

# Influence of illumination spectrum on dissociation kinetic of iron-boron pairs in silicon

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## 1 Introduction

Defects significantly impact semiconductor properties. Although minimizing device dimensions to nanometers shifts some focus from extensive to point defects, physical properties still rely heavily on the presence and distribution of these irregularities. Hence, many strategies for enhancing semiconductor structures, including radiation and temperature treatments or certain fabrication conditions, strive to decrease the defect concentration or neutralize its effects [1, 2, 3]. For instance, in the case of photovoltaic devices, we must understand and optimize the carrier properties tied to defects and impurities [1]. Such controlled alteration methods of the defective subsystem have been generalized under the term “defect engineering” and are extremely important from a practical standpoint.

Successful defect engineering hinges on an in-depth understanding of defect properties. Key factors are defect formation energy, transition energy levels, self-compensating effects, nonradiative recombination caused by defects, and the mechanism of reconstruction and diffusion [1]. Considering the extraordinary diversity of possible intrinsic and impurity defects, complete information on all of them is lacking even for silicon, which is the most studied semiconductor. Nevertheless, it must be noted that considerable data have been amassed on silicon, and have a solid understanding of some defects [4].

For instance, such defects are iron impurity, a common, detrimental, and often unavoidable contaminant in photovoltaic silicon [3, 5], and iron-boron pair. Specifically, iron atoms are known to be at the interstitial sites, and  $\text{Fe}_i^+$  are highly efficient recombination centers [6]. In p-type Si at room temperature, iron atoms are almost predominantly bound in complexes with dopants (B, Ga, Al, In). This defect demonstrates bistable behavior: the stable state is defined by the configuration in which the Fe occupies the first nearest tetrahedral interstitial site closest to the substituent atom, whereas, in the metastable configuration, Fe is at the second  $T_d$  interstitial site [7]. The energy levels associated with iron and its complexes, as well as the respective carriers capture cross-sections, are well-established [4, 8]. Among the acceptor-iron pairs, the complex FeB is the most thoroughly investigated, primarily due to the widespread use of Si:B in the fabrication of various devices, such as solar cells. However, it is worth mentioning that gallium is gaining increasing attention as an acceptor dopant whose incorporation, for instance, can help mitigate the light and elevated temperature-induced degradation [9].

The dynamics of FeB pairs are also examined. It's established that FeB pairs can be dissociated through illumination, minority carrier injection, and thermal treatment at 200 °C [10]. In the context of illumination, the dissociation rate  $R_d$  is influenced by the overall carrier generation rate  $G$  [11, 10, 12, 13]:

$$R_d = K \left( \frac{G}{N_{\text{FeB}}} \right)^2, \quad (1)$$

where  $N_{\text{FeB}}$  is the pair concentration,  $K$  is the constant of material. To achieve almost complete dissociation of the FeB pair, it is necessary for the illumination power to exceed 0.1 W cm<sup>-2</sup> [14]. The dissociation process of FeB pairs by electron capture unfolds in two stages [15, 10]: the initial stage involves

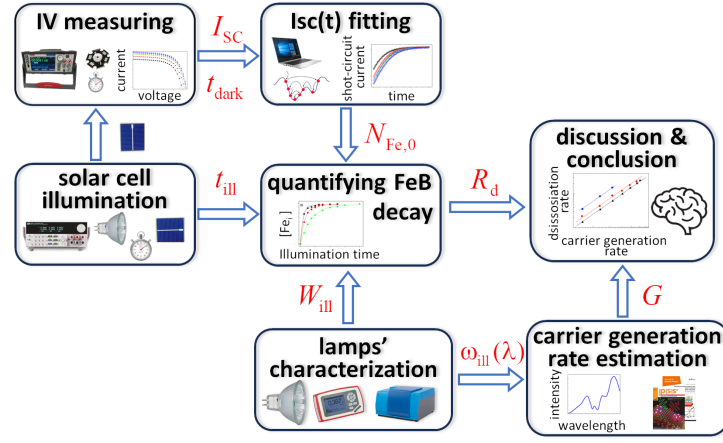


Figure 1: Investigation framework

the neutralization of Fe and the elimination of the Coulombic attraction between the pair components. The mechanism of the second stage is contentious; it may involve either the recharge of the iron ion or the recombination-enhanced defect reaction (REDR) triggered by electron-hole recombination. It should be noted that despite the extensive data on the properties of iron-related defects in silicon, intensive research persists. In particular, efforts focus on analyzing the impact of high-intensive illumination [16] or dopant compensation [17], alongside clarifying the second-stage mechanism of dissociation [5] or reassessing recombination parameters [18].

