



# Ph.D. Thesis

# Development of recombination lifetime measurement methods for photovoltaic applications

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> Budapest 2025

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## Acknowledgement

I would like to express my sincere gratitude to Professor Ferenc Simon, my supervisor, for his guidance throughout my research journey. His encouragement to adopt a rigorous scientific approach in my developmental work and his extensive insights into microwave systems have been crucial to my academic growth. His scientific vision and dedication have been a constant source of inspiration and motivation.

I am deeply indebted to my industrial consultant, Ferenc Korsós, for coordinating the research and development related to my thesis at Semilab. I am particularly grateful for the time he generously spent sharing his exceptional knowledge of recombination processes and the broader field of solar cell physics.

My sincere appreciation goes to Semilab Semiconductor Physics Laboratory Co. Ltd. for their support through the Cooperative Doctoral Fellowship. I am especially indebted to Dr. Tibor Pavelka, the CEO of Semilab, for creating the opportunity to conduct my research and for providing me with enlightening advice. I would like to extend my thanks to my colleagues at Semilab who directly contributed to my research. Foremost, I am grateful to Péter Tüttő for illuminating development goals from different perspectives with his comprehensive vision during scientific discussions, and for his significant contributions to sensor development. I extend my thanks to Gábor Paráda and Attila Tóth for their technical expertise and practical advices. I am grateful to Kinga Szőke and Gergely Havasi, who contributed to the experiments. My gratitude goes to Gyula Kómár and Dr. Krisztián Kis-Szabó for developing the measurement control systems. I am indebted to Enikő Balogh-Kis and Dr. Ferenc Ujhelyi for designing the homogenizing optics, and to Martin Kovács for his work on COMSOL simulations and result interpretation. I would like to thank Dr. Csaba Jobbágy for his support in chemical processes, particularly wafer etching and iodine-ethanol passivation.

I would also like to express my gratitude to Lingo Tang and Shaoyong Fu from Semilab China for their enthusiastic support in collecting customized, state-of-the-art samples from leading solar industry manufacturers.

I would like to express my sincere gratitude to Professor Sébastien Dubois and his team at CEA INES for providing high-quality silicon wafers.

I am very grateful to the former and current colleagues and students at BME, particularly Dr. András Bojtor, Dr. Sándor Kollarics, and Gábor Csősz, for their support in my research and for facilitating my integration into the academic group.

A special note of thanks goes to my fiancée, Virág, for her extraordinary patience and unwavering support during the intense months of dissertation writing.

Finally, I am profoundly grateful to my family, who prioritized supporting my studies above all else. Their unwavering belief in me and unconditional love have been the bedrock of my journey. I will be forever thankful for their support and encouragement. Project no. C1010858 has been implemented with the support provided by the Ministry of Culture and Innovation of Hungary from the National Research, Development and Innovation Fund, financed under the KDP-2020 funding scheme.

This thesis was supported by the K137852, 149457, 2022-2.1.1-NL-2022-00004, TKP2021-NVA-02 and TKP2021- EGA-02 grants of the NRDIF.

# Chapter 1 Introduction

Silicon has long been the cornerstone of modern electronics and remains essential in the manufacturing of integrated circuits (ICs) and solar cells (SCs). Its prominence stems from its favorable band gap, the uniquely low defect density of its native oxide layer that can be grown on the surface, and its high availability. Over the past seven decades, silicon has been the most extensively studied and best-understood crystalline material. Consequently, silicon is integral to the performance and scalability of a wide range of modern technologies, from microelectronics to renewable energy ecosystems.

The continuous development of solar cells over the past decades has resulted in a dynamic year-on-year increase in their contribution to global energy production. In 2023, solar energy became the leading source of newly installed energy capacity globally [1]. These achievements have been made possible by decades of research and development in materials science and semiconductor technology. The current record for photoelectric conversion efficiency in silicon-based single-junction solar cells is 27.3% [2], which approaches the recently updated theoretical efficiency limit [3, 4]. This limit considers the inevitable losses beyond the well-known Shockley-Queisser limit [5]. Therefore, any further enhancement necessitates an even more precise determination of the relevant physical parameters. Another significant trend is the shrinking gap between record efficiencies achieved in R&D laboratories and the efficiencies of solar cells manufactured on modern production lines [6]. Consequently, advanced and more accurate measurements are needed not only to achieve further efficiency records but also to reliably control manufacturing processes.

In solar cells, a dominant factor determining performance is the recombination rate of excess charge carriers. The bulk recombination rate within a silicon wafer, governed by the initial crystal quality and the concentration of defects and impurities, plays a central role in determining solar cell efficiency. Additionally, surface recombination, occurring at the p-n junction and near ohmic contacts, becomes a critical factor for solar cell devices as well. To further increase energy conversion efficiency, the reduction of recombination-related energy losses is a primary goal of research projects. For this purpose, the optimization of the Czochralski process for pulling monocrystalline silicon material [7–10] and the development of complex solar cell structures [11–15] are ongoing research topics.

While in solar cells, the recombination rate directly influences conversion efficiency, for ICs, the quality of the semiconductor-dielectric interface is of paramount importance. This interface governs the efficiency of charge transport and device operation. Impurities or localized defect-related energy states at this interface can severely degrade performance [16], making

careful control of the manufacturing process essential. Owing to the relationship between localized states at the interface and the corresponding recombination rate, these defects can also be detected using charge carrier recombination lifetime measurements [17].

Therefore, understanding and quantifying the recombination properties of silicon is essential for improving the performance of both ICs and SCs. A wide variety of measurement methods have been developed for the precise determination of charge carrier recombination lifetime. Compliance with modern manufacturing requirements demands the use of fast and non-contact technologies. Therefore, the most common measurement solutions currently rely on optical excitation and contactless detection sensitive to excess carriers. Commercial measurement systems utilize microwave reflectance [18, 19] or radio-frequency eddy current sensors [20, 21] for this purpose.

This work focuses on the development and applications of recombination lifetime measurement methods, primarily for the investigation of solar cell materials and structures, for both industrial quality control and research purposes. I believe that my work over the past four years has made a significant contribution to the photovoltaic research community and industry at three distinct levels. At the Budapest University of Technology and Economics (BME), my research group determined the recombination properties of novel photovoltaic materials [22] (in the perovskite family) using cryogenic carrier lifetime measurement systems, recording the recombination properties as a function of temperature [23, 24]. Beyond the development of the applied evaluation routines, I devised a method to verify the reliability of measurements. My work at Semilab Semiconductor Physics Laboratory Co. Ltd. (Semilab) has enabled the experimental determination of excess carrier lifetime and excess carrier mobility data for modern silicon-based solar cell structures with unprecedented accuracy. This was achieved through the development of a unique measurement method combining, for the first time, three independent carrier lifetime measurement principles. Finally, the reliable determination of the bulk lifetime of modern silicon material faces significant challenges. I established a new measurement and an iterative, simulation-based evaluation method to overcome these challenges. The method is already applied in commercial carrier lifetime measurement systems, providing rapid and precise information about possible contamination in an early production phase.

The structure of the thesis is as follows. In Chapter 2, I summarize the scientific background of the research. After presenting the operating principles of solar cells, I provide a compact overview of the most advanced cell structures. In this Chapter, recombination processes in semiconductors and the corresponding measurement methods are also discussed. In Chapter 3, I introduce the employed measurement techniques. Since my results also include the development of measurement methods, this Chapter summarizes previously existing methods and their implementation. I also provide details of the samples used to obtain my scientific results. Chapter 4 discusses my contribution to fundamental research on low-temperature carrier lifetime measurement techniques at the BME. In Chapter 5, I provide insights into my developments to gather accurate bulk lifetime data from thick silicon samples, even meeting the requirements of direct industrial applications. In the Chapter 6, I introduce the measurement setup developed for the characterization of silicon wafers used in modern solar cell structures. Finally, in Chapter 7, I summarize the results I have achieved along with the listing of the thesis points.

### Chapter 2

### **Theoretical background**

### 2.1 Solar cells

In this chapter, I summarize the theoretical background necessary to contextualize my results. First, a general overview of solar cell operation is presented, followed by an introduction to relevant modern solar cell structures. Then, I discuss charge carrier recombination processes in semiconductors and their general measurement methods. Finally, the importance of recombination lifetime measurements from a solar cell development perspective is highlighted.

#### 2.1.1 Principles of solar cell operation

#### The ideal solar cell

A solar cell operates by converting light energy into electrical energy through the photovoltaic effect. When the surface of a solar cell is radiated by photons, they are absorbed by a semiconductor material [25]. Photons with energy exceeding the band gap of the semiconductor material generate electron-hole pairs (EHPs) upon absorption. Lower-energy photons are not utilized, representing a primary source of theoretical losses. The excited electrons and holes rapidly lose energy, thermalizing to the bottom of the conduction band and the top of the valence band, respectively. The higher the energy of the absorbed photon, the greater the thermalization loss associated with this process. These losses are summarized in Figure 2.1a. It should be noted that not only ordinary semiconductor materials feature the photovoltaic effect (e.g. organic [26] or dye-sensitized solar cells[27]), but in my thesis I discuss only semiconductorrelated photovoltaic phenomena.

These inevitable losses determine the maximum theoretical efficiency of single-junction solar cells, known as the Shockley-Queisser limit [5]. Silicon is a suitable material for harvesting solar energy, as the Shockley-Queisser limit for the  $E_g = 1.12 \text{ eV}$  band gap of Si is around 32%, which is quite close to the "ideal" 1.3 - 1.5 eV band gap [29], theoretically enabling 33.5% conversion efficiency for the solar spectrum at the Earth's surface [30].

To surpass this limitation, multi-junction cells have been designed, consisting of stacked layers of materials with different band gaps. This structure absorbs a broader range of the solar spectrum. By capturing different wavelengths within different materials, these cells can minimize spectral losses and improve overall efficiency. Although the most advanced multi-junction cells can operate with efficiency above 40% [31] (actual world record is 47.6% from



Figure 2.1: (a) The usable portion of the solar irradiance spectrum and the related losses in a silicon solar cell [28]. (b) In a traditional silicon-based solar cell, the electric field created in a p-n junction is responsible for charge carrier separation.

2022 on a wafer-bonded four-junction concentrator solar cell [32]), they are used only for space applications, since their fabrication is very expensive. From the other side, the development of cost-efficient two-junction tandem solar cells is the hottest contemporary photovoltaic (PV) research topic [33, 34]. For example, a silicon-based bottom cell combined with a perovskite top cell is considered a promising candidate to replace the silicon single-junction cells in mass production [35–37].

In the next operational step, the generated EHPs must be separated by an electric field. In a traditional silicon-based solar cell, one surface of the p-type silicon wafer is highly doped with donor atoms to create a p-n junction. Near the p-n junction, the conduction and valence bands bend to compensate the mismatch of the chemical potentials (which is referred as Fermi-level in the semiconductor society) in the n and p type regions, which by definition manifests in a built-in electric field near the p-n junction in both material regions (Figure 2.1b.). The width of this region is traditionally described using the depletion approximation, in which the ionized donor atoms in a depleted region (that is free of delocalized charge carriers) around the p-n junction are responsible to create the built-in electric field. Minority carriers from each side of the p-n junction reaching this depletion region are swept to the other side by the built-in electric field. Electrons are driven toward the n-type side, while holes are pushed toward the p-type side. In these regions both types of charge carriers belong to the majority charge carriers thus these are protected from further recombination.

The final requirement that the solar cell must meet is the collection and extraction of the generated current. For this purpose, electrical contacts must be created, through which the electron and hole currents generated inside the solar can be connected to external electronic systems to produce electrical power.

#### **Realistic solar cells**

In realistic solar cells, several additional factors contribute to efficiency losses, further reducing the performance compared to the Shockley-Queisser limit. These losses can be broadly categorized as optical-, recombination-, or resistive-type losses. Optical losses occur because the cell cannot capture all the incident photons. Some portion of the light is reflected from the illuminated surface of the cell or pass through the cell without being absorbed. In  $100 - 180 \,\mu$ m thick Si wafers, typically used for solar cells, at wavelengths above 900 nm, the light absorption weakens, causing significant current loss [38]. In high-efficiency modern silicon solar cells, several methods are applied to enhance light absorption. To minimize reflection, solar cells are coated with anti-reflective layers (typically with silicon nitride), which reduce the amount of reflected light, allowing more light to enter the cell [39]. Texturing the surface of the cell (e.g., by using pyramidal or nanostructured patterns) is the other technique, further increasing the likelihood that photons are absorbed within the cell [40, 41].

Recently, popular methods involve materials that enhance energy efficiency by converting multiple lower-energy photons into a higher-energy one (up-conversion) [42, 43], while others capture high-energy photons and split them into two lower-energy photons (down-conversion) [44, 45]. These techniques are being researched as ways to better utilize the solar spectrum and reduce optical losses, thus circumventing the Shockley-Queisser limit.

Electron-hole pairs generated by absorbed photons can recombine before the built-in electric field separates them. When this occurs, they do not contribute to the current, leading to recombination-type losses. It has to be noted that recombination losses primarily reduce the operating voltage of the solar cell due to the reduction of excess carrier concentration. It decreases the splitting of Fermi levels of electrons and holes, which is the fundamental origin of the voltage in a solar cell [25]. Recombination can occur through various mechanisms, such as surface recombination (due to defects or imperfections at the cell surface), bulk recombination (within the silicon wafer), or recombination at interfaces between different materials in the cell. In Section 2.2.2, the physical mechanisms and typical solar cell recombination processes are summarized in detail.

Resistive losses occur due to the internal resistance of materials within the solar cell and at the interface between the semiconductor and metal contacts. Series resistance arises from resistance to current flow within the cell itself, while shunt resistance is caused by leakage currents that bypass the intended current path. The design of metal contacts and grid lines on the surface of the solar cell can reduce series resistance. Using low-resistivity materials for electrical contacts, interconnects, and front/back electrodes also helps to minimize the resistive losses. Materials like silver, copper, and conductive polymers are commonly employed. Advanced contact formation technologies, such as carrier-selective contacts, can be successfully applied to maximize the shunt resistance by conducting the current only in the preferred direction, similarly to the operation of diodes [46, 47].

For efficient operation, the solar cell must be designed to maximize the power conversion efficiency. Several strategies are applied to enhance the light absorption, ensure the effective separation and transport of charge carriers, and minimize the recombination rate. In some cases, improvements for one loss mechanism can aggravate other losses, therefore the careful optimization requires a holistic approach. For example, increasing the density of metal grids reduces the resistive losses but increases the optical losses as a larger portion of the solar cell surface is covered by metal [48]. Therefore, all effects must be considered together in the optimization of the solar cell architecture.

Several research and development projects focus on optimizing materials, device structures, and processing techniques. As technology advances, these solutions will continue to push the boundaries of solar cell performance, making the solar energy more viable and sustainable.

#### 2.1.2 Modern solar cell structures

The most important goal of the continuous development of solar cell structures is to reduce the recombination-related losses. Until 2017, most solar cells were manufactured from multicrystalline silicon wafers (polycrystalline material with typical grain size of several millimeters) [49], featuring low bulk recombination lifetime. Consequently, the simplest Back Surface Field (BSF) cell structures were sufficient to realize the inherent potential of the materials [50, 51].

With the improvement in the silicon crystal quality, first achieving more controlled grain structure in "high performance" multicrystalline cast ingots [52], then due to the transition to monocrystalline silicon wafers, the role of surface recombination became increasingly significant. During this period, the Passivated Emitter and Rear Cell (PERC) solar cell type began to displace traditional technology [53, 54].

After the significant improvement of the surface passivation properties, the recombination near the contacts became the main limiting factor. So-called passivated contact solar cells provide solutions to this problem [55]. This increased the significance of the previously developed Silicon Heterojunction (SHJ) cell type, which utilizes thin intrinsic amorphous silicon (a-Si) layers to passivate the silicon surface and to gently isolate the metal contacts [56, 57]. In the development of traditional p-n junction based cells, it led to the concept of the Tunnel Oxide Passivated Contact (TOPCon) structure, which reduces recombination at the rear contacts of a PERC-like structure [58]. A promising further development direction is Interdigitated Back Contact (IBC) solar cells, in which both the electron and hole contacts are placed on the back side of the cell [59, 60]. This allows the extension of TOPCon passivation to the emitter as well. In this section, I describe the operation of the PERC and SHJ cell types in more detail, as ingots and wafers related to these sample types were used during my research.

#### Passivated Emitter and Rear Cell (PERC) solar cell

Passivated Emitter and Rear Cell (PERC) type solar cells represent an advanced type of silicon solar cell and have been the workhorse of the solar cell industry for almost ten years. They feature improved efficiency compared to traditional solar cells by using a passivated rear surface to reduce recombination losses. The PERC solar cell structure is depicted in Figure 2.2. Its production steps include various key processes that enhance performance by minimizing recombination losses and improving light absorption.

In this section, I introduce the main manufacturing steps of PERC solar cells in more detail, since all more advanced homojunction cell concepts originated from PERC and employ the same or similar building steps. PERC solar cells are made from thin silicon wafers, typically sliced from a high-quality, low-defect p-type monocrystalline silicon ingot using wire sawing method [62]. Monocrystalline silicon is preferred for PERC cells due to its higher efficiency, which results from its excellent crystal quality with very low crystal-defect density [63]. In recent years, gallium has been used as a doping material for silicon ingots instead of boron due to the absence of light-induced degradation (LID) [64–67], a significant recombination mechanism related to boron-oxygen complexes.

The wafers are then chemically etched to remove surface scratches originating from the sawing process. Next, the wafer is etched in a crystal orientation preferred way, using an acid or alkaline solution (typically sodium hydroxide or potassium hydroxide) to create a textured surface with pyramidal (Figure 2.2.) or random features. This surface texture reduces light



Figure 2.2: Cross-sectional schematic diagram of the PERC structure based on a p-type Czochralski-grown (CZ) wafer with highly doped  $n^+$  emitter layer. Both surfaces are passivated by dielectric layers (SiN<sub>x</sub> and Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub>). The back contacts are screen-printed in thin fingers to reduce recombination losses. During the co-firing process, the aluminum diffuses into the wafer creating a local back surface field (LBSF), which repels the minority electrons from the surface. From *Review of status developments of high-efficiency crystalline silicon solar cells*, Liu Jingjing *et al.*[61].

reflection, and increases the virtual absorption path, which is important for longer wavelength photons allowing more light to be trapped within the wafer, which improves the cell efficiency.

To create the p-n junction necessary for the photovoltaic effect, phosphorus dopants are introduced into the front surface of the wafer. This involves placing the silicon wafer in a high-temperature furnace and exposing it to a phosphorus-containing gas (typically POCl<sub>3</sub>). At high temperatures (typically over 1000 °C), the phosphorus atoms diffuse into the upper few hundred nanometers of the silicon surface, creating the n-type emitter region of the solar cell.

In the next steps, the front and rear surfaces of the wafer are passivated to minimize surface recombination. A thin layer of silicon nitride  $(Si_3N_4)$  is typically deposited on the front surface using plasma-enhanced chemical vapor deposition (PE-CVD). This serves both as a passivation layer and an anti-reflection coating. The back surface is often passivated using a multilayer structure of AlO<sub>x</sub> and Si<sub>3</sub>N<sub>4</sub>, both deposited by PE-CVD [68, 69].

To form the metallic contacts, fine metal fingers (typically silver) are screen-printed onto the front surface of the wafer in a grid pattern, along with a busbar that connects the fingers to improve current collection. On the back surface, windows are first opened in the passivation layers using laser ablation. This is the so-called laser contact opening (LCO) process. Aluminium fingers are then screen-printed into these windows. Finally, the co-firing process takes place, where the wafer is subjected to high temperatures in a furnace (typically around 800 - 900 °C) to fuse the metal contacts to the silicon and ensure good electrical contact between the metal and the wafer. At the backside, during co-firing, the aluminum diffuses into the silicon, forming a low-resistance electrical contact. Furthermore, the increased, local Al doping creates a p+/p junction, which serves as a local back surface field (LBSF), and reduces recombination by repelling the minority electrons from the surface.

The PERC structure is inherently bifacial, designed to capture sunlight from both the front

and rear surfaces of the solar cell [70]. These cells are particularly effective in environments with high albedo (reflective surfaces like snow and sand), where reflected light contributes to a significant increase in power yield. The current efficiency record for such a structure is over 24% [71], while in mass production, the typical value lies between 22% and 23% [72].

Although PERC technology is still produced in large quantities, the transition to TOPCon technology as a dominant solar cell type happened very quickly during the last two years [73]. TOPCon represents the next generation of high-efficiency (Si-Si) homojunction silicon solar cells with enhanced passivation quality of the rear surfaces using a thin tunnel-oxide layer between the silicon wafer and metal contacts. This structure makes it possible to achieve an efficiency above 26.5% [74].

In summary, PERC technology is the first in mass production aiming to minimize the recombination lifetime related to both surfaces and inside the wafer. Therefore, carrier lifetime and its accurate determination for R&D and in different phases of the solar cell production is essential.

#### Silicon Heterojunction (SHJ) based solar cells

The next-generation high efficiency Silicon Heterojunction (SHJ) solar cells represent a different family of silicon solar cells. While in silicon homojunction cells (PERC, TOPCon, IBC) a diffused layer creates the p-n junction responsible for the charge-separating electric field, in SHJ cells it is achieved by the deposition of a semiconducting layer with a different band gap. SHJ cells combine the advantages of crystalline silicon (c-Si) with thin layers of amorphous silicon (a-Si) to create a heterojunction, since a-Si has a larger band gap, around 1.7 eV, than that of crystalline silicon (1.12 eV) [75]. Since the c-Si / a-Si interface has a very low defect density, it significantly reduces carrier recombination at the surfaces and improves the overall efficiency of the solar cell [56]. The cross-sectional schematic diagram of the cell structure is depicted in Figure 2.3.

SHJ cells are fabricated from n-type high-quality monocrystalline silicon wafers sliced from Czochralski-pulled ingots. After polishing and etching, the wafer thickness is typically around  $100 - 150 \,\mu$ m, with textured surfaces on both sides, as the solar panels are typically produced in a bifacial arrangement.

In the next step, thin a-Si layers are deposited on both surfaces by PE-CVD method to create the heterojunction and passivate the surface of the crystalline wafer. A-Si has many dangling bonds, which can lead to defects and carrier recombination. Hydrogen helps passivate these dangling bonds, improving the electronic properties of the a-Si layer and reducing recombination [77]. Hydrogen is introduced directly into the precursor gas mixture during the PE-CVD process. The most common precursor gases for a-Si deposition are silane (SiH<sub>4</sub>) or disilane (Si<sub>2</sub>H<sub>4</sub>), both of which contain hydrogen.

The PE-CVD deposition allows for the deposition of thin layers (typically under 10nm), which is crucial to minimize the optical losses. Photons absorbed in the a-Si do not entirely contribute to the current. First, a few nm thin intrinsic layer is deposited, which is followed by a p-doped emitter layer on the back surface and an n-doped layer on the front surface. In some cases, these amorphous layers are replaced by nanocrystalline (nc) silicon or nc silicon oxide [78], as can be observed on the front surface of the structure in Figure 2.3.

In SHJ solar cells, Transparent Conductive Oxide (TCO) layers play a crucial role in forming efficient electrical contacts between the a-Si layers and the metallic grids while allowing



Figure 2.3: Cross-sectional schematic diagram of the Silicon Heterojunction (SHJ) cell structure. From *Aluminum-Doped Zinc Oxide as Front Electrode for Rear Emitter Silicon Heterojunction Solar Cells with High Efficiency*, Daniel Meza *et al.*[76].

light to pass through to the underlying silicon layer. In most cases, indium tin oxide (ITO) is used as the TCO on both surfaces due to its excellent electrical conductivity and optical transparency [79]. This combination ensures that they can collect the electrical current generated by the cell without significantly blocking incoming light. These layers are typically deposited by physical vapor deposition (PVD) and due to the optimal refraction index, they also serve as an antireflection coating.

In the last step of SHJ cell production, metallic contacts are formed on both surfaces using screen-printed silver. The thermal treatment of the metallization, like all other processes, is performed at low temperatures (below 250°C) to prevent the crystallization of the a-Si layers. This also means that the electrical properties of the initial wafer are preserved.

Due to the excellent surface passivation achieved by a-Si layer stacks, the absolute efficiency record among single-junction solar cells is held by the SHJ structure, with 27.3% [80]. In mass production, efficiencies over 25% are typically achieved. The SHJ solar cell is also a promising candidate as a bottom cell for Si/perovskite tandem structures [81–83].

Currently, the silicon heterojunction interface provides the best recombination properties, therefore these structures have twofold importance. First, they are applied to make excellent solar cells; secondly, they enable to determine the bulk lifetime of the silicon wafers. Often the latter is not possible, as annealing processes above 500°C, such as thermal oxidation, introduce bulk defects in Si. However, since the a-Si layers acting as passivation layers are deposited at much lower temperatures, the as-grown recombination properties of the silicon material can be determined [84, 85].

