# Investigation of the spatial distribution of dangling bonds in light-soaked hydrogenated amorphous silicon

メタデータ	言語: eng
	出版者:
	公開日: 2017-10-03
	キーワード (Ja):
	キーワード (En):
	作成者:
	メールアドレス:
	所属:
URL	http://hdl.handle.net/2297/3507

# Investigation of the spatial distribution of dangling bonds in light-soaked hydrogenated amorphous silicon

Jiang-Huai Zhou\*

Department of Electrical and Computer Engineering, Faculty of Engineering, Kanazawa University, Kanazawa 920, Japan

Minoru Kumeda

Laboratory for Development of Engineering Materials, Faculty of Engineering, Kanazawa University, Kanazawa 920, Japan

Tatsuo Shimizu

Department of Electrical and Computer Engineering, Faculty of Engineering, Kanazawa University, Kanazawa 920, Japan (Received 17 January 1995; revised manuscript received 16 October 1995)

We report on a detailed investigation of the spatial distribution of dangling bonds (DB's) in light-soaked hydrogenated amorphous silicon (a-Si:H) films. The results for light soaking at different light intensities (3 W/cm<sup>2</sup> and 300 mW/cm<sup>2</sup>) show that the inverse power-law DB distribution,  $N_v(x) = C_v x^{-\alpha}$ , holds regardless of the light soaking intensity. Here,  $N_n(x)$  is the volume density of DB's at depth x measured from the surface, and  $C_v$  and  $\alpha \approx 0.6$ ) are constants. The nonuniform spatial distribution of DB's in light-soaked a-Si:H is thought to originate from a nonuniform distribution of photocarriers during light soaking rather than from an inhomogeneity of the material. The same annealing behavior of light-induced DB's was observed regardless of the thickness of the sample and regardless of whether the sample was light soaked from one side or from both sides. This result, together with the observation of identical spin characteristics, indicates that the light-induced DB's at various depths of a given *a*-Si:H sample are identical in nature. The surface DB density is found to be much less sensitive to light soaking than the bulk DB density and can be assumed unchanged if the lightsoaking intensity is not much higher than 300 mW/cm<sup>2</sup> and the light-soaking time is shorter than  $\sim 10$  h. We show that the conventional method of estimating the surface DB density is no longer appropriate for lightsoaked a-Si:H, due to the highly nonuniform distribution of DB's in the material. The nonuniform distribution of DB's can lead to significant disagreements between different techniques in quantifying the Staebler-Wronski effect and should therefore be taken into account in studies of the SW effect.

#### I. INTRODUCTION

In a recent publication,<sup>1</sup> we reported that the spatial distribution of dangling bonds (DB's) in light-soaked hydrogenated amorphous silicon (a-Si:H) is highly nonuniform and has an inverse power-law form,

$$N_v(x) = C_v x^{-\alpha},\tag{1}$$

where  $N_v(x)$  is the volume density of DB's at depth x, measured from the (illuminated) surface, and  $C_v$  and  $\alpha$  are constants.  $\alpha$  is found to be essentially independent of light soaking and is about 0.6. Another key conclusion reached in Ref. 1 is that the surface DB density is much less sensitive to light soaking than the bulk DB density.

Following the previous study,<sup>1</sup> we have carried out a detailed investigation of the distribution of DB's in lightsoaked a-Si:H. In the present paper, we report the experimental results, which include the results for different lightsoaking intensities, the data for light soaking from both the surface and substrate sides, and a comparison between the annealing behaviors of light-induced DB's in samples of various thicknesses. We discuss in detail the origin of the inverse power-law distribution of DB's and the implications of the DB distribution for the estimation of the surface DB density in light-soaked a-Si:H and for the photoconductivity measurements. The number of DB's in the samples were measured by electron-spin resonance (ESR). The experimental details can be found in Ref. 1 and will not be described in this paper. What we would like to add here is that the dangling-bond ESR signal of our samples has the usual asymmetric line shape, with a peak-peak width of ~7.5 G and a g value of ~2.0055, regardless of the light-soaking time and intensity.

## II. ON THE SPATIAL DISTRIBUTION OF DANGLING BONDS

The conclusions drawn in Ref. 1 were based on the results for light soaking at an intensity of 3 W/cm<sup>2</sup>. However, in studies of the Staebler-Wronski effect (SWE) (Ref. 2), the most commonly used light-soaking intensities are between 100 and 500 mW/cm<sup>2</sup>. Therefore, it is important to check whether the light-soaking intensity affects the distribution of DB's in light-soaked a-Si:H. To this end, we have carried out a similar experiment, using a light intensity of  $300 \text{ mW/cm}^2$ . Figure 1 shows, on a logarithmic scale, the areal density of DB's,  $N_a$ , as a function of sample thickness after light soaking at 300 mW/cm<sup>2</sup> for various light-soaking times. One can see that, after light soaking,  $N_a$  increases with sample thickness according to a power law, very similar to the result for light soaking at 3 W/cm<sup>2</sup> (Ref. 1). This indicates that the spatial distribution of DB's is also of an inverse power-law form for light soaking at 300 mW/cm<sup>2</sup>. By fitting the data for

