Light-induced degradation in *n*-type Czochralski silicon by boron-doping and thermal donor compensation

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In this paper, the behaviors of boron-oxygen complexes related to the light-induced degradation (LID) have been studied in *n*-type boron-doped Czochralski silicon (Cz-Si) with different levels of thermal donors (TDs) compensation. The degradation is found to have two stages, a fast- and a slow-process, similar to that in *p*-type boron-doped Cz-Si. In order to compare with the properties of LID in *p*-type Si, we systematically investigate the dependence of the saturated defect concentration (N_t^*) and the defect generation rate constant (R_{gen}) in our TD-compensated *n*-type Cz-Si on the carrier concentrations, i.e., the net electron concentration (n_0) and the excess hole concentration (Δp). Moreover, we determine the activation energy for the slow-process to be 0.4 eV. Based on our results, we suggest that the LID effect in boron-doped Si, either in *p*-type case or in *n*-type case by compensation, should be attributed to the same defects. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4759245]

I. INTRODUCTION

The commercial solar cells based on the conventional boron-doped Czochralski silicon (Cz-Si) usually suffer the light-induced degradation (LID) in efficiency as much as 3% relatively due to the formation of boron-oxygen (B-O) complexes. The B-O complexes, having strong recombination activity for carriers, can form in the case of minority carriers injection, whether by the sunlight illumination or by the application of a forward bias.[1,2] Their formation combines a fast- and a slow process, with activation energies of 0.23 eV and 0.48 eV, respectively.[3,4] By means of advanced lifetime spectroscopy, the energy level of the B-O complexes forming during the slow process is located at E_c-0.41 eV.[5] The early investigations have shown that in the conventional boron-doped silicon, the saturated defect concentration (N_t^*) of B-O complexes is proportional to the boron concentration (N_B) and to the square of the oxygen concentration.[6,7] So, Schmidt *et al.* proposed that the B-O complex might be composed of a substitutional boron atom and an oxygen dimer.[8] Later, it has been recognized that in *p*-type boron-doped Cz-Si with phosphorus compensation, the N_t^* and the defect generation rate constant (R_{gen}) are proportional to the hole concentration (p_0) and p_0^2, respectively, irrelevant with N_B.[9,10] Therefore, Voronkov *et al.*[11] proposed that the LID effect in Cz-Si starts from a grown-in latent B_iO_{2i} complex, composed of an interstitial boron atom (B_i) and an oxygen dimer. Macdonald *et al.*[12] extended the B_iO_{2i} model into *n*-type compensated silicon and indicated that the higher electron concentration (n_0) in samples will lead to a smaller value of N_t^*; however it is inconsistent with the experimental data reported by Rougieux *et al.*[13] Recently, Voronkov *et al.*[14] claimed that the degradation observed in *n*-type Cz-Si is attributed to a latent B_sO_{2i} complex and corresponds

to the fast process in *p*-type case, rather than the slow process. Nevertheless, more experimental results of LID in *n*-type compensated Cz-Si are still needed in order to further understand the B-O complexes in Cz-Si.

In this paper, we have studied the behaviors of B-O complexes in boron-doped *n*-type Cz-Si with thermal donors (TDs) compensation. The advantage of TD compensation is able to obtain a variation of carrier concentrations n_0 in Cz-Si with a constant N_B. The results show that the LID phenomenon in TD compensated *n*-type Si also contains a fast- and a slow-process as like in *p*-type case and has similar activation energy of 0.4 eV for the slow-process. It implies that the configurations of B-O complexes in compensated *n*-type Si are the same as that in *p*-type Si.

II. EXPERIMENTAL DETAILS

The starting samples were cut from the boron-doped Cz-Si wafer with resistivity of about 2 Ω cm. The N_B of 6.2×10^{15} cm^{-3} was calculated by four-point probe resistivity measurement after annealing at 650 °C for 30 min. The interstitial oxygen concentration is 1.07×10^{18} cm^{-3}, determined by Fourier transform infrared spectrometry (FTIR) with a calibration coefficient of 3.14×10^{17} cm^{-3} for 1107 cm^{-1}. All the samples were then subjected to annealing at 450 °C for 20–84 h with a protective argon atmosphere to generate different contents of TDs, in order to convert to *n*-type. Resistivity measured for each sample was directly converted into the electron concentration according to the ASTM-F723. Here, the influence of TD compensation on the carrier mobility was neglected, according to our calculation based on the Klaassen's model,[15] for the sum of the boron, and generated TD concentrations is less than 10^{16} cm^{-3}.