This study aims to investigate the effect of the light spectrum on the dissociation kinetics of FeB pairs in silicon. While pair dissociation is typically carried out using a halogen lamp [11, 5] or 904 nm laser [16, 10, 19], there is limited understanding of how the light source influences this process. By studying the impact of different illumination spectra on FeB dissociation, we aim to provide valuable insights for defect engineering and the efficient transformation of detrimental impurity iron atoms into a highly mobile interstitial state within the active region of a silicon device. Besides, such information, in our opinion, can help make the right choice between existing options for the second stage of pair decay.

Figure 1 illustrates the main stages of the research. Initially, the dissociation rate of FeB pairs under illumination of the solar cell with different integral intensities was determined. For this purpose, the dependence of the quantity of interstitial iron atoms formed over time under intense illumination was measured. The concentrations of formed pairs were measured by investigating the kinetics of short-circuit current. Three light sources from different manufacturers were used (further details are described in Section 1). To determine the carrier generation rate, spectra of sample illumination using various light sources were measured, taking into account the effects of light reflection, absorption by free carriers, and effective absorption depths. The obtained results led to the conclusion that the efficiency of light-induced dissociation increases with decreasing photon wavelength.

In Figure 1, the main stages of the research are illustrated. First step was determination of the dissociation rate of FeB pairs under illumination with different integral intensities. Three light sources from different manufacturers were used (further details are described in Section 4). To measure number of interstitial iron atoms formed over fixed time under strong illumination the kinetics of short-circuit current was used. The result is presented in Section 2.1. Section 2.2 deals with estimating the carrier generation rate using spectra of sample illumination and considering the effects of light reflection, absorption by free carriers, and effective absorption depths. The obtained results showed that the efficiency of light-induced dissociation increases with decreasing photon wavelength — see Section 2.3. Finally, we conclude this paper in Section 3.

## 2 Results and Discussion

### 2.1 Dissociation rate determination

The equilibrium between free  $\text{Fe}_i$  and  $\text{Fe}_i\text{B}_s$  is known to be determined by the following equations [20, 5, 10]



where  $R_a$  is the association rate. As a result, the concentration of interstitial iron atoms  $N_{\text{Fe}_i}$  depending on illumination time  $t_{\text{ill}}$  during light-induced dissociation can be described as follows [11, 12, 21]

$$N_{\text{Fe}_i}(t_{\text{ill}}) = \left( N_{\text{Fe,eq}} - N_{\text{Fe,tot}} \frac{R_d}{R_d + R_a} \right) \exp[-(R_d + R_a)t_{\text{ill}}] + N_{\text{Fe,tot}} \frac{R_d}{R_d + R_a}, \quad (3)$$

where  $N_{\text{Fe,tot}}$  is the total concentration of the impurity iron,  $N_{\text{Fe,eq}}$  represents the concentration of unpaired interstitial iron atoms in the equilibrium state (in darkness,  $N_{\text{Fe,eq}} = N_{\text{Fe}_i}(t_{\text{ill}} = 0)$ ). It's important to highlight that  $N_{\text{Fe,eq}}$  is significantly influenced by temperature and the Fermi level location [20]. Specifically, in the case of p-type Si with a hole concentration of  $1.36 \times 10^{15} \text{ cm}^{-3}$  (which corresponds to the base of the structure under investigation), at a temperature of  $T = 300 \text{ K}$ ,  $N_{\text{Fe,eq}}$  constitutes merely about 1% of  $N_{\text{Fe,tot}}$ , rendering it negligible for practical considerations. However, when the temperature rises to 340 K, the proportion of  $N_{\text{Fe,eq}}$  increases to approximately 14.5%.

