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Coherent femtosecond spectroscopy of exciton-continuum interaction in semiconductors

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Coherent Femtosecond Spectroscopy of Exciton-Continuum Interaction in Semiconductors

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Abstract

This thesis investigates the coherent interaction between exciton and continuum transitions in semiconductors and semiconductor quantum wells with transient four-wave-mixing spectroscopy.

The advent of ultrashort and, consequently, ultrabroadband laser pulses generated with Titanium:Sapphire lasers allows for the study of coherent properties of continuum transitions, exciton transitions and interactions among them in gallium arsenide (GaAs) and aluminum gallium arsenide (Al$_x$Ga$_{1-x}$As) semiconductors.

The linear optical properties of energetically degenerate exciton and continuum transitions in semiconductor quantum wells are investigated by means of linear absorption measurements. It is shown that energetically degenerate exciton and continuum transitions in semiconductor quantum wells form exciton Fano continua. The coupling between the degenerate exciton and continuum transitions is due to the bare Coulomb potential in the linear regime. The linear optical properties of exciton Fano continua can be described by the atomic physics Fano model.

The nonlinear optical properties of energetically degenerate exciton and continuum transitions in semiconductor quantum wells in the coherent regime are investigated by means of four-wave-mixing spectroscopy. It is shown that the nonlinear optical properties of exciton Fano continua strongly disagree with the predictions of the atomic physics Fano model due to strong many-body Coulomb coupling between exciton and continuum transitions in the coherent regime. These discrepancies manifest themselves in a pulse-width limited decay of the temporally integrated four-wave-mixing signal although the four-wave-mixing power spectrum shows narrow lines and the dephasing times are much longer than the pulse width. This many-body Coulomb coupling is also important for the coherent
dynamics of non-degenerate exciton and continuum transitions in semiconductors. To account for this many-body Coulomb coupling, a density dependent dephasing term has to be included into the semiconductor Bloch equations. This mechanism is also known as excitation induced dephasing.

Coherent optical properties of continuum transitions can be transferred to discrete exciton resonances due to the many-body Coulomb coupling by means of specially designed four-wave-mixing experiments. This mechanism can also be used in the reversed direction to transfer coherent properties of discrete excitons to continuum transitions. The transfer process can be understood in a common ground state picture of strongly interacting transitions. The underlying concept is applicable to other systems with a common ground state.

At higher carrier densities the many-body Coulomb coupling between exciton and continuum transitions becomes weaker and normal dephasing behavior dominates. The dephasing can be described accounting for carrier-carrier and carrier-phonon scattering. In this density regime, the nonlinear optical properties of semiconductors can be described by the semiconductor Bloch equations.

This thesis shows that the exciton continuum coupling due to many-body Coulomb interaction strongly affects the nonlinear optical properties of semiconductors in the coherent regime.
Kurzfassung

In der vorliegenden Arbeit wird die kohärente Wechselwirkung zwischen Exziton- und Kontinuumsanregungen in Halbleitern und Halbleiterquantenfilmen mit Hilfe des transienten Vierwellenmischens untersucht.

Ultrakurze und spektral extrem breite Laserpulse, die von einem Titan-Saphir-Laser erzeugt werden, ermöglichen die Untersuchung kohärenter Eigenschaften von Kontinuumsanregungen und Exzitonanregungen und die Untersuchung der kohärenten Wechselwirkung zwischen diesen Anregungen in Galliumarsenid (GaAs) und Aluminium-Galliumarsenid (Al_xGa_{1-x}As) Halbleitern.


Die vorliegende Arbeit zeigt, dass die Vielteilchen-Coulomb-Kopplung zwischen Exziton- und Kontinuumsanregungen die nichtlinearen optischen Eigenschaften von Halbleitern im kohärenten Regime stark beeinflusst.
1. Introduction and Motivation

Time-resolved optical spectroscopy is a powerful tool to investigate electronic and vibrational properties of atoms, molecules, and solids. In semiconductors, optical spectroscopic techniques like reflection, absorption, luminescence, and light-scattering techniques have made valuable contributions to the understanding of the physics of semiconductors [1-3]. Time-resolved optical spectroscopy allows the investigation of non-equilibrium, nonlinear, and transport properties of semiconductors [1-4]. For example, with time-resolved spectroscopy the study of coherent phenomena in semiconductors, scattering among quasiparticles, and the underlying physical interaction is possible. The strong interaction between optically excited quasiparticles in a semiconductor gives rise to very complex dynamics as compared to the dynamics of optically excited states in atoms or molecules. Despite this complexity, the interpretation of nonlinear optical experiments on semiconductors has become feasible with the development of the semiconductor Bloch equations [3, 5]. These equations take into account the Coulomb interaction between optically excited quasiparticles in a semiconductor. Many nonlinear experiments on semiconductors have been interpreted in the framework of the semiconductor Bloch equations [1].

The invention of the modelocked Titanium:Sapphire laser [6] has opened the door to study ultrafast coherent phenomena in gallium arsenide (GaAs). The continuous development of modelocked Titanium:Sapphire lasers has led to shorter and shorter pulse widths. The shortest pulse widths achieved today directly from a Titanium:Sapphire laser are in the range of only 5.5 fs [7-10]. Optical pulses with such a short duration have an ultrabroadband spectrum. Therefore, with these pulses the study of ultrafast continuum interaction in semiconductors is possible. As a rule of
thumb, coherent continuum interactions in a semiconductor are an order of magnitude faster than interactions involving only discrete transitions [11, 12]. Therefore, ultrafast and, consequently, ultrabroadband laser pulses are a necessary prerequisite for the investigation of continuum interactions in semiconductors.

In this work, we will study the coherent semiconductor-light interaction involving discrete exciton transitions, continuum transitions or both, by means of transient four-wave-mixing spectroscopy [1]. We will investigate the interaction between continuum transitions and exciton transitions in semiconductor bulk material and semiconductor quantum wells.

The lowest exciton transitions are separated from the continuum transitions by their binding energy. Therefore, they are discrete exciton transitions. Higher-order excitons in quantum wells, corresponding to higher electron and hole subbands, are embedded in the continuum transitions of lower electron and hole subbands. It has been theoretically shown that higher-order exciton resonances in quantum wells, quantum wires or bulk semiconductors with magnetic field are Fano resonances [13-16]. Fano resonances can be observed if a discrete state and an energetically degenerate continuum are quantum mechanically coupled and both types of states can be optically excited from a common ground state. This type of quantum interference is called Fano interference. A Fano resonance has a particular absorption spectrum, known as Fano profile [17]. The characteristic feature of a Fano resonance is the minimum of the absorption spectrum. This minimum lies well below the absorption of the continuum states. Fano resonances were experimentally observed in quantum wells [18, 19] and in bulk semiconductors under magnetic field [13, 20]. Fano interference in semiconductors has also been investigated in superlattices [21-24], doped semiconductors [25], and for intersubband transitions in doped quantum wells [26, 27].

The first nonlinear optical investigations of Fano resonances were performed in Ref. [28, 29] in bulk semiconductors under magnetic field. It was found that the coherent emission from the exciton Fano continuum is
strongly affected by interference effects between exciton and continuum transitions. These interference effects have their origin in many-body Coulomb interaction between exciton and continuum transitions. These investigations triggered the study of other types of exciton-continuum systems [30-32]. The coherent emission of all these exciton-continuum systems in the time-delay domain could not be described by the semiconductor Bloch equations [33]. Additional density dependent dephasing terms have to be included into the semiconductor Bloch equations to account for the interference effects between exciton and continuum transitions [33].

In this text, we perform the first nonlinear optical investigation of exciton Fano continua in quantum wells in the absence of a magnetic field [19]. We use the four-wave-mixing technique with a time resolution of 100-fs to study the coherent emission from exciton Fano continua in quantum wells. The linear optical properties of different exciton Fano continua are investigated and we demonstrate the similarities and differences between these Fano resonances. We show that the linear and nonlinear optical properties of Fano resonances in quantum wells without magnetic field are similar to the linear and nonlinear properties of Fano magnetoexcitons in bulk semiconductors. With an increased time resolution of 20-fs we show that the interference effect between exciton and continuum transitions gets weaker at higher carrier densities.

With the recently developed partially non-degenerate four-wave-mixing technique [30] we show for the first time that coherent properties of discrete exciton resonances can be transferred to exciton Fano continua and vice versa. This results in the observation of characteristic features of the discrete excitons at exciton Fano continua. On the other hand, characteristic features of the exciton continuum interference can be observed at discrete exciton resonances.

In the last chapter of this thesis we show that, at higher carrier densities, the many-body Coulomb coupling between exciton and continuum transitions gets weaker and normal dephasing determines the coherent
emission from bulk gallium arsenide (GaAs) and bulk aluminum gallium arsenide (Al\textsubscript{x}Ga\textsubscript{1-x}As) semiconductors. The dephasing in this density regime is due to electron-electron and electron-phonon scattering [34] and can be described by a simple relaxation rate approach in a first approximation [34].
2. Theoretical and experimental background

In this chapter the basic concepts used to describe coherent phenomena in semiconductors are reviewed. It begins with an introduction to the optical Bloch equations (OBE), a set of equations used to describe the interaction between an electrical field and an ensemble of independent two-level systems (2-LS) [35]. The ensemble of 2-LS could be homogeneously or inhomogeneously broadened. While many coherent experiments on semiconductors have been described in the framework of the OBE [1], today, coherent phenomena in semiconductors are usually described in terms of the so-called semiconductor Bloch equations (SBE) [3]. The SBE, in contrast to the optical Bloch equations, take into account the interaction among the optically excited quasiparticles in a semiconductor due to Coulomb coupling. The second part of this chapter focuses on this topic of SBE. The last part of this chapter deals with the experimental technique used to investigate the coherent response of a semiconductor immediately following excitation by an ultrashort laser pulse, namely, four-wave-mixing spectroscopy.

2.1 Coherent dynamics of non-interacting two-level systems - Optical Bloch Equations

Coherent phenomena in non-interacting or weakly interacting systems, such as atoms or molecules, are usually analyzed for an ensemble of independent two-level systems [1, 35]. This model assumes that the incident electric field is resonant or nearly resonant with one transition and far off resonance with other transitions that therefore do not contribute to the coherent response of the system. The model further neglects interactions between the two-level systems. The ensemble of non-interacting two-level systems can be homogeneously or inhomogeneously broadened. This simple picture describes the coherent response of a
semiconductor in its simplest approximation. Despite its simplicity this model is often used because discrete excitonic transitions in a high-quality semiconductor sample are mainly homogeneously broadened and can be considered as an homogeneously broadened ensemble of non-interacting two-level systems if Coulomb interaction is neglected. The continuum states of a semiconductor can be considered as inhomogeneously broadened in momentum space if Coulomb interaction is neglected and, therefore, can be described by an inhomogeneously broadened ensemble of non-interacting two-level systems.

Let us assume a homogeneously broadened ensemble of independent two-level systems with a ground state vector \(|a\rangle\) with ground state energy \(E_u\) and excited state vector \(|b\rangle\) with excited state energy \(E_h\). The density matrix formalism is suitable to describe the dynamics of such an ensemble and the density matrix in this case can be written as

\[
\rho = \begin{pmatrix} 
\rho_{bb} & \rho_{bu} \\
\rho_{ub} & \rho_{uu} 
\end{pmatrix},
\]

(2.1)

where \(\rho_{bb}\) is the probability of finding the system in the excited state, i.e. the population in the excited state, and \(\rho_{uu}\) is the probability of finding the system in the ground state, i.e. the population in the ground state. If the two-level system is closed, no population can disappear and the following expression holds:

\[
\rho_{uu} + \rho_{bb} = 1. \tag{2.2}
\]

The off-diagonal elements of the density matrix represent the coherence, i.e., the phase correlation between the excited and ground state or, in other words, the superposition of excited and ground state. It holds that

\[
\rho_{ub} = \rho_{bu}^*. \tag{2.3}
\]
The dynamics of a homogeneously broadened ensemble of two-level systems is described by the Liouville equation

\[
\frac{i\hbar}{\partial t} \rho = [H, \rho],
\]

(2.4)

where the square bracket is the commutator and \( H \) is the total Hamiltonian operator of the system containing the Hamiltonian of the unperturbed two-level system \( H_0 \), the interaction Hamiltonian \( H_{\text{int}} \), and the relaxation Hamiltonian \( H_{\text{rel}} \):

\[
H = H_0 + H_{\text{int}} + H_{\text{rel}}.
\]

(2.5)

The interaction Hamiltonian \( H_{\text{int}} \) describes the interaction between the ensemble of two-level systems and the incident electromagnetic wave and the relaxation Hamiltonian \( H_{\text{rel}} \) describes the processes that return the ensemble back to thermal equilibrium after excitation.

The unperturbed Hamiltonian is given by

\[
H_0 = \begin{pmatrix} E_b & 0 \\ 0 & E_a \end{pmatrix}
\]

(2.6)

and the state vector of the \( j \)-th system for an ensemble of two-level system can be written as the superposition of the two eigenstates \( |a \rangle \) and \( |b \rangle \)

\[
|\Psi_j(t)\rangle = C_{aj}(t)|a\rangle + C_{bj}(t)|b\rangle.
\]

(2.7)

The density matrix has then the following form

\[
\rho = \sum_j p_j \begin{pmatrix}
|C_{aj}|^2 & C_{aj} C_{aj}^* \\
C_{aj} C_{aj}^* & |C_{aj}|^2
\end{pmatrix}.
\]

(2.8)
$P_j$ describes the probability that the system is in the $j$-th state. In this form of the density matrix, the coherence of an ensemble of two-level systems becomes quite clear. If the amplitudes $|C_{aj}|^2$ and $|C_{bj}|^2$ are the same for all $j$'s, but the phases of $C_{aj}$ and $C_{bj}$ are randomly distributed between 0 and $2\pi$, the off-diagonal elements of the density matrix vanish and the ensemble is said to be incoherent. If there is a well-defined phase relationship between the phases for different two-level systems, the ensemble is said to be coherent.

The interaction Hamiltonian describes the interaction between the incident light field and the ensemble of two-level systems. In the dipole approximation it is given by

$$H_{int} = -\mathbf{d} \cdot \mathbf{E}(t),$$

where $\mathbf{d}$ is the dipole moment operator and $\mathbf{E}$ is the electric field of the incident light pulse. The components of the dipole operator are given by

$$\mathbf{d}_{sk} = -\epsilon \int u_n^* \mathbf{r} u_k d^3r,$$

where $u_n$ and $u_k$ are the wave functions in real space and $\mathbf{r}$ is the electron coordinate. The components of the interaction Hamiltonian $H_{int}$ are given by

$$\mu_{sk} = -\mathbf{d}_{sk} \cdot \mathbf{E}(t).$$

The diagonal elements of $\mu_{sk}$ are zero because the dipole operator has odd parity. If the incident electromagnet wave is a monochromatic plane wave with angular frequency $\omega$ and linear polarization in the direction $\varepsilon$, it can be described by the sum of two electromagnetic fields

with

\[ E^+(R,t) = \frac{1}{2} \varepsilon E_0 \exp(i[k \cdot R - \omega t]) \]  \hspace{1cm} (2.13)

and

\[ E^-(R,t) = E^+(R,t) . \]  \hspace{1cm} (2.14)

\( E_0 \) denotes the real electric field amplitude. In the rotating-wave approximation the fast oscillating phase term \( \exp(\pm(i\omega + (E_b - E_a) / \hbar)) \) is neglected and only the slowly varying term \( \exp(\pm(i\omega - (E_b - E_a) / \hbar)) \) is kept [1, 35]. In this approximation the components of the interaction Hamiltonian can be written as

\[ \mu_{ba} = e(r_{ba} \cdot \varepsilon) \frac{1}{2} E_0 \exp(i[k \cdot R - \omega t]) = \mu_{cb}^* . \]  \hspace{1cm} (2.15)

The expression \( e(r_{ba} \cdot \varepsilon)E_0 / \hbar \) is called the Rabi frequency and reflects the coupling between the electromagnetic wave and the ensemble of two-level systems.

The final term of the total Hamiltonian is the relaxation Hamiltonian. Relaxation processes bring an optically excited semiconductor back to thermal equilibrium and include recombination, collisions with phonons, impurity scattering, and scattering with other electronic states. There are different ways to describe relaxation processes in semiconductors, like the non-Markovian ansatz, the classical Boltzmann description, or the relaxation-rate approach [1]. We will comment here on the relaxation-rate approach because this one is used in this thesis.

In this approach, the relaxation to thermal equilibrium is described by constant phenomenological relaxation rates. The relaxation Hamiltonian is given by
\[ \left[ H_{\text{rel.}} \right]_{h_b} = -\rho_{b_b} / T_1 \] (2.16)

and

\[ \left[ H_{\text{rel.}} \right]_{b_a} = -\rho_{b_a} / T_2 . \] (2.17)

\( T_1 \) is the lifetime of upper state \( |b\rangle \) and \( 1/T_2 \) the sum of the half recombination rate \( 1/T_1 \) and the pure dephasing rate \( 1/T_2 \). \( 1/T_2 \) is often called the dephasing rate or transverse relaxation rate. The coherent experiments described in this thesis are analyzed within this approach.

The linewidth \( \Gamma_\ell \) of a homogeneously broadened optical transition, which is described by a Lorentz curve in the frequency domain, is related to the dephasing time \( T_2 \) by

\[ \Gamma_\ell = \frac{2\hbar}{T_2} . \] (2.18)

The evaluation of equation (2.4) with equations (2.6, 2.9, and 2.16, 2.17) yields the equation of motion of the density matrix:

\[ \frac{d}{dt}(\rho_{b_h} - \rho_{a_a}) = -\frac{i}{\hbar}(\mu_{ab}\rho_{ba} - \mu_{ba}\rho_{ab}) - \frac{1}{T_1}(\rho_{b_h} - \rho_{a_a}) , \] (2.19)

\[ \frac{d}{dt}(\rho_{b_a}) = -\frac{i}{\hbar}\mu_{ba}(\rho_{ba} - \rho_{ab}) - \frac{i}{\hbar}(E_b - E_a)\rho_{ba} - \frac{1}{T_2}\rho_{b_a} . \] (2.20)

These are the optical Bloch equations. These coupled equations of motion describe the time evolution of population \( \rho_{b_h} - \rho_{a_a} \) and phase correlation \( \rho_{b_a} \) between the excited and ground states of an ensemble of independent two-level systems after excitation with a short laser pulse.

