

Studying light-induced degradation by lifetime decay analysis: Excellent fit to solution of simple second order rate equation

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Abstract— Twenty different boron-doped Czochralski silicon materials have been analyzed for light induced degradation. The carrier lifetime degradation was monitored by an automated quasi steady state photoconductance setup with an externally controlled bias lamp for in-situ illumination between measurements. Logarithmic plots of the time resolved lifetime decays clearly displayed the previously reported rapid and slow decays, but a satisfactory fit to a single exponential function could not be achieved. We found, however, that both decay curves, for all the investigated samples, can be fitted very well to the solution of a simple second order rate equation. This indicates that the defect generation process can be described by second order reaction kinetics. The new information is used to discuss the role of holes in the defect reaction and the rate determining steps of the rapid and slow defect reactions.

Index Terms—Light-induced degradation, reaction kinetics, B-O defects, minority carrier lifetime, rate equations, Czochralski silicon

I. INTRODUCTION

A. The B-O defect

Boron-doped Czochralski silicon (Cz-Si) is known to be subject to light induced degradation (LID), decreasing the minority charge carrier lifetime (hereafter: lifetime) and thus the solar cell efficiency. It has been shown that a fast initial decay occurs during the first few minutes while a second slower decay proceeds until lifetime saturation [1],[2]. The two characteristic decays have been reported to be depending on two independent mechanisms, although both mechanisms are related to the simultaneous presence of boron and oxygen

[1]. According to Shockley-Read-Hall theory [3] the defect concentration can be directly related to carrier lifetime (τ), provided that the generated defects dominate the recombination behavior.

LID is often described in terms of a normalized defect concentration, N_t^* , which is related to the measured lifetime by the following equation:

$$N_t^*(t) = \frac{1}{\tau(t)} - \frac{1}{\tau_0}, \quad (1)$$

where t is time, τ_0 is the initial carrier lifetime before LID and $\tau(t)$ is the carrier lifetime after time t . Upon prolonged illumination at a sufficiently high carrier injection level [4], N_t^* approaches a saturated value $N_{t,sat}^*$ that has been found to be proportional to the boron concentration and the oxygen concentration squared ($N_{t,sat}^* \propto [B][O]^2$) [5]. A model by Schmidt et al. [5], where fast diffusing oxygen dimers capture substitutional boron atoms to form B_s-O_{2i} recombination centers, was the first complete model that took this into account and for years this was the prevailing model. In 2009, however, Macdonald et al. [6] discovered that in p-type material co-doped with boron and phosphorus, where the hole concentration p_0 is smaller than $[B]$, $N_{t,sat}^*$ was found to be proportional to p_0 and not to $[B]$. This finding invalidated the model of Schmidt et al, and a new model was later proposed by Voronkov and Falster [7],[8]. In a recent work by Forster et al. [9], however, where silicon co-doped with boron and gallium was investigated, it was found that the defect concentration was proportional to $[B]$ instead of p_0 . Accordingly, how the saturated defect concentration scales with $[B]$ and/or p_0 is still up for debate.

In the model by Voronkov and Falster [7],[8], a latent recombination center is already incorporated in the crystal before carrier injection. According to this model, the centers responsible for the rapid and slow lifetime decays are B_sO_{2i} and B_iO_{2i} respectively. The reaction model is similar for the two decays, but with some differences: In p-type silicon the activation of B_sO_{2i} , (the defect center responsible for the rapid lifetime decay), is explained through a series of steps where the quadratic proportionality observed between p_0 and lifetime is accounted for by the capture of two holes by a negative latent center (LC_{fast}^-) resulting in the formation of a positive latent center

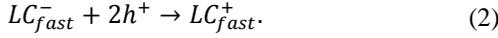
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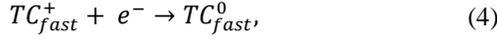
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The positive latent center is then reconstructed into a transient center (TC_{fast}) by overcoming an energy barrier



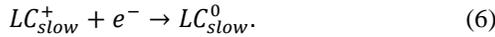
which due to recharging by injection of electrons forms a neutral transient center TC_{fast}^0



