Abstract

Ultrafast spatiotemporal control of photocarriers in doped semiconductors

Control of the spatial and temporal dynamics of photoexcited charge carriers at the surfaces and interfaces of semiconducting materials is pertinent to many of modern technologies such as solar cells, photodetectors and other optoelectronic devices. In inhomogeneous materials such as nanostructured materials, spatial variations in carrier dynamics are inherent via the material design and accordingly allow for improvements in device performance e.g. transport of electrons from donor to acceptor regions in photovoltaic devices. On the other hand, one can also create spatial inhomogeneity in the carrier dynamics in homogeneous systems by using a non-uniform photoexcitation profile. This would have the advantage that the control of the carrier dynamics could be more flexible and not tied down to the fixed material design. In principle, photoexcitation even with a simple Gaussian beam could result in nontrivial inhomogeneous carrier dynamics within the full-width-half-maximum (FWHM) of the optical spot. However, in typical ultrafast spectroscopy measurements performed thus far, one averages out the photocarrier response over the excitation spot and spatial variations therein are inaccessible.

In this thesis, we create nontrivial spatiotemporal dynamics of the photoexcited electrons in a homogeneous Zn-doped GaAs using the spatial intensity variation in a simple Gaussian photoexcitation beam. We image these dynamics using time-resolved photoemission electron microscopy (TR-PEEM) - a technique offering both high temporal and

spatial resolutions. In particular, we demonstrate the spatial redistribution of the photoexcited electrons in two different regimes: (I) the early time delays where we control the vertical transport of electrons to the sample surface and (II) at long time delays where we manipulate the lateral distribution of the electrons along the sample surface.

In the first study, we achieve spatial variations in the screening process of the intrinsic surface field, which influences the vertical drift of photoexcited electrons from the bulk to the sample surface. Combined with the occurrence of Auger recombination in regions of higher intensity, we see a depletion in the electron population at the center, with a gain just away from the center. We show control of these processes and how they affect the electron distribution on the sample surface in nontrivial ways.

In the second study, we show that at long time delays the spatially varying screening process leads to the creation of lateral fields along the sample surface. These highly local and spatially varying lateral fields then act upon the photoexcited electrons, eventually pulling them apart into two distinct distributions. Using a simple model that explains the experimental data, we also show the possibility of generating near-arbitrary lateral fields and thus controlling electrons on the sample surface in more general ways.

In conclusion, this thesis demonstrates the capability to create and control nontrivial spatiotemporal dynamics of the photoexcited electrons even in a homogeneous semiconductor by exploiting the intensity variation of an ultrafast light pulse. This capability could lead to a promising new handle for use in high-speed optoelectronic devices.