

**Dr. Daniel Dietze**

*Nonlinear Terahertz Spectroscopy of Semiconductor Heterostructures*

Supervisor: Karl Unterrainer

ABSTRACT

The main focus of this thesis has been the investigation of the interaction of terahertz (THz) radiation with two-dimensional semiconductor heterostructures beyond the regime of linear optics. The THz spectral region constitutes an especially rich area with many prospects in soft and condensed matter physics. Based on its non-ionizing character and its ability to penetrate a variety of non-conducting materials, THz spectroscopy and imaging techniques face a growing number of industrial and scientific applications. An important milestone in bringing the THz range to the customer has been the invention of the THz quantum cascade laser (QCL) in 2002. Quantum cascade lasers have the potential of being used as miniaturized THz sources in a variety of applications including quality control, environmental monitoring and astronomy. However, room temperature operation, a feature that is highly desired to meet industrial requirements, has not been achieved to date. Recently, THz time-domain spectroscopy (TDS) has been demonstrated to be the perfect tool for an in-depth investigation of the dynamics inside a lasing QCL. By scaling the incident THz field amplitudes to several tens of kV/cm, the pulses can be used to actively manipulate and control the intersubband excitations, giving rise to non-equilibrium states of matter. From the dynamics following the ultrafast excitation pulse, considerable insight into the microscopic properties of the sample can be gained. Examples of accessible parameters include the identification of dephasing and energy relaxation channels, as well as clarifying the role of multi-photon and collective excitations. During this work, various methods for the generation of intense single-cycle THz pulses have been investigated. Based on the coherent conversion of amplified near infrared laser pulses with pulse energies up to 4mJ, we have been able to achieve THz peak field strengths up to 40 kV/cm with bandwidths extending over 6THz. In addition, we have developed several schemes for increasing the THz yield from well-known emitter concepts. These include area sectioning, THz waveguides and dynamic phase-matching. As a proof-of-principle experiment, we have investigated non-equilibrium dynamics of intersubband (ISB) excitations subject to intense single-cycle THz pulses. The broadband excitation enabled the simultaneous driving of several adjacent transitions. The main results have been the observation of the undressing of collective excitations, coherent population transfer to higher states and the identification of a nonlinear contribution to the refractive index based on parametric multi-photon processes. An alternative route for probing light-matter interactions beyond the linear regime is provided by cavity quantum electrodynamics. The coupling of ISB transitions to a cavity leads to the formation of so-called ISB cavity polaritons as a new type of elementary excitation. These polaritons are subject to intense research worldwide as they may enable lasing without inversion and the generation of quantum vacuum radiation. To investigate THz ISB polaritons, we have chosen to replace the cavity by a planar THz metamaterial. Thereby, we could prove that the coupling strength between a single parabolic quantum well and the metamaterial is sufficient to open a gap in the polariton dispersion relation, which is a signature of ultrastrong coupling.