The off-diagonal elements \( \rho_{ba} \) and \( \rho_{ab} \) of the density matrix are related to the expectation value of the dipole operator and give rise to a macroscopic polarization or dipole moment density \( P \) given by
\[ P = N \text{Tr}\{d\rho\} = N(d_{aa}\rho_{ab} + d_{ab}\rho_{ba}). \] (2.21)

\(N\) is the number density of the ensemble of homogeneously broadened two-level systems. Therefore, the off-diagonal elements of the density matrix are often referred to as polarization. With this terminology the OBE can be interpreted as follows. Equation (2.19) describes the population in the ensemble. The population is driven by the polarization and the incident electric field and it relaxes with a time constant \(\tau_1\). Equation (2.20) describes the polarization of the ensemble. It is driven by the population and the incident electric field. The polarization decays with the time constant \(\tau_2\).

The macroscopic polarization \(P\) can be experimentally investigated because it is a source term in Maxwell's equation and gives rise to the radiation of an electric field \(E_r(t)\). Usually, the intensity \(I_r(t) \propto |E_r(t)|^2\) of the electric field is measured. For thin samples, the radiated electric field is proportional to the macroscopic polarization, \(|E_r(t)|^2 \propto |P(t)|^2\) [1]. Therefore, the time evolution of the macroscopic polarization can be determined from the time evolution of the measured intensity of the radiated electric field.

Inhomogeneously broadened two-level systems can also be described by the OBE if the distribution of the resonance frequencies of the different homogeneous systems is known. One has to integrate the results of the homogeneous systems with respect to the inhomogeneous linewidth. The phases of the different homogeneous systems evolve according to the different transition energies. This leads to a decay of the macroscopic polarization within the laser pulse width if the inhomogeneous linewidth is larger than the laser spectral width. The decay time is proportional to the inverse of the inhomogeneous linewidth, otherwise. The decay of the macroscopic polarization is not due to dephasing but due to interference effects. The decay can be reversed by means of the four-wave-mixing technique. This effect is referred to as photon echo [35] and the dephasing rates can be obtained by means of four-wave mixing [1, 36].
Although the OBE do not include any many-body effects, many coherent transient experiments in semiconductors (homogeneously or inhomogeneously broadened) are analyzed in this framework [1]. In order to include many-body effects the semiconductor Bloch equations have been developed. These equations include the Coulomb interaction between states in a semiconductor in the framework of the Hartree-Fock approximation. The next chapter will give a short description of these equations.

2.2 Coherent dynamics in semiconductors - Semiconductor Bloch Equations

The Coulomb attraction between electrons and holes modifies the linear and nonlinear optical properties of a semiconductor. In the linear regime the Coulomb attraction between electrons and holes leads to the formation of a series of bound electron-hole pairs, the well-known exciton [37]. This manifests itself in distinct peaks below the absorption edge in the linear absorption spectrum. The Coulomb interaction also enhances the absorption of continuum states of electron-hole pairs (Sommerfeld enhancement) [37]. The nonlinear optical properties of a semiconductor can be ascribed to band filling due to the Pauli exclusion principle, renormalization of the electron and hole energies, renormalization of the Rabi frequency, and screening of the Coulomb interaction [38].

If the Coulomb interaction is neglected, continuum electron and hole states can be regarded as inhomogeneously broadened in momentum space.

In many optical experiments with semiconductors [38] Coulomb interaction must be included to describe the nonlinear response of a semiconductor correctly. The optical Bloch equations have to be extended to include the Coulomb interaction between different electron and hole states. This has been done in recent years [3], and the resulting equations are called the semiconductor Bloch Equations (SBE):
\[
\frac{\partial P_i(t)}{\partial t} = -\frac{i}{\hbar} \left( e_{c,k} + e_{h,k} \right) P_i - i \left( f_{c,k}(t) + f_{h,k}(t) - 1 \right) \omega_{R,k} + \frac{\partial P_i(t)}{\partial t}_{\text{scatt}}, \tag{2.22}
\]

\[
\frac{\partial f_{c,k}(t)}{\partial t} = -2 \text{Im}(\omega_{R,k} P_i^\ast(t)) + \frac{\partial f_{c,k}(t)}{\partial t}_{\text{scatt}}, \tag{2.23}
\]

\[
\frac{\partial f_{h,k}(t)}{\partial t} = -2 \text{Im}(\omega_{R,k} P_i^\ast(t)) + \frac{\partial f_{h,k}(t)}{\partial t}_{\text{scatt}}, \tag{2.24}
\]

with

\[
\omega_{R,k} = \frac{1}{\hbar} \left[ \mu_{cv} + \sum_{q \neq k} V_{k-q} P_q(t) \right], \text{ renormalized Rabi frequency} \tag{2.25}
\]

and

\[
e_{h,k} = e_{h,k} - \sum_{q \neq k} V_{k-q} f_{h,q}(t), \; i = e, h, \text{ renormalized single particle energies} \tag{2.26}
\]

The single particle energies in the parabolic band structure approximation are given by

\[
e_{c,k} = \frac{E_{\text{gap}}}{2} + \frac{\hbar^2 k^2}{2m_e}, \; \text{electron energy} \tag{2.27}
\]

and

\[
e_{h,k} = -\frac{E_{\text{gap}}}{2} - \frac{\hbar^2 k^2}{2m_h}, \; \text{hole energy} \tag{2.28}
\]

The zero of the energy scale is in the center of the bandgap. \( P_i(t) \) denotes the polarization in state \( k \) and \( f_{c,k}(t), \; f_{h,k}(t) \) are the electron and hole distribution functions, respectively. \( V_q \) is the Fourier transform of the Coulomb potential. The crucial difference between the OBE and the SBE is that in SBE states with different wave vector \( k \) interact via the Coulomb potential. The Coulomb interaction renormalizes the electron and hole
energies (equation 2.26). Moreover, the Coulomb interaction renormalizes the interaction between the electric field and the semiconductor (equation 2.25, renormalized Rabi frequency) and additional driving terms for the population and polarization occur.

The relaxation terms in the SBE are the same as those in the OBE and the mathematical approaches to describe the relaxation terms are the same as those mentioned in Chapter 2.1. The complete set of SBE cannot be solved analytically for the general case. One has to solve them numerically, which is very time consuming even on the fastest computers available today [1].

2.3 Four-wave-mixing spectroscopy

In this chapter the measurement technique, which allows us to measure the dephasing of optically excited electrons and holes in a semiconductor, namely, four-wave-mixing spectroscopy (FWM), is described. The FWM spectroscopy is only sensitive for the coherent regime, i.e., the FWM signal vanishes if the excited electron and hole pairs have totally lost their phase correlation.

2.3.1 Four-wave-mixing spectroscopy – measurement principles

(a) Degenerate four-wave mixing

In Fig. 2.1 a sketch of the simplest form of a degenerate four-wave-mixing (DFWM) experiment in transmission geometry is shown [1, 39]. In a DFWM experiment all laser pulses have the same spectrum. In the two-beam DFWM configuration the pulse with wave vector \( \mathbf{k} \), arrives at a time \( t_i \) at the sample. It produces a macroscopic polarization \( \mathbf{P} \), which is initially in phase with the incident electromagnet wave, i.e. the polarization and the laser field are coherent. Scattering processes among the excited quasiparticles and between the excited quasiparticles and their
environment destroy this coherence. As mentioned before, the characteristic time in which the polarization decreases to \( \gamma \) is called dephasing time \( T_2 \) [40]. The pulse with wave vector \( k_1 \) arrives at the sample at a certain time \( t_2 \) delayed by \( \Delta t = t_2 - t_1 \) with respect to the first pulse. If the time delay \( \Delta t \) is smaller than the dephasing time \( T_2 \) of the polarization, an interference grating is generated. The pulse with wave vector \( k_1 \) can be self-diffracted from this grating along the phase-matched directions \( 2k_1 - k_1 \). The FWM signal in the direction \( 2k_1 - k_1 \) is free of any background light.

A more mathematical way of looking at the generation process of FWM signals is in terms of the nonlinear susceptibility. FWM experiments make use of the third-order susceptibility of the investigated material. The third-order susceptibility mediates the nonlinear interaction of the incident
laser pulses and a third-order polarization \( P^{(3)} \propto \chi^{(3)} E^3 \) is generated. This third-order polarization generates an electromagnetic wave which is emitted in the direction \( 2k_z - k_1 \). This constitutes the FWM signal. It can be calculated by an iterative solution of the OBE (2.19) and (2.20). In a first step the laser pulse with wave vector \( k_1 \) generates a linear first-order polarization. The interference grating generated by the remaining first-order polarization and the second laser pulse \( k_2 \) is in second order in the electric field. Finally, the diffracted FWM signal along \( 2k_z - k_1 \) is due to the third-order polarization.

There are three different techniques to measure the FWM signal: Temporally and spectrally integrated FWM (TI-FWM), spectrally resolved FWM (SR-FWM), and time resolved FWM (TR-FWM) (see Fig. 2.2). If the diffracted signal is temporally and spectrally integrated (TI-FWM), one measures the diffracted energy along \( 2k_z - k_1 \) as a function of the time delay \( \Delta t \) between the laser pulses \( k_1 \) and \( k_2 \) by means of a photo multiplier and lock-in technique. The spectrally resolved FWM signal (SR-FWM), the FWM power spectrum (FWM-PS) \( |P^{(3)}_{\mathrm{FWM}}(\omega, \Delta \nu)|^2 \), along \( 2k_z - k_1 \) is measured at fixed time delays \( \Delta \nu \) by means of a monochromator.
The TI-FWM signal is given by

$$\text{TI-FWM}(\Delta t) \propto \int |P^{(3)}_{\text{FWM}}(t, \Delta t)|^2 \, dt. \quad (2.29)$$

The time integration is performed by a slowly integrating photo detector. If the diffracted signal is temporally resolved (TR-FWM), one measures its temporal evolution, usually with a nonlinear optical cross correlation technique, along the direction $2k_x - k_i$ at a fixed time delay $\Delta t$. The temporally resolved FWM signal is given by

$$\text{TR-FWM}(t) \propto |P^{(3)}_{\text{FWM}}(t, \Delta t)|^2, \quad (2.30)$$

if the delayed gating pulse (see Fig. 2.2) can be approximated by a $\delta$-pulse. There is a certain relationship between the FWM-PS, $|P^{(3)}_{\text{FWM}}(\omega, \Delta t)|^2$, and the TR-FWM signal $|P^{(3)}_{\text{FWM}}(t, \Delta t)|^2$. The FWM-PS is connected to the autocorrelation function of the third-order polarization $P^{(3)}_{\text{FWM}}(t, \Delta t)$ by Fourier transformation. On the other hand, the TR-FWM signal is proportional to the amplitude of the third-order polarization $|P^{(3)}_{\text{FWM}}(t, \Delta t)|^2$. This shows that, for example, a narrow FWM-PS is always connected with a slow decay of the third-order polarization $P^{(3)}_{\text{FWM}}(t, \Delta t)$, i.e., a slow dephasing. The TR-FWM technique will not be discussed any further in this thesis because it was not used in the FWM experiments.

We will now briefly discuss the scattering processes in semiconductors. The following scattering processes destroy the phase of the optically excited electron-hole pairs [40, 41] and lead to a decay of the polarization:

- scattering among electronic excitations, e.g., exciton-exciton scattering, exciton-electron scattering or electron-electron scattering.
• scattering at impurities or other lattice defects, e.g. interface roughness scattering in quantum wells or alloy scattering in mixed crystals.
• scattering with phonons. This includes the scattering with optical phonons via the Fröhlich coupling or scattering with acoustic phonons via the deformation potential or the piezo-coupling [42].
• recombination. If all other phase destroying processes are absent, the phase will be lost in the recombination process. The recombination process is usually described by the lifetime $T_i$.

The FWM technique allows the determination of the time constants of the above listed scattering processes.

(b) Partially non-degenerate four-wave mixing

An extension of the standard fully degenerate FWM scheme is partially non-degenerate FWM (PND-FWM) [30]. In the PND-FWM scheme the beam with wave vector $k_1$ is passed through a spectral amplitude filter. The beam with wave vector $k_2$ is not filtered and remains broadband. The filter’s transmission is nonzero for only a part of the whole laser spectrum. In our set-up the filter consists of a pair of SF10 prisms.

Fig. 2.3 Spectral amplitude filter in double path configuration.
and a variable slit [43, 44]. It is used in a double path configuration. Figure 2.3 shows a schematic sketch of the amplitude filter. In the forward direction the prisms produce a spatial dispersion of the various wavelengths at the place of the slit. The bandwidth and the central wavelength are adjusted by varying the width and the position of the slit. The spectral resolution is enhanced by focusing the incident light by means of a lens. In the backward direction the two prisms reverse the spatial dispersion and the laser beam is again collinear when it leaves the spectral amplitude filter. The full width at half maximum (FWHM) of the filtered pulse can be adjusted from 6 meV to the full width of the unshaped pulse. The center of the filtered pulse spectrum can be varied in the range of the broadband unfiltered pulse.

It is important to note that in PND-FWM experiments only in that part of the spectrum where the filtered and the unfiltered pulse spectrally overlap an interference grating is generated. This has important implications on the FWM signals obtained by this technique and will be discussed in detail in Chapter 4.

2.3.2 Interpretation of four-wave-mixing results

The interpretation of FWM experiments is strongly dependent on the nature of the broadening of the sample under investigation. The first parts of this section discuss FWM in homogeneously and inhomogeneously broadened non-interacting two-level systems. Then, the effects of the Coulomb coupling between excited quasiparticles in semiconductors on the interpretation of FWM experiments will be discussed in the framework of the SBE. The last part of this chapter discusses why some FWM experiments in semiconductors cannot be described by the SBE alone and introduces the phenomenological terms that have to be included in the SBE.
(a) Homogeneously broadened, non-interacting, two-level systems

In a homogeneously broadened ensemble of non-interacting two-level systems all excited two-level systems have the same transition frequency. We assume that the laser is in resonance with this frequency. The homogeneous linewidth $\Gamma_h$ (FWHM) is related to the dephasing time $T_2$ by $\Gamma_h = \frac{2\pi}{T_2}$, for a line shape that is Lorentzian. For laser pulse widths much smaller than the relaxation times $T_1$ and $T_2$, the TI-FWM signal depends on the time delay $\Delta t$ in the following way [36]:

$$\text{TI-FWM} \propto \theta(\Delta t) \exp\left(\frac{-2\Delta t}{T_2}\right),$$

where $\theta$ denotes the Heaviside step function. This function accounts for the fact that, in an ideal two-level system, the FWM emission in direction $2\mathbf{k} - \mathbf{k}$ is zero if the time delay between the two pulses is negative. The spectrally resolved FWM signal, the FWM power spectrum (FWM-PS), is proportional to the third-order polarization

$$\text{FWM-PS} \propto |P_{\text{FWM}}^{(3)}(\omega, \Delta t)|^2.$$

Because the dephasing time $T_2$ is related to the homogeneous linewidth by $\Gamma_h = \frac{2\pi}{T_2}$, it can be deduced from the width of a Lorentzian FWM-PS. Thus, the dephasing time $T_2$ can be measured with TI-FWM and SR-FWM when the semiconductor can be modeled as a homogeneously broadened ensemble of non-interacting two-level systems.

(b) Inhomogeneously broadened, non-interacting, two-level systems

In an inhomogeneously broadened ensemble of two-level systems, each two-level system has a different transition frequency. The phases of the different frequency components evolve at different rates when excited by the first laser pulse $\mathbf{k}$. As a result, the macroscopic polarization decays to zero with a time either proportional to the laser pulse duration or inversely
proportional to the inhomogeneous linewidth $\Gamma_{in}$ depending on whether the inhomogeneous linewidth $\Gamma_{in}$ is larger or smaller than the linewidth of the laser pulse [1]. This decay of the macroscopic polarization is not due to dephasing but rather to destructive interference. The second laser pulse $k_2$ reverses the phases of the different frequency components if the time delay $\Delta t$ is smaller than the dephasing time $T_2$. When this is true, a photon echo is emitted at $\Delta t$ after the second laser pulse $k_2$ [1, 35, 36].

For the inhomogeneously broadened case, the TI-FWM signal depends on the delay time $\Delta t$ as [36]:

$$\text{TI-FWM} \propto \theta(\Delta t) \exp\left(-\frac{4\Delta t}{T_2}\right).$$

(2.33)

The dephasing time $T_2$ can be measured with TI-FWM experiments.

(c) Semiconductors: excitonic excitations

As mentioned in Chapter 2.2, Coulomb effects can strongly influence the coherent dynamics of optically excited electron-hole pairs and the OBE are no longer valid to describe these dynamics. Instead, the SBE, which account for the Coulomb coupling between excited carriers in a semiconductor, have to be used [1]. We will give here one example of the changes in the FWM signals due to Coulomb interaction because it is important for the interpretation of the FWM experiments in the chapters 3 to 5.

Let us assume that homogeneously broadened excitons at the bandedge are resonantly excited by a laser pulse and no additional continuum transitions are excited. FWM emission in direction $2k_1 - k_1$ at negative time delays $\Delta t < 0$ is generated due to the renormalized Rabi frequency [45]. This leads to a slow rise of the FWM signal at negative time delays. The slow rise of the TI-FWM signal is related to the dephasing time $T_2$ by [45, 46]:
The FWM emission at negative time delays vanishes for inhomogeneously broadened transitions [45]. The generation process of FWM signal at negative time delays is referred to as polarization scattering [45, 46]. The decay of the TI-FWM signal at positive time delays is not influenced by many-body Coulomb interactions and can be determined from equation (2.31).

\[ \text{TI-FWM} \propto \theta(-\Delta \gamma) \exp \left( \frac{4\Delta \gamma}{T_2} \right). \]  

(d) Semiconductors: continuum excitations

The semiconductor continuum can be considered as an inhomogeneously broadened ensemble of non-interacting two-level systems if Coulomb interaction is neglected. In this situation, one expects a photon-echo-like decay of the FWM signal if only continuum states of a semiconductor are excited [1]. This was experimentally proved by Lohner and coworkers [47] with TR-FWM spectroscopy at high carrier densities. In this situation, the FWM-PS has a smooth, unstructured shape, similar to the shape of the exciting laser pulse [47, 48]. Dephasing experiments on semiconductor continuum states have also been performed by Becker and coworkers and Bigot and coworkers [12, 49]. The obtained dephasing times increased with decreasing carrier density. However, in these experimental works, the photon-echo-behavior of the decay of the FWM signal was not checked.

