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ScienceDirect

Energy Procedia 8 (2011) 64–70

Energy Procedia

SiliconPV: 17-20 April 2011, Freiburg, Germany

Spatially resolved carrier lifetime calibrated via quasi-steady-state photoluminescence

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Abstract

The estimation of solar cell efficiency from minority carrier lifetime measurements requires precise and robust lifetime techniques. For multicrystalline wafers and solar cells this brings about the necessity of adequately averaged spatially resolved lifetime measurements. Materials such as e.g. multicrystalline upgraded metallurgical grade silicon frequently feature relatively low lifetimes, high trap densities, and several material parameters (charge carrier mobilities and net dopant concentration) that are not straightforwardly predictable or measurable. As this may substantially compromise conventional lifetime measurements, a lifetime technique which is unaffected by such restrictions is of general interest. We present a lifetime imaging technique based on a calibration of photoluminescence images via quasi-steady-state photoluminescence (QSSPL), which requires no a priori information about material parameters such as carrier mobilities and net dopant concentration. It is virtually unaffected by effects related to reabsorption of luminescence, by optical effects related to a sample's surface morphology, and by trapping. Injection dependence of lifetime is properly taken into account. We further accomplished a substantial upgrade of the hitherto existing sensitivity limit of QSSPL in terms of lifetime, to where average lifetimes down to one microsecond are now reliably measurable – yielding a sensitivity of spatially resolved lifetime well below one microsecond.

Keywords: *Carrier Lifetime, Photoluminescence*

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 doi:10.1016/j.egypro.2011.06.103

1. Introduction

This contribution compiles the findings of our recent publications concerned with the spatially resolved carrier lifetime calibration of silicon wafers and solar cells via QSSPL [1, 2] and with reliable QSSPL lifetime measurements down to the microsecond range [3]. For an elaborate account and a more detailed reference list the reader is referred to the respective journal articles.

Two roads for photovoltaics to enhance competitiveness are to be pursued: the first aims at boosting solar cell efficiencies, and the second attempts to drive down material cost while maintaining the electronic material quality. Low cost silicon feedstock – particularly multicrystalline upgraded metallurgical silicon – is very likely to play a significant role in silicon photovoltaics in the future. The optimization of solar cell concepts based on low cost silicon feedstock requires a high level of accuracy in carrier lifetime metrology. This is because the smaller minority carrier lifetime of silicon solar cell precursors is, the higher is the relative error when predicting solar cell current and voltage from inaccurate lifetime measurements. Concerning lifetime measurements on low cost silicon feedstock, there is a conflict between the requirements in terms of accuracy on the one hand, and the established state of the art of lifetime metrology on the other hand. This conflict stems mainly from the fact that well established carrier lifetime techniques require a priori information about carrier mobilities and/or dopant concentration. QSSPL – being a time dependent photoluminescence based lifetime technique – provides a solution here. The following sources of inaccuracy leave QSSPL virtually unaffected:

- Carrier mobility affects any lifetime technique sensitive to the product of mobility and lifetime – particularly the family of non-transient photoconductance measurement techniques.
- Trapping affects any lifetime technique sensitive to the sum of free minority and majority carriers rather than to their product (the latter is the case for luminescence measurements).
- Effects related to a sample's surface morphology, to reabsorption of band to band luminescence, and to free carrier absorption: these effects relate to any steady-state optical lifetime technique which is directly sensitive to signal magnitude.
- Dopant concentration is required for many photoconductance / luminescence based lifetime techniques.

QSSPL yields injection dependent minority carrier lifetime, it can be applied to both unmetallized and metallized cell precursors as well as finished solar cells. In spite of the stated advantages, the wide dissemination of QSSPL in photovoltaics has been hampered so far by a relatively poor sensitivity limit in terms of carrier lifetime. A systematic analysis of the sensitivity limiting mechanisms led to a substantial upgrade of sensitivity, to where reliable lifetime results are obtained down to carrier lifetimes in the range of a microsecond.

Here, the carrier lifetime calibration of (spatially resolved) photoluminescence images of silicon wafers and solar cells via QSSPL shall be sketched. Since the lateral resolution of a QSSPL measurement is on the order or larger than areas of homogeneous material quality in multicrystalline silicon, it has to be addressed how each element of a well defined active sample area contributes to the measured QSSPL signal and finally to the measured QSSPL lifetime. Thus, this paper gives account of an appropriate averaging procedure to determine spatially resolved carrier lifetimes from photoluminescence images (at a resolution of ~$100\times100\mu m^2$) via QSSPL. The proposed method properly accounts for injection dependence of lifetime, and it reliably yields local lifetimes well below one microsecond.

