

## Metastable-defect generation in hydrogenated amorphous silicon

Qing Zhang, Hideki Takashima, Jiang-Huai Zhou, Minoru Kumeda, and Tatsuo Shimizu

*Faculty of Technology, Kanazawa University, Kanazawa 920, Japan*

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We report on the effects of intense light soaking at room temperature (RT) and at 77 K on the defect density in hydrogenated amorphous silicon ( $a$ -Si:H). It is found that at short light-soaking times, light soaking at RT is more efficient in creating metastable defects than at 77 K. With increasing light-soaking time, however, 77-K light soaking causes the defect density to increase at a higher rate than does RT light soaking. There are signs that the saturated value of the defect density for 77-K light soaking is larger than that for RT light soaking. Qualitatively, a correlation exists between the increase in the defect density and the decrease in the photoconductivity; however, an inverse proportionality is not observed between the photoconductivity and defect density. For a given defect density, the photoconductivity is smaller for 77-K light soaking than for RT light soaking. The defects generated by 77-K light soaking are found to be stable at 77 K. However, significant annealing of defects occurs after raising the sample temperature to RT. Light-induced annealing of defects is also observed. We explain our results by adopting the views that there is a broad distribution of defect-annealing activation energies and that the defects with small annealing activation energies are more effective recombination centers than those with large annealing activation energies. We show that many other related experimental results can also be accounted for by the above views.

### I. INTRODUCTION

Although many models have been proposed to explain the light-induced metastable changes in hydrogenated amorphous silicon ( $a$ -Si:H), reported by Staebler and Wronski,<sup>1</sup> the physical processes involved in the metastable changes are still ill understood. Since almost all models are based on the results obtained at room temperature (RT) and, at the same time, consider the temperature at which light soaking is performed an important factor, it is expected that important insights into the light-induced effect can be gained by comparing the effects of light soaking at various temperatures. Santos and Jackson<sup>2</sup> have carried out light soaking of  $a$ -Si:H between 250 and 400 K and found that the lower the light-soaking temperature the higher the saturated value of the defect density. Han and Fritzsche<sup>3</sup> reported that although light soaking at 160 K decreases the mobility-lifetime product ( $\mu\tau$ ) of photocarriers in  $a$ -Si:H as efficiently as light soaking at RT, light soaking at 160 K causes the sub-band-gap absorption to increase much less than light soaking at RT. They, therefore, proposed that there are two kinds of light-induced metastable defects in  $a$ -Si:H. One kind mainly decreases the  $\mu\tau$  and the other increases the sub-band-gap absorption. Kumeda, Ohta, and Shimizu<sup>4</sup> studied the effect of light soaking on the photoluminescence (PL) in  $a$ -Si:H, and found that light soaking at a low temperature (LT) results in a larger fatigue in the PL than does light soaking at RT. However, the increase in the spin density is smaller for LT light soaking than for RT light soaking. They tentatively explained their results by suggesting that besides ESR centers, spinless nonradiative recombination centers are also created by the LT light soaking. Recent investigation by Yoshida and Taylor<sup>5</sup> supports the above suggestion. Moreover, Kumeda

*et al.*<sup>6</sup> observed that recovery in the defect density, after light soaking at LT and raising the temperature to RT, depends on the quality of the sample. They argued that a distribution of defect annealing activation energy is needed to explain their results. In a previous paper,<sup>7</sup> we reported that, compared with RT light soaking, light soaking at 77 K makes the photoconductivity degrade more. We suggested that the distribution of defect annealing activation energies is affected by light-soaking temperature and the difference in the recombination rates results from a difference in the distributions of defect annealing energies. Very recently, Stradins and Fritzsche<sup>8</sup> have studied the effects of light soaking at 4.2 and 80 K, and suggested that the defects having smaller annealing energies are more effective in decreasing the photocarrier lifetime than defects with larger annealing energies.

In this paper, we report experimental results on the metastable changes in the defect density and the photoconductivity in  $a$ -Si:H induced by light soaking at 77 K and at RT. We show that our result supports the suggestions that there is a broad distribution of defect annealing energies, and that the defects which have low annealing activation energies are the most effective recombination centers for photocarriers. We also give some discussions on the results obtained by other groups.