### 2.2 **Properties of charge carriers in silicon**

This section provides a comprehensive overview of the principal characteristics of charge carriers in silicon, with a particular emphasis on recombination properties and their measurement techniques. The discussion begins with an examination of charge carrier mobility and its existing models for silicon. Subsequently, the various recombination mechanisms in semiconductor materials are presented, along with strategies to mitigate their effects in silicon-based devices. Finally, the most significant carrier lifetime measurement methods are introduced, including how they are affected by other physical quantities and processes such as mobility and diffusion.

Throughout this dissertation, I employ concepts consistent with the conventional description of semiconductors. In this framework, the concentration of electrons in the conduction band (n) and holes in the valence band (p) are determined by the density of states function dictated by the band structure, and the Fermi-Dirac statistics [86].

Within the temperature range under investigation, I apply the approximation that in n-type doped silicon, the equilibrium electron concentration  $(n_0)$  is equivalent to the concentration of dopant atoms  $(N_{dop})$  [87]:

$$n_0 = N_{\rm dop}.\tag{2.1}$$

Furthermore, I assume that following sample excitation, the excess carrier concentrations (also referred to as injection level) of electrons and holes are equal at every point in the sample and at every moment:

$$\Delta n(x,t) = \Delta p(x,t) \tag{2.2}$$

This condition is satisfied in samples where the concentration of recombination-active defects and impurities is lower than  $\Delta n$  [88].

### 2.2.1 Charge carrier mobility in silicon

The Drude model establishes a relationship between the conductivity of semiconductors and the density of charge carriers [86]:

$$\sigma_{\rm e} = \frac{ne^2 \tau_{\rm rel}}{m_{\rm e}^*} = ne\mu_{\rm e}, \qquad (2.3)$$

where *e* represents the electron charge, *n* denotes the electron density,  $\tau_{rel}$  is the momentum relaxation time,  $m_e^*$  is the effective mass of electrons, and  $\mu_e$  represents the electron mobility. After defining the hole mobility  $\mu_h$  in a similar manner, the total conductivity can be expressed as:

$$\sigma = ne\mu_{\rm e} + pe\mu_{\rm h}.\tag{2.4}$$

This relation is particularly important as it links the charge carrier concentrations with the conductivity measured in photoconductance-based techniques.

The precise values of electron and hole mobilities are influenced by various scattering effects within the silicon crystal. The most significant processes include scattering by lattice phonons, dopant atoms or other impurities, and mobile charge carriers (e-e scattering). The momentum



Figure 2.4: Sum of electron and hole mobilities based on several mobility models for a sample with  $N_{dop} = 10^{15} \text{ cm}^{-3}$ .

relaxation times associated with individual scattering processes can be summarized using the Matthiessen rule:

$$\frac{1}{\tau_{\rm rel}} = \frac{1}{\tau_{\rm rel,e-ph}} + \frac{1}{\tau_{\rm rel,e-e}} + \frac{1}{\tau_{\rm rel,dop}}.$$
(2.5)

In lightly doped silicon without excess carriers, the most determinant mechanism is the scattering on phonons, quasi-particles associated to lattice vibration. Due to the difference between electron and hole effective masses, the corresponding charge carrier mobilities differ with al $cm^2$   $cm^2$ 

most a factor of 3. At room temperature  $\mu_e \approx 1400 \frac{\text{cm}^2}{\text{Vs}}$  and  $\mu_h \approx 470 \frac{\text{cm}^2}{\text{Vs}}$ .

Charge carrier mobility in monocrystalline silicon with common dopant atoms (B, P) has been extensively studied as a function of doping concentration [89–92], temperature[89, 91, 93], and injection level [94, 95]. Dannhauser [96] and Krausse [97] employed voltage measurements of p-n junctions to obtain  $\mu(\Delta n)$ . Klaassen synthesized previous models and provided the first physics-based analytical model describing majority and minority carrier mobility, incorporating impurity screening by charge carriers, electron-hole scattering, and the full temperature dependence of these processes [98–100]. Schindler later expanded this description to include compensated silicon [101].

Recent decades have seen significant efforts to develop fast, contactless methods for mobility determination. Three major works based on photoconductance measurements have been presented. Neuhaus et al. utilized the quasi-steady-state open circuit voltage method (QSS-V<sub>oc</sub>) [102]. Rougieux et al. introduced a contactless method combining transient and steady-state lifetime measurements of the quasi-steady-state photoconductance technique for samples with intermediate carrier lifetimes [103]. Using this technique, Zheng et al. provided a parameterization for  $\mu(\Delta n)$  for both n- [104] and p-type [105] silicon. Finally, Hameiri et al. determined  $\mu(\Delta n)$  using an independently calibrated photoluminescence measurement [106].

Figure 2.4. compares the sum of electron and hole mobilities for various models with  $\mu(\Delta n)$  parametrization, using n-type silicon with a doping concentration of  $N_{dop} = 10^{15} \text{ cm}^{-3}$ . The graph reveals that at low  $\Delta n$ , mobility remains nearly constant. However, above  $\Delta n =$ 

 $10^{15}$  cm<sup>-3</sup>, it decreases due to the enhanced effect of e-e scattering. As  $\Delta n$  increases, the discrepancy between these models becomes more pronounced.

In an excited silicon sample, electrons and holes are both present in high concentration. Since the mobility of electrons is larger than that of holes, they can spread faster. Similarly to the Dember effect [107], an internal electric field is induced due to this spatial separation. The drift currents originating from this internal field effectively decelerate the electron motion and accelerate the hole motion. Consequently, this phenomenon can be treated as a drift with a modified coefficient, known as the ambipolar mobility [108–110]:

$$\mu_{\rm amb} = \frac{n+p}{\frac{n}{\mu_{\rm b}} + \frac{p}{\mu_{\rm e}}}.$$
(2.6)

 $\mu_{amb}$  accounts for the Coulomb interaction between holes and electrons.

The Einstein relation provides the connection between the mobility and diffusion coefficient *D*:

$$D_{\rm i} = \mu_{\rm i} \frac{k_{\rm B}T}{e},\tag{2.7}$$

where  $k_{\rm B}$  is the Boltzmann constant and the subscript "i" stands for "e", "h" or "amb". This relation arises from the fact that the same scattering mechanisms determine the drift and diffusion motions of the charge carriers. The ambipolar diffusion coefficient describes the motion of a quasi-neutral group of holes and electrons together, which is a crucial simplification for considering diffusive motion of charge carriers during carrier lifetime measurement.

#### 2.2.2 Charge carrier recombination in silicon

The recombination lifetime is one of the most important physical parameters of a semiconductor material as it may provide information about the presence of harmful defects and contaminations [85, 111, 112]. Furthermore, in the case of solar cell applications, it directly relates to the efficiency of the final device [113, 114]. Therefore, the proper measurement of recombination lifetime is essential to the thorough characterization of modern semiconductor devices.

Even in dark thermal equilibrium, charge carriers are continuously generated by thermal excitation and recombine with each other through various processes. If excess EHPs are generated in the semiconductor instantaneously, the recombination rate increases, and the decay of the charge carrier density can be characterized by the recombination lifetime. In more general terms, the recombination lifetime is defined as [115]:

$$\tau = \frac{\Delta n}{R},\tag{2.8}$$

where  $\Delta n$  is the excess minority carrier concentration or as it is called in solar cell terminology: injection level, and *R* is the recombination rate.

The charge carrier diffusion length refers to the average distance that a charge carrier can diffuse within a semiconductor material before recombination occurs. It is closely related to recombination lifetime:

$$L_{\rm D} = \sqrt{D \cdot \tau},\tag{2.9}$$



Figure 2.5: The main charge carrier recombination processes in semiconductors. A) Radiative recombination, B) the Auger process, C) the Shockley-Read-Hall mechanism. From *Sustainable Developments by Artificial Intelligence and Machine Learning for Renewable Energies*, Sourav Sadhukhan *et al.*[116].

where D is the diffusion coefficient of the minority charge carriers. In solar cells, the diffusion length is an important parameter, as it relates to the probability that the minority charge carriers reach the junction before recombining.

The rate of most recombination processes depends on the charge carrier density. The exact dependence  $(R(\Delta n))$  is determined by the number of charge carriers involved in a single recombination event.

The three major recombination mechanisms are summarized and illustrated in Figure 2.5. After a photon is absorbed, the induced excess electrons and holes rapidly thermalizes to the band edges. The type of a recombination event relates to the mechanism how the band gap equivalent energy is emitted during the recombination event.

#### **Radiative recombination**

Radiative recombination is the most direct way of recombination, where an electron from the conduction band moves to an empty electron state in the valence band, "jumps into a hole state", thus an electron and a hole recombine, while a photon is emitted with a photon energy close to the band gap. Since one electron and one hole are involved in a single recombination event, it implies the following charge carrier dependence of its recombination rate:

$$R_{\rm rad} = A \cdot n \cdot p = A \cdot (n_0 + \Delta n) \cdot \Delta n, \qquad (2.10)$$

where A is the radiative recombination coefficient, and we assume an n-type material with  $n_0$  doping concentration.

Since this is an elementary recombination process, its rate is determined by the band structure of the crystal, therefore its existence and the related recombination loss is unavoidable. Although this phenomenological approach and the related formula is simple, the deeper physics behind is integrated in the value of the radiative recombination coefficient [117, 118]. In case of indirect band gap materials,  $R_{rad}$  is small, since this recombination event requires the involvement of phonons as well to satisfy both the energy and momentum conservation requirements. Therefore, in silicon, its rate is insignificant compared to other recombination mechanisms. However, radiative recombination in silicon is important and constantly researched [119–121]. The emitted photons can be captured by sensors and cameras providing an excellent method to monitor or image the carrier concentration (imaging photoluminescence) or provide information about the distortion of the band structure in given positions (spectral photoluminescence) [122–124].

#### Auger recombination

In the Auger process, an electron and a hole recombine directly and release their energy to a third carrier (electron or hole), which is first excited to a higher energy state, then relaxes back to the bottom of the conduction band (or to the top of the valence band). Therefore, the rate of Auger recombination,  $R_{Aug}$ , depends on the third carrier concentration as well. The general expression for the Auger recombination rate is:

$$R_{\text{Aug}} = C_n \cdot n^2 p + C_p \cdot np^2 = C_n \cdot (n_0 + \Delta n)^2 \Delta n + C_p \cdot (n_0 + \Delta n) \Delta n^2, \qquad (2.11)$$

where  $C_n$  and  $C_p$  are the Auger recombination coefficients describing the mechanisms for when the third carrier is an electron (as shown in Figure 2.5.) or a hole, respectively.

Similarly to radiative recombination, the strength of the Auger process also relates to the crystal structure, and the doping concentration (which is intended and controlled in most cases) and is independent of structural defects or contaminations. Therefore, these two recombination processes are collectively called intrinsic recombination. My personal remark is that this is unfortunate naming used in PV society, incorrectly implying that they relates to intrinsic material properties, although their rate depends strongly on intentional doping concentration. A better choice would be unavoidable processes or non-defect related recombination processes. Since this recombination is typically dominant at high  $\Delta n$  in silicon, the accurate values of these coefficients are a constant research topic [125–127]. In 2012, Richter et al. provided a parametrization for intrinsic recombination [128], which has been fine-tuned recently by Black et al. [129] and Niewelt et al. [4] for Auger recombination, and Fell for the radiative part [121].

#### **SRH** recombination

The Shockley-Read-Hall (SRH) process is a recombination mechanism that occurs via energy states (trap states) in the band gap [130]. These trap states can be caused by local disturbance of the perfect periodicity of crystal, such as grain boundaries, dislocations or other crystal defects or impurities in the silicon crystal and on the surface of the crystal. In SRH recombination, an electron from the conduction band and a hole from the valence band are captured by the same trap state, and after recombination, the energy is released typically as thermal energy in the form of a phonon. The rate of SRH recombination,  $R_{SRH}$ , is given by:

$$R_{\rm SRH} = \frac{np - n_{\rm i}^2}{\tau_{\rm n} \left( p_1 + p \right) + \tau_{\rm p} \left( n_1 + n \right)},\tag{2.12}$$

where  $n_i$  is the intrinsic carrier concentration,  $\tau_n$  and  $\tau_p$  are the minority carrier lifetimes for electrons and holes for the given defect, respectively. These lifetime values are related to the capture-cross sections,  $\sigma_n$  and  $\sigma_p$  and the density of the trap state,  $N_T$ :

$$\tau_{\rm n} = \frac{1}{N_{\rm T} \cdot \sigma_{\rm n} \cdot v_{\rm th}},\tag{2.13}$$



Figure 2.6: The bulk recombination lifetime is dominated by different elementary processes at low and high injection levels. From *Lifetime Spectroscopy—A Method of Defect Characteriza-tion in Silicon for Photovoltaic Applications*, Stefan Rein [134].

$$\tau_{\rm p} = \frac{1}{N_{\rm T} \cdot \sigma_{\rm p} \cdot v_{\rm th}},\tag{2.14}$$

where  $v_{\text{th}}$  is the thermal velocity of charge carriers.  $n_1$  and  $p_1$  are additional terms related to the effective recombination time and the positions of the defect levels in the band gap, influencing the recombination rate.

$$n_1 = N_{\rm C} \cdot \exp\left(\frac{E_{\rm T} - E_{\rm C}}{kT}\right),\tag{2.15}$$

$$p_1 = N_{\rm V} \cdot \exp\left(\frac{E_{\rm V} - E_{\rm T}}{kT}\right),\tag{2.16}$$

where  $E_{\rm T}$  is the energy of the trap level.  $E_{\rm C}$  and  $E_{\rm V}$  are the energies at the bottom of the conduction band and the top of the valence band, respectively. While  $N_{\rm C}$  and  $N_{\rm V}$  are the effective density of states in the conduction band and valence band near these energy levels. As a result, the rate of the SRH recombination process depends on the four main parameters of the given defect: the density, the capture cross sections and the trap energy level. Therefore, there are defects more "harmful" for the device operation, typically with large capture cross sections, compared to others. For instance in p-type silicon, most contaminating transition metal atoms cause strong SRH recombination rate [131, 132], while the SRH recombination activity of interstitial oxygen is negligible, despite its concentration in Cz grown silicon being ~ 1000 times larger than the doping concentration itself [133].

Typically, the different recombination processes occur simultaneously. In this case, the total recombination rate is the sum of individual contributions considering more SRH recombination centers:

$$R_{\text{total}} = R_{\text{rad}} + R_{\text{Aug}} + R_{\text{SRH},1} + R_{\text{SRH},2} + \dots$$
(2.17)

Then the effective carrier lifetime,  $\tau_{eff}$ , is given by

$$\frac{1}{\tau_{\rm eff}} = \frac{1}{\tau_{\rm rad}} + \frac{1}{\tau_{\rm Aug}} + \frac{1}{\tau_{\rm SRH,1}} + \frac{1}{\tau_{\rm SRH,2}} + \dots$$
(2.18)

In silicon samples for solar cells, the dominant recombination processes are typically defectrelated SRH at low injection levels and Auger recombination at high injection levels as depicted in Figure 2.6. with  $\tau_{\text{eff}} = \tau_{\text{bulk}}$  as only bulk recombination processes were considered.

#### Surface recombination

Surface recombination refers to the process in which charge carriers (electrons and holes) recombine at the surface or interface of a semiconductor material, rather than in the bulk. This phenomenon is particularly important in semiconductor devices, such as solar cells, since their thickness is smaller than the diffusion length. Therefore, surfaces often significantly influence the overall device performance.

The large variety of dangling bonds on the surface and other structural imperfections manifest in surface states, which have energy levels located within the band gap of the semiconductor [135]. Therefore, their recombination properties can be described with the Shockley-Read-Hall model as well. However, instead of one discrete energy level, there is a spectrum of energy states at the surface. The practical approach to mathematically handle the surface recombination phenomenon in a universal way applies the parameter called surface recombination velocity (SRV) *S* [136, 137]. Its definition is similar to the carrier lifetime integrating all recombination events corresponding to the surface [138]:

$$S = \frac{R_{\rm surf}}{\Delta n_{\rm surf}},\tag{2.19}$$

where  $\Delta n_{\text{surf}}$  is the surface excess carrier density and  $R_{\text{surf}}$  is the recombination rate on the surface. S has dimensions of m/s (the majority of the literature uses cm/s), which stems from the fact that  $R_{\text{surf}}$  is a surface quantity with dimensions of m<sup>-2</sup>s<sup>-1</sup>.

#### **Recombination and diffusion**

So far, the recombination rates were handled as local quantities. However, in practice, carrier generation and recombination events in different locations are linked to each other due to the movement of the carriers, which must be taken into account for our experiments. In the general case, the continuity equation determines the change of excess electron concentration during the measurement[115]:

$$\frac{\partial \Delta n(x,t)}{\partial t} = G(x,t) - R(x,t) - \nabla (J_{\text{n,diff}} + J_{\text{n,drift}}), \qquad (2.20)$$

where G(x,t) is the generation rate, R(x,t) is the recombination rate, and  $J_{n,diff}$  and  $J_{n,drift}$  are the diffusion and drift current densities. Several realistic assumptions can be made to simplify this equation.

First, material homogeneity is assumed, so the doping level, carrier lifetime and mobility do not change in the investigated volume of the sample.

Secondly, external electric field is not taken into account. Although an electric field is a crucial part of the operation of solar cells, in crystalline silicon solar cells this field is localized in the few 100 nm thick depletion region near to the surface. This volume is negligible compared to the total thickness of the cells, which is typically  $100 - 200\mu$ m. Using these assumptions, the continuity equation of electrons and holes can be simplified as

$$\frac{\partial \Delta n(x,t)}{\partial t} = G(x,t) - R(x,t) + D_{\rm e} \frac{\partial^2 \Delta n(x,t)}{\partial x^2} + \Delta n(x,t) \mu_{\rm e} E_{\rm int}, \qquad (2.21)$$

$$\frac{\partial \Delta p(x,t)}{\partial t} = G(x,t) - R(x,t) + D_{\rm h} \frac{\partial^2 \Delta p(x,t)}{\partial x^2} - \Delta p(x,t) \mu_{\rm h} E_{\rm int}, \qquad (2.22)$$

where  $D_e$ ,  $D_h$ ,  $\mu_e$  and  $\mu_h$  are the diffusion coefficient and mobility for electrons and holes, respectively.  $E_{int}$  indicates the internal electric field arising from the spatially different distribution of electrons and holes. The corresponding drift current can be incorporated into the diffusion term using the ambipolar diffusion coefficient. In this case Equation 2.21 and 2.22 can be merged assuming  $\Delta n(x,t) = \Delta p(x,t)$ :

$$\frac{\partial \Delta n(x,t)}{\partial dt} = G(x,t) - R(x,t) + D_{\text{amb}} \cdot \frac{\partial^2 \Delta n(x,t)}{\partial x^2}.$$
(2.23)

This simplified continuity equation is the basis of the carrier lifetime measurement. The surface recombination velocity *S* can be considered as a boundary condition at the edge of the depletion zone  $(x_{depl})$ :

$$D \cdot \frac{\partial \Delta n(x,t)}{\partial x} \bigg|_{x=x_{\text{depl}}} = S \cdot \Delta n(x = x_{\text{depl}}, t).$$
(2.24)

In general case, the continuity equation cannot be solved analytically. However, for the measurement of thin wafers with thickness  $W < L_D$ , a particular solution can be found [115]. During a typical carrier lifetime measurement, the decay of  $\Delta n$  is observed after a short illumination, so the generation part can be omitted:

$$\frac{\partial \Delta n(x,t)}{\partial t} = -\frac{\Delta n(x,t)}{\tau} + D_{\rm amb} \frac{\partial^2 \Delta n(x,t)}{\partial x^2}, \qquad (2.25)$$

which has a solution form:

$$\Delta n(x,t) = A\cos(\alpha x)e^{-\beta t}, \qquad (2.26)$$

where

$$\beta = \frac{1}{\tau} + \alpha^2 D_{\text{amb}}.$$
 (2.27)

This expression enables us to categorize recombination processes according to the location where they occur:

$$\frac{1}{\tau_{\rm eff}} = \frac{1}{\tau_{\rm bulk}} + \frac{1}{\tau_{\rm surf}},\tag{2.28}$$

With  $\tau_{\text{eff}} = \frac{1}{\beta}$  the overall, effective recombination lifetime, and  $\tau_{\text{surf}} = \frac{1}{\alpha^2 D_{\text{amb}}}$  is the surface recombination lifetime. The unkown  $\alpha$  parameter can be determined using the boundary condition (Equation 2.24):

$$\tan\left(\frac{\alpha W}{2}\right) = \frac{S}{\alpha D}.$$
(2.29)

In two limiting cases, the expression can be further simplified. First, in the case of reduced surface recombination  $(S \rightarrow 0)$ :

$$\tau_{\rm surf}(S \to 0) = \frac{W}{2S}.\tag{2.30}$$



Figure 2.7: Effective lifetime ( $\tau_{eff}$ ) versus wafer thickness (*d*) as a function of surface recombination velocity  $s_r$ .  $D = 30 \text{ cm}^2/\text{s}$ . From *Semiconductor Material and Device Characterization*, Dieter K. Schroder [138]

While in the opposite case  $(S \rightarrow \infty)$ :

$$\tau_{\rm surf}(S \to \infty) = \frac{W^2}{\pi^2 D}.$$
(2.31)

This is the so-called diffusion-limited case typically observed in as-cut or polished surfaces, when the surface recombination is limited by the minority carrier diffusion process.

Tüttő et al. proposed that in the general case, the sum of the former two terms can be used with reasonably small error [139]:

$$\tau_{\rm surf} \approx \frac{W}{2S} + \frac{W^2}{\pi^2 D}.$$
(2.32)

The study of these recombination mechanisms is critical for the development of highefficiency devices. Figure 2.7. depicts the thickness dependence of  $\tau_{eff}$  for different as a function of SRV.

A common strategy to mitigate surface recombination in solar cells is surface passivation. Passivating the surface involves coating it with materials (such as silicon dioxide, silicon nitride, or organic layers) that reduce the density of surface states, thereby lowering the surface recombination velocity.

#### **2.2.3 Recombination lifetime measurement principles**

Recombination lifetime is a local property of the silicon crystal. However, due to the movement of charge carriers, its effect on devices is not localized to where the recombination occurs. During carrier lifetime characterization, an apparent effective carrier lifetime ( $\tau_{eff}$ ) is measured (Equation 2.28), which in some cases provides direct information regarding the electrical performance of the final device (e.g. measurement on solar cell structures before metallization). In other cases, the measurement procedure must be adjusted or additional sample treatments have to be applied to obtain the information of interest. Such treatment is the surface passivation of the sample in order to obtain the carrier lifetime within the wafer.

To evaluate and understand the recombination dynamics in silicon materials and devices, several experimental techniques have been developed. They are essential for optimizing the material quality and the device performance, especially for photovoltaic applications. Technically, any carrier lifetime characterization technique is based on the same phenomenon: the injection of excess charge carriers, and the monitoring of their actual density  $\Delta n$ . In the general case, Equation 2.23 describes the evolution of  $\Delta n(t)$ , where we should consider the  $\Delta n$  dependence of each recombination processes and the carrier diffusion. To simplify the discussion, the average of the carrier density in the depth is taken, so  $\Delta n_{av}(t) = \frac{\int_0^W \Delta n(x,t) dx}{W}$ . Using the formulas for  $\tau_{eff}$  in Equation 2.28, the impact of surface recombination and diffusion process can be handled mathematically using a single  $\tau_{eff}$  value. Therefore, we get a simple approximate formula for  $\Delta n_{av}(t)$ :

$$\frac{\partial \Delta n_{\rm av}(t)}{\partial t} = G - R(\Delta n_{\rm av}(t)) = G - \frac{\Delta n_{\rm av}(t)}{\tau_{\rm eff}(\Delta n)},\tag{2.33}$$

where G is the generation rate of the optical injection and  $R(\Delta n)$  is the total recombination rate of the carriers. Rearranging this equation, the general expression of the carrier lifetime measurement can be determined:

$$\tau_{\rm eff}(\Delta n_{\rm av}) = \frac{\Delta n_{\rm av}}{G - \frac{\partial \Delta n_{\rm av}}{\partial t}}.$$
(2.34)

From this point, to simplify the equations,  $\Delta n := \Delta n_{av}$  always refers to the average injection level, while if the local carrier density needs to be applied, it will be indicated as  $\Delta n(x)$ .

In this study, I discuss only contactless carrier lifetime measurement technique, meaning EHP generation by an external light source and  $\Delta n$  sensing by electromagnetic fields.

#### Sensing the carrier density using electromagnetic field

Free carriers can be sensed using electromagnetic (EM) fields, since the dielectric function  $\varepsilon_{\rm r}(\omega)$ , and therefore the refractive index (*n*) and the extinction coefficients ( $\kappa$ ) are influenced by the actual density of free carriers. Based on the Drude theorem for electric fields periodic in time with  $\omega$  angular frequency [86]:

$$\varepsilon_{\rm r}(\omega) = (n + i\kappa)^2 = \varepsilon_{\rm r,0} - \frac{\omega_{\rm p}^2}{\omega(\omega + i/\tau_{\rm rel})}, \qquad (2.35)$$

where  $\omega_p^2 = \frac{ne^2}{\varepsilon_0 m^*}$  is the plasma frequency, *n* is the density of free carriers,  $\tau_{rel}$  is the momentum relaxation time and  $\varepsilon_{r,0}$  is the relative permittivity corresponding to given material but without free charge carriers.

