

## Light-induced change in defect-band photoluminescence of doped hydrogenated amorphous silicon

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A much larger decrease of the defect-band photoluminescence than that of the main band is observed in phosphorus-doped *a*-Si:H after light soaking. Accompanying this change, the dark conductivity, the photoconductivity, and the sub-band-gap absorption all increase. The experimental results coherently favor a larger increase of donor states than dangling-bond states during light soaking. A possible decrease of neutral-dangling-bond density (under illumination) is also indicated.

### I. INTRODUCTION

It is now generally recognized that all the metastable effects in *a*-Si:H have a common origin related to the deviation of the Fermi level from equilibrium position.<sup>1,2</sup> However, many microscopic mechanisms still remain as possibilities, such as weak-bond-to-dangling-bond conversion involving hydrogen motion<sup>3,4</sup> or not involving hydrogen motion,<sup>5,6</sup> weak-bond-to-dangling-bond and -floating-bond conversion,<sup>7</sup> mechanism of charge-trapping-induced transitions,<sup>8-10</sup> and mechanism involving electron sensitizing centers.<sup>11</sup> Here we present experimental results on heavily doped *a*-Si:H from photoluminescence (PL) and conductivity measurements. The PL spectra of P-doped *a*-Si:H generally consist of two bands, one at 0.75 eV, and the other at 1.2 eV.<sup>12,13</sup> The main-band luminescence at 1.2 eV is attributed to radiative recombination between trapped electrons and holes in the corresponding tail states.<sup>13</sup> The efficiency is influenced by several nonradiative-recombination mechanisms, one of which is the electron tunneling to dangling bonds. On the other hand, the luminescence band at 0.75 eV is generally attributed to optical transitions involving dangling bonds. One may expect that the defect-band luminescence grows with increasing dangling-bond density while the main-band luminescence decreases. In our observations, both PL bands decrease with light soaking in heavily P-doped *a*-Si:H. We suggest that the efficiency of defect-band luminescence is determined by a competition between the radiative transition from the mobility edge to neutral dangling bonds and nonradiative electron tunneling from tail states and donor states to dangling bonds. The observed fatigue of the defect-band PL then can be explained by a larger increase of donors than dangling-bond defects during light soaking, while the fatigue of the main-band PL is still attributed to the increase in dangling-bond density. The result also implies a decrease of neutral-dangling-bond density during light soaking. These conclusions are further supported by the

observed increases in conductivity and photoconductivity. Furthermore, the fatigue of the defect band is nearly independent of the temperature at which the light soaking was done. All these results provide strong constraints on the possible mechanisms of metastable effects.

The experimental results are presented in Sec. III and will be discussed in Sec. IV (photoluminescence in IV A, photoconductivity in IV B, mechanism of metastable changes in IV C). The conclusions are outlined in Sec. V.

### II. EXPERIMENT

The samples are glow-discharge deposited doped and undoped *a*-Si:H made at the Solar Energy Research Institute (SERI) (Golden, CO) and at Chronar, with thicknesses of about 0.5  $\mu\text{m}$ . The *n*-type samples, with doping ratios of 0.1–1%, show an activation energy of about 0.2 eV for the dark conductivity.

The photoluminescence is excited by an Ar-ion laser at 488 nm. A PbS photoconductive detector cooled to dry-ice temperature is used for photon detection. The influence of light soaking on the luminescence efficiency was recorded in several ways: One is to record the PL signal versus time while the laser is illuminating the sample. We have taken care to be sure that any change in PL is not due to possible change in sample or detector temperature, or to optics instability. The PL from a GaAs standard was occasionally checked and recorded for this purpose, and was found to be stable within 1% during an hour of illumination. The other technique is to compare the PL of the light-soaked area with that of a fresh area using a much lower laser intensity.<sup>6</sup> For undoped *a*-Si:H, one can observe a small dip of the luminescence peak at the previously light-soaked position while scanning the sample.

For the conductivity measurements, the electrodes were made either by evaporation in vacuum following sample deposition or by silver painting. Optical-

absorption spectra before and after light soaking were measured by the constant photocurrent method,<sup>14</sup> without moving the sample. Light-induced changes in photoconductivity were either recorded during PL measurement, or recorded separately during light soaking by a halogen lamp with a similar photon flux as the laser.