2. Theory

Photoluminescence imaging via a CCD camera provides a spatially resolved steady-state distribution of luminescence intensities of an optically excited silicon wafer. The method sketched in the following aims at an interpretation of spatially resolved luminescence intensity in terms of minority carrier lifetime, which is accomplished here through a QSSPL calibration.

2.1. Fundamentals of QSSPL

A QSSPL measurement is distinguished by the simultaneous recording of both the time modulated (preferably sinusoidal) optical irradiation intensity upon a well defined specimen area and the corresponding photoluminescence intensity as a function of time. Our perception of the quasi-steady-state condition is that the timescale of the excitation modulation (modulation period T) has to be much greater than effective carrier lifetime τ .

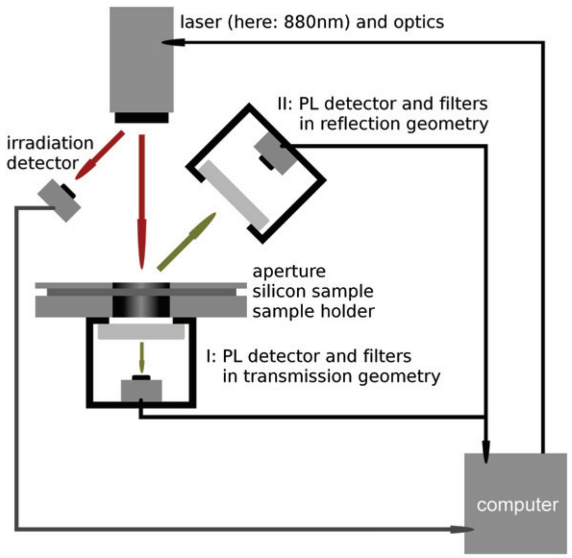

Figure 1. Depiction of the experimental setup of QSSPL. A time modulated laser irradiates a silicon wafer or solar cell. The irradiation intensity is recorded as a function of time in an irradiation detector, while the photoluminescence intensity is recorded in another – encapsulated – detector which can be mounted either beneath the sample (I: in the case of unmetallized cell precursors) or toward the irradiated sample side (II: for metallized wafers or solar cells). Both the relative irradiation and photoluminescence intensity are read into a computer and evaluated in terms of injection dependent carrier lifetime.

2.2. Injection dependent effective carrier lifetime from QSSPL

With the time dependent generation rate $G(t)$, the excess carrier density $\Delta n(t)$ obeys the continuity equation

$$\frac{\partial \Delta n(t)}{\partial t} + \frac{\Delta n(t)}{\tau(\Delta n(t))} = G(t), \qquad (1)$$

while $\tau(\Delta n(t))$ is the injection dependent effective minority carrier lifetime. As in the steady-state case, the measured QSSPL intensity $\Phi(t)$ reads

$$\Phi(t) = A\Delta n(t)(\Delta n(t) + N), \tag{2}$$

while N denotes net dopant concentration, and the factor A incorporates the coefficient of radiative recombination as well as all relevant measurement setup and optical sample properties. Under quasi-steady-state conditions, it can be shown with Eqs. (1) and (2) that effective minority carrier lifetime can be extracted from the time shift between maxima of irradiation and luminescence intensities [4]. An equivalent way to determine effective minority carrier lifetime from QSSPL (albeit applicable only under low injection conditions without knowledge of N) is known as the self-consistent lifetime calibration by Trupke et al. [5]. Figure 2 depicts both a QSSPL measurement and its interpretation in terms of lifetime.

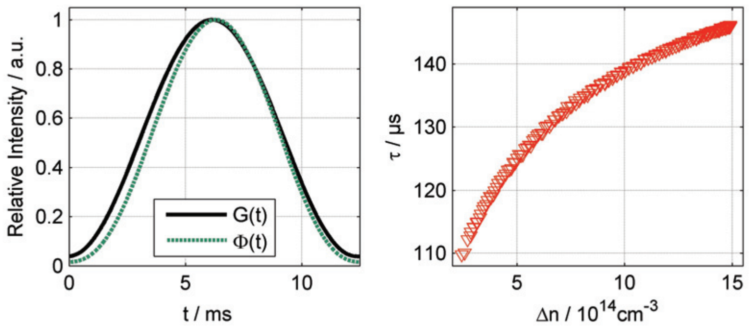

Figure 2. (left) Example of a QSSPL measurement on a silicon wafer (modulation frequency: 80Hz); (right) Interpretation of the QSSPL measurement at the left in terms of injection dependent minority carrier lifetime (self-consistent evaluation).

