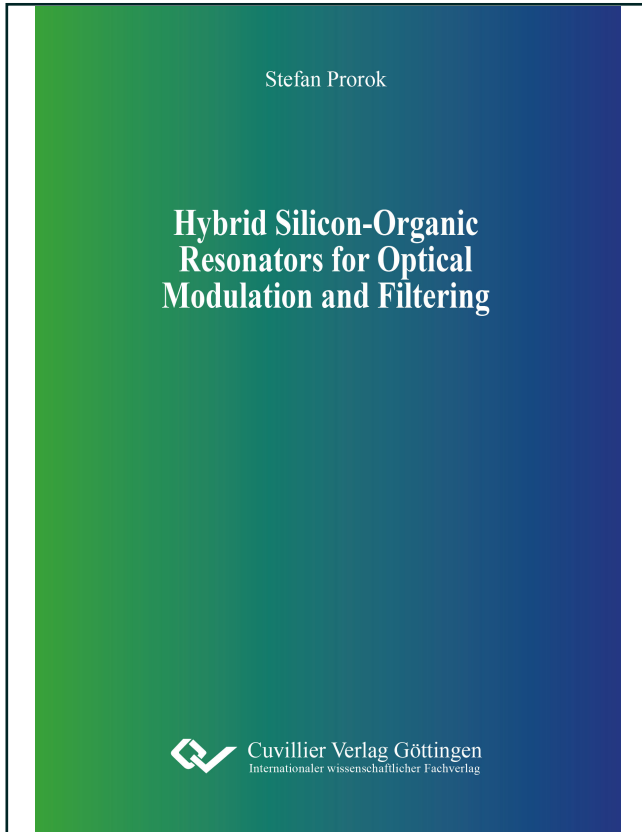




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**Hybrid Silicon-Organic Resonators for Optical  
Modulation and Filtering**



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# Chapter 1

## Introduction

Nowadays we live in a world where high speed network connection has become both vital and self-evident for us. Most urban areas in the industrialized world offer a dense coverage of Wifi or high-speed mobile service and the majority of households are equipped with broad band internet access. At the same time reliable internet access has become a strategic asset. Companies need it to communicate with customers and suppliers, states utilize it to gather intelligence data and private persons use it to maintain professional and personal networks.

All of this would not have been possible without major breakthroughs in optical communication. Compared to copper based communication optical systems feature larger available bandwidth, negligible cross talk between adjacent fibers and low propagation losses. Due to the expensive equipment optical communication systems were originally only used for long haul communication.

In the last couple of years the trend of *big data* has led to an economic pressure to develop optical communication systems for a rack to rack and chip to chip applications. The reason for this lies in the ongoing concentration of computing power in data centers. In modern data centers 25-35% of the electrical energy is consumed by interconnects between different racks and chips, not for computation [1, 2]. All major suppliers of server hardware (>80% market share [3]) are engaged in research activity in the area of optical transceivers revealing the importance of this problem [4, 5, 6, 7].

Even though optical transceivers, which are integrated on silicon chips, are already commercially available dense integration of optical components on the chip level remains challenging. This holds in particular for the electro-optic modulator that should fulfill the following four requirements:

- Large electrical modulation bandwidth
- Low electrical modulation amplitude
- Tolerance with respect to fabrication imperfections
- Small geometric footprint

In this thesis it will be shown how hybrid silicon-organic electro-optic modulators can be used to meet these requirements. Technological and scientific challenges of this material system will be discussed.

### **Outline of this Thesis**

This thesis focuses on the advancement of polymer infiltrated silicon photonic structures as integrated electro-optic modulators and filters. An emphasis is placed on modulators based on photonic crystal resonators, which have the potential to achieve highest integration densities and low power consumption at the same time. The thesis is structured as follows:

Chapter 2 introduces the physical background of this thesis. Basic concepts of nonlinear organic materials, photonic crystals and the numerical methods employed in this thesis are being presented.

Chapter 3 addresses the trimming of silicon ring resonators. This topic is of high technological relevance since so far random process fluctuations have strongly limited the commercial applicability of silicon ring resonators. It is shown that bleaching of a dye doped polymer cladding can be used to compensate these random process fluctuations and thereby allow a higher integration of ring resonators.

Chapter 4 describes how the bleaching approach can be extended to photonic crystal waveguides. It will be shown that by spatially resolved bleaching photonic

crystal waveguides can be modified in a flexible way. Using this method it is possible to define band edge filters and cavities with high quality factors.

Chapter 5 provides a detailed analysis of different aspects that determine the quality factor in photonic crystal heterostructures. Different approaches are presented, which significantly improve the quality factor in such structures.