### III. RESULTS

#### A. Photoluminescence

The PL spectra of the doped samples consist of two bands: the defect band at 0.75 eV and the main band at 1.2 eV, which is in agreement with previous studies. During light soaking, we found that the luminescence of both bands decreases. The fatigue of the main band is similar to the fatigue of the main band of undoped *a*-Si:H. At 78 K and under a laser excitation of 120 mW/cm<sup>2</sup>, the fatigue in 10 min is a drop by about 6% of the initial luminescence intensity. The defect band, however, exhibits a much larger decrease. As illustrated in Fig. 1, the decrease is about 22% in 8 min.

From Fig. 1, it is clear that the relative decrease of the defect-band PL does not obey a simple power law. In agreement with Redfield and Bube,<sup>15</sup> we found that it is better to fit the time-dependences data to a stretched exponential of the form

$$\frac{I_{\text{PL}}(t) - I_{\text{PL}}(s)}{I_{\text{PL}}(0) - I_{\text{PL}}(s)} = \exp \left[ - \left( \frac{t}{\tau} \right)^\beta \right], \quad (1)$$

where  $I_{\text{PL}}(0)$  is the PL intensity  $I_{\text{PL}}(t)$  at  $t=0$ ,  $I_{\text{PL}}(s)$  is the saturation value. A plot according to Eq. (1), which plots the logs of both sides of Eq. (1), is shown in Fig. 2.  $\beta$  is the slope of such a plot. For clarity, only data at 78 K are shown. This behavior brings to mind the finding of Jackson and Moyer<sup>16</sup> for the generation of defects by carrier accumulation. They found that  $\tau$  is activated with an activation energy of about 1 eV and  $\beta$  is proportional to temperature. In the present case, the temperature dependence of  $\tau$  is shown in Fig. 3. The activation energy of  $\tau$

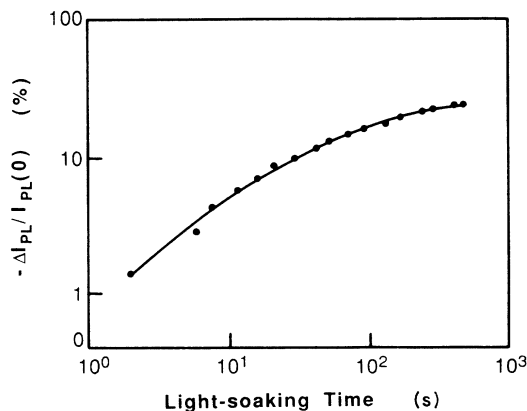


FIG. 1. Fatigue of defect-band luminescence during laser light soaking at 78 K for a sample corresponding to 1% gas-phase phosphorus doping.

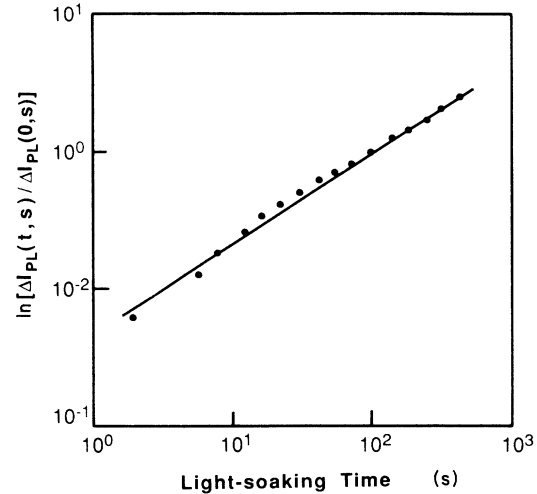


FIG. 2. Time-dependent fatigue of defect-band luminescence at 78 K fitted to a stretched exponential.

is less than 0.01 eV. Furthermore, we found that  $\beta=0.66\pm 0.02$  is actually temperature independent. This difference indicates that the mechanism of the metastable effect may not involve hydrogen motion.

Similar to the temperature dependence of  $\tau$ , the relative decrease of the defect-band PL is also slightly smaller at higher temperature. This is a general observation for the five P-doped samples investigated here.

Figure 4 shows the relative decrease  $-\Delta I_{\text{PL}}/I_{\text{PL}}$  versus laser intensity used for light soaking, measured at 78 K. Data for an undoped sample is shown there for comparison. Since the luminescence efficiency of the defect band is low, the data  $\Delta I_{\text{PL}}/I_{\text{PL}}$  is less accurate at the lower laser intensities. However, compared to the relative fatigue of the main band of undoped samples, it is clear that the fatigue of the defect band shows a stronger intensity dependence.