7267

© 1996 The American Physical Society



FIG. 1. Sample-thickness dependence of the areal density of dangling bonds in *a*-Si:H light soaked at 300 mW/cm<sup>2</sup> for various light-soaking times: (A) annealed, (1) 2 h, (2) 24 h, (B) light soaked from the film side for 24 h and then from the substrate side for 2 h. The solid lines are fits to the data assuming an inverse power-law DB distribution in the bulk and a depth-independent DB density in the surface layer.

light soaking from the surface side only (data 1 and 2 of Fig. 1), we found the exponent  $\alpha$  of the inverse power-law DB distribution to be 0.6, which is identical to that for light soaking at 3 W/cm<sup>2</sup>. Thus, we conclude that the distribution of DB's in light-soaked *a*-Si:H has the inverse power-law form described by Eq. (1) and does not depend on the light-soaking intensity.

Another important result of Fig. 1 is that the areal DB density at 0.01  $\mu$ m is hardly changed even after 24-h light soaking at 300 mW/cm<sup>2</sup>, indicating that the surface DB density is little changed by light soaking at  $300 \text{ mW/cm}^2$ . This result should be compared with that for light soaking at 3  $W/cm^2$ , where the surface DB density is increased by about a factor of 4 for a comparable light-soaking time (20 h).<sup>1</sup> However, the conclusion holds for both light-soaking intensities that the surface DB density is much less sensitive to light soaking than the bulk DB density. As already stated in Ref. 1, the distribution of DB's in the near-surface region (between 0.01 and 0.05  $\mu$ m) cannot be determined unambiguously in this work. To obtain the surface DB density, we assumed in fitting the experimental data that the DB distribution in the near-surface region is of the same inverse power-law form as that deep in the bulk. One might then wonder how much the calculated surface DB density may depend on the distribution of DB's in the near-surface region. We show in the Appendix that, in fact, a rational choice of the DB distribution in the near-surface region has little effect on the calculated surface DB density. Specifically, the calculated surface DB density varies by only about 30%, when the DB density in the near-surface region is changed from the same inverse power-law distribution as that deep in the bulk to depth independent. Another factor that affects the calculation of the surface DB density is the thickness of the surface layer, which is taken to be 0.01  $\mu$ m in this work.<sup>3,4</sup> We would like to point out, however, that the choice of the thickness of the surface layer affects only the absolute magnitude and not the relative change of the surface DB density. It is, thus, clear



FIG. 2. Sample-thickness dependence of the areal density of dangling bonds in *a*-Si:H light soaked at 3 W/cm<sup>2</sup>. Triangles: light soaked for 1200 min. Circles: light soaked from the surface side for 1200 min and then from the substrate side for 300 min. The lines are fits to the data.

from the above discussion that the conclusion drawn here on the sensitivity of the surface DB density to light soaking is valid, regardless of the thickness of the surface layer and the distribution of DB's in the near-surface region.

The data shown by the circles in Fig. 1 (data B) were obtained after subjecting the samples to an additional light soaking from the substrate side for 2 h following the 24-h light soaking from the surface side. The immediate impression is that for samples thicker than  $\sim 1 \ \mu m$ , the areal DB density is somewhat increased, whereas for thinner samples, no change of the areal DB density can be detected within the experimental error. In fact, the effect of additional light soaking from the substrate side can be better discussed if we look at the case of light soaking at  $3 \text{ W/cm}^2$ , because a systematic study was carried out for this case. We subjected half of the samples to an additional 300-min (5-h) light soaking from the substrate side after 1200-min (20-h) light soaking from the surface side. We find, as shown in Fig. 2, that the areal DB density is increased at all thicknesses, and the increase is much larger than it would be for the same additional light soaking from the surface side.

The thickness dependence of the areal DB density for light soaking from both the surface and substrate sides can also be well represented by a power law, as shown by the dashed line in Fig. 2. However, the corresponding distribution of DB's is no longer of an inverse power-law form. The power-law behavior of data B can be explained, in a handwaving manner, as follows. Light soaking of a sample from both the surface and substrate sides produces a DB distribution that has an inverse power-law form from both the surface and interface of the sample. This, naturally, gives rise to an apparent power-law thickness dependence of the areal DB density. It is interesting to see that the increase in  $N_a$  caused by the additional light soaking from the substrate side appears to increase with increasing sample thickness. This result is consistent with an inverse power-law DB distribution for single-side light soaking. During the surface-side light soaking, defect generation on the second half of the film is more substantial in thin than in thick samples, thus further light soaking from the substrate side will have a larger effect on thick than on thin samples.

