Dielectric response and its light-induced change in undoped *a*-Si:H films below 13 MHz

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The low frequency (<13 MHz) dielectric response and its light-induced change in undoped *a*-Si:H were investigated in detail. The dielectric constant ε (the real part) in this range decreases with illumination time, following a stretched exponential law similar to that found for other light-induced changes. The saturation relative change was about 0.1–0.2 % for the measured samples. The change is fading away either after repeated illumination-annealing training or by aging at room temperature. The present results indicate some rearrangement of the whole Si network caused by light soaking. [S0163-1829(98)07604-8]

I. INTRODUCTION

The light-induced degradation of *a*-Si:H [Staebler-Wronski effect (SWE)] (Ref. 1) has been drawing much attention since its discovery because it makes a big obstacle toward a fruitful application of the material on one hand, and the effect involves interesting physics in a metastable system on the other. One of the key problems is whether the effect asks for an instability of the whole system, or it only needs some rearrangement of individual bonds, as proposed by most earlier models.[2–5] There is now increasing data showing that the mechanism of SWE is not so simple as was thought earlier. A nuclear magnetic resonance experiment[6] shows that most of the hydrogen takes part in the light-induced enhancement of local motion. A direct evidence of the light-induced increase of Si-H bonds[7] has been given by tracing the change of Si-H stretching mode at 2000 cm^{-1}. The result was questioned by H. Fritzsche,[8] who argued that the change was caused rather by a structural change than by the increase of Si-H bond number. Indeed, there are some indications of such a structural change.[9] Therefore, it would be interesting to look for some more direct evidence of the possible structural change and to see if the structural change alone can account for the light-induced increase of the absorption at the Si-H 2000 cm^{-1} stretching mode observed in Ref. 7.

The absorption of the Si-H vibration modes can be expressed by[10]

$$\int \frac{\alpha}{\omega} d\omega = \frac{N_s}{A}, \tag{1}$$

where α is the absorption coefficient, N_s is the number of Si-H bonds, and A varies as the inverse of the oscillator strength and is related to e^*, the effective charge of the Si-H dipole via the relation[10]

$$A = (cnm\omega_0)/2\pi e^{*2}, \tag{2}$$

where c is the speed of light, n the refractive index, m the reduced mass, and ω_0 the angular frequency of the mode. Generally, parameter A depends on the local-field effect, the screening of the *a*-Si network, and the dipole moment of the Si-H bonds. In a simplified model all of these factors can be related to an effective dielectric constant ε_m by a modification of expression (2) (Ref. 10),

$$A = \frac{(1+2\varepsilon_m)^2(\varepsilon_m)^{1/2}}{9\varepsilon_m^2}. \tag{3}$$

Usually, ε_m is equated to the dielectric constant of the pure Si network. This is an acceptable approximation when the Si-H bond density is not large or clustering is not important. Otherwise, ε_m should also include contributions of Si-H bonds. So, it would not be a bad approximation by equating ε_m to the total dielectric constant ε of the *a*-Si:H film. Therefore, the measurements of ε should shed light on the question of possible light-induced structural change.

Up to now, almost all of the measurements of dielectric response were done by using the optical ellipsometric method.[11] While the method has many advantages, the results are sensitive to the surface condition of samples and the accuracy is not expected to be able to trace the possible subtle change of ε by light soaking. Low-frequency measurements were made by many authors,[12,13] but most of these measurements were devoted either to the space charge effect at the metal-semiconductor interface without taking notice on the dielectric response in the bulk, or only to the frequency-dependent loss of the material.[14]

The present work reports the detailed investigation of the low-frequency dielectric constant of undoped *a*-Si:H as well as its light-induced change. The paper is organized as fol-

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lows. After a description of experimental procedure in Sec. II, we will give the low-frequency dielectric constant ε from our measurements in Sec. III. In Sec. IV, we will show the light-induced change of ε and discuss the results in connection with the observation of the light-induced change in Si-H absorption. The paper will be ended with a brief summary.

II. EXPERIMENTAL

There are two main obstacles in getting reliable data of the low-frequency dielectric response of dipoles and bound electrons in a semiconductor: the contribution of conduction electrons in the conduction band and at the Fermi level, and the interference of space charge at the interfaces. A simple estimation as well as experimental results show that the contribution of conduction electrons is not a serious problem for undoped a-Si:H at room temperature. So great care should be taken to subtract the space charge effect, starting from the sample preparation.