The light-induced changes of both bands can be annealed out by heating the sample to 150 °C.

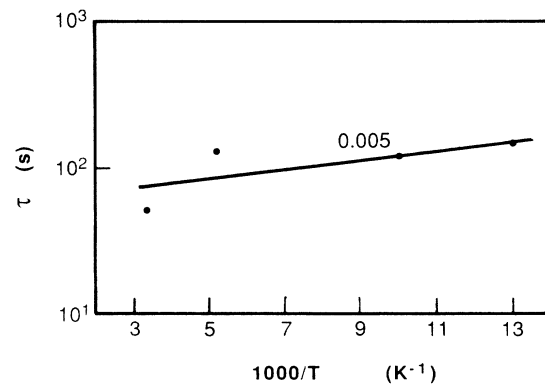


FIG. 3. Temperature dependence of time constant  $\tau$ , which is defined in Eq. (1). Fitting the data to an exponential form (straight line in the figure), the activation energy of  $\tau$  is about 0.005 eV.

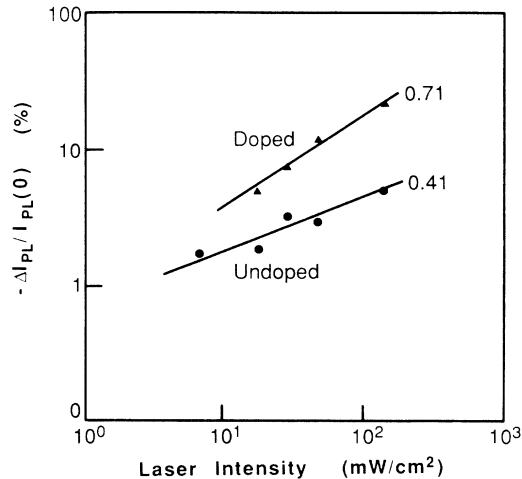


FIG. 4. Relative change of photoluminescence at 0.7 eV for a P-doped sample, measured at 78 K for a light-soaking time of 7 min. Photoluminescence at 1.3 eV for an undoped sample is shown here for comparison. The numbers in the figure represent the slopes.

### B. Conductivity and optical-absorption spectra

For heavily doped *a*-Si:H, a light-induced increase of photoconductivity (PC) and dark conductivity was reported.<sup>17</sup> Here we carried out the experiment at lower temperatures, 78, 193, and 294 K. The photoconductivity response was measured across a series resistor  $R$ , the  $RC$  time constant of the circuit being about 10  $\mu$ sec. For the heavily doped samples, the response clearly shows both a fast process (ms) and a slow process. At low temperature, the fast process dominates, while at room temperature the slow process dominates. This point is illustrated in Fig. 5 for PC response under AM1 excitation at 198 K. Figure 5(a) shows the PC response during the first minute after light on or off; the fast and slow processes are visible. Figure 5(b) shows another even slower process where PC increases gradually in the first 10 min and saturates afterwards. We attribute this increase to light-induced structural change. However, the PC response at room temperature is too slow to extract from it the structure-change-related PC increase. The slow increase of PC as in Fig. 5(b) during the 1–10 min time slot amounts to 3–13 %, depending on sample and temperature.

After light soaking at 294 K for 1 h, the dark conductivity shows an increase of 10%, which is measured a day later so that the photocarriers have decayed away. Light soaking at 78 K increases the dark conductivity factor of 2, while no change in the activation energy could be found.

For the doped samples, the absorption spectra were measured by the constant PC method at 78 or 100 K. Skumanich *et al.*<sup>18</sup> found from photothermal deflection spectroscopy an increase of about 30% in the defect density after a 1-h 1-W/cm<sup>2</sup> light soaking, for doping levels from 1 ppm to 1%. However, we found a much smaller increase, which may be due to the 10 times lower light in-