Chapter 6 is focused on the experimental realization of integrated silicon electro-optical modulators. Electro-optic phase modulation in slotted waveguides and amplitude modulation in a photonic crystal cavity are demonstrated for low frequencies. Electro-optic modulation with GHz frequency is shown in a photonic crystal waveguide.

In chapter 7 the high field poling process with silicon electrodes is analyzed in order to identify possible reasons for the low electro-optic coefficients in slotted waveguides. The electrical conduction mechanisms in polymer thin films on silicon substrate have been studied and a theoretic model for the current conduction is presented. The results are complemented by electro-optic measurements utilizing a modified Teng-Man method. It is shown that the low poling efficiency on silicon substrates is to a large extent due to the electronic properties of the silicon surface. These properties can be influenced by interface modifications. It is shown that the modification of the silicon interface by a 5 nm thick layer of  $\text{Al}_2\text{O}_3$  leads to a significant improvement of the electro-optic coefficients.

Chapter 8 summarizes the results of this work and gives an outlook on future research.



# Chapter 2

## Basic Concepts and Numerical Methods

This chapter provides an overview on the state of the art in the field of silicon based electro-optical modulators. Furthermore basic theoretical concepts of nonlinear electro-optic polymers and photonic crystals are introduced. The numerical methods that were used in this work are discussed briefly.

### 2.1 State of the Art in Silicon Electro-Optic Modulators

Several different physical effects can be utilized in order to create silicon based electro-optic modulators. Silicon has a reported thermo-optic coefficient of  $1.85 \cdot 10^{-4}/\text{K}$  at room temperature [8]. Hence, moderate temperature changes yield a refractive index change in the order of  $10^{-3}$ , which is sufficient for modulation. However, thermo-optic devices show switching speeds in the order of 10 to 100  $\mu\text{s}$ , which is not suitable for fast electro-optic modulation [9]. Another effect to be mentioned is the Franz Keldysh effect, which describes a change in the absorption spectrum of the material upon applied electric field. In undoped silicon the changes in absorption and refractive index due to the Franz Keldysh effect are below  $0.1/\text{cm}$

and  $10^{-5}$  respectively [10], which is too small for efficient electro-optic modulation.

The most common way to achieve electro-optic modulation in silicon is by means of the plasma dispersion effect of free charge carriers. In semiconductors as silicon the refractive index and absorption can be changed by altering the amount of free charge carriers in the material. Free charge carriers lead to enhanced linear material losses due to free carrier absorption. At the same time free charge carriers reduce the refractive index of silicon [10]. Early works on electro-optic modulators using free carrier absorption have been performed by Treyz et al. in 1991 [11]. The electrical bandwidth of this modulator was limited to 20 MHz.

The first electro-optic modulators using the refractive index change due to the plasma dispersion effect have been realized by Lorenzo et al. in 1987 [12, 13]. However, for a long time the speed of silicon modulators has been limited to electric bandwidths below 1 GHz. In 2004 Liu et al. realized the first GHz electro-optic modulation in a silicon waveguide [14]. The works of Lorenzo and Liu relied on the injection of charge carriers into a weakly doped core region.

Using charge injection in order to change the refractive index of a silicon structure imposes limitations on the modulation frequency. While the depletion of charge carriers is a field assisted process and can be as fast as 7 ps the injection is a diffusion process, which has time constants in the order of nanoseconds [15, 16]. In 2007 the first modulator was realized that relied only on the depletion effect [17]. This modulator featured a modulation band width of 30 GHz. Since then a lot of effort has been done in order to further optimize the modulation band width. Several modulators with modulation speeds up to 50 GHz have been shown in the last couple of years [18, 19, 20].

Despite the progress in the field of silicon modulators a fundamental challenge remains. For modulators with large modulation bandwidth high doping concentrations are required, which increase the insertion loss of the modulators. One way to remedy this problem is to combine silicon waveguides with a second material. In 2005 Baer-Jones et al. introduced the concept of a slotted silicon waveguide that is infiltrated by a nonlinear electro-optic polymer [21]. In their concept the light is confined by two silicon rails, which are separated by a slot of 200 nm or less. A

significant part of the optical power, however, is guided in the low refractive index polymer.

