Chapter 1

Introduction

The globalized world of today is unimaginable without optical lightwave communication technologies. The exchange of information in a fast and flexible way is of substantial importance for the current economy. The first generation of lightwave systems, operating at 0.8 µm, became commercially available in 1980 [San78]. They provided a bit rate of 45 Mb/s and a repeater spacing of about 10 km. During the subsequent two generations, the dispersion minimum of silica single-mode fibers at 1.3 µm and InGaAsP-laser sources have been utilized to enhance the performances of optical communication systems by orders of magnitude [Sip95]. The fourth generation of systems, which represents the current state of the art technology, operates at 1.55 µm, benefiting from a loss minimum of silica fibers in this particular spectral domain [Miy79]. Wavelength-division-multiplexing (WDM), allowing synchronous signal transmission, and Erbium-doped optical amplifiers, providing simultaneous multichannel amplification, gives rise to transoceanic systems, which can transmit information at a total bit rate of more than 1 Tbit/s over a distance of 10000 km [Kah01], [Hei00], [Agr04].

Processing of the data streams is ensured by supplying additional components like modulators, photodetectors, WDM-elements and opto-optical switches. For the discussion in the present thesis, optical modulators and filters are of major interest. Such filters are primarily employed in current communication systems to address one particular WDM-channel. Since the characteristic distance between two adjacent channels is about 0.8 nm (@ 1550 µm), high spectral selectivities have to be guaranteed. In this sense, active data processing is provided by tunable optical filters, given the fact, that the spectral location of the pass-band of such structures can be manipulated, e.g., thermal-optically [Iod03], electro-optically (EO) [Kam74] or strain-induced [Mal03]. Typically, these devices are composed of high-Q resonators or waveguides and based on the widespread planar semiconductor technology [Man99], [Alm04]. A similar functional principle is used in optical modulators, where the spectral properties of the devices are modified by external voltages [Liu04a]. In such active components, the electrical signals to be transmitted are basically impressed on optical carrier waves. Since beyond 10 GHz, copper interconnects become bandwidth limited due to frequency-dependent losses, down-scaling of optical modulators potentially lead to effective on-chip and chip-to-chip communication concepts based on lightwave technology. These integration concepts, which emphasize the importance of nanostructured devices, represent the most promising approaches to overcome the intrinsic problems of metalized interconnects used in current chip architectures [Moh04], [Her04].

Clearly, the maximum working frequency defines the performance of the modulator considered, therefore representing the bottleneck in current communication systems. Direct modulations of the laser sources are not reasonable at high bitrates ($\geq$ 10 Gb/s), since variations of the input currents impose undesired phase shifts and, thus, deformations of the signals to be impressed [Agr04]. In today's networks, optical modulators, which do not exhibit such drawbacks, are realized by electro-absorption (EA) based devices [Dah04] and Mach-Zehnder-Interferometers (MZI) [Dal03]. The EA-effect utilizes a change of the optical absorption, induced by an applied voltage. In detail, the size of the electronic band gap of the employed semiconductor quantum well is reduced in the presence of an external electric field. Thus, photons of energies slightly smaller than the band gap cannot travel through semiconductor, whereas they can pass in the absence of the field (Franz-Keldysh-effect) [Agr04]. These devices suffer from undesired wavelength selectivities, which cause problems in mass production, and large modulator chirps due to additional induced refractive index changes. These crucial issues do not emerge in active MZI-modulators, which utilize the Pockels-effect. This second-order nonlinear optical (NLO) effect is a three-wave mixing process based on the interaction of two optical waves with a quasi-static electric field. Due to the fact, that the employed NLO-media are non-resonant in the spectral regime of operation, externally induced refractive index changes based on Pockels-effect are inherently ultrafast [Lee02b]. The fabricated modulators generally consist of interferometers, where the refractive index in each arm can be modified by applied voltages. Consequently, the degree of interference and therefore the output intensity is controlled externally by the electric field strength. Typically, these devices have been fabricated from lithium niobate ($LiNbO_3$) single crystals, exhibiting a relatively large EO-response [Kam74]. For instance, waveguiding structures are realized in this material by e.g. titanium diffusion, leading to permanent local refractive index changes [Das03]. However, the large difference between the dielectric permittivity in the low-frequency ($\approx$ 100 GHz) and optical ($\approx$ 200 THz) regimes, gives rise to strong mismatches between optical and microwave fields, reducing respective interaction lengths significantly. Relatively large modulation voltages are required, since the electric field strength inside the NLO-medium is reduced strongly due to the large dielectric permittivity of $LiNbO_3$ additionally implying potential power dissipation. So far, no feasible nanostructuring technique with acceptable aspect ratios has been reported excluding $LiNbO_3$-modulators for applications in integrated optics.