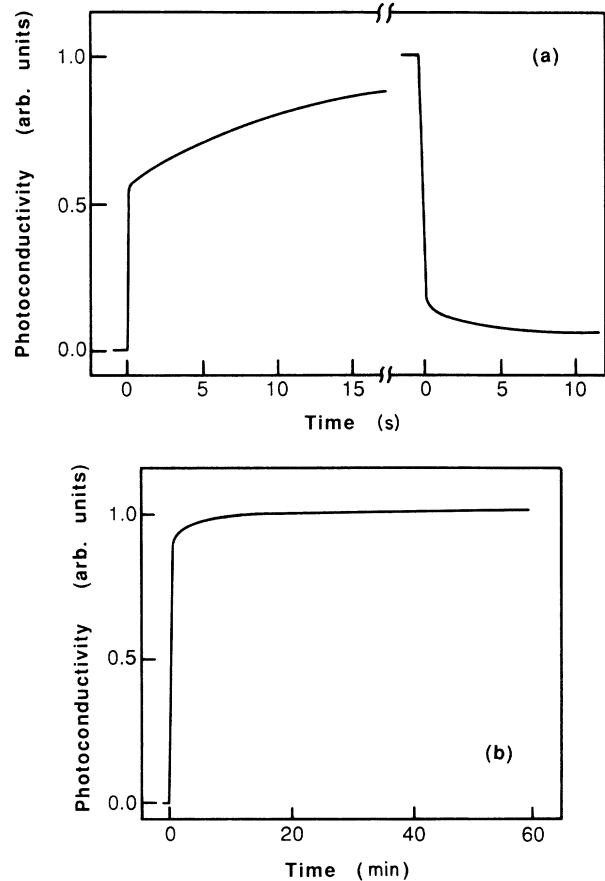


FIG. 5. Photoconductivity response of a heavily P-doped sample to AM1 at 198 K. (a) During the first minute following light-on or light-off, a fast and a slow process are visible, and (b) a gradual increase in the first 10 min, which is related to light-induced structure change.

tensity we used for light soaking. Figure 6 shows a typical result: the increase of absorption coefficient in the sub-band-gap region is about 6%, i.e., about  $7 \times 10^{16}$  defects/cm<sup>3</sup> were generated. For the undoped samples, on the other hand, we found from the absorption spectra an increase nearly the same as reported by Skumanich

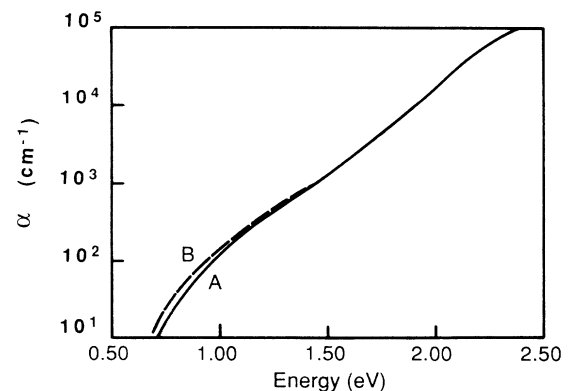


FIG. 6. Light-induced change in absorption spectra of 1% gas-phase, phosphorus-doped *a*-Si:H, measured at 100 K by the constant photoconductivity method. *A* is the annealed state, *B* is the light-soaked state.

*et al.*<sup>18</sup> and Dersch *et al.*<sup>19</sup>

In summary, we have observed fatigue of both luminescence bands in heavily P-doped *a*-Si:H; and the fatigue of the defect band is the more pronounced of the two. Accompanying these changes, we found increases in defect density, in dark conductivity, and in photoconductivity.

#### IV. DISCUSSION

Two results are well established:<sup>12,20,21</sup> (1) the main-band luminescence at 1.2–1.4 eV decreases with doping for doped samples or decreases with increasing dangling-bond density for undoped samples; (2) the defect-band luminescence peaks at a doping level of about  $10^{-4}$  for doped samples and peaks at a defect density of about  $10^{18}/\text{cm}^3$  for undoped samples. That is to say, when the doping level or the defect density is too high, both luminescence bands decrease with increasing doping or defect density. Therefore, our observation that both PL bands decrease in heavily doped *a*-Si:H during light soaking can be explained phenomenologically as a light-induced increase in either doping level or dangling-bond defect density.

As discussed by Branz,<sup>9</sup> the increase of dark conductivity after light soaking is an indication of increased donor concentration. This point suggests the analogy between light soaking and increasing doping level. We have made a detailed comparison between the effects of increasing doping level and the effects of light soaking, as shown in Table I. Besides the common points we just mentioned, there are two major differences. One is that the ratio of the defect-band PL to the main-band PL increases with doping while this ratio decreases with light soaking; the other is that the  $\mu\tau$  product from PC for heavily doped samples decreases with doping while it increases with light soaking.