Photon-echo-like behavior of the decay of the FWM signal was not only observed in undoped semiconductors at high carrier densities [12, 34, 47, 49] but also in modulation doped GaAs/AlGaAs quantum wells [50]. In both cases, the Coulomb coupling was reduced due to screening of the Coulomb interaction due to optically excited carriers or carriers resulting from the
modulation doping [50]. The reduced Coulomb interaction makes the independent transition picture applicable.

At lower carrier densities, the description of a semiconductor continuum as an ensemble of non-interacting, two-level systems is questioned because the screening of the Coulomb interaction is reduced and, in turn, the Coulomb interaction is increased. Experiments have shown that the excitonic transitions at the bandedge, if they are excited, determine the FWM-PS at lower carrier densities [31-33, 51]. This is also true if the excitonic transitions are excited together with a large amount of continuum transitions [31, 32, 52]. In this excitation situation, exciton-continuum interaction becomes important for the interpretation of FWM experiments and the assumption of non-interacting, two-level systems is no longer valid.

If, together with discrete excitonic transitions at the bandedge many continuum states of the semiconductor are excited, effects are observed [33, 52], which cannot be described by the SBE [33]. These effects manifest themselves in a pulse-limited decay of the TI-FWM signal even though, surprisingly, the FWM-PS is narrow. This phenomenon was first observed at higher-order magnetoeexcitons in bulk semiconductors [28, 29] and was referred to as destructive quantum interference (DQI). In this thesis we describe the first observation of DQI at higher-order excitons in quantum wells without magnetic field [19]. In these cases, the excitonic transitions are embedded in the continuum transitions of lower-order excitons and continuum transitions are always excited with them. A detailed experimental analysis of DQI of higher-order excitons in quantum wells will be given in the next chapter.

The fast decay of the TI-FWM signal and the narrow linewidths of the FWM-PS is not explained by the SBE. The SBE predict that both the spectral width and the decay of the FWM signal in the time delay domain at the exciton resonance should be governed by the excitonic dephasing rate [33]. Thus, a fast pulse-width limited decay of the TI-FWM implies a
broad FWM-PS. To account for the discrepancy between the narrow spectral width and the fast decaying TI-FWM signal, a phenomenological dephasing rate, which increases linearly with the excited carrier density, is introduced into the SBE [30, 33]. This concept is known as excitation-induced dephasing (EID) [53]. The dephasing rate \( \gamma \) is then given by:

\[
\gamma(n(t)) = \gamma + \gamma' n(t) ,
\]

(2.35)

where \( n \) is the carrier density. If the density-dependent polarization decay is phenomenologically introduced into the SBE, the fast decay of the FWM signal in the time delay domain can be modeled when the FWM-PS is narrow [33]:

\[
\left. \frac{\partial P_k(t)}{\partial t} \right|_{\text{coinc}} = -\left( \gamma + \frac{\gamma'}{2} \sum_{q,j} f_{jq}(t) \right) P_k(t) .
\]

(2.36)

With this extension of the SBE an additional source term for the polarization of a state \( k \) arises. This term is given by \(-\frac{\gamma}{2} \sum_{q} (f_{eq} + f_{hq}) P_k\). As a consequence, the carrier population (sum over states \( q \)) can modify the polarization of state \( k \), e.g., the population in the continuum can modify the polarization of the exciton [31-33]. The additional source term due to EID can also be understood as a many-body coupling between excitonic and continuum states due to the Coulomb interaction. The coupling of different states can also be described by a multi-level system with a common ground state [30]. While this phenomenological ansatz makes possible the modeling of the fast decay of the FWM signal in the time delay domain when the FWM-PS is narrow, a microscopic explanation is not yet available [33, 53]. Implications of this phenomenological ansatz are discussed in following chapters.

The photon-echo-like behavior of the coherent emission from undoped semiconductors at higher carrier densities and the DQI behavior at lower carrier densities implies a transition from DQI to photon-echo-like
behavior. At lower carrier densities Coulomb interaction is not screened. With increasing carrier density the screening of the Coulomb interaction is enhanced and, in turn, the Coulomb interaction is decreased. Therefore, at lower carrier densities the excitonic and continuum transitions in a semiconductor are coupled due to many-body Coulomb interaction mediated by EID. At higher carrier densities the coupling is weakened and the semiconductor continuum can be described by an inhomogeneously broadened ensemble of weakly or non-interacting two-level systems. The transition from DQI to dephasing is not fully understood and a microscopic many-body description is not available. It depends on several factors like carrier density, sample temperature, and the dimensionality of the system (bulk material or quantum well).

2.3.3 Experimental tools of four-wave-mixing spectroscopy in semiconductors

To investigate dephasing processes in semiconductors, a light source producing pulses with widths smaller or at least comparable to the dephasing times of the excited electron-hole pairs, is needed. This means that the light pulses must have a width in the range of 10 fs to several hundreds of fs. Throughout this work commercially available and home-built Ti:sapphire lasers were used as light sources. The wavelength range of the Ti:sapphire laser spans from 650 nm to 1050 nm and overlaps the absorption edge of GaAs. The commercially available Ti:sapphire laser (Spectra Physics, Tsunami) emits pulse trains with a repetition rate of about 100 Mhz. Each pulse has a full width at half maximum (FWHM) of 100 fs. The distance between adjacent pulses is 10 ns. The pulse energy is around 10 pJ if an average output power of 1 mW is assumed. The commercial Ti:sapphire laser system is pumped by an argon-ion laser. The home-built Ti:sapphire laser allows for pulse widths of only 5.5 fs [7]. This laser system is pumped by a frequency-doubled diode pumped Nd:YVO4 (Nd3+ ions doped in a yttrium vanadate
crystalline matrix) laser. Again, the repetition rate is about 100 Mhz. The pulse generation process in both lasers is due to the Kerr-effect [6]. The Kerr-lens mode-locking process in the home-built Ti:sapphire laser is assisted by a semiconductor saturable absorber mirror (Sesam) [54, 55], which leads to a very stable output power and to self-starting of the pulse generation process. The shortest pulses used in the experiments described in this thesis have a FWHM of about 15 fs. This corresponds to a spectral width of about 90 meV. The group delay dispersion inside and outside the laser cavity has to be controlled with prism pairs [56] and double-chirped mirrors [57, 58] to achieve these short pulses and to maintain the pulse duration at the position of the sample. For pulses with a FWHM of 100 fs, special dispersion control outside the laser cavity is not necessary.

For the FWM experiments the laser pulse is split into two pulses with a 50/50 beam splitter (Fig. 2.2). Therefore, both pulses are coherent and approximately equal in power. The time delay between the two pulses is controlled with a variable delay stage driven by a stepper motor with a resolution of 1 μm per step (FWM experiments with pulses with a FWHM of 100 fs) or 0.1 μm per step (FWM experiments with pulses with a FWHM of 15 fs) making the time delay adjustable in 6.67 fs or 0.67 fs steps.

The emitted FWM signal (Fig. 2.2) is detected either as function of the time delay with a photo multiplier tube (PMT) with lock-in technique (TI-FWM) or spectrally resolved with a monochromator at fixed time delays (SR-FWM). The PMT in our experiments is a GaAs(Cs) type PMT with a dark current of less than 0.1 nA at room temperature. The dark current is reduced by a factor of 2 to 3 by cooling the PMT with a Peltier element to −15° C. Cooling of the PMT is necessary because the power of the emitted FWM signals is on the order of a few pW.

We use a sum frequency lock-in technique, where both incident beams are chopped. The chopping frequency is usually in the low kHz regime (1-3kHz). With this technique we achieve a maximal signal to noise ratio (SNR) better than 1000.
For the spectral resolution of the FWM signal, we use a 46-cm monochromator with a CCD (charge-coupled device) array as the detector. The CCD array is cooled to 140 K to reduce the number of dark counts. The spectral resolution of the monochromator is about 0.1 meV for an entrance slit with a width of 100 μm and a grating with 1200 grooves per millimeter.

The FWM experiments are performed in the transmission geometry (Fig. 2.2). The semiconductor samples are grown by molecular beam epitaxy (MBE) on 500 μm thick GaAs substrates. The substrate has to be removed by chemical wet etching [59] to allow for transmission experiments because the absorption coefficient of gallium arsenide (GaAs) is in the 10^4 cm⁻¹ range [59]. First, the substrate is thinned to 100 μm with a bromine methanol solution. The sample is then glued on a sapphire disc and etched with a H_2O_2:NH_4OH solution to the first etch stop layer. The next two etch stop layers are removed by a HCl solution and a weak H_2O_2:NH_4OH solution (pH value 7.05), respectively. When the substrate and all etch stop layers have been removed the sample is antireflection coated on the front side. This reduces the Fabry-Perot oscillations of the transmitted light by a factor of 10.

Some of the FWM experiments in this work were performed at low temperatures. The cooling of the semiconductor sample reduces the carrier-phonon scattering rate and the coherent phenomena, which would be normally obscured by this scattering process at higher temperatures, could be observed. An Oxford Instruments liquid helium cryostat is used in some of the following FWM experiments. With this instrument the sample temperature can be varied from 1.8 K to room temperature.
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3. Coherent dynamics of Fano resonances of higher-order excitons in semiconductor quantum wells

Fano resonances generally occur in the linear absorption spectra of higher-order excitons in lower dimensional semiconductors [14, 15, 18, 19, 28, 60]. The Fano interference is caused by the Coulomb coupling between the excitons of higher-order subbands to the continuum of lower-order subbands. In this chapter we will give a short review of the atomic physics Fano-resonance model first proposed by [17] with additional homogeneous broadening. The next part deals with the experimental observation of Fano resonances in semiconductor quantum wells with linear absorption spectroscopy. In Chapter 3.3 the nonlinear optical properties of Fano resonances in semiconductor quantum wells are investigated with four-wave-mixing spectroscopy with 100-fs pulses. It is shown that destructive quantum interference, i.e., pulse-width limited decay of the FWM signal in the time delay domain although the dephasing time is much longer than the pulse width, is an intrinsic feature of Fano resonances in semiconductor quantum wells at lower carrier densities. The FWM experiments at lower carrier densities demonstrate that the signature of multi-subband excitation is a distinctly structured FWM power spectrum. At higher carrier densities the FWM spectra broaden and dephasing begins to dominate the decay of the FWM signal in the time-delay domain. This indicates that destructive quantum interference looses its influence on the decay of the TI-FWM signal at higher carrier densities. However, it is difficult to decide whether destructive quantum interference or strong dephasing is the underlying physical reason for the fast decay of the TI-FWM signal due to the limited time resolution.

In Chapter 3.4 nonlinear optical properties of Fano resonances in semiconductor quantum wells are investigated with an increased time resolution. With this increased time resolution we could show that
destructive quantum interference looses its influence at higher carrier densities.

3.1 Atomic physics Fano model

Fano interference occurs if a discrete transition is coupled to an energetically degenerate continuum of transitions. The discrete and the continuum transition need to be optically excitable, i.e., the optical matrix element between excited and ground states must not vanish, and both transitions must have a common ground state. If all these prerequisites are fulfilled Fano interference manifests itself in an asymmetric line shape of the linear absorption spectrum. The most obvious feature of a Fano resonance is a minimum in the linear absorption spectrum. This minimum can reach zero if any additional homogeneous or inhomogeneous broadening is neglected. The minimum can be situated either energetically below or above the absorption line of the uncoupled discrete transition. We will now give a brief overview of the theoretical Fano model [17, 61]. The discrete state $|d_0\rangle$ is energetically degenerate to a continuum of states $|c(E)\rangle$. Both states are optically excitable from a common ground state $|g\rangle$. The transition energy of the discrete state is $E_0$. The quantum mechanical coupling between the states $|d_0\rangle$ and $|c(E)\rangle$ is given by $V$. The wave function and the transition energy of the discrete state are modified due to the coupling to the continuum. The new transition energy is given by $\Omega = E_0 + E_{coup}$ and the new wave function of the discrete state will be denoted by $F(E)$. Moreover, the coupling leads to an interference of the optical transition amplitudes. At a certain photon energy $E$ the optical transition amplitudes interfere destructively and the minimum in the linear absorption profile occurs. The minimum lies well below the continuum absorption value.
The Fano absorption profile can be calculated by means of degenerate perturbation theory [17, 61]. This leads to a new Fano eigenstate $F(E)$ with an energy dependent optical matrix element $\mu_{F(E)g}$. This is indicated in Fig. 3.1. On the left-hand side the discrete state and the energetically degenerate continuum are depicted. The coupling $V$ is symbolized by an arrow. On the right-hand side the Fano eigenstate, which results from the coupling between the discrete state and the continuum of states, $F(E)$ is symbolized by the shaded area. The resulting Fano absorption profile $A(q,\varepsilon)$ can be described by the following expression:

$$A(q,\varepsilon) = \frac{|\mu_{F(E)g}|^2}{|\mu_{c(E)g}|^2} = \frac{(q + \varepsilon)^2}{1 + \varepsilon^2},$$

(3.1)

where $\mu_{c(E)g}$ is the optical matrix element of the transition from the ground state to the continuum of states. $\varepsilon$ is a normalized energy given by $\varepsilon = 2(E - \Omega)/\Gamma$, where $\Omega$ is the transition energy of the modified discrete state and $\Gamma$ the broadening of the modified discrete state due to the coupling to the continuum of states. $\Gamma$ is related to the coupling $V$ by $\Gamma = 2\pi|V|^2$. The Fano parameter $q$ is proportional to the ratio of the dipole matrix element $\mu_{dc}$ (transition from ground state to modified discrete state) to the dipole matrix element $\mu_{c(E)g}$ (transition from the ground state to the continuum of states):
The Fano q-parameter also determines the position of the minimum in the Fano absorption profile. For $q>0$ the minimum of the Fano absorption profile is shifted down in energy and for $q<0$ it is shifted up in energy with respect to the maximum of the absorption line. In Fig. 3.2 some examples of Fano absorption profiles $A(q,\epsilon)$ are plotted for the Fano q-parameters $q = 0.5$, $q = 1$, $q = 2$, and $q = -1$. No additional homogeneous or inhomogeneous broadening is assumed. It holds that $A(q,\epsilon) = A(-q,-\epsilon)$.

We will now comment on the lifetime of the Fano eigenstate. The Fano eigenstate will decay with a time constant inversely proportional to $\Gamma$, although the discrete transition is not homogeneously broadened at all. This can be seen from the following calculation. The susceptibility $\chi(\omega)$ of the Fano eigenstate is given by

$$q = \frac{\mu_{le}}{\pi V^* \mu_{l(E)q}}.$$
\[ \chi(\omega) = \int d\omega_0 \left| \frac{\mu_{E(\omega_0)}}{\omega_0 - \omega - i\gamma} \right|^2, \] (3.3)

where \( \gamma \) is the additional homogeneous broadening of the discrete transition. After some calculations and with the relations \( v = (\Omega - \omega) / \Gamma \) and \( g = \gamma / \Gamma \) we obtain

\[ \chi(v) = \left| \mu_{jk} \right|^2 \frac{2}{\Gamma} \left[ 1 - \frac{2i}{q} + \frac{g + iv}{q^2} \right]. \] (3.4)

If we now assume that \( \gamma = 0 \), i.e. no homogeneous broadening of the discrete state, the susceptibility can be written as

\[ \chi(\omega) = \left| \mu_{jk} \right|^2 \frac{2}{\Omega - \omega - i\Gamma} \left[ 1 - \frac{2i}{q} + \frac{iv}{q^2} \right]. \] (3.5)

In the limit \( q \to \infty \) we recover the Lorentz profile and it becomes clear that the Fano eigenstate decays with a time constant inversely proportional to \( \Gamma \), although no homogeneous broadening is assumed. The lifetime of the Fano eigenstate is due to the coupling to the continuum of states.

The linear polarization, which is given by \( P(\omega) = \chi(\omega)E(\omega) \), also decays with time constant \( 1/\Gamma \). In the \( \delta \)-function approximation the third-order polarization, i.e., the quantity which determines the signal in FWM experiments, can be expressed by the first order polarization in the framework of the optical Bloch equations [62]. Therefore, the atomic physics Fano model predicts that in FWM experiments the dephasing time of a Fano resonance should be proportional to \( 1/\Gamma \). In the Chapter 3.3 we show that exciton Fano continua in semiconductors behave differently.

### 3.2 Fano resonances in the linear absorption spectra of quantum wells
Semiconductor quantum wells can possess several electron and hole subbands, depending on the properties of the confinement potential. In interband optics, the subband structure leads to the observation of several excitonic Rydberg series, each one corresponding to an optically allowed transition between an electron and a hole subband. The exciton resonance corresponding to the energetically lowest \( n = 1 \) electron and hole subbands is well separated from continuum transitions by its binding energy and, therefore, is a discrete transition. In contrast, higher-order excitons corresponding to electron and hole subbands with \( n > 1 \) are embedded in continuum states of lower electron and hole subband pairs.

In semiconductors, Coulomb coupling between higher-order excitons and lower-order continuum states has theoretically been shown to result in Fano interference and in the formation of Fano resonances in quantum wells and wires [14-16]. The so-called \( |F(E)\rangle \) exciton Fano resonance is a structured continuum, consisting of a higher-order exciton coupled to energetically degenerate continuum states by the bare Coulomb potential [14]. These structured continua are the true eigenstates of one- and two dimensional semiconductors above the band edge.