Glass substrates covered with SnO_2 or an indium and tin oxides (ITO) transparent conducting layer were used in order to investigate the light-induced change. To make a good Ohmic contact, a thin (several hundred angstrom) n^+ layer was deposited first. The undoped a-Si:H film was then deposited by the standard plasma-enhanced chemical-vapor-deposition method described in Ref. 7. To decrease the effect of the contact barrier and to avoid any possible leakage, a thin layer of SiO_2 was then deposited before making the upper Al electrode. The SiO_2 layer was proved very useful in filling up pinholes and preventing possible leakage, yet it did not contribute appreciable resistance to the structure. The sample finally forms a $SnO_2/n^+/i/SiO_2/Al$ configuration.

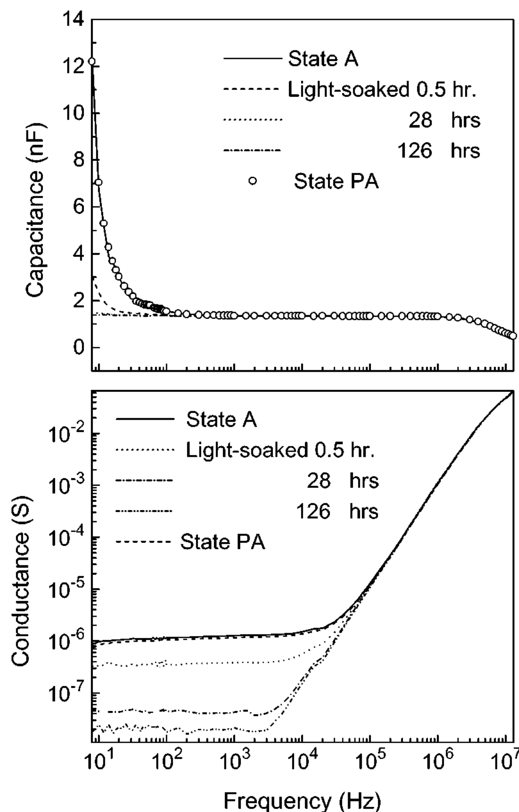

FIG. 1. Capacitance C and conductance G of sample 1 for different light-soaking time. State A is the initial annealed state; State PA the postannealed state.

A sample without the SiO_2 layer was simultaneously made for dc measurements. The results show that the activation energy is 0.68 eV for the sample in the annealed state (state A), and increases to 0.77 eV after light soaking (state B). The dark conductivity at room temperature is 280 nS in state A and decreases to 30.0 nS for state B.

In order that the bulk property dominates, the films should be as thick as possible. The thickness, however, is limited by the growth condition as well as by the depth of the effective light soaking if we want to investigate the light-induced change. The thicknesses of the samples were chosen to be 5–10 μm. Since the accurate information of the thickness is important in deducing ε of the material, we have made careful measurements for each sample. For the sample that we have studied in detail and whose results will be shown below, the average thickness is 8.5 μm. This is obtained by measuring the thicknesses in many different places of the sample, using the Dektek II profilmeter. The results show some thickness fluctuation of about ± 0.2 μm. Taking an arithmetic average of the thicknesses, we estimate that it brings an uncertainty of less than $\pm 2.4\%$ in calculating the absolute dielectric constant of the sample. This uncertainty does not influence the investigation of the change of ε under exposure to light.

The measurements of optical index show that there is at most several percent difference between crystalline Si and a-Si:H.[15] Therefore, we can only expect a very small change of ε by light soaking, if any. To trace this subtle change, the sample was put in a thermostat whose temperature was always kept at 297 K by a computer to within 5 mK. Since our results give a temperature coefficient of less than 10^{-3} K^{-1}, the thermostat eliminates any drift of ε from temperature fluctuation to less than 10^{-5}.