These two differences imply that the reaction mechanism during light soaking is different from the reaction mechanism during equilibrium deposition.<sup>23</sup> In the following, we will illustrate these two differences in more detail. In IV A, we shall explain the fatigue of both PL bands. The fatigue of the defect-band luminescence is attributed to the decreased density ratio of neutral dangling bonds to donors; while the fatigue of the main-band luminescence is still attributed to the increase in dangling-bonds density. In IV B, we shall show that the increase of photoconductivity during light-soaking results

TABLE I. Comparison between the effects of increasing doping and the effects of light soaking in the heavily doped region.

Quantity Manner	Dark				
	$I_{\text{PL}}$ (0.75)	$I_{\text{PL}}$ (1.2)	conduct- ivity	$\mu\tau$ product	$I_{\text{PL}}(0.75)/$ $I_{\text{PL}}(1.2)$
Doping	↓ <sup>a,b</sup>	↓ <sup>a,b</sup>	↑	↓ <sup>a,c</sup>	↑
Light- soaking	↓	↓	↑	↑	↓

<sup>a</sup>From Ref. 12.

<sup>b</sup>From Ref. 20.

<sup>c</sup>From Ref. 22.

from more light-induced donors than dangling bonds. Finally, in IV C, we shall discuss the mechanism involved in the light-soaking process.

In the discussion below, we follow Adler's notation to denote a defect center by  $A^x_n$ , where  $A$  is  $T$  or  $P$ , representing Si or phosphorus, respectively;  $n$  is the coordination number, and the symbol  $x$  (+, − or 0) on the upper right corner represents the charge states of the center. We also represent the three charge states of dangling bonds by  $D^+$ ,  $D^0$ , and  $D^-$ . For each species  $A^x_n$ ,  $[A^x_n]$  denotes its density. For heavily P-doped *a*-Si:H, the density of neutral dangling bonds  $[D^0]$  is negligibly small in thermal equilibrium. However, this is not the case when the sample is under light illumination. As illustrated by light-induced electron-spin-resonance (LESR) experiments,<sup>20</sup> about 10% of the dangling bonds are in the neutral configuration; therefore, hereafter we refer to  $[D^0]$  as the neutral-dangling-bond density under light illumination.

##### A. Fatigue of both luminescence bands

Although there are several detailed models for defect-band luminescence, we will adopt the model suggested by Street *et al.*<sup>24</sup> In this model, the defect-band luminescence represents a transition from the mobility edge  $E_c$  to neutral dangling bonds. For the calculation of the luminescence efficiency, several possibilities exist. Here we favor the earlier suggestion of Street, i.e., transition of electrons into any dangling bonds can proceed either radiatively or nonradiatively.<sup>24</sup> We assume that electron transition from mobility edge to  $[D^0]$  is radiative, while electron tunneling from tail states and donor states to  $[D^0]$  is nonradiative. As a result, the transition rate is proportional to the optical transition probability  $b^0$ , and to the neutral-dangling-bond density  $[D^0]$ . Denote the tail-state density by  $N_{ct}$ , and the donor-state density by  $[P^+_4]$ ; then the PL efficiency of the defect band is (see the Appendix)

$$\eta_{0.75} \propto b^0[D^0]/(b_n N_{ct} + b^+[P^+_4] + b^0[D^0]), \quad (2)$$

where  $b_n$  and  $b^+$  are appropriate constants. Equation (2) ensures that the defect-band luminescence shows the same thermal quenching as that of neutral dangling bonds.<sup>26</sup> In addition, Eq. (2) can also represent the doping dependence of the defect-band PL quite well. At low doping levels, the tunneling from tail states dominates, PL increases with doping since  $[D^0]$  increases. At high doping levels, tunneling from donor states can dominate because at high doping levels ( $\geq 10^{-3}$ ) most electrons occupy the donor states rather than tail states.<sup>27</sup> Now, PL could decrease with increasing doping since the ratio  $[D^0]/[P^+_4]$  decreases (e.g., see data in Ref. 20). Applying Eq. (2) to the case of light-induced changes for heavily doped samples, we arrive at the conclusion that light induces a decrease of the ratio  $[D^0]/[P^+_4]$ .