### II. EXPERIMENT

High-quality undoped  $a$ -Si:H films used in our study were deposited on silica substrate at 300 °C by decomposing SiH<sub>4</sub> in a hot-wall glow-discharge system which was used to reduce the impurity content in the films.<sup>9</sup> The film thicknesses were about 3  $\mu$ m. Strong light soaking of the samples was carried out using a Xe lamp with an IR-cut filter. The light intensity was 3.8 W/cm<sup>2</sup>, which cor-

responds roughly to a carrier generation rate of  $1.5 \times 10^{22} \text{ cm}^{-3} \text{ s}^{-1}$ . The same light source was also used for the photoconductivity measurements but the light intensity was reduced to  $6 \text{ mW/cm}^2$ . A powerful electrical fan was used to restrain temperature rising during light soaking at RT.

For light soaking at 77 K, a constant and exact temperature can be achieved by immersing the sample in liquid  $\text{N}_2$ . The reversibility of light induced metastable changes in the defect density and photoconductivity was verified by annealing the samples at  $200^\circ\text{C}$  in the dark for 5 h in flowing  $\text{N}_2$  gas and then cooling to RT at a rate less than  $2^\circ\text{C/min}$ . When the photoconductivity was measured at RT, the sample was kept in vacuum. The ESR measurements were performed in the X band. The bulk spin density (defect density) was obtained by subtracting the surface spin density, which is not appreciably affected by light soaking,<sup>10</sup> from the measured total spin density.

### III. RESULTS

For convenience of description, we first introduce the symbol,  $N_S(T1|T2)$ , to designate the defect density, where  $T1$  denotes the temperature at which the sample was light soaked and  $T2$  denotes the temperature at which the defect density was measured (by ESR). Thus,  $N_S(77 \text{ K}|77 \text{ K})$  is the defect density after 77 K light soaking and measured at 77 K without raising the temperature, and  $N_S(77 \text{ K}|RT)$  is the defect density after 77 K light soaking and measured at RT. Figure 1(a) shows the

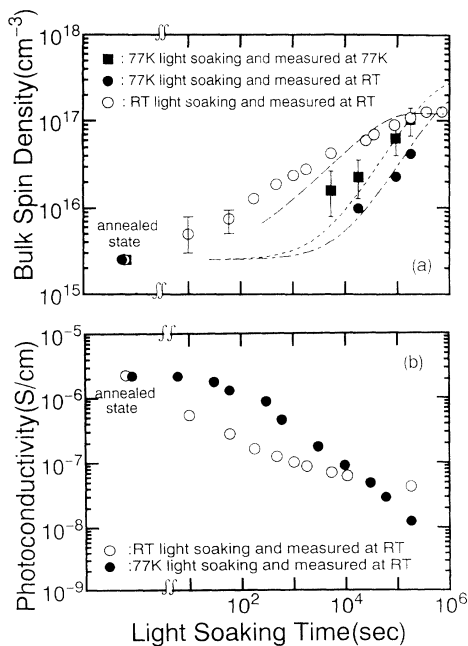


FIG. 1. Light-induced defect density (a) and photoconductivity (b) plotted against light-soaking time. Open circles stand for light soaking at RT and measured at RT, closed squares for light soaking at 77 K and measured at 77 K without raising the temperature, and closed circles for light soaking at 77 K and measured at RT. The dashed curves in (a) are fitting results using Eq. (3). The photoconductivities were measured at RT in vacuum with a light intensity of  $6 \text{ mW/cm}^2$ .