These TD compensated *n*-type samples were etched to remove the surface damage and cleaned by the RCA process. The silicon-nitride (SiN$_x$) films were deposited on both the sample surfaces to minimize the surface recombination. A

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200 °C/10 min annealing at dark was used to annihilate the B-O complexes in the samples to reach the initial annealed state before the light soaking. Then, the B-O complexes were generated under the illumination of halogen light of 30 mW/cm^2, in the temperature range of 50–90 °C which is controlled by a heating plate. And these defects were also generated under the illumination of the pulse laser of 904 nm with pulse width of 0.2 μs and pulse interval of 62.5 ms, which is the setup of microwave photo-conductance decay (MWPCD).

The effective carrier lifetime (τ) of the sample was measured by the MWPCD technique and the normalized defect concentration N_t was derived from the measured lifetime using the following equation:

$$N_t \equiv \frac{1}{\tau} - \frac{1}{\tau_0} = \left(\frac{1}{\tau_{SRH}} + \frac{1}{\tau_{res}} \right) - \frac{1}{\tau_{res}}, \quad (1)$$

where τ_0 is the lifetime in the annealed state, and τ_{SRH} and τ_{res} are the lifetime limited by the B-O complex and the residual defects, respectively. Since the excess carrier concentrations injected by the laser of MWPCD are in the range of 10^{16} cm^{-3} in our samples, which is approximate at the high injection level, Eq. (1) can be expressed as

$$N_t = \frac{r_p r_n}{r_p + r_n} N_{BO}, \quad (2)$$

where N_{BO} is the concentration of B-O complexes, and r_p and r_n are its capture rate of hole and electron, respectively. As can be seen, the normalized defect concentration is proportional to the concentration of B-O complexes.

III. RESULTS AND DISCUSSION

Fig. 1 demonstrate the effective lifetime τ degradation triggered by the halogen light and the pulse laser as a function of the illumination time t in (a) and (b), respectively. A reference sample of gallium-doped Cz-Si coated with SiN$_x$ film shows that no lifetime degradation happens under both the illumination conditions, meaning that the degradation in our TD-compensated n-type Si is due to the defect generation in the bulk. Note that in both the figures, the generation processes are not fully completed, and the generated B-O complexes only reach to the extent of about 90% of the saturated concentrations within our experimental time. If let the samples have the full degradation, time of more than 48 h and 10^6 s is needed under the illumination of halogen light and pulse laser, respectively.

It can be seen that the degradation behaves similarly between the halogen light illumination (at low injection level) and the pulse laser illumination (at high injection level). Both the degradation in Figs. 1(a) and 1(b) consist of two processes, i.e., a fast- and a slow-process. In the case of halogen light illumination, it is difficult to accurately determine the initial fast-process, yet it has sufficient accuracy to measure the characteristics of the slow-process. So we can employ it to induce the effective lifetime degradation at different temperatures, in order to obtain the activation energy for the generation of B-O complex. And in the case of laser
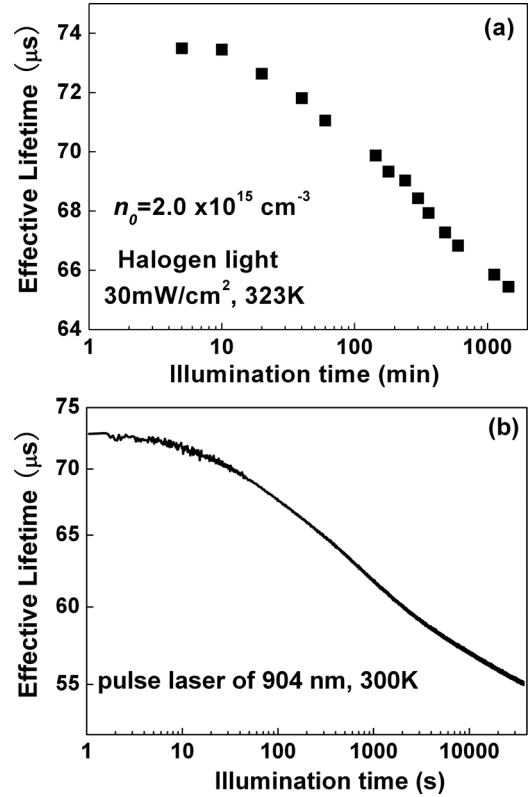