We attribute the main-band PL to radiative recombination between electrons trapped in conduction-band tail and donor states, and holes trapped in valence-band tail states. According to Street,<sup>13</sup> the efficiency of the main-band PL is

$$\eta_{1,2} = \exp[-4\pi(R_c)^3 N_s / 3], \quad (3)$$

where  $R_c$  is a critical radius within which the trapped electrons will tunnel to dangling bonds nonradiatively; while  $N_s$  is the dangling-bond density. We see immediately from this equation that the fatigue of the main band may be due to light-induced increase of  $N_s$ . From Eqs. (2) and (3), the ratio of defect-band efficiency to that of main band is

$$r \propto b^0 [D^0] \exp[4\pi(R_c)^3 N_s / 3] (b_n N_{ct} + b^+ [P^+_4]). \quad (4)$$

The dangling bond and donor densities in as-deposited, doped Si:H are determined by auto-compensation reaction under equilibrium,<sup>23</sup> and  $[P^+_{4}] \approx [D^-]$ . As a result, this ratio increases with doping for as-deposited samples. It will decrease with light soaking if the increase of  $\exp[4\pi(R_c)^3 N_s / 3]$  is less than the decrease of  $[D^0]/[P^+_{4}]$ . Equations (2)–(4) are in agreement with the experimental results, which show a decrease of  $r$  by 17%, a decrease of  $[D^0]/[P^+_{4}]$  by 22%, and an increase of  $N_s$  by 6%. Considering the decrease of  $r$  from a different angle, it may reflect decreases of both  $[D^0]$  and  $\exp[4\pi(R_c)^3 N_s / 3]/[P^+_{4}]$ . The decreases of  $[D^0]$  and  $[D^-]/[P^+_{4}]$  are suggested by the increase of photoconductivity, as will be discussed below. The point that light soaking produces more donors than dangling bonds is also inferred by Stutzmann *et al.* from LESR measurements.<sup>3</sup>

### B. Photoconductivity

We have observed that light soaking induces an increase in photoconductivity. In this section, we shall show that this is related to more light-induced P donors than dangling bonds. Furthermore, considering the bimolecular nature of photocarrier recombination, the hole density (including those in  $D^-$ , i.e.,  $[D^0]$ ) decreases after light soaking. These points are illustrated by the following mathematics: Denote the electron density in the conduction band and conduction-band tail states by  $n$ , and denote the density of holes in valence band and valence-band tail states by  $p$ . Then the charge neutrality condition is  $n + [D^-] = p + [P^+_{4}]$ . Or by denoting  $N_s \equiv [D^0] + [D^-]$  and  $[P^{0,+}_4] \equiv [P^0_4] + [P^+_{4}]$ ,

$$P = N + N_s - [P^{0,+}_4], \quad (5)$$

where  $N = n + [P^0_4]$  is the total electron density above the Fermi level, and  $P = p + [D^0]$  is the total hole density below it. Under illumination, let us assume that  $N_{op}$  electrons and  $P_{op}$  holes are generated. Then the charge neutrality condition under illumination becomes

$$P_{op} = N_{op} + \Delta(N_s - [P^{0,+}_4]), \quad (6)$$

where  $\Delta(N_s - [P^{0,+}_4])$  represents a possible light-induced structural change. In the steady state, where the carrier generation rate  $G$  is balanced by recombination, we have

$$G = RP_{op}N_{op} \quad (7a)$$

and

$$N_{op} = \sqrt{G/R'}, R' = R \{1 - \Delta([P^{0,+}_4] - N_s)/N_{op}\} \\ \equiv R(1-x) \quad (7b)$$

or denoting  $\sqrt{G/R}$  by  $N_{op}$  of the annealed state  $A$ ,

$$(N_{op})_{LS} = (N_{op})_A (1+x/2), \quad (7c)$$

where  $R$  is the average recombination coefficient, and the subscript  $_{LS}$  denotes the value during the process of light soaking.

Note that electron hopping is the dominant transport mechanism in heavily P-doped  $a$ -Si:H,<sup>28</sup> and PC is therefore proportional to  $N_{op}$ . Equation (7b) produces the right intensity dependence of photoconductivity.<sup>22</sup> From Equation (7c), we can explain the increase of PC during light soaking by a structural reaction which induces an increase of  $[P^{0,+}_4] - N_s$ . This idea can be further checked quantitatively.  $N_{op}$  from LESR is about  $2 \times 10^{17} \text{ cm}^{-3}$ .<sup>20</sup> From Equation (7c), a 3–13% increase in  $N_{op}$  gives  $\Delta([P^{0,+}_4] - N_s) = 6\text{--}26\% N_{op}$ , i.e.,  $\Delta[P^{0,+}_4] \approx 10^{17} \text{ cm}^{-3}$ . Considering the coarseness of the model, this result agrees with Stutzmann *et al.* quite well.<sup>3</sup> Furthermore, Eq. (7a) implies a decrease of  $P_{op}$  and/or  $[D^0]$  after light soaking. Therefore, the point that light soaking induces more donors than dangling bonds is further supported by the photoconductivity results.

