

Long-Term Metastability of Light-Induced Defects in Hydrogenated Amorphous Silicon — Further Investigations

by

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Abstract

The peak intensity of the electron spin resonance signal due to silicon-dangling bonds in a-Si:H has been measured at room temperature as a function of time during 6.5×10^3 days after the intense pulsed illumination was turned off. The decay curve is fitted by an exponential function with a decay constant of 393 days and reaches a steady-state value smaller than the value taken before illumination. Such a long-term metastability of light-induced dangling bonds is interpreted in term of reconstruction of the amorphous network occurring through hydrogen motion.

Keywords: Amorphous silicon; Defects; ESR; Intense pulsed illumination; Long-term metastability of light-induced defects

1 Introduction

In a previous paper [1], we reported experimental results of ESR (electron spin resonance) peak intensity (the peak to peak height of the derivative of the ESR absorption signal) of the silicon-dangling bonds created by an intense pulsed illumination, measured at room temperature as a function of time until 3400 days after an intense pulsed illumination was turned off. We extend this measurement until 6.5×10^3 days after an intense pulsed illumination. The result is reported

in this paper and is interpreted in terms of photo-structural change in the amorphous network.

2 Experimental details

The a-Si:H samples were prepared at 120 °C on substrates of fused silica, using a plasma-enhanced chemical vapour deposition reactor with a mixture of SiH₄ (10 volume %) and H₂ (90 volume %) gases. Two films were used for the purpose of this study, in which one of them is called 15y15 and the other called 15y15-1 and 15y15-2, i.e. two samples were used by cutting the film with two pieces for different pulsed illumination [1].

In our experiments, intense pulsed illumination was performed at 10 K, using a YAG optical parametric oscillator laser system, in which a pulse has a width of 10 ns, a repetition frequency of 11 Hz and a power density of 100 mJ/cm², operating at a photon energy of 2.48 eV (wavelength 500 nm) [2]. The a-Si:H films were illuminated for 30 min.

Electron spin resonance (ESR) measurements were carried out at room temperature using a JES-FA100 spectrometer (JEOL) operating at 9.4 GHz.

3 Results and Discussion

First, we summarize the experimental results reported in a previous paper [1] and the interpretation on them, as follows:

(1) After an intense pulsed illumination, the ESR peak intensity (the peak to peak height of the derivative of the ESR absorption signal measured at room temperature) of the silicon-dangling bond increased twice as large as before the intense pulsed illumination. This means that silicon-dangling bonds are created by an intense pulsed illumination on the surface of the sample with the penetration depth of intense pulsed illumination [3].

(2) After an intense pulsed illumination, the decay of the ESR peak intensity, $I(t)$, is measured during 3400 days and the decay is found to follow the following equation,

$$I = I_A + I_1 \exp(-t/t_1), \quad (1)$$

$$I_A = I(\infty), \quad (2)$$

where $I_A = 23$, $I_1 = 104$, and $t_1 = 393$ days. Before illumination (in the dark), we obtain $I = 59$. For $t = \infty$, we obtain $I = 23$. This means that after illumination, the peak intensity tends to saturate at 23, that is, the pulsed illumination results in a partial annealing of dangling bonds with 61% of the dangling bond density in the dark, keeping the sample at room temperature after illumination.

(3) It is known that two types of silicon-dangling bonds, that is, the normal dangling bond and the hydrogen-related dangling bond, that is, a hydrogen atom is associated with a silicon atom having a dangling bond [4, 5]. According to an analysis of the ESR signal shape [6], we conclude that the dangling bond created by an intense pulsed illumination is the normal dangling bond.

As shown above, the ESR peak intensity at $t = \infty$, I_A , becomes lower than that before intense pulsed illumination. This result can be explained as follows: The intense pulsed illumination creates free hydrogen atoms, so that they terminate some silicon-dangling bonds. As a result, after

the intense pulsed illumination, the number of the silicon-dangling bonds decreases from that of silicon-dangling bonds before intense pulsed illumination (in the dark). We conclude that intense pulsed illumination gives rise to photo-structural change in the amorphous network of a-Si:H.

In this study, we measure the ESR peak intensity in the same sample of a-Si:H as that used in our previous paper until $t = 6.5 \times 10^3$ days.

Figure 1 shows the decay of ESR peak intensity as a function of time, t , after the intense pulsed illumination is turned off. The first two experimental points obtained for samples 15y15-1 and 15y15-2 in Figure 1 at $t = 8$ days and 107 days are plotted, with the illumination intensity and the illuminated area being corrected for comparison with other experimental points for sample 15y15, because samples 15y15-1 and 15y15-2 were prepared under the same condition as that for sample 15y15. As shown in Section 1, experimental points are fitted with a curve a (eq. (1)). Two points at $t = 6.3 \times 10^3$ days and 6.5×10^3 days are new ones measured in this study. These two points are also fitted to the curve a within experimental errors. This shows a long-term metastability of light-induced defects (silicon-dangling bonds).