The frequency-dependent capacitance and conductance were measured by a HP4192A LF impedance analyzer from 5 Hz to 13 MHz in a four-lead configuration. The measuring signal level across the sample was kept at 100 mV in all measurements without dc bias on. The nominal accuracy of the instrument is 0.15%, but the stability of the readings is much better, allowing us to track a small change with reproducible results. The reliability of the measurements was checked by a precision air-spaced capacitor of 548 pF and a mica capacitor of 1.47 nF. The capacitance of the former is strictly frequency independent and is ideal for checking the transfer function, while the mica capacitor had a capacitance very close to the value of our sample and the test was essentially a measurement of the dielectric response of mica. For both capacitors the readings were fairly flat up to 1 MHz, but bending somewhat upward above $\sim$3 MHz. It turned out that the readings in this frequency range were sensitive to the contacts as well as the layout of the measurement circuit. However, this small uncertainty does not influence the results presented here that are drawn basically from the data between 10^4 and 10^6 Hz. Since we were interested in the possible light-induced change of the sample dielectric constant, the reproducibility of the measurements was of vital importance. This was checked by measuring the frequency dependence of the sample's capacitance at least five times for every state, and each data point represented an

average of 10 readings. The results show that diversity between different curves was at most less than 0.1 pF, i.e., a relative uncertainty of 7×10^{-5}. The data were also checked by a more accurate LCR meter HP4284 below 1 MHz, confirming the reliability of the results.

The light soaking was carried out by illuminating the sample with white light of 200 mW/cm^2. After successive illumination for several times, the sample was annealed at 200°C in vacuum for one hour to see if the light-induced change is reversible.

III. DIELECTRIC RESPONSE OF *A*-Si:H BELOW 13 MHZ

The capacitance C and conductance G for one of the a-Si:H samples is shown in Fig. 1. Below $\sim$1 kHz there is a strong frequency-dependent behavior, which is obviously caused by the space charge effect at the interfaces. A rapid increase of G and a decrease of C are also found above $\sim$100 kHz, which most likely originates from the effect of contact resistance as we shall see later.

A. The space charge effect at the low frequency end

First we will discuss the contribution of space charge effect, using the equivalent circuit shown in Fig. 2.[15] C_B and R_B are the capacitance and resistance of the bulk sample, and C_I and R_I correspond to the values in the space charge region. It is easy to see that the measured capacitance and conductance are expressed by

$$C=\frac{G_B^2C_I+G_I^2C_B+\omega^2C_IC_B(C_I+C_B)}{(G_B+G_I)^2+\omega^2(C_I+C_B)^2}, \tag{4}$$

$$G=\frac{(G_I+G_B)(G_IG_B-\omega^2C_IC_B)+\omega^2(C_I+C_B)(C_IG_B+C_BG_I)}{(G_B+G_I)^2+\omega^2(C_I+C_B)^2}, \tag{5}$$

where $\omega=2\pi f$ is the angular frequency of the measuring signal. $G_I=R_I^{-1}$ and $G_B=R_B^{-1}$. C_I dominates when $\omega C_B<G_B$, i.e., when the bulk behaves as ''resistive.'' This is so especially at very low frequencies because C_I roughly follows[16]

$$C_I=C_0\left[1+Ln\frac{\psi_s}{(E_c-E_\omega)-E_a}\right]^{-1}, \tag{6}$$

where $C_0=\varepsilon/L_0=\sqrt{q\varepsilon N_0}$, L_0 is the Debye length, N_0 the state density at Fermi energy, ψ_s the built-in potential, E_c the lowest energy of the conduction band, E_ω the cutoff energy, and $E_a=E_c-E_F$ the activation energy. When ω increases, C_I approaches C_B. We can estimate the cutoff frequency ω_c where the space charge effect vanishes. According to Eq. (6), ω_c is determined by

$$E_a=kTLn\left(\frac{1}{\omega_c\tau_0}\right). \tag{7}$$

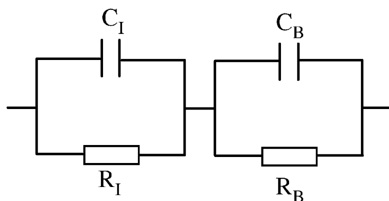

FIG. 2. Equivalent circuit for a sandwich sample with a barrier layer; C_I and R_I are the capacitance and resistance of the space charge region, C_B and R_B the capacitance and resistance of the bulk sample.

The activation energy E_a is about 0.68 eV for our samples. τ_0, the relaxation time of electrons, should have a very broad distribution. By taking a value at the short end, e.g., 10^{-14} sec,[16] we find $f_c=\omega_c/2\pi\approx200$ Hz. We cannot rely too much on this simple model. Fortunately, since the thickness of the sample is 8.5 μm, while the space charge region spans at most 0.2~0.3 μm,[12] the small uncertainty in C_I can be neglected when ω increases to a value where $\omega C_B\gg G_B$. For example, from Fig. 1 we see that $\omega C_B\sim10^{-5}$S at 10 kHz, but G_B is only $\sim2\times10^{-6}$ S at the frequency. Using expression (4), a simple calculation shows that an uncertainty of 1% in C_I leads to an uncertainty in C_B of less than 0.01%. Therefore, we are very safe to deduce the low-frequency dielectric response of a-Si:H when we refer to the data in the frequency range higher than, say, 10^4 Hz.

