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A unified parameterization of the formation of boron oxygen defects and their electrical activity

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Abstract

The magnitude of light-induced degradation of solar cells based on Czochralski grown silicon strongly depends on material properties. We have performed experiments to describe the activation and recombination activity of boron oxygen defects in boron compensated *n*-type silicon. Compensated *n*-type material enables flexible assessment of charge carrier influences on the defect that cannot be distinguished on *p*-type material. The results can be generalized to *p*-type material and thus provide valuable insights to the defect. Our measurements demonstrate the two-level defect nature of the slow-formed boron oxygen defect component and allow the study of the dopant dependency of the defect concentrations. Our findings strongly support a revision of the existing model of the defect composition.

Based on the experimental results and literature data we have created a parameterization of the lifetime limitation in silicon due to BO defects. Established findings from literature for uncompensated *p*-type silicon are taken into account and ensure general validity. The parameterization is useful to discuss BO defect influences and can serve to predict material properties after LID.

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Keywords: LID; boron oxygen defects; parameterization; lifetime limitation; Czochralski silicon

1. Introduction

The widely used Czochralski growing method introduces large amounts of oxygen into the silicon crystal. In combination with the standard dopant species boron, the introduced oxygen forms a defect addressed as the BO defect. This defect is transformed into a recombination active configuration under illumination, causing a decline of minority charge carrier lifetime that is known as light-induced degradation (*LID*). Many studies have suggested methods to cure this degradation effect [1, 2] *(and references therein).* However, the process is empirically optimized and often incomplete because the mechanism is not sufficiently understood, yet.

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It is widely accepted in literature that the magnitude of LID shows a close correlation to the oxygen content incorporated in the silicon bulk. Therefore the Czochralski crystal growth process has been optimized in order to minimize oxygen contamination, reducing the problem of LID.

In order to minimize the detrimental effects of LID in silicon based devices for a given material, it is crucial to be able to assess and predict the degradation. Knowledge of the resulting material properties allows e.g. the adaption of structure designs or an evaluation of financial impacts of further optimization the material quality. The available studies regarding this [3, 4] do not take recent findings on the recombination activity and on special features of compensated silicon into account. We have developed a new parameterization of the lifetime limitation due to BO defects based on results both from literature and recent studies conducted at Fraunhofer ISE. Light soaking studies were conducted on compensated *n*-type silicon, allowing us to derive new parameterizations of the recombination activity and activation kinetics of BO defects. The conclusions drawn on the compensated *n*-type material can be transferred to *p*-type material, providing valuable insights on the defect that are not easily assessable otherwise. The main advantage of compensated *n*-type regarding studies of the BO defect is the possibility to decouple the boron concentration from the hole concentration, which can itself be controlled via illumination. The combination of the injection dependent lifetime parameterization and the kinetic model allows extraction of defect concentrations on both *n*- and *p*-type silicon from experiments that are more physically meaningful than the concentrations determined before. The determined effective concentrations are now related to the actual concentration of defect centers and can serve as a measure in defect studies.

2. Experiment

2.1. Sample preparation and light soaking

The light soaking experiments were performed on samples cut from different positions of two compensated *n*-type crystals doped with both boron and phosphorus and wafers from several conventional boron doped crystals. The net doping concentration $n_0 = [P] - [B]$ of the *n*-type samples varies from $1.1 \cdot 10^{15}$ and $3.6 \cdot 10^{16}$ cm^{-3}, the *p*-type samples feature a similar variation of the doping concentration $p_0 = [B]$ from $1.1 \cdot 10^{15}$ to $1.4 \cdot 10^{16}$ cm^{-3}. FTIR measurements of the interstitial oxygen concentration resulted in $[O_i]$ of $5\text{-}7 \cdot 10^{17}$ cm^{-3} for the *n*-type and $3.1\text{-}8.5 \cdot 10^{17}$ cm^{-3} for the *p*-type samples. Lifetime samples were processed featuring wet chemical etching, a $POCl_3$ gettering step followed by an emitter etch back and surface passivation with dielectric films. Silicon-rich oxynitride-stacks deposited by PECVD [5] were used to passivate the *n*-type samples and Al_2O_3 deposited via *plasma activated atomic layer deposition* for *p*-type samples. The *n*-type samples underwent a short high temperature step to activate the surface passivation and the *p*-type samples were annealed at 425°C for 25 minutes to achieve optimal surface passivation. After annealing in the dark high lifetimes in the millisecond regime were achieved on all samples, proving the high surface passivation quality.

