FS-TIME-RESOLVED SPECTROSCOPIC ELLIPSOMETRY

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Implementing time-resolved ellipsometry to study effects of free charge carriers induced by pulsed laser excitation has been tried first in the 1970's [1]. However, for a long time, experimental limitations and single wavelength probes limited the ellipsometric approaches to indirect investigations of pump-induced charge carriers in the near-infrared. Today's possibilities to generate continuum white light pulses allow even spectroscopic ellipsometry with time resolution in the fs-range. Still, a number of experimental challenges as spectral fluctuations and group velocity delay remain. We demonstrate a new setup based on an amplified Ti:sapphire laser which is used to generate continuum white light in a CaF_2 crystal as probe while its fundamental, doubled or tripled frequency are applied as pump. Time-resolved spectroscopic ellipsometry measurements have been carried out in *polarizer-sample-compensator-analyzer* configuration by obtaining transient reflectance-difference spectra at different compensator azimuth angles.

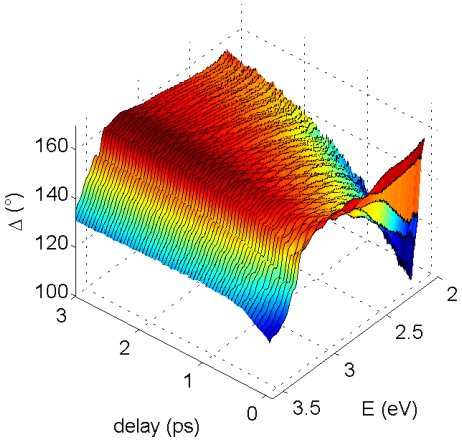

Fig. 1. Transient ellipsometric angle Δ for a *c*-plane oriented ZnO single crystal showing the response of plasma absorption as well the damping of the excitonic and higher transitions.

We demonstrate first measurements of ZnO single crystals, films and resonators as well as Ge single crystals. In Ge, depending on the pump wavelength, band filling effects or renormalization are observed at the E1 and E1+Δ1 transitions which take place at the L point in the momentum space. In ZnO, the dynamics of UV-excited hot charge carriers can be monitored by their induced plasma absorption as well as damping of optical transitions of excitons and above the band gap. Carrier thermalization, recombination and lattice heating can be distinguished as processes on different spectral ranges and time scales ranging from fs to ns. A particular challenge to model transient ellipsometry data arises from the depth gradient of the density of excited charge carriers due to the pump laser absorption.

Keywords: dynamics; pump-probe; hot charge carriers

References

[1] D. H. Auston, C. V. Shank, Phys. Rev. Lett. **32** (1974) 1120.