This approach is applicable to understand and estimate the  $\Delta n$  sensitivity of a sensor using the electromagnetic field of a given frequency, either measuring reflectance or absorption. However, it is not precise enough to describe the response of realistic sensors and thus to directly calculate  $\Delta n$  from the measured signal.

In the following, I systematically categorize carrier lifetime measurement methods according to key illumination and detection parameters.

#### Methods classified by the EM field's frequency for $\Delta n$ sensing [115]:

- Radiofrequency (RF): f = 1 100MHz, mostly using RF coils and eddy current method.
- Microwave frequency (MW) f = 1 100GHz, with MW antennas.
- Infrared light (IR):  $\lambda = 1 10\mu m$ , typically detecting the absorption of IR photons.

#### Methods based on optical excitation type

The evaluation and the operation mode of the measurement, corresponding to a given carrier lifetime technique, is determined by the parameters of the optical excitation. The light source, based on wavelength range, can be:

- spectral: lamp, or customized multi-color LED arrays,
- monochromatic: lasers and LEDs.

#### Methods classified by the duration of the light excitation (T)

- short pulse (typically  $T < 1\mu s$ ): mostly semiconductor and solid-state lasers, or optically chopped light sources,
- steady-state (SS) : LEDs, continuous wave (CW) lasers, lamps.

#### Methods classified by the operation mode of the light excitation

Light intensity of steady-state light sources can be modulated in different ways.

• Steady-state (SS) mode:  $T >> \tau_{eff}$ 

 $\Delta n$  is recorded under steady-state conditions, so  $\frac{\partial \Delta n}{\partial t} = 0$ . Therefore,

$$\tau_{\rm eff}(\Delta n) = \frac{\Delta n}{G}.$$
(2.36)

#### • Transient or decay mode: $T << \tau_{eff}$

 $\Delta n(t)$  is recorded after the excitation is terminated, so G = 0. Therefore,

$$\tau_{\rm eff}(\Delta n) = \frac{\Delta n}{-\frac{\partial \Delta n}{\partial t}}.$$
(2.37)

• Harmonic modulation:  $G(t) = G_0 \sin(2\pi t/T)$ 

 $\tau_{\text{eff}}$  can be determined either from the amplitude (V<sub>0</sub>) or the phase shift ( $\phi$ ) of the recorded  $V(t) = V_0 \sin(2\pi t/T + \phi)$  signal [115]. This method is not used in this work.

#### • Small perturbation (SP) method: $G(t) = G_{SS} + \delta G$ if t < T; and $G(t) = G_{SS}$ if t > T.

In this case, after the small  $\delta G$  perturbation is switched off, the recombination follows the dynamics characterized by the so-called "differential" or "small perturbation" carrier lifetime [110, 140, 141]:

$$\tau_{\rm eff,d}(G) = \frac{\partial \Delta n(G)}{\partial G}.$$
(2.38)

From these "components" the recipe to build a carrier lifetime measurement configuration is: choosing one of the  $\Delta n$  sensing options, adding one excitation light option and modulate the intensity corresponding to the required operation mode.

# 2.2.4 Photoconductance-based measurement techniques relevant for this work

#### Quasi-Steady-State Photoconductance (QSSPC)

The Quasi-Steady-State Photoconductance (QSSPC) method, developed by Sinton Instruments, is one of the most widely used techniques for determining minority carrier lifetimes in solar cell silicon samples [20, 142, 143]. This technique utilizes a **flash lamp** to generate EHPs, and the excess conductance is inferred from the **RF coil** inductively coupled to the sample. Two separate methods are used in this technique based on the range of the carrier lifetime to be measured.

For low lifetime values, when the recombination is much faster than the decay of the flash lamp (around  $100 \,\mu$ s), the Steady-State evaluation is used according to Equation 2.36. *G* is constantly monitored by a calibrated photodiode, and  $\Delta n$  is calculated from the photoconductance signal using predefined mobility model data. Errors that may appear in both values are directly transmitted to the determined carrier lifetime as well.

If the carrier lifetime is longer compared to the decay of the illumination, monitoring the light intensity is not necessary, since carrier lifetime is determined from the decay of the detected conductance according to Equation 2.37. This evaluation called transient photoconductance, or photoconductane decay (PCD) method. This evaluation method is less sensitive to uncertainties arising from the charge carrier mobility data used for the calculation of  $\Delta n$ .

#### Microwave-detected Photoconductance Decay (µPCD)

The Microwave-detected Photoconductance Decay ( $\mu$ PCD) method is a contactless, timeresolved technique that provides direct measurement of carrier lifetime through the decay of photoconductance after a **short laser pulse** excitation [18]. Unlike QSSPC,  $\mu$ PCD enables to investigate all samples with  $\tau > 100$  ns with the same PCD evaluation method based on function fitting.

 $\mu$ PCD is widely employed in the characterization of silicon wafers, thin films, and other materials used in semiconductor devices. The technique is especially valuable for analyzing carrier lifetime patterns, as it can be integrated into mapping platforms due to the small spot illumination and detection. Therefore, it is typically applied for contamination detection in the wafer quality control process in the production of integrated circuits.



Figure 2.8: Illustration of small perturbation QSS- $\mu$ PCD method. From QSS- $\mu$ PCD measurement of lifetime in silicon wafers: advantages and new applications, Marshall Wilson *et al.* [144].

#### Quasi-Steady-State Microwave-detected Photoconductance Decay (QSS-µPCD)

The Quasi-Steady-State Microwave-detected Photoconductance Decay (QSS- $\mu$ PCD) method is an advanced **small perturbation** technique developed by Semilab, which applies a continuous wave (CW) laser to generate a steady-state condition and a small energy pulse to perturb the excess carrier concentration at every steady-state level, as depicted in Figure 2.8. [144, 145]. This technique directly measures the differential lifetime,  $\tau_{eff,d}$  (Equation 2.38) which differs from the  $\tau_{eff}$  recombination lifetime introduced previously and will be called "actual" lifetime,  $\tau_a$ , during the application of this method. Measuring the differential lifetime in several steady-state conditions, the actual lifetime can be calculated as follows:

$$\tau_{\rm a}(G_{\rm SS}) = \frac{1}{G_{\rm SS}} \int_0^{G_{\rm SS}} \tau_{\rm diff}(G'_{\rm SS}) \, dG'_{\rm SS}, \tag{2.39}$$

where  $G_{SS}$  is the generation rate corresponding to the illumination intensity adjusted by the CW laser power. Using the actual carrier lifetime, the injection level can be calculated using the steady-state approximation:

$$\Delta n_{\rm SS} = G_{\rm SS} \cdot \tau_{\rm a}. \tag{2.40}$$

This technique also requires the accurate value of the generation rate, which not only depends on the applied laser, but also on the optical properties of the investigated sample. However, its advantage is that mobility data is not needed for the evaluation.

In summary, each of the presented techniques offers a unique approach to measuring charge carrier recombination in silicon. The selection of an appropriate method depends on the specific application, whether it is evaluating bulk material quality, investigating surface recombination dynamics, or searching for contamination. These methods play a pivotal role in optimizing the

performance of silicon-based devices, particularly in photovoltaic applications, where minimizing recombination is critical for maximizing efficiency.

#### Alternative techniques to characterize the recombination lifetime

In addition to photoconductance decay methods, various other techniques are routinely employed to measure carrier lifetimes in the photovoltaic industry. Some of these techniques are presented below, along with their principles, advantages, and limitations.

#### Surface Photovoltage (SPV)

The surface photovoltage (SPV) method involves measuring changes in the surface potential induced by photoexcitation [146–148]. When a light pulse generates excess carriers in the semiconductor, the resulting change in surface potential is monitored. This change is directly related to the excess carrier concentration near the surface. By measuring the SPV signal with light sources of different wavelengths, the diffusion length and carrier lifetime can be calculated.

SPV is a non-contact and non-destructive technique that enables the determination of bulk recombination properties without passivating the surface. However, it requires the presence of a built-in potential, such as that found in a p-n junction at the surface. In addition, the sample must be much thicker than the diffusion length for accurate measurement, a condition that is not met in the case of high-quality thin silicon wafers used in solar cell production. Nevertheless, SPV is intensively used in microelectronic industry due to its unique feature of providing  $L_D$  at very low  $\Delta n$ . For specific metal contaminants this provides better sensitivity to detect them [148–150].

#### **Photoluminescence (PL)**

Photoluminescence (PL) is another widely used technique based on the radiative recombination of photoexcited carriers. In this method, a light source, such as a laser, excites the material, and the emitted photons from radiative recombination are detected and analyzed. The PL technique is highly sensitive and non-invasive, making it suitable for detecting defects and impurities in semiconductor materials. Since detection is based on radiative recombination, the measured signal is proportional to the product of electron and hole densities (Equation 2.10). However, charge carrier concentrations are governed by the net effect of all recombination processes, so PL measurements also provide information about their collective behavior.

PL measurements can be performed using steady-state and transient methods. In steadystate PL measurements, the entire wafer or a thick line across it is typically illuminated with a CW laser, and the emitted light is measured using a camera. Therefore, it is a useful tool for visual inspection of defects in silicon wafers. Therefore, the main application of this technique is to record high-resolution images of recombination-active defects in a very rapid way [122, 124]. However, accurate calibration of the measured signal is challenging due to the complex optical factors involved.

In contrast, photoluminescence decay (PL decay) measures the time-dependent decay of photoluminescence following a short excitation pulse [151–153]. By using a pulsed laser to excite carriers and fast detection equipment to monitor the emitted light, this method directly

quantifies carrier lifetime. PL decay provides detailed insights into both bulk and surface recombination contributions.

#### **Open-Circuit Voltage Decay (OCVD)**

The open-circuit voltage decay (OCVD) technique is particularly useful in device structures like solar cells. It measures the decay of open-circuit voltage after illumination is turned off. Under open-circuit conditions, a light source generates photocarriers, and the subsequent voltage decay reflects the excess carrier density over time, providing an estimate of the carrier lifetime [154]. This technique is simple, cost-effective, and directly applicable to solar cells and diodes. However, it is limited to devices with a well-defined junction.

#### Free Carrier Absorption (FCA)

Finally, the free carrier absorption (FCA) method measures changes in infrared (IR) absorption caused by free carriers in the semiconductor [155]. This method can be also classified as photoconductane measurement based on the Equation 2.35. A short pulse or modulated light source in the visible or near-infrared (NIR) range is used to generate excess carriers in the semiconductor sample through interband absorption. This laser provides the necessary photoexcitation to create electron-hole pairs in the material. Another IR laser, typically in the mid-IR range, is used to probe the free carrier density [156, 157]. The wavelength of the probe laser is chosen such that it corresponds to the absorption band of the free carriers without interacting with the lattice or other intrinsic features of the material.

Each of these techniques provides valuable information about carrier dynamics and recombination mechanisms, with their applicability depending on the material system and specific measurement requirements.

#### 2.2.5 Applications of carrier lifetime measurement

Charge carrier recombination measurements are critical tools for characterizing and optimizing semiconductor materials, particularly silicon. These measurements provide essential insights into material properties, including defect and contamination densities, and the surface quality. As recombination rates directly impact the efficiency and performance of silicon-based devices, understanding and controlling these rates is crucial for various applications, from material qualification to process optimization and cutting-edge research. This section explores the primary applications of charge carrier recombination measurements in silicon.

#### **Qualification of silicon ingots**

Silicon ingot qualification is a vital step in the production of high-quality silicon wafers, which serve as the foundation for semiconductor devices, particularly in the photovoltaic and microelectronics industries. Charge carrier recombination measurements are essential in assessing the quality of silicon ingots before they are sliced into wafers for further processing. These measurements provide information on the defect density, minority carrier lifetime, and overall crystalline quality.

In both the photovoltaic and microelectronic industries, monocrystalline silicon ingots are typically grown using the Czochralski (Cz) process [158]. These ingots can exhibit variations in quality depending on growth conditions and the presence of impurities or defects [159, 160]. This variability is even more pronounced in high-speed industrial solutions used in the photovoltaic industry, such as Recharged Czochralski (RCz) [161, 162] and Continuous Czochralski (CCz) methods [7, 163], where multiple ingots are pulled from the same crucible. By measuring recombination rates, manufacturers can identify regions of high defect density within the ingot, which can affect the performance of devices made from these wafers. Longer minority carrier lifetimes typically correspond to fewer recombination centers, suggesting higher material purity and fewer defects.

Specific types of defects within the ingot, such as oxygen precipitates, carbon-related defects, and metal impurities, can be identified by carrier lifetime measurements using appropriate measurement protocols [111, 112, 123, 164, 165]. Identifying and quantifying these defects at an early stage can reduce production costs by ensuring that only high-quality silicon material is used in wafer production.

Measurement methods such as Quasi-Steady-State Photoconductance (QSSPC) and eddycurrent based Photoconductance Decay (ePCD) are commonly used to determine recombination lifetime in silicon ingots. The deep photogeneration achieved by infrared laser excitation allows for rapid assessment of bulk recombination in the ingot, which is critical for ensuring that the measured quantity is related to the crystalline quality rather than surface recombination.

#### **Industrial process control**

Process control is crucial for maintaining the consistency and quality of semiconductor devices during large-scale production. Charge carrier recombination measurements are widely used in industrial process control to monitor material properties, identify process variations or process-related contamination, and to indicate critical failures during production.

In solar cell production, after silicon ingots are sliced into wafers, they undergo a series of processes, such as doping, etching, and passivation. Each of these steps can introduce defects or alter their size or structure, which in turn affects the recombination behavior. A frequent issue is the formation of oxygen precipitates during the high temperature processing steps [85, 111, 166]. Charge carrier recombination measurements can be used to continuously monitor the effects of these processes.

By incorporating charge carrier recombination measurements into the industrial process control workflow, manufacturers can continually refine their processes. For example, PCD measurements are routinely applied to control the PE-CVD deposition in HJT cell productions. They are capable to detect the changes in temperature, pressure, or precursor gas flow during deposition processes.

In conclusion, charge carrier recombination measurements are an essential tool in industrial process control, enabling the monitoring and optimization of semiconductor manufacturing processes to ensure consistent product quality and improved yields.

#### **Research and development**

In the field of research and development (R&D), charge carrier recombination measurements are extensively used to explore new materials, develop advanced device architectures, and to optimize the performance of emerging technologies. These measurements determine the recombination properties of novel materials and help researchers develop strategies to mitigate recombination losses, thus improving device performance.

In the development of new materials for solar cells, photodiodes, and other semiconductor devices, understanding the recombination characteristics is critical. For example, researchers investigate novel perovskite-based structures, such as perovskite-silicon tandem cells [167, 168]. Charge carrier recombination measurements help assess the impact of defects, interfaces, and material properties on carrier lifetime and recombination rates in these new materials. By analyzing recombination rates in these novel materials and structures, researchers can identify pathways to improve efficiency.

In photovoltaic R&D, understanding and reducing recombination is crucial to increase the efficiency of high-efficiency solar cells, such as those based on PERC, SHJ, or IBC technologies. Researchers use recombination measurements to understand how new passivation techniques, surface treatments, or novel materials affect recombination and, consequently, the device performance.

By regularly measuring recombination rates in silicon ingots, manufacturers can optimize the conditions under which the ingots are grown. For instance, variations in pulling rate, temperature, gas composition, and crucible refilling during crystal growth can affect defect density and, consequently, recombination behavior [8–10, 169, 170]. Recombination measurements provide feedback that can be used to fine-tune these parameters to improve the quality of the resulting material.

In conclusion, charge carrier recombination measurements are indispensable tools in R&D, enabling researchers to explore new materials, optimize device performance, and develop innovative technologies. By providing valuable insights into material properties, defect structures, and carrier dynamics, these measurements facilitate the development of next-generation semiconductor devices with higher efficiencies and improved performance.

### 2.3 Recombination processes in modern solar cells

Recombination lifetime is a critical factor influencing the efficiency of modern solar cell structures, including PERC, TOPCon and SHJ technologies. Longer recombination lifetimes enhance solar cell performance by increasing the voltage that can be extracted from the solar cells and the overall efficiency. Advanced cell designs rely heavily on strategies to minimize recombination losses in the bulk and at the surfaces.

#### 2.3.1 Bulk defects in silicon crystal

In silicon-based solar cells, bulk lifetime refers to the sum of recombination processes existing in the bulk crystalline silicon wafer (Equation 2.18). Besides the unavoidable intrinsic recombination, it is mostly related to impurities and lattice defects that enter the silicon during the crystal growth process. The most significant families of the critical defects are the metallic contamination and the presence of silicon oxide precipitates. The vast majority of modern solar cells are made from monocrystalline silicon grown using the Czochralski method. In this process, a monocrystalline rod is slowly pulled out from molten silicon contained in a quartz crucible. Despite the high purity of the melt, a significant amount of oxygen (in concentrations of a few ppm) is present, which dissolves into the silicon from the crucible walls. Detrimental defects originate from oxygen impurities [63, 85], some frequent types of which are presented below.

Not only the growth process but also the cell fabrication procedure can contaminate the wafer or change the microstructure of existing defects. This effect is even more significant in the production of homojunction solar cells (PERC, TOPCon), where high-temperature processing steps enable the formation of oxygen-related harmful defects and the diffusion of surface contamination into the wafer. In contrast, the low-temperature fabrication process of SHJ cells ( $< 250^{\circ}$ C) preserves the high-quality silicon substrate properties, avoiding the introduction of new defects and maintaining long recombination lifetimes.

**Oxide precipitates (OP)** are inclusions of SiO<sub>2</sub> with a size of a few nanometers [171]. The nucleation of OP typically takes place around 700 °C during the cooling of crystallization or high-temperature solar cell processing steps. The density and size of OP depend on the thermal history of the sample, as after nucleation, the growth rate or dissolution depends on the time span the sample spends at high temperature above 900 °C. The nucleation of OP can be enhanced in the presence of silicon vacancies. The mechanism behind the formation, evaluation, and dissolution of OPs is thoroughly described by the Voronkov model explaining the formation of enlarged OPs during solar cell processing with a two-step model [172, 173]. These precipitates strain the lattice and act as strong recombination centers, reducing the bulk recombination lifetime in silicon. Murphy et al. presented thorough studies about the recombination mechanism of OP and the parametrization of the corresponding energy states in both n- and p-type silicon [111, 112, 174].

**Thermal donors (TD)** are nano-chains of oxygen and silicon atoms in the Si crystal and act like donor atoms with double ionization, as their name suggests [175, 176]. The formation temperature lies in the range between 300°C to 550°C, with a peak growth rate at 450°C. In addition to the donor behavior, the recombination activity of TD was proven by Hu et al. [177], mostly at high TD densities (>  $10^{15}$  cm<sup>-3</sup>) [85, 178, 179].

Light and Elevated Temperature-Induced Degradation (LeTID) is a phenomenon observed in silicon solar cells, particularly in PERC and TOPCon architectures [180, 181]. It results in a significant, reversible reduction in solar cell performance when exposed to light and elevated temperatures during operation. LeTID manifests when cells are exposed to temperatures typically ranging from 50 °C to 75 °C under illumination. The degradation can occur over hours to hundreds of hours of exposure. Following the degradation phase, partial or full recovery is observed during continued light soaking or annealing.

The origin of LeTID is still under debate [182, 183], but it is agreed that hydrogen-related defects are involved. The hydrogen is introduced into the silicon during cell processing and diffuses to other impurities or lattice defects [184, 185]. Such complex structures form into

recombination centers or are activated during light soaking at elevated temperatures [186].

Although the concentration of interstitial oxygen in the Cz silicon is very large, typically  $10^{17} - 10^{18} \text{ cm}^{-3}$ , their direct recombination activity is weak. The oxygen related issues correspond to other complex defects formed from the interstitial oxygen. However, their evolution can be mitigated by controlling the crystal growth process.

Compared to oxygen, the amounts of **metallic contaminants** are orders of magnitude less, roughly around  $10^9 - 10^{11}$  cm<sup>-3</sup> in modern materials. Nevertheless, their impact to the PV cell performance can be significant. These contaminants may originate from the crucible walls or from the feedstock silicon of the Czochralski process. The most frequent metallic impurities such as Fe, Cu, Ni, and Cr form deep-level traps in the band gap, acting as efficient recombination centers [132, 187, 188]. The distribution of impurities alongside the ingot follows the Scheil-Pfann equation [186], which describes the segregation of impurities during directional solidification:

$$C_{\rm S} = kC_0(1-g)^{k-1} \tag{2.41}$$

where  $C_S$  is the impurity concentration in the solid, k is the segregation coefficient,  $C_0$  is the initial impurity concentration in the melt, and g is the fraction of melt that has solidified.

To mitigate their impact, two primary strategies are employed to trap or remove these impurities.

First, during internal gettering, defects such as dislocations, oxide precipitates, or stacking faults act as sites for trapping impurities during high-temperature annealing [189, 190]. However, this is not suitable for solar cell production as unintended recombination centers, such as metallic decorated oxide precipitates, can be introduced.

External gettering involves the use of external layers or processes to attract and remove metallic impurities from the bulk silicon, typically moving them to regions away from the active device area [191, 192]. In homojunction solar cell production, the phosphorus (top) and aluminum (back) diffused layers bind the metallic impurities during the diffusion process and reduce their recombination activity by forming electrically inactive compounds.

#### **2.3.2** Surface recombination and passivation methods

Surface passivation is a crucial technique for reducing recombination losses in modern solar cell architectures by neutralizing dangling bonds or creating electric fields to repel carriers (detailed in Section 2.1.2). Figure 2.9. presents the different nanostructures of silicon surface for (100) and (111) orientation.

In PERC solar cells, surface recombination is addressed by applying dielectric passivation layers on the rear side as well. Atomic layer deposited (ALD) aluminum oxide with a silicon nitride capping layer is used to saturate dangling bonds and create a field effect that repels minority carriers, reducing recombination [193–195]. These layers are partially opened to create electrical contact with the silicon wafer. Limitations such as Auger recombination in the heavily doped emitter regions restrict PERC efficiencies as well.

In TOPCon solar cells, the recombination lifetime benefits significantly from the use of advanced passivated back contact [58]. A thin tunnel oxide layer and a doped polysilicon layer



Figure 2.9: Silicon surface with  $P_b$ ,  $P_{b0}$  and  $P_{b1}$  dangling bond centers. Some bonds are terminated by O or H atoms. From *Semiconductor Material and Device Characterization*, Dieter K. Schroder [138].

form the key structural elements at the rear side, minimizing recombination at the contact interface. The tunnel oxide passivates the full silicon surface while the polysilicon layer maintains excellent conductivity with minimal recombination losses. The longer carrier lifetimes associated with these technologies lead to higher and improved performance under real-world conditions.

SHJ solar cells achieve some of the highest efficiencies among commercial technologies [2]. These cells utilize thin amorphous silicon layers to minimize surface recombination. The amorphous silicon acts as an exceptional passivating layer, virtually eliminating surface recombination losses [56, 57]. Moreover, SHJ cells avoid direct metal contacts on silicon, reducing recombination at the interfaces. TOPCon and SHJ cells use thin passivation layers over the entire wafer surface, so these technologies are collectively known as passivated contact cell types.
## Chapter 3 Samples and Experimental methods

This chapter provides a comprehensive overview of the experimental techniques and corresponding measurement tools employed to investigate key recombination properties of silicon materials. The primary focus is on charge carrier recombination lifetime measurement techniques applied to various sample types: silicon ingots, wafers, and small-size samples for research and development purposes. Additional measurement techniques, such as resistivity measurements and thickness characterization, are utilized for proper calibration of lifetime measurement tools and to ensure an accurate evaluation. The preparation methods and main properties of the investigated samples are also discussed.

### **3.1** Microwave-detected photoconductance decay ( $\mu$ PCD)

Microwave-Detected Photoconductance Decay ( $\mu$ PCD) is a well-established method for PCD measurements [18, 139]. This technique is widely employed in the semiconductor industry for quality control, process monitoring, and characterization of silicon wafers. The operation of Semilab commercial  $\mu$ PCD systems based on carrier generation induced by a short pulse of 904 nm laser light, which increases the conductivity of the material.

Following the light pulse, the decaying conductance is monitored by detecting the microwave reflectivity of the sample. This is accomplished using a ring-shaped microwave antenna operating near its resonance frequency. At this predefined frequency, the change in reflected power during the decay is approximately proportional to the change in the conductance of the wafer. Consequently, the measured microwave reflectivity decay is fitted with an exponential curve, and the resulting time constant is recorded as the measured effective lifetime.

The compact measuring sensor includes all microwave circuit components, as shown in Figure 3.1. The laser pulse is guided from the semiconductor laser source to the MW antenna via an optical fiber. The measured signal is then transferred to a data acquisition card. To enhance the signal-to-noise ratio, multiple curves are recorded and averaged by this card for each measurement.