variations of the defect densities,  $N_S(77 \text{ K}|77 \text{ K})$ ,  $N_S(77 \text{ K}|RT)$ , and  $N_S(RT|RT)$ , with light-soaking time. It can be seen that at the early stage of light soaking, the defect density increases more slowly at 77 K than at RT. With increasing light-soaking time, however, the increase rates of  $N_S(77 \text{ K}|77 \text{ K})$  and  $N_S(77 \text{ K}|RT)$  become larger than that of  $N_S(RT|RT)$ . At about  $10^5$  sec, the value of  $N_S(77 \text{ K}|77 \text{ K})$  approaches the value of  $N_S(RT|RT)$ . It appears that  $N_S(77 \text{ K}|77 \text{ K})$  will eventually exceed  $N_S(RT|RT)$  at very long times. An important point to note in Fig. 1(a) is that  $N_S(77 \text{ K}|RT)$  is significantly smaller than  $N_S(77 \text{ K}|77 \text{ K})$ . Light soaking at 77 K for about 50 h makes  $N_S(77 \text{ K}|77 \text{ K})$  increase from the annealed state value,  $2.5 \times 10^{15} \text{ cm}^{-3}$  to  $1.0 \times 10^{17} \text{ cm}^{-3}$ . The defect density then remains unchanged at 77 K (within a relative accuracy of 20%), at least up to two days. However, when the sample was taken out of liquid  $\text{N}_2$ , the defect density drops sharply to  $4.2 \times 10^{16} \text{ cm}^{-3}$  within about 1 h at RT. It then again remains unchanged, at least for several days at RT (within a relative accuracy of 15%).

The photoconductivity degradations for 77 K and RT light soakings are shown in Fig. 1(b). The photoconductivities were measured at RT with a probing light intensity of  $6 \text{ mW/cm}^2$ . It is found that the degradation rate of the photoconductivity for 77 K light soaking is smaller than that for RT light soaking at short light-soaking times, but after a few hundred seconds the degradation rate for 77 K light soaking becomes larger. A crossover occurs at about  $10^4$  sec between the photoconductivity curves for 77 K and RT light soakings, consistent with the expectation that the defect density for 77 K light soaking will eventually exceed that for RT light soaking at sufficiently long times. It should be pointed out, however, that although the degradation behavior of the photoconductivity in Fig. 1(b) correlates quite well with the increase of the defect density in Fig. 1(a), the photoconductivity is not simply inversely proportional to the defect density. In Fig. 2, the photoconductivity is plotted against the defect density for both  $N_S(77 \text{ K}|RT)$  and

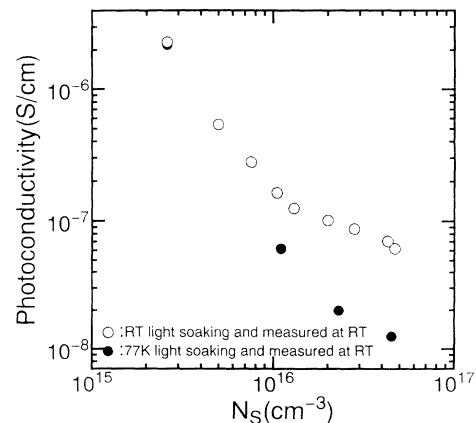


FIG. 2. Photoconductivity versus the defect density for 77-K light soaking (closed circles) and for RT light soaking (open circles). Both defect density and photoconductivities were measured at RT. The probing light intensity for photoconductivity measurement was  $6 \text{ mW/cm}^2$ .

$N_S(\text{RT}|\text{RT})$ . From this figure, one can see that for a given defect density, 77-K light soaking decreases the photoconductivity more than does RT light soaking. A similar result has been reported earlier.<sup>7</sup>