After the cessation of illumination, only the process of association occurs, and the time dependence of  $\text{Fe}_i$  concentration can be expressed as follows [20, 22]:

$$N_{\text{Fe}_i}(t_{\text{dark}}) = (N_{\text{Fe},0} - N_{\text{Fe,eq}}) \times \exp(-R_a t_{\text{dark}}) + N_{\text{Fe,eq}}, \quad (4)$$

where  $t_{\text{dark}}$  is the time after strong illumination stopping,  $N_{\text{Fe},0}$  is the concentration of interstitial iron atoms formed after illumination,  $N_{\text{Fe},0} = N_{\text{Fe}_i}(t_{\text{dark}} = 0) = N_{\text{Fe}_i}(t_{\text{ill}})$ .

The study examined the dependence of  $N_{\text{Fe},0}$  in silicon solar cells on illumination time  $t_{\text{ill}}$  using different illumination intensities  $W_{\text{ill}}$  (200 – 750 mW) and light sources (three halogen lamps, labeled as Orion, Osram, and GE, and described in detail in Section 4). The experiments were conducted at a temperature of 340 K. The values of  $N_{\text{Fe},0}$  were determined using a methodology [23, 21] based on the study of the kinetics of short-circuit current  $I_{SC}$  under low-intensity monochromatic illumination. Specifically, after strong illumination with a duration of  $t_{\text{ill}}$ , the current-voltage characteristic (IV) of the solar cell was measured every 21 seconds over a time  $t_{\text{dark}}$  interval of approximately 3000 seconds.

Figure 2a shows some typical IV curves. It can be seen that upon cessation of illumination, there exists a gradual augmentation in both the short-circuit current and the open-circuit voltage. This phenomenon is indicative of a decrease in the recombination activity of the defective subsystem, which is a result of the transition of interstitial iron to a bound state with an acceptor. Moreover, at the end of the measurement interval, the minute changes in the IV curves denote that the selected interval of 50 minutes is sufficient to complete the association.

Figure 2b illustrates the dependencies  $I_{SC}(t_{\text{dark}})$  after illumination with different intensities. As previously shown [21], the magnitude of the change in  $I_{SC}$  after the dark recovery period inherently correlates with the concentration of  $\text{Fe}_i$  formed as a result of light-induced dissociation of  $\text{FeB}$  pairs. From examining the presented data, it is evident that escalating  $W_{\text{ill}}$  leads to an augmentation in the dissociation efficiency. Concurrently, the recovery time remains insensitive to the illumination parameters, which conforms to expectations, given that the latter is determined by  $R_a$  — see Equation (4).

It should be noted that besides  $N_{\text{Fe},0}$  values, the methodology used allows for the estimation of the energy of  $\text{Fe}_i$  migration  $E_m$  and bulk lifetime  $\tau_{\text{other}}$ , which arises from recombination channels other than Fe-related defects and intrinsic recombination. The obtained value  $E_m = (0.650 \pm 0.005) \text{ eV}$  coincides with that wellknown value [12, 24, 25]. This coincidence confirms that the investigated processes are indeed associated with rebuilding, as described by Equation (2). The value of  $E_m$  allows for the estimation