FIG. 1. The plots of the effective lifetime τ in a TD-compensated n-type Cz-Si sample as a function of the illumination time t, under the illumination of (a) halogen light and (b) pulse laser, respectively.

illumination, large amounts of data points of τ can be recorded during the entire degradation process, so the fast- and the slow-process can be exactly extracted from the effective lifetime degradation curve. Therefore, the setup of laser illumination at room temperature is especially suitable to explore the degradation of fast-process in our compensated samples.

Fig. 2 shows the generation of B-O complex during the pulse laser illumination, extracted from the data in Fig. 1(b). As mentioned, the generation is composed of a fast- and a slow-process, in which the fast-process dominates in the initial period of 1000 s and then the slow-process follows. It can be described by two exponential functions, in the form of
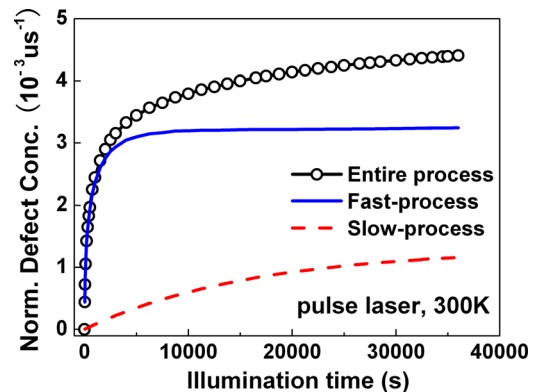

FIG. 2. The normalized defect concentration N_t derived from Fig. 1(b) using Eq. (1). The entire process (circle) is resolved into a fast-process (solid line) and a slow-process (dash line) by fitting with Eq. (3).

$$N_t(t) = N_t^* [1 - \exp(-(R_{gen}t)^n)], \qquad (3)$$

where exponent n is a constant. For the slow-process, $n = 1$ and the $R_{gen, slow}$ is about 6.2×10^{-5} s^{-1}. While for the fast-process, the N_t can be precisely fitted with $n = 0.5$ and the $R_{gen, fast}$ of 1.6×10^{-3} s^{-1}. Similarly, this asymptotic degradation phenomenon containing a fast- and a slow-process also occurs in the conventional p-type Si.[3,4] Therefore, we infer that the origins of the fast-process and the slow-process in p- and n-type Cz-Si might be the same, respectively.

By fitting the generation of B-O complexes using Eq. (3), we can obtain the correlation of saturated defect concentration N_t^* of the entire process as well as the fast- and slow-process, with the net carrier concentrations n_0 in TD-compensated n-type Si, as presented in Fig. 3. It shows that the fast-process accounts for the major part of the entire process, and the slow-process plays a less significant role in the generation process. However, the proportion between the fast- and slow-process will change with the injection level. Besides, it can be seen that the N_t^* of both the fast- and slow-process nonlinearly increases with an increase of n_0 in the samples. This correlation between N_t^* and n_0 in n-type TD compensated samples could be ascribed to several reasons. (1) Since the samples were first subjected to 650 °C annealing and the subsequently 450 °C annealing for different time, accompanied with the generation of TDs, the oxygen dimer may increase with the duration of annealing until it reaches to its equilibrium concentration.[16] Then, the N_t^* increases with the concentration of oxygen dimer ([O$_2$]) in the TD compensated samples.[8,17] (2) The effective defect concentration measured by the MWPCD depends on the electron concentration. As the injection level $\Delta n/n_0$ decreases with the increase of n_0 in the samples, the measured N_t gradually deviates from Eq. (2) and increases with the increasing n_0. Yet, it is interesting to note that only a slightly increase of N_t^* occurs in the samples with n_0 less than 3×10^{15} cm^{-3}, in which Eq. (2) is approximately complied with. (3) The electron is one crucial factor for the generation of B-O complex, which limits the reaction. However, it has been reported that the N_t^* is independent on the light intensity of 0.1 to 100 mW/cm^2 in p-type boron-doped Si, in-