Experimentally, Fano absorption profiles have been observed in quantum wells [18] by means of photoluminescence excitation (PLE) and in bulk semiconductors under magnetic field [13, 20]. The linear optical properties of these coupled exciton-continuum resonances are well understood [13-16, 18] and calculations show that in semiconductor quantum wells Fano resonances with positive and negative \( q \)-parameter occur [61].

We will discuss here the linear optical properties of Fano resonances in a 160 Å wide GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As multiple quantum well and in a 500 Å wide GaAs/Al\(_{0.3}\)Ga\(_{0.7}\)As multiple quantum well. Higher-order excitonic resonances in these quantum wells show a Fano absorption profile with different \( q \)-parameter. The nonlinear optical properties of the Fano resonance in the 500 Å wide quantum well will be discussed in the following chapter.
The different excitonic transitions in a quantum well are denoted by the subbands that are involved in the optical transition of this particular resonance. The electronic subbands are abbreviated by e and the heavy- and light-hole subbands by hh and lh, respectively. In an infinitely deep quantum well only transitions between electron and hole subbands with the same quantum number are allowed. In a finitely deep quantum well this condition is weakened and additional transitions are allowed if \( |n_{\text{electron}} - n_{\text{hole}}| \) is an even number [2, 63]. For example, the transition from the third heavy-hole subband (hh3) to the first electron subband (e1) is allowed and will be abbreviated by e1hh3.

In 1989 H. Chu and coworkers [60] theoretically predicted that the e1hh3 resonance in a quantum well shows a Fano absorption profile. The minimum of this Fano absorption profile should lie energetically below the maximum absorption, which

![Absorption spectrum](image)

**Fig. 3.3** Linear absorption spectrum of a 160 Å GaAs/Al0.3Ga0.7As multiple quantum well at 10 K. The dotted curve is a Fano fit to the e1hh3 absorption line with the parameters \( q = 3.2 \), \( \Gamma = 0.71 \) meV, and \( \Omega = 1.5554 \) eV.

...corresponds to a positive Fano q-parameter. The first experimental proof of this prediction was reported by Oberli and coworkers in 1994 by means of
photoluminescence excitation (PLE). We will show here that the observation of a Fano resonance is also possible with linear absorption measurements, a technique, which is similar to the PLE technique. Fig. 3.3 shows the linear absorption spectrum of a 160 Å GaAs/Al\textsubscript{0.3}Ga\textsubscript{0.7}As multiple quantum well at a temperature of 10 K. The spectrum consists of several exciton resonances which correspond to transitions between electron (e) and heavy-hole (hh) subbands and electron and light-hole (lh) subbands. The e1hh1 exciton absorption line at 1.528 eV and the e1lh1 exciton absorption line at 1.5315 eV show a Lorentz absorption profile because these transitions are not energetically degenerate to a continuum of states. Thus, they are discrete transitions. However, the e1hh3 exciton absorption line, which is energetically degenerate to the continuum of the hh1 and the lh1 subbands, shows a Fano absorption profile. The coupling between the discrete transition and the degenerate continuum is due to Coulomb interaction. The minimum of this absorption profile lies well below the continuum absorption and, as predicted, energetically below the maximum absorption. The fit is obtained by means of the Fano absorption profile $A(q,\Gamma)$. No additional homogeneous broadening is assumed [64]. The fitting parameters are $q = 3.2$, $\Gamma = 0.71$ meV, and $\Omega = 1.5554$ eV. The positive Fano q-parameter accounts for the fact that the absorption minimum lies energetically below the absorption maximum.

A different example of a Fano absorption profile in a quantum well is shown in Fig. 3.4. The figure shows the linear absorption spectrum of a GaAs/Al\textsubscript{0.3}Ga\textsubscript{0.7}As multiple quantum well sample at 7 K temperature. It contains ten 500 Å broad GaAs/Al\textsubscript{0.3}Ga\textsubscript{0.7}As quantum wells. Again, the spectrum comprises several exciton resonances which correspond to transitions between electron (e) and heavy hole (hh) subbands with the same quantum number $n$, labeled
Coherent dynamics of Fano resonances...

Fig. 3.4 Linear absorption spectrum of a 500Å wide quantum well at $T = 7\, K$. The dashed curve is a Fano-profile fit to the absorption line of the Fano resonance $e3hh3$. The parameters are $q = -2.1$, $\Gamma = 0.58\, \mathrm{meV}$, and $\Omega = 1.5339\, \mathrm{eV}$.

as $e1hh1$, $e2hh2$, $e3hh3$. The lowest light hole (lh) exciton resonance, $e1lh1(1.5126\, \mathrm{eV})$, is found energetically below the $e1hh1\, (1.5141\, \mathrm{eV})$ exciton. This is because mechanical strain at low temperatures has shifted both the hh and the lh valence bands to lower energies where the shift of the lh band is larger [65] and overcompensates the difference in quantization energies of the $e1hh1$ and $e1lh1$ excitons. Again, the linear absorption spectrum also shows several exciton resonances with $n_{\text{electron}} \neq n_{\text{hole}}$. The lowest exciton transitions $e1hh1$ and $e1lh1$ have symmetric Lorentzian line shapes with a FWHM of only 0.4 meV, demonstrating small inhomogeneous and homogeneous broadening in this high-quality sample. The higher-order exciton resonances are structured exciton Fano continua with an asymmetric line shape which is the signature of Fano interference between exciton transitions and energetically degenerate continuum transitions [17], e.g. the $e2hh2$ resonance at 1.521 eV and the $e3hh3$ resonance at 1.5335 eV. The different types of transitions are coupled by the bare Coulomb potential [14]. The $e3hh3$ excitonic absorption line in the 500 Å quantum well shows a clear signature of a Fano resonance. This time, the minimum of the absorption lies energetically above the maximum
absorption, which correspond to a negative Fano q-parameter. The dashed curve is again a Fano-profile fit with no additional homogeneous broadening [64]. The parameters are q = - 2.1, Γ = 0.58 meV, and Ω = 1.5339 eV. The negative Fano q-parameter accounts for the fact that the minimum absorption lies energetically above the maximum absorption. The calculated Fano absorption profiles fit very well the observed resonances. This reconfirms the results of previous work [14-16, 18, Chu, 1989 #1549].

Thus, the linear optical properties of Fano resonances in semiconductor quantum wells are well understood and they can be described by the atomic physics Fano model. However, much less is known about the coherent nonlinear optical properties of quantum wells for multi-subband excitation involving the lowest discrete exciton transition and exciton Fano continua. Therefore, we investigated the nonlinear optical properties of the e3hh3 exciton Fano continuum by means of degenerate FWM technique. The results will be given in the following chapter.

3.3 Coherent dynamics in quantum wells for multi-subband excitation

This chapter deals with the transient coherent emission from the 500 Å wide quantum well for multi-subband excitation, i.e., the coherent dynamics of higher-order excitons is investigated. These higher-order excitons are energetically degenerate to the continuum of lower-order subbands. Spectrally resolved transient four-wave mixing (FWM) experiments with 100-fs pulses were performed over a wide range of carrier densities. These experiments will shed light on the nonlinear optical properties of two-dimensional exciton Fano continua in quantum wells.

The coherent emission from exciton Fano continua has been studied for one-dimensional magnetoeexcitons in bulk GaAs in [28]. Here, we
investigate for the first time the coherent properties of two-dimensional exciton Fano continua in quantum wells at zero magnetic field [19]. We note that the lowest Lorentz exciton shows a distinctly different behavior in coherent nonlinear optical experiments under a strong magnetic field compared to the zero-field case [66]. Therefore, it is not a priori possible to draw the conclusion that the nonlinear optical properties of magnetoexciton Fano continua are similar to the nonlinear optical properties of exciton Fano continua in quantum wells at zero field.

Moreover, our experiments address the question how coupling between the lowest Lorentzian exciton and the structured exciton Fano continua in the nonlinear regime affects the coherent emission from a quantum well. The interaction between the lowest Lorentzian exciton and continuum transitions has been studied for the unstructured "flat" continuum of the lowest subband [30-33, 47, 51] and for the magnetoexciton system [29] but not for exciton Fano continua in quantum wells at zero magnetic field. We will point out the similarities and differences between the different exciton-continuum systems.

The results of transient FWM experiments for multi-subband excitation of a quantum well at lower and intermediate carrier densities are presented in section 3.3.1. We determine the dephasing times of the different resonances from the width of the emission lines in the FWM spectra. Our data show that the decay of the FWM emission from quantum well exciton Fano continua versus the time delay between the excitation laser pulses is not due to dephasing at these densities. This effect is attributed to quantum interference between exciton and continuum states, which has its origin in many-body exciton-continuum coupling. Similar results and conclusions were obtained in [28, 29] for magnetoexciton Fano continua, showing that this effect is of general importance for exciton Fano resonances in semiconductor quantum wells. This effect was not observed with Fano resonances in biased semiconductor superlattices [24]. Here, the decay of the TI-FWM signal is due to dephasing and agrees well with the times expected from the FWM linewidth. In section 3.3.2, we will discuss
the properties of the coherent emission from quantum wells for multishubband excitation at higher carrier densities.

**Experimental**

We used degenerate four-wave mixing as the appropriate experimental tool. This technique was already described in Chapter 2.3 of this work. The four-wave-mixing experiments were performed with bandwidth-limited 100 fs pulses from a Ti:sapphire laser in the standard two-beam configuration with collinearly polarized excitation pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. Both pulses had equal intensities. The excitation pulses are separated in time by a variable time delay $\Delta t$, which is controlled by a stepper motor. The nonlinear interaction in the sample gives rise to a FWM signal emitted in the phase-matching direction $2\mathbf{k}_2 - \mathbf{k}_1$. We detect the FWM signal either spectrally integrated as a function of the time delay $\Delta t$ ($\propto \int |P^{(3)}(\omega, \Delta t)|^2 d\omega$) where $P^{(3)}(\omega, \Delta t)$ is the nonlinear polarization in third order) or spectrally resolved ($\propto |P^{(3)}(\omega, \Delta t)|^2$) at fixed time delays $\Delta t$. The spectrally resolved FWM measurements as well as linear absorption experiments were performed with a monochromator and a CCD array providing an overall spectral resolution of 0.1 meV.

To allow for transmission experiments, the GaAs substrate of the sample was removed by chemical wet-etching. All experiments were performed at the temperature $T = 7$ K.

**3.3.1 Coherent dynamics at lower and intermediate carrier densities**

(a) Experimental results

In the FWM experiments, the excitation laser spectrum, centered at 1.530 eV, is tuned slightly below the $e_3^3h_3^3$ resonance to excite simultaneously the lowest $e_1h_1$ and $e_1h_4$ excitons and higher-order exciton Fano continua, as shown in Fig. 3.5.
Fig. 3.5 Linear absorption spectrum of the 500 Å wide quantum well at a temperature of 7 K. The exciting laser spectrum is tuned slightly below the $e3hh3$ exciton Fano resonance.

Power spectra of the FWM signal are plotted in Fig. 3.6 for different time delays $\Delta t$ between $-100$ fs and $167$ fs and a carrier density $N_0 \approx 3 \cdot 10^{10}$ cm$^{-2}$ per excitation pulse. Unlike the excitation pulse spectrum, the FWM spectra show a distinct structure which arises from the discrete exciton transitions at the bandedge and from exciton transitions which are embedded in the continuum. This distinct structure is the signature of multi-subband excitation at lower carrier densities. The largest emission line in the FWM spectrum at 1.514 eV originates from the $e1hh1$ exciton. A significant contribution to the FWM spectrum at 1.534 eV can be traced back to the $e3hh3$ exciton Fano continuum.
Fig. 3.6 Four-wave-mixing power spectrum for multi-subband excitation of the 500 Å quantum well. Time delay from \(-100\) fs to \(+167\) fs. The strong emission at \(1.514\) eV results from the discrete e1hh1 exciton resonance, the emission at \(1.534\) eV from the e3hh3 exciton Fano resonance. Carrier density \(N_0 = 3 \cdot 10^{10}\) cm\(^{-2}\) (lowest density), temperature 7 K.

Very small contributions from the e2hh2 transition at 1.521 eV and from transitions with \(n_{\text{electron}} \neq n_{\text{hole}}\) are also visible, e.g. the e1hh5/e1lh3 transition at 1.524 eV. All emission lines in the FWM spectrum are much narrower than the laser spectrum. Since the dominating e1hh1 and e3hh3 resonances are well separated from the adjacent resonances, these FWM resonances can be analyzed more quantitatively.

In Fig. 3.7, we have plotted the full width at half maximum (FWHM) of the e1hh1 Lorentzian resonance and the e3hh3 Fano resonance versus time delay for the excitation density \(N_0 = 3 \cdot 10^{10}\) cm\(^{-2}\) (open symbols), as obtained from Fig. 3.6. The filled symbols mark the FWHM of the resonances at the carrier density 3\(N_0\) and the dashed line is the width of the e1hh1 resonance in the linear absorption spectrum in Fig. 3.4, i.e., at zero carrier density. We note that the FWHM of both resonances is essentially independent of the time delay but increases with increasing excitation.
density. The density dependence of the linewidth of the e1hh1 resonance in

![Graph](image)

**Fig 3.7** Full width at half maximum of the e1hh1 resonance (circles) and the e3hh3 resonance (squares) in FWM power spectra. Open symbols: carrier density \( N_0 = 3 \times 10^{10} \text{ cm}^{-2} \), filled symbols: carrier density \( N = 3N_0 \). The dashed line is the FWHM of the e1hh1 exciton resonance in the low-temperature linear absorption spectrum.

The density range between zero and \( 3N_0 \) demonstrates that density-independent inhomogeneous broadening makes only a small contribution to the linewidth for densities of \( N_0 \) and larger in this high-quality sample. The measured line widths for \( N_0 \) and \( 3N_0 \) are dominated by homogeneous broadening, which is enhanced at higher carrier densities due to increased carrier-carrier scattering. Two features of the data are noteworthy. (i) For a fixed carrier density, the homogeneous linewidth of the e3hh3 exciton Fano resonance is always larger than the homogeneous linewidth of the e1hh1 Lorentzian exciton. The average FWHM of the e1hh1 resonance is 0.87 meV for carrier density \( N_0 \) and 1.29 meV for \( 3N_0 \). The average FWHM of the e3hh3 resonance is 1.58 meV for \( N_0 \) and 2.13 meV for \( 3N_0 \). This result reflects that, for Fano resonances, the coupling \( \Gamma \) to the energetically degenerate continuum is an additional
intrinsic contribution to the total homogeneous broadening, which adds to
the natural homogeneous broadening \( \gamma \) resulting from quasi-particle
scattering [64]. (ii) The Fano coupling \( \Gamma \) is expected to decrease with
increasing carrier density [64] because \( \Gamma \) is due to Coulomb interaction
[14], which is weakened by screening at elevated carrier densities.
However, we observe that the total homogeneous linewidth of the c3hh3
exciton Fano resonance increases with increasing carrier density.
Consequently, we conclude that the natural homogeneous broadening \( \gamma \)
shows a strong increase with increasing carrier density which
overcompensates the decrease of the Fano coupling \( \Gamma \). The same
conclusion as for exciton Fano continua in quantum wells was reached for
Fano magnetoexcitons [64].

Since inhomogeneous broadening is negligible at the carrier densities \( N_0 \)
and \( 3N_0 \), we can estimate the dephasing times of the different resonances
from the FWHM of the emission lines in the FWM spectrum. A single
dephasing time characterizes each resonance for a given carrier density
since the FWHM of the resonances does not show a significant
dependence on the time delay. We use the fundamental relation
\( \tau = T_\gamma / 2 = h / \Delta E \) where \( \tau \) is the decay time, \( T_\gamma \) the dephasing time, and \( \Delta E \)
the FWHM of the emission lines [29]. From this relation and from the
average FWHM, we can estimate dephasing times of 1.5 ps and 1.0 ps for
the e1hh1 exciton at the excitation density \( N_0 \) and \( 3N_0 \), respectively. For
the e3hh3 resonance, we obtain dephasing times of 0.83 ps at \( N_0 \) and
0.61 ps at \( 3N_0 \). We note that LO phonon emission does not limit the
dephasing time of the e3hh3 transition since the small subband
separations in the 500 Å quantum well prohibit this process.

Figure 3.8 shows the spectrally and temporally integrated FWM signals (TI-
FWM) for multi-subband excitation and carrier densities \( N_0 \) and \( 3N_0 \). The
spectrally integrated FWM signals as a function of the time delay between
the excitation pulses follow the excitation laser pulse shape over almost
two orders of magnitude, i.e., the signals decay with a time constant of less than 100 fs.

![Graph](image)

**Fig. 3.8** Spectrally and temporally integrated FWM-signal (TI-FWM) versus time delay $\Delta t$ for multi-subband excitation. Dashed line, carrier density $N_0 = 3 \cdot 10^{10}$ cm$^{-2}$; solid line, carrier density $3N_0$. Temperature $T=7$ K.

Importantly, for the excitation density $N$, none of the exciton resonances has a shorter dephasing time than the e3hh3 exciton, i.e. the e3hh3 exciton has the greatest linewidth. This dephasing time is 0.83 ps, much longer than the pulse-width limited decay of the spectrally integrated FWM signal in the time delay domain. Consequently, we conclude that the pulse-width limited decay of the spectrally integrated FWM signal is not caused by dephasing for multi-subband excitation at the lowest density $N_0$. We refer to this effect as destructive quantum interference (DQI). It is characterized by the following phenomenological features: (i) The decay of the FWM signal in the time delay domain is pulse width-limited although the dephasing times of the involved transitions are much longer than the pulse width. (ii) The pulse-width limited decay dominates the shape of the FWM signal in the time delay domain and any slowly decaying contribution
to the TI-FWM signal is much smaller than expected from the weight of the narrow resonances in the FWM power spectrum.

For the density $3N_s$, dephasing is faster and the difference between the dephasing times and the decay time of the spectrally integrated FWM signal becomes smaller. The signal to noise ratio (SNR) of the FWM signal for the densities $N_s$ and $3N_s$ is better than 50. Therefore, any slowly decaying tail of the TI-FWM signal must be smaller than 2% of the maximum of the TI-FWM signal. When DQI is effective, we would expect a much larger contribution than 2% from the distinct resonances to the total FWM power spectrum. This is indeed the case and we will show this in the next paragraph.