The impact of a nonuniform spatial distribution of defects is especially strong upon measurements where the motion of nonequilibrium carriers is along the thickness of the sample. Measurements employing diode or sandwich structures are typical examples. In fact, the effect of a nonuniform spatial distribution of defects on the measurements of a-Si:H p-i-n devices has already been observed and pointed out by other researchers.<sup>5</sup> Moreover, since the spectrum of the Xe lamp resembles closely the spectrum of the sun light, the defect distribution obtained in this work should have direct implications for a-Si:H solar cells.

The results of this work also have some bearings on the value of the so-called saturated density of metastable defects in a-Si:H.<sup>6,7</sup> Although it is still highly controversial as to whether the saturation is due to the exhaustion of the sites (precursors) that can be converted into metastable defects by light soaking or to a balance between defect generation and annealing, some researchers have come to the conclusion that a density of  $\sim 1 \times 10^{17}$  cm<sup>-3</sup> represents the upper limit for metastable-defect generation in a-Si:H.<sup>6,8–10</sup> This view has been strongly criticized in a recent paper of Stutzmann, Rossi, and Brandt<sup>11</sup> who find that, with pulsed light soaking, the density of metastable DB's in a-Si:H can be increased above  $1 \times 10^{18}$  cm<sup>-3</sup> and argue that the saturation is a result of a balance between metastable-defect generation and annealing.<sup>12</sup> Although the present study cannot discriminate between the two saturation mechanisms, our results demonstrate (see Fig. 2 of Ref. 1) that the density of light-induced DB's in *a*-Si:H can be as large as  $1 \times 10^{18}$  cm<sup>-3</sup>, in agreement with the finding of Stutzmann, Rossi, and Brandt.<sup>11</sup>

# III. ORIGIN OF THE INVERSE POWER-LAW SPATIAL DISTRIBUTION OF DANGLING BONDS IN LIGHT-SOAKED a-Si:H

Regardless of the model, the presence of photoexcited carriers is essential for the generation of light-induced DB's in a-Si:H. The fact that the generation efficiency of lightinduced DB's increases with light-soaking intensity suggests that the nonuniform spatial distribution of DB's observed in our light-soaked a-Si:H samples originated from a nonuniform distribution of photocarriers during light soaking, rather than from an inhomogeneity of the films.<sup>13</sup> Unfortunately, to determine the distribution of photocarriers is difficult. It requires the knowledge of not only the carrier generation, but the carrier recombination and diffusion as well, both recombination and diffusion depending strongly on the lightsoaking time. As a rough guide, we calculated the spatial distribution of photocarrier (pair) generation rate for the soaking light used in this work. In the calculation, we ignored the fine details in the spectrum of the soaking light and approximated the energy distribution of the number density of incident photons to the distribution shown by the solid line in the inset to Fig. 3. The optical absorption constants were taken from the literature.<sup>14</sup> We also ignored the photons below 1.65 eV,<sup>15</sup> and assumed that the quantum efficiency is energy independent. The calculated distribution of photocarrier generation rate is shown by the solid line in Fig. 3. One can see that the photocarrier generation rate is only weakly dependent on depth below 0.01  $\mu$ m and has roughly an in-



FIG. 3. Spatial distributions of photocarrier generation rate, calculated using the energy distributions of the number density of incident photons shown in the inset. Solid line: for the spectrum of the soaking light used in this work. Dashed line: for a uniform energy distribution of the number density of incident photons.

verse power-law depth dependence above 0.03  $\mu$ m, with the power being about 1.4. Obviously, the calculation is crude in nature. However, we find that the distribution of photocarrier generation rate is a weak function of the spectrum of soaking light, as long as the light is white. For example, the generation rate calculated for a uniform energy distribution of the number density of incident photons (dashed line in Fig. 3) is not fundamentally different. Although a quantitative comparison between the distribution of photocarrier generation rate and that of DB's is difficult, the calculation appears to support the claim that a nonuniform distribution of photocarriers is responsible for the nonuniform distribution of DB's in light-soaked a-Si:H samples. The implication of the above discussion is clear: The spatial distribution of DB's in lightsoaked *a*-Si:H depends on the spectrum of the soaking light. Indirect evidence for this has been obtained recently from photoconductivity measurements.<sup>11</sup>

Although a nonuniform spatial distribution of photocarriers seems to be responsible for the inverse power-law spatial distribution of DB's observed in our light-soaked *a*-Si:H samples, other possibilities, such as an inhomogeneity of the films,<sup>13</sup> cannot be completely ruled out. To clarify this point, it is necessary that a similar investigation be carried out with light soaking, using uniformly absorbed band-gap light. Unfortunately, such a light source is not readily available, owing mostly to the difficulty in obtaining a light intensity that is high enough to be useful.