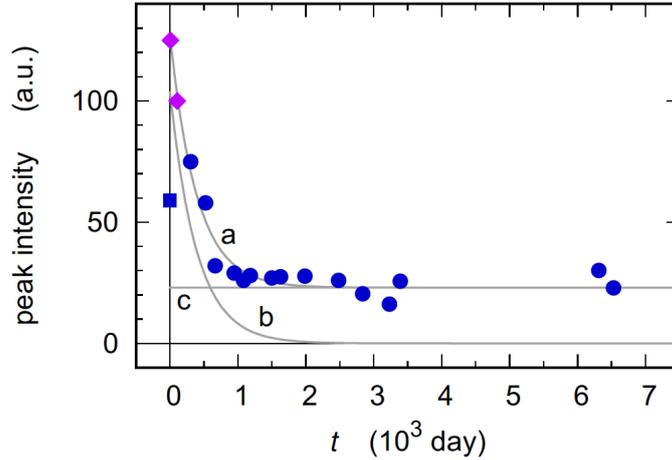


Fig. 1 The decay of ESR peak intensity as a function of time, t , after intense pulsed illumination is turned off. The two experimental points (diamond symbols) are those taken at $t = 8$ days and 107 days for a-Si:H samples 15y15-1 and 15y15-2, respectively (see the text). Other points (circular symbols) correspond to those taken for a-Si:H sample 15y15. The square symbol at $t = 0$ corresponds to the peak intensity taken before intense pulsed illumination (in the dark). The solid curve (a) is the one fitted to the experimental points and given by eq. (1) with $I_A = 23$, $I_1 = 104$, and $t_1 = 393$ days (see the text). The solid curve (b) is the one corresponding to the second term of eq. (1). The Solid line (c) is the constant one, I_A , corresponding to the first term of eq. (1).

In Figure 2, an observed ESR signal (violet colour) at $t = 6.3 \times 10^3$ days is shown along with that (black colour) taken at $t = 306$ days. The red curve shows the calculated one due to normal dangling bonds with the following values of ESR parameters fitted to the observed one: $g_{//} = 2.0058$, $g_{\perp} = 2.0061$, $\sigma_{//} = 1.6$ G, and $\sigma_{\perp} = 5.5$ G, in which $g_{//}$ and g_{\perp} are the g -values parallel and perpendicular to the principal axis with axial symmetry, respectively and $\sigma_{//}$ and σ_{\perp} are the standard deviations of the Gaussian spin packet for directions parallel and perpendicular to the principal axis, respectively. A spectrum observed at $t = 6.3 \times 10^3$ days has noises, but it is almost similar to that observed at 306 days within experimental errors.

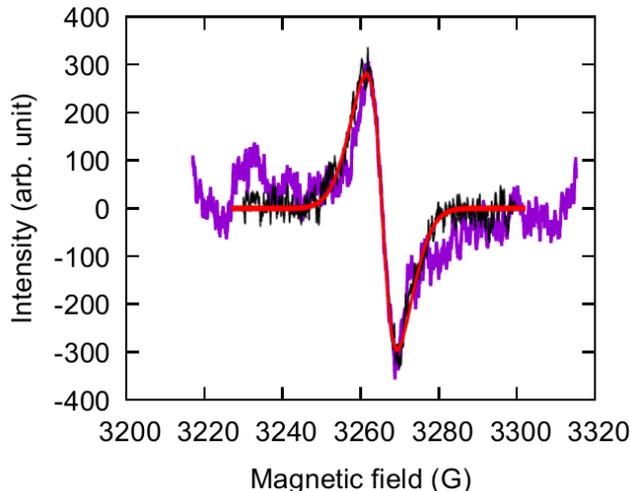


Fig. 2 The ESR spectra (violet colour and black colour) for a-Si:H sample 15y15 taken at $t = 6.3 \times 10^3$ days and at $t = 306$ days after intense pulsed illumination is turned off, respectively. The solid curve (red colour) is the calculated one due to normal dangling bonds with the following values of ESR parameters fitted to the observed one: $g_{//} = 2.0058$, $g_{\perp} = 2.0061$, $\sigma_{//} = 1.6$ G, and $\sigma_{\perp} = 5.5$ G. See the text for the notations of $g_{//}$, g_{\perp} , $\sigma_{//}$, and σ_{\perp} .

4 Conclusion

In a previous paper, we conclude that reconstruction of amorphous network occurring through hydrogen motion may be responsible for such a long-term metastability of light-induced dangling bonds in a-Si:H. The results shown in this paper support this conclusion. In particular, the motion of free hydrogen atoms plays an important role in the photo-structural change in a-Si:H under intense pulsed illumination.

References

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