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Summary

Frequency conversion in two-dimensional photonic structures

With the advent of nanotechnology it became possible to structure semiconductor materials on a wavelength and subwavelength scale. This opened a door to novel materials called photonic structures with exciting linear and nonlinear optical properties. Semiconductor photonic crystals and nanowires are examples of these photonic structures. Photonic crystals are periodic dielectric structures with features on a wavelength and subwavelength scale with the periodicity in one, two, or all three spatial directions. Nanowires are essentially one-dimensional structures with diameters ranging from a few to several hundred nanometers and typical lengths of several micrometers. They can be grown in a random as well as in a periodic fashion on a suitable substrate.

This thesis presents an experimental study of second harmonic generation in two-dimensional aluminum gallium arsenide photonic crystal slabs (Chapters 3–6) and second harmonic generation in ensembles of aligned gallium phosphide nanowires (Chapter 2). The motivation behind using these III-V semiconductor photonic structures for frequency conversion is their relatively large nonlinearity. The extra dispersion due to the special arrangement of dielectric material in these novel structures may be used to compensate a phase mismatch in the nonlinear process.

Gallium phosphide nanowires are randomly grown on a gallium phosphide substrate by epitaxy at elevated temperatures using $\sim$ 20 nm gold droplets as a catalyst. As-grown nanowires have a diameter determined by the size of gold droplets and a length determined by the growth time. A typical length of these wires is several micrometers. The high length-to-width aspect ratios of the nanowires combined with the high refractive index of gallium phosphide can lead to strong birefringence. In Chapter 2 we investigate if this birefringence can be used to phase-match second harmonic generation. The enhancement of the second harmonic signal is only effective for nanowires that are much longer than the coherence length. However, long nanowires ($> 10\ \mu$m) have a significantly reduced birefringence due to the bending of the wires. We also discuss a number of experiments that aim at separating the second harmonic generated in the substrate from the second harmonic generated in nanowires that are shorter than the coherence length. Unfortunately, we were not able to separate these two contributions.

Chapter 3 describes the fabrication of freestanding, two-dimensional photonic crystal slabs with a square lattice of holes perforated in a ~ 150 nm thick $Al_{0.35}Ga_{0.65}As$ slab. Samples that are investigated in this chapter have a hole radius of $r \sim 150$ nm, a lattice constant $a = 890$ nm, and a surface area of $\sim 300 \times 300\ \mu m^2$. The freestanding slabs are supported by a GaAs substrate. The linear optical properties of these structures are investigated by measuring the reflection spectra as a function of angle of incidence. These spectra show dispersive, asymmetric lineshapes superimposed on top of a slowly oscillating background. The lineshapes can be understood in terms of the Fano model. Within this model, the interference between a direct (non-resonant) and an indirect (resonant) contribution gives rise to the asymmetric lineshape in the spectra. The direct contribution is due to the Fresnel reflection of the incident light from the slab, and the resonant contribution is due to the coupling of the incident light to a leaky waveguide mode via diffraction from the lattice. By using the Fano model it is possible to obtain the dispersion relation and the quality factor of the resonance from the experimental reflection spectra. The nonlinear optical properties of the photonic crystal slabs are investigated by measuring the nonlinear reflection spectra as a function of angle of incidence. We show that the resonant coupling of a pulsed laser at a wavelength of 1.535 μm can significantly enhance the second harmonic signal. A second harmonic enhancement of more than 4500 × compared to the non-resonant contribution is measured when the pulsed laser beam is tuned into resonance with one of the leaky modes of the structure.

Freestanding photonic crystal slabs are not flat, but they buckle because of a small lattice mismatch (0.05%) between the $Al_{0.35}Ga_{0.65}As$ material of the slab and the GaAs substrate. In Chapter 4, we present a novel technique to transfer a freestanding photonic crystal slab to a transparent gel layer. In contrast to the freestanding structures, the transferred structures are almost perfectly flat, and they allow for transmission measurements. Most importantly, the resonant features in the measured reflection spectra of a structure on a gel are much more visible than those of a freestanding structure. Therefore, the transferred structures are more attractive for investigating the resonant coupling of light to photonic crystal slabs. In addition, we study in more detail an avoided crossing between two modes in experimental transmission spectra. At the avoided crossing one of the modes becomes long-lived (subradiant) and the other mode becomes short-lived (superradiant). For the subradiant mode we measure quality factors as high as $Q = 300$. This value is limited by the finite size of the structure, which suggests an excellent optical quality of the photonic crystal slabs transferred to the gel substrate.

The linear reflection and transmission spectra of a two-dimensional photonic crystal slab consist of asymmetric Fano lineshapes due to the resonant coupling of light to leaky modes of the photonic crystal slab via diffraction from the photonic lattice. The coupling of a continuum of modes to a single resonant channel leads to interference between the direct (non-resonant) and the indirect (resonant) channel. This is described by the Fano model. The generally accepted picture is that for a lossless system the sign of a real-valued parameter q of the Fano model determines the asymmetry of the lineshape. For a lossless and symmetric air-slab-air structure, the sign of q changes if the amplitude reflection coefficient of the direct contribution goes through zero. As a consequence, the asymmetry of the Fano lineshape in the reflection spectra for p-polarized light can be reversed by tuning the angle of incidence through Brewster's angle for the symmetric system. In Chapter 5 we show that for a lossless and asymmetric air-slab-gel structure it is also possible to change the asymmetry of the Fano lineshape by angle tuning without reaching the condition of zero amplitude. However, this requires a complex-valued parameter q and shows that the picture of a real-valued parameter q that controls the asymmetry is not complete. More importantly, this demonstrates that a Fano type resonance, characterized by a complex-valued parameter q is not necessarily a sign of microscopic processes of decoherence and/or dephasing.

Chapter 6 studies in more detail second harmonic generation from photonic crystal slabs transferred on a gel substrate. In contrast to Chapter 3, we use a collimated laser beam, and we measure the second harmonic signal in transmission instead in reflection as a function of angle of incidence. Compared to Chapter 3 we go a step further in understanding the importance of leaky modes at both the fundamental and second harmonic frequency for the nonlinear process of second harmonic generation. The main features of the experimental second harmonic signal as a function of angle of incidence are explained in terms of a relatively simple coupled mode theory. Unlike full numerical calculations, our model offers direct insight into the underlying physical mechanism. Furthermore, the model does not assume parameters of an ideal two-dimensional photonic crystal slab. Instead, it uses measured dispersion relations and quality factors of relevant modes as well as the measured non-resonant second harmonic signal. This makes the model applicable to less-than-perfect structures with a finite size and a number of fabrication imperfections. At normal incidence, both the fundamental and the second harmonic field resonantly couple to leaky modes of the structure, and we measure second harmonic enhancements as large as 10000 × compared to the non-resonant contribution. In addition, the measurements convincingly show the effect of the resonant coupling of the second harmonic wave to a leaky mode of the structure. The angular width of the measured second harmonic signal is a factor of 1.6 smaller than the width deduced from the linear optical properties of the leaky mode at the fundamental frequency. More obviously, two additional satellite peaks appear at angles of incidence of ± 9.1°. These satellites can be explained by considering the resonant coupling of both the fundamental beam and the second harmonic beam to leaky modes of the photonic crystal slab. This accounts for the reduced width of the measured second harmonic signal at normal incidence and for the two satellite peaks, and shows the importance of a double resonant condition for efficient second harmonic generation from a photonic crystal.