#### IV. ESTIMATION OF THE SURFACE DANGLING-BOND DENSITY IN LIGHT-SOAKED *a*-Si:H

Considerable disagreements exist as to how the defect density in the surface layer is affected by light soaking. Most researchers have found little or small increase in the surface defect density (compared to the change of the bulk defect density) after light soaking.<sup>13,16,17</sup> Our study also shows that the surface layer is much less sensitive to light soaking than the bulk. However, recently Ganguly and Matsuda<sup>18</sup> have reported that both the surface and bulk DB densities in their *a*-Si:H samples are increased by about 10 times after



FIG. 4. Sample-thickness dependence of the areal density of neutral dangling bonds in *a*-Si:H, light soaked at 3 W/cm<sup>2</sup> for various light-soaking times: (A) annealed, (1) 2 min; (2) 20 min, (3) 180 min, (4) 1200 min, (B) light soaked from the surface side for 1200 min and then from the substrate side for 300 min. The lines are fits to the data and are discussed in detail in Ref. 1 and in this paper.

 $\sim$ 3000-h light soaking with AM1 illumination (100 mW/cm<sup>2</sup>). In the following, we show that the controversy over the surface defect density probably resulted from the method used to estimate the surface defect density of light-soaked *a*-Si:H, and highlight the impact of a highly nonuniform defect distribution on the estimation of the surface defect density.

To determine the surface defect density, one usually plots on a linear scale the areal defect density as a function of sample thickness for a convenient range of sample thicknesses and fits the data with a linear function. The surface defect density is then deduced from the zero-thickness intercept of the linear fit.<sup>16,18</sup> It is important to note, however, that the underlying assumption in this conventional method of estimating the surface defect density is that the distribution of defects is substantially uniform throughout the bulk of the samples. Thus, the conventional method is defective by nature when applied to light-soaked *a*-Si:H, where the DB's are highly nonuniformly distributed.

To demonstrate how misleading the conventional method can be when used to estimate the surface defect density of light-soaked a-Si:H, we plot in Fig. 4 the data of Fig. 1 of Ref. 1 and data B of Fig. 2 on a linear scale. As is clear from Fig. 4, if one collects data only from samples thicker than 1  $\mu$ m (which is usually the case, as a good signal-to-noise ratio is difficult to achieve with thinner samples) and uses only three or four sample thicknesses as in some previous studies,<sup>16,18</sup> the relationship between the areal DB density  $N_a$ and sample thickness d can easily be mistaken as being linear for light-soaked samples. The nearly linear thickness dependence of  $N_a$  at large sample thicknesses is expected, as the inverse power-law DB distribution [Eq. (1)] predicts a weak thickness dependence of  $N_a$  at large sample thicknesses. However, if one takes the entire thickness range into consideration, the thickness dependence of  $N_a$  for lightsoaked samples is actually a power law and is thus highly nonlinear. Taking data 4 (obtained after 1200-min light soaking at 3 W/cm<sup>2</sup>) in Fig. 4 as an example, a forced linear fit to the data for  $d>1 \ \mu m$  would easily lead one to claim that both the surface and bulk DB densities are increased by some 20 times, whereas the surface DB density is actually increased by about a factor of 4 and the bulk DB density by one to three orders of magnitude, depending on the depth (see Fig. 2 of Ref. 1). Examination of the data for other light-soaking times (except for the shortest) leads to qualitatively the same result. It is, therefore, clear that the zerothickness intercept of a linear thickness dependence forcibly taken for samples thicker than 1  $\mu$ m has little to do with the true surface DB density of light-soaked a-Si:H; the surface DB density so determined contains a large contribution from and even can be dominated by the DB's in the near-surface bulk region (<1  $\mu$ m), where the DB density largely exceeds the value set by the slope of the forced linear fit to the data for thick samples. We think that this may be the reason why Ganguly and Matsuda<sup>18</sup> reached the conclusion that the surface and bulk DB densities are increased by the same factor in their light-soaked a-Si:H samples, although the lightsoaking conditions (both intensity and time) in their study were quite different from that in the present work.

It should be pointed out, however, that the above discussion on the conventional method applies primarily to the cases where sufficient light soaking (i.e., high light intensity and/or long light-soaking time) has been carried out. For weak or short light soaking, the conventional method does not necessarily give rise to a significant overestimation of the surface defect density. Because in such a case, the number of defects created in the near-surface bulk region may be too small to outnumber the defects in the surface layer. As a result, the estimated surface defect density is still dominated by the defects in the surface layer, the density of which is hardly changed by the light soaking.

The most important point here is perhaps that, in principle, the conventional method is inappropriate for the estimation of the bulk and surface DB densities of light-soaked *a*-Si:H. This method overestimates the surface DB density of light-soaked *a*-Si:H. In addition, the bulk and surface DB densities estimated by the conventional method depend on the thickness range used for the estimation. For example, compared with using the thickness range of 1–10  $\mu$ m, an estimation using the thickness range of 1–5  $\mu$ m leads to a larger bulk and smaller surface DB density. It is worthwhile to mention that, for strong and/or long light soaking, the bulk and surface DB densities are always found to be increased by almost the same factor when the thickness range of 1–10  $\mu$ m is used.