A system, which does not exhibit such intrinsic drawbacks, is represented by NLO-polymers [Eic89b], [Bur94], [Dal03], [Mar97]. These amorphous organic materials basically consist of high-glass temperature polymers, doped or functionalized by NLO-active chromophores. These chromophores, which are composed of π-electron systems enclosed by acceptor and donor endgroups, naturally contain unharmonic intramolecular potentials, due to the asymmetric molecule compositions [Gue00]. In order to transfer the microscopic nonlinearity into macroscopic EO-activity, a non-centrosymmetric orientation of the chromophore ensemble is typically achieved by aligning the NLO-constituents along a predefined direction. Extremely high EO-coefficients up to 100 pm/V and even more have been obtained in polymeric systems, which are about a factor 4 larger than that of $LiNbO_3$ [Luo03], [Luo02], [Ma01], [Gar04], [Zha00]. The dielectric permittivities at low and optical frequencies are roughly equal and independent of wavelength. Moreover, polymeric materials generally show very low waveguide losses, large transparency ranges, moderate refractive indices, and they can be deposited as thin films on any flat surfaces, especially on silicon wafers [Dal03]. Currently, fast polymeric MZI-modulators with very low switching voltages have been realized [Zha01], [Shi00b], [Dal99a], [Che97], whereas promising laboratory experiments indicate an extension of the operation frequency in the THz-domain [Lee02b]. Also, first devices are commercially operating at 40 Gb/s are commercially available [Pac05].

However, these structures own intrinsic dimensions of a few centimeters, excluding them from potential applications in integrated optics. In this sense, an auspicious concept is represented by Photonic Crystals (PCs), since they provide an enormous integration density, required for ultracompact optical devices. PCs, which were introduced by Russell [Rus86b], Zengerle [Zen87], Yablonovitch [Yab87], and John [Joh87], apply the concept of using periodic dielectric functions in space in more than one dimension. Such regular refractive index modulations lead to band structures of the same dimensionality, comparable to that of electrons, resulting from the periodic atomic potentials [Yab93]. In both cases, the eigenfunctions are given by Bloch functions with wavelengths on the scale of the periodicity. If the dielectric lattice geometry is chosen appropriately, a frequency range opens in the band structure, blocking light of respective wavelengths [Joa97]. This range is called the photonic band gap (PBG), and is one of the most important characteristics of PCs, because it allows to manipulate the propagation of light within a few lattice constants. By doping PCs with isolated defects, allowed states may be created inside the PBG, resulting in high-Q cavities [Sau05], [Aka05], [Ryu03], [Aka03] and straight waveguides [Lin98], [Joh00], [Son00]. Moreover, guiding of light around sharp corners has been demonstrated both, theoretically and in the experiment [Aug03], [Chu02], [Tal02]. Superprisms with enormous angle and spectral selectivity, based on the anisotropic isofrequency dispersion surface of PCs, can be realized [Not02], [Bab02], [Kos98]. Dispersion compensation in such structures is provided by coupled cavities [Sug02] and single straight defect waveguides [Pet05].

