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Fabrication and Characterization of Quasi-Phasematched NLO-Waveguides using Liquid Crystalline Polymers

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Side chain polymers were investigated for the realisation of nonlinear film and rib waveguides. Cyanobiphenyl and azo-chromophores have been studied as nonlinear optical side groups by measuring the nonlinear optical coefficients, the refractive indices and the transparency range. Periodically poled waveguides using electrode and Corona poling in combination with corrugated surfaces have been fabricated. First experience and results for the realisation of quasi-phase-matched NLO-waveguides for second-harmonic generation using these polymers are presented.

Keywords: Second harmonic generation; optical frequency conversion; NLO; waveguides; quasi-phasematched; liquid crystalline polymers

Second harmonic generation with guided waves has been demonstrated using side-chain polymers with NLO-Chromophores e. g. like DANS¹ or DR1². Large first order nonlinearities and small refractive indices at the second harmonic wavelength have been observed, compared with inorganic and semiconductor single layer devices^{3,4}. Optical nonlinearities in such polymeric waveguides have found a successful application in fast electro-optical modulation using the Pockels effect. On the other hand, the realization of

efficient optical frequency conversion like SHG into the green and blue spectral range is still an outstanding problem, which has not been solved. In fact, huge absorption of the nonlinear azo-chromophores below 500 nm limits the effective interaction length of these materials for such applications.

In the present paper, results obtained with new NLO-sidegroups which exhibit an improved transparency range are discussed. The opportunities for the fabrication of nonlinear waveguide devices comprising quasi-phase matching (QPM) have been proved with these new materials.

Figure 1 shows the investigated acrylate copolymer with cyanobiphenyl (CN) as optically nonlinear side group. For simplicity, some of the following results have also been obtained with similar guest-host systems, using the same chromophores in a polymethylmethacrylat (PMMA) host. These polymers have an absorption maximum at 300 nm.

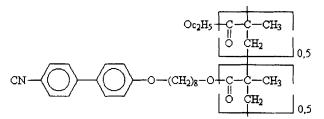


Figure 1: Copolymer with an acrylate backbone and cyanobiphenyl (CN) as nonlinear optical side group.

The extinction coefficients for green (e.g. 532 nm) and blue (e.g. 405 nm) light are about 200 times smaller compared to the values obtained with the above mentioned azo-chromophores⁵. Thin film layers (= 2 μ m) were spincoated on ITO-coated glass substrates and poled at δ 300 V/ μ m for 30 min near the glass transition temperature at 110°C using a *Corona*⁶ setup. The nonlinear effective coefficients d_{eff} have been determined with an Q - switched Nd:YAG laser (λ = 1064 nm, pulse duration 10 ns, repetition rate 1 - 10 Hz) using the Maker fringe method and comparing with a quartz reference (d₁₁= 0.4 pm/V)⁷. Figure 2 shows the dependence of the nonlinear coefficient

on the loading concentration of the SH active side group. Nonlinear coefficients up to $d_{33} \approx 1.1$ pm/V have been obtained with the copolymer with 50% load of NLO-chromophores. From these results coefficients up to $d_{33} \approx 2.25$ pm/V can be expected for the cyanobiphenyl homopolymer, which is presently developed.

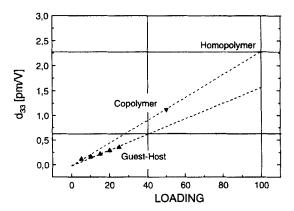


Figure 2: Nonlinear coefficient d₃₃ versus loading concentration of the NLO side group.

Although the nonlinear coefficients of the above mentioned azo-polymers are about 20 times larger than the coefficient of the CN polymer, the huge absorption at 500 nm limits the effective interaction length for SHG generation of green or blue light in these materials. With regard to the improved transparency window the nonlinear coefficient of CN is of the same magnitude as well established inorganic crystals like KTP, which is still quite good.

For the realisation of NLO waveguides a low index buffer and cladding material is required Furthermore, the buffer material should be processable by standard lithographic technology. For this purpose perfluorcyclobutane (PFCB) has been chosen as a buffer and cladding material. The refractive indices of all polymers were determined with m-line spectroscopy (Fig. 3).

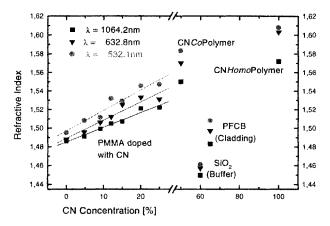


Figure 3: Refractive indices of the nonlinear polymers (CN), the cladding polymer (PFCB) and silicondioxide

Waveguide devices were fabricated by multilayer spin coating onto silicon substrates with a SiO₂ buffer layer. PFCB was coated as a low refractive index buffer polymer. An approximately rectangular groove was etched via reactive ion etching with a mixture of SF₆ and O₂ into this layer. The grooves were then filled with the waveguiding NLO-polymer. The upper cladding was placed above the waveguide to separate the guided mode fields from the absorbing poling electrodes. Titanium or Aluminium electrodes were deposited (LPCVD) and wet-etched. In the last step the wafer was diced with a diamond blade saw into pieces for endfire coupling.

The channels have been designed as so called rib-waveguides shown in Figure 4. For single mode operation, the rib-waveguide dimensions {w,h,t} have to satisfy the relations⁸:

$$h < 2t$$
; $\frac{w}{h} \le 0.3 + \sqrt{\frac{t/h}{1-(t/h)^2}}$

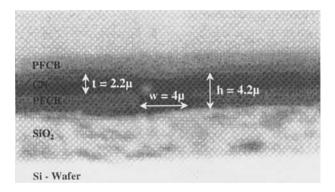


Figure 4: Rem picture of a rib waveguide prepared on a Si-substrate with a SiO2 buffer layer and a upper polymer cladding layer (PFCB).

