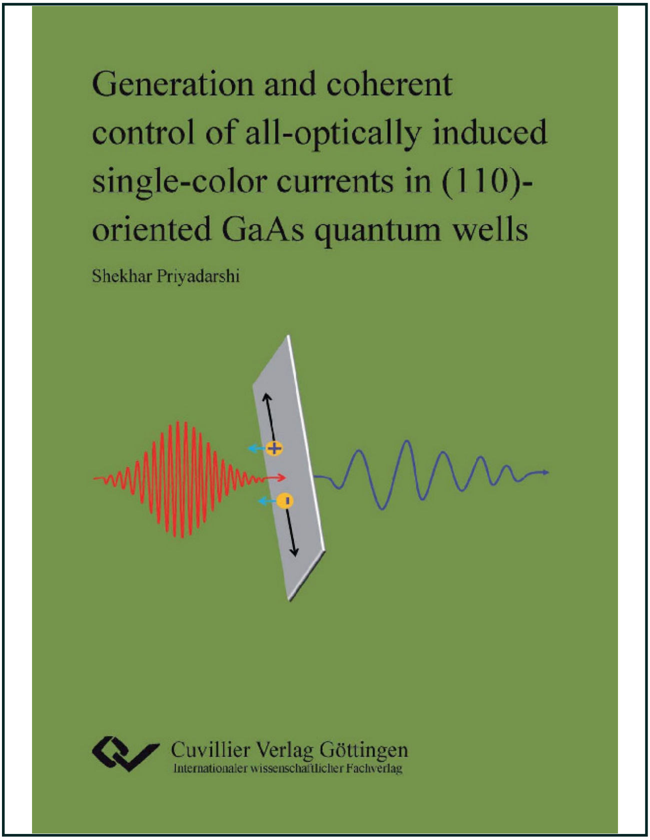
Shekhar Priyadarshi (Autor)

Generation and coherent control of all-optically induced single-color currents in (110)-oriented GaAs quantum wells



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1 Introduction

Optical excitation of semiconductors with a photon energy above the fundamental bandgap leads to the generation of electron and hole pairs (EHPs) [1]. Normally, the excited EHPs are distributed symmetrically in the real and momentum space of the semiconductor. Therefore, such excitation alone doesn't lead to any polarization of the carriers, which, in turn causes a current flow. An antenna structure on the surface of the semiconductors which allows one to apply an external electric field is usually employed to obtain a macroscopic polarization of photo-excited carriers. This device constitutes a so-called photoconductive switch [2]. Such device, when excited with ultrashort optical pulses, emits terahertz (THz) radiation.

However, in certain classes of semiconductor crystals a photo-induced macroscopic polarization can also be obtained without any external electric field. In this case one relies on nonlinear optical effects which lead to an asymmetric carrier distribution in real or momentum space [3, 4]. This asymmetry in real or momentum space results in current transients which are known as all-optically induced currents (AOICs) [5]. The generation of ultrashort AOICs yields broadband THz radiation [5, 6] which can be used for THz spectroscopy [7] and ultrafast wireless communication [8]. At the origin of AOICs and the consecutive THz generation is the lack of inversion symmetry in certain classes of semiconductor crystals. Hence, AOIC depends on the point-group symmetry of the semiconductor crystals and can be used for the investigation of an unknown crystals' point-group symmetry [3, 9-11]. Moreover, certain types of AOIC depend on the spin-selection rules [4, 12] and involve the transport of carriers with certain spin orientation. Such currents can be classified as normal charge currents, spin polarized currents, and pure spin currents. Normal charge currents consist of carriers with equal distribution of spin-up and spin-down orientations, which flow in the same direction. Spin-polarized currents consist of carriers moving in the same direction and having the same spin orientation. Pure spin currents consist of spin-down carriers moving in one direction and the same number of spin-up carriers moving in the opposite direction. The study of such currents is important for the emerging field of spintronics [4, 13-18] and quantum computing [19, 20]. Finally, AOICs flowing in semiconductor crystals can be coupled into coplanar strip-lines, yielding ultrashort voltage pulses. This can be employed for metrological purposes [21-26], since the ultrafast dynamics of AOIC facilitate the shaping of the voltage pulses via optical pulse-shaping [27-32]. This might enable one to pre-compensate the dispersion the voltage pulses experience during propagation over a transmission line.

The AOIC investigated in this thesis can be linked to a second-order difference-frequency mixing process [5, 33, 34]. The difference-frequency mixing of the same optical wavelength, which is called single-color difference-frequency mixing, may induce a constant polarization in the crystal. This process is known as *optical rectification* [35]. The optical rectification process is a dominant process for below bandgap excitation and has been widely studied in several non-centrosymmetric crystal structures [36]. However, for above-bandgap excitation additional types of AOIC exist. They are known as single-color shift and injection currents [5, 37-42]. These currents, which will be called shift and injection currents throughout this thesis, open a new chapter in AOIC research and will be discussed in detail.