Lifetime evolution under controlled temperatures and intensities was monitored with modulated photoluminescence and QSSPC measurements. Compensation effects on the charge carrier mobility were taken into account and corrected for.

A second type of experiments was conducted specifically to study the fast initial lifetime decay due to the formation of the fast-formed component of the BO defect FRC. In order to achieve a high temporal resolution of lifetime measurements while maintaining defined illumination conditions, the samples were illuminated with a harmonically modulated laser and the resulting photoluminescence response was measured. We applied a self-consistent approach described in [6] to evaluate the PL response over the full modulation period to determine injection dependent lifetime curves with a repetition frequency of 40 Hz. The photoluminescence signal was sampled with a sampling rate of 40 kHz, giving a very high temporal resolution of the current injection density under illumination. After the first rapid degradation multiple cycles were averaged in order to improve the signal quality and reduce the amount of data. Knowledge of the injection density was found to be of crucial importance for the case of LID under modulated illumination, as the degradation rate was found to depend on the charge carrier concentrations.

2.2. Injection dependent lifetime

We have demonstrated in recent publications, that the slow-formed BO defect component (*SRC*) is formed or activated in compensated *n*-type silicon, as well [7, 8]. A thorough study of the injection dependent lifetime of the SRC in *n*-type can be found in Ref. [7]. The crucial result from this study is, that the SRC does not follow the simple Shockley-Read-Hall statistics [9, 10] for defects with one energy level in the silicon band gap. We have demonstrated, that the previously common assumption of one deep donor level located at 0.41 eV below the conduction band edge can in fact only describe a narrow range of *p*-type material with a sufficient precision. In order to describe the full injection dependent lifetime of SRC in other material, a more advanced parameterization including a second energy level – an acceptor level at 0.26 eV above the valence band edge – is necessary. It had been shown before, that the FRC exhibits a similar behavior [11] and an advanced lifetime formulation for such defects has been established [12]. The applied method of advanced lifetime spectroscopy does not by itself provide absolute values of the capture coefficients of the defect levels. However, it is possible to determine the relations between the capture coefficients, allowing them to be determined relative to one coefficient. For SRC we have determined the values in relation to the hole capture of the donor level $\alpha_{d,h}$ The other coefficients then result to be $\alpha_{d,e} = 9.5\times\alpha_{d,h}$, $\alpha_{a,e} = 0.54\times\alpha_{d,h}$ and $\alpha_{d,h} > 211\times\alpha_{d,h}$, respectively. For more details on the parameterization, we refer to Ref. [7].

Our latest experiments further support the indication, that measured lifetimes are best described, when both FRC and SRC are regarded as two-level defects. Examples of the improved data description utilizing the two-level parameterizations are shown in Fig. 1.

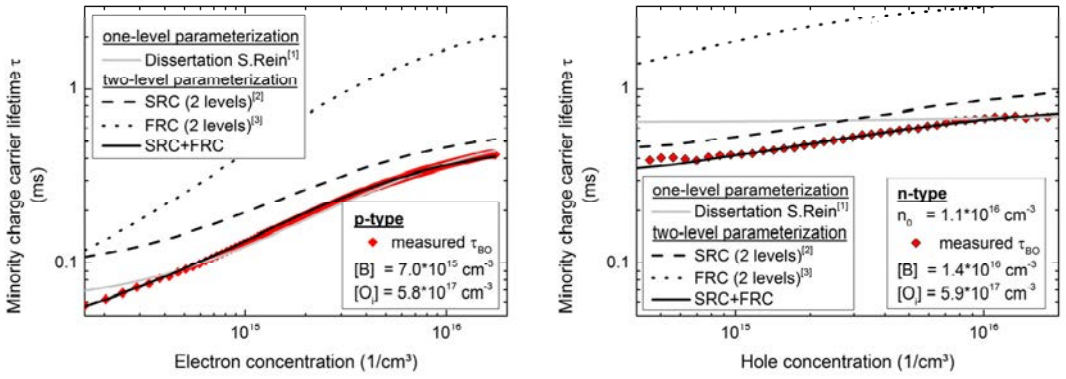

Fig. 1: Exemplaric injection dependent lifetime curves of samples limited by the BO defect. (left) p-type sample, (right) n-type sample The symbols denote experimentally determined BO lifetime limitations (derived from measurements in annealed and degraded state).