# V. ANNEALING BEHAVIOR OF LIGHT-INDUCED DANGLING BONDS

The annealing characteristics of light-induced DB's in a-Si:H have been studied extensively. However, to our knowledge, no attempt has been made to compare the annealing of light-induced DB's in samples of various thicknesses. Such a comparison is desirable, because a thickness-dependent annealing behavior would imply that the properties of light-induced DB's depend on their positions in the film, despite the fact that the light-induced DB's have



FIG. 5. Average density of light-induced dangling bonds as a function of annealing time at 125 °C. (a) Results for samples lightsoaked at 3 W/cm<sup>2</sup> for 1200 min from the surface side only. The sample thicknesses are, from top to bottom, 0.13, 0.39, 1.15, 2.2, 4.6, and 8.7  $\mu$ m. (b) Results for samples light soaked at 3 W/cm<sup>2</sup> for 1200 min from the surface side and for another 300 min from the substrate side. The sample thicknesses are, from top to bottom, 0.05, 0.2, 0.63, 1.63, 3.2, and 6.5  $\mu$ m.

identical spin characteristics (i.e., ESR line shape, width, and *g* value).

Figure 5 shows the annealing behavior of light-induced DB's at 125 °C for all 12 samples studied in the work. The average density of light-induced DB's was obtained by subtracting the average density of DB's in the annealed state (200 °C, 2 h) from that measured for a given annealing time at 125 °C. In order to minimize the effect of the large experimental error in the measured density of DB's in the annealed state, we used the average DB density calculated from the annealed-state distribution of DB's (distribution A in Fig. 2 of Ref. 1) to obtain the average density of light-induced DB's for a given thickness. Figure 5(a) shows the results for the six samples that were light soaked for 1200 min at 3 W/cm<sup>2</sup> from the surface side only, and Fig. 5(b) shows the results for the other six samples, which were subjected to a further 300min light soaking from the substrate side. For comparison, the same log scale is used in Figs. 5(a) and 5(b). One can see from Fig. 5 that, within the uncertainties of the measurements, the same annealing behavior is observed regardless of the thickness of the sample and whether the sample was light soaked from one side or from both sides. This result, together with the spin characteristics, is a clear indication that the light-induced DB's at various depths of a given a-Si:H sample are identical in nature. In particular, there seems no difference between light-induced DB's generated in the surface layer and those in the bulk, because such a difference would have been seen in the thinnest samples where the light-induced DB's in the surface layer account for some 50% of the total number of light-induced DB's in the samples. The identical nature of light-induced defects at various depths also supports the view that the nonuniform spatial distribution of DB's observed in light-soaked a-Si:H is an extrinsic feature.

# VI. EFFECT OF THE DISTRIBUTION OF DANGLING BONDS ON THE PHOTOCONDUCTIVITY

Although carrier recombination in a-Si:H is far from being fully understood, it remains unchallenged that the DB is the dominant recombination center in the material. Most of the studies so far on carrier recombination in a-Si:H assume that the recombination centers are spatially uniformly distributed, regardless of whether the sample is in the annealed or light-soaked state. However, the fact that the density of DB's in light-soaked a-Si:H has an inverse power-law depth dependence implies that the carrier lifetime in light-soaked samples depends strongly on the position of the carriers and increases with increasing depth.

The photoconductivity is probably the most convenient tool for studying carrier recombination (and thus defect density) in *a*-Si:H. Indeed, in most cases, the SWE is studied by measuring the photoconductivity as a function of light soaking (light-soaking time, intensity, temperature, etc.). However, the photoconductivity itself is only a relative measure of the recombination, as it depends critically on external factors, such as the probe light intensity, probe photon energy, and sample thickness. One quantity that provides an absolute measure of the recombination is the photoresponse (assuming that the quantum efficiency is photon-energy indepenphotoresponse dent). The is defined as the photoconductivity<sup>19</sup>  $\sigma_n$  divided by the photon absorption rate<sup>20</sup> G. Figure 6(a) shows the photoresponse of the 8.7- $\mu$ m sample at room temperature as a function of light-soaking time for two probe photon energies:  $h\nu = 1.77$  eV ( $\lambda$  $=7000\pm55$  Å) and  $h\nu=2.36$  eV ( $\lambda=5260\pm40$  Å). Using the optical-absorption data in the literature,<sup>14</sup> the absorption depths for the two photon energies are estimated to be about 6 and 0.11  $\mu$ m, respectively. The sample had a coplanar electrode configuration, with the aluminum electrodes evaporated on the top of the film. The photoconductivity was measured with the probe light incident upon the surface. The intensity was 2 mW/cm<sup>2</sup> for both probe lights. Light soaking of the sample was done in the same manner as for the ESR samples<sup>1</sup> and the light-soaking intensity was 3  $W/cm^2$ . The clearest feature in Fig. 6(a) is perhaps that the photoresponse for  $h\nu = 1.77$  eV is always larger than that for  $h\nu = 2.36$  eV. However, we would like to draw the reader's attention to the fact that the difference between the two photoresponses is increased after light soaking. In Fig. 6(b), we plot the ratio of the photoresponse for  $h\nu = 1.77$  eV to that for  $h\nu = 2.36$  eV, as a function of light-soaking time. One can see that the ratio increases sharply after the initial 2-min light soaking, followed by a slow decrease with increasing light soaking.