B. Results at the high-frequency end

Now let us turn to the results in the high-frequency end. We see a rapid increase of G accompanied by a corresponding decrease of C (Fig. 1). The frequency-dependent conductivity in amorphous semiconductors has been investigated a great deal.[14,17–19] The frequency dependence is generally ascribed to the response to an applied ac field of electron states deep within the principal energy gap in the region of the Fermi level. It takes the form[17,18]

$$G_{\mathrm{ac}}=a\omega^s, \tag{8}$$

where a is a constant, the exponent s usually less than unity for most amorphous semiconductors. When we plot our G data in log-log scale according to Eq. (8), we find the exponent in our case is around 2 when $f>100$ kHz. Long[18] ar-

gued that the term is very likely caused by contact resistance $R_0 = G_0^{-1}$. Indeed, if R_0 is small but still cannot be neglected, i.e., $G_0 \gg G_B$, the measured conductance G_m takes the form

$$G_m = \frac{G_B + \frac{\omega^2 C_B^2}{G_0}}{1 + \frac{\omega^2 C_B^2}{G_0^2}}. \tag{9}$$

G_0 also leads to an apparent frequency dependence of measured capacitance, C_m. Again, in the case of $G_0 \gg G_B$,

$$C_m = \frac{C_B}{1 + \frac{\omega^2 C_B^2}{G_0^2}}. \tag{10}$$

We see that G_0 results in an apparent frequency dependence of C_m, especially at the high-frequency end, in accordance with what we observed (Fig. 1). From Eq. (10) we get

$$C_B \approx C_m + \frac{\omega^2 C_m^3}{G_0^2}, \tag{11}$$

where we neglected higher orders in C_m, assuming $\omega^2 C_m^4 / G_0^2 \ll 1$. Since we are interested in C_B, which reflects the bulk property of the sample, we should recover its true value from the measured data, C_m. This can be done by fitting the G_m and C_m data with Eqs. (9)–(11). However, the simple treatment was proved to be satisfactory only below ~ 1 MHz. Therefore, considering the space charge effect at the low-frequency end and the G_0 effect at high-frequency end, we restrict ourselves in the region of $10^4 - 10^6$ Hz in estimating the contribution to C_B from the gap states near the Fermi level, C_{ac}. Using the Kramers-Kraning transform, we may get[18]

$$C_{\text{ac}} = \frac{G_{\text{ac}}}{\omega} \tan\left(\frac{s\pi}{2}\right) = a\omega^{s-1} \tan\left(\frac{s\pi}{2}\right), \tag{12}$$

where a and s can be obtained by fitting the C_B data to the equation

$$C_B = C_\infty + C_{\text{ac}} = C_\infty + a\omega^{s-1} \tan\left(\frac{s\pi}{2}\right), \tag{13}$$

where C_∞ is the nondispersive contribution due to high-frequency atomic and dipolar vibrational transitions. For the data shown in Fig. 1, the best-fitting parameters are $G_0 = 0.06$ S, $s = 0.54$, and $a = 2.24 \times 10^{-10}$. Although this is only a very rough estimation limited by the data accuracy, it does show at least above, say, $f = 10^5$ sec^{-1}; C_{ac} is negligibly small in comparison with C_m. Thus, we conclude that the dominant contribution to C_m around 10^5 Hz comes from dipoles and the polarization of the Si network. The experimental data lead to a dielectric constant $\varepsilon = 13.5$, about 15% larger than the optical value. To our knowledge, this is the first report about the low-frequency ε of a-Si:H.