To check the dependence of the saturated values of the defect density and of the photoconductivity on light-soaking temperature, we carried out the following experiment. First, the sample was light soaked at RT until saturation was reached, resulting a defect density of  $N_S(\text{RT}|\text{RT}) = 1.3 \times 10^{17} \text{ cm}^{-3}$ . Then, the sample was light soaked further at 77 K for another 50 h. It was found that the defect density was increased to  $1.9 \times 10^{17} \text{ cm}^{-3}$  at 77 K. After raising the temperature to RT, the defect density was decreased to  $1.5 \times 10^{17} \text{ cm}^{-3}$ , which was still larger than the saturated value for RT light soaking. Correspondingly, the photoconductivity dropped to  $4.4 \times 10^{-8} \text{ S/cm}$  after the RT light soaking and further to  $1.8 \times 10^{-8} \text{ S/cm}$  after the additional light soaking at 77 K. As a last step, the sample was light soaked again at RT for 3 h. It was observed that the defect density and photoconductivity return to the initial saturated values for RT light soaking. The changes of the defect density and photoconductivity in the above experiment are illustrated in Fig. 3(a) and 3(b), respectively. The results of Fig. 3(a) and 3(b) suggest that the saturated values of both the defect density and photoconductivity depend on light-soaking temperature; the lower the light-soaking temperature, the higher the saturated defect density and the lower the saturated photoconductivity. It is also evident that RT light soaking can recover the metastable changes produced by 77 K light soaking, indicative of light-induced annealing.

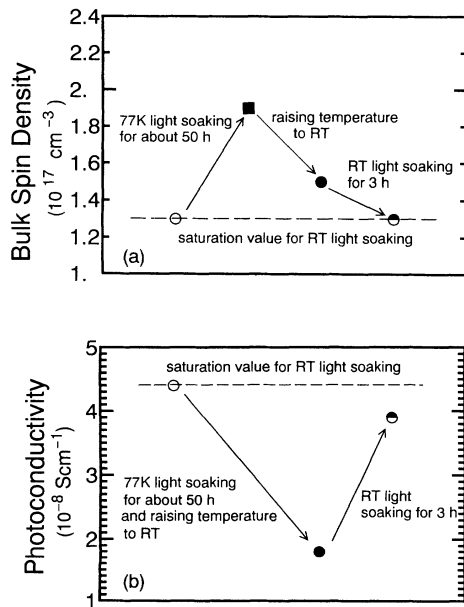


FIG. 3. Effects of the various treatments indicated in the figure on the defect density and photoconductivity. The defect density was initially saturated by light soaking at RT. (a) Defect density; (b) photoconductivity.

#### IV. DISCUSSION

Usually, thermal annealing of light-induced defects at RT or below is considered negligible, based mainly on the fact that no appreciable recovery occurs at such a temperature after cessation of light soaking at that temperature. However, it is evident from Fig. 1(a) and Fig. 3(a) that RT annealing can be significant if the light soaking is carried out below RT. To account for the annealing effect observed at RT, we adopt the view that there is a much broader distribution of defect annealing activation energies than that proposed by Hata and Wagner.<sup>11</sup> The lifetime ( $\tau$ ) of a metastable defect, which is the time required for the defect to be annealed out, increases exponentially with the annealing activation energy  $E_A$ ,

$$\tau(E_A) = \tau_0 \exp \left( \frac{E_A}{k_B T} \right), \quad (1)$$

where  $\tau_0$  is assumed to be constant,  $1 \times 10^{-10}$  sec, for simplicity.  $k_B$  is the Boltzmann constant, and  $T$  is the temperature. An important implication of Eq. (1) is that even at relatively low temperatures (say below RT), annealing of defects can occur, provided that the annealing activation energies are sufficiently small. The fact that no appreciable annealing effect is observed if the sample is kept at the light-soaking temperature or below can be understood as follows. Those metastable defects that have small annealing activation energies are already annealed out during light soaking, and those that remain after light soakings are the metastable defects that have large annealing activation energies. In order for significant annealing to occur, the sample temperature must be raised above the light soaking temperature. This is exactly what is observed experimentally.