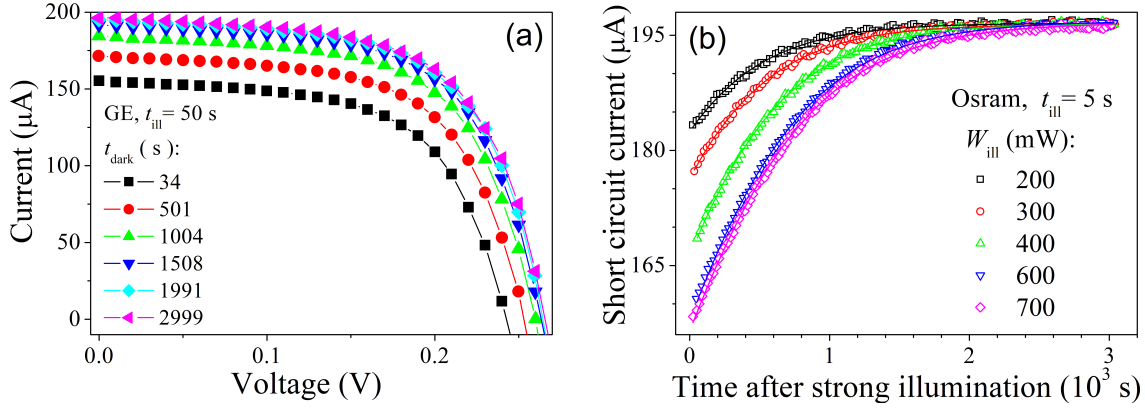


Figure 2: Typical current-voltage characteristics measured under low-intensive (LED) illumination across different periods following exposure to strong light (halogen lamp) (panel a) and short circuit current plotted as a function of the time after high-intensive illumination (panel b). The marks are the experimental results and the lines on panel b are the curves fitted according to [23, 21]. Light sources: GE (a), Osram (b).  $t_{\text{ill}}$ , s: 50 (a), 5(b).  $W_{\text{ill}} = 400$  mW (a).  $T = 340$  K.

of the recombination rate [10, 12, 24]:

$$R_a^{-1} = 5.7 \times 10^5 \frac{\text{s}}{\text{K cm}^3} \times \frac{T}{p} \exp\left(\frac{E_m}{kT}\right). \quad (5)$$

Thus, in our case,  $R_a = (1.68 \pm 0.03) \times 10^{-3} \text{ s}^{-1}$ .

Regarding the value of  $\tau_{\text{other}}$ , it was found to significantly exceed the lifetime associated with Shockley-Read-Hall (SRH) recombination on Fe-related defects. Notably, according Möller *et al.* [10], such a condition is essential for the accurate determination of the constant  $K$ , which is included in Equation (1). The dependencies of the concentration of interstitial atoms on illumination time are shown in Figure 3. From the data, it's evident that the pair dissociation rate is significantly influenced by the illumination intensity. This effect is consistent across all utilized light sources. Nonetheless, the  $W_{\text{ill}}$  value is not the exclusive determining factor for the pair dissociation rate, as demonstrated in Figure 3d. For instance, when using the GE source, pair dissociation occurs most efficiently. With Osram, the process unfolds more slowly, and illumination with Orion, under otherwise identical conditions, proves to be the least effective in terms of altering the state of FeB pairs.

The  $N_{\text{Fe},0}(t_{\text{ill}})$  dependencies were fitted using the equation

$$N_{\text{Fe},0}(t_{\text{ill}}) = A \exp(-t_{\text{ill}}/\tau_{\text{dis}}) + N_{\text{Fe,fit}}, \quad (6)$$

where  $\tau_{\text{dis}}$  is the characteristic dissociation time, and  $N_{\text{Fe,fit}}$  concentration of dissociated pairs at saturation. The fitting results, shown in Figure 3 as lines and listed in the Table 1, include the coefficients of determination  $R^2$ . The high values of  $R^2$  (greater than 0.99) confirm the suitability of the chosen approximation formula.

The comparison of Equations (3) and (6) reveals a relationship between the fitting parameters and defect characteristics, specifically:

$$\tau_{\text{dis}}^{-1} = R_a + R_d, \quad (7)$$

$$N_{\text{Fe,fit}} = N_{\text{Fe,tot}} \frac{R_d}{R_d + R_a}. \quad (8)$$

Table 1 suggests that regardless of the light source used and  $W_{\text{ill}}$ , the observed total concentration of impurity iron atom is consistently  $N_{\text{Fe,tot}} = (8.7 \pm 0.1) \times 10^{12} \text{ cm}^{-3}$ . This stability supports the accuracy of the analysis. However, the FeB dissociation rate may vary significantly for the same intensity value depending on the light source used.

According to Wijaranakula [20], at the specified value of  $N_{\text{Fe,tot}}$ , the equilibrium (in darkness) concentrations of interstitial iron atoms  $N_{\text{Fe,eq}}$  and FeB pairs  $N_{\text{FeB}}$  at  $T = 340$  K are  $1.3 \times 10^{12} \text{ cm}^{-3}$  and

