Defects in hydrogenated amorphous silicon created by intense pulsed illumination at low temperature and the decay of their density

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Abstract. We present density of dangling bonds of silicon created in a-Si:H films by illumination of intense pulsed light at low temperature. The density of the photo-created dangling bonds decreases at room temperature with increasing time from illumination with a decay time estimated to be approximately $4.2 \times 10^6$ s (48 days). The results are compared with the intensity and lifetime of defect PL in a-Si:H films measured at various time from the pulsed illumination.

1. Introduction

Light-induced creation of defects in a-Si:H, which is known as the Staebler-Wronski effect [1], is an interesting phenomenon. Dominant defects in a-Si:H are dangling bonds of silicon (Si DBs). Si DBs in a-Si:H are detected by EPR measurements as a signal observed at $g = 2.0055$. The light induced creation of the Si DBs is observed as increase of the intensity of the EPR signal after illumination of intense light. However the mechanism of the defect creation has not been fully understood. Si DBs in a-Si:H act as non-radiative recombination centres as has been shown by optically detected magnetic resonance measurements [2]. The light-induced creation of the Si DB causes decrease of photoluminescence (PL) of the principal peak at 1.3 – 1.4 eV in intensity. On the other hand, there are defects which act as radiative recombination centres in a-Si:H, as indicated by observation of defect PL of the energy less than 0.9 eV depending on the defect density. The illumination causes the increase of the defect PL in intensity [3], indicating that the radiative defects are also created by illumination together with the Si DBs.

We have investigated the photo-induced phenomena in a-Si:H by monitoring PL. Pulsed illumination at low temperature causes significant enhancement of defect PL [4] and is suitable for the detailed study of the radiative defects responsible for the defect PL. However, the properties of the photo-created defects may be affected by the condition of illumination such as intensity of the light and the temperature of the a-Si:H film during the illumination. Further study of the properties of the photo-created defects is desired for the case of pulsed illumination at low temperature. In this paper we present our recent results of EPR measurements for a-Si:H after the pulsed illumination together with those of defect PL.
2. Experiment
The a-Si:H films used in this study were prepared in a capacitively coupled glow discharge reactor from a mixture of SiH₄ and H₂. The illumination was done at 8 – 10 K by a pulsed light of 2.48 eV from a YAG-OPO laser system for 0.5 h with a repetition frequency of 11 Hz. The intensity of the pulsed light was approximately 2 × 10⁻² mJ/cm². The a-Si:H films were kept at room temperature after the illumination.

The EPR measurements were performed for the a-Si:H films prepared at the substrate temperature of Tₛ = 120 °C and those after illumination of the pulsed light. The frequency and power of the microwave were 9.45 GHz and 1.0 mW, respectively. The intensity of the signal from Si DBs at g = 2.0055 was plotted as a function of the time from the illumination. In order to plot the intensity of the signal from photo-created DBs, the contribution of native DBs, estimated from the measurements of the a-Si:H film as grown, was subtracted.

The defect PL of 0.83 eV was measured under excitation of continuous wave light of 1.95 eV (635nm) for the a-Si:H films prepared at Tₛ = 250 °C after illumination of the pulsed light of 2.48 eV. Non-exponential decay of the PL in a-Si:H is understood as a superposition of exponential decays with lifetime distribution [5]. The broad featureless lifetime distributions of the defect PL were observed by means of frequency resolved spectroscopy (FRS). Characteristic lifetimes, τ₁/₂, which correspond to the intermediate values of the lifetime distributions [6, 7], were also estimated from the results of the FRS measurements. The intensities, I, and τ₁/₂ of the defect PL were obtained at various temperatures in the range of 10 – 200 K.

3. Results and Discussion
First we present the results of the density of Si DB after the illumination obtained from EPR measurements. The a-Si:H film used for EPR measurements contains a number of native Si DBs since it was prepared at low Tₛ. The intensity of the EPR signal obtained for the a-Si:H film before illumination, which represents the number of the native Si DBs, Nₛ₀, was subtracted from those obtained for the a-Si:H films after illumination, Nₛ. Thus the number of photo-created DBs, Nₛ – Nₛ₀, is plotted as a function of the time from the illumination, t. Figure 1(a) shows decay of the number of the photo-created Si DBs in the a-Si:H film measured for several months after the illumination. The result indicates that Si DBs in the a-Si:H film created by the pulsed illumination at low temperature are partially annealed out at room temperature. We have also found from EPR measurements of the a-Si:H film for several years after the pulsed illumination that some photo-created Si DBs have a much longer decay time. The result in figure 1(a) is fitted by least squares method with an equation

\[ Nₛ - Nₛ₀ = a \exp(-t/b) + c, \tag{1} \]

where a and b are constants characterizing an exponential decay and c is a constant approximately describing other components of much longer decay time. We obtained a curve with b ≃ 4.2 × 10⁶ s (48 days) and c/a ≃ 0.23. The curve is also shown by a broken line in figure 1(a).

Figure 1(b) shows the number of the photo-created Si DBs including those measured for several days after illumination. A significant decrease of the number of Si DBs at room temperature with increasing time after the illumination is seen. Due to the difference of the condition of the illumination, e.g. the area of the illuminated surface, the number of the photo-created DBs for the results in figure 1(b) is actually smaller than that for the results in figure 1(a) and the error in figure 1(b) seems to be larger than that in figure 1(a). However, the most of the results in figure 1(b) are likely described by (1) with the same b and c/a as above, which is also shown by a broken line in figure 1(b).

Next we present the intensity and characteristic lifetime of the defect PL measured at various temperatures in the range of 12 – 200 K. The measurements were done for a high quality a-Si:H
Figure 1. (a) Decay of the number of the photo-created Si DBs in the a-Si:H film obtained for several months after the illumination, and (b) that obtained for shorter time after illumination. The curve of (1) with $b \approx 4.2 \times 10^6$ s (48 days) and $c/a \approx 0.23$ is also shown by a broken line.

Figure 2. (a) Intensity and (b) characteristic lifetime of the defect PL of 0.83 eV in a-Si:H after illumination of the pulsed light. The results were measured at (i) $9 \times 10^4$ s, (ii) $3.5 \times 10^5$ s and (iii) $1.12 \times 10^6$ s after illumination.

film prepared at $T_s = 250$ °C. The defect PL from the a-Si:H film as grown is too weak to obtain the FRS results, because of the low defect density. Therefore the FRS signal from the a-Si:H film after illumination is due to photo-created radiative defects. Figures 2(a) and (b) show the intensity and the characteristic lifetime of the defect PL of 0.83 eV, respectively, measured at different time after illumination and plotted as functions of the temperature, $T$. The results do not strongly depend on the time after illumination, although the time after illumination is still shorter than $b$ obtained from the above EPR results. The characteristic lifetime observed at low temperature is approximately $20 \mu$s and much smaller than that we have previously reported.
for various a-Si:H films [8]. The short lifetime may be due to high density of Si DBs which act non-radiative recombination centres.

4. Conclusions
We have observed that Si DBs in the a-Si:H film created by the pulsed illumination at low temperature are partially annealed out at room temperature. The decay time of the photo-created DBs is estimated to be $4.2 \times 10^6$ s (48 days). The intensity and lifetime of defect PL due to radiative defects have also been obtained at various time after illumination. However, the results of the defect PL do not strongly depend on the time after illumination.

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