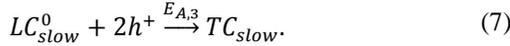
and finally the recombination active center RC_{fast}



The defect center responsible for the slow lifetime decay also starts from a latent complex, but this time the substitutional boron is replaced by interstitial boron, B_iO_2 . The latent center is now denoted LC_{slow} . In the reaction sequence for the slow decay the first step is a recharge of the latent center into a neutral state by the assistance of injected minority carriers



The second step is a temporary recharging to a double positive state by two holes, resulting in a transient center TC_{slow}



The final step is controlled by overcoming a free energy barrier between TC_{slow} and a slow recombination active center RC_{slow} and is thus governed by the attempt frequency and the Boltzmann energy



Voronkov et al. suggests that in most practical cases it is the final step that is rate determining for the process[7].

In the work by Schmidt et al [5], [10] the defect generation rate R_{gen} , for both the rapid and the slow decay, has been determined by fitting a single exponential function to the measured lifetime degradation data by the following equation:

$$N'_t(t) = N_0 [1 - \exp(-kt)], \quad (9)$$

where N'_t is the concentration of latent defects, N_0 is the initial latent defect concentration and k is the rate constant. A rate equation used to describe the defect generation process shows the effect of changing the concentration of the reactant on the rate of the reaction. A fit like this serves as a solution of a first order rate equation, given by

$$-\frac{dN'_t}{dt} = k(N'_t). \quad (10)$$

In other work [11], [12] it has, however, been acknowledged that a satisfactory fit between this function and the measured decays is not always obtained. In these works a double exponential function has been used to fit the lifetime decay data. Such a function can however not be directly related to

the kinetics of the defect generation reaction and thus gives less physical understanding of the process.

In the present work we show that both the rapid and the slow lifetime decay can be described far better by a second order rate equation than a first order rate equation. Information on the defect reaction kinetics extracted from the second order rate equation fit are used to discuss the rate determining steps of the rapid and slow defect reactions.

II. EXPERIMENTAL

The samples used in this study are boron doped Cz-Si wafers with a boron concentration ranging from 2×10^{15} - 11×10^{15} cm^{-3} . A boron doped FZ-Si sample was used as a reference. Prior to the measurements, the wafers were cleaned with RCA [13] and thereafter received a double side passivation by plasma-enhanced chemical vapor deposited (PECVD) hydrogenated amorphous silicon. The surface recombination velocity is estimated to be below 20 cm/s. In order to ensure that no defect centers are generated before the start of the measurements, the samples were deactivated by an annealing at 200 °C for 15 min on a hotplate in the dark [10] with dummy wafers above and below to avoid contamination.

The normalized B-O defect densities N_t^* were determined by time resolved lifetime measurements. The lifetime degradation was monitored by an automated quasi-steady state photoconductance (QSSPC) setup with an externally controlled bias lamp for in-situ illumination between measurements. The bias light source was a halogen lamp supplying the sample with an intensity of approximately 50 mW/cm^2 . The temperature of the sample was monitored by a resistance temperature detector coupled to the sample stage to ensure that the temperature change of the stage and sample due to illumination by the lamp was too small to significantly influence the measured lifetime. All measurements were performed at $T = 303 \pm 2$ K.

The normalized defect concentration, N_t^* , was calculated from the measured lifetime according to (1). The lifetimes were measured at an injection level corresponding to one tenth of the doping level for each sample. A doping-independent injection level would be problematic since the doping concentration changes the lifetime dependency on the injection level substantially as seen in Fig. 1.

As an indicator of the mismatch between the measured data and the best-fit functions a calculation of the mean deviation χ^2 was performed (a lower value for χ^2 indicates a better match to the experimental data). χ^2 is defined as

$$\chi^2 = \frac{1}{N} \sum_i \left(\frac{y - y_i}{\sigma_i} \right)^2, \quad (11)$$

where y is the fitted value for a given point, y_i , is the measured data value for the point and σ_i is the standard deviation of y_i . σ_i is estimated based on algebraic propagation of errors since we do not have multiple repetitions of the measurement for all samples. N is the number of data points minus the number of adjustable coefficients in the fit functions. A Levenberg-Marquardt algorithm is used to search for the minimum value

of χ^2 by an iterative process performed by the computer program IGOR [14].