dependent on the injected electron concentration Δn.[18] And the N_t^* also seems to have no correlation with n_0 in boron-phosphorus compensated n-type Si.[13] It shows that either in p- or in n-type Si, the N_t^* is insensitive to the electron concentration, so this explanation can be excluded. Nevertheless, the exact mechanism of N_t^* depending on n_0 in our TD-compensated Si has to be further investigated.

In order to investigate the dependence of the N_t^* on the hole concentration, which is the majority carrier in p-type Si and the minority carrier in our n-type Si, the samples were illuminated by varying the laser intensities I from 6×10^{12} to 1.5×10^{13} photons/pulse, as shown in Fig. 4. After the completely degradation, the N_t^* appears to be proportional to I, showing both the N_t^* of fast- and slow-process are proportional to the excess hole concentration (Δp), for Δp is proportional to I. Besides, we can find that the N_t^* generated under the laser illumination is larger than that under the halogen light illumination due to its larger injected Δp in Si, by comparing the final τ in Figs. 1(a) and 1(b). It is well known that the N_t^* in p-type Si is dependent on p_0,[6,7,9,10] so we suggest that this proportionality of N_t^* on the Δp in our TD compensated n-type Si should be identical to the p_0 dependence of N_t^* found in p-type Si. Therefore, it supports our consideration that there are the same defects contributing to the LID effect in both p- and n-type Cz-Si. Yet unfortunately, the explanation of the proportionality of saturated defect concentration on the hole concentration is still in controversy.[8,11,16]

Here, we propose a model based on a latent center (LC) of the B$_s$O$_{2i}$ complexes. The O$_{2i}$ is trapped by the B$_s$ atom around a distance of ~1 nm (R_c) in the Si zigzag chains. The LC concentration (N_{LC}) can be determined by the mass action law as follows:

$$\frac{N_{LC}}{N_B[O_{2i}]} \approx \frac{c}{\rho} \exp\left(\frac{E_b}{k_B T}\right), \qquad (4)$$

where c is the possible O$_{2i}$ sites around per B$_s$ atom (c is about 20 in the case of $R_c = 1$ nm), ρ the Si lattice site density, k_B the Boltzmann's constant, and E_b the binding energy of LC. The value of E_b should be larger than 0.7 eV. Since the O$_{2i}$ mobiles above 350 °C[19] and is gradually frozen below this temperature. By using Eq. (4), the ratio of $N_{LC}/[O_{2i}]$ at some critical
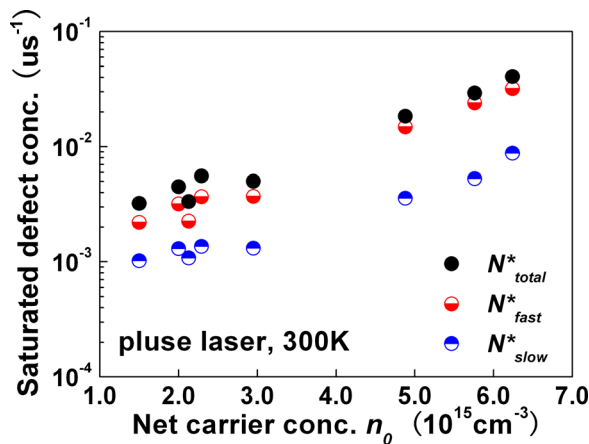

FIG. 3. The plots of the saturated defect concentration N_t^*, including for the fast- and slow-process, as a function of the net carrier concentrations n_0 in TD-compensated n-type Si.
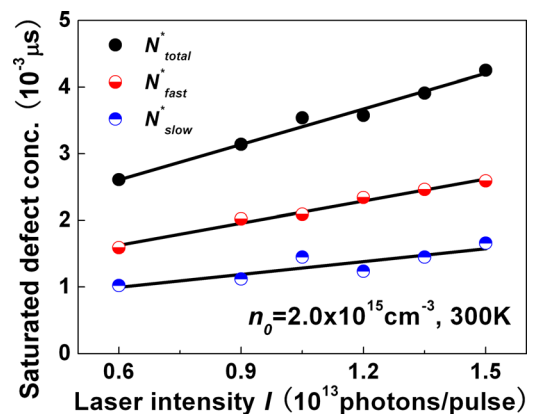