2.3. QSSPL lifetime calibration of photoluminescence images

The spatially resolved intensity φ_i of pixel i of a (steady-state) photoluminescence image relates to local minority carrier lifetime τ_i as

$$\varphi_i = aG\tau_i(G\tau_i + N). \tag{3}$$

Here, G is the steady-state optical carrier generation rate per volume, and the calibration factor a again accounts for the coefficient of radiative recombination and any relevant setup and optical sample properties. If a and N were known, τ_i could be directly extracted from a photoluminescence image φ_i, but this is generally not the case. So a has to be determined by inserting τ_i into the following equation as the positive solution of Eq. (3) for each of the n pixels, which contribute to a QSSPL measurement:

$$\tau_{QSSPL} = \frac{\sum_{i=1}^{n} \tau_i S_i \left(2G^2\tau_i^2 + G\tau_i N\right)}{\sum_{i=1}^{n} S_i \left(2G^2\tau_i^2 + G\tau_i N\right)}. \tag{4}$$

This relation is derived in full detail in [1]. S_i denotes the relative sensitivity of the QSSPL detector with respect to the sample position corresponding to pixel i .

Under low injection conditions, the proportionality of carrier lifetime and photoluminescence intensity enables a significant simplification. The local photoluminescence intensity φ_{QSSPL} to be assigned to the measured QSSPL carrier lifetime $\tau_{QSSPL}(G)$ then reads

$$\varphi_{QSSPL}\left(\tau_{QSSPL}(G), \Delta n << N\right) = \frac{\sum_{i=1}^{n} S_i \varphi_i^2}{\sum_{i=1}^{n} S_i \varphi_i}, \tag{5}$$

which corresponds to an intensity weighted average over local photoluminescence intensities. In this case, local carrier lifetime is

$$\tau_i\left(\Delta n << N\right) = \frac{\tau_{QSSPL}(G)}{\varphi_{QSSPL}} \varphi_i . \tag{6}$$

3. Tackling the Microsecond Range with QSSPL

Several measures have to be taken in order to ensure a sufficient sensitivity limit of QSSPL in terms of carrier lifetime. As QSSPL is most sensitive to the phase shift of two modulated light intensities with respect to each other, particular attention must be paid to any phase changes throughout acquisition and processing of the measured signals.

3.1. Requirements for microsecond lifetime measurements

- Sufficient filtering of laser irradiation in photoluminescence detector: a combination of (radiation free) dielectric optical long pass filters and (radiation free) absorption long pass filters is deemed mandatory.
- Sufficient suppression or filtering of radiation within the relevant band to band luminescence spectrum, either generated directly in the light source, or originating from optical activity along the light path (fibers and optics), must be ensured.
- The phase delays imposed upon the photocurrents during the amplification process, either through RC interaction between diode capacitance and resistive elements of the amplifier, or caused by the finite response time of the operational amplifiers in use, must be considered.
- Consideration of an additional phase delay imposed upon the photocurrent of the photoluminescence detector when measuring with silicon photodiodes: due to the different spectra of silicon band to band luminescence and laser irradiation, this delay amounted to almost 30µs in our setup (elaborated in [3]).

3.2. Sensitivity evaluation – state of the art

In [3], we showed that our experimental setup at Fraunhofer ISE features sufficient filter properties in order to guarantee reliable carrier lifetime measurements down to the range of 100ns (given net dopant concentrations equivalent to <3Ωcm). When using silicon photodiodes, the uncertainty of lifetime due to phase delays related to the third and fourth items in the previous section amounts to ±500ns. With the use of InGaAs photodiodes, which are not exposed to a phase delay due to different light spectra of luminescence and laser irradiation, this uncertainty can be substantially reduced to ±200ns, which we deem a sufficient precision for a lifetime measurement of one microsecond.

4. Experimental Results

Spatially resolved carrier lifetime measurements were performed on both multicrystalline silicon wafers (plasma cleaned and upgraded metallurgical grade) and solar cells. They were carried out at injection conditions equivalent to solar irradiance (880nm photon current density of $2.55 \cdot 10^{17}$cm^{2}s^{-1}).

4.1. Results on unmetallized cell precursors

Representative samples of multicrystalline wafer classes – comprising plasma cleaned and compensated upgraded metallurgical grade wafers – were investigated. Samples are phosphorus gettered (emitter etched back) and feature a silicon nitride surface passivation. The obtained spatially resolved lifetimes of silicon wafers were compared to a spatially resolved carrier lifetime calibration via QSSPC, as also detailed in [1].