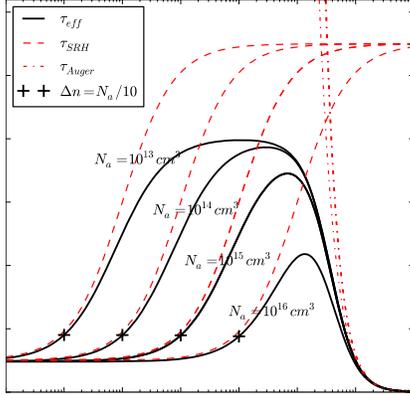


Fig. 1. Simulation showing the impact of the doping concentration on lifetime as a function of carrier injection level. Comparing the lifetime of samples with different doping concentrations measured at a fixed injection level of e.g. 1×10^{15} may lead to systematic error.

III. RESULTS

Logarithmic plots of the time resolved lifetime decays clearly display the previously reported rapid and slow decays (see Fig. 3). Other groups have reported that degradation typically occurs during the first hours of illumination [8],[15],[16]. In this work we experience that only samples with a hole concentration p_0 above $9 \times 10^{15} \text{ cm}^{-3}$ saturate within 24 hours. This is shown in Fig. 2 where the time where τ has reached 99% of the total lifetime loss is plotted as a function of p_0 . Saturation times were also compared to oxygen concentrations but no correlation could be seen.

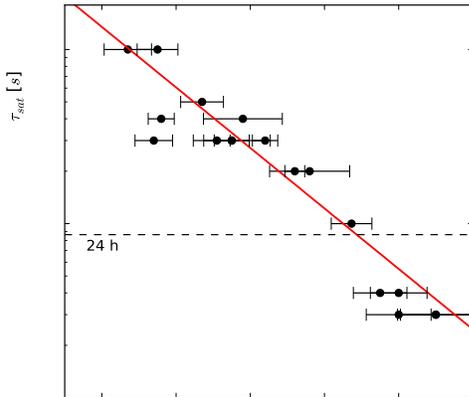


Fig. 2 The time [s] where τ has reached 99% of the total loss as a function of hole concentration. After 24 hours only the samples with a hole concentration above $9 \times 10^{15} \text{ cm}^{-3}$ have reached saturation. The red line is a best fit power

curve of the data and may serve as an empirical expression predicting the approximate lifetime saturation time t_{sat} in seconds: $t_{sat} = 9 \times 10^{43} [p_0]^{-2.45}$. The oxygen concentration is not measured for all samples but for those that are measured it ranges from $7 \times 10^{17} - 1 \times 10^{18} \text{ cm}^{-3}$. All measurements are performed at $T = 303 \pm 2 \text{ K}$

As mentioned in the introduction other groups [11], [12] have commented that the measured decays can be difficult to fit with a single exponential decay and a double exponential function has therefore been used to fit the decay data. In this work we have found that for both the rapid and the slow decay, for all the investigated samples, a much better fit could be obtained if (10) is replaced by a rate equation of *second* order, so that

$$-\frac{dN'_t}{dt} = 2k(N'_t)^2 \quad (12)$$

The carrier lifetime as a function of time is then no longer given by an exponential, but as a sigmoidal function on the following form:

$$\tau(t) = \frac{\tau_0 + \tau_\infty \left(\frac{t}{w}\right)}{1 + \left(\frac{t}{w}\right)}. \quad (13)$$

Here τ_∞ is the defect saturated lifetime and w is the half-life, i.e. the time where the initial lifetime, τ_0 , is halved:

$$w = \frac{1}{2k} \frac{\tau_\infty^2}{\tau_0 - \tau_\infty} \quad (14)$$

Note that for the slow degradation τ_0 is in fact the lifetime where the rapid degradation has saturated. A full derivation of (13) from the second order rate equation is given in the appendix.

An example showing the best fit curves from the solutions of the first order (9) and second order (13) rate equations is given in Fig. 3.