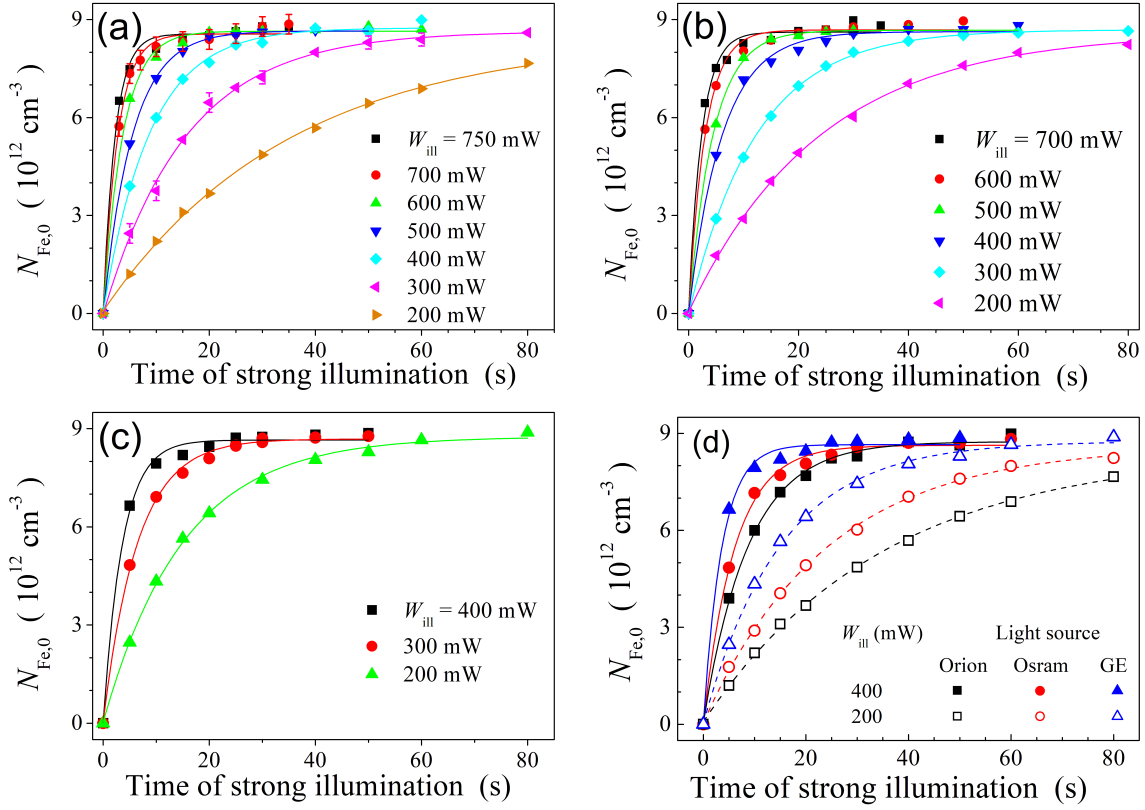


Figure 3: The relationships between the concentration of FeB pairs following intense illuminations of varying intensities and the illumination duration. Light source: Orion (a), Osram (b), GE (c). Panel d highlights variations in the dissociation of pairs induced by different light sources. The marks are the experimental results, the lines are the fitted curves using Equation (6).  $T = 340$  K.

Table 1: Fitting results of experimental dependencies  $N_{\text{Fe},0}(t_{\text{ill}})$  using Equation (6) and defect parameter estimation using Equations (7-8).