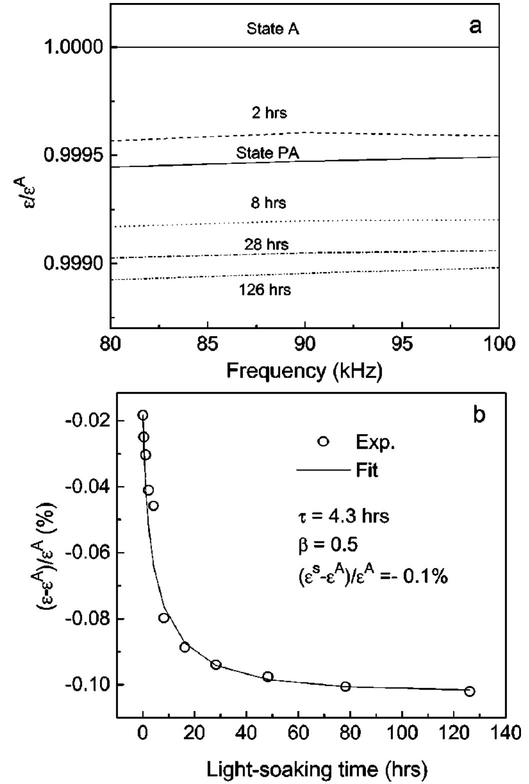

FIG. 3. (a) The relative dielectric constant $\varepsilon/\varepsilon^A$ between 80 and 100 kHz for state A and different light-soaking time; ε^A is the ε value in state A. The curve state PA represents the $\varepsilon/\varepsilon^A$ data after postannealing for 1 h at 200 °C. (b) Light-induced decrease of relative dielectric constant $\varepsilon/\varepsilon^A$ at 10^5 Hz as a function of light-soaking time. The fitting curve is according to Eq. (14) with parameters shown in the figure.

IV. LIGHT-INDUCED CHANGE OF DIELECTRIC CONSTANT ε

As we mentioned in the beginning of the paper, the most interesting question is the possible light-induced structural change, which may leave its fingerprint on the dielectric constant ε.

Altogether five samples deposited in the same run have been measured. Among them, four samples did show some light-induced change in ε. The typical result is shown in Fig. 3. We see that ε decreases with illumination time [Fig. 3(a)]. The decrease of ε approximately follows a stretched exponential rule [Fig. 3(b)]. The exponential rule can be expressed as

$$\frac{\varepsilon - \varepsilon^A}{\varepsilon^A} = \frac{\varepsilon^S - \varepsilon^A}{\varepsilon^A} \left\{ 1 - \exp\left[-\left(\frac{t}{\tau}\right)^\beta \right] \right\}, \tag{14}$$

where ε^A is the ε value in state A and ε^S is the saturation value in state B. For the data shown in Fig. 3 we get $\tau = 4.3$ h and $\beta = 0.5$. The saturated $(\varepsilon^A - \varepsilon^S/\varepsilon^A) \approx 0.1\%$. Annealing at 200 °C for 1 h can reverse the greater part of the change [see Fig. 3(a)].

A very interesting phenomenon was that the light-induced decrease in ε weakened when we measured one of the samples (3) again a week later. Although the light-induced decrease of ε was still there, the saturated relative change of

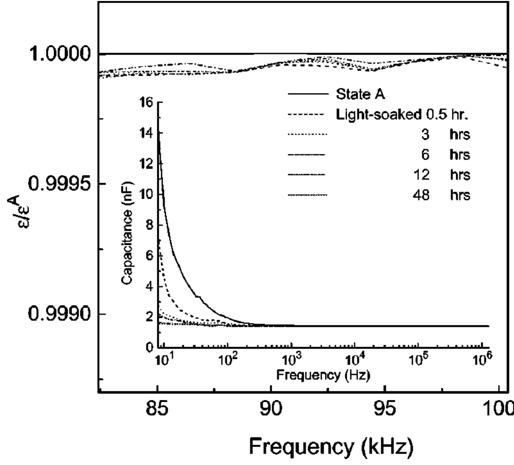

FIG. 4. The relative dielectric constant $\varepsilon/\varepsilon^A$ of sample 4 in the frequency range of 80–100 kHz for state A and different light-soaking time. The sample was measured after putting it on the shelf for six months.

ε decreased to 0.07%, in contrast to the value of 0.14% obtained in first measurement. When we measured the sample for the third time six months later, the light-induced change was gone. To check if the phenomenon was only a fortuity, we remeasured another one (4) of the four samples. Again, the ε change disappeared for this sample, though the change in space charge effect by illumination still existed (Fig. 4). Then we took one sample (5), which was not measured before but was put on the shelf for ten months. This time we did not find any light-induced change in ε within the resolution of our measurements (10^{-4}). These experiments show that the light-induced change in ε can be faded away either by repeated illumination-annealing training or by the aging effect.

Two questions then arise. Does the ε change observed here reflect a structural change of the Si network, or is it simply caused by some local change of electron states, resulting in a change of C_{ac}? If the ε change does come from the structural change, is it enough to account for the light-induced increase of Si-H bond absorption at 2000 cm^{-1} observed in Ref. 7?