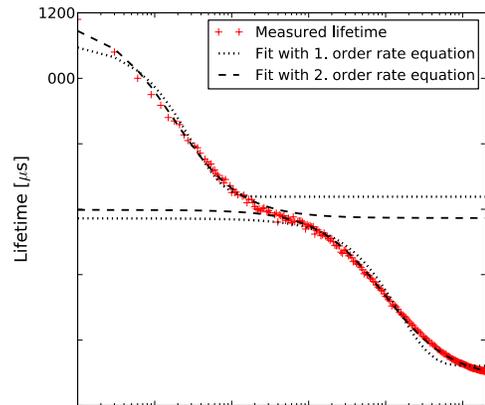


Fig. 3 Plot showing the best-fit solutions of two different rate equations for an experimental LID lifetime decay curve.

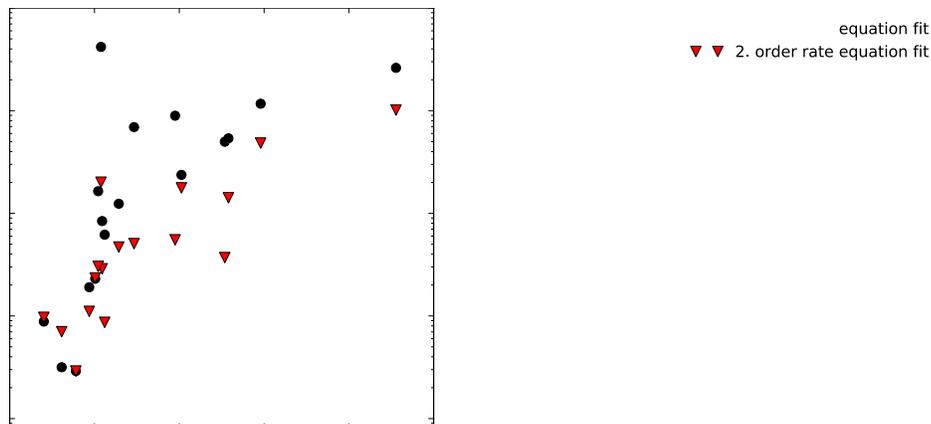


FIG. 4. The mean deviation χ^2 for the two different fit functions is plotted for each sample as a function of initial lifetime before light exposure. a) Rapid lifetime decay b) Slow lifetime decay.

Fig. 4a and 4b shows a comparison of the χ^2 values of the fit using the first and second order rate equations for the rapid and the slow decays, respectively. The χ^2 values are given as a function of the initial lifetime τ_0 before any light exposure for each sample. χ^2 is increasing with initial lifetime for both the rapid and the slow decay. This is mainly because we need a large dynamic injection range through the entire lifetime decay. High lifetime samples have a larger lifetime span from start to end and so the scatter in the lifetime against injection level becomes larger.

For the slow lifetime decay (Fig. 4b) the mean deviation χ^2 is substantially lower for the second order fit compared to the first order fit for all investigated samples. This is also true for the rapid lifetime decay (Fig. 4a) except for the samples with the lowest initial lifetimes. This is the region of the plot where the initial lifetime is low and a hypothesis is that the initial lifetime of these samples are limited by some other defect, and that the presence of this defect influences the kinetics of the rapid lifetime decay.

Comparisons of the χ^2 values for the rapid and the slow decay show that χ^2 is in general higher for the rapid decay fits. This is because the number of data points is limited. More measurements during the rapid decay will alter the outcome since the lifetime, at this point, is sensitive to even the short flash from the QSSPC measurement (<1 s).

The half width w in (14) is the time responsible for 50 % of the lifetime degradation. Accordingly, this parameter reveals information on the kinetics of the generation process. In Fig. 5 the half width of the slow decay is plotted as a function of p_0 for the investigated samples. The plot indicates an approximate quadratic decrease of the half width with p_0 using the exponent as a free parameter. Lifetime decay analysis has also been performed on a set of compensated p-type boron and phosphorus doped Cz-Si samples and the results showed that w for these samples falls far outside the indicated trend lines indicated in Fig. 5 when plotted against $[B_s]$ instead of p_0 . Hence, the results from this investigation points towards a

correlation between the kinetics of the defect reaction rate and p_0 rather than $[B_s]$. This is in accordance with the work of Macdonald et al. from 2009 [6].