2.3. Modelling of the degradation kinetics

The measured temporal evolution of the charge carrier lifetimes of the *n*-type samples under long-term illumination was studied in detail in Ref. [8]. A careful assessment of the hole concentration under illumination p_{illum} taking the complex lifetime limitation due to the formation of FRC and SRC into account was identified to be crucial. The formation rates of both defect components R_{SRC} and R_{FRC} were found to depend on this quantity. While a quadratic dependence of R_{SRC} on p_{illum} was proven by the good quantitative matching between measurements and simulation (e.g. in Fig. 2), the coarse temporal resolution in the early stages of degradation complicated definite statements on the FRC formation.

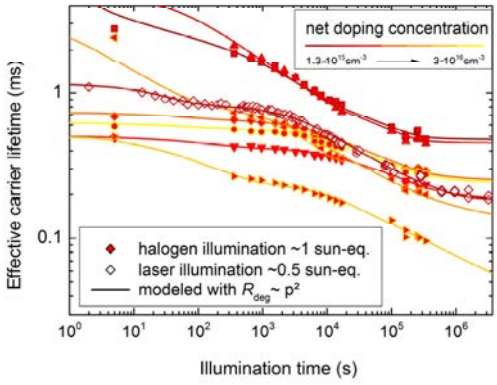

Fig. 2: Temporal evolution of the minority charge carrier lifetime of compensated n-type samples under illumination. The lines represent modelling of the defect formation according to Ref. [8].

Modelling the temporal evolution of the lifetime under illumination allows the determination of defect concentrations from the magnitude of lifetime degradation. The results of the studies on compensated *n*-type silicon presented in Ref. [8] are shown in Fig. 4 and discussed in Section 3.1.

The experiment conducted under modulated laser illumination provided a greatly enhanced temporal resolution without loss of precise knowledge of $p_{illum}(t)$ that would have occurred with most common lifetime measurement techniques. Due to the high frequent variation of the illumination intensity, a simple determination of p_{illum} from the generation rate and the lifetime is not possible in this case. However, the photoluminescence signal was sampled with a kHz frequency and selfconsistently evaluated, thus providing $p_{illum}(t)$ quasi-continuously.

Usually, BO defect formation rates R_{BO} are determined from exponential fits to the temporal evolution of the effective defect concentration $N_t^* = 1/\tau_{eff} - 1/\tau_0$ according to eq. 1.

$$N_t(t) = N_{t,\max} \cdot \left(1 - \exp\left[-t \cdot R_{BO}\right]\right) \tag{1}$$

However, this approach does not provide physically meaningful results for non-constant degradation rates - especially for the case of the strongly fluctuating rate that is expected in our experiment due to the modulated hole concentration $p_{illum}(t)$. Therefore we evaluated the data utilizing a variable transformation from the time regime t to a time-p_{illim}^{A}-regime t^* with fixed exponent A, allowing simple exponential fits to provide more precise formation rates.

$$\begin{aligned} N_t(t, p_{illum}(t)) &= N_{t,\max} \cdot \left(1 - \exp\left[-t \cdot R_{BO}(t, p_{illum}(t))\right]\right) \\ t \quad &\Rightarrow \quad t^* = \int_0^t t \cdot p_{illum}{}^{A}(t)\, dt \\ R_{BO}(t, p_{illum}(t)) \quad &\Rightarrow R_{BO^*}(t^*, A) \\ N_t(t^*) &= N_{t,\max} \cdot \left(1 - \exp\left[-(t^* \cdot R_{BO}\right]\right) \end{aligned} \tag{2}$$

We took into account that both FRC and SRC are formed simultaneously, resulting in the superposition of two such formation equations. Good description of the data was achieved, when the data was analyzed assuming factors

of A=2 for both FRC and SRC and the rates R_{BO} were chosen in analogy to results from p-type silicon as presented in Ref. [4]. An example is shown in Fig. 3.