It would be easy to answer the first question if we knew exactly how large C_{ac} is and how it changes with illumination. As mentioned in Sec. III, C_{ac} is negligibly small in comparison with C_m. In fact, C_{ac} is of the order of ~ 1 pF at $f = 10^5$ Hz, that is, $\sim 0.1\%$ of C_m. Unfortunately, it is much more difficult to determine the light-induced change of C_{ac} because of the limited accuracy of the experimental data. Nevertheless, there are reasons in favor of a structural change. First, our experiments and previous work[19] show that G_{ac} increases with illumination, though in our case the changes were not so large as to give a functional dependence. The increase of G_{ac} with illumination is reasonable, considering the possible increase of gap states near the Fermi level by light soaking. Therefore, the ε change observed here is in the direction opposite to what should be expected from the change of C_{ac}. Secondly, because the light-induced change in space-charge-related capacitance is directly related to the electron states in the material, if the ε change is caused by the change of electron states, it should be observed as long as the effect in the low-frequency range exists, in contrast to what we have found here. Thus, the present measurements give strong support to a light-induced structural change of the whole Si network. The structural relaxation for a relatively long period or by illumination-annealing training can make the network of a-Si:H more stable, which may weaken the light-induced rearrangement of the a-Si:H network, which was why the light-induced change in ε could not be detected for the long-term reserved samples. As for the second question, regarding whether the ε change is large enough to account for the light-induced increase of the Si-H bond absorption α, a simple estimation shows that the ε change is far from enough to give a change observed in Ref. 7. From Eqs. (1) and (3), we have

$$\alpha_i = \int \frac{\alpha d\omega}{\omega} = \frac{N_s}{A} = \frac{9\varepsilon_m^2 N_S}{(1+2\varepsilon_m)^2 \varepsilon_m^{1/2}}, \tag{15}$$

where α_i is the integrated absorption of the Si-H bonds. Assuming that the light-induced change of α_i is only caused by the change of A or ε_m, then, by differentiation (15), we get

$$\frac{d\alpha_i}{\alpha_i} = \frac{dA}{A} = \left(\frac{4\varepsilon_m}{1+2\varepsilon_m} - \frac{3}{2}\right)\frac{d\varepsilon_m}{\varepsilon_m}. \tag{16}$$

Taking $\varepsilon_m \sim \varepsilon = 13.5$, Eq. (16) becomes

$$\frac{d\alpha_i}{\alpha i} \approx -0.43\frac{d\varepsilon}{\varepsilon}. \tag{17}$$

The measurements in Ref. 7 give a total increase of $d\alpha_i/\alpha_i$ as large as 1%. To account for this change, we need a decrease in ε of about 2.3%, more than an order larger than the saturated ε change observed in present measurements. Therefore, the light-induced enhancement of Si-H absorption at a 2000 cm^{-1} peak could be closely related to some structural change, accompanied by an increase in the Si-H bond number, but it is unlikely that it is caused merely by a structure-related change of the effective dielectric constant. Recently, a model was proposed that explained the light-induced increase of absorption at 2000 cm^{-1} by the transformation of some monohydrides precipitating in hydrogen-rich clusters into isolated Si-H bonds.[20] Similar to some earlier models, the model proposed in Ref. 20 is essentially also based on a change of some individual bonds instead of the whole network. It is not clear if such interchange between isolated and clustered Si-H bonds can account for the light-induced change of ε found here as well as its fading away with elapsed time of samples.

V. CONCLUDING REMARKS

We have measured the low-frequency dielectric constant ε of undoped a-Si:H films and traced its change with light-soaking. Careful analysis of the data leads to $\varepsilon = 13.5$, a value substantially larger than the optical one. This is not unusual as the optical measurements only feel the response

of electrons. As is shown in Sec. III, electrons in deep states near the Fermi level only contribute a very small part of ε.

Our measurements of dielectric constant ε as a function of light-soaking time show that the light-induced structural change does exist in a-Si:H films. The process is stronger for freshly made samples. The change is fading away either by repeated illumination-annealing training, or by putting the samples on the shelf for a long enough time, indicating that the structure becomes more stable in the process. Although our results confirm a light-induced structural change, the latter does not lead to a change in ε that is large enough to account for the light-induced increase in Si-H bond absorption at 2000 cm^{-1}.

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