$W_{\text{ill}}$ [mW]	Light source	fitting parameters			defect parameters	
		$\tau_{\text{dis}}$ [s]	$N_{\text{Fe,fit}}$ [ $10^{12} \text{ cm}^{-3}$ ]	$R^2$	$R_d$ [ $10^{-3} \text{ s}^{-1}$ ]	$N_{\text{Fe,tot}}$ [ $10^{12} \text{ cm}^{-3}$ ]
750	Orion	$2.2 \pm 0.2$	$8.6 \pm 0.1$	0.993	450	8.6
700	Orion	$2.7 \pm 0.2$	$8.7 \pm 0.1$	0.995	370	8.7
	Osram	$2.4 \pm 0.2$	$8.6 \pm 0.1$	0.992	410	8.6
600	Orion	$3.7 \pm 0.2$	$8.65 \pm 0.06$	0.998	270	8.7
	Osram	$3.0 \pm 0.2$	$8.69 \pm 0.08$	0.995	330	8.7
500	Orion	$5.5 \pm 0.2$	$8.65 \pm 0.04$	0.999	180	8.7
	Osram	$4.5 \pm 0.1$	$8.7 \pm 0.1$	0.998	220	8.8
400	Orion	$8.8 \pm 0.3$	$8.74 \pm 0.06$	0.998	110	8.8
	Osram	$6.1 \pm 0.2$	$8.63 \pm 0.08$	0.997	160	8.7
	GE	$3.6 \pm 0.3$	$8.7 \pm 0.1$	0.996	280	8.7
300	Orion	$15.7 \pm 0.6$	$8.6 \pm 0.1$	0.998	62	8.8
	Osram	$12.4 \pm 0.1$	$8.69 \pm 0.02$	0.999	79	8.8
	GE	$6.5 \pm 0.2$	$8.69 \pm 0.05$	0.998	150	8.8
200	Orion	$35 \pm 3$	$8.5 \pm 0.3$	0.998	27	8.8
	Osram	$24 \pm 1$	$8.6 \pm 0.1$	0.999	40	8.9
	GE	$15.1 \pm 0.5$	$8.7 \pm 0.1$	0.999	65	8.8

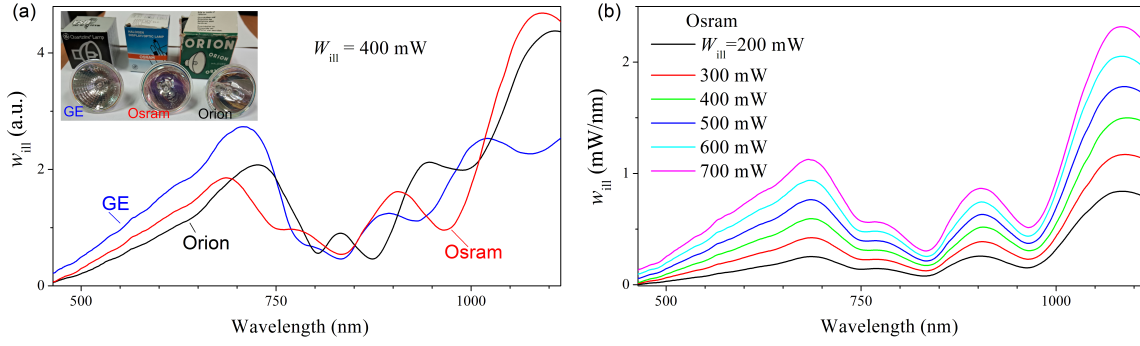


Figure 4: The spectra of sample illumination in the case of using different light sources with the same integral intensity  $W_{\text{ill}} = 400 \text{ mW}$  (panel a) and a single source (Osram) at various  $W_{\text{ill}}$  values (panel b). The inset shows photos of light sources.

$7.4 \times 10^{12} \text{ cm}^{-3}$ , respectively. The values of  $N_{\text{Fe,eq}}$  and  $N_{\text{FeB}}$  were used to estimate the minority carrier diffusion length  $L_n$  in the base of the used solar cell. It was assumed that the dominant recombination processes are SRH recombination at  $\text{Fe}_i$  and  $\text{FeB}$  and intrinsic recombination. The required electron mobility  $\mu_n$  value were taken from Klaassen [26], the capture cross sections and energy levels for  $\text{Fe}_i$  and  $\text{FeB}$  from Rougieux *et al.* [8], the coefficients of band-to-band radiation recombination and Auger recombination from Niewelt *et al.* [27] and Black & Macdonald [28], respectively. The calculated value was found to be  $L_n = 80 \text{ }\mu\text{m}$ , which is remarkably close to the value of  $86 \text{ }\mu\text{m}$  obtained from the study of temperature dependencies of short-circuit current — see Supplementary materials.