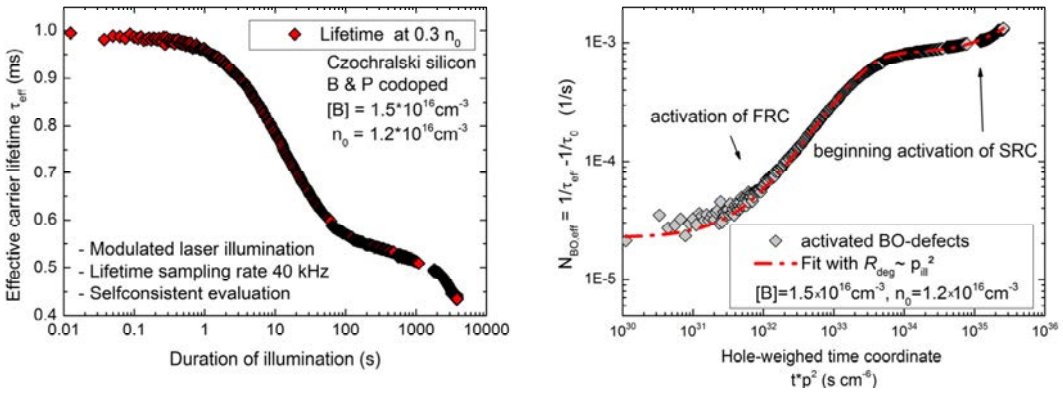

Fig. 3: Example of the transformation of the lifetime data (recalculated to effective defect concentrations $N_{BO,eff}$) from the t-regime to the t*-regime for the case A=2. The resulting high fit quality proves that the formation rate R_{FRC} exhibits a quadratic dependence on p_{illum}.

The exponent A=2 in the transformed time regime t* corresponds to a quadratic dependence of $R_{BO}(t)$ on p_{illum}, i.e. $R_{BO} = R_{BO*} * p_{illum}^2$. It should be noted, that for the case of low sample lifetimes a deviation from the quadratic dependence was observed that we attribute to the occurrence of a backwards transition reaction, as discussed in Ref. [11].

3. Parameterization of the BO lifetime limitation

3.1. Relevance of the injection dependent lifetime

The common approach to study the BO defect has been by means of an effective defect concentration N_t* derived from the difference in lifetime before and after activation of the defect. It has been convention to compare this data only for lifetimes evaluated at 10% of the dark hole concentration p_0 because this minimizes the effect of injection dependent charge carrier lifetime. This approach allows comparison of effective defect concentrations on most p-type materials, because the injection dependent lifetime in p-type material affected by the BO defect is dominated by SRC recombination, which is in turn dominated by an energy level deep in the silicon band gap. This causes the injection dependence to be similar for a range of dopant concentrations. However, the two-level natures of both FRC and SRC imply the injection dependent lifetime to exhibit a complex reaction to the doping level. We have experimentally found this effect for lowly doped p-type silicon and especially for n-type material [7, 13]. This affects the determination of effective defect concentrations from lifetime measurements, as the same absolute defect concentration in samples of different doping concentration would result in different determined effective concentrations. Thus this effect can hinder the identification of underlying trends and mechanisms.

Knowledge of the injection dependence of the lifetime allows the determination of defect concentrations that are independent of the injection level. We have determined effective defect concentrations from the light soaking experiments on compensated n-type silicon mentioned above. When the injection dependent lifetime is taken into account and the influence of the interstitial oxygen concentration [O_i] is normalized, the resulting SRC concentrations [SRC] show no clear dependence on the net doping concentration n_0, as shown in Fig. 4, or the boron concentration [B] (*not shown*). This contradicts the involvement of interstitial boron atoms B_i in the composition of SRC that was suggested in Ref. [11]. Due to the negative charge of B_i in n-type silicon, an involvement of B_i in the formation of SRC at moderate temperatures should create a linear dependence of [SRC]/[O_i] to n_0.

The close value range for the three studied crystals suggests that $[SRC]/[O_i]^2$ could be even the same or similar in all compensated *n*-type crystals. As the studied crystals were likely grown under similar conditions (*as they were intended for photovoltaic research*) an important precursor of the defect could have been formed alike in all three crystals.

