The latter, and the fact that the calculated capture rate of electrons and holes is varied from sample to sample or even in the same sample with light soaking, may explain the experimental result[15,16,24–27] that the photoelectric properties of a-Si:H do not have a good correlation with the defect density N_d.

It is believed that the role of the dbs formation in a-Si:H is to relieve strains from the fourfold coordinated amorphous silicon overconstrained structure. Since a threefold coordinated db is located in a microvoid, the neighbor atoms have some empty space to occupy and the structure is locally more flexible to accommodate internal strains of the disordered structure. Thus, it is reasonable to conclude that some bonds close to the dbs are bent, and so, the db with the bent or strained bonds may act as a cluster of states for the capture process, increasing the effective capture rates of the dbs for electrons and holes, respectively. It is expected that the distribution of strains in the films, which is mainly defined during deposition, and depends on the light soaking history of the sample, may determine the effective capture rate in the dbs. As a result, the capture probability of the dbs is expected to be sensitive to the structural disorder, giving rise to

the widespread of the calculated capture rates of the carriers in the series of samples in Fig. 5, and the increase of the capture rate near the surface and interface of the films in Fig. 6.

V. CONCLUSIONS

The effective capture rates of electrons and holes by the defects measured by OBMPC technique under a constant photoconductivity condition may vary by 1–2 orders of magnitude from sample to sample and does not represent any systematic dependence on the defect density in undoped *a*-Si:H films.

The capture probability of the defects does not represent a significant dependence on the trap depth in the energy gap. The capture rates near the surface or interface are higher than in the bulk of the films.

From the present results, it is concluded that the effective capture cross section of the dbs that dominate electron trapping and recombination above the midgap of *a*-Si:H is not only determined from the charge state of the dbs but also depends on the local atomic environment and the local structural disorder. Since the dbs relieve strains from the disorder structure, it is suggested that strained bonds exist near the dbs. The strained bonds and the db may act as a cluster of states enhancing the capture rates and as a result the capture process is sensitive to the local structural disorder. This may explain the fact that the calculated capture rates significantly vary in the films studied.

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