For integrated optics, finite two dimensional (2D) PCs play a key role, because planar slab structures may be structured fairly easily, using current fabrication technology. The in-plane propagation of light is governed by its interaction with a

2D-lattice of air-holes etched through the slab, whereas vertical confinement is facilitated by total internal reflection. Slab waveguide cores thus are required to have a higher refractive index than substrate and cladding. For stability reasons and ease of fabrication, planar PCs should have non-air substrates and hence are in most cases vertically asymmetric. Choosing an air-hole array, the functionality of the PCs is limited to one polarization, nevertheless showing an in-plane, direction independent stop gap for triangular lattices [Joh99], [Joa95]. Most 2D-PCs realized today are based on semiconductor materials (GaAs, InP or Si), achieving a large horizontal refractive index contrast and thus a wide PBG [Tan04], [Mon01], [Vla04]. Waveguide cores for single mode operation are around 0.25 µm thick, resulting in a strong coupling mismatch between standard optical single-mode fibers and PCs. Therefore, polymeric PCs become attractive, since larger core thicknesses of about 1.5 µm and smaller refractive indices correspond much better to the dimensions and index of single-mode silica fibers [Kee05], [Sch05a], [Bou04], [Lig01]. Moreover, the smaller refractive index of polymers potentially reduce the influence of Rayleigh-scattering due to incorporated irregularities.

Beside applications in EO-switches, polymeric systems may be also employed to generate light of desired wavelengths by the use of Second-Harmonic-Generation, since they potentially exhibit very high nonlinear coefficients. Due to the fact, that the current integrated laser systems do not cover the entire visible spectral regime, PCs consisting of NLO-polymers may be applied to generate electro-magnetic waves of the missing wavelengths by frequency doubling of infrared light emitted by GaAs-lasers. Therefore, integrated coherent light sources may be fabricated by combining periodic dielectric waveguide structures with on-chip semiconductor lasers based on nonlinear three-wave mixing processes. This concept provides the realization of supplementary devices to state of the art integrated systems, creating light of particular wavelengths, which is not accessible by e.g. GaN- or GaAs-lasers.

The main motivation of this doctoral thesis was to investigate PC-structures consisting of polymeric NLO-materials. Thus, the issues PCs and NLO-polymers previously introduced are combined within the scope of this work. Due to the relatively low refractive index contrast between dielectric and air, the properties of these novel microstructures are to a great extend unexplored so far, since the entire PC-community almost concentrates their investigations on semiconductor materials. The primary objective of this dissertation is to give a comprehensive study of the linear and especially NLO-properties of polymeric PC-structures. Both, experimental and theoretical aspects, will be elaborated and discussed qualitatively and, in particular, quantitatively, showing fundamental properties and potential applications. In this sense, the central questions to be addressed in the context of this thesis are:

- What are the fundamental characteristics of polymeric NLO-PCs?
- Are high-Q cavities feasible in polymeric PCs?

- Could the potentially high hyperpolarizability be effectively exploited for NLO-phenomena?
- How do irregularities affect the optical properties of polymeric NLO-PCs?
- Can polymeric NLO-PCs may be used as electro-optical switches and for efficient frequency doubling?
- Do polymeric PCs represent a better alternative to semiconductor systems?

The thesis is structured as follows: In the first three subsequent chapters (Chap. 2, Chap. 3, and Chap. 4), the underlying theory and simulation methods, the employed materials and the measurement techniques are reviewed precisely. Chapter 5 discusses the theoretical and simulated results. The elaboration begins by evaluating the linear and NLO-characteristics of one-dimensional PCs, where a self-developed code based on the Transfer-Matrix-Method has been utilized [Pen94]. Subsequently, the main focus is concentrated on the optical properties of polymeric PC-line-defect- and PC-ridge-waveguide-resonators, simulated by a program code commercially available. The entire discussion is primarily aimed at the quality factor and the resonance transmittance of the cavities. The investigations presented in the next chapter (Chap. 6) are dedicated to the experimental results gained during the time of the doctoral thesis. First, the fundamental transmission properties of polymeric PCs consisting of Teflon substrates are discussed, where mirror losses and Q-factors are determined. Then, the concept of quasi-air-bridge PCs, based on the application of ultralow refractive index substrates, is discussed intensively. The next part of this chapter introduces a scheme, which may be used to compensate for systematic fabrication induced inaccuracies by UV-photobleaching. Finally, the results referring to electro-optically tunable PCs are presented. A detailed experimental and theoretical investigation reveals, that EO-modulation based on the Pockels-effect has been observed in a PC-system for the first time. Chapter 7 concludes the dissertation and gives a detailed outlook for future investigations of polymeric PC-structures.

