PITFALLS OF TIME-RESOLVED SPECTROSCOPIC ELLIPSOMETRY

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Time-resolved spectroscopic ellipsometry measurements have been carried out at ZnO single crystals and a film using a pump-probe setup employing UV pump and white light probe pulses. Here, we will discuss experimental details and strategies such as the choice of polarization optics in order to minimize the chirp induced by the group velocity delay, and limiting factors for the time resolution. Fluctuations of the white light spectrum are compensated by the use of two chopper plates. Further difficulties result from non-ideal optical components. Different calibration strategies have been tried when obtaining ellipsometric data from the measured intensity spectra.

The ellipsometric approach is superior to transient reflectance or transmittance experiments because it allows unambiguous distinction of effects on the real and imaginary part of the dielectric function. Furthermore, measuring transient depolarization can serve as monitor for unresolved processes faster than the time resolution of the instrument. In principal, time-resolved ellipsometry does even enable studying the dynamics of anisotropies. However, a crucial point in obtaining true dielectric function data of a pumped sample is application of an appropriate model. In particular, artificial observation of gain, indicated by a negative ε_2, is possible when neglecting that the density of excited charge carriers is highest close to the surface and decreases towards the bulk (Fig. 1). Such stimulated emission cannot be observed in reflection experiments [1], because the detected light has an opposite wave-vector component normal to the interface. Though, this aspect is hard to identify when modeling bare reflectance measurements [2].

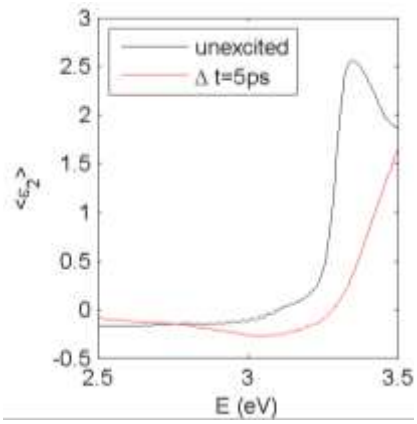

Fig. 1. As measured pseudo-dielectric function of a *c*-plane oriented ZnO single crystal before and 5ps after arrival of the UV pump pulse. Seemingly, gain is observed. (Small offset due to surface roughness)

Keywords: pump-probe; instrumentation; dynamics; modeling

References

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