FIG. 4. The plots of the saturated defect concentration N_t^* (black circle), including for the fast- (half-down circle) and the slow-process (half-up circle), with the laser intensity I.

temperature $T = 350\,°C$ can easily be estimated. Typically, the N_B is in the order of $10^{16}\,cm^{-3}$ in the solar cells, and then the ratio is larger than 1. This indicates that all the O_{2i} should be trapped by B_s atoms forming the LC. Hence, the grown-in N_{LC} is only proportional to $[O_{2i}]$, irrelevant to N_B.

Furthermore, it has been revealed that the O_{2i} has two energy-favored structures by the first-principle calculation, i.e., the staggered- (O_{2i}^{st}) and the squared-dimer (O_{2i}^{sq}).[20,21] Note that all the O_{2i} should be in the neutral O_{2i}^{st} configuration existing in the LC after the ingot cooling.[22] However, according to Adey's proposal,[23] only those positively charged O_{2i}^{+} can diffuse towards B_s atoms by Bourgoin-Corbett mechanism, forming the recombination centers. In fact, Murin *et al.* systematically investigated the absorption bands of the positively charged dimers by FTIR at low temperature, but they cannot find their signals in the IR spectra.[22] The reason may be that only a very small portion of the O_{2i}^{st} are ionized and their concentrations are too low to be detected by FTIR.

Theoretical calculation shows that O_{2i}^{st} has two deep donor levels (E_1 and E_2) very close to the top of valence band,[24] so the portion of the different charge states can be approximately determined as[25]

$$\frac{[O_{2i}^{st+}]}{[O_{2i}^{st0}]} \approx \frac{p}{p_1}, \tag{5a}$$

$$\frac{[O_{2i}^{st++}]}{[O_{2i}^{st0}]} \approx \frac{p^2}{p_1 p_2}, \tag{5b}$$

where p_1 and p_2 are the characteristic concentrations at the level E_1 and E_2, respectively, which can be present as $p_{1,2} = N_v \exp[-(E_{1,2} - E_v)/k_B T]$ (N_v is the effective state density in the valence band and E_v is the energy at the top of valence band). Therefore, combining Eqs. (4) and (5a), the saturated defect concentration can be given by

$$N_t^* = \frac{p}{p_1}[O_{2i}]. \tag{6}$$

From Eq. (6), it can be seen that the saturated defect concentration N_t^* is proportional to p. If assuming the value of E_1-E_v in the range of 0.05–0.1 eV, and taking the typical value of p and $[O_{2i}]$ to be $1 \times 10^{16}\,cm^{-3}$ and $1 \times 10^{14}\,cm^{-3}$, the value of N_t^* will be in the range of 2×10^{11}–$1.5 \times 10^{12}\,cm^{-3}$.

Besides the correlation of N_t^* with the carrier concentration, we can also obtain the dependence of R_{gen} of B-O complexes on the electron concentration n_0 by fitting with Eq. (3). In Figs. 5(a) and 5(b), these dependences for the fast- and the slow-process are shown, respectively. It can be seen that the $R_{gen, fast}$ is linearly proportional to the n_0, while the $R_{gen, slow}$ is independent on it. This indicates that the capture of electron plays an important role in the initial fast-process of B-O complex formation, yet it should not be very important for the slow-process. Compared with the conventional *p*-type Si, it has been reported that the $R_{gen, slow}$ of B-O complexes increases linearly with the light intensity below 1 mW/cm² and then saturates at a higher illumination level.[8,15] It means that the capture of electron will not be the rate-