The coherent emission from the Lorentzian e1hh1 exciton is at least 60% of the total FWM emission for the excitation densities $N_s$ and $3N_s$. The contribution from the e3hh3 resonance is about 10% of the total FWM emission at these densities. The relative contribution of every exciton resonance to the whole FWM signal does not change with the time delay $\Delta t$. Therefore, the pulse-width limited decay of the total, spectrally and temporally integrated FWM signal versus time delay implies that neither the coherent emission from the e1hh1 exciton nor the coherent emission from the e3hh3 exciton Fano continuum show a pronounced slowly decaying tail which would reflect the dephasing time of the respective resonances. For multi-subband excitation at the lowest density $N_s$, the coherent emission from both the e1hh1 and the e3hh3 resonances decays much faster in the time delay domain than predicted by the dephasing times of the respective transitions. Consequently, the decay of the coherent emission from the e1hh1 exciton and from the e3hh3 Fano continuum in the time delay domain is not due to dephasing for multi-subband excitation at the density $N_s$.

In contrast, the decay of the coherent emission from the e1hh1 exciton versus time delay reflects the dephasing time if only the Lorentzian excitons at the band edge are excited, as expected [1]. The Lorentzian excitons can be excited without additional excitation of continuum
transitions. This cannot be achieved for exciton Fano continua. Here, always continuum transitions are simultaneously excited together with the exciton resonance. In Fig. 3.9 both cases are compared for the carrier density $N_\text{c}$. The solid curve shows the FWM signal for excitation of exciton Fano continua. A pulse-width limited decay is observed. For excitation of the lowest excitons (center of the laser spectrum is tuned to 1.504 eV) the well known quantum beating is observed [67, 68]. The beating period of 2.69 ps corresponds very well to the spectral distance of the $e_1hhl$ and $e_1lh1$ excitons (see Fig. 3.5). The dephasing time is $T_2 = 2.8$ ps. This corresponds to the line widths of the SR-FWM signal shown in Fig. 3.10. Thus, for excitation of the lowest Lorentzian excitons without continuum excitation the normal dephasing behavior is observed as expected from a homogeneously broadened ensemble of two-level systems.

**Fig. 3.9** Spectrally and temporally integrated FWM signal (TI-FWM) versus time delay $\Delta t$. Solid curve, excitation of exciton Fano continua, laser is slightly tuned below the $e_3hh3$ resonance. Dashed curve, excitation of the lowest $e_1hh1,e_1lh1$ Lorentzian excitons without any continuum transitions. Carrier density $N_0 \approx 3 \times 10^{10}$ cm$^2$. Temperature $T = 7$ K.
Fig: 3.10 Four-wave-mixing power spectrum (FWM-PS) for excitation of the Lorentz excitons at the bandedge. Time delay $\Delta t = 0$. The FWHM of the e1lh1 exciton is 0.35 meV and the FWHM of the e1hh1 exciton is 0.7 meV. The FWHM does not change with time delay. Carrier density $N_a = 3 \times 10^{10} \text{cm}^{-2}$. Temperature $T = 7 \text{ K}$.

We conclude that the simultaneous excitation of the Lorentzian exciton is not the reason for the discrepancy between the decay in the time delay domain and the time scale of dephasing observed for the e3hh3 exciton Fano continuum. This discrepancy is an inherent property of exciton Fano continua in quantum wells. As already mentioned, this discrepancy was not observed with Fano resonances in biased semiconductor superlattices [24] due to the spatial separation of discrete and continuum transitions.

The discrepancy between the decay in the time delay domain and the time scale of dephasing for exciton Fano continua in quantum wells are attributed to quantum interference between exciton and continuum transitions. The coupling between the exciton and the continuum transitions is due to a many-body Coulomb interaction. We refer to this effect as to destructive quantum interference (DQI). The same behavior as for exciton Fano continua in quantum wells at zero magnetic field was found for magnetoexciton Fano continua [28, 29]. This shows that destructive quantum interference is of general importance for
one- and two-dimensional exciton Fano continua in semiconductors, regardless of the application of a magnetic field.

The coherent emission from the Lorentzian exciton is also affected by destructive quantum interference in the multi-subband experiments at the density $N_s$ in which exciton Fano continua are excited. A pulse-width limited decay in the time delay domain is observed although the dephasing time of the Lorentzian exciton is much longer than the pulse width. Similar results were obtained at low densities for the simultaneous excitation of the exciton and an energetically separated broadband flat continuum of the same subband [30-33] and for the interaction between Fano magnetoexciton continua and Lorentzian magnetoexcitons [29]. This comparison shows that the discrepancy between the decay in the time delay domain and the time scale of dephasing of the lowest exciton is of general importance in exciton-continuum systems, irrespective of the properties of the continuum. As a consequence, destructive quantum interference dominates the decay of the FWM emission in the time delay domain for multi-subband excitation in quantum wells at low and intermediate densities.

(b) Discussion

In this section, we will discuss that the atomic physics Fano model and the SBE cannot describe the effect of DQI. A carrier density dependent dephasing rate has to be included into SBE to account for DQI. This mechanism is called excitation induced dephasing (EID) [33, 53].

With regard to the mechanism of destructive quantum interference, we first note that the effect for a single Fano continuum cannot be explained in the framework of the atomic physics Fano model [17] even though this model can quantitatively describe the linear absorption line shape, as shown in Chapter 3.2. The atomic physics Fano model describes the formation of exciton Fano resonances due to the coupling between exciton and continuum transitions by the bare Coulomb potential. For a single Fano
continuum, the atomic physics Fano model predicts a decay of the FWM emission in the time delay domain which is determined by the dephasing time [29], in disagreement with the experimental result. To further illustrate this point, we recall that the atomic physics Fano model [17] can be transformed into a system consisting of a ground state optically coupled to \( N \) excited states where the line shape of the resonance is reflected by the energy dependence of the optical matrix element between the ground and the excited states [17, 62]. In such an \( N \)-level system, the dynamics of the coherent emission is the same in the real time and the time delay domain for short-pulse excitation [62]. Consequently, a pulse-width limited decay of the spectrally integrated FWM signal versus time delay implies a pulse-width limited decay of the coherent emission in real time. Due to the Fourier relation between energy and real time (see Chapter 2), this simple model predicts a FWM spectrum which is as broad as the excitation laser spectrum in this situation, in disagreement with the experimental data. Thus, the atomic physics Fano model cannot describe the DQI behavior.

We have shown that DQI is not observed if only discrete Lorentzian excitons are excited (Fig. 3.9; 3.10) This confirms the results obtained in [11, 69]. The DQI effect requires the simultaneous excitation of excitonic and continuum transitions. The coupling between excitonic and continuum transitions is due to many-body Coulomb interaction in the nonlinear regime. This type of coupling is necessary for DQI to occur. This type of coupling is not included in the atomic physics Fano model because this model describes the formation of Fano resonances due to the bare Coulomb potential in the linear regime.

This type of many-body coupling between the excitonic and continuum transitions is also not included in the framework of SBE because the SBE show a strict correlation between the decay in the time-delay domain and the width of the FWM spectrum [31, 33].

An obvious shortcoming of the above models (atomic physics Fano model and SBE) is the assumption of a constant coupling strength between discrete and continuum transitions. In semiconductors, the intersubband
coupling results from Coulomb interaction [14], which is weakened by screening with increasing carrier density. In fact, for magnetoexciton Fano continua it has been experimentally observed that the Fano coupling $\Gamma$ decreases with increasing carrier densities [64]. Therefore, the assumption of constant Fano coupling is only justified for linear absorption experiments where indeed the absorption profiles of exciton Fano continua in quantum wells and in bulk semiconductors under magnetic field can be described by the expressions of the atomic physics Fano model [13, 18].

Besides the weakening of the Fano coupling $\Gamma$, the natural homogeneous broadening $\gamma$ of exciton Fano continua strongly increases with increasing carrier density in nonlinear optical experiments due to carrier-carrier scattering. This has been shown for exciton Fano continua in quantum wells by the analysis of the experimental data in Fig. 3.7 and has also been observed for magnetoexciton Fano continua [64]. The density dependence of the homogeneous broadening of exciton Fano continua suggests to consider excitation-induced dephasing (EID) [53]. Excitation-induced dephasing results from screening of the Coulomb interaction in nonlinear optical experiments and manifests itself by a density-dependent dephasing rate [53].

EID has been considered in order to model FWM experiments where only the lowest exciton at the band edge and an energetically separated broadband continuum of the lowest subband were excited [30-33]. In such single-subband experiments at low densities, a pulse-width limited decay of the FWM emission in the time delay domain was observed [30-32]. The corresponding FWM spectrum showed only a narrow line at the spectral position of the exciton from which a dephasing time much longer than the pulse width could be deduced [30-32]. These experimental results were modeled introducing a density-dependent dephasing rate in the semiconductor Bloch equations [31-33]. As a consequence, the carrier population in the continuum states can modify the polarization of the exciton [31-33], (see Chapter 2.3). This is the essence of the nonlinear many-body exciton-continuum coupling. It has also been pointed out that
the many-body coupling between exciton and continuum states due to EID can be described by a common ground state picture in which quantum interference effects naturally occur [30].

Further theoretical studies including EID on a phenomenological level have shown that the FWM spectrum consists of a spectrally narrow line at the discontinuity of the linear absorption spectrum [33]. Therefore, for the excitation of structured exciton Fano continua, it is expected that the FWM spectrum comprises narrow emission lines corresponding to the exciton Fano resonances. The decay of the FWM signal in the time delay domain is expected to follow the excitation pulse [33]. The experimental observations in Figs. 3.6 and 3.8 are in agreement with the EID model. These results suggest to attribute the destructive quantum interference effect observed for exciton Fano continua in quantum wells and bulk semiconductors under magnetic field [28, 29] to the many-body exciton-continuum coupling resulting from EID [19].

3.3.2 Coherent dynamics at higher carrier densities

We extended the studies of the nonlinear optical coherent response of the 500 Å quantum well to the carrier density \(10^9 \text{ cm}^{-3}\). This density is approximately equal to the Mott density calculated for GaAs quantum wells with infinitely high barriers [70]. We expect that the destructive quantum interference at this higher density is weakened due to the increased screening of the Coulomb interaction.

Power spectra of the FWM emission at this density are plotted in Fig. 3.11 for time delays between -33 fs (bottom) and 167 fs (top). The spectra show contributions from the Lorentzian \(e1hh1\) and \(e1lh1\) excitons below 1.517 eV, from the \(e2hh2\) and \(e3hh3\) Fano continua at 1.521 eV and 1.534 eV, respectively, as well as from transitions with \(n_{\text{electron}} \neq n_{\text{hole}}\). Compared to the data for the density \(N_0\) in Fig. 3.6, all emission lines are much broader and the FWM spectra are much less structured. Moreover,
the shape of the high-density FWM spectra significantly depends on the time delay, in contrast to the results for the density $N_0$.

![Power spectrum of the four-wave-mixing signal (FWM-PS) for multi-subband excitation for time delays $\Delta t = -33, +33, +100, \text{ and } +167 \text{ fs.}$](image)

*Fig. 3.11* Power spectrum of the four-wave-mixing signal (FWM-PS) for multi-subband excitation for time delays $\Delta t = -33, +33, +100, \text{ and } +167 \text{ fs.}$ Carrier density $10N_0 = 3 \cdot 10^{11} \text{ cm}^{-2}$, temperature $7 \text{ K.}$

The data in Fig. 3.11 show that the Lorentzian excitons below the band edge contribute less to the total FWM emission at the largest positive time delay $\Delta t = 167 \text{ fs}$ than at earlier time delays. This behavior was also observed for bulk semiconductors [34, 48, 51, 71] (see Chapter 5). These results have been explained by excitonic polarization scattering at negative time delays. Polarization scattering is only effective on homogeneously broadened transitions and gives rise to a FWM signal from the homogeneously broadened excitonic transitions at negative time delays. The inhomogeneously broadened continuum transitions do not contribute to the FWM signal at negative time delays. At positive time delays, both exciton and continuum transitions contribute to the FWM signal due to phase space filling. This leads to a decrease of the excitonic FWM signal relative to the continuum contribution to the FWM signal [48, 51, 71]. This
interpretation is based on the assumption that the flat continuum can be treated as an inhomogeneously broadened ensemble of uncoupled two-level systems. We will show later that this assumption is reasonable at higher carrier densities. With regard to the shape of the high-density FWM spectra, we recall that the natural homogeneous broadening $\gamma$ of exciton Fano resonances strongly increases with increasing density due to dephasing resulting from carrier-carrier scattering. Likewise, screening at higher carrier densities weakens the Coulomb Fano coupling. This has been directly experimentally observed for magnetoexciton Fano continua [64]. Both effects result in a loss of the structure of the Fano absorption spectrum which smears out and broadens with increasing density [64]. The same effect is seen in the high-density FWM spectra in Fig. 3.11, which do not show the distinct structure of the FWM spectra at the density $N_0$ due to the increased homogeneous broadening.

Thus, the diminished contribution of the Lorentzian excitons to the total FWM signal at larger positive time delays is a general feature of the Lorentzian excitons in bulk semiconductors and semiconductor quantum wells at higher carrier densities. The similar behavior of the flat continuum in bulk semiconductors and the exciton Fano continua in quantum wells indicates that the exciton Fano continua can be treated as an inhomogeneously broadened ensemble of non-interacting two-level systems at higher carrier densities. Since DQI is due to a strong many-body Coulomb coupling between different transitions it should be less effective in exciton Fano systems at higher carrier densities.

The temporally and spectrally integrated FWM signal (TI-FWM) as a function of the time delay is shown in Fig. 3.12. It is pulse-width limited for the density $10N_0$, as for lower densities. This means that also in this high carrier density case one prerequisite for DQI is fulfilled. However, at this carrier density the FWM power spectrum consists of several lines, which are much broader and much less structured than for the low-density case. At larger positive time delays the FWM spectra extend over a range comparable to the spectral width of the excitation laser pulse (Fig. 3.11).
Therefore, the discrepancy between the fast decay in the time-delay domain and the dephasing times obtained from the widths of the FWM power spectrum is weakened. This and the 100-fs time resolution of the experiment makes it impossible to decide whether the DQI effect or fast dephasing leads to the pulse width-limited decay of the TI-FWM signal.

![Fig. 3.12 Spectrally and temporally integrated four-wave-mixing (TI-FWM) signal versus time delay $\Delta t$ for multi-subband excitation. Pulse-width limited decay. Carrier density $10N_0 = 3 \times 10^{11} \text{cm}^{-2}$. Temperature $T = 7 \text{ K}$.](image)

In the next paragraphs we present a more quantitative analysis of the FWM spectra to substantiate the statement that at higher carrier densities continuum transitions can be treated as an inhomogeneously broadened ensemble of two-level system and, in turn, DQI looses its importance. The relative contributions of the $e1hh1$ and the $e3hh3$ transitions are plotted versus time delay for carrier densities $N_0$ (circles), $3N_0$ (triangles), and $10N_0$ (squares) in Fig. 3.13. As in the low carrier density case, we define the relative contribution of a resonance as the area of the resonance in the FWM spectrum divided by the total area of the spectrum. The relative contribution of the different resonances does not significantly depend on the time delay for the densities $N_0$ and $3N_0$, as already mentioned in section 3.3.1. At the highest density $10N_0$, the $e1hh1$ resonance looses
strength at large time delays, as discussed for Fig. 3.11. This is not reflected in Fig. 3.13 because a quantitative analysis of the e1hh1 resonance was not performed at large time delays since it seemed too uncertain due to the increased spectral overlap of the broadened emission lines. For the same reason, we did not attempt a quantitative analysis of the e3hh3 emission line at 10N₀. However, the data in Fig. 3.13 clearly demonstrate that the relative contribution of the Lorentzian e1hh1 exciton drops from 75 % to 60 % and below 50 % with increasing carrier density for a fixed time delay. The relative contribution of the e3hh3 Fano continuum stays constant at about 10 % at N₀ and 3N₀. The decrease of the e1hh1 contribution and the constant e3hh3 contribution imply that the

![Fig. 3.13](image)

**Fig. 3.13** Relative contribution of the e1hh1 transition and the e3hh3 transition to the total four-wave-mixing emission versus time delay $\Delta t$. Carrier density $N_n = 3 \times 10^{16}$ cm$^{-2}$ (circles), $3N_n$ (triangles), and $10N_n$ (squares), temperature 7 K.

contributions from the e2hh2 Fano continuum and from transitions with $n_{\text{electron}} \neq n_{\text{hole}}$ are enhanced as the carrier density increases.

This quantitative analysis of the FWM spectra supports the statement that the continuum transitions gain relative strength, as expected, if the continuum transitions can be modeled as an inhomogeneously broadened ensemble of uncoupled two-level systems.
We will now underline the statement that at higher carrier densities the flat continuum in bulk semiconductors behaves similar to the exciton Fano continua in quantum wells. With respect to the data in Fig. 3.13, we note that the contribution from the Lorentzian e1hh1 exciton to the FWM emission is rather large, given the small overlap of the laser spectrum with this resonance. This is at least partially due to the large oscillator strength of the lowest exciton as compared to the exciton Fano continua [47]. This excitonic enhancement of the lowest exciton [72] is already seen in the linear absorption spectrum. The nonlinear exciton-continuum coupling [33, 51, 73] and the longer dephasing time [74] may also contribute to the intense FWM emission at the spectral position of the lowest exciton. The data in Fig. 3.13 indicate that the excitonic enhancement of the lowest exciton is reduced as the carrier density approaches the Mott density. This reflects the reduction of the oscillator strength of the lowest exciton due to the decrease of the binding energy [47]. This mechanism is not equally effective for higher-order exciton Fano resonances, which shows the continuum nature of these transitions. In fact, the same relative decrease of the FWM contribution from the lowest exciton and the increase of the continuum contribution with increasing density have been observed in single-subband experiments on the lowest exciton and a flat continuum [47]. For multi-subband excitation, the time delay dependence of the shape of the FWM spectrum is only observed at the highest density. The relative decrease of the coherent emission from the lowest exciton at positive time delays is reminiscent of the results for single-subband excitation of the lowest exciton and a flat continuum [34, 51, 71, 73]. The comparison between the single- and the multi-subband excitation results indicates that, at high carrier densities, the exciton Fano continua show a similar behavior as a flat continuum.