To demonstrate the validity of the above arguments, we carried out a computer simulation by assuming a distribution of annealing activation energies of defects produced by 77-K light soaking as shown by the solid curve in Fig. 4. This distribution is essentially a Gaussian dis-

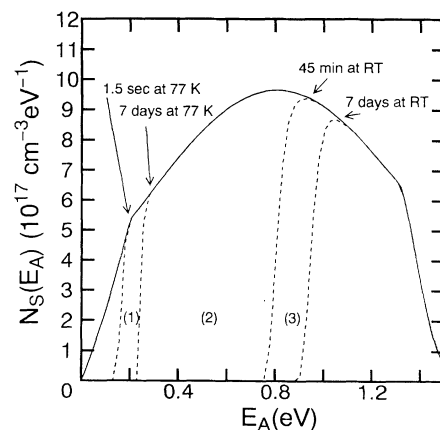


FIG. 4. Schematic illustration of the distribution of annealing activation energies for metastable defects produced by 77-K light soaking (solid curve), and the evolution of the defect density at 77 K and at RT, plotted by dashed curves. For details, see text.

tribution peaked at 0.8 eV, but with some modifications at low and high energies so that the defect density approaches zero at zero activation energy and at above 1.5 eV. At present, we have no evidence for the distribution used in the present simulation except that it is consistent with our observed recovery behaviors of the defects. We tried to explain the recovery behaviors using only two different annealing activation energies<sup>3</sup> and also using a simple Gaussian distribution, but either of them appears to be difficult to explain the recovery behaviors. We simulated the time dependence of the defect density by using

$$\frac{dN_S(E_A)}{dt} = -\frac{N_S(E_A)}{\tau(E_A)}, \quad (2)$$

i.e.,  $N_S(E_A)$  changes the time  $t$  as  $\exp(-t/\tau(E_A))$ . These results are shown by dashed curves in Fig. 4. We found that after 77-K light soaking, the defect density,  $N_S = \int_0^\infty N_S(E_A) dE_A$ , decreases by only 4.3% in one week, if the sample is kept at 77 K [region (1)]. However, after the sample temperature is raised to RT, half of the remaining defects are annealed out in 45 min [region (2)]. Keeping the sample at RT for another week leads to only a 27% further decrease in the defect density [region (3)]. Obviously, the  $N_S$  depends critically on the form of the distribution. However, our calculation demonstrates convincingly that a distribution of defect annealing activation energies is capable of accounting for the observed results.

Next, we discuss the generation kinetics of the light-induced defects. According to the weak-bond breaking model, the defect generation rate should be directly proportional to the product of the density of trapped electrons ( $n_t$ ) and the density of trapped holes ( $p_t$ ), as proposed by Stutzmann, Jackson, and Tsai<sup>12</sup> and to the density of the weak bonds which are convertible to defects. The rate of light-induced annealing should be directly proportional to the density of free electrons ( $n_f$ ) and to the light-induced defect density.<sup>13</sup> In principle, both thermal and light-induced annealing should be included in the rate equation. However, since our interest is only in the defects that remain after cessation of light soaking, we can neglect the thermal annealing term in the rate equation. The fact that the thermal annealing term is temperature dependent can be effectively taken into account of by treating the number of precursors, i.e., the weak bonds, to be temperature dependent. Consequently, the rate equation can be written as

$$\frac{dN_S}{dt} = A(N_T - N_S)n_t p_t - B(N_S - N_0)n_f, \quad (3)$$

where  $N_T$  and  $N_0$  are, respectively, the density of precursors and the density of defects in the annealed state, and  $A$  and  $B$  are constants.  $A$  is determined by the recombination characteristics and defect formation probability.  $B$  is associated with the free-carrier capture probability of the defects. We obtained the time dependences of  $n_t$ ,  $p_t$ , and  $n_f$  from the photoconductivity measured during light soaking, instead of using the approximations  $n_t$ ,  $p_t$ ,  $n_f \propto 1/N_S$ , which have been shown to be inconsistent with our experimental results (see, for example, Fig. 2).