Oxygen concentration measurements performed by Fourier transform infrared spectroscopy were performed on a selection of the samples. No correlation between the oxygen concentration and the half width w could be seen, indicating that the oxygen is not playing a role on the kinetics of the defect generation. This is in accordance to the work by Bothe et al. [10].

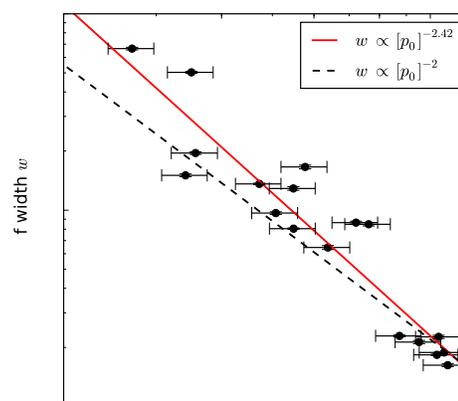


Fig. 5 Half width w for all measured slow lifetime decays as a function of the hole concentration. For the best fit power curve (solid line) w is proportional to $[p_0]^{-2.42}$. The function corresponding to a decrease proportional to $[p_0]^{-2}$ is indicated by the dashed line.

IV. DISCUSSION

A. 2. order reaction kinetics

The purpose of a kinetic study is to obtain information concerning the reaction mechanism. Agreement between a

specific rate equation and the experimental data, is then consistent with, but not a proof of that type of reaction. The second order rate equation used to describe the lifetime degradation in this work shows that the rate of the reaction is changing with the square of the latent defect concentration $[N_t']^2$. A second order rate equation often indicates that two species of the same type react together to form one product species. The rate equation of a reaction with a multi-step mechanism cannot, however, in general, be deduced from the stoichiometric coefficients of the overall reaction. The equation may involve fractional exponential coefficients, or it may depend on the concentration of an intermediate species. Nevertheless, the fit to the LID decay by a second order rate equation might indicate that two defect species of the same kind react in the process of forming the recombination active defect. It is then the concentration of these reactants that acts as N_t' and is depleted during the course of the reaction. The full reaction still involves steps where holes and electrons play a role, and the complete picture of the defect generation process is still missing.

A conclusion we can draw, however, is that a reaction between two individual reactant molecules of different species cannot be the case, as the solution of a second order rate equation with two different reactants cannot fit the measured data.

B. The role of p_0

From Fig. 5 we saw that for the slow lifetime decay there is a correlation between the half-life w and the hole concentration p_0 . According to (14) w is inversely proportional to k and proportional to $(\tau_\infty^2/\tau_0 - \tau_\infty)$. In our analysis we do not find any correlation between p_0 and $(\tau_\infty^2/\tau_0 - \tau_\infty)$. We therefore conclude that it is mainly k that is influenced by p_0 , giving $k \propto p_0^n$, with n being between 2 and 2.5. This opens for the possibility of two potential roles of p_0 in the defect reaction; The first scenario is that p_0 is embedded in the rate constant as an additional reactant, and that k acts as a pseudo rate constant for instance as $k = p_0^2 k'$, where k' is the actual rate constant. A requirement for embedding a reactant in a pseudo rate constant in this way is that its concentration must not change during the reaction, i.e. it must be in great excess ($p_0 \gg N_t'$). p_0 is commonly occurring in the order of $1 \times 10^{15} - 1 \times 10^{17} \text{ cm}^{-3}$ in boron-doped silicon and N_0 is estimated to be less than $1 \times 10^{13} \text{ cm}^{-3}$ [17], hence the requirement is met. Note that a reactant embedded in the pseudo rate constant still influences the rate of the reaction, but it means that the second order kinetics is describing changes in N_t' exclusively. We can, of course, not rule out that there are other reactants embedded in k , meaning that we cannot exclude other reactants as contributors to the defect. They must, however, also be in great excess.

