Femtosecond spectroscopy of photo-induced processes

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Ultrafast charge carrier interactions at semiconductor interfaces are the functional basis for a variety of electronic and optoelectronic devices of technological significance. The development of high-speed electronic circuits for telecommunications [1] and next-generation high efficiency solar cells [2] profits from the fundamental understanding of the charge carrier dynamics at the semiconductor boundaries. The real-time investigation of photo-induced processes in bulk semiconductors and functionalized materials such as thin molecular and nano-structured semiconductor films invokes a variety of experimental techniques to elucidate the specifics of the respective samples.

The dynamics of some essential photo-induced reactions in different semiconductor materials comprising charge transfer, carrier relaxation and carrier transport investigated by femtosecond (fs) pump-probe spectroscopy are reviewed and illustrated with recent results.

Transient absorption using fs broadband white light continuum as a probe was applied to investigate the electron injection dynamics of dye sensitized colloidal and nano-rod TiO_2 and ZnO films [3]. The simultaneous measurement of absorption spectra and time traces enables a detailed temporal study of the energetic states involved in the photo-induced electron transfer and early recombination mechanisms. High time resolution is achieved with the use of specially designed sub-20 fs non-collinearly phase matched optical parametric amplifiers (NOPAs) [4] operating at high repetition rates and ultralow photon flux. An injection time < 200 fs was measured for the dye/ZnO system representing the shortest overall injection time on any dye-sensitized ZnO colloid system currently recorded. This injection time agrees well with theoretical predictions concerning the electron injection of such systems and is much shorter than many previous experimental reports. Terahertz (THz) spectroscopy using fs generation techniques is a very useful tool to study carrier transport phenomena in these hybrid systems. The direct measurement of the sub-ps resolved temporal evolution of the complex dielectric function gives insight into the charge carrier mobility.

Time-resolved two-photon-photoemission (TR-2PPE) was employed to investigate the temporal relaxation of optically excited electrons at the In-rich reconstructed InP(100) surface. High-energy carriers were generated at laser pump energies chosen to populate hot electron bulk states or the well known C2 surface state [5] via near resonant direct optical excitation. The different relaxation pathways arising from the two generation schemes involve a complex interplay between Γ, X and L intervalley scattering and the transient bulk-phonon mediated occupation of an additional surface state at lower energies, C1 [6]. The dynamics of these processes were recorded

with a novel experimental setup using two NOPAs enabling two-colour pump-probe experiments in the linear regime. These experiments provide useful information in understanding the fundamental dynamics of devices on the basis of this semiconductor medium such as solar cells and high-speed switching circuits.

References:

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