Abstract

The present PhD thesis studies the potential of organic PR polymer composites as dynamic recording medium for holographic optical coherence imaging (HOCI) of biological tissue samples. Photorefractive (PR) media undergo a refractive index modulation via space-charge-field formation and electro-optic nonlinearity when exposed to nonuniform illumination, setup by two coherently interfering laser beams. The resulting index modulation creates a phase grating or hologram that is able to store phase and amplitude of optical light waves. The hologram recording process in PR media is dynamic, that is it can be erased by uniform illumination and rewritten again. This makes these materials suitable for real-time imaging applications such as HOCI. The advantage of holographic recording compared to other state-of-the-art imaging techniques including confocal microscopy or optical coherence tomography (OCT) is its whole-field image formation mechanism, which enables high data parallelism, and consequently image acquisition with high frame rates and without the need for point-like scanning.

In practice, to compete with other techniques high recording speed and detection sensitivity in the near infrared (NIR) region of the spectrum is crucial. In this spectral region, biological tissue features low absorption, meaning high penetration depth. With respect to the PR recording medium these requirements imply a fast grating buildup and high diffraction efficiency. In the past, it has been possible to dramatically increase the sensitivity of PR polymer composites based on two conceptional different strategies that have been pursued. The chemical strategy is to understand and adapt the structure/property relationship of the different functionalities comprising the PR composite to the experimental needs. This led to the development within our group of the currently most NIR sensitive PR polymer composite reported in the literature, which is based on the hole conducting polymer host PF6-TPD, sensitized by the fullerene derivative PCBM. Alternatively, the PR performance can be tuned by physical means. In this particular case pre-

illumination (PI) has been shown to be an efficient tool to improve the response time owing to the pre-generation of additional charge carriers. However, also negative effects such as an increase of the grating build time have been reported when applying PI.

In the present thesis, the PI effect is systematically studied in a poly(N-vinylcarbazole)-based photorefractive composite by illuminating it within the absorption band of the electro-optical (EO) chromophore. Under these conditions both, mobile holes and electrons are generated. Additionally, PI outside the absorption range of the chromophore, in the red spectral region, was carried out. The holographic performance was investigated at 830 nm by two-beam coupling and degenerate four-wave mixing.

When illuminating the material at a wavelength of very strong absorption a thin charge carrier layer is formed and the material is flooded with carriers from the side of the device after turning on the electric field. This should alter the recording dynamics by changing the trapping landscape at the recording wavelength and by providing additional mobile carriers. Because the resulting charge carrier layer ($\approx 1\ \mu$m) is thin compared to the total thickness of the device (105 μm) grating recording in the presence of the PI wavelength, even at high intensities, is feasible. PI with 405 nm and 532 nm improved the recording speed by a factor of $\sim$2 and $\sim$2.5, respectively. Additionally, under PI conditions competition between positive and negative charges leads to sign inversion of the two-beam coupling gain coefficient during recording. The chromophore radical ions DMNPAA$^{\cdot -}$ and MNPAA$^{\cdot -}$ are identified as the photogenerated active species. Their life time is determined by introducing variable temporal delays between PI and hologram recording. The magnitude of the PI effect was observed to depend on the PI wavelength, PI intensity and the external electric field.

PI at 633 nm and 830 nm improves the diffraction efficiency but leads to a reduction of the recording speed. This can be explained by the concept of *optical trap activation* (OTA). Only at high external fields the effect of OTA is overcome because of the strongly field-dependent charge-carrier-photogeneration efficiency.

Despite a slight improvement of the recording speed the PVK-based material features insufficient sensitivity to be employed as recording medium for holographic imaging. Contrary to the PVK medium, the outstanding sensitivity and device life time of the PF6-TPD-based composite permitted to integrate the material into an imaging system based on coherence-gated holography or HOCI. Comprehensive experiments with different short-coherence light sources allowed to identify the image domain, instead of the also widely used Fourier domain, as the recording geometry guaranteeing the best image resolution. The reason for this choice is the problem of *beam walk-off* that occurs in off-axis geometries and leads to a significant reduction of the interaction region in the intersection area of the write beams.

Different two-dimensional (2D) and three-dimensional (3D) test targets and tissue models were investigated including resolution targets, a polymer foam and spherical rat tumors. The samples to be analyzed were first imaged in a transillumination geometry. This method, however, prevents the possibility for depth-resolved imaging. Thus HOCI was employed to permit the acquisition of in-depth images. In this case light backscattered from the sample is coherently detected within the PR polymer. At a total write beam intensity of $\sim$10 mW/cm^2 and a fringe spacing of 4 μm holographic recording in the polymer achieves a frame exposure time of $\sim$1 s corresponding to a sensitivity of $\sim$10 cm^2/J. An axial resolution of 18 μm and a transverse resolution of 30 μm, up to a depth of 600 μm (1.2 mm of total path) was obtained when imaging the polymer foam. The HOCI data set of the rat tumor demonstrate a penetration depth of $\sim$800 μm with a dynamic range of $\sim$20 dB. The field of view of the hologram reconstructions was $\sim$1 mm.