According to Zhou and Elliott,<sup>14,15</sup> the photoconductivity of *a*-Si:H is dominated by free-electron transport down to about 60 K. We took the electron microscopic mobility as  $\mu = 13 \times (300/T) \text{ cm}^2/\text{sec V}$  (Ref. 16) and obtained  $n_f$  from the photoconductivity ( $\sigma_p$ ) during light soaking using the relation  $n_f = \sigma_p / e\mu$ . In order to determine  $n_t$ , first, the quasi-Fermi level for electrons ( $E_{fn}$ ) can be obtained from  $n_f = k_B T N_C \exp[(E_{fn} - E_C)/k_B T]$ , where  $E_C$  is the conduction band edge. Given that conduction band-tail states can be described by  $g(E) = N_C \exp[-(E_C - E)/k_B T_C]$ , where  $T_C$  was chosen as 310 K and  $N_C$  as  $1 \times 10^{21} \text{ cm}^{-3} \text{ eV}^{-1}$ ,  $n_t$  can be simply calculated by  $n_t = \int_{-\infty}^{E_{fn}} g(E) dE = (k_B T_C N_C) \times [\sigma_p / (e\mu k_B T N_C)]^{T/T_C}$ . Further assuming  $n_t \approx p_t$ , Eq. (3) can be solved directly by using the observed photoconductivity during light soaking at 77 K and RT shown in Fig. 5. It is interesting to note that the photoconductivity is hardly changed during the 77 K light soaking, indicating that recombination is dominated by the band-tail states.<sup>14,15</sup>

The fitted results are shown in Fig. 1(a) by the dashed curves. The values of the parameters used in the fitting are as follows.  $N_T$  was taken to be  $2 \times 10^{17}$ ,  $6 \times 10^{17}$ , and  $2.5 \times 10^{17} \text{ cm}^{-3}$  for RT light soaking, 77-K light soaking and measured at 77 K, and 77 K light soaking but measured at RT, respectively.  $A$  is  $1.5 \times 10^{-41}$  and  $3.5 \times 10^{-33} \text{ cm}^6/\text{sec}$  for the 77 K and the RT light soaking, respectively.  $B$  is  $4 \times 10^{-19} \text{ cm}^3/\text{sec}$  for both light-soaking temperatures. If  $A$  is thermally activated, the activation energy is about 0.2 eV.

Although the fitting is not very satisfactory, the shape of the time dependence of the defect density can indeed be produced by Eq. (3). In the fitting, the saturated values of the defect density were chosen in such a way that the value for RT light soaking is  $1.3 \times 10^{17} \text{ cm}^{-3}$ , in line with our observation. The value for 77-K light soaking was assumed to be  $3 \times 10^{17}$ , based on the observation that the saturated defect density for 77-K light soaking is larger than that for RT light soaking.

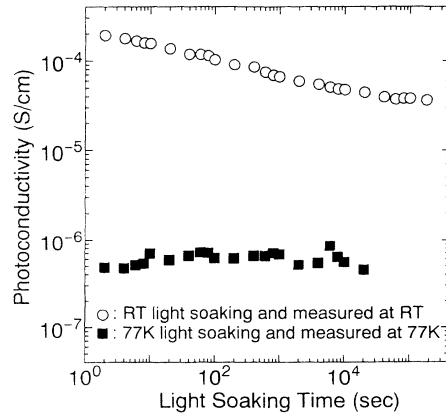


FIG. 5. Photoconductivity as a function of time measured during 77-K light soaking (closed squares) and measured during RT light soaking (open circles). The light intensities for both light-soaking temperatures were about  $3.8 \text{ W}/\text{cm}^2$ .

Based on Eq. (3), we can qualitatively explain the different behaviors for 77 K and RT light soakings shown in Fig. 1(a). The fact that  $A$  is much smaller for 77-K light soaking than for RT light soaking is the main reason for the smaller creation rate of defect for 77-K light soaking than for RT light soaking at the early stage of light soaking.  $n_i \propto (\sigma_p)^{T/T_c}$  as mentioned above. Therefore, according to the photoconductivity degradation shown in Fig. 5,  $n_i$  and  $p_i$  decrease quite remarkably by RT light soaking, while  $n_i$  and  $p_i$  by 77-K light soaking do not appreciably change. Hence the defect creation rate for RT light soaking becomes smaller as the light soaking proceeds. With increasing defect density, the contribution of light-induced annealing increases and finally competes with light-induced creation. Since the value of  $n_f$  is much smaller at 77 K than at RT, the effect of light-induced annealing at 77 K should be much smaller than that at RT. This also makes the saturated defect density for 77-K light soaking larger than that for RT light soaking.