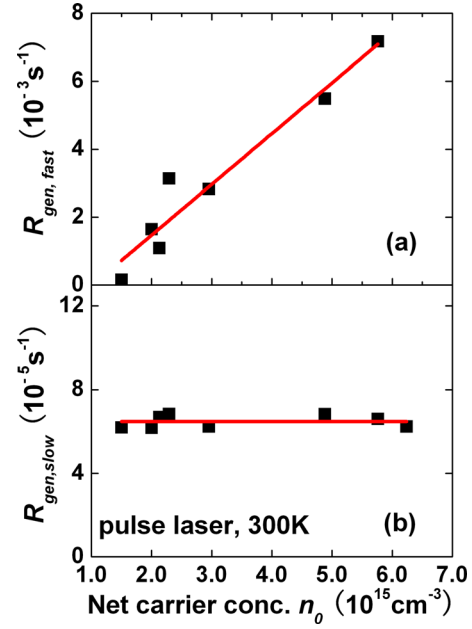FIG. 5. The generation rate constants of B-O complexes as a function of the electron concentrations n_0 in TD-compensated *n*-type Cz-Si. (a) The fast process ($R_{gen, fast}$); (b) The slow processes ($R_{gen, slow}$).

limited step for the generation of slow-forming B-O complexes when the electron concentration is above $10^{12}\,cm^{-3}$, which is the concentration induced by the light with intensity of 1 mW/cm². So if considering the same recombination center contributes to the slow-process in both *p*- and *n*-type Cz-Si, the $R_{gen, slow}$ should be independent on n_0 in *n*-type Si, for the electron is the majority carrier and its concentration is far above $10^{12}\,cm^{-3}$ in *n*-type Si. We can see that this conclusion well agrees with the result in Fig. 5(b).

Furthermore, the dependences of R_{gen} for the fast- and the slow-process on the laser intensity I are shown in Figs. 6(a) and 6(b), together with the fits of a power law $R_{gen} \sim I^a$. For the fast-process, $a = 1.6 \pm 0.3$, and for the slow-process, $a = 2.1 \pm 0.1$, indicating that the R_{gen} of both processes are approximately quadratically dependent on the laser intensity, i.e., the injected Δp in samples. As well known, the R_{gen} for the fast- and slow-process in *p*-type Si are also proportional to the squared hole concentration p_0^2.[3,9,10] It demonstrates that the R_{gen} in *p*- and *n*-type Si exhibit the same dependence on the hole concentration.

The p^2 dependence of R_{gen} has already been proposed in Palmer's and Voronkov's models. However, the explanations in these two models are totally different.[11,16] In our consideration, the recombination centers are generated by the O_{2i} jumping from the bounded sites of LC to the nearest sites of B_s atoms. The generation rate can be written as

$$\frac{dN_t}{dt} = R_{gen}[O_{2i}]. \tag{7}$$

Note that the N_B is not included in Eq. (7) since all the O_{2i} are trapped by B_s atoms in the LC. On the other hand, the O_{2i} diffusion follows the sequential reactions:[23,24,26]

(a) $O_{2i}^{st+} + h^+ \rightarrow O_{2i}^{st++}$,
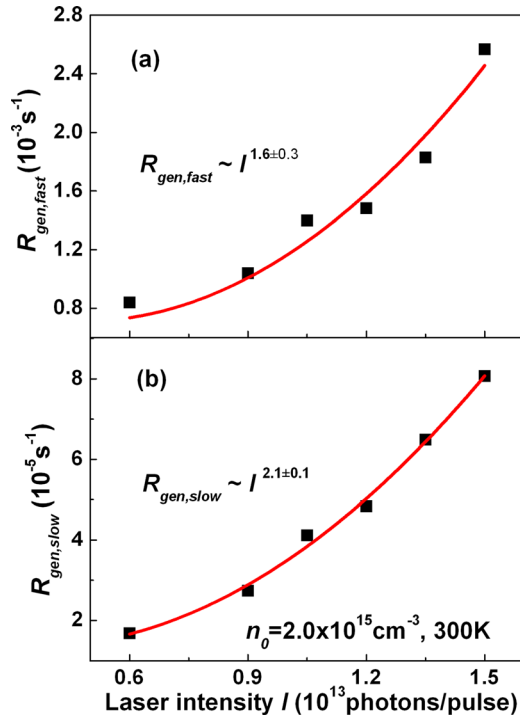(b) $O_{2i}^{st++} \rightarrow O_{2i}^{sq++} (reconfiguration)$,

FIG. 6. The plots of generation rate constants for (a) the fast-process ($R_{gen,\,fast}$) and (b) the slow-process ($R_{gen,\,slow}$) as a function of laser intensity I. The fitting lines indicate a quadratic dependence of R_{gen} on the I.