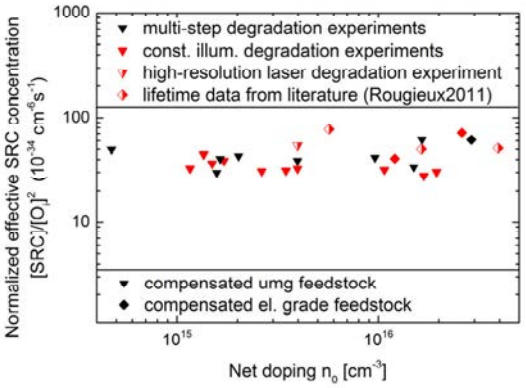

Fig. 4: Defect concentrations determined from the magnitude of degradation via the kinetic fits to light soaking experiments as shown in Fig. 2.

3.2. Parameterization

In order to predict the lifetime limitation due to BO defects for a given material, we have combined the parameterizations of the injection dependent lifetime from Refs. [11] and [7] with trends established and discussed in literature. The resulting parameterization was then tested and partially adapted to results from the light soaking experiments mentioned in section 2 of this contribution. It should be noted, that the studied *p*-type samples originated from conventional uncompensated silicon crystals. The compensated *n*-type samples were also included in the studies to determine the injection dependence and the defect concentration level in compensated *n*-type.

All samples underwent a phosphorus gettering step, which is known to affect the lifetime limitation due to BO defects. Usually the effective concentration is reduced by a factor of 2-3 but provides more comparable data between similar crystals, indicating that the high temperature during the $POCl_3$ diffusion might compensate for different cooldown conditions during crystal growth. Figure 5 shows projections of the parameterization of the expected lifetime at a fixed injection level of 10% of the net doping concentration. The equations utilized for the parameterization are given in the appendix of this contribution. The values are normalized to $[O_i] = 10^{17}$ cm^{-3} based on the assumption that both [*FRC*] and [*SRC*] have a quadratic dependence on $[O_i]$, as established e.g. in Ref. [4]. Therefore the lifetime of a material with a given oxygen concentration would be calculated by dividing the given value by a factor of $([O_i]/10^{17}\ \text{cm}^{-3})^2$.

The dashed and dotted lines in the graphs indicate the limitation due to FRC and SRC, respectively. The generally higher lifetime limit of FRC implies that the recombination is dominated by SRC for the given injection level for net doping levels from 10^{14} to 10^{17} cm^{-3}. However, the different slopes of the curves indicate a shifting importance of the defect components with varying net doping concentration that is caused by the different influences on their injection dependent lifetimes. The growing importance of FRC in low resistivity *p*-type silicon might become relevant for application of such material. It is not yet clear, what defects limit the lifetime after the process to cure BO defects (*i.e. permanent recovery* [1, 2, 14]) and whether both defect components are deactivated or only SRC. Thus FRC could become more relevant, especially as certain solar cell concepts could benefit from higher base material doping, once the BO defect limitation is reduced.

The parameterization for *p*-type material can be regarded as an update to Ref. [3] and is based on the trends established in Ref. [4]. These established trends have partially been reconsidered in terms of the effect due to the injection dependence as discussed in section 3.1. Based on Refs. [15, 16] we have assumed the linear dependence to the boron concentration to be actually caused by the dependence of N_t* on the hole concentration p_0 for most *p*-type Czochralski grown crystals. Materials incorporating significant amounts of other acceptors, the boron concentration might be more significant instead as indicated by Refs. [17, 18]. We consider this an uncommon case and have therefore neglected it for this parameterization. The grey area around the total BO limitation indicates an uncertainty range of the parameterization that has been estimated from the scatter of data points in Ref. [4] and the uncertainty that we estimated for our own data used as a benchmark for the parameterization. A detailed study on the uncertainties of the underlying dependencies - *i.e. linearity to p_0 and quadratic dependence on* [O_i] *for both defect components* – was beyond the scope of this work.

The parameterization for compensated *n*-type material is based on the findings and data from Refs. [7, 8, 11, 19, 20]. It should be noted that this data base is small related to the many thorough studies on *p*-type material. Only few further studies on LID in compensated *n*-type exist and only those taken into account provided enough information concerning the samples and measurements for us to include the results. The most important findings from our studies concerning the BO defect in compensated *n*-type are summarized here:

- The same defect components FRC and SRC as in *p*-type are activated in compensated *n*-type upon illumination.
- There is no apparent trend in [*FRC*] or [*SRC*] on [B] or n_0 once they are normalized by their oxygen content.