The increase of the ratio of the two photoresponses after light soaking is consistent with the nonuniform spatial distribution of DB's in light-soaked *a*-Si:H samples described in this paper and in Ref. 1. After the initial 2-min light soaking, the density of DB's in the bulk region of  $x < 1 \ \mu m$  is in-



FIG. 6. (a) Photoresponses for the probe lights of  $h\nu$ =1.77 and 2.36 eV. (b) Ratio of the two photoresponses as a function of light-soaking time. The sample thickness was 8.7  $\mu$ m. The light-soaking intensity was 3 W/cm<sup>2</sup> and probe light intensities were 2 mW/cm<sup>2</sup>. (A) Data in the annealed state, (B) data obtained after light soaking for 1200 min from the surface side and for another 300 min from the substrate side.

creased much more than the density of DB's in the region of  $x > 1 \mu m$ . Given the absorption depths for the two photon energies, the photoresponse for  $h\nu = 1.77$  eV will be decreased much less than the photoresponse for  $h\nu = 2.36$  eV, thus resulting in a large increase in the ratio of the photoresponses after light soaking. With increasing light-soaking time, the DB density is dominated by light-induced DB's throughout the sample and the functional form of the DB distribution remains basically unchanged. As a result, the two photoresponses are expected to decrease at about the same rate, causing little change in the ratio of the photoresponses. The slow, yet steady, decrease of the photoresponse ratio with light soaking is somewhat unexpected, and the reason for this is not clear. It seems that a more accurate description of the recombination is necessary to account for this slow decrease.

Another important result of Fig. 6(a) is that, after light soaking for 1200 min from the surface side, an additional 300-min light soaking from the substrate side causes a further decrease in the photoresponse for  $h\nu=1.77$  eV, but little change in the photoresponse for  $h\nu=2.36$  eV. This is responsible for the deep drop in the ratio of the photoresponses (labeled *B*) in Fig. 6(b). According to the discussion in Sec. II, additional light soaking from the substrate side increases mainly the density of DB's in the second half of the sample. Since very few photons for  $h\nu=2.36$  eV can penetrate to this half of the sample, little change will be seen in the corresponding photoresponse. In contrast, for  $h\nu=1.77$  eV, the photocarriers are by and large uniformly distributed in the sample, and thus the photoresponse for this photon energy will be further decreased.

The difference between the two photoresponses in the annealed state can be understood by considering carrier recombination through the high-defect-density surface layer. The large difference between the absorption depths for the two photon energies implies that recombination through defects in the surface layer is more significant in the case of  $h\nu$  = 2.36 eV than in the case of  $h\nu$ =1.77 eV. This leads, of course, to a smaller photoresponse for  $h\nu$ =2.36 eV.

Similar results were obtained with thinner samples. However, the effect of the nonuniform distribution of DB's decreases with decreasing sample thickness, as one might have expected. In fact, for samples thinner than about 0.5  $\mu$ m, the ratio of the two photoresponses is independent of lightsoaking time within the experimental error. Given the absorption depth for  $h\nu$ =2.36 eV (which is about 0.11  $\mu$ m), this result suggests that carrier diffusion needs to be considered for an accurate account of the recombination and photoconductivity.

#### VII. SUMMARY AND CONCLUSIONS

We have carried out further investigation of the spatial distribution of dangling bonds (DB's) in light-soaked a-Si:H films. The results for light soaking at different light intensities (3 W/cm<sup>2</sup> and 300 mW/cm<sup>2</sup>) show that the inverse power-law DB distribution,  $N_{\nu}(x) = C_{\nu} x^{-\alpha}$ , holds regardless of the light-soaking intensity. Here,  $N_{v}(x)$  is the volume density of DB's at depth x measured from the surface, and  $C_n$ and  $\alpha ~(\approx 0.6)$  are constants. The nonuniform spatial distribution of DB's in light-soaked samples is thought to originate from a nonuniform distribution of photocarriers during light soaking rather than from an inhomogeneity of the films. The surface DB density is found to be much less sensitive to light soaking than the bulk DB density. In particular, the surface DB density can be assumed unchanged if the light-soaking intensity is not much higher than 300 mW/cm<sup>2</sup> and the lightsoaking time is shorter than  $\sim 10$  h. The same annealing behavior of light-induced DB's was observed regardless of the thickness of the sample and regardless of whether the sample was light-soaked from one side or from both sides. This result, together with the observation of identical spin characteristics, indicates that the light-induced DB's at various depths of a given *a*-Si:H sample are identical in nature.