## 2.2 Carrier generation rate estimation

The dissociation rate of  $\text{FeB}$  pairs during light-induced decay is well known to be dependent on the carrier generation rate — see Equation (1). Our subsequent objective involved determining the values of  $G$  for various light sources. The measured dependencies of spectral intensity  $w_{\text{ill}}$  of illumination incident on the sample under diverse conditions, are shown in Figure 4. It is crucial to highlight that our focus is specifically on the light reaching the sample; hence, the spectrum is altered not only by the infrared transparency of the lamp reflector but also by absorption in the fiber utilized to transmit the light flux to the solar cell. Analogous modifications to the illumination spectra have been observed previously [29]. Figure 4a displays discrepancies in the illumination spectra obtained from different light sources, attributed to variations in the operational temperatures of the halogen lamps and differences in reflectors (photos of the lamps are in the inset of Figure 4a). It is important to note that the upper limit of the spectra in Fig 4 is limited by the silicon bandgap, which, according to Passler [30], corresponds to 1.11 eV at 340 K. Furthermore, Figure 4b demonstrates the change in the Osram spectrum with integral intensity increasing. Notably, in addition to the expected increase in the curve's area, a minor spectrum shift towards shorter wavelengths is observed. These behaviour is typical for all used light sources.

Carrier generation rate was estimated as follows:

$$G = \int g(\lambda) d\lambda, \quad (9)$$

where spectral carrier generation rate  $g$

$$g = \frac{w_{\text{ill}} \lambda}{hc} \frac{(1 - R) A_{\text{bb}}}{S d_{\text{eff}}}, \quad (10)$$

where  $n_{\text{ph}} = \frac{w_{\text{ill}} \lambda}{hc}$  is the spectral photon flux,  $R$  is the coefficient of reflection,  $A_{\text{bb}}$  is the fraction of the band-to-band transitions,  $S$  is illuminated square of sample,  $d_{\text{eff}}$  is the effective width of carrier generation.

In calculating the value of  $R$ , we employed an approach [31], which accounted for the presence of antireflective and passivating layers on the front surface of the sample, as well as the effects of multiple reflections. The resulting spectral dependence of  $R$  is shown in Figure S3 of the Supplementary materials.

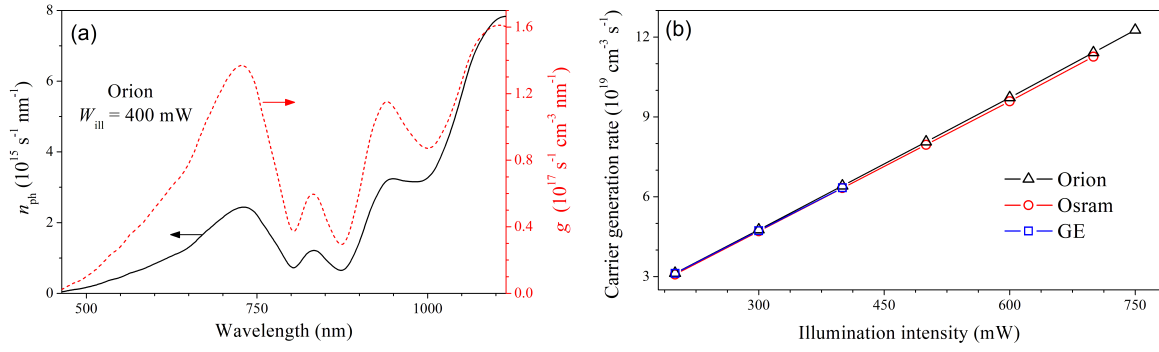


Figure 5: (a) Photon flux spectral density (left axis, solid line) and carrier generate rate spectral density (right axis, dashed line). Orion light source,  $W_{ill} = 400$  mW. (b) Dependencies of carrier generation rate on illumination intensity for different light sources.

The expression for the e-h pair generating fraction of the Lambertian absorptance in a solar cell can be written as [32]:

$$A_{bb}(\lambda) = \frac{\alpha_{bb}}{\alpha_{bb} + \alpha_{fca}} \frac{(1 - T_r)(1 + T_r)n_r^2}{n_r^2 - (n_r^2 - 1)T_r^2}, \quad (11)$$

with

$$\begin{aligned} T_r &= (1 - x) \exp(-x) + x^2 E_1(x), \\ x &= (\alpha_{bb} + \alpha_{fca})d, \\ E_1(x) &= \int_x^\infty t^{-1} \exp(-t) dt, \end{aligned}$$

where  $\alpha_{bb}$  is the absorption coefficient due to e-h pair generation by band-to-band transitions;  $\alpha_{fca}$  is the absorption coefficient due to free carrier absorption;  $n_r$  is the refractive index;  $d$  is the width of the device.