The similar behavior of the flat continuum in bulk semiconductors and exciton Fano continua in quantum wells indicate that DQI looses importance for exciton Fano continua at higher carrier densities. We have shown that there is no clear discrepancy anymore between the pulse-width
limited dynamics in the time delay domain and the dephasing times obtained from the widths of the FWM spectra. This can be due to the limited time resolution of 100-fs or due to strong dephasing, which takes place on a time scale comparable to the pulse width. However, we have indicated that quantum interference effects become less important and dephasing becomes more important at higher carrier densities.

Summarizing the findings for the high carrier density case, we can state that the distinct structure of the FWM spectra, which is the signature of multi-subband excitation at lower densities, is washed out at high carrier densities. With increasing density, the emission lines in the FWM spectra broaden and resemble more and more the spectra obtained for single-subband excitation of the lowest exciton and a flat continuum. At high carrier densities, the FWM spectra for multi-subband and single-subband excitation show similar features with respect to their time delay dependence and with respect to the density dependence of the contributions from the lowest exciton and the continuum transitions. These findings are reasonable in view of the increased dephasing. Due to this strong dephasing at high carrier densities, quantum interference effects lose their importance and the description of the continuum transitions by an inhomogeneously broadened ensemble of non-interacting two-level systems becomes more applicable.

We will now comment on the transition from DQI to normal dephasing, i.e., the connection between the low carrier density data presented in section 3.3.1 and the high carrier density data presented in this section. The carrier density at which dephasing determines the decay of the TI-FWM signal and, in turn, DQI looses its influence is difficult to define in general. Of course, the transition exists because photon-echo-like behavior in bulk semiconductors was observed in bulk GaAs at higher carrier densities [71]. The photon-echo-like behavior is clear evidence that continuum transitions can be treated as an inhomogeneously broadened
ensemble of non-interacting two-level systems. Consequently, the DQI looses its influence at higher carrier densities. This was also shown for bulk GaAs in Ref. [52].

We have shown that exciton Fano continua show a similar behavior as a flat continuum at higher carrier densities. The DQI effects are also similar in bulk systems and exciton Fano systems at lower carrier densities. Therefore we expect a transition from DQI at lower carrier densities to dephasing dominated behavior at higher carrier densities in exciton Fano systems. Actually, a dephasing dominated behavior was observed in quantum wells at higher carrier densities [49].

In the DQI regime the coupling between the excitonic and the continuum transitions is mediated by excitation induced dephasing (EID) [30, 53]. This many-body Coulomb exciton-continuum coupling is weakened at higher carrier densities due to enhanced screening of the Coulomb interaction. Therefore, at higher carrier densities the description with an inhomogeneously broadened ensemble of non-interacting two-level systems becomes more applicable and the dephasing behavior becomes dominating. These considerations explain why DQI is observed in bulk GaAs and quantum wells at low carrier densities [19, 28, 31, 32, 75] and normal dephasing at high carrier densities [12, 34, 49, 76] (see Chapter 5).

There is no general theory, which can predict the density range of the transition from DQI behavior in a strongly coupled exciton-continuum system to the dephasing dominated behavior in an uncoupled or weakly coupled system. Only a microscopic many-body theory can describe this transition in detail. Such a theory has, to the best of our knowledge, not yet been worked out. The density range of this transition depends, e.g., on the dimensionality of the system, i.e., bulk or quantum well, the width of the quantum well, the sample temperature, and the width of the exciting laser spectrum.

The density range for the transition from the DQI regime to the dephasing dominated regime in this 500 Å wide quantum well sample at a temperature of 7 K cannot specified due to the lack of sufficient time.
resolution. However, we have indicated that a high carrier densities dephasing increases and DQI looses its importance. The experimental observation of the transition from DQI to dephasing in a semiconductor quantum well will be given in the next section. The increased time resolution allows the observation of this transition.

3.4. Dephasing and destructive quantum interference in semiconductor quantum wells

In the preceding sections destructive quantum interference (DQI) was the determining factor of the decay of the TI-FWM signal at low carrier densities if continuum transitions were involved. At high carrier densities the decay of the TI-FWM signal was still fast. We indicated that at high carrier densities DQI should lose its importance. However, it was unclear if the observed fast decay of the TI-FWM signal was due to strong dephasing, which could not be resolved due to the limited time resolution, or due to DQI.

In this section we clearly resolve the transition from DQI to normal dephasing due to the increased time resolution and signal-to-noise ratio (SNR). We show that DQI manifests itself by a sharp decrease of the TI-FWM signal immediately following the laser pulse at lower carrier densities. At larger positive time delays the TI-FWM signal is very weak due to DQI and decays due to dephasing.

We study the coherent FWM emission from a 160 Å wide GaAs/Al$_{0.3}$Ga$_{0.7}$As multiple quantum well at 10 K by means of degenerate FWM as a function of carrier density and time delay. The quantum well structure is the same as already described in Chapter 3.2.

The FWM experiments were performed with 20-fs pulses from a Ti:sapphire laser in the standard two-beam configuration with collinearly polarized excitation pulses with wave vectors $k_1$ and $k_2$. Both pulses had equal intensities and spectra. The excitation pulses are separated in time
by a variable time delay $\Delta t$. The FWM signal emitted in the phase-matching direction $2k_j - k_i$ is detected either spectrally integrated as a function of the time delay $\Delta t$ or spectrally resolved at fixed time delays $\Delta t$. The spectrally resolved FWM measurements as well as linear absorption experiments were performed with a monochromator and a CCD array providing an overall spectral resolution of 0.1 meV.

To allow for transmission experiments, the GaAs substrate of the sample was removed by chemical wet-etching. All experiments were performed at the temperature $T = 10$ K.

The excitation conditions are shown in Fig. 3.14. The laser spectrum is centered at 1.556 eV and has a FWHM of 67 eV (dashed curve). The main features of the linear absorption spectrum (solid line) are, as already has been shown, the discrete exciton resonances $e_1hh_1$, $e_1lh_1$ at the bandedge and the exciton Fano continua $e_1hh_3$ (1.5556 eV), $e_2hh_2$ (1.5813 eV), $e_2lh_2$ (1.6031 eV), and $e_3hh_3$ (1.6686 eV). The lowest exciton transitions $e_1lh_1$ and $e_1lh_1$ have symmetric Lorentzian line shapes with a FWHM of only 1.0 meV, 0.7 meV respectively, demonstrating small inhomogeneous and homogeneous broadening in this high-quality sample at zero carrier density. The $e_1hh_3$ exciton Fano continuum shows an asymmetric absorption profile, characteristic for a Fano resonance (see Chapter 3.2).

The laser pulse excites simultaneously the exciton Fano continua and the discrete exciton resonances at the bandedge, i.e., many subbands are excited simultaneously. The excitation of the discrete resonances, in this case, is stronger than compared to the excitation of the discrete resonances in the 500 Å wide quantum well (see Fig. 3.5).

Figure 3.15 shows the spectrally and temporally integrated FWM signals (TI-FWM) for multi-subband excitation at carrier densities $N_1 = 2 \cdot 10^{10}$ cm$^{-2}$ (dotted curve), $N_2 = 9 \cdot 10^{10}$ cm$^{-2}$ (solid curve), and $N_3 = 2.5 \cdot 10^{11}$ cm$^{-2}$ (dashed curve) in the time delay interval from -80 to +60 fs.
Fig. 3.14 Solid line: Linear absorption spectrum of the 160 Å quantum well at 10 K. Dashed line: Exciting laser pulse spectrum for the degenerate FWM experiment.

The solid gray line is the auto correlation curve of the incident laser pulses. The rising edges of the TI-FWM signals are not pulse-width limited. This could be due to excitonic contributions to the FWM signal at negative time delays because similar results have been observed in FWM experiments exciting both excitonic and continuum transitions in bulk semiconductors [34, 51, 71] (see also Chapter 5). These results have been explained by excitonic polarization scattering [45, 46]. The weak density dependence of the rising edges of the TI-FWM signals is not yet understood.

In contrast, the dotted curve (lowest carrier density $N_i$) shows a pulse-width limited decay at positive time delays in the time delay interval from 20 fs to 60 fs. This could easily be seen when the TI-FWM signal is compared with the auto correlation function of the incident laser pulses. The decay time constant is $\tau = 11$ fs. We recall that a pulse-width limited decay is one prerequisite for DQI.
The increased time resolution allows a determination of the decay time constants, which was not possible in Chapter 3.3. The TI-FWM curves with carrier density \( N_i \) and \( N_i' \) decay with a time constant of 20 fs and 23 fs, respectively, in the time delay interval from 20 fs to 60 fs. The decay time constants increase with increasing carrier density. Thus, the decay of the FWM signal at higher carrier densities is not pulse-width limited and therefore one prerequisite of DQI is not fulfilled. This shows that DQI looses its influence at higher carrier densities. This is in agreement with observations made by Weimer and coworkers [52] and with the observations described in Chapter 3.3 of this thesis. To experimentally prove that DQI is the determining factor of the decay of the TI-FWM signal at the lowest carrier density we have performed spectrally resolved FWM experiments.

Power spectra of the FWM signal are plotted in Fig. 3.16 for different time delays \( \Delta t \) between -19 fs and 128 fs. The carrier density is \( N_i = 2 \cdot 10^{10} \text{cm}^{-2} \) per excitation pulse. This is the lowest carrier density. Unlike the
excitation pulse spectrum, the FWM spectra show a distinct structure which arises from the discrete exciton transitions at the bandedge and from exciton transitions which are embedded in the continuum. The largest emission lines in the FWM spectrum at 1.527 eV originates from the e1hh1 exciton and from the e1lh1 exciton at 1.531 eV. A small contribution to the FWM spectrum at 1.555 eV can be traced back to the e1hh3 exciton Fano continuum. The FWHM of the e1hh1 and e1lh1 resonances are independent of the time delay for the carrier density \( N_i = 2 \cdot 10^{10} \text{cm}^{-2} \).

**Fig. 3.16** Four-wave-mixing power spectrum for multi-subband excitation of the 160 Å quantum well. Time delay from −19 fs to +128 fs. The strong emission at 1.527 eV results from the discrete e1hh1 exciton resonance, the emission at 1.531 eV from the discrete e1lh1 exciton resonance, and the emission at 1.555 eV from the e1hh3 exciton Fano resonance. Carrier density \( N_i = 2 \cdot 10^{10} \text{cm}^{-2} \) (lowest density), temperature 10 K.
The density dependence of the linewidth of the e1hh1 and the e1lh1 resonances in the density range between zero and \( N_i = 2.5 \times 10^{11} \text{ cm}^{-2} \) demonstrates that density-independent inhomogeneous broadening makes only a small contribution to the linewidth for densities of \( N_i = 2 \times 10^{10} \text{ cm}^{-2} \) and larger in this high-quality sample. This is shown in Fig. 3.17. Again, the dashed curve shows the FWM-PS for the highest carrier density. The thick solid curve is the FWM-PS at the moderate density and the dotted curve the FWM-PS at the lowest density. All FWM-PS have been taken at a time delay of \( \Delta t = 40 \text{ fs} \). The solid gray line is the absorption spectrum at zero density. The FWHM of the FWM-PS are always larger than FWHM of the absorption spectrum. This shows that the linewidths for carrier densities of \( N_i = 2 \times 10^{10} \text{ cm}^{-2} \) and higher are dominated by homogeneous broadening, which is enhanced at higher carrier densities due to increased carrier-carrier scattering.

**Fig. 3.17** Spectrally resolved FWM signal at the time delay \( \Delta t = 40 \text{ fs} \) for the highest carrier density \( N_i = 2.5 \times 10^{11} \text{ cm}^{-2} \) (dashed line), moderate density \( N_i = 9 \times 10^{10} \text{ cm}^{-2} \) (solid line), and the lowest density \( N_i = 2 \times 10^{10} \text{ cm}^{-2} \) (dotted line). The gray solid line is the absorption spectrum at zero density. All data were taken at \( T = 10 \text{ K} \).
Since inhomogeneous broadening makes only a small contribution to the total broadening at the lowest carrier densities $N_\text{c} = 2 \cdot 10^{10} \, \text{cm}^{-2}$, we can estimate the dephasing times of the e1hh1 resonance from the FWHM of the e1hh1 emission line in the FWM spectrum. A single dephasing time characterizes each resonance for a given carrier density since the FWHM of the resonances does not show a significant dependence on the time delay. We use the fundamental relation $\tau = T_2 / 2 = \hbar / \Delta E$ where $\tau$ is the decay time, $T_2$ the dephasing time, and $\Delta E$ the FWHM of the emission lines [29]. From this relation and from the FWHM of 2.5 meV for the e1hh1 resonance, we can estimate dephasing times of 520 fs for the e1hh1 exciton at the lowest carrier density $N_\text{c} = 2 \cdot 10^{10} \, \text{cm}^{-2}$. The e1lh1 resonance is not considered because its contribution to the total FWM-PS is small, especially at higher carrier densities. This dephasing time is significantly larger than the dephasing time of 22 fs obtained from the TI-FWM signal in the time delay interval from 20 to 60 fs (see Fig. 3.15) if homogeneous broadening is assumed. Thus, one requirement for DQI, namely a pulse-width limited decay of the FWM signal in the time delay domain if the dephasing time of the resonance is much longer than the pulse width, is fulfilled. The second requirement, namely that the pulse-width limited decay dominates the shape of the FWM signal in the time delay domain and that any slowly decaying contribution is much smaller than expected from the weight of the slowly dephasing resonance in the FWM-PS, is also fulfilled. We will show this now. The weight of the e1hh1 resonance to the total FWM-PS, i.e., the area of the e1hh1 resonance divided by the total area of the FWM-PS, at the lowest carrier density is 77 %. The slowly dephasing contribution to the FWM in the time delay domain at the lowest carrier density is only about 4 %. This can be seen from Fig. 3.18.
Fig. 3.18 Spectrally and temporally integrated FWM signal versus time delay $\Delta t$ for the excitation conditions shown in Fig. 3.14. Dashed line, highest carrier density $N_i = 2.5 \times 10^{11} \text{cm}^{-2}$; solid line, moderate carrier density $N_i = 9 \times 10^{10} \text{cm}^{-2}$; dotted line, lowest carrier density $N_i = 2 \times 10^{10} \text{cm}^{-2}$. Temperature $T = 13$ K. Again, the solid gray line is the auto correlation curve of the laser pulses.

Fig. 3.18 shows the temporally and spectrally integrated FWM signal in a larger time delay interval from $-250$ to $500$ fs. The marking of the curves is the same as in Fig. 3.15 and the auto-correlation function of the laser pulses is plotted as the gray solid curve. Every TI-FWM curve shows pronounced beating modulation. The beating periods match well the energy splittings between the $e_{1hh1}/e_{1hh3}$ and $e_{1hh1}/e_{2hh2}$ resonances seen in the linear absorption spectrum. Clearly, the decay of the TI-FWM signal at the lowest carrier density (dotted curve) shows a pulse-width limited decay at positive time delay around zero time delay. The slowly decaying contribution to this TI-FWM signal at larger positive time delays is small and not stronger than 4% of the maximum TI-FWM signal. This shows that DQI dominates the decay of the TI-FWM signal at the lowest carrier density.

At larger time delays than 140 fs the TI-FWM signal is very weak due to DQI and normal dephasing is the dominant factor of the decay of the TI-
FWM signal (Fig. 3.18). This indicates that DQI suppresses the TI-FWM signal at positive time delays comparable to the pulse width. The dephasing times are $T_2 = 180$ fs for the highest carrier density $N_i = 2.5 \cdot 10^{11}$ cm$^{-2}$, $T_2 = 282$ fs for $N_i = 9 \cdot 10^{10}$ cm$^{-2}$, and $T_2 = 490$ fs for $N_i = 2 \cdot 10^{10}$ cm$^{-2}$. These dephasing times were obtained from fits to the TI-FWM curves for time delays from 140 to 500 fs. We have assumed homogeneous broadening, which is reasonable, as was shown before. The dephasing time constants decrease with increasing carrier density, as expected for normal dephasing. The dephasing times calculated with the relation $\tau = \frac{T_1}{2} = \frac{\hbar}{\Delta E}$ from the FWHM of the FWM-PS at larger time delays ($\Delta t = 80$ fs) are $\tau_1 = 146$ fs (highest density), $\tau_2 = 280$ fs (moderate density), and $\tau_3 = 520$ fs (lowest density).

The agreement between the decay time constants of the TI-FWM signals and the dephasing times calculated from the FWHM of the FWM-PS is very good. Thus, dephasing is the dominating factor of the decay of the TI-FWM signal for time delays larger than 140 fs in the carrier density range from $N = 2 \cdot 10^{10}$ cm$^{-2}$ to $N = 2.5 \cdot 10^{11}$ cm$^{-2}$.

Summarizing this section, we have clearly shown that DQI is important at lower carrier densities and at positive time delays comparable to the pulse width. It loses its influence with increasing carrier density. This could be experimentally proved due to the increased time resolution. The underlying mechanism is the same as described in Chapter 3.3. This is a many-body Coulomb coupling between excitonic and continuum transitions due to EID. At higher carrier densities Coulomb interaction is screened and, in turn, the density dependent EID coupling decreases. This explains why DQI becomes less important at higher carrier densities. The increased signal to noise ratio allows us to observe normal dephasing at larger time delays. At these time delays the FWM signal is very weak due to DQI and the decay time constants of the TI-FWM curves agree with the FWHM of the FWM-PS.
4. Transfer of coherence properties between interband transitions in semiconductor quantum wells

The coupling between excitonic and continuum transitions can be investigated in more detail by means of partially non-degenerate four-wave mixing (PND-FWM) [30]. In PND-FWM, the laser pulse with wave-vector $\mathbf{k}_1$ is filtered by a spectral amplitude filter [44] (see section 2.3.1). This type of FWM was first introduced by Cundiff and coworkers in 1996 [30] to investigate non-degenerate interactions in semiconductors quantum wells and in magneto-exciton systems. In Ref. [77] PND-FWM was used to investigate coherently coupled homogeneous subsystems in an inhomogeneously broadened excitonic ensemble. PND-FWM suppresses the signatures of inhomogeneous broadening and reveals the coherent coupling between different transitions in quantum wells [77]. We show that the PND-FWM technique enables one to transfer coherence properties between interband transitions in quantum wells. With PND-FWM coherent emission from the continuum can be generated at time delays at which the emission is absent if degenerate FWM is used. The experimental results can be understood in a common ground state picture of strongly coupled transitions.