Chapter 2

Basic Theory and Simulation Techniques

The objective of the present work is the elaboration of PCs, which are composed of polymeric slab waveguides. In this chapter, the explanation of the simulated and experimental results is provided by a detailed introduction of PCs of different dimensionality and geometry. In particular, the basic equations of linear and nonlinear optics are introduced followed by a discussion of the asymmetric dielectric slab waveguide. The theoretical background of one- and two-dimensional PCs is presented in the next part showing the basic relationships and calculation techniques. Finally, the characteristics of resonant structures are discussed generally and on the concrete example of the Fabry-Perot resonator.

Since nearly all materials utilized in the thesis are of organic origin, the diagrams of this chapter refer to polymer parameters. Especially the parts discussing the microscopic properties are dedicated to functionalized polymer systems.

2.1 Maxwell Equations

In the non-relativistic domain, all electro-magnetic (EM) phenomena are fundamentally explained by the four Maxwell equations supplemented by two consecutive material expressions. This introductory section recalls the basic relationships of electrodynamics forming the theoretical background of the present thesis. Any expressions derived in the work can be attributed to this set of equations. In this sense the book of Jackson should be mentioned [Jac82], which represents one of the best introductions to EMs and optics.

Basically, the phenomenon of light is classically described by traveling waves consisting of electric and magnetic fields. These waves, which are mainly characterized by their amplitude, polarization, phase and k-vector, are general solutions of

Maxwell Equations (MEs):

$$\begin{aligned}\nabla \cdot \mathbf{D} &= \rho_e & (2.1)\\ \nabla \cdot \mathbf{B} &= 0 & (2.2)\\ \nabla \times \mathbf{H} &= \frac{\partial}{\partial t}\mathbf{D} + \mathbf{J} & (2.3)\\ \nabla \times \mathbf{E} &= -\frac{\partial}{\partial t}\mathbf{B} & (2.4)\end{aligned}$$

with the dielectric displacement field $\mathbf{D}$, the electric charge density ρ_e, the magnetic induction $\mathbf{B}$, the magnetic field $\mathbf{H}$, electric current density $\mathbf{J}$ and the electric field $\mathbf{E}$. These four first-order partial differential equations, which have been proposed by Maxwell in 1865,[1] form the mathematical background covering the entire spectrum of EM-phenomena in the classical wave picture. To account for interaction between light and matter, two constitutive expressions including the electric and magnetic properties of the respective media have to be supplemented to MEs:

$$\begin{aligned}\mathbf{B} &= \widetilde{\mu}\mu_0\mathbf{H} & (2.5)\\ \mathbf{J} &= \widetilde{\sigma}\mathbf{E} & (2.6)\end{aligned}$$

with the relative permeability tensor $\widetilde{\mu}$, the permeability of vacuum μ_o and the electrical conductivity $\widetilde{\sigma}$. The materials of interest for the current discussion are assumed to be of dielectric origin meaning that they are non-ferromagnetic ($\widetilde{\mu} \approx \widetilde{1}$) and non-conducting ($\widetilde{\sigma} \approx 0 \Rightarrow \mathbf{J} = 0$) in the relevant wavelength regime. Additionally, no macroscopic charge density should be localized inside the materials ($\rho_e \approx 0$). This leads to the definition of the dielectric displacement field $\mathbf{D}$, which represents the shifted charge density in a medium under the influence of an electric field:

$$\mathbf{D} = \epsilon_0\mathbf{E} + \mathbf{P} \tag{2.7}$$

with the macroscopic polarization $\mathbf{P}$ and the permittivity of vacuum ϵ_0. Substituting this expression into the MEs results in the essential wave equation (WE) for the electric field:

$$\Delta\mathbf{E} - \frac{1}{c_0^2}\frac{\partial^2}{\partial t^2}\mathbf{E} = \frac{1}{\epsilon_0 c_0^2}\frac{\partial^2}{\partial t^2}\mathbf{P} \tag{2.8}$$

with c_0 equal to the speed of light in vacuum. This second-order partial differential expression describes an electrical field $\mathbf{E}$, which is generated by the polarization $\mathbf{P}$.

Elementary solutions of the WE are plane waves ($\mathbf{E} = \mathbf{E}_0 \exp(i(\mathbf{kr} - \omega t))$) consisting of the wave vector $\mathbf{k}$, which is perpendicular to the phasefronts and the angular frequency ω. The polarization of an EM wave is defined by the direction of the electric field amplitude vector $\mathbf{E}_0$. Considering a plane wave in vacuum propagating along the x-direction of a cartesian coordinate system, the complex field

[1] Maxwell substituted $\mathbf{J}$ by $\mathbf{J} + \frac{\partial}{\partial t}\mathbf{D}$ in Eq. 2.3. This crucial replacement gives rise to traveling wave solutions and therefore electrodynamic phenomena.

amplitude $\mathbf{E}_0$ has two independent components ($\mathbf{E}_0 = (0, E_y(x,y,z), E_z(x,y,z))$). By definition an optical wave is called linearly polarized, if the phase difference between the two elements is a multiple of π or one of the components is zero, since the electric field always oscillates in the same plane. For a finite but temporal constant phase difference the head of the electric field vector moves generally on an ellipse defining the elliptical polarization state. Especially in the case of $|E_y| = |E_x|$ and a phase difference of $\pi/2$, the polarization is called circular, since the ellipse degenerates to a circle. A temporal statistical fluctuating phase difference results in unpolarized light, which is the typical polarization state of light emitted from thermal lamps.

Additionally, it should be noted that the WE (Eq. 2.8) is linear in $\mathbf{E}$ and $\mathbf{P}$. Thus, the solutions, which are indeed the EM-fields, can be represented as superpositions of basis functions, in particular as linear combinations of plane waves. This feature is one of the essential characteristics of this equation and will be utilized several times in the thesis.

2.2 Linear Optics

If the electric field strength of an incident EM-wave is small compared to corresponding intra-atomic forces, the interaction of light and matter can be described by a linear relationship between the macroscopic polarization and the electric field. This domain, which is called the regime of linear optics, always plays an important role in every day life, spectroscopy and low-intensity experiments.

2.2.1 Refractive Index and Indicatrix

The refractive index n is the key parameter characterizing the optical properties of a dielectric medium, since it determines the phase velocity of light inside a material. Within this section, the basic macroscopic properties of this optical constant are derived precisely.

An EM-wave, which propagates through a dielectric medium, induces a microscopic polarization into this material, since the bound electrons, which interact with the electric field, are deflected from their equilibrium state. The average of the microscopic dipole moment per unit volume is the macroscopic polarization $\mathbf{P}$, which was introduced in Eq. 2.7. Generally, this polarization may be a consequence of induced electronic, ionic and/or permanent dipoles. In the optical regime the polarization is of pure electronic origin, since only electrons are capable of following the fast electric field oscillations ($\nu \approx 200\,\mathrm{THz}$) due to their low intrinsic mass.

In the case of small electric field strengths compared to interatomic ones, the bound electrons follow the external electric field sinusoidally forming harmonic oscillators on atomic scales. Fundamentally, these oscillators consist of movable charge densities (the electrons in the outer shells) and corresponding spatially fixed atomic