The parameterization of the BO lifetime limitation of compensated *n*-type silicon is therefore based on the assumption, that a constant concentration of BO defects is present in all studied *n*-type materials. This assumption is drawn from the results presented in Fig. 4. The uncertainty range indicated in Fig. 5 was created from the scatter of the [*SRC*] data shown in Fig. 4 and [*FRC*] (*not shown*). A normalized concentrations of SRC of 45×10^{-34} cm^{-6}s^{-1} was chosen for the parameterization, as it provides a good median of the values determined from the kinetic study in Ref. [8]. The data points included in the figure were derived from the injection dependent lifetimes at the end of the light soaking experiment shown in Fig. 2. It is apparent from the kinetic fits (*indicated by the lines in Fig. 2*), that the samples were not fully degraded at this time. This is also reflected by their normalized higher lifetimes compared to the prediction of the parameterization. Their large error bars result from very unprecise determination of [O_i] and the assumption of additional measurement uncertainties due to the compensation.

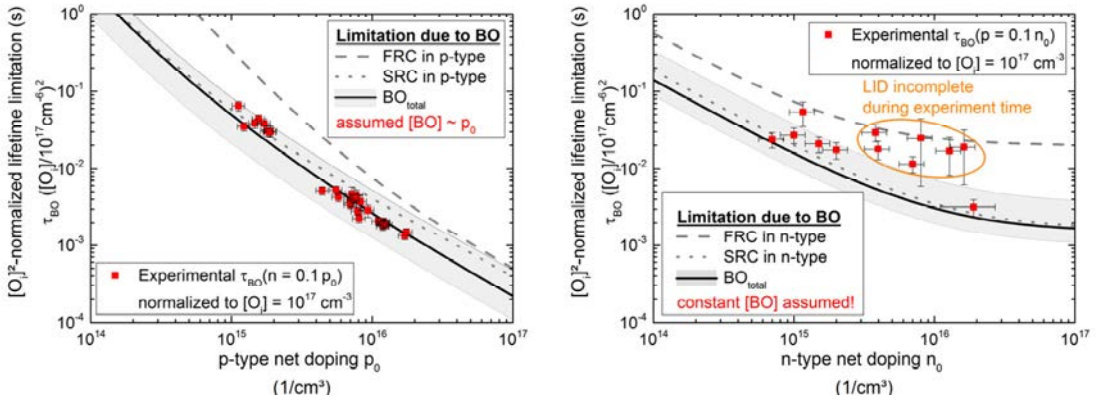

Fig. 5: Parameterization of the lifetime limitation due to BO defects for a fixed injection density of 10% of the net doping. (left) for *p*-type material doped (or significantly codoped) with boron (right) for B-compensated *n*-type material. The values are normalized to an interstitial oxygen concentration [O_i] of 10^{17} cm^{-3} assuming a quadratic dependence. In order to predict the lifetime limitation for a material of a given net doping and [O_i], the lifetime value must therefore be divided by a factor of ([O_i]/10^{17} cm^{-3})². The data points show results from light soaking studies performed at Fraunhofer ISE.

4. Conclusion

We have recapitulated the results from recent studies on LID in compensated *n*-type silicon caused BO defects conducted at Fraunhofer ISE. We report very good agreement of measurement data when both defect components FRC and SRC are simulated according to the two-level Shockley-Read-Hall statistics from Ref. [12] with the parameterizations for FRC [11] and SRC [7]. Taking the injection dependent lifetimes of BO defects into account, it is possible to derive effective defect concentrations that can be used to develop and test defect models. This is demonstrated for the example of compensated *n*-type samples, where we do not find the dependence of [*SRC*] that was to be expected from the model suggested in Ref. [11], contradicting the involvement of interstitial boron atoms in the SRC.

An experiment based on light soaking with modulated laser illumination has been demonstrated and we have shown that the evaluation of the resulting data supports the theory that FRC activation is driven by the hole concentration under illumination. Namely a quadratic dependence has been found, indicating the involvement of two holes in the process.

The results concerning the injection dependent lifetimes of samples limited by BO defects have been combined with trends and findings from literature to create an empirical parameterization of the lifetime limitation for given materials. We have discussed the approach that was used to derive the parameterization and some of the implications that can be drawn from the parameterization.