In contrast to silicon nonlinear polymers can exhibit a very large Pockels effect. Due to recent advances in molecular engineering Pockels coefficients of more than 300 pm/V have been achieved [22]. This value is 10 times higher than that of lithium niobate, which is still the standard material for inorganic electro-optic applications [23]. Furthermore, polymers are compatible to standard complementary metal-oxide-semiconductor (CMOS) technology and can be readily integrated into existing fabrication processes for electronic circuits [24]. Witzenz et al. have shown theoretically that EO polymers in slotted waveguides can have a significantly smaller device footprint as compared to modulators using plasma dispersion as modulation effect [25]

So far the most compact electro-optic modulator based on silicon slotted waveguides and electro-optic polymer reached a value of  $V_\pi l$  as low as 5 Vmm but the reported device has only a modulation bandwidth of 1 kHz [26]. Slotted waveguide modulators with 3 GHz modulation bandwidth and a  $V_\pi l$  of 8 Vmm have been realized by Ding et al. [27]. For comparison, the most compact silicon modulator based on the plasma dispersion effect has a  $V_\pi l$  of 0.36 Vmm, which is about 14 times smaller than the best slotted waveguide based modulator [28]. The main reason for the comparatively large value of  $V_\pi l$  in slotted waveguides is the small in device electro-optic coefficient of the polymer. This aspect will be covered in detail in the chapters 6 and 7.

In order to further reduce the modulator footprint the interaction between the optical wave and the change in refractive index must be enhanced. One way of achieving this is to let the light propagate through a material or waveguide with slow group velocity. Monat et al. have reported on the enhancement of free carrier absorption and Kerr nonlinearities in silicon slow light waveguides [29]. Lin et al. have used slotted slow light waveguides in order to enhance the electro-optic effect [30]. They have reported on an increase of the electro-optic modulation signal by 23 dB. The resulting modulator exhibited a  $V_\pi l$  of 0.56 Vmm. On the other hand, the slow light effect limits the available modulation band width [31]. Li et al.



have systematically studied the bandwidth limitations for silicon photonic crystal waveguides [32]. They found that by engineering the properties of the photonic crystal lattice waveguides with low dispersion and group velocities as low as  $\frac{c}{49}$  can be created, which have optical bandwidths of 9.5 nm.

Another way of increasing the refractive index sensitivity of a modulator is to use resonant structures. Ring resonators with embedded pn-junctions as well as hybrid polymer silicon ring resonators have been used as electro-optic modulators [33, 34]. While the modulation amplitude obtained for resonant structures are comparable to those found in integrated interferometers the device footprint of a resonant modulator can be as small as 12  $\mu m$  [33]. In resonators the required modulation voltage is inversely proportional to the quality factor  $Q$  of the cavity, therefore in principle the required modulation voltage can be arbitrarily low [35]. This property makes optical resonators interesting for densely integrated electro-optic modulators.

## 2.2 Second Order Nonlinear Effects in Organic Materials

In order to understand the physical principles behind the electro-optic effect in nonlinear polymers it is most intuitive to first look at the properties of these materials at the molecular level. The knowledge obtained from the molecular analysis can then be transferred to the macroscopic level where it is possible to operate with effective material parameters.

### 2.2.1 Microscopic Origin of the Electro-Optic Effect

Most state of the art nonlinear polymers are either guest-host systems or copolymers in which chromophore molecules are embedded in an inactive polymer matrix [36, 37, 38]. The chromophore molecules exhibit a donor and an acceptor part. These two parts of the molecule are connected by a conjugated chain of carbon bonds that allows charge carriers to move along the molecule. Due to the



high mobility of charge carriers along the conjugated chain chromophore molecules are highly polarizable. The molecular polarization  $p$  induced by an external electric field  $E_n$  can be written as

$$p = \alpha_{ij}E_i + \beta_{ijk}E_iE_j + \gamma_{ijkl}E_iE_jE_k\dots \quad (2.1)$$

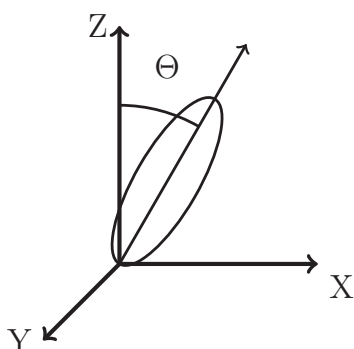
where  $\alpha_{ij}$  describes the linear polarizability of the molecule. The terms  $\beta_{ijk}$  and  $\gamma_{ijkl}$  are tensors, which describe the first and second order hyperpolarizabilities of the molecule that give rise to first and second order non-linear effects. In this thesis the discussion will be limited to the first order hyperpolarizability. Higher order nonlinear effects are not considered.

In order to create materials with a strong first order nonlinearity the chromophore molecules should exhibit a large hyperpolarizability  $\beta$ . Oudar and Chemla have proposed a model that relates  $\beta$  to the charge transfer inside the chromophore molecule with

$$\beta(-\omega_3; \omega_1, \omega_2) = 2 \frac{e^2(\mu_e + \mu_g)\mu_{ge}^2}{\Delta E^2} \cdot \frac{\omega_{ge}^4(3\omega_{ge}^2 + \omega_1\omega_2 - \omega_3^2)}{(\omega_{ge}^2 - \omega_1^2)(\omega_{ge}^2 - \omega_2^2)(\omega_{ge}^2 - \omega_3^2)}. \quad (2.2)$$

$\mu_g$  and  $\mu_e$  denote the dipole moments of the ground state and the excited state.  $\mu_{ge}$  and  $\Delta E$  describe the transition dipole moment and the energy difference between ground state and excited state.  $\omega_{ge}$  is the resonance of the dipole transition [39, 40].  $\omega_1$  and  $\omega_2$  are the frequencies of the external electric fields  $E_n$  and  $\omega_3$  is the frequency of the resulting nonlinear polarization  $p$ .