The  $\mu$ PCD measurement can be integrated into various characterization platforms optimized for specific target applications. For this study, I used two Semilab products utilizing the  $\mu$ PCD technique for different purposes.



Figure 3.1: Schematic illustration of the Semilab  $\mu$ PCD measurement setup used for the characterization of silicon wafers. Source: Semilab Engineering

### **3.1.1** The $\mu$ PCD carrier lifetime mapping system

The WT-2000 is a table-top measurement platform capable of performing a variety of measurements on solar cells and silicon wafers, including  $\mu$ PCD (Figure 3.2.). The system is typically used to record maps by scanning the sample surface with the measurement head. This approach enables the identification of lateral inhomogeneities and contaminated areas.

### **3.1.2** QSS- $\mu$ PCD measurement system to record $\tau(\Delta n)$

Semilab WT-1200A is a single-point, table-top system optimized for injection-leveldependent carrier lifetime measurement of photovoltaic wafers and solar cells. It employs the QSS- $\mu$ PCD technique, implemented using multiple illumination sources and the  $\mu$ PCD measuring sensor. The schematic of the measurement setup is illustrated in Figure 3.3.

The WT-1200A enables QSS- $\mu$ PCD measurement from both sides of the sample. The topside option consists of a regular 904 nm pulsed laser and a halogen lamp for steady illumination. Both sources are guided through the measuring head with optical fiber, resulting in a small spot ( $\approx 1 mm$ ) illumination.

The rear-side option provides an advanced QSS- $\mu$ PCD measurement using large spot illumination. In addition to the regular 904nm pulsed laser, a second, 980nm continuous wave second laser is used to create steady state conditions. The adjustable position of the optical fiber allows the enlargement of the laser spot size up to 15 mm. It reduces the effect of lateral spread3.2. LASER AND EDDY-CURRENT-SENSOR-BASED PHOTOCONDUCTANCE DECAY (E-PCD) MEASUREMENT



Figure 3.2: Semilab WT-2000 measurement platform used for lifetime mapping of silicon wafers. Source: Semilab

ing during the measurement. This feature mitigates potential distortions in measured lifetime for samples with high diffusion length. Consequently, the lower measurement option provides very accurate measurements on samples with long carrier lifetime as well [145].

### Laser and eddy-current-sensor-based photoconductance 3.2 decay (e-PCD) measurement

This section provides a detailed explanation of the eddy-current-sensor-based photoconductance measurement technique, which is the primary characterization method used in this thesis. In contrast to the flash-lamp-based QSSPC technique, Semilab tools utilize lasers as primary illumination sources. This technique, known as laser e-PCD or simply e-PCD, allows for rapid and non-destructive assessment of carrier dynamics within silicon ingots and wafers, providing information regarding the recombination lifetime, defect density, material quality, and the charge carrier mobility.

The eddy-current detection is based on the interaction between an alternating magnetic field and free charges (electrons or holes) in a conductive material (Section 2.2.3. An oscillating magnetic field near the material surface induces circulating currents (eddy currents) within the material. These currents generate their own magnetic fields, which oppose the original magnetic field according to Lenz's law. The magnitude of eddy currents and their interaction with the magnetic field is influenced by the electrical conductivity (or resistivity) of the material as well as its thickness and other electromagnetic properties.

In Semilab systems, an alternating current (AC) is passed through a coil placed near the sample, and the response of the eddy currents is measured through changes in the impedance of the



Figure 3.3: Schematic illustration of QSS- $\mu$ PCD measurement setup realized in Semilab WT-1200A equipment. From: *Improved QSS-\muPCD measurement with quality of decay control: Correlation with steady-state carrier lifetime*, Marshall Wilson *et al.* [145].

coil. The depth at which the electromagnetic field penetrates and induces eddy currents depends on the skin depth, determined by the electrical resistivity of the material and the frequency of the applied magnetic field:

$$\delta = \sqrt{\frac{2\rho}{\mu\omega}},\tag{3.1}$$

where  $\rho$  is the resistivity of the material,  $\mu$  is the permeability (for silicon, it is almost equal to vacuum permeability), and  $\omega$  is the angular frequency of the applied field. The resistivity of silicon typically used in solar cell applications ( $\approx 1 \Omega$ cm) results in a skin depth of around 1 cm, which implies that the entire depth of thin wafers ( $W < 200 \,\mu$ cm) can be properly sensed, while it is questionable for thick samples, ingots.

Electrically chopped CW lasers are used as primary illumination sources in these systems. The use of lasers offers several benefits due to their good controllability, good beam propagation qualities and monochromatic nature. The main advantage is that lasers can be switched very quickly, allowing measurement and evaluation of all relevant samples with  $\tau > 1 \,\mu$ s using the pure PCD method (Section 2.2.3). In addition, variable pulse length and energy enable to set different steady-state conditions within the samples. The monochromatic nature simplifies calculations involving penetration depth, supporting wavelength optimization for different systems.

For carrier lifetime measurements of silicon ingots and slugs of a few cm thickness, I used the Semilab WT-1200IL tool [21]. This non-contact, hand-held device employs the e-PCD technique optimized for rapid characterization of monocrystalline silicon ingots in industrial environments.

This tool also uses an electrically chopped CW laser for the photogeneration of charge carriers. The lower limit of detectable characteristic decay time is determined by two factors: the laser switching time and the response time of the RF-circuit. Using this hardware configuration,



Figure 3.4: Semilab WT-1200IL carrier recombination lifetime tool for silicon ingots. Source: Semilab

we were able to reliably record photoconductance decay (PCD) times as short as 800ns on a highly contaminated silicon sample. Consequently, this system can reliably characterize samples with carrier lifetimes from  $2 - 3 \mu s$  using transient mode evaluation. Further details about the illumination source are presented in Chapter 5, as the optimization of the tool was based on my simulation results. The system is presented in Figure 3.4.

### **3.3** Carrier lifetime measurement setup for R&D purposes

In the research laboratory of the Physics Department, András Bojtor developed a carrier lifetime measurement setup allowing to apply wide range of measuring parameters [23, 24]. In this setup, the sample is placed on a coplanar waveguide (CPW) [196]. The CPW has a conducting strip on the front surface, where the measuring signal propagates. This strip is isolated from other grounded conducting areas on the surfaces by a thin gap. An electromagnetic field is formed in the gap, which is suitable for investigating the reflectance of a semiconductor sample. The schematic drawing of the CPW with its electromagnetic field is depicted in Figure 3.5.

The CPW is positioned on the cold finger of a helium-based cryostat (M-22, CTI-CRYOGENICS), capable of cooling down to 10 K. A photo of this setup is shown in Figure 3.6. The sample is optically accessible through a window at the top of the cryostat. Multiple laser sources can be used for sample excitation based on its optical properties (band gap, absorption depth) and the targeted measurement method.

For PCD measurements, a Q-switched diode-pumped solid-state laser (NL201-2.5k, Ekspla) can be applied at two wavelengths, 532 nm and 1064 nm. The short (< 10 ns) and high energy (0.3 mJ) laser pulses generate a large density of excess carriers even in low lifetime samples, while the high repetition rate (up to 2500 Hz) helps improve the signal-to-noise ratio. To synchronize the laser pulses and data acquisition, we split the laser beam using a thin glass sheet



Ground (on backside of substrate)

Figure 3.5: Coplanar waveguide (CPW) used as a microwave sensor. From *RF and Microwave Engineering: Fundamentals of Wireless Communication*, Frank Gustrau [197].



Figure 3.6: CPW with a Si sample placed on the cold finger of the cryostat.

and directed the light to a photodetector (DET36A/M, Thorlabs). This detector is fast enough (time resolution up to  $\approx 14$  ns) to detect the short laser pulses of both wavelengths.

We also used a CW laser for steady-state measurements with a wavelength of 1064 nm.

While most components enable PCD measurements in a wide frequency range, we used microwave components in the presented studies. The block diagram of this microwave and optical system is depicted in Figure 3.7. We applied a 10 GHz, 13 dBm microwave signal using a microwave source (MKU LO 8-13 PLL, Kühne GmbH). This signal is split by a hybrid coupler (Micronde R433721) into two parts. The reference part goes to the IQ mixer through a rectangular waveguide-based phase shifter (DKX 1). The other part is directed towards the sample through an isolator (T-8S43U-20, Teledyne Microwave Solutions) to prevent reflected signals from returning to the generator. After the isolator, a rectangular waveguide-based "magic tee" is used with a phase shifter (DKX 1) and an amplitude modulator (TZC 504 Variable Attenuator, TKI). Both are rectangular waveguides with a movable wall. The purpose of this subsystem is to eliminate or minimize the dark reflectance from the CPW and the sample. This way, only the



Figure 3.7: Block diagram of the measurement system. For PCD measurements, we only use the IQ mixer and the oscilloscope for data acquisition. From András Bojtor's PhD Dissertation [198].

photogeneration-based signal is directed to the IQ-mixer. In some cases, this signal is amplified with a low-noise amplifier (LNA, made by Janilab Inc.) to increase the signal-to-noise ratio.

The IQ mixer (IQ-0618LXP, Marki Microwave Inc.) is used to create the difference signal between the reference signal and the signal reflected from the sample. To prevent DC signals from reaching the mixer, two rectangular connectors (SM-WR90) insulated by plastic sheets are used before both inputs. The I and Q outputs of the IQ mixer are then recorded with a high-bandwidth digital oscilloscope (Tektronix MDO3024). The signal from the photodetector is also directed to this oscilloscope to trigger the measurement.

### **3.4** Evaluation of PCD curves

This section presents a comprehensive overview of the standard evaluation procedure for the Photoconductance Decay (PCD) method, applicable across various measurement setups and detection techniques. While certain applications, such as rapid silicon wafer mapping, may utilize a simplified approach of extracting a single lifetime value through exponential function fitting, more sophisticated analyses of recombination properties necessitate the determination of the excess-carrier-concentration-dependent recombination lifetime function,  $\tau(\Delta n)$ .

The importance of obtaining the  $\tau(\Delta n)$  function is particularly pronounced when examining exotic sample types, where recombination lifetime can exhibit order-of-magnitude variations during decay, primarily due to charge carrier trapping phenomena. This function provides

crucial insights into the complex recombination dynamics occurring within the material.

The objective of this evaluation procedure is to derive the  $\tau(\Delta n)$  function from the observed decay curve. This process is presented in Figure 3.8. step-by-step for a high lifetime n-type sample. In the standard PCD measurement protocol, multiple sequential excitation pulses are typically employed to enhance measurement accuracy. The subsequent signal decay curves are averaged, resulting in a significant improvement in the signal-to-noise ratio. The number of averages is typically optimized based on the measured carrier lifetime and the signal magnitude to balance the measurement time and the accuracy. The equilibrium state signal is recorded prior to excitation as it provides the conductivity and doping level.

The determination of carrier lifetime necessitates the numerical differentiation of the decay curve, as described by Equation 2.34. With regard of the sensitivity of numerical differentiation to noise, the elimination of external disturbances is crucial for an accurate evaluation. To address this challenge, I developed an adaptive averaging method that minimizes the distortion effects of periodic noise while preserving the integrity of the decay signal (blue triangles in Figure 3.8a.). This enables the robust evaluation of carrier lifetime, particularly in cases of significant periodic noise or when dealing with samples exhibiting complex decay behaviors.

The subsequent phase in the evaluation process involves the computation of excess charge carrier concentration ( $\Delta n$ ) decay from the averaged signals as it is presented in Figure 3.8b.

For eddy current measurement techniques, the measured signal can be easily calibrated to the sheet conductance ( $\sigma_s$ ) of silicon wafers. Recent research by Black et al. [199] demonstrated that accurate calibration requires the consideration of the wafer thickness (*W*), particularly when it approaches the skin depth of the eddy currents. To avoid the inaccuracy arising from the skin depth correction, calibration wafers of similar thickness to the sample under investigation were employed.

In the investigation of novel materials, standardized resistance samples of equivalent dimensions are often unavailable for precise calibration. In such cases, a common approach involves determining a single lifetime value by fitting an exponential function to the measured decay curve. However, this method can be significantly compromised if the antenna's calibration function exhibits non-linear behavior.

To address these challenges and enhance measurement accuracy for non-standard samples, I developed a novel calibration method combining steady-state and transient measurement methods. This innovative approach aims to mitigate the effects of non-linear antenna responses and improve the reliability of lifetime measurements for a wide range of materials and sample configurations. The detailed exposition of this new calibration technique, including its theoretical foundation, experimental validation, and practical implementation, is presented in Chapter 4.

The standard formula to calculate the density of excess carriers from the sheet conductance is:

$$\Delta n = \frac{\Delta \sigma_{\rm s}}{W \cdot e \cdot \mu_{\rm sum}(\Delta n, N_{\rm dop})},\tag{3.2}$$

where *e* is the electron charge,  $\Delta \sigma_s$  is the excess sheet conductance, and  $\mu_{sum} = \mu_e + \mu_h$  is the sum of electron and hole mobilities.

The charge carrier mobility is not a constant parameter but rather a function of the dopant atom concentration and the excess charge carrier density. Consequently, an iterative calcula-



Figure 3.8: Evaluation procedure of photoconductance decay measurement for a high lifetime n-type sample with  $1e15 \text{ cm}^{-3}$  doping concentration. (a) Adaptive averaging of measured data points reduces the distortion effects of external noise. (b) Decay of  $\Delta n$  is computed using mobility data. The recombination lifetime  $\tau$  curve, calculated from the numerical differentiation of the  $\Delta n(t)$ , in the function of (c) time and (d) injection level.

tion is used to accurately determine  $\Delta n$ . The mobility shift of dopant-related free carriers also contributes to the observed conductance in the injected case [200]. Considering this effect, the expression for  $\Delta n$  is modified as follows:

$$\Delta n = \frac{\sigma_s / W - N_{\rm dop} \cdot e \cdot \mu_{\rm maj}(N_{\rm dop}, \Delta n)}{e \cdot \mu_{\rm sum}(N_{\rm dop}, \Delta n)},\tag{3.3}$$

where  $N_{dop}$  is the doping concentration and  $\mu_{maj}$  is the mobility of majority charge carriers.

Numerous theoretical and empirical models are available to determine the mobility of electrons and holes in silicon [89, 91–100]. Among these, I employ Klaassen's semi-empirical model [98–100], which not only accounts for carrier concentration but also incorporates temperature dependence, providing a more accurate representation of charge carrier mobility across a wide range of operating conditions.

The determination of  $\Delta n$  in thick silicon samples is challenging, as it will be discussed in Chapter 5.

The concluding phase of the evaluation process involves the numerical differentiation of the  $\Delta n(t)$  curve, followed by the computation of the recombination lifetime corresponding to each data point based on Equation 2.34 (Figure 3.8c and d).

### **3.5** Further measurement methods

### **3.5.1** 4-point probe sheet resistance measurement

The four-point probe (4PP) method is a widely used technique to measure the resistivity of semiconductor materials [138]. The main applications of this technique is the resistivity measurement of thick samples and wafers with known thickness, while for thin films it measures the sheet resistance value. The four-point probe setup consists of four equally spaced probes, that are brought into contact with the surface of the semiconductor sample. The two outer probes are used to source DC current, while the two inner probes measure the resulting voltage drop with very large input resistance. In this way, the effect of contact resistance can be eliminated from the measurement result. The resistivity or sheet resistance is calculated using the measured current, voltage, and geometric factors. For thick samples (t >> d, where t is the sample thickness and d is the probe spacing) the resistivity is given as:

$$\rho = 2\pi d \frac{V}{I},\tag{3.4}$$

where *I* is the applied current and *V* is the measured voltage. For thin films ( $t \ll d$ ) the primary measured quantity is the sheet resistance:

$$\rho_s = \frac{\pi}{\ln(2)} \frac{V}{I}.\tag{3.5}$$

The Semilab FPP-1000 system was applied using a conventional 4PP head from Jandel inc. to calibrate our eddy current sensor used in the e-PCD technique.

### 3.5.2 Thickness measurement

The eddy current signal is calibrated to the sheet conductance of silicon wafers. Thus, accurate knowledge of the thickness is necessary to determine the charge carrier concentration. For this purpose, we measured all silicon wafers with Mitutoyo Litematic VL-50S thickness tester.

### **3.6** Studied samples

For my research, I used industrially relevant silicon ingots and thin wafers sliced from them. In the solar industry, both gallium-doped p-type and phosphorus-doped n-type silicon wafers are used to manufacture solar cells. The former is typically characterized by lower resistivity and recombination lifetime due to the small segregation coefficient of Ga atoms [201]. For n-type materials, the continuous improvement in the purity of manufacturing processes is accompanied by an increase in bulk recombination lifetime. This, along with the increased charge carrier concentration, poses new challenges for accurate determination of  $\tau(\Delta n)$ .

Overall, it can be stated that the industry and associated research and development require lifetime measurement methods that operate reliably across a wide range of lifetimes and resistivities. The investigated silicon crystals and cell manufacturing processes were provided by leading Si wafer and solar cell manufacturers, ensuring that our results are relevant for state-of-the-art Si materials.



Figure 3.9: Thickness of 1 cm x 1 cm adjacent silicon wafers after etching time  $t_{\text{etching}}$ .

### Silicon wafers for validation of novel calibration method

The primary objective of the low-temperature measurement system developed in the university research laboratory is to investigate novel materials. However, prior to this, I analyzed the accuracy of the system through calibration measurements, for which silicon wafers, with known charge carrier properties, were an obvious choice.

For the calibration study presented in Chapter 4, I cut adjacent pieces of 1 cm x 1 cm size from the center of long-lifetime  $\tau_b > 1$  ms semiconductor-grade Si wafers using a diamond cutter, ensuring the comparability of results. The doping of the p-type wafer is  $N_{dop} = 1.5 \cdot 10^{15}$  cm<sup>-3</sup>, while for the n-type wafer, it is  $N_{dop} = 5 \cdot 10^{14}$  cm<sup>-3</sup>. The relatively low  $N_{dop}$  reduces the influence of the Auger recombination. The initial thickness of the samples was 760  $\mu$ m.

The samples were etched in a mixture of HF, HNO<sub>3</sub>, and CH<sub>3</sub>COOH acids to obtain wafers of different thicknesses. The thickness of the samples removed from the acid mixture at different times is shown in Figure 3.9. Following the acid etching, the surface is unpassivated, therefore the recombination is very intense there, resulting in an estimated surface recombination velocity of  $S = 10^6 \frac{\text{cm}}{\text{s}}$  [202].

### Thick silicon samples for $\tau_{\rm b}$ characterization

In Chapter 5, I examined 1 - 2 cm thick slugs cut from ingots, as is most commonly done in industrial quality control. Initially, I studied Ga-doped samples as follows. Due to the low segregation coefficient of Ga atoms, the resistivity and lifetime can vary significantly within a single ingot. Furthermore, the metallic impurities accumulate in the tail sections of the ingots [186], which can significantly reduce the recombination lifetime in p-type silicon. Therefore, I examined slug samples from the seed-end (or top), middle, and tail-end sections of the ingots (Figure 3.10a.).

These ingots are most commonly manufactured using the Recharged Czochralski (RCz) process for cost-effectiveness, during which up to 10 ingots can be pulled from the same crucible keeping some melt before re-filling. Therefore, this results in varying levels of contamination and recombination lifetime between ingots. I investigated 4 ingots within a single RCz process (Figure 3.10b.).



Figure 3.10: (a) 1 - 2 cm thick slugs from different sections of Ga-doped Si ingot were examined. (b) The ingots were selected from different pulls of the Recharged Czochralski (RCz) process. (c) Adjacent wafers passivated by a stock of a:Si layers served as reference  $\tau_b$  samples.

In the next step, we studied n-type slugs, although we have managed to obtain only a few industrial samples in the high carrier lifetime range, so far. However, these already provided significant information regarding the accuracy of our method.

### **Passivated cell structures**

Thin wafers (approximately  $150 \,\mu$ m in thickness) were examined using the e-PCD measurement setup optimized for silicon wafers. These wafers were passivated using hydrogenated amorphous silicon (a-Si:H) layers providing excellent surface passivation. The passivation process involved depositing a thin intrinsic a-Si layer on the p/n-type wafer, followed by a thinner n/p-type a-Si layer. This structure, symmetrically formed on both sides (Figure 3.10c.), provides optimal passivation of the wafer. Moreover, the low-temperature deposition of a-Si layers ensures the preservation of the bulk properties. Such "lifetime samples" do not operate as solar cells, but are suitable to obtain  $\tau_b$  avoiding the distorting effects of surface recombination.

Wafers were extracted from the immediate vicinity of the slugs presented in the previous subsection to establish a lower limit for  $\tau_b$ , owing to the superior surface passivation quality. These results are presented in Section 5.3.3. Furthermore, one of these wafers and one high-lifetime n-type wafer were analyzed using the combined lifetime measurement technique, with the results detailed in Section 6.4.

### Chapter 4

# Calibration and validation of $\mu$ PCD measurement setup for research purposes

In this chapter, I summarize my contribution to the development of low-temperature PCD measurement setup in the research laboratory of BME. The purpose of the measurement setup (referred to in Section 3.3) is to investigate the photoconduction of novel materials (particularly perovskite-structured materials) as a function of temperature. During the investigations conducted with this measuring device, it was frequently questioned whether the observed strongly non-exponential decay was a characteristic of the recombination properties of the sample or an artifact of the measurement system [24]. To resolve this, I developed and performed a validation measurement procedure on silicon samples and, recognizing the nonlinear behavior of the system, I devised a novel calibration method. Through these validation measurements, I determined the ambipolar diffusion coefficient  $D_{amb}$  of silicon at low  $\Delta n$  down to 120 K. The corresponding publication is under preparation [O1].

There are no well-defined sample standards for recombination lifetime measurements. For thick, bulk samples, the measured lifetime is greatly influenced by charge diffusion and surface recombination, as I explain in more detail in Chapter 5. Even for thin wafers, alongside bulk recombination  $\tau_b$ , the surface recombination velocity *S* and sample thickness *W* are equally important parameters in the measured lifetime. In this case, using the approximation of Tüttő et al. [139], the measured effective lifetime  $\tau_{eff}$  is given by:

$$\frac{1}{\tau_{\rm eff}} = \frac{1}{\tau_{\rm b}} + \frac{1}{\frac{W}{2S} + \frac{W^2}{\pi^2 D_{\rm amb}}}.$$
(4.1)

In expression 4.1, the parameters  $\tau_b$  and *S* are difficult to control, so to select an ideal test sample, the uncertainty in these must be minimized. For this purpose, I used long  $\tau_b$  silicon wafers with high *S*, which is ensured by the etched surface. Figure 4.1. shows that above  $\tau_b = 1$  ms and in the *S* range of  $10^5 - 10^6$  cm/s corresponding to the as-cut surface after slicing, the characteristic time associated with the diffusion process  $\tau_{diff}$  determines  $\tau_{eff}$ , thus enabling reproducible measurements in which:

$$\tau_{\rm eff} \approx \tau_{\rm diff} = \frac{W^2}{\pi^2 D_{\rm amb}}.$$
(4.2)



Figure 4.1: Measurable effective lifetime  $\tau_{eff}$  as a function of bulk lifetime  $\tau_b$  for various wafer thicknesses. At high  $\tau_b$  and *S*, the diffusion-limited lifetime is determinant.

The thickness W in this expression is easily measurable, while  $D_{amb}$  can be determined from literature mobility models and the Einstein relation (Equation 2.7). I used the Klaassen model [98–100] describing the charge-carrier mobility in silicon, which is proven to be reliable (see later in Chapter 6).

To validate the accuracy of the measuring device, I performed measurements on 1 cm x 1 cm pieces of a silicon wafer of different thicknesses using a 1064 nm laser. The detailed properties of the samples and their preparation method are described in Section 3.6.

Figure 4.2a. shows a decay curve measured on a  $665 \,\mu\text{m}$  thick sample, and Figure 4.2b. depicts the apparent lifetime  $\tau_{app}$  calculated from it without the calibration of the measured signal:

$$\tau_{\rm app}(\Delta V) = \frac{\Delta V}{-\frac{\partial \Delta V}{\partial t}}.$$
(4.3)

It is evident that at the beginning of the transient, the decay exhibits a strongly non-exponential character, characterized by an increased  $\tau_{app}$  at high  $\Delta V$ . After a short time period, the exponential nature of the decay is restored, which corresponds to the characteristic time of the diffusion process.

Although  $D_{amb}$ , and thus  $\tau_{diff}$ , may change at higher injection levels, this would at most result in a change of a few tens of percent, which would not justify the order of magnitude increase in lifetime. We thus identified the non-linear response of the microwave system as the cause of the observed phenomenon, the system thus requires further calibration. However, since the development aim is to examine various experimental samples, which often have the most



Figure 4.2: (a) Non-exponential decays are observed at high  $\Delta V$ , (b) resulting in a significantly increased  $\tau_{app}$ . After a time period, the exponential decay is restored with an exponent corresponding to  $\tau_{diff}$ .

diverse structures and geometries, the procedure used for silicon cannot be applied here.

