1.8 Nonlinear Optical and Electro-Optic Investigations of Organic Crystals

(F. Pan, Ch. Bosshard, S. Follonier, G. Knöpfle, R. Spreiter and U. Meier)

This year most of our nonlinear optical work on organic crystals concentrated on the salt DAST and the hydrazone derivative DANPH.

Due to the growth of large size DAST crystals up to 20×20×5 mm^3, a-, b- and c-plates could be cut and polished for linear, nonlinear optical and electro-optical investigations. The dispersion of the principal refractive index n_1 was re-measured and extended to wavelengths down to 700 nm and up to 1535 nm. In the same spectral range we performed electro-optical experiments. Our interferometric electro-optic measurements were in good agreement with experiments by field-induced birefringence with crossed polarizers (±45°). As expected from the molecular arrangement, the electro-optical coefficient r_{11} is quite large with values of 47±8 pm/V at λ = 1535 nm and 77±8 pm/V at λ = 800 nm. We found that our experimental results deviate from the expected theoretical expectation both in magnitude and in the wavelength dispersion. For a better understanding of this results, further measurements are in progress.

For the first time to our knowledge phase-matched SHG was observed in single crystals of DAST. In this experiments we used a (100) plate that was illuminated with light at λ =1542 nm. For an s-polarized fundamental wave a type-I phase-matched p-polarized second-harmonic wave was generated with an effective nonlinear optical coefficient of d_{eff}= 8±2pm/V. This d_{eff} is based on the contributions for the small second order NLO coefficients, d_{23}, d_{13} and d_{15}. More interesting phase-matching possibilities based on the NLO coefficient d_{26} (=130 pm/V at 1542 nm) exist. From the dispersion of the refractive indices of DAST, the phase-matching conditions were calculated, leading to effective nonlinear optical susceptibilities up to 56 pm/V at λ=1.542 μm for type II phase-matching. New crystal cuts optimized for further phase-matching measurements are under preparation.

Combining the nonlinear optical susceptibilities and the dispersion of the refractive indices of DANPH, phase-matching configurations for optic parametric oscillation, sum frequency mixing as well as sum frequency generation at λ=1.542 μm could be obtained with effective nonlinear optical susceptibilities up to 170 pm/V. When pumping in the near infrared (using e.g. a Ti-Sapphire laser), a broad tuning range from 800 nm up to 3 μm can be reached by changing the external angle of incidence of the crystal by less than 12 degrees. Crystals of larger size are still expected to allow a direct measurement of the electro-optic and further nonlinear optical coefficients of this material.