4.1 Partially non-degenerate four-wave-mixing scheme and experimental setup

We use the standard two-beam geometry in the PND-FWM experiments. Both laser pulses are linear and parallel polarized. The wave vectors of the two pulse are denoted, as usual, by $\mathbf{k}_1$ and $\mathbf{k}_2$. The FWM signal, in direction $2\mathbf{k}_1 - \mathbf{k}_2$, is measured either temporally and spectrally integrated versus time delay $\Delta t$ between the excitation laser pulses (TI-FWM) or
spectrally resolved at fixed time delays $\Delta t$ (SR-FWM). A sketch of the experimental setup is shown in Fig. 4.1. The details of the spectral amplitude filter are described in section 2.3.1.

![Fig. 4.1 Sketch of the experimental setup for partially non-degenerate four-wave-mixing (PND-FWM).](image)

A Ti:sapphire laser generates 16-fs pulses with a 82 meV wide spectrum (full width at half maximum, FWHM) centered at 1.55 eV. The laser pulse with wave vector $k_i$ is passed through a spectral amplitude filter, which consists of two SF10 prisms in double-path configuration and an adjustable slit [44]. This set-up allows us to change the center wavelength and the FWHM of pulse spectrum $k_i$. If not mentioned otherwise, the filtered narrow band pulses $k_i$ have a spectral FWHM of about 9 meV and a temporal width of about 200 fs. The pulse with wave vector $k_j$ is not spectrally filtered and remains broadband.

The transfer of coherence properties between interband transitions was investigated in a GaAs/Al$_{0.3}$Ga$_{0.7}$As multiple quantum well sample. It comprises 160 Å wide wells and 150 Å wide barriers, repeated 30 times. As usual, to allow for FWM experiments in transmission geometry, the sample was glued on a sapphire disk and the GaAs substrate was removed by chemical wet etching. The sample is antireflection coated at the front
4.2 Transfer of coherence properties in quantum wells: Experimental results

In this chapter the results of the PND-FWM experiments on the 160 Å wide GaAs/Al$_{0.3}$Ga$_{0.7}$As quantum well are described. The linear absorption spectrum of this quantum well at a temperature of 10 K is shown in Fig. 4.2. The spectrum comprises heavy hole (hh) and light hole (lh) excitonic transitions corresponding to different electron (e) and hole subbands with subband index n. The e(n=1)hh(n=1) exciton transition at 1.528 eV has a FWHM of 1.0 meV, demonstrating small inhomogeneous broadening in this high-quality sample. The other resonances can be assigned to the e1lh1 (at 1.5315 eV), the e1hh3 (at 1.556 eV), the e2hh2 (at 1.581 eV), and the e2lh2 (at 1.603 eV) excitonic transitions.

![Absorption Spectrum](image)

*Fig. 4.2 Linear absorption spectrum of the 160 Å quantum well sample at a temperature T = 10 K.*

The double-peak structure of the e2hh2 transition results from mixing with the e2lh1-(2p) exciton transition [60]. Without mixing the e2lh1-(2p)
The exciton transition is forbidden [60] due to the different parity of the envelope functions [2, 63]. The suffix (2p) denotes a higher excitation level of the exciton. The notation scheme is the same that is usually used for electronic transitions in the hydrogen atom.

We recall that the higher-order excitons, such as e2hh2, are coupled to the energetically degenerate continua of lower-order subband pairs and form exciton Fano continua [13, 18]. The e1hh1 and e1lh1 transitions are discrete excitonic transitions.

We used two different excitation conditions in the PND-FWM experiments with the 160 Å quantum well. They are shown in Fig. 4.3. In the left panel of Fig 4.3 the narrow band laser pulse with wave vector $k_1$ excites the e1hh1/e1lh1 discrete excitonic transitions, whereas the broadband laser pulse with wave vector $k_1$ excites all transitions from the e1hh1 discrete excitonic transition to the e2lh2 exciton Fano continuum.

![Excitation conditions for PND-FWM experiments. In the left panel the e1hh1/e1lh1 discrete excitonic transitions are excited by the narrow band laser pulse. In the right panel the e2hh2 exciton Fano continuum is excited by the narrow band laser pulse. The broad band laser pulse is the same in both cases.](image)

The other excitation condition is shown in the right panel. Here, the narrow band laser pulse with wave vector $k_1$ excites the e2hh2 exciton Fano continuum. The broadband laser pulse with wave vector $k_1$ is not changed throughout all PND-FWM experiments. The broadband pulse $k_1$ produces a pulse energy fluence of 2.1 µJ/cm², whereas the fluences of pulse $k_1$ are 0.18 µJ/cm² and 0.17 µJ/cm² at the e1hh1 and e2hh2
resonances, respectively. This corresponds roughly to a carrier density in the spectral region where both laser pulses overlap of $N = 1.5 \cdot 10^{10}$ cm$^{-2}$. The pulse fluences are not changed throughout the PND-FWM experiment.

The spectrally resolved PND-FWM results at a fixed time delay $\Delta t$ are shown in Fig. 4.4 for narrow band laser excitation of the $e_2hh_2$ exciton Fano continuum (a) and for narrow band laser excitation of the discrete $e_1hh_1/e_1lh_1$ excitonic transitions (b). The data are taken at time delays $\Delta t = 60$ fs (a) and at $\Delta t = 30$ fs (b). These time delays correspond to the maximum of the FWM emission.

In Fig. 4.4(a) the narrow band pulse $k_1$ excites the $e_2hh_2$ exciton Fano continuum. The spectrally resolved FWM signal shows peaks at the $e_1hh_1$
and e1lh1 discrete exciton resonances and at the spectral positions of the e1lh3 and e2lh2 exciton Fano continua. The e1lh1 emission dominates the FWM spectrum although the e1lh1 exciton is not excited by the narrow band laser pulse \( k_1 \). This surprising feature can be understood in the framework of a common ground state picture [30] and will be discussed in detail in Chapter 4.3. We refer to coherent emission as "indirect emission" if only the broad band pulse \( k_2 \) has frequency components at the spectral position of the emission. By the same token, "direct emission" is generated if both pulses \( k_1 \) and \( k_2 \) have frequency components at the spectral position of the emission. This means that the indirect emission is dominating in this case. We observe both direct and indirect emission. This is due to the large oscillator strength of exciton resonances [2] which are inherently involved in a Fano continuum. Strong indirect emission at the lowest exciton resonance was also observed in Ref. [30] if an unstructured continuum without embedded excitons was excited by the narrow band laser pulse. This observation demonstrated strong interaction between non-degenerate interband transitions in the coherent regime. Direct emission from the unstructured continuum was not observed in Ref. [30] due to the reduced oscillator strength of the continuum transitions as compared to the oscillator strength of the exciton Fano continuum.

We observed direct and indirect coherent emission also for excitation of the discrete excitons e1lh1/e1lh1 at the bandedge. Data for this excitation condition are shown in Fig. 4.4(b). Coherent emission from all resonances between the band edge and the e2lh2 exciton Fano continuum is observed. Fig. 4.4(b) demonstrates that non-degenerate interaction between interband transitions can generate indirect coherent emission upshifted in energy with respect to the direct emission. The observation of direct and indirect coherent emission for excitation of discrete excitons and exciton Fano continua allows for a complete analysis of the dynamics. A complete analysis of the dynamics is presented, to the best of our knowledge, for the first time in this work. Direct and indirect coherent emission can also be generated in discrete exciton systems [30, 77]. However, a complete
analysis of the dynamics was not performed [30, 77]. The coherent emission versus time delay is shown in Fig. 4.5, exciting with the narrow band pulse \( k \), at \( e_{2hh2} \) (a) or \( e_{1hh1}/e_{1lh1} \) (b). The FWM emission has been integrated over a 25 meV wide spectral window centered either at the \( e_{1hh1} \) or the \( e_{2hh2} \) resonance. The spectral windows are defined by interference filters whose transmission characteristics ensure that the coherent emission from the different transitions is measured separately.

![Figure 4.5](image)

**Fig. 4.5** (a) Temporally and spectrally integrated four-wave mixing signal versus time delay for narrow band laser pulse \( k \), excitation at the \( e_{2hh2} \) exciton Fano continuum (a) or at the \( e_{1hh1}/e_{1lh1} \) discrete exciton resonances (b). Thick solid lines: coherent emission at the \( e_{1hh1}/e_{1lh1} \) exciton resonances. Dashed lines: coherent emission at the \( e_{2hh2} \) exciton Fano continuum. The thin solid line in (b) is a fit to the dashed curve (see text). The dotted curve in (a) and (b) is the crosscorrelation of the narrow band laser pulse with the broad band laser pulse.
The dashed lines in Figs. 4.5(a) and 4.5(b) show the coherent emission around the e2hh2 exciton Fano continuum. The thick solid lines in Figs. 4.5(a) and 4.5(b) show the coherent emission around the e1hh1/e1lh1 discrete excitons at the bandedge. For excitation at e2hh2, i.e., at the exciton Fano continuum, the FWM traces at both emission positions show a pulse-width limited decay, characteristic for exciton Fano continua at lower carrier densities as was shown in Chapter 3.3 of this thesis. In contrast, for excitation at e1hh1/e1lh1, i.e., at the discrete excitonic transitions at the bandedge, the decay of the FWM signal at positive time delays is well resolved at both detection positions, as shown in Fig. 4.5(b). The dotted curve in both figures (a) and (b) depicts the cross correlation of the narrow band laser pulse k, with the broad band laser pulse k,. The cross correlation is smooth and has no side pulses. This shows that the beating structure seen TI-FWM signal is a true physical effect and is not affected by the pulse shapes. For excitation at the bandedge, we observe a pronounced beating modulation for detection around e1hh1 and around the exciton Fano continuum e2hh2. The beat detected around e1hh1 can be fitted with a beat period of 1.14 ps, which well matches the energy splitting of 3.5 meV between the e1hh1 and e1lh1 discrete exciton resonances seen in linear absorption. Consequently, for excitation and detection around e1hh1, the hh-lh beating of the lowest excitons is observed, characteristic for the coherent dynamics of these discrete exciton transitions [67, 68]. The thin solid line in Fig. 4.5(b) shows a fit to the dashed curve (emission at e2hh2) with the beat period of 1.14 ps, which was obtained from a fit to the solid curve (emission at the band edge e1hh1/e1lh1). All other fit parameters were taken from the fit to the emission curve around the exciton Fano continuum e2hh2. There is an excellent agreement between the beating period of the TI-FWM curve around the exciton Fano continuum e2hh2 and the discrete excitons e1hh1/e1lh1. This shows that the hh-lh beat of the lowest exciton transitions is observed at the spectral position of the e2hh2 exciton Fano.
continuum. Fig. 4.6 highlights this surprising hh-lh beating at the e2hh2 position.

![Graph showing spectrally resolved four-wave mixing signal versus time delay.](image)

**Fig. 4.6** Spectrally resolved four-wave mixing signal versus time delay. Narrow band laser pulse excitation at the e1hh1/e1lh1 discrete excitonic transitions as in Fig 4.5(b). The gray shaded curve shows the spectrally integrated four-wave mixing signal versus time delay. The lh-hh quantum beat is clearly visible.

Moreover, for excitation at e1hh1, the direct and the indirect coherent emission show very similar decay times of 0.32 and 0.40 ps, respectively. Figures 4.5 and 4.6 clearly demonstrate that coherence properties of the discrete excitons can be detected in the coherent emission from exciton Fano continua (Fig. 4.5(b)) and vice versa (Fig. 4.5(a)). Due to this transfer of coherence properties, indirect coherent emission from the exciton Fano continuum can be generated at time delays larger than the pulse width, which is impossible for direct emission [19, 28].

In addition to the transfer of the beating period and the decay time of the TI-FWM signal at positive time delays, the slow rise of the TI-FWM signal at negative time delays is also transferred. Fig. 4.5(b) shows clearly the slow rise of the TI-FWM signal if compared to the cross correlation curve.
In contrast, no time resolved rise can be detected for excitation of the exciton Fano continuum $e_2hh_2$ as can be seen in Fig. 4.5(a), i.e., a pulse width limited rise is observed. The slow rise of the TI-FWM signal at negative time delays was observed for excitation of the lowest excitonic transitions in degenerate FWM experiments [45, 46]. It was explained by polarization scattering which is only effective for homogeneously broadened transitions [45]. For inhomogeneously broadened transitions the effect of polarization scattering vanishes. The slow rise of the TI-FWM signal at negative time delays was not observed for excitation of exciton Fano continua in degenerate FWM experiments [19, 28] (see section 3.3.1). This explains why for excitation of the $e_2hh_2$ exciton Fano continuum with the narrow band laser pulse $k_i$ no slow rise of the TI-FWM signal, neither for direct emission nor for indirect emission, is observed.

Summarizing the experimental observations, we have shown that both the discrete excitons and the exciton Fano continuum $e_2hh_2$ show the lh-hh quantum beat for excitation with the narrow band laser pulse $k_i$ at the discrete excitons $e_{1h1}/e_{1l1}$. Both the discrete excitons and the exciton Fano continuum $e_2hh_2$ show a pulse width limited decay for excitation with the narrow band laser pulse $k_i$ at the exciton Fano continuum $e_2hh_2$. The transfer of coherence properties between spectrally separated transitions in PND-FWM can be understood in the common ground state picture of strongly interacting transitions. This model was put forward to explain the generation of indirect coherent emission in semiconductors [30]. A detailed description of the implications of this model on the interpretation of the PND-FWM experiments will be given in the next chapter.
4.3 Transfer of coherence properties in semiconductor quantum wells: Common ground state model

We briefly recall the generation process of the FWM signal for the totally degenerate FWM technique. Let us assume that inhomogeneous broadening can be neglected. The laser pulse with wave vector $k_1$ arrives at the sample and creates a coherent polarization. The second pulse with wave vector $k_2$ arrives delayed at the sample with a time delay $\Delta t$ with respect to the first pulse and creates a population grating if permitted by the coherence properties of the transitions excited by pulse $k_1$, e.g., if the polarization excited by pulse $k_1$ has not yet dephased when pulse $k_2$ is applied. A part of this second pulse is then self-diffracted off this population grating. This constitutes the FWM signal. One can also think of this part of the second pulse as a third pulse that is applied at the same time [39].

Now, let us return to the partially non-degenerate FWM experiment. In the PND-FWM scheme only in that spectral region where both the narrow band laser pulse $k_1$ and the broad band laser pulse $k_2$ overlap a population grating is generated if permitted by the coherence properties of the transitions excited by the narrow band laser pulse $k_1$. Again, to simplify the discussion let us neglect the inhomogeneous broadening at this time. This population grating is generated in the excited states and in the ground state level [36, 39]. In the common ground state model, all frequency components in the broadband spectrum $k_2$ can couple to the population grating in the ground state. This part of the broadband laser pulse $k_2$ can be thought of as the third pulse in a three pulse FWM experiment.

Therefore, pulse $k_2$ can generate nonlinear polarization in third order in the electric field at transitions involving excited states which have not been excited by pulse $k_1$. This third-order polarization gives rise to coherent emission which is frequency-shifted with respect to the spectrum of $k_1$. This coherent emission constitutes the indirect emission. For inhomogeneously broadened systems, a similar mechanism has been
discussed in the context of accumulated [78] and two-color photon echoes [79, 80].

The intensity of the emitted indirect and direct emission immediately after the second pulse is applied depends on the strength of the generated population grating [36, 39]. This strength is determined by the coherence properties of the transitions excited by pulse \( k_i \), since only in this spectral region a population grating is generated. For example, the population grating will lose its strength quickly if the polarization of these transitions rapidly dephases or its strength will oscillate in time if allowed by the transitions excited by the laser pulse \( k_i \). It is important to note that there is no coherent emission if the strength of the population grating is zero. Therefore, the coherence properties of the transitions excited by pulse \( k_i \) determine the amplitude \( A \) of the direct and indirect coherent emission, independent of the coherence properties of the emitting transitions. The amplitude \( A \) is the intensity of the coherent emission directly after pulse \( k_i \) has been applied. In this way, coherence properties are transferred between spectrally separated transitions. The amplitude of the coherent emission depends on the time delay \( \Delta t \) between the laser pulses \( k_i \) and \( k_j \): \( A = A(\Delta t) \). The decay \( f(t) \) of the coherent emission in real time \( t \) is independent of this amplitude \( A(\Delta t) \) and only depends on the emitting transitions. In TI-FWM experiments the energy of the coherent emission is measured [1, 36], i.e., the coherent emission is temporally integrated due to the use of a slowly integrating detector (see Chapter 2.3). Therefore, the TI-FWM signal can be written as

\[
\text{TI-FWM}(\Delta t) = \int A(\Delta t) f(t) dt = A(\Delta t) \int f(t) dt = A(\Delta t) \cdot C .
\]  

(4.1)

Now it is obvious that the real-time decay \( f(t) \) only determines the height of the TI-FWM signal, whereas its shape is given by the amplitude \( A(\Delta t) \), i.e., the strength of the population grating. That is the reason why the transfer of coherence properties between spectrally separated transitions is observed in the \( \Delta t \)-domain, as demonstrated in Figs. 4.5 and 4.6. This
common ground state model can be extended to any multi-level system with a common ground state and, therefore, PND-FWM can be used in these systems to transfer coherence properties of the excited transitions to the emitting ones.