### 4.1 Novel calibration method

In this section, I present a novel calibration method that is suitable for measuring systems with non-linear responses or for studying novel materials. As part of this, I performed a quasi-steady-state photoconductance (QSS-PC) measurement prior to the transient measurement, during which I illuminated the sample with a harmonically modulated CW laser. The laser wavelength was  $\lambda = 1064$  nm, similar to the pulse laser used in the transient measurement, to ensure identical absorption properties in both measurements. The modulation angular frequency was low enough for the sample to remain in a quasi-steady state throughout the measurement ( $\omega << 1/\tau_{eff}$ ). Part of the light was coupled to a photodiode. The maximum laser power  $P_{max}$  was also measured with a power meter, making it possible to calculate the generation rate in the sample:

$$G_{\rm qss}(t) = \frac{P_{\rm max}}{2\frac{hc}{\lambda}A_{\rm spot}W} (1 + \sin(\omega t)), \qquad (4.4)$$

where *h* is the Planck constant, *c* is the speed of light, and  $A_{spot} = 1 \text{ mm}^2$  is the size of the illuminated spot.  $G_{qss}(t)$  and the microwave response signal  $\Delta V$  are shown in Figure 4.3a. during the measurement. Since the illuminated spot is very similar to the one used during the transient measurement due to the common optical elements, the same signal level corresponds to the same injection level. This allows for an estimation of the  $G_{qss}(\Delta V)$  relationship (Figure 4.3b.).

Exploiting the fact that the sample goes through the same states during both transient and QSS measurements, we can equate the carrier lifetimes corresponding to each signal level:

$$\tau_{\rm PCD}(\Delta V) = \tau_{\rm QSS}(\Delta V). \tag{4.5}$$



Figure 4.3: (a) Quasi-steady-state photoconductance measurement with slowly harmonically modulated illumination intensity. (b) The simultaneous measurement of light intensity and  $\Delta V$  enables the determination of a calibration function.

$$\frac{\Delta n}{-\frac{\partial \Delta n}{\partial t}}\bigg|_{V^*} = \frac{\Delta n}{G}\bigg|_{V^*}.$$
(4.6)

Utilizing the equality of the denominators:

$$G(V^*) = -\frac{\partial \Delta n}{\partial t}\bigg|_{V^*} = -\frac{\partial \Delta n}{\partial \Delta V}\bigg|_{V^*} \cdot \frac{\partial \Delta V}{\partial t}\bigg|_{V^*}.$$
(4.7)

The derivative of the  $\Delta n(\Delta V)$  calibration function appears in the expression:

$$\frac{\partial \Delta n}{\partial \Delta V}\bigg|_{V^*} = \frac{G(V^*)}{-\frac{\partial \Delta V}{\partial t}\bigg|_{V^*}}.$$
(4.8)

Figure 4.4a. shows that up to a signal of about 1 mV, this derivative is constant, so the calibration function is linear. However, after this point, a strong non-linearity appears. Numerically integrating the data, the derivative yields the calibration curve (Figure 4.4b.).

Using the calibration, the  $\Delta n(t)$  decay curve can be obtained (Figure 4.5a.), which now shows a much more exponential character as expected. The  $\tau(\Delta n)$  curve calculated from this decay exhibits a similar character to  $\tau_{\text{diff}}(\Delta n)$  derived from the Klaassen mobility model (Figure 4.5b.). The  $\Delta n$  dependence seen here is mainly caused by the dependence of  $D_{\text{amb}}$  on  $(\Delta n)$ .

By applying this method, the order-of-magnitude change observed in the  $\tau_{app}(\Delta V)$  function has disappeared. The method can be further refined if both the transient and quasi-steady-state measurements are performed with the same laser source, and thus the same illuminated volume.



Figure 4.4: (a) The novel calibration method enables the determination of the derivative of the  $\Delta n(\Delta V)$  calibration function. (b) After numerical integration, the calibration function is constructed.



Figure 4.5: (a) After accurate calibration of the signal, an exponential  $\Delta n(t)$  decay is observable. (b) The  $\tau_{\text{eff}}$  shows good similarity to the trend of  $\tau_{\text{diff}}$  calculated by the Klaassen mobility model.

### 4.2 Validation measurements on silicon wafers

Following the calibration of the measurement system, I performed measurements on p- and n-type samples of various thicknesses to examine the accuracy of the system. Each curve was evaluated around the injection level of  $\Delta n \approx 10^{14} \text{ cm}^{-3}$ , where a reliable value can still be extracted from the decay curve, but  $D_{\text{amb}}(\Delta n)$  does not change significantly. Figure 4.6. shows that  $\tau_{\text{eff}}$  indeed scales with the square of the thickness for both samples. The slope of the curve, after unit conversions, gives the  $D_{\text{amb}}$  corresponding to the low injection level.

$$D_{\text{amb,n}}(\Delta n = 10^{14} \,\text{cm}^{-3}) = \frac{1}{7.91 \cdot 10^{-3} \pi^2} \frac{\text{cm}^2}{\text{s}} = 12.8 \frac{\text{cm}^2}{\text{s}}$$
(4.9)

$$D_{\text{amb},p}(\Delta n = 10^{14} \,\text{cm}^{-3}) = \frac{1}{3.20 \cdot 10^{-3} \pi^2} \frac{\text{cm}^2}{\text{s}} = 31.6 \frac{\text{cm}^2}{\text{s}}$$
(4.10)



Figure 4.6: The measured  $\tau_{eff}$  is indeed proportional to the square of sample thickness for a p-type sample with  $N_{dop} = 1.5 \cdot 10^{15} \text{ cm}^{-3}$  and an n-type sample with  $N_{dop} = 5 \cdot 10^{14} \text{ cm}^{-3}$ . The slope of each curve yields the low injection  $D_{amb}$  with good agreement to the Klaassen mobility model [98, 99].

According to the Klaassen mobility model, these values are  $D_{\text{amb,n}}(\Delta n = 10^{14} \text{ cm}^{-3}) = 13.18 \frac{\text{cm}^2}{\text{s}}$  and  $D_{\text{amb,p}}(\Delta n = 10^{14} \text{ cm}^{-3}) = 30.33 \frac{\text{cm}^2}{\text{s}}$ , which in both cases represents a difference of less than 5%, which confirms the precision of the PCD measurement if this self-consistent calibration method of the recorded signal is used.



Figure 4.7: The low injection  $D_{\text{amb}}$  fits well with the value from the Klaassen mobility model for an n-type sample with  $N_{\text{dop}} = 5 \cdot 10^{14} \text{ cm}^{-3}$  and  $W = 760 \,\mu\text{m}$ , across a wide temperature range.

I also performed temperature-dependent measurements on a  $W = 760 \,\mu$ m thick sample, the results of which are shown in Figure 4.7. It is clearly visible that down to 120 K, our measurement results closely follow the diffusion-limited lifetime derived from the Klaassen model [98, 99]. Below this temperature, a different process begins to dominate in the measurable life-

time. This is likely caused by the activation of a charge trapping process, but investigating this was not the aim of this work.

My research in the BME labs led to a method which provides the calibration of the recorded signal to  $\Delta n$  in a self-consistent way. The reliability of this method was confirmed using etched silicon samples, since they have a very precisely known effective carrier lifetime. However, the method is applicable for any other novel semiconductor materials with less known material properties. A further important feature of this technique is its universality. It is a common issue with microwave-detected PCD measurements, that  $\Delta n$  can be hardly determined if the illuminated area is smaller than the sensed surface area, for which the MW reflectance signal is typically calibrated [203]. However, the adaption of my self-consistent method is not limited by such phenomena as the calibrating quasi-steady-state measurement is recorded on the same illuminated surface. This method can be applied for other  $\Delta n$  sensing principles as well.

### Chapter 5

### **Recombination dynamics of photo-induced carriers in thick silicon samples**

This Chapter summarizes my own work aimed at improving the lifetime measurements on bulk Si samples motivated by direct benefits in industrial applications. I investigated the influence of measuring system parameters to the accuracy, and found an optimal way to mitigate surface recombination. These results were published in [O2]. The most important result is the unprecedentedly accurate determination of  $\tau_b(\Delta n)$ , which was achieved by a simulation-based evaluation process for PCD transient. It enables to characterize the bulk recombination processes of very high quality Si ingots which is not possible in other ways. The corresponding publication is under preparation [O3].

Monocrystalline silicon ingots produced by the Czochralski (Cz) method constitute the primary raw material for solar cell manufacturing. Characterizing the purity of these ingots is of crucial importance during the initial production phase to ensure that only high-quality material is processed further, while contaminated sections are identified and discarded. The detection of metallic impurities is particularly critical for integrated circuit applications [204, 205], as these contaminants can precipitate during high-temperature device fabrication processes, potentially compromising the performance. Their detection is also very important for solar cell applications, since they act as strong SRH recombination centers significantly reducing the efficiency of solar cells. This consideration becomes even more significant in the context of the recharged Czochralski (RCz) method, where multiple crystal rods are consecutively pulled from the same melt [162]. Moreover, the accelerated crystal growth rates employed in RCz can lead to increased incorporation of oxygen-related defects, which may further diminish the potential efficiency of the resultant solar cells [7].

Injection-level-dependent carrier lifetime (ILDCL) measurement is a widely used characterization technique for crystalline silicon. It is generally accepted that accurate bulk carrier lifetime values can be acquired only on wafers with very high-quality surface passivation. To obtain recombination parameters identical to as-grown material properties, it is mandatory to use low-temperature surface passivation methods, such as wet-chemical passivation or deposition using low-temperature processes like amorphous silicon stacks. These methods, however, can be time-consuming and may not be suitable for rapid quality control for the PV industry. Measuring thick samples without surface passivation offers an alternative approach. In this case, the surface recombination can be significantly reduced, enabling the detection of carrier lifetimes close to the bulk value [21, 206, 207]. This is particularly advantageous for characterizing silicon ingots or thick slabs in production environments, where quick and non-destructive measurements are essential. The reduced impact of surface recombination in thick samples can be attributed to several factors.

Previous studies indicated that the eddy-current-based photoconductance decay (e-PCD) method is particularly well-suited for carrier lifetime measurements on silicon ingots due to its large sensitivity depth. However, the accurate evaluation of e-PCD measurements presents significant challenges.

The accuracy of the quasi-steady-state photoconductance (QSSPC) method has been investigated in numerous studies for both its transient and quasi-steady-state (QSS) modes. Swirhun et al. initially showed the temporal evolution of the excess carrier profile during the transient decay and its subsequent impact on the measured effective lifetime [207]. Subsequently, Goodarzi et al. demonstrated that the utilization of various long-pass optical filters mitigates the influence of unpassivated surfaces in both transient [208] and QSS modes [209]. Despite extensive optimization efforts, their experimental results revealed an unsatisfactory correlation between different methods within their common measurement range. Significant discrepancies, ranging from 40% to 70%, were observed across different segments of the same ingot. This observation clearly demonstrates the substantial benefit of developing a unified method capable of covering the entire relevant lifetime range. Our approach, which employs a fast-switching laser for the photoconductance decay (PCD) method, aims to realize this objective.

This chapter presents a comprehensive overview of the development of the laser and eddycurrent-based photoconductance decay (e-PCD) method for characterizing silicon ingots. Initially, we address the challenges inherent to measuring thick samples. Subsequently, we discuss the optimization of the measurement system, with particular emphasis on the illumination source. The third section introduces an integrated model that combines charge carrier dynamics and detector sensitivity to simulate the transient lifetime measurement process in its entirety. Using this sophisticated simulation, I developed an iterative evaluation procedure to extract  $\tau_b(\Delta n)$  from measured decay curves. Finally, I demonstrate the practical applicability of this method on gallium-doped RCz and high-quality n-type Cz silicon slugs, which represent the most commonly used raw materials in contemporary solar cell production.

### 5.1 Challenges of PCD measurement of thick samples

### 5.1.1 Inhomogeneous charge carrier profile and surface recombination

The measurement of thick samples is an opportunity to extract the bulk lifetime ( $\tau_b$ ), but it also introduces several challenges. The primary difficulties arise from the inhomogeneous excess carrier concentration profile ( $\Delta n(x)$ ) and its continuous evolution during the measurement. The rate of surface recombination is dependent on the  $\Delta n(x,t)$  profile. A one-dimensional approach is applicable to investigate the problem, as the illumination spot size is significantly larger than the diffusion length. To analyze the  $\Delta n(x,t)$  depth profile (where x=0 represents the front surface), the continuity equation for excess electrons must be employed, assuming  $\Delta n = \Delta p$  [115]:

$$\frac{\partial \Delta n(x,t)}{\partial t} = G(x,t) - R(x,t) + D_{\text{amb}} \cdot \frac{\partial^2 \Delta n(x,t)}{\partial x^2}, \qquad (5.1)$$

where  $D_{\rm amb}$  is the ambipolar diffusion coefficient, which accounts for the Coulomb interaction between holes and electrons (see Section 2.2.1.). Drift currents originated from an external electric field are not considered in this model, as the electric field decays within the Debye length [210] under the surface, which is below  $1 \mu m$  in moderately doped silicon. The effect of the electric field near to the surface is incorporated into the surface recombination velocity (SRV). At the sample surfaces, the SRV boundary condition (Equation 2.24) must be satisfied. Typically, the SRV on as-grown or as-cut surfaces of thick silicon samples falls within the range of  $S \approx 10^4 - 10^6$  cm/s, indicating very rapid recombination [138, 202].

In moderately doped silicon, band-to-band transitions dominate light absorption below 1100 nm wavelength. Consequently, the electron-hole pair photogeneration rate as a function of depth (G(x)) can be expressed using the absorption coefficient  $\alpha$ :

$$G(x) = (1 - R)\Phi\alpha e^{-\alpha x},\tag{5.2}$$

where R and  $\Phi$  represent the reflectance from sample surface and photon flux, respectively.

The excess-carrier dependence of  $\tau_b$  and  $D_{amb}$  increases the complexity of solving Equation 5.1. To address these challenges, a comprehensive one-dimensional simulation of the charge carrier dynamics was developed. In this simulation, the bulk recombination rate comprises the intrinsic recombination rate (utilizing the model proposed by Richter et al. [128]) and a defect-related recombination rate ( $\tau_{SRH}$ ). The  $D_{amb}(\Delta n)$  dependence is also incorporated into the simulations, considering the injection-level dependence of electron and hole mobility using data from Dannhauser [96] and Krausse [97], and the Einstein relation (Equation 2.7).

To illustrate the evolution of  $\Delta n(x)$  over time, a simulation with a constant  $\tau_{\text{SRH}} = 1 \text{ ms}$  distribution was performed. The simulation assumed the use of a 1064 nm laser source with an absorption coefficient  $\alpha = 1/0.085 \text{ cm}$  and photon flux  $\Phi = 2 \cdot 10^{19} \text{ cm}^{-2} \text{s}^{-1}$ . The simulated profiles are depicted at several time points after the termination of illumination in Figure 5.1a. Near the surface, the excess carrier concentration is very low due to the rapid surface recombination. The peak injection level is typically located a few mm below the surface. In the deeper regions, an exponential decrease is observed due to the absorption and diffusion processes.

Due to the strong inhomogeneity of  $\Delta n(x)$ , determining an average excess carrier concentration  $\Delta n_{avg}$  is challenging. Bowden suggested using a weighted mean [206]. If handling  $\frac{\Delta n(x)}{\Delta N_{total}}$  as a probability distribution function, then  $\Delta n_{avg}$  is defined as its expected or mean value:

$$\Delta n_{\rm avg} = \frac{\int_0^W (\Delta n(x))^2 dx}{\int_0^W \Delta n(x) dx}.$$
(5.3)

This approach enables the determination of a spatially averaged injection level considering the near-surface volume with high  $\Delta n$ . Such an approximation is essential for defining the injection level in thick samples for which carrier lifetime is reported, although these values may differ from those obtained from thin wafers with excellent surface passivation. A spatially averaged lifetime can be estimated based on the decay of  $\Delta n_{avg}$  providing a representative measure of the overall characteristics of the sample. It enables meaningful comparisons across different measuring parameters as it will be presented in the followings.



Figure 5.1: (a) Simulated  $\Delta n(x,t)$  depth profiles for a thick (W = 5 cm) silicon sample corresponding to  $\tau_{\text{SRH}} = 1 \text{ ms.}$  (b) The increase of  $\tau_{\text{surf}}$  during the decay due to the changing profile shape.

An effective width  $W_{\text{eff}}$  can also be defined for the average depth of the profile as the total excess carrier concentration  $\Delta N_{\text{total}}$  divided by  $\Delta n_{\text{avg}}(t)$ :

$$W_{\rm eff} = \frac{\Delta N_{\rm total}}{\Delta n_{\rm avg}} = \frac{\left(\int_0^W \Delta n(x) dx\right)^2}{\int_0^W (\Delta n(x))^2 dx}.$$
(5.4)

The  $\Delta n_{\rm avg}$  and  $W_{\rm eff}$  are depicted in Figure 5.1a. for the steady-state condition.

The surface recombination lifetime introduced in Equation 2.28 can be determined by integrating Equation 5.1 over the sample thickness during the decay phase (G = 0), which yields the decay rate of  $\Delta N_{\text{total}}$ :

$$\frac{\partial \Delta N_{\text{total}}(t)}{\partial t} = -\frac{\Delta N_{\text{total}}(t)}{\tau_{\text{b}}} + D_{\text{amb}} \cdot \left[\frac{\partial^2 \Delta n(x,t)}{\partial x^2}\right]_{x=0},$$
(5.5)

where the last term gives the surface recombination rate after division by  $\Delta N_{\text{total}}$ :

$$\tau_{\text{surf}} = \frac{\Delta N_{\text{total}}(t)}{D_{\text{amb}} \cdot \left[\frac{\partial^2 \Delta n(x,t)}{\partial x^2}\right]_{x=0}}.$$
(5.6)

Figure 5.1b. illustrates the surface recombination lifetime during the decay for the same simulation parameters. As the near-surface charge carriers recombine faster, the excess charge carrier profile shifts deeper toward the bulk, which reduces the effect of front surface recombination. The actual rate of surface recombination is strongly dependent on the  $\tau_b(\Delta n)$  function through the bulk diffusion process, since a simple correction function cannot be composed.

### 5.1.2 Varying depth sensitivity of the eddy-current detection

The evaluation of the measured PCD curves represents a significant challenge in accurately calculating the average injection level from the signal. This difficulty arises primarily



Figure 5.2: Effective depth of eddy-current detection sensitivity. Below  $3\Omega$  cm the skin depth of the probing field limits the effective depth. Above this value, the geometry of the coil does not allow deeper penetration than 0.4 cm.

from the complex interaction between the inhomogeneous charge carrier depth profile and the radio-frequency (RF) magnetic field employed to sense  $\Delta n$ . The depth of the excess carrier concentration profile,  $\Delta n(x)$ , often coincides with or exceeds the penetration depth of the RF field, requiring consideration of carrier diffusion into or out of the sensed volume. To calibrate the depth sensitivity of the coil used in the measurement, a set of thin wafers (approximately  $300 \,\mu$ m) were measured using both the eddy-current sensor and four-point-probe technique. A nearly linear relationship was observed across the relevant sheet conductance ( $\sigma_{sh}$ ) range. As the result of the calibration,  $\sigma_{sh}$  is calculated from the recorded eddy-current signal. However, for thick samples such as ingots or slabs (> 1 cm thickness), the sensitivity of the eddy-current detection is limited by the penetration depth of the RF wave. For these samples, an effective depth of sensitivity was determined as:

$$d_{\text{sense}} = \rho_{4\text{pp}} \cdot \sigma_{\text{sh,sensed}}, \tag{5.7}$$

where  $\rho_{4pp}$  is the resistivity measured by four-point-probe technique, and  $\sigma_{sh,sensed}$  is the sensed sheet conductance calculated from the recorded eddy-current signal from the thick samples, but using the sheet conductance calibration obtained using wafers.

Figure 5.2. illustrates the effective depth of sensitivity as a function of resistivity. In the low resistivity range, the effective depth increases with increasing skin depth, while above  $3\Omega$ cm, it remains constant due to the limiting factor of the coil geometry.

A particularly challenging aspect of this measurement technique is the dynamic nature of the skin depth. As the excess carrier concentration evolves during the measurement, the effective conductivity of the sample changes, leading to a dynamic skin depth effect. This results in a time-dependent penetration depth of the RF field throughout the measurement, further complicating the interpretation of the eddy-current signal. Previous studies have investigated the sensitivity of eddy-current techniques in e-PCD tools. Black et al. demonstrated the necessity of considering silicon wafer thickness due to decreasing sensitivity within the sample. They proposed a model incorporating a skin-effect based correction, though its applicability to thick samples is limited. Swirhun et al. identified a dark resistivity-dependent sensitivity depth of the RF coil in a commercial RF-PCD system [207]. While they noted the issue of decreased sensitivity depth in illuminated samples, their simulations were confined to low injection conditions to avoid distortion.

In conclusion, the accurate measurement of carrier lifetime in thick silicon samples using eddy-current detection remains a complex task. It demands a comprehensive understanding of the interplay between the inhomogeneous carrier profile, the dynamic skin depth effect, and the depth-dependent sensitivity of the eddy-current sensor. The approach presented in Section 5.3.1 is based on the parametrization of the depth sensitivity function, which accounts for the impact of the  $\Delta n(x)$  depth profile, offering a more comprehensive solution to these challenges.

### 5.2 Influence of system parameters to the e-PCD results

### 5.2.1 Impact of excitation parameters

The Semilab WT-1200I commercial e-PCD measurement tool was optimized to enhance the accuracy of  $\tau_b$  determination. At the time of development, the most commonly manufactured PERC solar cell structures utilized Ga-doped p-type silicon with typical carrier lifetimes ranging from 100 to 1000  $\mu$ s. Furthermore, emerging passivated contact structures (HJT, TOP-Con) employed high-lifetime (> 1 ms) n-type wafers. Consequently, it became necessary to determine silicon ingot lifetimes in a wide range.

Until 2020, Semilab WT-1200I system [21] applied a 980 nm laser source to illuminate the ingots in a 2.2 cm diameter spot, delivering an average photon flux of  $3 \cdot 10^{18} \text{ cm}^{-2} \text{s}^{-1}$ . The full width at half maximum of the Gaussian-like light distribution corresponded to the size of the light aperture (referred to as the spot size).

I investigated, whether the accuracy of the measurements can be enhanced by using alternative light source options. I focused on two primary objectives: minimizing the influence of surface recombination and maximizing the achievable charge carrier density. There are three basic parameters of a switchable laser light source: the wavelength of photons, the intensity of the beam and the duration of the illumination. First, I evaluated the influence of the photon wavelength.

### The impact of the excitation wavelength to $\Delta n(x,t)$

I employed the one-dimensional computer simulation of charge carrier dynamics, introduced in the previous chapter. To simplify the analysis, the injection-level dependence was considered only for intrinsic recombination processes, while defect-related mechanisms were treated as injection-independent:

$$\frac{1}{\tau_{\rm b}(\Delta n, N_{\rm dop})} = \frac{1}{\tau_{\rm SRH}} + \frac{1}{\tau_{\rm intr}(\Delta n, N_{\rm dop})}.$$
(5.8)

This approach allowed for studying the time evolution of the surface recombination rate and the assessment of achieved  $\Delta n_{avg}$  corresponding to realistic system and material parameters.



Figure 5.3: (a) Simulated steady-state injection levels,  $\Delta n_{\text{avg}}^{\text{SS}}$  in the function of excitation wavelength for different  $\tau_{\text{SRH}}$  values, (b) and the direct comparison of the 980 nm and 1064 nm laser sources.

Achieving the excess carrier density of  $10^{15} \text{ cm}^{-3}$  is important, as this is the standard  $\Delta n$  value for reporting carrier lifetimes in the industry. Indeed, increasing photon flux enhances the steady-state injection level  $\Delta n_{\text{avg}}^{SS}$ , however, above  $10^{16} \text{ cm}^{-3}$  the Auger-process becomes dominant, mitigating this effect. Moreover, radiation safety regulations in commercial systems impose constraints on the maximum allowable photon flux. All simulations utilized the maximum photon flux permitted by the 3B laser class  $2 \cdot 10^{19} \text{ cm}^{-2} \text{s}^{-1}$ ) for this system's optical design.

The wavelength of the excitation laser determines the absorption coefficient and, consequently  $\Delta n_{avg}^{SS}$ . Simulations were conducted across a wavelength range of 900 to 1100 nm for various  $\tau_{SRH}$  values. The results are illustrated in the Figure 5.3a. At shorter wavelengths, excess carriers are generated near the surface, leading to stronger surface recombination rates limiting  $\Delta n_{avg}^{SS}$ . From the other side, at longer wavelengths, the deeper penetration results in excess carriers being generated in a larger volume, which also reduces the average injection level. As  $\tau_{SRH}$  and the corresponding diffusion length increase, the maximum value of  $\Delta n_{avg}^{SS}$  shifts towards longer wavelengths. Above  $\tau_{SRH} > 1$  ms carrier lifetime, the excitiation wavelength range from 1050 to 1070 nm appears to be optimal. I found that the surface recombination is particularly significant in the initial part of the decay curve, causing strongly varying carrier lifetimes. Longer wavelengths, with their deeper penetration, reduce the impact of surface recombination during this critical initial period (as discussed below in relation to Figure 5.4.).