An important implication of the nonuniform distribution of DB's is that the conventional method of estimating the surface DB density is no longer appropriate for light-soaked a-Si:H. We have shown that this method overestimates the surface DB density in light-soaked a-Si:H. Moreover, the surface DB density so estimated for light-soaked a-Si:H depends on the thickness range used for the estimation. The nonuniform distribution of DB's can lead to significant disagreements between different techniques in quantifying the Staebler-Wronski effect and should, therefore, be taken into account in studies of the SWE. In particular, since the spectrum of the Xe lamp resembles closely the spectrum of the sun light, the DB distribution obtained in this work should have direct implications for a-Si:H solar cells.

#### ACKNOWLEDGMENTS

The authors would like to thank Dr. A. Morimoto for discussions. This work was partly supported by the New Sunshine Project of the Ministry of International Trade and Industry of Japan. Let us divide the sample into three regions along the thickness: the surface layer of  $x \le x_1$  ( $x_1=0.01 \ \mu m$ ), the near-surface region between  $x_1$  and  $x_2$  ( $x_2=0.05 \ \mu m$ ), and the deep bulk region of  $x \ge x_2$ , which is covered by the experimental data. Assume that the density of DB's in the surface layer,  $N_s$ , is depth independent. For the near-surface region, we assume an inverse power-law distribution of DB's,

$$N_{vs} = C_{vs} x^{-\delta}, \tag{A1}$$

where  $N_{vs}$  is the density of DB's in this region, and  $C_{vs}$  and  $\delta$  are constants. An inverse power-law functional form is chosen for the distribution of DB's in the near-surface region, because the degree of dependence of the DB density on depth can be conveniently varied by varying the power exponent  $\delta$ . For instance, a depth-independent DB density is obtained by setting  $\delta$  to zero.

For a given sample of thickness  $d \ge x_2$ , the areal density of DB's can be directly calculated using  $N_s$  and Eqs. (1) and (A1):

$$N_{a} = N_{s}x_{1} + \frac{C_{vs}}{1 - \delta} \left( x_{2}^{1 - \delta} - x_{1}^{1 - \delta} \right) + \frac{C_{v}}{1 - \alpha} \left( d^{1 - \alpha} - x_{2}^{1 - \alpha} \right).$$
(A2)

Experimentally, we find

$$N_a = C_a d^\beta = \frac{C_v}{1 - \alpha} d^{1 - \alpha}.$$
 (A3)

From Eqs. (A2) and (A3), we have

$$N_{s}x_{1} = \frac{C_{v}}{1-\alpha} x_{2}^{1-\alpha} - \frac{C_{vs}}{1-\delta} (x_{2}^{1-\delta} - x_{1}^{1-\delta}).$$
(A4)

- \*Present address: Joint Research Center for Atom Technology, c/o National Institute for Advanced Interdisciplinary Research, 1-1-4 Higashi, Tsukuba-shi, Ibaraki 305, Japan. Electronic address: zhou@jrcat.or.jp
- <sup>1</sup>J.-H. Zhou, M. Kumeda, and T. Shimizu, Appl. Phys. Lett. **66**, 742 (1995).
- <sup>2</sup>D. L. Staebler and C. R. Wronski, Appl. Phys. Lett. **31**, 292 (1977); J. Appl. Phys. **51**, 3262 (1980).
- <sup>3</sup>T. Shimizu, X. Xu, H. Kidoh, A. Morimoto, and M. Kumeda, J. Appl. Phys. **64**, 5045 (1988).
- <sup>4</sup>H. Yan, A. Morimoto, M. Kumeda, T. Shimizu, and Y. Yonezawa, in *Amorphous Silicon Technology—1992*, edited by M. J. Thompson, Y. Hamakawa, P. G. LeComber, A. Madan, and E. A. Schiff, MRS Symposia Proceedings No. 258 (Materials Research Society, Pittsburgh, 1992), p. 247.
- <sup>5</sup>R. A. Street, J. Non-Cryst. Solids 164-166, 643 (1993).
- <sup>6</sup>H. R. Park, J. Z. Liu, and S. Wagner, Appl. Phys. Lett. **55**, 2658 (1989).
- <sup>7</sup>Z. Y. Wu, J. M. Siefert, and B. Equer, J. Non-Cryst. Solids 137&138, 227 (1991).
- <sup>8</sup>D. Redfield and R. H. Bube, Appl. Phys. Lett. **54**, 1037 (1989).
- <sup>9</sup>N. Hata, G. Ganguly, S. Wagner, and A. Matsuda, Appl. Phys. Lett. **61**, 1817 (1992).