It is worth comparing in detail the relative changes in the defect density and photoconductivity caused by additional light soaking at 77 K. From Figs. 3(a) and 3(b), the defect density measured at RT increases by a factor of  $1.5 \times 10^{17} / 1.3 \times 10^{17} \sim 1.2$ , while photoconductivity decreases by a factor of  $4.4 \times 10^{-6} / 1.8 \times 10^{-6} \sim 2.5$ . These results are consistent with the result shown in Fig. 2. As mentioned earlier, RT light soaking mainly creates defects which have large annealing activation energies, while 77-K light soaking results in the addition of defects which have smaller annealing energies. It is due to the low defect annealing energies that the additional defects are annealed out more easily than the defects produced by RT light soaking.<sup>17</sup> The strong effect of additional 77-K light soaking on the photoconductivity suggests that the defects with small annealing activation energies act as more effective recombination centers for photocarriers.

Finally, we would like to point out that the suggestions that there exists a broad distribution of defect annealing energies and that the defects which have small annealing energies are the most effective recombination centers explain not only our results but related results obtained by other researchers. A similar suggestion has been made by Shepard *et al.*<sup>18</sup> and Shimizu *et al.*<sup>19</sup> for RT light-soaking case. By investigating the light soaking stage and the annealing stage for 4.2 K and RT light soaking, Stradins and Fritzsche<sup>8</sup> also proposed a similar suggestion. The results of Santos and Jackson,<sup>2</sup> which are qualitatively similar to the present results in the behavior of the saturated value of light induced defects as a function of light-soaking temperature, although their temperature range is different from ours, can be easily understood in the light of the broad distribution of annealing activation energies, as we have discussed. If one considers that  $\mu\tau$  corresponds to the photoconductivity and the sub-band-gap absorption to the defect density, the results of Han and Fritzsche<sup>3</sup> can be understood under the above suggestion without introducing two kinds of light-induced

defects created at low temperature and at RT. Because in our case, the defect density,  $N_S(77\text{ K}|RT)$  is much smaller than the density,  $N_S(RT|RT)$ , at about  $10^4$  sec, while the photoconductivity degradations due to the 77 K and the RT light soakings reach a similar level for this light-soaking time. The consistency indicates that their data can be explained by the above suggestions.

It should be noted that after LT light soaking without raising the temperature the sample which shows a larger increase in the defect density has a larger fatigue in the photoluminescence.<sup>4,6</sup> After the sample is exposed to RT, the sample which has a larger recovery in the defect density also has a larger recovery in the fatigue. These results indicate that the increase in the defect density is related to the fatigue in photoluminescence. We still can explain the phenomena using a broad distribution of annealing activation energies. The defects with small annealing activation energies probably are also effective nonradiative recombination centers at LT. As small annealing activation energy defects can only remain at LT after they are produced, it is natural that LT light soaking causes a larger fatigue in the photoluminescence than can RT light soaking.

## V. CONCLUSIONS

At the early stage of light soaking, the creation rate of metastable defects for light soaking at 77 K is smaller than the creation rate for light soaking at room temperature. As light soaking proceeds, the rate for 77-K light soaking becomes larger than that for RT light soaking. Although the photoconductivity decreases with the increase of the defect density, the photoconductivity is not inversely proportional to the defect density. For a given defect density, 77-K light soaking makes the photoconductivity degradation larger than RT light soaking does.

The density of defects produced at 77 K remains constant at 77 K at least for two days, but decreases to almost half after raising the temperature to RT for about 1 h, and then remains unchanged for at least several days at RT. The defect density can be further increased after saturation is reached for RT light soaking, by additional light soaking at 77 K. A light-induced annealing effect is observed.

The above results suggest that (1) a broad distribution of annealing activation energies for the metastable defects is necessary to explain our results; (2) compared with RT light soaking, 77-K light soaking makes the distribution extend to low energies; (3) the defects with smaller annealing activation energies are more effective recombination centers than those with larger annealing energies.

## ACKNOWLEDGMENTS

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