Considering these factors, and the availability of commercial laser sources, a 1064 nm wavelength laser was selected for replacing the original 980 nm laser. This way the penetration depth increased with a factor of 8 (from 0.1 mm to 0.8 mm). Comparison of maximum  $\Delta n_{avg}^{SS}$  across the entire  $\tau_{SRH}$  range of interest revealed a crossover point around  $\tau_{SRH} \approx 30 \,\mu$ s (Figure 5.3b.). Above this lifetime, where the majority of samples in the industry and in material research labs are found, the 1064 nm laser proves to be more beneficial. The target excess carrier density of  $10^{15} \,\mathrm{cm}^{-3}$  is achieved for  $\tau_{SRH}$  values above approximately  $\approx 15 \,\mu$ s, which aligns with the lower limit for mono-Si used to manufacture solar cells with acceptable efficiency. Therefore, based on the simulation of  $\Delta n(x,t)$ , I was able to define the optical excitation wavelength for PCD measurements of thick samples.

### Simulation of U(t) recorded by the e-PCD measurement

The theoretically achievable accuracy of the measurement setup with both wavelengths was investigated by simulations of  $\Delta n(x,t)$ . The measured photoconductance signal  $\Delta U(t)$  was considered to be proportional to the sheet density of "sensed" excess carriers  $\Delta N_{\text{sensed}}(t)$ , since when calibrating the eddy-current sensor, we received a linear calibration function to  $\sigma_{\text{sh}}$ . Then, we employed an exponentially decaying depth sensitivity function:

$$\Delta U(t) \propto \Delta N_{\text{sensed}}(t) = \int_0^W \Delta n(x, t) \cdot \exp\left(-\frac{x}{d_{\text{sensed}}(\rho)}\right) dx,$$
(5.9)

where W is the sample width and  $d_{\text{sensed}}(\rho)$  is the resistivity-dependent sensing depth presented in Figure 5.2. While this approximation assumes linearity between  $\Delta U(t)$  and  $\Delta N_{\text{sensed}}(t)$ , it is important to note that in general cases, this simplification may not hold due to the inhomogeneous  $\Delta n(x)$  depth profile and injection-level dependent carrier mobilities. However, for the purposes of these simulations, which focuses on results at  $\Delta n = 10^{15} \text{ cm}^{-3}$ , the use of a depthindependent  $d_{\text{sensed}}(\rho)$  is justified as  $\Delta n < N_{\text{dop}}$  (typically,  $N_{\text{dop}} > 5 \cdot 10^{15} \text{ cm}^{-3}$ ). The detailed explanation of the injection-dependent depth sensitivity function is presented in 5.3.1.

In both practical measurements and simulations, the laser pulse duration is set to achieve steady-state conditions. The practical evaluation of PCD transients begins with estimating the steady-state injection level,  $\Delta n_{\text{avg,meas}}^{SS}$ , based on the bulk lifetime and the laser parameters ( $\Phi$  and  $\alpha$ ) [206]:

$$\Delta n_{\rm avg}^{SS} = \frac{\alpha \Phi L^2}{(1 + \alpha L)^2 D_{\rm amb}},\tag{5.10}$$

where  $D_{amb}$  and L are the ambipolar diffusion coefficient and diffusion length, respectively. During the transient decay, the average injection level  $\Delta n_{avg,meas}(t)$  is considered proportional to the signal in both simulated (using Equation 5.9) and measured cases:

$$\Delta n_{\rm avg}(t) = \frac{\Delta n_{\rm avg}^{SS}}{\Delta U_{\rm max}} \cdot \Delta U(t).$$
(5.11)

The measured lifetime  $\tau_{\text{meas}}$  is then calculated using the standard dynamic definition:

$$\tau_{\rm meas}(t) = -\frac{n_{\rm avg,meas}(t)}{\frac{\partial n_{\rm avg,meas}(t)}{\partial t}}.$$
(5.12)



Figure 5.4: Simulation of the  $\Delta n(x,t)$  decay of a gallium doped silicon slab with both 980nm and 1064nm laser sources. (a) Excess carrier depth profiles in steady-state conditions and  $\Delta t = 150 \,\mu s$  time after the illumination is terminated. The depth of the maximal injection level moves towards the bulk as the near-surface carriers recombine rapidly at the surface. (b) The simulated measurable lifetime  $\tau_{\text{meas}}$  in the function of time elapsed. (c)  $\tau_{\text{meas}}$  is more accurate with the 1064 nm laser using the simplified injection level calculation. (d) This is generally true for a wide range of  $\tau_{\text{SRH}}$ . Simulation parameters:  $N_{\text{dop}} = 10^{16} \,\text{cm}^{-3}$ ,  $\tau_{\text{SRH}} = 300 \,\mu s$ ,  $W = 5 \,\text{cm}$ and  $\Phi = 2 \cdot 10^{19} \,\text{cm}^{-2} s^{-1}$  for both wavelengths.

### Assessment of the achievable accuracy of e-PCD based on simulations of U(t)

Simulations were performed for a p-type,  $N_{dop} = 10^{16} \text{ cm}^{-3}$  gallium-doped Si ingot with unpassivated surfaces and  $\tau_{SRH} = 300 \,\mu\text{s}$ . These parameters correspond to a reasonable material quality to manufacture PERC cells. The sample was considered thick enough (5 cm), so the recombination rate at the back surface is negligible. The two laser wavelengths (980 nm and 1064 nm) were compared, using the same photon flux of  $\Phi = 2 \cdot 10^{19} \,\text{cm}^{-2} \text{s}^{-1}$ . The results are illustrated in Figure 5.4. The 1064 nm laser produces a deeper carrier profile (Figure 5.4a.), reducing the influence of surface recombination. This is evident in the time-dependent measured lifetimes (Figure 5.4b.), which increases with elapsed time due to the deepening carrier profile and the decreasing contribution of Auger-recombination. On Figure 5.4c.  $\tau_{meas}$  is plotted in the function of  $\Delta n_{avg}$  and compared to  $\tau_b$ . At  $\Delta n = 10^{15} \,\text{cm}^{-3}$  the measured lifetime is approxi-



Figure 5.5:  $\tau_{\rm sim}(\Delta n = 10^{15} \,{\rm cm}^{-3})$  is independent of the front surface recombination velocity,  $S_{\rm front}$ , in the typical range between  $10^3 - 10^6 \,{\rm cm/s}$  for thick silicon samples. Simulation parameters:  $N_{\rm dop} = 10^{16} \,{\rm cm}^{-3}$ ,  $\tau_{\rm SRH} = 3 \,{\rm ms}$ ,  $W = 5 \,{\rm cm}$  and  $\Phi = 2 \cdot 10^{19} \,{\rm cm}^{-2} {\rm s}^{-1}$ .

mately 75% of the bulk lifetime with the 1064 nm laser, compared to only 50% with the 980 nm laser for  $\tau_b \approx 300 \,\mu$ s. Simulations were repeated for various  $\tau_{SRH}$  values to assess measurement accuracy over a wide range (Figure 5.4d.). The ratio of  $\tau_{meas}(10^{15} \,\mathrm{cm}^{-3})$  to  $\tau_b(10^{15} \,\mathrm{cm}^{-3})$  was analyzed, revealing that the 1064 nm laser produces measured lifetimes exceeding 80% of the bulk lifetime for  $\tau_b = 100 \,\mu$ s, while this ratio is continuously decreasing to 60% for  $\tau_b = 10 \,\mathrm{ms}$ . In theory this result may enable a simple correction of the measured lifetime to bulk lifetime at a given injection level.

The dependence of measured lifetime on surface recombination velocity ( $S_{\text{front}}$ ) was examined for a wide range in ingot-level measurements (Figure 5.5.). Using the 1064 nm laser, the simulated measurement outputs showed minimal variation within the typically relevant range of  $10^4$  cm/s to  $10^6$  cm/s, indicating that the e-PCD measurement is relatively insensitive to the actual value of surface recombination velocity. This may be important in cases when there is a surface etching step prior to the e-PCD measurement.

#### Assessment of the achievable accuracy of e-PCD based on experiments

I evaluated the possible benefits of the laser optimization for e-PCD measurements on silicon ingots across a wide range of carrier lifetimes, experimentally. Semilab WT-1200I/IL systems were used to compare PCD measurement using the original 980 nm laser source with an optimized 1064 nm source. To isolate the effects of wavelength and light power, measurements with the optimized setup were performed at both low  $(3 \cdot 10^{18} \text{ cm}^{-2} \text{s}^{-1})$  and high  $(2 \cdot 10^{19} \text{ cm}^{-2} \text{s}^{-1})$  photon flux levels. To illustrate the significance of both power and wavelength optimization, I analyzed the measured  $\tau_{\text{meas}}(t) = -U(t)/\frac{\partial U(t)}{\partial t}$  values as a function of  $\Delta t$  for three representative samples in Figure 5.6., each exhibiting different ILDCL characteristics.

First, we examined a high-purity n-type sample with  $20\Omega$ cm resistivity, featuring negligible ILD at low injection levels (Figure 5.6a.). This behavior is more common in n-type sili-



Figure 5.6: Temporal evolution of measured  $\tau_{\text{meas}}$  during PCD measurement on thick silicon slabs. (a) High-quality n-type silicon slab exhibiting negligible injection-level dependence (ILD) demonstrates comparable  $\tau_{\text{meas}}(\Delta t)$  increase irrespective of laser wavelength and power. (b, c) Samples with pronounced ILDCL, increased wavelength and laser power makes the determination of higher  $\tau_{\text{meas}}$  possible closer to  $\tau_{\text{b}}$ . (Low power:  $\Phi = 3 \cdot 10^{18} \text{ cm}^{-2} \text{s}^{-1}$ , high power:  $\Phi = 2 \cdot 10^{19} \text{ cm}^{-2} \text{s}^{-1}$ ).

con due to the generally higher capture cross-coefficients of electrons compared to holes for metallic contaminants, such as interstitial iron. The results showed no significant difference between  $\tau_{\text{meas}}(\Delta t)$  curves recorded using different parameters. However, at  $\Delta t = 0 \,\mu$ s, the carrier lifetime was shorter when measured with the 980 nm laser compared to the 1064 nm laser, attributable to the strong impact of surface recombination. Additionally, this initial lifetime was shorter when operating the 1064 nm laser at high power compared to low power, due to Auger recombination reducing the bulk lifetime at higher injection levels. In summary, the negligible ILD resulted in similar maximum measurable lifetimes, regardless of laser power properties, indicating that ingot and slab purity can be reliably assessed even with the original 980nm laser source.

Results from bulk samples exhibiting moderate and strong ILD (Figs. 5.6b. and c., respectively) clearly demonstrates the importance of light excitation parameters in the measurement results. Both samples showed an initial increase in effective lifetime, similar to Figure 5.6a., followed by a decrease in  $\tau_{\text{meas}}$  due to ILD. This behavior is typically caused by defects or contaminating atoms, which act as SRH centers limiting the lifetime at low injection levels (as it was presented in Figure 2.6.). The longer wavelength and higher laser power resulted in increased maximum lifetime due to the higher injection level achieved. The difference between results obtained with the 980 nm and 1064 nm lasers correlated positively with the strength of the ILD, confirming the importance of optimized laser excitation for industrial applications where samples with strong ILD are frequently encountered.

#### Investigation of the e-PCD measurement accuracy using surface passivation

The influence of surface recombination at various injection levels was experimentally investigated using 2 cm thick Czochralski-grown monocrystalline silicon slabs. Measurements were conducted using the optimized laser e-PCD tool on samples with both as-sliced and chemically passivated surface preparations. Nine gallium-doped and two phosphorus-doped slabs were se-



Figure 5.7:  $\tau_{\text{meas}}$  recorded on as-sliced surfaces of Czochralski-grown monocrystalline silicon slabs approaches  $\tau_{\text{meas}}$  after chemical surface-passivation. The time required for this convergence increases with higher lifetime values. At the standard excess carrier density of  $\Delta n = 10^{15} \text{ cm}^{-3}$  (denoted by yellow lines) and below, the discrepancy remains under 20% for (a) low, (b) medium and (c) high lifetime samples.

lected to cover the entire industrially relevant lifetime range. Chemical passivation was achieved through a thorough process, which involved chemical damage etching (removing a 30  $\mu$ m thick Si layer from the top surface) followed by the application of an iodine-ethanol solution. For chemically passivated samples, the front surface recombination velocity ( $S_{\text{front}}$ ) was estimated to be between 50 cm/s and 200 cm/s, with 100 cm/s used in the 1-D simulation to calculate  $\Delta n_{\text{avg}}^{\text{SS}}$  for the passivated samples. This passivation ensures that  $\tau_{\text{meas}}$  closely approximates  $\tau_{\text{b}}$  for thick samples. Figure 5.7. presents detailed laser e-PCD results from three selected samples representing different lifetime ranges.

When decreasing  $\Delta n$  from high injection levels, the data exhibits an initial increase in lifetime, attributed to the decreasing rates of surface and Auger recombination (See in Figure 2.6.). The longer the bulk lifetime and the diffusion length, the longer the period with strong influence of surface recombination, as illustrated in Figures 5.7b. and c. This phenomenon is evident when comparing lifetime curves from passivated and as-sliced samples. In the low lifetime range (Figure 5.7a.), the curves show negligible difference, while in the millisecond lifetime range, they converge only when  $\Delta n$  decreases an order of magnitude.

Current industrial practice typically reports carrier lifetime values at  $10^{15} \text{ cm}^{-3}$ . Therefore, we conducted a detailed comparison of  $\tau_{\text{meas}}(\Delta n = 10^{15} \text{ cm}^{-3})$  values (Figure 5.7., yellow markers). As shown in Figure 5.8., the measured lifetime values did not deviate more than 20% between as-cut surfaces ( $\tau_{\text{raw}}$ ) and chemically passivated surfaces ( $\tau_{\text{pass}}$ ) for any of the samples. This excellent correlation between results obtained from as-sliced and passivated samples demonstrates the usefulness of the optimized laser e-PCD technique in mitigating surface recombination effects, even for unpassivated samples.

This optimization of the laser source enhances the accuracy and reliability of e-PCD measurements. The use of a longer wavelength (1064 nm) and sufficient laser power significantly reduces the impact of surface recombination when calculating the carrier lifetime at  $\tau_{\text{meas}}(\Delta n = 10^{15} \text{ cm}^{-3})$ . This approach enables the recording of carrier lifetimes and injection levels very close to  $\tau_{\text{b}}$ , even when using simple evaluation routines. These findings indicate the robustness of the optimized laser e-PCD method for accurate carrier lifetime measurements



Figure 5.8: A robust correlation is observed between  $\tau_{\text{meas}}$  recorded on as-cut and chemically passivated surfaces at  $\Delta n = 10^{15} \text{ cm}^{-3}$  across the entire lifetime range of interest. For sample exhibiting the lowest lifetime, comparison is made at  $\Delta n = 10^{14} \text{ cm}^{-3}$ , since  $\Delta n = 10^{15} \text{ cm}^{-3}$  is not reached for such low lifetime.

across a wide range of silicon materials without any surface treatment. This capability is particularly valuable for industrial applications, where rapid and reliable characterization of as-cut silicon ingots and slabs is essential for quality control and process optimization in photovoltaic manufacturing.

### **5.3** Obtaining $\tau_{\mathbf{b}}(\Delta n)$ from e-PCD measurements

This section presents the modeling of the depth sensitivity in eddy-current measurements for samples with inhomogeneous charge carrier depth profiles. The aim is the accurate evaluation of transients providing  $\tau_b(\Delta n)$  recorded by the e-PCD technique.

### 5.3.1 Parametrization of the depth sensitivity of eddy-current sensing

During a typical e-PCD measurement, the light source generates a strongly inhomogeneous excess carrier depth profile  $(\Delta n(x))$  near the surface of the sample. The proximity of the many charge carriers at the surface significantly raises the eddy-current signal compared to a deeper  $\Delta n(x)$  profile. In this case, the accurate calculation of the average injection level  $\Delta n_{avg}$  from the signal is challenging. The contribution of excess carriers to the measured signal depends on their spatial distribution and the sensitivity decay of the probing electromagnetic field in the sample.

Commercially available e-PCD systems often rely on static depth sensitivity functions,  $\varphi(x)$ , which assume constant material properties [207]. While in Chapter 5.2, I presented an alter-



Figure 5.9: Cross-sectional diagram of the three-dimensional finite element model. The maximum of the simulated eddy-current density lies below the coil, 7.5 cm from the center of the sample. Based on the depth distribution a one-dimensional depth sensitivity function was created.

native solution with an equilibrium resistivity-dependent depth sensitivity  $\varphi(x, \rho)$  (Equation 5.9. However, at high excitation levels, where the  $\Delta n(x)$  becomes comparable to or exceeds the doping density, significant variations occur in the skin depth, consequently, in the depth sensitivity. These variations change dynamically during the PCD measurement. To address this issue, I developed a more generalized depth sensitivity function ( $\varphi(x, \sigma(x))$ ) [211]. This function accounts for the local conductivity changes due to excess carriers and enables accurate reconstruction of the measured signal:

$$U(t) \propto \int_0^\infty \sigma(x,t) \cdot \varphi(x,\sigma(x,t)) dx, \qquad (5.13)$$

### **3D** finite element simulations

The interaction between the probing electromagnetic field and the charge carriers inherently involves a three-dimensional problem. To model this interaction, a three-dimensional finite element (3D-FE) simulation was accomplished using *COMSOL Multiphysics* software. The simulation accounted for the geometry of the RF coil, sample dimensions, and dynamic changes in conductivity during measurement. The 3D-FE simulations provided valuable insights into eddy current density distributions and their dependence on material properties (Figure 5.9.). These simulations were validated by comparing computed signals with experimental measurements of silicon slugs with known resistivity under non-illuminated conditions. The excellent correlation between the simulated and measured signals confirmed the reliability of the 3D-FE model.

#### Development of a simplified 1D model

While 3D-FE simulations provide accurate predictions, they are computationally intensive and unsuitable for real-time analysis during measurements. To address this limitation, a onedimensional (1D) approximate model was developed. The empirical depth sensitivity function,  $\varphi(x, \sigma(x))$ , was constructed based on the parametrization of eddy current density distribution of the 3D-FE simulated steady-state  $\Delta n(x)$  profiles. The calibration of this model involved application of homogeneous resistivity etalon samples of varying thicknesses to determine key parameters of  $\varphi(x, \sigma(x))$ . Validation was performed using samples illuminated by controlled laser intensities to generate known steady-state excess carrier profiles. For such  $\Delta n(x)$  profiles the measured and simulated eddy signals were compared to each other to fine-tuning the model. This method provides sufficient accuracy for practical applications while being computationally efficient.

### **5.3.2** Iterative simulation based measurement evaluation

In Section 5.2 I presented that the measurement accuracy can be significantly enhanced through optimal selection of illumination sources. However, precise determination of  $\tau_b(\Delta n)$  across the full injection level range remains challenging due to several confounding factors. The detected decay rate underestimates the bulk lifetime due to the influence of surface recombination, particularly in the early phase of the decay. Moreover, the continuously deepening inhomogeneous excess carrier profile and the simultaneously evolving depth sensitivity preclude analytical calculation of the bulk lifetime. This complexity is further compounded when considering the ILD of physical parameters such as the bulk carrier lifetime or diffusion coefficient. This chapter presents my approach to overcome these obstacles by procreating a novel evaluation method. It should enable the extraction of  $\tau_b(\Delta n)$  from PCD transients recorded on silicon ingots or slugs, without requiring any surface treatment.

The approach integrates charge-carrier dynamics and eddy-current detection sensitivity models to fully simulate the transient lifetime measurement process. At its core, the method solves the continuity equation for excess carrier density ( $\Delta n$ ) in its full complexity, as it was presented in Section 5.2, incorporating the most recent Auger recombination model [4] and the injection level dependent mobility model proposed by Klaassen [98, 99]. The sample thickness (W) is adjustable, while the surface recombination velocity is set to 10<sup>6</sup> cm/s. Illumination parameters, such as pulse length, energy, and absorption coefficient, are also adjustable, with default settings reflecting realistic parameters used in the measurement tool. From the computed excess charge carrier profile, a conductivity distribution is determined:

$$\sigma(x,t) = e \cdot |N_{dop} \cdot \mu_{maj}(N_{dop}, \Delta n) + \Delta n \cdot \mu_{sum}(N_{dop}, \Delta n)|.$$
(5.14)

The implied measured voltage signal is then calculated using the depth sensitivity function of the eddy-current detection ( $\varphi(x, \sigma(x, t))$ ):

$$U(t) = \beta \int_0^W \sigma(x,t) \cdot \varphi(x,\sigma(x,t)), \qquad (5.15)$$

where  $\beta$  is an instrumental factor determined through calibration with high resistivity thin wafers.



Figure 5.10: The schematic summary of the iterative evaluation process to obtain  $\tau_b(\Delta n)$  on thick silicon samples.

The integration of charge carrier dynamics and depth-sensitivity models enables comprehensive simulation of the PCD measurement process. To determine the  $\tau_b(\Delta n)$  curve from the measured signal decay, an iterative evaluation process based on the e-PCD measurement simulation was developed. Figure 5.10. schematically illustrates this simulation method.

The evaluation utilizes two primary measurement input parameters: the measured dark signal  $(U_0)$  and an offset-corrected signal decay curve  $(\Delta U(t))$ . The former provides the doping concentration  $(N_{dop})$ , while the effective measured lifetime  $(\tau_{eff,meas})$  can be calculated from the latter as the function of elapsed time. Additional required sample parameters include thickness and doping type.

To initiate the PCD measurement simulation, an initial estimate of the  $\tau_{\rm b}(\Delta n)$  function is necessary. As a first approximation, in addition to the unavoidable Auger recombination, a constant  $\tau_{\rm SRH,0} = 1.5 \cdot \max(\tau_{\rm meas}(\Delta t))$  is used (see Figure 5.4c.), accounting for the typical reduction in measurable lifetime due to surface recombination:

$$\frac{1}{\tau_{\rm b}(\Delta n, N_{\rm dop})} = \frac{1}{1.5 \cdot \max(\tau_{\rm meas}(\Delta t))} + \frac{1}{\tau_{\rm Auger}(\Delta n, N_{\rm dop})}.$$
(5.16)

Following the simulation, the implied signal decay U(t) is calculated as described by Equa-
tion 5.15. Based on the decay of the simulated signal, an effective lifetime can be calculated:

$$\tau_{\text{eff,implied}}(t) = \frac{U(t)}{-\frac{\partial U(t)}{\partial t}}.$$
(5.17)



Figure 5.11: Iterative evaluation process steps to obtain  $\tau_{\rm b}(\Delta n)$  on thick silicon samples for a 1.5 cm thick p-type sample with  $N_{\rm dop} = 2.4 \cdot 10^{16} \,\mathrm{cm}^{-3}$ . Simulated decay of (a)  $\Delta n_{\rm avg}$  and (b) eddy-current signal  $\Delta U$ . (c) The simulated measurable  $\tau_{\rm eff}$  fits well on the measured curve from the second iteration step. The bulk lifetime correction factor  $\alpha$  in the function of (d) elapsed time and (e)  $\Delta n_{\rm avg}$ . (f) The  $\tau_{\rm b}(\Delta n)$  result does not change significantly from the second iteration step.

This function is then compared to  $\tau_{\text{eff,meas}}$  as a function of time, and their ratio ( $\alpha$ ) serves as a correction factor for  $\tau_{b}(\Delta n)$ . The corrected bulk lifetime is subsequently used as input for the next iteration of the PCD simulation, refining the accuracy of the  $\tau_{b}(\Delta n)$  determination.

The iterative simulation process is illustrated step-by-step in Figure 5.11. for a 1.5 cm thick p-type sample with  $N_{dop} = 2.4 \cdot 10^{16} \text{ cm}^{-3}$ . Following the complex simulation, Figure 5.11a. shows the decay dynamics of  $\Delta n_{avg}$ , while Figure 5.11b. displays the decay of the measurable signal.

Figure 5.11c. presents  $\tau_{eff}$ , determined from the measured signal decay using equation 5.17. The result obtained from the first iteration step differs significantly from the measured curve. While the simulated lifetime continuously increases due to the diminishing effects of the Auger process and surface recombination, the measured lifetime exhibits a decreasing trend after an initial phase.

Figure 5.11d. shows the relative difference between the two curves as a function of elapsed time. During the first simulation, this value varies between 0.5 and 1.25. The same parameter can be examined as a function of  $\Delta n_{\text{avg}}$  (Figure 5.11e.), which enables the correction of the  $\tau_{\text{b}}(\Delta n)$  function (Figure 5.11f.). Finally, this function serves as input parameter for the subsequent simulation.