Damping of the waveguides has been determined by cut-off experiments. The waveguide losses are typically in the range of 3...5 dB/cm and the coupling losses 3 dB (input plus output facet).

In NLO-waveguides with normal dispersion the fundamental and second harmonic wave propagate in the same direction. In general, the two waves walk out of phase after propagation over a distance l_c and the generated secondharmonic is backconverted into the fundamental. As a result the SHG power oscillates like $sin^2(\Delta\beta z/2)$ along the propagation direction z without any net accumulation, with $\Delta \beta = \pi/I_c$ is the phase mismatch. Since conventional techniques well-known from birefringent bulk NLO-materials can not be applied with waveguides to compensate the phase mismatch, another approach has to be used. Presently so-called quasi-phase matching (QPM) is one of the most promising techniques in that context. Usually, QPM is achieved by inverting the sign of the optical nonlinearity in a periodical manner along the wave propagation direction with a period $\Lambda = 2l_c = \mathcal{N}2(N^{2\omega} - N^{\omega})$, where N^{ω} and $N^{2\omega}$ are the effective refractive indices of the fundamental and the second harmonic wave.

In order to apply the correct grating period Λ the phase mismatch has been determined experimentally with the above discussed waveguides. For this purpose, 40 channels of different length have been prepared by cutting the exit edge of the substrate including a small angle with respect to the input edge. Recording SHG for the different channels yields Maker-fringes from which the coherence length l_c has been determined (Fig. 5).

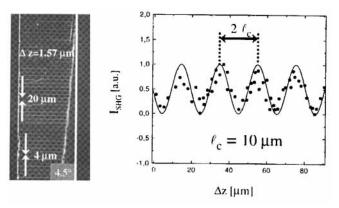


Figure 5: Corona poled rib waveguides of different length (left), Maker fringes and determination of the coherence length l_c in rib waveguides (right).

For the realization of quasi-phasematched rib waveguides we are following two approaches.

First, rib waveguides with metallic (Ti or Al) electrodes are prepared (Fig. 6). Each sample contains several groups of waveguides with the waveguide width w varied from $1.5 \, \mu m$ to $6 \, \mu m$ in steps of $0.5 \, \mu m$. The samples were fabricated with interdigital finger electrodes of different grating periods ranging from

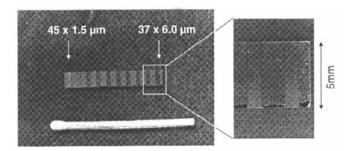


Figure 6: Microscope picture of a rib wave-guide. The channel width w varies from left to right from 1.5 μm to 6.0 μm

14 μm to 28 μm. Using planar electrodes in combination with high voltage, the achievable electric poling field is limited. If the applied voltage exceeds a threshold value of approx. 65 V/μm, the sample is damaged by short circuit currents originating from defects inside the layers or at the contactinterfaces. Comparing the SHG-signal obtained in samples poled as described above with that of identical but Corona poled samples, the electric field in the sample produced by the Corona discharge can be estimated up to 300V/μm.

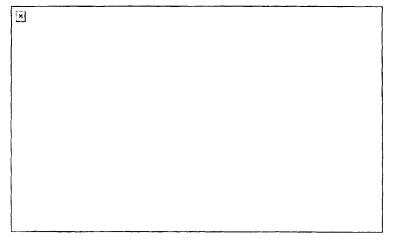


Figure 7: Preparation of periodically poled waveguides using Corona poling in combination with corrugated surfaces

Figure 7 shows the second approach for the preparation of periodically poled rib-waveguides using Corona poling in combination with corrugated surfaces. In that case the upper cladding is covered with a photoresist layer, which is then wet-etched to realize corrugation grating periods ranging from 15 μ m to 30 μ m. Due to the corrugated polymer surface, the Corona discharge leads to a periodic poling of the NLO-chromophores. Scanning a $\Lambda=30~\mu$ m sample with a focused (3 μ m) Nd:YAG laser beam along the rib waveguide shows the corresponding modulated SHG-Signal (Fig. 8), which clearly exhibits the periodic poling.

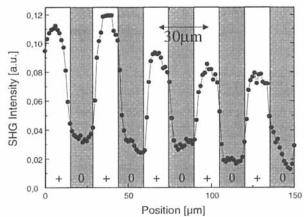


Figure 8: SHG Intensity in a Periodically Structured Sample using Corona Poling (+ areas poled, 0 areas not poled)

In summary, cyanobiphenyl co- and homopolymers as well as guest-host systems using PMMA as host have been realised and characterised as interesting NLO materials for SHG into the green and blue spectral range. The refractive indices of the polymers were determined with m-line spectroscopy. Nonlinear coefficients of $d_{33} \approx 1.1 \text{ pm/V}$ have been obtained with a 50% coploymer using a Nd:YAG fundamental wave, accompanied by a clearly improved transparency window down to 400 nm. The QPM-period has been

experimentally determined to $\Lambda = 20 \, \mu m$ for a 4 μm wide rib-waveguide by Marker fringes. Cut-off experiments yield waveguide losses of 3...5 db/cm and coupling losses of 3 db. Based on these results poled rib-waveguides comprising quasi-phase matching have been fabricated with $\Lambda = 15...30 \, \mu m$, which are presently under investigation.

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