### C. Mechanism involved in light-induced changes

Up to now, we have not considered any detailed mechanism for the light-induced changes. As discussed in Secs. IV A and IV B, the experimental results favor those mechanisms which induce more P-donor states than dangling-bond states. In addition to a mechanism which produces only dangling bonds, a new mechanism to activate P donors during light soaking is desirable. Furthermore, we have also presented in Sec. III A the stretched exponential time dependence of the fatigue, with a weakly temperature-dependent time constant. All these observations provide strong constraints on the possible mechanisms. For example, the small activation energy of less than 0.01 eV for the time constant cannot be explained simply by the hydrogen motion model.<sup>3</sup> It seems to us that the carrier trapping and bond-switching model of Branz<sup>9</sup> is an appealing mechanism. We outline here the main points of this suggestion.

Let us consider the hole-trapping reaction, which induces a decrease of  $[D^0]$  and an increase of  $[P^+_{4}]$ :<sup>9</sup>



Upon light soaking, the hole density is much higher than its thermal equilibrium density. Then reaction (8) is pushed to the right-hand side. The generation rate of  $P^+_{4}$  should be proportional to the difference between its present density and the steady-state density  $[P^+_{4}(s)]$ . The generation rate should also be proportional to the density of holes, the density of  $D^0$ , and the hole-trapping coefficient  $b_p$ . From Sec. IV B, we know  $p[D^0] \propto G$ . Furthermore, there is experimental evidence that the trapping coefficient decays with time as a power law,<sup>29</sup> i.e.,  $b_p \propto bt^{-a}$ . So we expect

$$d[P^+_4]/dt \propto bGt^{-a}([P^+_4(s)] - [P^+_4]) . \quad (9)$$

This equation will give a stretched, exponential time dependence of  $[P^+_4]$ , allowing us to interpret the PL fatigue with  $\beta = 1 - a$ . Another advantage of the trapping model is that it permits us to explain the slightly smaller  $\tau$  at higher temperatures, since the capture coefficient  $b_p$  is somewhat smaller at higher temperatures.<sup>30</sup>

## V. CONCLUSIONS

Light-induced effects in heavily P-doped *a*-Si:H were investigated. Based on the experimental results, we suggest the following.

(1) The increase of donors is more than the increase of dangling-bond defects.

(2) In spite of the increase of total dangling-bond density, the density of neutral dangling bonds  $[D^0]$  decreases in the presence of light illumination.

(3) The hole-trapping reaction may be an essential reaction during light soaking in heavily P-doped materials.

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## APPENDIX

Here we derive Eq. (2), i.e., to find the fraction of recombination events at dangling bonds which are radiative. We assume that the electron transition from the mobility edge to  $D^0$  is radiative, while electron tunneling from localized states to  $D^0$  is nonradiative. From another point of view, we may consider neutral dangling bonds as recombination sites causing radiative recombination of diffusing electrons, and nonradiative recombination of trapped electrons. Denote the electron density above the mobility edge by  $n_f$ , the radiative recombination rate constant by  $b^0$ , then the radiative rate is  $n_f b^0 [D^0]$ . Denote the electron density in tail states and donor states by  $n_t$ , the average nonradiative recombination time by  $\tau$ , then the nonradiative recombination rate is  $n_t / \tau$ . Therefore, the luminescence efficiency of the defect band is

$$\eta_{0.75} = n_f b^0 [D^0] / (n_f b^0 [D^0] + n_t / \tau) . \quad (A1)$$

Under steady-state illumination,  $n_t$  will be proportional to  $n_f$ ,<sup>25</sup> or more specifically,  $n_t / n_f \propto f_1 N_{ct} + f_2 [P^+_4]$ , where  $f_1$  and  $f_2$  are two temperature-dependent factors. With this in mind, Eq. (A1) is the same as Eq. (2).

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