It is evident that from the second step onward, only minor modifications are observed in the simulated decay curves and the calculated  $\tau_{\text{eff}}(\Delta t)$  function. The deviation of the latter from the measured value is less than 2% for all  $\Delta n$  values.

This novel evaluation method represents a significant advancement in the characterization of silicon ingots and other thick semiconductor materials. By providing more accurate and detailed information about bulk lifetime and its ILD, it can contribute to improved quality control in silicon ingot production, a better understanding of recombination mechanisms in semiconductor materials, and more precise material selection for high-efficiency solar cells.

The evaluation method was tested on several thick (>1 cm) mono-Si ingots using the optimized e-PCD with 1064 nm laser source. Samples were selected from a wide range of lifetimes and resistivities to investigate the generality of the method. Measurements were performed on ingots with both as-cut and polished surfaces to demonstrate the method's independence from surface quality within this high surface recombination velocity (SRV) range.

### 5.3.3 Results on industrial Ga-doped thick slugs

Ga-doped mono-Si samples were selected from different pulls of the rechargeable Czochralski (RCz) process, including tail, middle, and seed end sections of each ingot, as it was presented in Section 3.6. This approach allowed testing the method on ingots with varying purity levels due to the impurity segregation (Section 2.3). Additionally, thin wafers were sliced from adjacent parts of the ingot, and p+/i/Cz-Si wafer/i/p+ symmetrical lifetime test structures were fabricated using a-Si:H layers of different thicknesses. The results were assumed as control values for  $\tau_b(\Delta n)$ .

Bulk lifetime values typically ranged from  $100\,\mu$ s to 1 ms, with high-quality surface passivation achieved on four samples, as illustrated in Figure 5.12. Results from different regions of the same ingot are presented. The lifetime in the top (Figure 5.12a.) and middle (Figure 5.12b.) sections is nearly identical, indicating the absence of harmful seed-end-related defects caused by oxgen precipitation in that region [85]. However, the lifetime decreases significantly at the



Figure 5.12: Results of 1.5 cm slugs from Ga-doped ingots are presented. The lifetime in the (a) top, (b) middle and (c) tail part of the same ingot shows a decreasing lifetime during the crystal pulling process due to the segregation of metallic impurities. (d) Another tail-end ingot from an earlier RCz process pull reveals a higher lifetime, attributed to a lower concentration of metallic impurities. The iterative simulation-based evaluation yields carrier lifetime curves fitting well with results from passivated adjacent wafers, while using the conventional (conv.) evaluation method without surface recombination correction, 30-50% discrepancy is observed at high  $\Delta n$ .

tail-end (Figure 5.12c.) due to the segregation of metallic impurities. Examining another tailend ingot from an earlier RCz process pull (Figure 5.12d.) reveals a higher lifetime, attributed to a lower concentration of metallic impurities.

The iterative simulation-based evaluation yields lifetime values at high  $\Delta n$  levels 30 - 50% higher than those obtained using the conventional method without surface recombination correction, presented in Section 5.2. The discrepancy continuously decreases during the decay. The 20% deviation observed around  $\Delta n = 10^{15} \text{ cm}^{-3}$  is in good agreement with our previous simulation results. Notably, these results align well with measurements from passivated wafers, validating the accuracy of our evaluation technique.

## 5.3.4 Results on high quality n-type slugs

In addition to the Ga-doped samples, we evaluated our method on long carrier lifetime ntype Cz-Si slugs, typically utilized as raw material in passivated contact solar cell structures.



Figure 5.13: After an initial, redistribution decay phase, a symmetrical  $\Delta n(x)$  profile develops, which makes the lifetime analytically predictable..

These samples, also approximately 1-2 cm thick, exhibited lifetimes typically in the 1-10 ms range. This results in an extended minority carrier diffusion length reaching the millimeter range. Such samples with long  $\tau_b$  behave analogously to wafers regarding the  $\Delta n(x,t)$  decay characteristics. Consequently, after an initial period, an approximately symmetrical charge carrier profile is established (Figure 5.13.), allowing the measurable effective lifetime to be analytically calculated using a similar expression to Equation 2.28:

$$\frac{1}{\tau_{\rm eff,li}} = \frac{1}{\tau_{\rm b,li}} + \frac{W^2}{D_{\rm p,li} \cdot \pi^2},$$
(5.18)

where W represents the sample thickness, and  $\tau_{eff,li}$ ,  $\tau_{b,li}$ , and  $D_{p,li}$  denote the effective lifetime, bulk lifetime, and minority hole diffusion coefficient, respectively, at low injection levels. This analytical expression provides a straightforward method to validate our simulation results in cases of constant  $\tau_{b,li}$ .

The measured slugs can be categorized into two groups. The first group comprised highquality samples exhibiting a saturated effective lifetime at low injection levels (Figure 5.14a.). Following an initial redistribution phase during which the  $\Delta n(x)$  profile becomes symmetric (Figure 5.13.), the decay can be characterized by a single  $\tau_{eff,li}$  value. We typically determined this  $\tau_{eff,li}$  value at approximately  $10^{14}$  cm<sup>-3</sup>, where the influence of Auger recombination becomes negligible. For such samples, the bulk lifetime can be calculated using the diffusionlimited lifetime formula (see Equation 5.18). Both the measured low-injection effective lifetime and the calculated bulk lifetime are depicted using solid lines in Figure 5.14a. The simulated bulk lifetime aligns perfectly with this theoretical value at low injection levels.

When measuring more contaminated silicon slugs with lower  $\tau_b$ , the analytical formula for diffusion lifetime becomes inapplicable due to the typical decrease in lifetime at low injection levels (Figure 5.14b.). In this scenario, the significance of our simulation-based evaluation becomes more apparent, as the discrepancy between measured and bulk lifetime continuously diminishes during the decay.

Overall, it is evident that the deviation between traditional and simulation-based evaluation can be substantial in this high-lifetime range due to the long diffusion length of excess charge



Figure 5.14: (a) A silicon slug of 1.5 cm thickness with constant carrier lifetime at low injection level demonstrates the accuracy of our simulation-based evaluation method. (b) For more contaminated samples, the importance of the novel evaluation is observable as the difference between the bulk and measured lifetime is continuously changing during the decay.

carriers. These results confirm the findings of our previous simulation, which indicated an inaccuracy of approximately 20% at  $10^{15}$  cm<sup>-3</sup> in this range. However, those studies did not account for the inhomogeneous depth sensitivity of the eddy-current sensor and only considered very thick samples. Our current results clearly demonstrate that sample thickness is also a crucial parameter for precise determination of  $\tau_b(\Delta n)$ .

I have developed the first evaluation method to obtain the complete  $\tau_{\rm b}(\Delta n)$  curve from PCD measurements by compensating for the effect of surface recombination. This evaluation method is based on the most detailed PCD measurement simulation ever reported, involving the dynamically changing sensitivity depth profile of the eddy-current-based sensing. This phenomenon for PCD measurements was neither handled nor reported prior to my work. The 3D finite element simulation helped to understand the phenomenon and provided artificial reference data to "calibrate" the a simplified phenomenological 1D model, I created the simulation-based approach appropriate for real-time measurement, even for industrial applications. My method was tested on various p- and n-type samples. Results from Ga-doped slugs evaluated using this novel approach showed excellent agreement with neighboring passivated wafers. For high-lifetime n-type samples, the wafer-like behavior provided an analytical proof for our method. These findings confirm the reliability of this novel bulk lifetime measurement technique, which offers more accurate characterization of silicon raw material prior to slicing. The application of this technique paves the way for identifying harmful defects or estimating final solar cell efficiency at the ingot level. Furthermore, the reliable determination of the carrier lifetime in very pure silicon material featuring long lifetime is really challenging. Above 10 ms bulk lifetime, any of the surface passivation techniques are not good enough to obtain  $\tau_{\rm b}$  from wafers. E-PCD measurement enhanced by my evaluation is probably the only method which can provide  $\tau_{\rm b}(\Delta n)$ in the as-grown state of the material. This enables to deliver important material researches of silicon growth methods. Related collaboration projects are ongoing with industrial partners and research laboratories.

## Chapter 6

# Simultaneous measurement of charge carrier lifetime and mobility

In this chapter, I summarize my development work on a novel carrier lifetime measurement technique combining three lifetime methods for silicon wafers. The combined technique enables to determine  $\tau(\Delta n)$  with enhanced accuracy, compensating the limitations of each individual methods. In addition, the sum of the excess carrier mobilities as a function  $\Delta n$  is also determined, which is particularly important for non-standard wafer types, for which known mobility models are not valid. The presented results were published in [O4, O5].

In the rapidly evolving field of photovoltaics, accurate characterization of carrier lifetime is crucial for controlling and optimizing solar cell manufacturing processes. Measurements on thin silicon wafers offer greater precision compared to bulk measurements, as depth-related effects such as material inhomogeneity, diffusion, or depth-sensitive detection do not pose significant challenges. This absence of depth-related issues enables a more localized and precise determination of injection-level dependent lifetime  $\tau(\Delta n)$ . In modern cells fabricated from high-purity materials, the bulk diffusion length typically exceeds the wafer thickness. Consequently, the measured effective lifetime evaluates both bulk lifetime and surface passivation quality, providing information on their combined effect on the expected cell efficiency.

Recent years have seen significant advancements in surface passivation techniques, including Si heterojunction methods and tunnel oxide-based approaches (Section 2.1.2). These improvements have drastically reduced recombination losses, resulting in substantially higher carrier lifetime ( $\tau$ ) and injection level ( $\Delta n$ ) values in state-of-the-art solar cells. This progress necessitates a re-evaluation of the precision of commonly used carrier lifetime measurement methods and their corresponding empirical models, particularly at elevated injection levels.

One key parameter is the charge carrier mobility (Section 2.2.1), which is also necessary to accurately evaluate the photoconductance decay (PCD) lifetime measurement curves. While the charge carrier mobility in monocrystalline silicon has been extensively studied as a function of doping concentration and temperature, relatively few experimental studies have addressed its injection level dependence. The most frequently cited experimental dataset, provided by Dannhauser [96] and Krausse [97], utilizes voltage measurements of p-n junctions to determine  $\mu(\Delta n)$ . In the past two decades, three major photoconductance-based studies explored this relationship: Neuhaus et al. employed the quasi-steady-state open circuit voltage method (QSS-Voc) [102], Rougieux et al. presented a contactless method combining transient and steady-state lifetime measurements using QSSPC for samples with intermediate carrier lifetimes [103], and Hameiri et al. determined mobility using an independently calibrated photoluminescence measurement [106].

This chapter explores the development and application of an innovative carrier lifetime measurement technique that combines multiple methods, based on two of my articles. The first study integrated PCD and small perturbation photoconductance decay (SP-PCD) techniques, offering unprecedented accuracy and reliability over a wide range of  $\Delta n$  [O4]. After addressing temperature stability issues caused by high-power laser usage, the steady-state photoconductance (SS-PC) method was also incorporated into the setup. This combination mitigates uncertainties associated with individual methods and enables simultaneous measurement of charge carrier concentration, mobility, and lifetime [O5]. This advancement addresses the long-standing challenge of accurately determining carrier mobility, especially at high injection levels where existing models show significant discrepancies. The method provides a novel, contactless approach to determine the sum of electron and hole mobilities, providing reliable basic material parameters for both standard and non-standard silicon wafers. Throughout this chapter, I explore the theoretical foundations, experimental setups, and key findings from these studies. I discuss how these new approaches overcome limitations of traditional measurement techniques and offer new possibilities for material characterization in photovoltaic research and development.

These developments represent significant progress in the characterization of silicon for photovoltaic applications. By providing more accurate and comprehensive data of charge carrier properties, these techniques enable better understanding and optimization of solar cell performance. The methods described here are particularly relevant for evaluating high-efficiency cell structures and advanced materials, where precise knowledge of carrier dynamics is essential for pushing the boundaries of solar cell efficiency.

## 6.1 Employed measurement methods and their limitations

As highlighted in the introduction, our measurement setup developed for unified carrier lifetime measurement integrates three principal photoconductance-based carrier lifetime measurement methods (Section 2.2.3). These techniques primarily measure distinct physical quantities and, consequently, employ different approaches to ultimately derive the  $\tau(\Delta n)$  function. The evaluation routines associated with these methods incorporate empirical physical models and approximations, which may potentially compromise their accuracy. In this section, we examine the potential sources of inaccuracy inherent to each method.

## Photoconductance decay (PCD) and Steady-State Photoconductance (SS-PC)

The fundamental principle underlying photoconductance decay (PCD) evaluation for silicon wafers involves measuring the change in sheet conductance ( $\sigma_{sh}$ ) following the excitation with a light pulse. Subsequently, the excess carrier concentration decay  $\Delta n(t)$  is calculated using a mobility model, taking into account the mobility shift of doping-related carriers as described in 3.3. The carrier lifetime is then determined from the decay of excess carriers, as expressed by:



Figure 6.1: (a) Simulation results on the same transient decay measurement using  $\mu(\Delta n)$  data by Zheng [104] and Dannhauser [96] for evaluation. (b) 20% discrepancy for  $\Delta n$  and 4% for  $\tau$  is observable.

$$\tau(\Delta n) = \frac{\Delta n}{-\frac{\partial \Delta n}{\partial t}},\tag{6.1}$$

Utilizing a continuous wave (CW) laser or a sufficiently long laser pulse enables the examination of the sample under pure steady-state conditions. In this case, a single lifetime value can be obtained at the given  $\Delta n$ :

$$\tau(\Delta n_{\rm SS}) = \frac{\Delta n_{\rm SS}}{G_{\rm SS}} = \frac{\Delta n_{\rm SS}}{\Phi_{\rm ph} \cdot OF},\tag{6.2}$$

where G represents the generation rate within the silicon wafer, which is related to the incident photon flux  $\Phi_{ph}$  through the optical factor (*OF*). This factor defines the ratio of generated electron-hole pairs to incident photons and introduces uncertainty due to varying optical properties of different sample structures. In the case of long-wavelength measurements, sample thickness can also influence the *OF* due to significant light transmission.

Since some solar cell structures contain thin layers of high conductance (diffusion layer or transparent conductive oxide), their contribution to  $\sigma_{sh}$  must be taken into account. Furthermore, both PCD and SS-PC methods rely on accurate carrier mobility models to translate  $\sigma_{sh}$  into  $\Delta n$ , which can be problematic, particularly at high injection levels. Figure 2.4. illustrates the sum of excess electron and hole mobilities  $\mu_{sum}(\Delta n)$ , demonstrating significant discrepancies between various models at high injection levels.

To emphasize the impact of model deviations on the determined  $\Delta n$  and  $\tau$  values, two of these models are compared in detail through simulation. Evaluating the same PCD measurement of a sample with  $\tau_{\text{SRH}} = 1 \text{ ms}$  and  $N_{\text{dop}} = 10^{15} \text{ cm}^{-3}$ , the resulting  $\tau(\Delta n)$  curves appear similar (Figure 6.1a.). However, the difference in  $\Delta n$  can reach 20% in the initial part of the decay. While the discrepancy between  $\tau$  values at the same  $\Delta n$  is less significant, it still exceeds 4% (Figure 6.1b.). In SS-PC measurements, the uncertainty in  $\Delta n$  directly translates to an equivalent uncertainty in the evaluated lifetime. The uncertainty associated with carrier mobility models is most pronounced at high injection levels ( $\Delta n$ ). However, as the injection level decreases, the significance of this uncertainty diminishes.

I need to emphasize the importance of the accuracy of the  $\Delta n$  values. The predicted performance of the solar cells can be calculated from  $\Delta n$  and not  $\tau$ , since the so-called implied open-ciruit voltage, iV<sub>oc</sub> is calculated from it:

$$iV_{\rm oc} = \frac{k_{\rm B}T}{q} \frac{\Delta n (\Delta n + N_{\rm dop})}{n_i^2}.$$
(6.3)

This formula is based on the approximation that the cell voltage in steady-state condition is originated from the quasi-Fermi level splitting of electrons and holes [25]. The 20% uncertainty in the carrier mobility data and so in the calculated  $\Delta n$ , which may appear at high injection levels using conventional PCD measurements, causes 5 - 10 mV uncertainty in the calculated  $iV_{oc}$  result.

### Small Perturbation Photoconductance Decay (SP-PCD)

The small perturbation photoconductance decay (SP-PCD) measurement method, based on the principle of the QSS- $\mu$ PCD technique (Section 2.2.3) was previously implemented by Wilson et al. [144, 145]. This method employs CW laser illumination to establish a steady-state condition with an excess carrier density ( $\Delta n_{SS}$ ), which is subsequently perturbed by a lowintensity, pulsed light source. This approach enables the measurement of the differential or small perturbation lifetime  $\tau_d$ . By repeating this measurement at various steady-state levels, the actual lifetime ( $\tau_a$ ) is calculated by integrating  $\tau_d$  over a range of generation rates ( $G_{SS}$ )):

$$\tau_{\rm a}(G_{\rm ss}) = \frac{1}{G_{\rm ss}} \int_0^{G_{\rm ss}} \tau_{\rm d}(G'_{\rm ss}) dG'_{\rm ss}. \tag{6.4}$$

The excess carrier density is then determined using the established formula:

$$\Delta n(G_{\rm ss}) = G_{\rm ss} \cdot \tau_{\rm a}(G_{\rm ss}). \tag{6.5}$$

In this chapter, this method is referred to as the small perturbation photoconductance decay method (SP-PCD).

The primary advantage of this method is its independence from sensor calibration or mobility models for  $\Delta n$  determination, as well as its insensitivity to surface conductive layers. However, a precise knowledge of optical losses is crucial for an accurate determination of injection level.

To ensure accurate  $\tau_d$  results at a given steady-state injection level, the perturbing laser power ( $G_p$ ) must be significantly lower than the steady-state light source. This constraint maintains the small perturbation nature of the measurement, resulting in purely exponential transient curves and indicating the reliability of the recorded  $\tau_d$  value. Employing a higher  $G_p$  leads to inaccurate  $\tau_d$  measurements due to significant changes in injection level during measurement, violating the small perturbation requirement. This limitation results in a low signal-to-noise ratio at low  $\Delta n$ , necessitating multiple transient measurements and averaging, thereby increasing measurement time. Consequently, the accuracy of this method is limited at low  $\Delta n$ .



Figure 6.2: Schematic illustration of the measurement setup for silicon wafers.

## 6.2 Combining the PCD, SS-PC and SP-PCD measurement principles

To address the limitations of individual techniques, an integrated measurement system was developed, that combines PCD, SS-PC, and SP-PCD techniques. The schematic representation of this carrier lifetime measurement setup is shown in Figure 6.2. The system is constructed using components of other commercial Semilab measurement systems, similar to those used in ingot tester tools, and incorporates multiple laser sources to enable advanced photoconductance measurement methods.

The system employs a high-intensity 915 nm CW laser with a power output exceeding 40W as the steady-state light source for SS-PC and SP-PCD methods. This laser provides sufficient penetration depth into silicon  $(1/\alpha \approx 39 \,\mu\text{m})$ , while its transmission remains minimal (<2%) for typical silicon wafers, ensuring high *OF*. A second laser, a 980 nm modulated CW laser, serves as the light source for PCD measurements and as the "small perturbation" illumination source for SP-PCD when operated at reduced power. Although the transmission of the 980 nm laser may exceed 10–20% depending on wafer thickness and surface conditions, its generation rate is not used in calculations. Moreover, the more uniform initial distribution of  $\Delta n(x)$  provided by the deeper penetration reduces the redistribution time required at the beginning of transients for samples with lower lifetimes. Both lasers are coupled into a shared homogenizing optical system via a combined optical fiber. This configuration produces a homogeneous illumination spot with a diameter of 3 cm, which minimizes distortions caused by lateral carrier spreading.

The detection system consists of a coil integrated into a printed circuit board positioned below the sample through a window in the holder. Illumination and detection are synchronized by a computer to ensure precise timing and data acquisition. The wafer holder is equipped with active cooling on both sides of the sample using a ventilation system to mitigate heating effects during high-intensity illumination. This thermal management ensures stable measurement conditions and reduces temperature-related influences on carrier lifetime measurements.

A schematic illustration of the measurement setup is presented in Figure 6.2., while Figure



Figure 6.3: Photo of the measurement setup for silicon wafers.

6.3. shows a photograph of the actual setup. Together, these figures provide a comprehensive overview of the design and functionality of the system.

## 6.2.1 Influence of the geometry of the light excitation

In general, the excess carrier distribution exhibits non-uniformity in both depth (z) and lateral (r) directions, with its profile evolving during PCD measurements. To accurately evaluate recorded decay curves, this inhomogeneity must be taken into account. However, for highlifetime solar wafers, where the excess carrier diffusion length significantly exceeds the wafer thickness and surface passivation is adequate,  $\Delta n(z)$  becomes sufficiently uniform to maintain accuracy in the derived carrier lifetime in all the three measurement methods. This section presents an investigation of lateral spreading and its contribution to measurable lifetime through both simulation and experimental results.

The impact of the size of the illuminated area on the accuracy of SP-PCD measurement was previously examined by Wilson et al. [145], who concluded that the spot size must be significantly larger than the carrier diffusion length to mitigate the effects of lateral spreading. To determine the necessary homogeneous spot size for accurate carrier lifetime and injection level measurements, we developed a simulation of one-dimensional (radial) carrier spreading. The ambipolar transport equation was employed in the following form:

$$\frac{\partial \Delta n(r,t)}{\partial t} = G(r,t) - \frac{\Delta n(r,t)}{\tau(\Delta n)} + D_{\text{amb}}(N_{\text{dop}},\Delta n) \cdot \nabla_r^2(\Delta n(r,t)), \tag{6.6}$$

where G(r,t) represents the generation rate (constant if  $t < t_0$  and  $r < r_0 = 1.5$  cm, otherwise zero),  $D_{\text{amb}}(N_{\text{dop}}, \Delta n)$  denotes the ambipolar diffusivity, and  $\tau$  is calculated from a  $\Delta n$ -independent defect-related  $\tau_{\text{SRH}}$  carrier lifetime and the Auger recombination lifetime.

Figure 6.4. presents the simulated PCD curves and the radial excess carrier profiles for an n-type sample with  $N_{dop} = 10^{15} \text{ cm}^{-3}$ . The profiles in Figure 6.4b. indicate that the inner



Figure 6.4: Simulated results of the PCD transient using optimized system parameters: (a) PCD curves, (b) radial carrier density profiles at given times during the PCD measurement, (c) comparison of the measurable and nominal carrier lifetime, (d) ratio between measurable and nominal lifetime during the PCD measurement, showing less than 1% difference at any  $\Delta n$ .

area with a radius of 1 cm remains unaffected by lateral diffusion throughout the transient PC measurement. Assuming a Gaussian-like lateral sensitivity distribution of the RF sensor centered at 0.65 cm radius, the measurable carrier lifetime can be determined. The comparison in Figure 6.4d. demonstrates that the theoretically measurable lifetime closely matches the initial carrier lifetime at any injection level, with differences below 1% during the PCD measurement. Further simulations predicted less than 2% systematic error in determined  $\Delta n$  for samples with  $\tau < 10 \text{ ms.}$ 

Based on these simulation results, a homogenizing optics was designed using collimator lenses and a diffuser. The final optics illuminates the sample surface with a 3 cm diameter spot, maintaining photon flux variation below 10% within the spot (Figure 6.5.). The eddy current coil, with 1.3 cm in diameter, is optimized to sense excess carriers only in the area with a homogeneous lateral excess carrier profile.



Figure 6.5: Characteristics of the optimized illumination system: (a) Intensity distribution at the sample plane, showing nearly 10% homogeneity across the illuminated area. (b) Photograph of the 3 cm diameter homogeneous light spot with the silhouette of the sensor overlaid.



Figure 6.6: Comparison of PCD measurements with different illumination conditions: (a) Measurements using inhomogeneous illumination, showing the influence of lateral spreading phenomena at different illumination intensities. (b) Measurements after optimization with homogeneous illumination, demonstrating perfectly merged transients regardless of illumination intensity.

Experimental PCD measurements comparing the homogenizing optics to a setup without it, but with a similar effective spot size, are presented in Figure 6.6. Thin Si wafers with long bulk lifetimes and decent passivation were used, allowing consideration of only the effect of the radially spreading  $\Delta n(r)$ . Measurements with different illumination intensities demonstrate the influence of lateral spreading phenomena in the case of inhomogeneous illumination (Figure 6.6a.). Insufficient light spot uniformity results in recorded  $\tau(\Delta n)$  curves that differ depending on the laser power used to generate excess carriers. During excess carrier decay, lateral diffusion leads to shorter measured carrier lifetimes, especially in the initial part of the transient.

In contrast, using the optimized optics with a homogenized light spot not only increases lifetime values but also results in perfectly merged PCD curves recorded at different laser powers (Figure 6.6b.). This experimental validation confirms the successful optimization of the measuring setup, providing lifetime accuracy within 1% across the entire range of interest.