(c) $O_{2i}^{sq++} + e^- \rightarrow O_{2i}^{sq+}$,
(d) $O_{2i}^{sq+} \rightarrow O_{2i}^{st+} (reconfiguration)$,
(e) $O_{2i}^{st+} \rightarrow O_{2i}^{st+} (rotation)$.

Among these reactions, steps (a) and (b) are the rate-limiting steps. Since in step (a), the O_{2i}^{st+} has to capture a hole by overcoming the repulsive Coulomb potential and then in step (b), the O_{2i}^{st++} reconfigures to the O_{2i}^{sq++} with a barrier E_a of about 0.3 eV. The reconfiguration rate is $\nu = \nu_0 \exp(-E_a/kT)$, and ν_0 is the jump-attempt frequency. Hence, the generation rate can also be described as

$$\frac{dN_t}{dt} = [O_{2i}^{st++}]\nu. \qquad (8)$$

Combining Eqs. (5b), (7), and (8), the R_{gen} is equal to

$$R_{gen} = \frac{p^2}{p_1 p_2} \nu_0 \exp\left(-\frac{E_a}{k_B T}\right). \qquad (9)$$

Thus, the R_{gen} is proportional to the square of the hole concentration p^2 rather than the N_B^2.

Fig. 7 shows the Arrhenius plot of the defect generation rate constant $R_{gen,slow}$ for the slow-process under the halogen light illumination. The $R_{gen,slow}$ determined under the halogen light illumination is also independent on n_0, the same to the result obtained under the laser illumination shown in Fig. 5(b). As seen from the figure, the activation energy of $E_{gen,slow} = 0.40 \pm 0.03$ eV for the generation of B-O complex is characterized by the slope of the fitting line, and the corresponding pre-exponential factor is $80\,\mathrm{s}^{-1}$. The similar values of $E_{gen,slow}$ in the conventional p-type Si also have been reported to be about 0.4 eV.[4,8,19] This convinces us that the
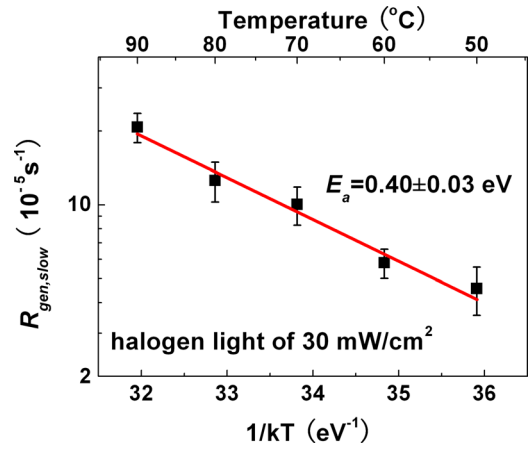

FIG. 7. The Arrhenius plots of the generation rate constants $R_{gen,\,slow}$ of B-O complexes in TD-compensated *n*-type Cz-Si. The slope of fitting line gives an activation energy of 0.40 eV.

slow-process in both *p*- and *n*-type Si should originate from the same recombination center.

IV. CONCLUSIONS

In summary, the behaviors of B-O complexes in *n*-type boron-doped Cz-Si with TD compensation have been investigated. It is found that the LID contains two stages, a fast- and a slow process, similar to that in *p*-type case. The saturated defect concentration (N_t*) increases nonlinearly with the net electron concentration (n_0) in our samples, and it strongly depends on the excess hole concentration (Δp) generated by the illumination. Moreover, the defect generation rate constant (R_{gen}) for the fast-process is proportional to n_0, while R_{gen} for the slow-process is independent on it. By varying the laser intensity, we find that the R_{gen} for both processes are quadratically dependent on the Δp. Besides, the activation energy for the slow degradation process is determined to be about 0.4 eV. Based on these findings, we suggest that the degradation in both *p*- and *n*-type Cz-Si is due to the same recombination centers generating under the light illumination.

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