Ultrafast Spectroscopy, Microscopy, and Nanoscopy in Optoelectronic Materials

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Optoelectronic materials operate at the intersection of photonics and electronics. Their function relies on a complex interplay of many dynamic processes that are difficult to disentangle with conventional methods as various spectroscopic signatures often overlap. Nanostructured samples pose additional challenges for time-resolved spectroscopy due to their spatial heterogeneity. In this talk, several new approaches will be discussed that provide information on dynamic processes with both spectral-temporal as well as spatial resolution. Exemplary applications will focus on materials of interest in optoelectronics.

The first technique, coherent two-dimensional (2D) spectroscopy, is sensitive to quantum-mechanical coherences and populations and provides detailed information on the couplings and dynamics of complex systems. It is particularly well suited to analyze energy transfer processes that are relevant both in natural photosynthesis as well as in artificial light harvesting of organic optoelectronic materials. Recent work includes the study of electronic couplings, the character of delocalized excitonic states, and exciton transfer in a porphyrin dimer, perylene-based dimers, and carbazole dendrimers.

The second technique provides direct spatial-temporal information for analyzing nanostructured materials such as plasmonic waveguides. The development of “spectral-interference microscopy” allows us, for example, to measure group velocities and dispersion properties of ultrashort plasmon pulses propagating through plasmonic “nanocircuits”. With such precise knowledge of multiple interfering modes we can control the routing of these pulses into different output ports.

In the third technique, 2D spectroscopy is generalized to provide additional subdiffraction spatial resolution. This “2D nanoscopy” was motivated by the desire to measure coherences and transport phenomena directly in space and time. We use a four-pulse excitation sequence generated by a pulse shaper, and detect photoelectrons using photoemission electron microscopy (PEEM) with ≈50 nm spatial resolution. Thus the optical diffraction limit can be circumvented for the detection. 2D Fourier transformation of the photoelectron yield provides 2D spectra for each spatial position. We applied this technique recently to investigate thin-film solar cells. Nanostructured interfaces lead to localized photonic modes and thus to enhanced efficiencies.

X-ray investigations of HIOS:
state of the art structures, chemical and optical control of growth

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We will summarize recent progress in x-ray investigations of HIOS. Starting with the surface preparation of bare ZnO, we discuss the structure of ultrathin self assembled monolayers and also multilayer films. We show that by extending situ real time x-ray measurements during growth to both specular and diffuse scattering, we can - for the first time in molecular growth - get all the parameters for a theoretical KMC description of temperature, rate and time dependence in thin film formation. Using this real time x-ray technique for understanding growth, we study the influence of a molecular dipole moment on the self-assembly and show that also laser light can be used as control parameter in molecular thin film growth.