Figure 6.7: (a) The illustration of the comprehensive measurement sequence, detailing: the application timing of the two lasers, (b) the changes in excess sheet conductance during the measurement procedure to obtain  $\sigma_{ss}(G_{ss})$  and  $\tau_d(G_{ss})$  plots, (c) the individual PCD measurement using the same modulated 980 nm laser that functions as the  $\tau_d$  probing laser for SP-PCD, but at higher intensity, (d) the determination of  $\tau_{PCD}(\Delta n)$  from the measured sheet conductance decay.

#### 6.2.2 Measurement procedure and temperature control

The measurement process integrates steady-state and small perturbation measurements within a single sequential cycle, as illustrated in Figure 6.7a. and b. The procedure comprises the following steps:

- 1. Activation of a high-intensity laser at generation rate  $G_{ss,1}$
- 2. Recording of steady-state photoconductance  $\sigma_{ss}$  upon signal stabilization
- 3. Application of a low-intensity light pulse  $(G_{p,1})$  to sample  $\tau_d$
- 4. Repetition of the sequence at the same laser intensity to enhance signal-to-noise ratio
- 5. Repetition of the measurement procedure at various steady-state excitation levels

This approach enables the acquisition of  $\sigma_{ss}(G_{ss})$  and  $\tau_d(G_{ss})$  plots.

Given that a single  $\tau_d$  sampling process can take several seconds, sample heating becomes a significant concern, particularly at high illumination intensities. To mitigate this issue, we implemented advanced ventilation and smart laser control. After each  $\tau_d$  sampling, the highintensity laser is switched off for a cooling period ( $t_{cooling}$ ). The duration of this cooling period



Figure 6.8: (a) Temperature rise during steady-state measurements at different illumination intensities without cooling. (b) Significantly reduced temperature variations after implementing ventilation and smart SS laser control.

is adjusted according to the applied intensity ( $G_{ss}$ ) and length ( $t_{ss}$ ) of the steady-state light pulse (Figure 6.7a.).

The effectiveness of this temperature management approach is demonstrated in Figure 6.8. Without laser control, continuous illumination with steady-state light can cause the wafer temperature to rise by tens of degrees (Figure 6.8a.), significantly altering lifetime and mobility measurements. By employing ventilation and smart laser control, temperature variations are constrained to less than  $4^{\circ}$ C even at very high light intensities (Figure 6.8b.), ensuring more reliable measurements.

The rapid PCD measurement is conducted separately after the combined SS-PC and SP-PCD measurements, as depicted in Figure 6.7c. and d. For PCD and SS-PC evaluation, the eddy current sensor signal is converted to sheet conductance based on a calibration using wafers of known resistivity (determined by 4PP method) and thickness. To minimize uncertainty, the thickness of the  $\sigma_{sh}$  calibration wafers closely matched that of the tested samples, addressing skin-effect-related issues described in [199].

This integrated approach ensures accurate and consistent measurements across various illumination intensities while minimizing thermal effects that could compromise data quality.

## 6.3 Advantages of the combined measurement method

In the preceding sections, I outlined the fundamental carrier lifetime measurement methods, their limitations across various  $\Delta n$  regimes, and presented the physical implementation of their integration into a single experimental setup. This section explores how this integration enhances the accuracy of the SP-PCD method, ultimately leading to the determination of  $\Delta n$ -dependent carrier mobility. This determination, in turn, improves the precision of both PCD and SP-PCD methods, allowing them to mutually enhance the accuracy of one another by providing physical parameters for the other method.



Figure 6.9: Comprehensive evaluation protocol for the SP-PCD measurements. (a) Determination of optical factor through generation rate matching with PCD method. (b) Pure exponential decay curve is obtained following a perturbing light pulse with suitably small intensity. (c-d) Determination of the initial actual lifetime  $\tau_1$  from the transient PC measurement. (e-f) Calculation of the accurate injection level and actual lifetime values for  $\Delta n > 10^{15} \text{ cm}^{-3}$ .

Figure 6.9. provides a comprehensive overview of the evaluation protocol for the SP-PCD measurement, supported by the PCD method. The subsequent subsections detail each step in the calculation of  $\tau(\Delta n)$  using the SP-PCD method, as demonstrated by the corresponding subfigures.

### 6.3.1 Determination of the optical factor

Precise optical factor data is essential for calculating  $G_{ss}$  from light intensity, typically determined through optical measurements. However, our integrated setup allows for more accurate data acquisition. Since the PCD measurement utilizes the same calibrated sensor, the measured signal and  $\sigma_{sh}$  corresponds to identical injection levels and generation rates, regardless of the specific measurement method employed.

An implied generation rate,  $G_{imp}(\Delta n) = -\frac{\partial \Delta n}{\partial t}$ , can be derived by evaluating the photoconductance decay. This should yield the same generation rate as the steady-state light source  $G_{ss}$ , which physically produces the same  $\sigma_{sh}$ . Assuming that individual PCD measurements are sufficiently accurate in certain injection level regimes, we can enhance the precision of the optical factor by taking the ratio of  $G_{imp}W$  (where W is the sample thickness) and the steady-state photon flux  $\Phi_{ph}$  (for SP-PCD and SS-PC measurements) corresponding to the same  $\sigma_{sh}$  (signal level):

$$OF = \frac{G_{\rm imp}W}{\Phi_{\rm ph}} \bigg|_{\sigma_{\rm sh,PCD} = \sigma_{\rm sh,SS}}$$
(6.7)

Given that the  $G_{imp}$  value in Equation 6.7 depends on the mobility model, we perform this adjustment at a suitably low injection level where  $\mu(\Delta n)$  mobility models exhibit good agreement and are therefore considered reliable. Figure 6.9a. illustrates an example of the *OF* determination process.

### 6.3.2 Resolving the contradiction of the SP-PCD method

The SP-PCD method relies on maintaining a small perturbation relative to the steady-state carrier density to determine the differential lifetime ( $\tau_d$ ) from the exponential decay of the signal. When the change in  $\Delta n$  generated by the perturbation light source is sufficiently small during the  $\tau_d$  sampling compared to the steady-state carrier density  $\Delta n_{SS}$ , variations in mobility and lifetime during sampling can be neglected (Wilson et al., 2012). Consequently,  $\tau_d$  corresponding to a given steady-state generation rate can be determined from a single exponential fit of the signal decay after switching the perturbation laser off, without relying on mobility models or PC sensor calibration. However, a larger perturbation ( $G_p$ ) leads to inaccurate  $\tau_d$  measurements due to significant changes in injection level, violating the small perturbation requirement. In contrast, a smaller  $G_p$  results in a low signal-to-noise ratio, making the determination of reliable  $\tau_d$  time-consuming and complicated.

To resolve this issue, we conducted a comprehensive study to determine the optimal  $G_p$  range using various samples and steady-state generation levels ( $G_{ss}$ ). We established that the optimal  $G_p$  range was approximately 10-20% of  $G_{ss}$  (Figure 6.10.). To ensure accuracy, we implemented a conservative 10%  $G_p/G_{ss}$  ratio for all  $\tau_d$  samplings during SP-PCD measurements. Under these conditions, the recorded SP-PCD transient exhibits purely exponential behavior, as demonstrated in Figure 6.9b.

The smallest steady-state laser intensity yielding accurate SP-PCD measurements defines the initial evaluation point ( $G_{ss,1}$ ). At this point,  $\Delta n_{ss}$  is approximately  $10^{15}$  cm<sup>-3</sup> due to the constraint of the  $G_p/G_{ss}$  ratio. In this range,  $\tau_{SRH}$  typically changes due to similar dopant concentration, rendering the actual lifetime ( $\tau_1$ ) at this injection level undefined. This uncertainty may reduce the precision of actual lifetimes at higher injections due to the integrated calculation of  $\tau$ . To address this, we derive  $\tau_1$  from the PCD measurement at the same  $\sigma_{sh}$ :

$$\tau_1(G_{\rm ss,1}) = \tau_{\rm PCD}(\sigma_{\rm sh,1}) \tag{6.8}$$

This approach matches lifetimes at equivalent signal levels (Figure 6.9c. and d). At this injection level,  $\tau_{PCD}$  is considered accurate due to minimal mobility changes.



Figure 6.10: Optimization of differential lifetime measurements. The graph shows measured differential lifetime as a function of probing laser power. The optimal range, where results are both accurate and have low noise, was found to be 10-20% of the steady-state laser intensity.

For steady-state illumination levels above  $G_{ss,1}$ , we calculate the actual lifetime through numerical integration of  $\tau_d$ :

$$\tau_{\text{SP-PCD}}(G_{\text{ss}}) = \tau_1 + \frac{1}{G_{\text{ss}} - G_{\text{ss},1}} \int_{G_{\text{ss},1}}^{G_{\text{ss}}} \tau_d(G'_{\text{ss}}) dG'_{\text{ss}}$$
(6.9)

After determining  $\tau_{\text{SP-PCD}}(G_{\text{ss}})$ , we calculate the accurate injection levels for  $\Delta n > 10^{15} \text{ cm}^{-3}$  using Equation 6.5 (Figure 6.9e.). This approach enables the determination of  $\tau_a(\Delta n)$  without relying on external mobility data (Figure 6.9f.).

### 6.3.3 Determination of charge carrier mobility

The combination of steady-state photoconductance (SS-PC) and small perturbation photoconductance decay (SP-PCD) methods provides valuable information about excess carrier mobility. Following the SP-PCD measurement sequence illustrated in Figure 6.9., one can record  $\Delta n(G_{ss})$  without relying on mobility models for the high injection level regime. Additionally, as shown in Figure 6.7b., the sheet conductance (and thus the conductivity  $\sigma(G_{ss})$ ) can be recorded simultaneously, allowing for the construction of  $\sigma(\Delta n)$ . From this purely experimental relationship, the sum of excess minority and majority mobilities  $\mu_{sum}(\Delta n)$  can be calculated.

Previous studies estimated  $\mu_{sum}(\Delta n)$  as the ratio of excess conductivity to injection level [104–106]:

$$\mu_{\text{sum}}(N_{\text{dop}},\Delta n) = \frac{\Delta\sigma}{e\cdot\Delta n}.$$
(6.10)

However, recent findings indicate that the correct formula for  $\sigma(\Delta n)$  must account for the mobility shift of majority carriers:

$$\sigma = e \cdot \left( \mu_{\text{sum}}(N_{\text{dop}}, \Delta n) \cdot \Delta n + \mu_{\text{maj}}(N_{\text{dop}}, \Delta n) \cdot N_{\text{dop}} \right).$$
(6.11)



Figure 6.11: (a) The ratio of  $\mu_n$  and  $\mu_p$  using the ratio of effective masses and the parametrization by Klaassen [98, 99]. (b)  $\mu_{sum}(\Delta n)$  measured by the multi-method carrier lifetime technique and evaluated using different  $f(N_{dop}, \Delta n) = \frac{\mu_n}{\mu_p}$  values.

Equation 6.11 incorporates both  $\mu_{sum}$  and the majority carrier mobility  $\mu_{maj}$ . Since photoconductance-based measurements cannot separate electron and hole conductance, we introduced the ratio of electron and hole mobilities  $f(N_{dop}, \Delta n) = \frac{\mu_n(N_{dop}, \Delta n)}{\mu_p(N_{dop}, \Delta n)}$ . This allows us to express  $\mu_{sum}$  from the measured  $\sigma(\Delta n)$  curve using  $f(N_{dop}, \Delta n)$  for p-type samples:

$$\mu_{\text{sum}}(N_{\text{dop}},\Delta n) = \frac{\sigma}{e \cdot \left(\Delta n + \frac{1}{1 + f(N_{\text{dop}},\Delta n)} N_{\text{dop}}\right)}.$$
(6.12)

For n-type samples, the inverse of the f value is used in the formula.

We estimated the  $f(N_{dop}, \Delta n)$  factor using two approaches. First, without using any theoretical mobility models, assuming f = 2.25, equal to the ratio of effective masses of electrons and holes. (using the effective mass values used for the calculation of density of states [27]). Second, using the semi-empirical mobility model proposed by Klaassen [98, 99] (Figure 6.11a.).

The mobility results computed using Equation 6.12 and applying these two  $f(N_{dop}, \Delta n)$  calculation principles are plotted in Figure 6.11b. for the same p-type sample used in Figure 6.9. Compared to uncorrected calculations, this compensation yields an increase of 2-5%  $\mu_{sum}$  assuming a constant f factor, with an additional increase of 3% using the Klaassen model-based correction, primarily at lower injection levels where the actual f value deviates significantly from 2.25.

It is important to note that this mobility calculation method is only applicable for ordinary recombination processes without significant carrier trapping phenomena. In materials where carrier trapping leads to inaccurately long evaluated lifetimes from PCD measurements, particularly in the low injection regime, this method may not be suitable. The monocrystalline samples used in our experiments do not exhibit perceptible trapping behavior, making them ideal for accurate carrier mobility determination.

To illustrate how the three combined lifetime evaluation methods complement and support each other, culminating in the calculation of the mobility sum, a Venn diagram has been constructed (Figure 6.12.). This diagram visually represents the interrelationships between the applied methods and formulas.



Figure 6.12: Venn diagram summarizing the applied carrier lifetime methods and formulas used to determine the mobility sum.

## 6.4 Results

We conducted measurements on various samples using the described method. To demonstrate the capabilities of the integrated system, we present results obtained from a-Si:H passivated p- and n-type mono-Si wafers. Figure 6.13a. and b. compare the carrier lifetime results of the SP-PCD method with those of the original PCD and SS-PC methods. For the latter two methods, we applied the parametrization of the widely used Dannhauser-Krausse mobility data. The consistency among the results obtained using these three techniques validates the reliability of the integrated system.

Figure 6.13c. and d. illustrate the sum of electron and hole mobilities as a function of the injection level. We compare these  $\mu_{sum}(\Delta n)$  results with the most widely used mobility models for lifetime measurements at room temperature (298 K). Our experimental mobility results from the "ideal" mono-Si p- and n-type wafers closely align with the Dannhauser-Krausse model, even at high injection levels. The minor deviations between mobility values account for the slight differences between SP-PCD and SS-PC lifetimes observed in Figure 6.13a. and b.

The excellent agreement among lifetime results from the three measurement methods, coupled with the close correspondence between mobility results and existing models, further confirms the reliability of our measurement setup and the presented combined technique.

We also compared our measured  $\mu_{sum}(\Delta n)$  curves to the model proposed by Zheng et al. [104], who employed a similar combined carrier lifetime approach using a flash-lamp system in



Figure 6.13: Comparison of carrier lifetime and mobility results: Carrier lifetime measurements using SP-PCD, PCD, and SS-PC methods for (a) p-type and (b) n-type passivated wafers. Sum of electron and hole mobilities as a function of injection level, compared with common mobility models for (c) p-type and (d) n-type passivated wafers.

its QSS and PCD modes. Our results demonstrate a closer alignment with other literature data [96, 97]. Moreover, our laser-based photoconductance measuring routine offers more universal applicability to virtually any typical modern PV wafer structures.

In conclusion, I presented a carrier lifetime measurement setup that integrates three distinct lifetime measurement principles: PCD, SS-PC, and SP-PCD, for the first time. This combination enables, to my knowledge, the most reliable and accurate determination of carrier lifetime, injection level, and mobility in silicon samples typically used in state-of-the-art solar cells. The use of this method leads to an excellent agreement between the lifetime and injection level results obtained from the three methods using the parametrization of Dannhauser-Krausse [96, 97] mobility data for PC measurements. Furthermore, the mobility results from standard mono-Si wafers closely align with predictions from previously published mobility models at room temperature. These findings collectively validate the reliability of our unified method, which was the focus of my work.

This technique can be applied for modern solar cell structures with conducting layers (highly doped surface emitter and rear side layers in PERC structures, highly doped poly-Si layer in TOPCon or transparent conductive oxide layer in HJT structures), where the sheet resistance of these layers must be known for accurate  $\Delta n$  calculation. This data is not always available

and not always easy to determine. The measurements using my calibration method enables to determine  $\tau(\Delta n)$  even on such wafer structures.

This measurement technique and platform also facilitate precise and comprehensive characterization of non-standard wafer types, such as wafers from different casting methods, compensated silicon or containing thermal donors, where current models can be inadequate.

## Chapter 7 Summary and Thesis Points

This dissertation focuses on the investigation of the recombination properties of silicon. Silicon-based semiconductor devices are the cornerstones of modern technology, and their continuous development is supported by a deeper understanding of charge carrier dynamics. The ongoing improvements in the efficiency of silicon-based solar cells, as well as the approach toward the Shockley-Queisser limit, necessitates a re-evaluation of previously employed empirical models and measurement techniques. In Chapter 1, I highlight the role of recombination lifetime in this process.

In Chapter 2, I provide a summary of the theoretical background necessary to contextualize my results. First, I describe the current status of solar cell technologies, with particular emphasis on state-of-the-art cell types. The characterization of the base material and the applied structures are the primary objective of my measurement technique developments. It is followd by the discussion of the dynamic properties of charge carriers in silicon. Recombination processes and diffuse motion of the charge carrier, and the related, commonly used characterization techniques are described in detail. Finally, I connect these topics by focusing on the recombination processes that most significantly influence the efficiency of silicon solar cells, such as the presence of impurities and surface recombination.

In Chapter 3, I present the equipments used for the measurements and introduce the devices that served as the foundation for my researches and developmentes regarding measurement techniques. In this chapter, I also describe the most important properties of the samples under investigation.

Chapters 4 through 6 summarize my key scientific achievements and the related measurement technique developments. Beyond presenting my own results, I emphasize the broader scientific significance of my work.

The novel, self-consistent calibration method introduced in Chapter 4 can be universally applied to study recombination in novel materials, thereby contributing to a better understanding of charge carrier dynamics in perovskite crystals in the ongoing research at BME.

The simulation-assisted evaluation presented in Chapter 5 is already utilized in the PV industry to enhance the reliability of quality control. Furthermore, it is the first method capable of characterizing actual bulk recombination processes in very high-quality silicon ingots without the distortion of surface recombination. Both industrial and R&D projects are ongoing to exploit this feature of the method.

The experimental method described in Chapter 6 enables determining the carrier lifetime versus injection level with unprecedented precision. This allows for the precise predictions of

the solar cell voltage prior to contact formation (eliminating the 5 - 10 mV uncertainty that has existed so far using standard methods). Furthermore, this technique enables carrier mobility measurements. The reliability of the method was confirmed using samples with well-described transport characteristics. In the same time, our results confirmed the validity of theoretical carrier mobility models for ordinary samples. However, this also provides important information for rather exotic materials, where existing mobility models are not valid. The potential of this method is being further explored through ongoing international collaboration projects.

My findings have already triggered superimposed researches. By modifying the combined carrier lifetime experimental setup, temperature-dependent carrier lifetime curves were recorded, leading to a successful MSc thesis by Gergely Havasi under my supervision.

My own results are summarized in the following Thesis points:

- I developed a self-consistent calibration method for a research-grade microwave-detected photoconductance decay measurement setup. This method is independent of the sample and the illumination spot size. I validated the method by measuring silicon wafers with different thicknesses and bare surface, where the measurable lifetime is limited by the diffusion process of charge carriers. This test also enabled to determine the low injection mobility of minority charge carriers down to 120K. [O1]
- 2. I have conducted a detailed investigation into the time-dependent behavior of surface recombination during photoconductance decay (PCD) measurements on thick silicon samples. I found that the distorting effect of the surface recombination on the measurement continuously diminishes during the decay of excess charge carrier density. I explored the severity of the surface recombination related distortions as a function of the wavelength of the optical excitation. Based on this analysis, I determined that the optimal wavelength range for characterizing thick silicon samples by PCD methods lies between 1050 - 1070 nm. I investigated the depth sensitivity of eddy-current-based measurements in the presence of inhomogeneous carrier distributions. This led to a phenomenological model that offers sufficient accuracy for practical applications. Charge carrier simulations and surface passivation experimental tests led to consistent results. The discrepancy between the measurable effective lifetime and the reference bulk lifetime value at the industrial standard  $10^{15}$  cm<sup>-3</sup> injection level is below 20% without using corrections of the surface recombination phenomena. [O2]
- 3. I developed a complex simulation method of the photoconductance decay (PCD) measurement of thick silicon slugs considering the charge carrier dynamics and the depth sensitivity of the eddy current sensor as well. I reconstructed the bulk lifetime from the measured decay curves using this simulation iteratively. This way I created a new evaluation method including the correction of surface recombination and the inhomogeneous depth sensitivity. I tested the method on p-type photovoltaic silicon samples, and found a very good agreement between the evaluated bulk lifetime and the lifetime results measured on well-passivated neighboring wafers. Measurements on n-type samples in the transitional thickness range also confirmed the accuracy of the method. This way, I characterized such samples in this early production phase with outstanding accuracy. [O3]
- 4. I integrated the photoconductance decay (PCD) and the small perturbation photoconductance decay (SP-PCD) carrier lifetime measurement methods into one measurement

setup. These techniques complement each other for accurate measurement in terms of injection level ranges. Therefore, their combination allows the investigation of passivated silicon wafers in a wide injection level range  $(10^{13} \text{ cm}^{-3} - 10^{17} \text{ cm}^{-3})$ . This self-consistent method does not require the exact knowledge of the optical properties of the sample and carrier mobility data. I investigated the injection-dependent lifetime of high quality silicon wafer structures used for modern solar cell types with the highest theoretically possible accuracy. [O4]

5. I realized steady-state photoconductance (SS-PC) carrier lifetime measurements on silicon wafers by complementing the measurement setup with a temperature stabilization method. Combining this technique with the transient PC measurement, I determined the charge carrier mobility of crystalline silicon in p- and n-type solar cell structures as well with higher accuracy than earlier studies, which applied less sophisticated methods. I compared the mobility results to accepted and widespread injection-dependent mobility models. [O5]

## **List of Publications**

- [O1] D. Krisztian *et al.*, "Novel calibration method for mw-pcd measurement," *in preparation*, 2025.
- [O2] D. Krisztian, F. Korsos, I. Saegh, G. Parada, M. Kovács, Z. Verdon, C. Jobbágy, P. Tüttő, X. Dong, H. Deng, S. Wang, and X. Chen, "Improved accuracy of eddy-current sensor based carrier lifetime measurement using laser excitation," *EPJ Photovoltaics*, vol. 13, p. 3, 01 2022.
- [O3] D. Krisztian *et al.*, "Determination of bulk carrier lifetime of silicon ingots using iterative simulation-base evaluation of pcd measurement," *in preparation*, 2025.
- [O4] D. Krisztian, F. Korsos, E. Kis, G. Parada, and P. Tüttő, "Integrated measurement of the actual and small perturbation lifetimes with improved accuracy," *AIP Conference Proceedings*, p. 110005, 01 2023.
- [O5] D. Krisztian, F. Korsos, and G. Havasi, "Simultaneous measurement of charge carrier concentration, mobility, and lifetime," *Solar Energy Materials and Solar Cells*, vol. 260, p. 112461, 09 2023.

## Other publications not included as thesis points

- B. Sánta, Z. Balogh, A. Gubicza, L. Pósa, D. Krisztián, Gy. Mihály, M. Csontos, A. Halbritter, Universal 1/f type current noise of Ag filaments in redox-based memristive nanojunctions. Nanoscale, 11, 10.1039/C8NR09985E (2019)
- B. Sánta, Z. Balogh, L. Pósa, D. Krisztián, T. Török, D. Molnár, Cs. Sinkó, R. Hauert, M. Csontos, A. Halbritter, *Noise Tailoring in Memristive Filaments*. ACS applied materials & interfaces, 10.1021/acsami.0c21156 (2021)
- L. Pósa, Z. Balogh, D. Krisztián, P. Balázs, B. Sánta, R. Furrer, M. Csontos, A. Halbritter, *Noise diagnostics of graphene interconnects for atomic-scale electronics*. npj 2D Materials and Applications, 5. 57. (2021)
- A. Bojtor, D. Krisztián, F. Korsós, S. Kollarics, G. Paráda, T. Pinel, M. Kollár, E. Horváth, X. Mettan, H. Shiozawa, B. Márkus, L. Forró, F. Simon, *Millisecond-Scale Charge-Carrier Recombination Dynamics in the CsPbBr*<sub>3</sub> *Perovskite*. Advanced Energy and Sustainability Research, 5. 10.1002/aesr.202400043 (2024)

- A. Bojtor, D. Krisztián, F. Korsós, S. Kollarics, G. Paráda, M. Kollár, E. Horváth, X. Mettan, B. Márkus, L. Forró, F. Simon, *Dynamics of Photoinduced Charge Carriers in Metal-Halide Perovskites*. Nanomaterials, 14. 1742. 10.3390/nano14211742 (2024)
- A. Bojtor, D. Krisztián, G. Paráda, F. Korsós, S. Kollarics, G. Csősz, B. Márkus, L. Forró, F. Simon, A Versatile System for Photoconductance Decay Measurement Across a Wide Range of Semiconductor Materials. 10.48550/arXiv.2411.16892. (2024)
- G. Havasi, D. Krisztián, F. Korsós, S. Fu, *Implied J-V Curves Recorded at Elevated Temperatures Using Light Controlled Heating*. SiliconPV Conference Proceedings, 2. 10.52825/siliconpv.v2i.1336 (2024)

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