$$d_{eff}(\lambda) = \frac{\left( \int_0^d \Delta n dx \right)^2}{\int_0^d \Delta n^2 dx}, \quad (12)$$

where  $\Delta n$  is the increase in minority carrier density due to illumination

$$\Delta n(x) = \frac{\alpha_{bb} n_{ph} L_n^2 q}{(\alpha_{bb}^2 L_n^2 - 1) k T \mu_n} \left[ \exp\left(-\frac{x}{L_n}\right) - \exp(-\alpha_{bb} x) \right]. \quad (13)$$

$d_{eff}$  [33]

$A_{bb}$  [32]

$\alpha_{bb}$ , refractive index  $n$  [34]

$\alpha_{fca}$  [35]

### 2.3 Effect of illumination spectrum on FeB pair decay

AIPAdv 3 082124 [16]

Effect of Dopant Compensation on the Behavior of Dissolved Iron and Iron-Boron Related Complexes in Silicon InterJPhotoener 2015 154574 [17]

JApplPhys 116 024503 [10]

PhysStatSolA 216 1900253 [12]

PhysStatSolRRL 15 2100520 [5]

PR ApplPhysLett 85 p5227 5229 [11]

PR JApplPhys 98 083509 [36]



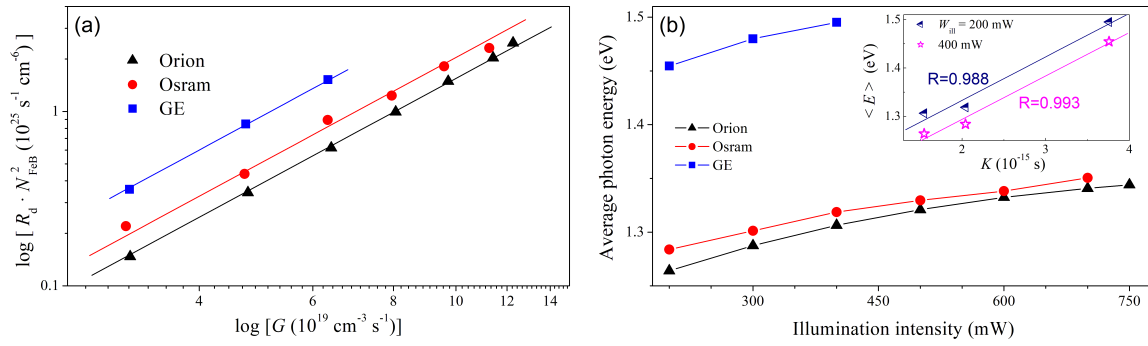


Figure 6: (a) FeB pair dissociation rate plotted as  $R_d \cdot N_{\text{FeB}}^2$  over the light induced generation rate. The solid lines show the quadratic dependence according to Equation (1). (b) Dependencies of average photon energy on illumination intensity for different light sources. The inset shows prefactor  $K$  vs average photon energy for the different light sources and illumination intensities. The lines are linear fitted curves. Coefficients of correlation are shown as well.

SolStPhenom 242 p230 [19]

JApplPhys 95 p1021 [14]

PR PhysStSolRRL 6 p1 [13]

2%

$3.8 \times 10^{-15} \text{ s}$  GE  $2.0 \times 10^{-15} \text{ s}$  Osram  $1.5 \times 10^{-15} \text{ s}$  Orion

$$\langle E \rangle = \frac{\int \frac{hc}{\lambda} n_{\text{ph}}(\lambda) d\lambda}{\int n_{\text{ph}}(\lambda) d\lambda}. \quad (14)$$

### 3 Conclusion

As explained in Refs.[6,21], the local vibrational energy released from a recombination event at the defect center can promote defect reactions such as dissociation and diffusion.

### 4 Experimental Section

The temperature was maintained constant using a PID (proportional-integral-derivative controller) algorithm which is implemented in the software which serves the system.

Therefore, the temperature of the wafers was regulated and controlled through a thermoelectric cooling system based on the Peltier effect

*First part of experimental section:*

*Second part of experimental section:*

#### Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest** The authors declare no conflict of interest.

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