A slight systematic deviation of lifetime for plasma cleaned multicrystalline wafers is observed (cf. Table 1). Lifetime is slightly higher in the case of a QSSPL calibration. Within the specified uncertainties, this finding is in good agreement with very recent studies on carrier mobility. There, carrier mobility is expected to be ~5% lower in plasma cleaned multicrystalline silicon than in standard material due to crystal defects acting as mobility barriers. Thus, we expect QSSPC to slightly underestimate effective carrier lifetime here.

Table 1. Comparison of arithmetically averaged effective carrier lifetimes ‹τ› as obtained from a QSSPC and a QSSPL calibration. Representative plasma cleaned and upgraded metallurgical grade multicrystalline wafers were investigated. Each value shown here represents an average value of five calibrations carried out on different wafer spots respectively. The specified uncertainty refers to the variance of these five calibrations.

Class of multicrystalline wafer material		plasma cleaned	upgraded metallurgical grade
‹τ› self-consistent QSSPL	/ µs	117 ± 8	44 ± 3
‹τ› QSSPC	/ µs	102 ± 6	27 ± 2

The very pronounced systematic deviation of lifetime for the compensated upgraded metallurgical grade wafer (cf. comparison in Table 1 and lifetime map in Figure 3a) could be quantitatively explained as a QSSPC error by the combined mobility lowering effect of crystal defects and compensation [1].

4.2. Results on solar cells

Figure 3b depicts a map of effective carrier lifetimes of an upgraded metallurgical grade silicon solar cell. Solar cell lifetime results as obtained here were compared to measurements of open circuit voltage U_{oc} at identical injection conditions on the basis of the relation

$$U_{oc} = \frac{k_B T}{q} \ln\left(\frac{1}{n_i^2} \left(\Delta n + \frac{n_i^2}{N} \right) \left(\Delta n + N \right) \right). \tag{7}$$

Net dopant concentration was determined via FTIR. This method is unaffected by the above addressed carrier mobility related issues. Further details are to be found in [2]. The average value of the estimated open circuit voltage according to Eq. (7) is $U_{oc} = 614 \pm 2\text{mV}$, which is well in accordance with the average value of measured open circuit voltage of $U_{oc} = 613.7 \pm 0.4\text{mV}$.

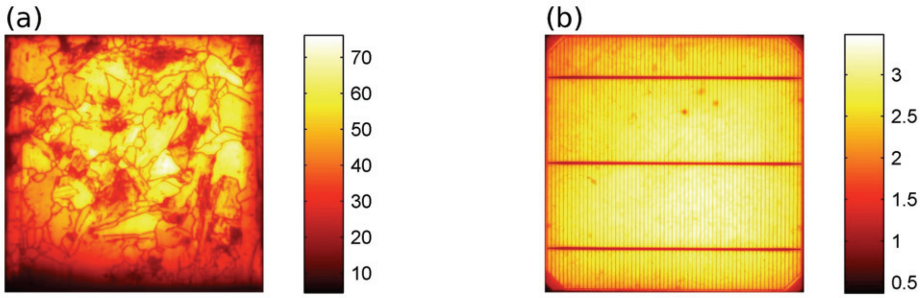

Figure 3. (a) Effective carrier lifetime image of a multicrystalline upgraded metallurgical grade silicon wafer as calibrated via QSSPL; (b) Effective carrier lifetime image of a multicrystalline upgraded metallurgical grade silicon solar cell as calibrated via QSSPL. Feedstock from near to the brick bottom causes a relatively low effective lifetime of about 2.7µs.

5. Conclusion

With the lifetime technique presented herein, the scope of quantitative carrier lifetime measurements based on quasi-steady-state photoluminescence is expanded to silicon solar cells and metallized or unmetallized solar cell precursors of an arbitrary lateral distribution of electronic material quality. The necessity and feasibility of a respective calibration procedure was experimentally demonstrated. The advantages of the presented technique compared to conventionally used techniques – particularly for low cost silicon feedstock – were addressed: the presented technique is virtually unaffected by trapping, carrier mobility, dopant concentration, surface morphology, and light absorption effects. It properly accounts for injection dependence of lifetime. It enables reliable lifetime measurements down to an average value of one microsecond, while the minimum of detectable spatially resolved lifetime is even an order of magnitude lower. Effective lifetime results of solar cells are very well compatible with measurements of open circuit voltage. Facing these favorable properties, the method proposed here seems suitable for quality control of practically any multicrystalline silicon material.

Acknowledgement

This work was funded by the Fraunhofer-Gesellschaft within the project Silicon BEACON.

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