The other possible explanation for the role of p_0 in the defect reaction is for p_0 to act as a catalyst. This hypothesis can be regarded to support the model of Voronkov et al., presented in the introduction, in the sense that they propose that there are two activation barriers for each decay that must be overcome in order to complete the defect. A catalyst

will thus work to decrease the activation barrier between one of the intermediate steps in the reaction and thereby decrease the half-width of the degradation decay with increased catalyst concentration. Note that the first explanation model where p_0 is embedded in a pseudo rate constant does not contradict Voronkov's model. Several groups have already reported the involvement of two holes/two boron atoms in the reaction kinetics [5-9].

C. Rate determining step

The measured overall rate of a reaction is controlled by the rate of the slowest step, the rate-determining step. The rate constant, k , will thus be designated this particular step. According to the data in Fig. 5 and the discussion in chapter IV-B k is strongly correlated with p_0 . Relating this to the slow defect reaction model by Voronkov et al. [7],[8], summarized in the introduction, these results point towards step 2 (7) as the rate determining step. This, however, contradicts the suggestion by Voronkov et al., which proposes the last step as rate limiting. Step 1 (6) can under any circumstances not be rate-determining since the rate of the defect reaction is independent of Δn [4].

For the fast defect the reaction steps 1, 2 and 3, described in (2)-(4) can all be excluded as rate determining steps because of step 3; In this step the presence of excess electrons cause a shift of the electron quasi-Fermi level across the energy level of the defect center in its passive state [4], [10]. This cannot be the rate determining step [4], but it also means that none of the earlier steps can be rate determining since the reaction does not start until the excess carriers are present.

V. CONCLUSION

A range of boron-doped Czochralski silicon materials have been analyzed with regards to light induced degradation. Curve fitting of the measured data to a single exponential (the solution of a first order rate equation) was attempted but a much better fit was achieved by a solution of a second order rate equation indicating that the defect generation process can be described by second order reaction kinetics. The rate equation describing our system shows that the rate of the reaction is changing with the square of the latent defect concentration $N_t'^2$. The concentration of holes is identified to control the half-life of the degradation process and is either playing a role as a reactant in excess or as a catalyst.

APPENDIX I

Derivation of eq. 7 from the second order rate equation

Initial arguments:

$$N_0 = N_t^* + N_t' = \text{constant} \quad (15)$$

$$\begin{aligned} t = 0: N_t^* &= 0, N_t' = N_0 \\ t = \infty: N_t^* &= N_0, N_t' = 0 \end{aligned} \quad (16)$$

where N_0 is the initial latent defect concentration, N_t^* is the

concentration of generated defects, N_t' is the concentration of latent defects. N_0 is equal to saturated defect concentration, which can be calculated the following way:

$$N_0 = N_{t,sat}^* = \frac{1}{\tau_\infty} - \frac{1}{\tau_0} \quad (17)$$

From the initial arguments and Eq. 1 we obtain the following relation:

$$N_t'(t) = N_0 - N_t^* = \frac{1}{\tau_\infty} - \frac{1}{\tau(t)} \quad (18)$$

A second order rate equation is a simple differential equation:

$$-\frac{dN_t'}{dt} = 2k(N_t')^2 \quad (19)$$

Using the boundary conditions in (16), the solution becomes

$$N_t'(t) = \frac{1}{\frac{1}{N_0} + 2kt} \quad (20)$$

Inserting for $N_t'(t)$ from Eq. 15 gives:

$$N_t^*(t) = N_0 - \frac{1}{\frac{1}{N_0} + 2kt} \quad (21)$$

and inserting for N_0 (Eq. 17) and $N_t^*(t)$ (Eq. 1) in Eq. 21, gives the final result:

$$\tau(t) = \frac{\tau_0 + \tau_\infty \left(\frac{t}{w}\right)}{1 + \left(\frac{t}{w}\right)} \quad (22)$$

where w is defined as

$$w = 2k \frac{\tau_\infty^2}{\tau_0 - \tau_\infty} \quad (23)$$

This expression can be successfully fitted to both the rapid and the slow lifetime degradation decay for all investigated samples.

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