Equation 2.2 contains two fractions. The first fraction only depends on the dipole moments and the energy difference between ground state and excited state. This part of the hyperpolarizability is frequency independent. The second fraction only depends on the resonance frequency of the dipole transition. This part of the hyperpolarizability is frequency dependent and models the dispersion of the hyperpolarizability. As a certain frequency separation between the dipole resonance and the optical fields is necessary in order to avoid linear losses increasing the frequency independent part of the hyperpolarizability is the cardinal goal in the synthesis of novel nonlinear materials.



**Figure 2.1:** shows the schematic of a chromophore molecule inside the laboratory coordinate system  $XYZ$ . The molecule is depicted by an ellipsoid and the axis of the conjugated  $\pi$  system is denoted by an arrow.

More recent studies have shown that the bond length alteration (BLA) between single and double carbon bonds is correlated with the hyperpolarizability of chromophore molecules [41]. Quantum mechanical calculations have revealed that many common chromophore molecules had BLAs, which were too large. The BLA is mainly determined by the ground state polarization induced by the donor and acceptor part of the chromophore molecule but also the host material can influence the BLA [42]. The optimization of structure parameters such as the BLA have lead to a significant improvement of the attained hyperpolarizabilities [43]. However, Kuzyk has pointed out that the structure parameter guided approach is a heuristic one and does not necessarily lead to optimal results. Molecules with close to optimal nonlinearities therefore require full quantum mechanical calculations [44].

In general, chromophore molecules can have different dipole transitions in different directions of the molecule. However, since many chromophore molecules for nonlinear applications exhibit a rod-like structure, it is often valid to assume only one dominant dipole moment that is aligned with the conjugated  $\pi$ -system of the chromophore. Under this assumption the hyperpolarizability tensor also has only one dominant component  $\beta_{zzz}$ , which is aligned with the axis of the conjugated  $\pi$ -system. Figure 2.1 shows how the chromophore orientation is defined in the laboratory coordinate system  $XYZ$ .

Provided that then hyperpolarizability can be described sufficiently well by the dominant component  $\beta_{zzz}$  the macroscopic second order susceptibility tensor of the material  $\chi^{(2)}$  contains two independent components, which are given by

$$\chi_{ZZZ}^{(2)} = N f_L(\omega_1) f_L(\omega_2) f_L(\omega_3) \beta_{zzz} \int_V \cos^3(\Theta) f(\Theta) d\Theta \quad (2.3)$$

and

$$\chi_{ZXX}^{(2)} = \frac{1}{2} N f_L(\omega_1) f_L(\omega_2) f_L(\omega_3) \beta_{zzz} \int_V \sin^2(\Theta) \cos(\Theta) f(\Theta) d\Theta \quad (2.4)$$

$f(\Theta)$  is the distribution function of the chromophore molecules.  $N$  and  $f_L$  are the number density and the local field factors for the fields external electric fields  $E_i(\omega_1)$  and  $E_j(\omega_2)$  and the nonlinear polarization  $p$  [45]. For quasi static electric fields the local field factor is given by the Onsager expression

$$f_L(\omega) = \frac{\epsilon(n^2 + 2)}{(n^2 + 2\epsilon)} \quad (2.5)$$

where  $\epsilon$  is the dielectric constant and  $n$  is the refractive index. For optical frequencies the local field factor is better described by the Lorentz field factor

$$f_L(\omega) = \frac{n^2(\omega) + 2}{3} [46]. \quad (2.6)$$

If the chromophore molecules are isotropically oriented the second order susceptibility becomes zero. Thus the material exhibits no first order nonlinearity at the macroscopic scale. In order to obtain a macroscopic first order nonlinearity it is necessary to break the centrosymmetry of the system by aligning the chromophore molecules.

The most common way to achieve an alignment of the chromophore molecules along one direction is to introduce a poling step. Different types of poling are known such as contact poling via electrodes, corona poling where the field is applied by a plasma and laser induced poling [47, 48, 49]. However, all of these methods involve heating the polymer up to the glass transition temperature and applying a strong electric field  $E_p$ . At the glass transition temperature the chromophore molecules become mobile. The torque of the applied electric field acts on the chromophores ground state dipole moment and induces a uniaxial alignment of the