Acknowledgements

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Appendix A. Equations and parameters of the shown lifetime limitation parameterization

The lifetime limitation due to a two-level BO defect with an acceptor level E_a and a donor level E_d in dependence of the ratio of holes to electrons $Y=p/n$ for *n*-type or electrons to holes for *p*-type, $X=p/n$ according to Ref. [12]:

$$\tau_{h,n-type} = \frac{1+\left(\frac{\frac{Q_d n_d (1-Y)}{n_0}+Y}{Q_d+\frac{p_d(1-Y)}{n_0}}\right)+\left(\frac{1+\frac{p_a(1-Y)}{Q_a n_0}}{\frac{n_a(1-Y)}{n_0}+\frac{Y}{Q_a}}\right)}{N_{BO}\left(\frac{\alpha_{a,e}}{Q_d+\frac{p_d(1-Y)}{n_0}}+\frac{\alpha_{d,h}}{\frac{n_a(1-Y)}{n_0}+\frac{Y}{Q_a}}\right)} \quad \text{(A1 a)}$$

$$\tau_{e,p-type} = \frac{1+\left(\frac{\frac{Q_d n_d (1-X)}{p_0}+1}{Q_d X + \frac{p_d(1-X)}{p_0}}\right)+\left(\frac{X+\frac{p_a(1-X)}{Q_a p_0}}{\frac{n_a(1-X)}{p_0}+\frac{1}{Q_a}}\right)}{N_t\left(\frac{\alpha_{d,e}}{Q_d X + \frac{p_d(1-X)}{p_0}}+\frac{\alpha_{a,h}}{\frac{n_a(1-X)}{p_0}+\frac{1}{Q_a}}\right)} \tag{A1b}$$

with

$$n_a = N_C \cdot \exp\left[\frac{-(E_C - E_a)}{k_B T}\right] \quad , \qquad n_d = N_C \cdot \exp\left[\frac{-(E_C - E_d)}{k_B T}\right] \tag{A2a,b}$$

$$p_a = N_V \cdot \exp\left[\frac{-(E_a - E_V)}{k_B T}\right] \quad , \qquad p_d = N_V \cdot \exp\left[\frac{-(E_d - E_V)}{k_B T}\right] \tag{A2c,d}$$

$$Q_a = \frac{\alpha_{a,e}}{\alpha_{a,h}} \quad , \qquad Q_d = \frac{\alpha_{d,e}}{\alpha_{d,h}} \tag{A3a,b}$$

Values used in the parameterization presented in the contribution:
FRC (*adapted from* [11])
$E_d = E_C$ - 0.27 eV
$E_a = E_V$ + 0.30 eV
$\alpha_{d,h} = 1/65\ \alpha_{d,e}$
$\alpha_{a,e} = 1/481\ \alpha_{d,e}$
$\alpha_{a,h} = 0.135\ \alpha_{d,e}$

SRC (*from* [7])
$E_d = E_C$ - 0.41 eV
$E_a = E_V$ + 0.26 eV
$\alpha_{d,e} = 9.5\ \alpha_{d,h}$
$\alpha_{a,e} = 0.54\ \alpha_{d,h}$
$\alpha_{a,h} > 211\ \alpha_{d,h}$

Defect concentrations N_{BO}: (*based on* [4, 7])
$N_{SRC,p\text{-}type}(p_0, [O_i]=1\times10^{17}\ \mathrm{cm^{-3}}) = ([O_i]/10^{17}\ \mathrm{cm^{-3}})^2/25 * 1.2\times10^{-4} * (p_0/10^{16}\ \mathrm{cm^{-3}})$ 1/s
$N_{FRC,p\text{-}type}(p_0, [O_i]=1\times10^{17}\ \mathrm{cm^{-3}}) = ([O_i]/10^{17}\ \mathrm{cm^{-3}})^2/25 * 0.7\times10^{-4} * (p_0/10^{16}\ \mathrm{cm^{-3}})$ 1/s

$N_{SRC,n\text{-}type}([O_i]=1\times10^{17}\ \mathrm{cm^{-3}}) = ([O_i]/10^{17}\ \mathrm{cm^{-3}})^2 * 0.2 * 45\times10^{-6}$ 1/s
$N_{FRC,n\text{-}type}([O_i]=1\times10^{17}\ \mathrm{cm^{-3}}) = ([O_i]/10^{17}\ \mathrm{cm^{-3}})^2 * 150\times10^{-6}$ 1/s

The projection in Fig. 5 was calculated for a fixed injection level of 10% of the net doping concentration, i.e. X (*p*-type) = 1/11 = Y (*n*-type).

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