The continuity of the DB density at  $x = x_2$  requires

$$C_{vs}x_2^{-\delta} = C_v x_2^{-\alpha}. \tag{A5}$$

From Eqs. (A4) and (A5), one obtains

$$N_{s} = \left[\frac{\alpha - \delta}{1 - \alpha} + \left(\frac{x_{1}}{x_{2}}\right)^{1 - \delta}\right] \left(\frac{C_{v} x_{2}^{1 - \alpha}}{1 - \delta}\right) / x_{1}.$$
 (A6)

Given the distribution of the photocarrier generation rate (Fig. 3), it seems unlikely that the DB density in the nearsurface region has a stronger depth dependence than the DB density deep in the bulk or  $\delta > \alpha$ . On the other hand, a stronger depth dependence of the DB density in the near-surface region will lead to even a smaller relative increase in the surface DB density, in greater favor of our claim that the surface DB density is much less sensitive to light soaking than the bulk DB density. Therefore, we will consider only the case of  $\delta \leq \alpha$ . To demonstrate how the distribution of DB's in the near-surface region affects the surface DB density, we choose data 3 in Fig. 1 of Ref. 1 (obtained after 180-min light soaking at 3 W/cm<sup>2</sup>) for our calculation. For this set of data,  $\alpha = 0.6$  and  $C_v = 3.74 \times 10^{14}$  cm<sup>-2.4</sup>. Taking  $x_1 = 0.01 \ \mu \text{m}$  and  $x_2 = 0.05 \ \mu \text{m}$ , we obtain, for  $\delta = 0.6, 0.4$ , 0.2, 0, the surface DB density  $N_s = 3.72 \times 10^{18}$ ,  $4.16 \times 10^{18}$ ,  $4.52 \times 10^{18}$ ,  $4.82 \times 10^{18}$  cm<sup>-3</sup>, respectively. Note that  $\delta = 0.6$ corresponds to the case of the same inverse power-law DB distribution throughout the bulk and  $\delta=0$  to the case of a constant DB density in the near-surface region. However, the surface DB densities in the two cases differ by only about 30%. It is thus clear that the calculated surface DB density is essentially independent on the distribution of DB's in the near-surface region between 0.01 and 0.05  $\mu$ m, so long as the DB density in the near-surface region does not, with increasing depth, increase significantly or decrease much faster than the DB density deep in the bulk.

- <sup>10</sup>J. Kocka, O. Stika, and O. Klima, Appl. Phys. Lett. **62**, 1082 (1993).
- <sup>11</sup>M. Stutzmann, M. C. Rossi, and M. S. Brandt, Phys. Rev. B 50, 11 592 (1994).
- <sup>12</sup>C. F. O. Graeff, R. Buhleier, and M. Stutzmann, Appl. Phys. Lett. 62, 3001 (1993).
- <sup>13</sup>M. Stutzmann, W. B. Jackson, and C. C. Tsai, Phys. Rev. B **32**, 23 (1985).
- <sup>14</sup>L. Ley, in *The Physics of Hydrogenated Amorphous Silicon (II)*, edited by J. D. Joannopoulos and G. Lucovsky, Topics in Applied Physics Vol. 56 (Springer-Verlag, New York, 1984), p. 141.
- <sup>15</sup>The cutoff photon energy of 1.65 eV is chosen because (a) a standard *a*-Si:H sample has an optical gap of about 1.7 eV and (b) photons below the optical gap are inefficient for dangling-bond generation [but, see, J. F. Tian, D. S. Jiang, B. R. Zheng, L. Huang, and G. L. Kong, Solid State Commun. **57**, 543 (1986)]. Also, the assumption of an energy-independent quantum efficiency is reasonable only for photon energies greater than the optical gap.
- <sup>16</sup>T. Shimizu, H. Kidoh, M. Matsumoto, A. Morimoto, and M. Kumeda, J. Non-Cryst. Solids **114**, 630 (1989).
- <sup>17</sup>Z. E. Smith, V. Chu, K. Shepard, S. Aljishi, D. Slobodin, J.

Kolodzey, S. Wagner, and T. L. Chu, Appl. Phys. Lett. **50**, 1521 (1987).

- $^{18}\mbox{G}.$  Ganguly and A. Matsuda, Phys. Rev. B 49, 10 986 (1994).
- <sup>19</sup>By photoconductivity, we mean the average photoconductivity  $\sigma_p$  calculated from the photocurrent  $i_p$  by using the equation

 $\sigma_p = i_p L/VS$ , where V is the applied voltage, L is the electrode spacing, and S is the cross section of the sample.

<sup>20</sup> The photon absorption rate is the average number of photons absorbed per unit time per unit volume. It is equal to the average photocarrier generation rate if the quantum efficiency is unity.