The shift current, also known as *linear photogalvanic current* and *above-bandgap rectification current*, arises due to a spatial shift of the photo-induced carriers in real space. It should be noted that the shift current is considerably different than optical rectification as the optical rectification involves virtual carriers while the shift current consists of real carriers. Recently, shift currents gained interest because of their capability to generate ultrabroadband THz radiation [43-45]. The study of shift currents can be a probe to carrier dynamics in the crystal, therefore, its interband and intraband excitations have been studied in several bulk crystals and lower dimension crystals [41, 44, 46-49].

The injection current, also known as *circular photogalvanic current*, occurs in crystals having a spin splitting of the subbands. The spin splitting arises from bulk inversion asymmetry or structure inversion asymmetry and is linear in the momentum [4]. Thus, carriers excited with the same excess energy but different spins have different momenta. This leads to an asymmetric distribution of carriers in momentum space. The asymmetric distribution of the carriers is equivalent to a net current injection in real space. Macroscopically injection currents result from quantum interference of the absorption pathways of two orthogonal polarization components of the optical beam. Such quantum interference process has also been widely studied for third-order nonlinear processes where the crystal is excited with two optical fields: one optical field excites the crystal resonantly while the other optical field excites the crystal via two photon absorption [37-39, 41, 50-57]. Such quantum interference processes will not be investigated in this thesis. The study of injection currents might be used to probe spin-related properties of the bandstructure of semiconductor. Therefore, such currents have been widely studied in several bulk and low dimensional crystals in order to explore their possible applications in spintronics [4, 15, 37-39, 58-60].

The shift and injection currents were first recognized as photogalvanic effects in ferroelectric materials [61, 62] but it was soon recognized that these currents may also occur in non-centrosymmetric crystals without any polar axis [47]. All initial measurements employed a near infrared or visible excitation of crystals, in which an interband excitation of carriers takes place. Recently the development of band gap engineering facilitating the growth of lower dimensional structures like GaAs QWs allows for intersubband excitation of AOICs. These studies were mainly done in the far-infrared and THz spectral regimes [63]. However, interband excitation of AOIC in such lower dimensional structures was also attempted which shows promising potential [39, 41]. The work presented in this thesis is based on such interband excitation of GaAs QWs.

In this thesis, the generation and control of shift and injection currents in (110)-oriented GaAs QWs will be discussed. This particular orientation of the GaAs QWs allows for the generation of shift and injection currents with normal incidence of the optical excitation pulses, hence allows one to use a simple experimental setup for the current generation and detection. The excitation of shift and injection currents with a femtosecond optical pulse results in the emission of THz radiation. The time-resolved detection of the THz radiation using electro-optic sampling is used to characterize these currents.

The thesis has been divided into seven chapters. In chapter 2, a simple microscopic description of shift and injection currents will be given.

In chapter 3, the properties of the GaAs QW samples and the free-space THz setup employed in the experiments will be described.

In chapter 4, a study of the dependence of shift and injection currents on the excitation photon energy will be presented. A shift current component which only exists in the QW region will be investigated in QWs with different well width. *Enhancements of the shift current are observed at photon energies which correspond to light-hole exciton resonance energies in the QWs*. Apart from the current enhancement of the shift current, the experiments show a current inversion between currents excited near the n=1 light-hole exciton (lhX) and near the higher order lhX energies. The phenomena can be explained in terms of the different rates of intersubband scattering of carriers from light-hole (lh) to heavy-hole (hh) states [64]. A similar study of the wavelength dependence of an injection current component in the QWs reveals a tremendous impact of *hh-lh bandmixing* on the injection current. It will be shown that the bandmixing allows for the excitation of initially dark interband transitions which results in an injection current flowing in the opposite direction as compared to the direction of injection current resulting from the allowed interband transitions. This results in an *oscillatory dependence of the injection current on the excitation photon energy* [65].

In chapter 5, it will be shown that shift and injection currents can be employed for symmetry investigations of non-centrosymmetric semiconductors. The investigation of shift current components which are unique for a particular crystal class will be shown to be useful for the specification of the point-group symmetry of the quantum well samples. *It is found that the nominally symmetric quantum wells are asymmetric structures with different left and right interfaces* [66]. Due to this finding it is also possible to investigate an injection current component which doesn't exist in symmetric QWs. The excitation photon energy dependence of this injection current component is found to be different than the dependence of the injection current component studied in chapter 4.

In chapter 6, the generation of coherent alternating photocurrents in the plane of the QW samples will be demonstrated. These alternating photocurrents were triggered during the excitation of shift currents. They result from a simultaneous excitation of heavy- and light-hole excitons with a broadband optical pulse. Such generation of charge oscillations between heavy- and light-hole states has already been demonstrated along the growth direction of GaAs QWs [43, 67-69]. *In this work, however, alternating photocurrents will be shown to exist in the plane of the GaAs QWs as well*. This observation proves the existence of an *in-plane transition dipole moment (TDM) between heavy and light-hole states for in-plane wavevector in (110)-oriented GaAs QWs* [70].

An outlook for future work will be given in chapter 7.