As already mentioned, in real time $t$, the emitting transitions determine the decay of the indirect coherent emission and, in turn, its spectrum. Therefore, for excitation around the discrete e1hh1/e1lh1 excitons, the beating seen in the time delay domain around the e2hh2 exciton Fano continuum in Fig. 4.5(b) is not reflected by an energy splitting in the spectrum around the e2hh2 exciton Fano continuum in Fig. 4.4(b). For excitation at the e2hh2 exciton Fano continuum, the fast decay of the coherent emission around the e1hh1 discrete exciton, Fig. 4.5(a), is not reflected by its line width, Fig. 4.4(a). The line width has a value of 4.2 meV which corresponds to a decay time of 309 fs (Lorentzian line shape assumed). Such a slow decay is not observed in the TI-FWM measurement, Fig. 4.5(a). The TI-FWM curve decays with a decay time constant of 50 fs. Thus, in both excitation schemes the dynamics in the $\Delta t$-domain is determined by the transitions excited by the laser pulse $k_i$ and not by the emitting transitions.

The pulse width limited decay for excitation with the narrow band laser pulse $k_i$ at the e2hh2 exciton Fano continuum is not due to dephasing but due to destructive quantum interference (DQI). We recall that DQI can be viewed as a destructive quantum interference between exciton and continuum transitions due to a nonlinear many-body Coulomb coupling. The effects of DQI can be seen in the Figs. 4.7 and 4.8. In Fig. 4.7 the excitation conditions for the narrow band laser pulse are shown. The absorption spectrum is also given. The broad band laser pulse is the same as in Fig. 4.1. The different spectral widths of the two narrow band laser pulses are reflected by the different fast decay of the TI-FWM in Fig. 4.8. The TI-FWM signal is integrated over the whole spectral range of the broad band laser pulse because all resonances show the same dynamics.
Fig. 4.7 Dotted line: absorption spectrum of the 160Å wide quantum well at 10 K. Solid and dashed line: different narrow band laser pulse spectra.

Fig. 4.8 Ti-FWM for excitation with the narrow band laser pulse \( \mathbf{k} \) at the \( e^{2}hh2 \) exciton Fano continuum. The excitation laser pulse \( \mathbf{k} \) of the dashed curve excites more continuum transitions than the laser pulse of the solid curve (Fig. 4.7).

The faster decaying curve in Fig. 4.8 belongs to the broader laser spectrum in Fig. 4.7 and vice versa. This reflects the spectral widths of the continuum states contributing to the signal. Thus, the decay of the Ti-FWM signal is not caused by dephasing of the \( e^{2}hh2 \) exciton Fano continuum but rather by DQI.

The discussion in Chapter 3 and the experimental observation of dephasing dominated Ti-FWM [12, 34, 49] shows that DQI looses its influence at higher carrier densities. This will be investigated in the next
chapter where we investigate the dephasing of continuum transitions in bulk semiconductors.

Summarizing the findings of this chapter, we have shown that in the PND-FWM the dynamics in the time delay domain are determined by the properties of the spectral region where both laser pulses overlap. For excitation with the narrow band laser pulse \( k \), at the elhh1/e1lh1 discrete excitonic transitions at the band edge this results in hh-lh quantum beats at the e1hh1/e1lh1 excitons and a the exciton Fano continuum e2hh2. For excitation with the narrow band laser pulse \( k_n \) at the exciton Fano continuum e2hh2 we observe a pulse width limited decay (DQI) at the Fano continuum and at the discrete excitonic transitions at the band edge. These experimental results show that the transfer of coherence properties between spectrally separated interband transitions in a semiconductor quantum well is possible by means of properly designed four-wave-mixing experiments. The results can be understood in a common ground state picture.
5. Dephasing of continuum transitions in bulk semiconductors

In the two preceding chapters destructive quantum interference was the determining factor of the decay of the Ti-FWM signal if continuum transitions were involved. In this chapter we encounter a carrier density regime where DQI looses its influence and dephasing processes determine the decay of the Ti-FWM signal if, again, the main part of the excited transitions are continuum transitions. The transition from DQI dynamics to two-level system dynamics was discussed in detail in Chapter 3.3.

In this chapter we study the coherent emission from bulk semiconductors by spectrally resolved (SR-FWM) and temporally and spectrally integrated four-wave mixing (TI-FWM) with pulses with a full width at half maximum (FWHM) of 16 fs. The carrier density regime ranges from $N = 1 \cdot 10^{16} \text{cm}^{-3}$ to $N = 6 \cdot 10^{17} \text{cm}^{-3}$. In this density regime the decay of the coherent emission in the time delay domain ($\Delta \tau$-domain) is governed by dephasing processes.

Dephasing in bulk semiconductors was investigated with broadband sub-10-fs pulses already in 1988 by Becker and coworkers [12]. In that work the coherent emission was not spectrally resolved and therefore the influence of excitonic transitions and continuum transitions to the overall dephasing could not be estimated. Moreover, the investigated carrier density regime extended from $N > 1 \cdot 10^{17} \text{cm}^{-3}$ to several $10^{18} \text{cm}^{-3}$, i.e. it is at the upper border of the density range investigated in this work.

The dephasing of continuum transitions in semiconductor quantum wells was investigated in [49]. In that work the coherent emission was not spectrally resolved, either.

The lack of spectral resolution rose the question whether the decay of the coherent emission is due to dephasing of continuum or excitonic transitions. This becomes especially important in view of the fact that the excitonic contribution to the FWM signal is much stronger than the continuum contribution [74].
The dephasing times in the bulk material and in the quantum well were attributed to carrier-carrier scattering due to the strongly screened Coulomb interaction [12, 49]. Carrier-phonon scattering processes were not considered [12, 49].

We have overcome some of these limitations because we spectrally resolve the coherent emission and we consider carrier-phonon scattering as an additional scattering process which cannot be neglected at lower carrier densities. This allows us to unambiguously relate the decay of the FWM signal at positive time delays to the dephasing of the continuum transitions, irrespective of the excitation of excitonic transitions. We use a simple relaxation rate approach to model the density dependent decay times of the FWM signal.

5.1 Dephasing of continuum transitions in bulk GaAs and Al_{0.06}Ga_{0.94}As – experimental results

In this chapter we study the decay of the coherent FWM emission from excitonic and continuum transitions in bulk GaAs and Al_{0.06}Ga_{0.94}As semiconductors in dependence of the carrier density and the excess excitation energy with respect to the band edge. In order to measure the decay of the coherent emission we use degenerate FWM in the standard two-beam configuration at room temperature. The pulse length (FWHM) is 16 fs and the spectrum of the laser pulses is centered at 1.54 eV and has a FWHM of 75 meV. The two excitation pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are parallel polarized. The FWM signal, in direction $2\mathbf{k}_2 - \mathbf{k}_1$, is detected either spectrally integrated versus the time delay $\Delta t$ between the excitation laser pulses or spectrally resolved at fixed time delays $\Delta t$. Both investigated samples have a thickness of 200 nm. To allow for transmission experiments, the samples were glued on sapphire disks and the GaAs substrate was removed by chemical wet etching. The samples are
antireflection coated at the front side to reduce the influence of Fabry-Perot oscillations in the transmission experiments.

The use of the GaAs and the Al_{0.06}Ga_{0.94}As sample allows us to vary the excitation conditions even though the laser spectrum is the same in all experiments. This is illustrated in Fig. 5.1. In the GaAs sample, the peak of the excitation laser spectrum lies 120 meV above the band edge. The band edge is not excited and excitonic transitions are not expected to contribute to the FWM signal. In order to investigate the influence of excitonic transitions

![Absorption spectra](image)

**Fig. 5.1** Linear absorption spectra (shaded area) of the Al_{0.06}Ga_{0.94}As sample (left panel) and the GaAs sample (right panel) at room temperature. The excitation laser spectrum (solid line) is the same for both samples.

on the FWM signal, the Al_{0.06}Ga_{0.94}As sample is used whose band edge overlaps with the low-energy flank of the excitation laser spectrum. The peak of the excitation laser spectrum lies 40 meV above the band edge of this sample. It is important to note that the excess excitation energy with respect to the band edge is increased by 80 meV in the GaAs sample as compared to the Al_{0.06}Ga_{0.94}As sample.

The crucial experimental point of continuum dephasing measurements is a sufficient time resolution. Therefore, we will first comment on the time resolution that can be achieved with sech^2 laser pulses with a FWHM of 16 fs. A sech^2 pulse shape is usually assumed in Kerr-lens modelocked lasers [81]. A 16-fs sech^2 laser pulse has a falling and rising edge of 4.7 fs. If such a pulse is convoluted with an exponential decay with a time constant of 10 fs the convolution decays with a decay constant of 10.4 fs.
Thus, 16-fs pulses have a sufficient time resolution to measure decay constants of only 10 fs.

Figure 5.2 shows the spectrally integrated FWM signal from the GaAs sample versus time delay $\Delta t$ for different carrier densities from $2 \cdot 10^{16}$ cm$^{-3}$ to $6 \cdot 10^{17}$ cm$^{-3}$. A carrier density dependent decay is observed for positive time delays which becomes slower for decreasing carrier density. This proves that at least the decays of the FWM signals for carrier densities lower than the maximum density are clearly resolved and reflect the dephasing of the coherent emission. The dashed lines represent fits to an exponential function $\propto \exp(-\Delta t/\tau)$ with decay times $\tau$ which vary from 15.4 fs to 10.4 fs for carrier densities between $2 \cdot 10^{16}$ cm$^{-3}$ and $6 \cdot 10^{17}$ cm$^{-3}$. The fastest decay time of 10.4 fs is very close to the time resolution of the experiment. The rise of the FWM signal is determined by the pulse width for all carrier densities.

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**Fig. 5.2** Spectrally integrated four-wave mixing signals (solid lines) from the GaAs sample versus time delay at room temperature. Carrier densities $N = 6 \cdot 10^{17}$ cm$^{-3}$ (steepest slope), $N = 7 \cdot 10^{16}$ cm$^{-3}$, and $N = 2 \cdot 10^{16}$ cm$^{-3}$. The dashed lines are exponential fits. Inset: Linear absorption spectra at room temperature (shaded) and excitation pulse spectrum (solid).
In contrast, the FWM signals from the Al\textsubscript{0.06}Ga\textsubscript{0.94}As sample, plotted in Fig. 5.3 for the same carrier densities as in Fig. 5.2, show a density dependent rise at negative time delays. The comparison between the results from the GaAs and the Al\textsubscript{0.06}Ga\textsubscript{0.94}As sample suggests that the density dependent rise of the FWM signal from the Al\textsubscript{0.06}Ga\textsubscript{0.94}As sample is due to the excitonic transitions. The rise becomes slower with decreasing carrier density. The same density dependence is found for the decays at positive time delays which can be fitted to exponential functions $\propto \exp(-\Delta t/\tau)$ with time constants $\tau$ varying from 15.3 fs to 11.0 fs for carrier densities from $2 \cdot 10^{16}$ cm\(^{-3}\) to $6 \cdot 10^{17}$ cm\(^{-3}\) (dashed lines in Fig. 5.3).

![Figure 5.3](image-url)

**Fig. 5.3** Spectrally integrated four-wave mixing signals (solid lines) from the Al\textsubscript{0.06}Ga\textsubscript{0.94}As sample versus time delay at room temperature. Carrier densities $N = 6 \cdot 10^{16}$ cm\(^{-3}\) (steepest slope), $N = 7 \cdot 10^{16}$ cm\(^{-3}\), and $N = 2 \cdot 10^{16}$ cm\(^{-3}\). The dashed lines are exponential fits. Inset: Linear absorption spectra at room temperature (shaded) and excitation pulse spectrum (solid).

Spectrally resolved FWM was performed to analyze the contributions to the coherent emission. Figure 5.4 shows the results for time delays from -42 fs to +54 fs for the simultaneous excitation of continuum and excitonic transitions in the Al\textsubscript{0.06}Ga\textsubscript{0.94}As sample. The carrier density is $N = 2 \cdot 10^{16}$ cm\(^{-3}\), corresponding to the curve with the slowest decay in Fig. 5.2. An excitonic
contribution to the FWM signal is clearly visible at the band edge and dominates the FWM power spectrum for time delays from -32 fs to +11 fs. For time delays $\Delta t > 11$ fs the FWM power spectrum broadens and the excitonic contribution is no longer dominant. The excitonic contribution vanishes for time delays $\Delta t > 22$ fs. The data in Fig. 5.2 clearly show that the excitonic transitions give rise to the FWM signal at negative time delays, but yield only a negligible contribution at positive time delays. Similar results have been obtained in FWM experiments with 100-fs pulses exciting both excitonic and continuum transitions [51, 71]. These results have been explained by excitonic polarization scattering [45, 46], which, at negative time delays, gives rise to an enhancement of the homogeneously broadened excitonic contribution to the FWM signal relative to the inhomogeneously broadened continuum contribution since it is only effective for homogeneous transitions [48, 51, 71]. At positive time delays, phase space filling results in FWM emission from both the continuum and the excitonic transitions, leading to a relative decrease of the excitonic contribution [48, 51, 71].

![Spectrally resolved four-wave mixing signal from the Al$_x$Ga$_{1-x}$As sample for different time delays at room temperature, carrier density $N = 2 \cdot 10^{19}$ cm$^{-3}$.](image_url)
With increasing carrier density, the excitonic contribution to the coherent emission from the $\text{Al}_{x}$-$\text{Ga}_{1-x}$-$\text{As}$ sample decreases compared to the continuum contribution (Fig. 5.5), as observed previously [47, 48]. This was also observed in [19] and in Chapter 3 of this work, where the discrete excitonic transitions at the bandedge loose strength compared to the continuum transitions. The diminished influence of the discrete excitonic transitions suggests that the continuum transitions of the investigated bulk semiconductors can be treated as an inhomogeneously broadened ensemble of non-interacting two-level systems. Moreover, the not pulse width limited TI-FWM signal in the Fig. 5.2 and 5.3 at positive time delays indicates that two-level system dynamics is applicable.

As expected, there is no excitonic contribution in the coherent FWM emission from the GaAs sample, as shown in Fig. 5.6 at the carrier density $N = 4.2 \cdot 10^{10} \text{cm}^{-2}$. The FWM power spectra from the GaAs sample hardly change their shape with varying time delay and carrier density.
We conclude that the decay of the spectrally integrated FWM signal for positive time delays unambiguously reflects the dephasing of the continuum transitions in the whole density range from $1 \cdot 10^{16} \text{cm}^{-3}$ to $6 \cdot 10^{17} \text{cm}^{-3}$, irrespective of the excitation of excitonic transitions. This confirms the interpretation of the results in [12].

Fig. 5.6 Spectrally resolved four-wave mixing signal from the GaAs sample for different time delays at room temperature, carrier density $N = 4 \cdot 10^{9} \text{cm}^{-3}$.

We will now discuss the dephasing of the continuum in more detail. Figure 5.7 shows the decay times $\tau$ of the coherent emission as a function of the carrier density for the GaAs (filled circles) and the Al$_{0.1}$Ga$_{0.9}$As (open squares) sample. These decay times have been extracted from exponential fits at positive time delays to a large set of FWM traces, and unambiguously reflect the dephasing of the continuum. The data for the highest density are not included in Fig. 5.7 since they are close to the time resolution of the experiment.
5.2 Dephasing of continuum transitions in bulk GaAs and Al\textsubscript{0.06}Ga\textsubscript{0.94}As – discussion

The TI-FWM data (Fig. 5.2 and 5.3) clearly show a density dependent decay for positive time delays and the FWM-PS which go with them show unambiguously that this decay is due to continuum transitions. The density dependent decay of the TI-FWM signal proves that dephasing is dominating in the investigated density regime. Destructive quantum interference (DQI) is not observed. This also means that the exciton-continuum coupling due to EID can be neglected. Since this coupling mechanism can also be described with a multi-level system with a common ground state [30], this description cannot be correct and the continuum
transitions have to be regarded as an inhomogeneously broadened ensemble of weakly or non-interacting two-level systems. In such a system dephasing is dominant.

As possible dephasing mechanisms, Coulomb scattering and LO phonon interaction should be considered. Scattering with acoustic phonons is slower than LO phonon scattering [74] and, therefore, is not expected to be important in FWM experiments on the 10-fs time scale. Other scattering processes like intervalley scattering or alloy scattering can also be ruled out. Intervalley scattering can be neglected because even the energetically highest electrons excited by our laser pulses are too low in energy for resonant intervalley scattering [83] to happen in the GaAs and the $A_{1x}Ga_{1-x}As$ sample. Consequently, intervalley scattering does not have to be considered in our experiments. Alloy scattering in $A_{1x}Ga_{1-x}As$ is known to make contributions of less than 1 meV to the homogeneous broadening [84]. Therefore, it is negligible on the 10-fs time scale.

Figure 5.7 shows that the density dependence of the decay times is very weak for both samples. Decreasing the density by one order of magnitude from $2 \times 10^{17}$ cm$^{-3}$ to $2 \times 10^{16}$ cm$^{-3}$ results in an increase of the decay times of less than a factor 1.4. We will now compare these results to previous work on bulk GaAs, performed in the high-density regime from $2 \times 10^{17}$ cm$^{-3}$ to $7 \times 10^{18}$ cm$^{-3}$, and reported in Ref. [12]. First, we note that our measurement at $N = 2 \times 10^{17}$ cm$^{-3}$ reproduces the result obtained in Ref. [12] at this density. This confirms that (i) the time resolution in our experiments is sufficient to determine the decay time at densities $N \leq 2 \times 10^{17}$ cm$^{-3}$, and (ii) that the continuum dephasing decay times in [12] are not affected by the excitation of the band edge, as expected from our spectral analysis in Figs. 5.4 and 5.5. The measurements in the high-density regime have shown that the decay time is given by $\tau_{\text{HD}}(\text{fs}) = 6.8\left[N(10^{18}\text{cm}^{-3})\right]^{-0.3}$, for densities $N$ from $2 \times 10^{17}$ cm$^{-3}$ to $7 \times 10^{18}$ cm$^{-3}$ [12]. This relation has been explained accounting for carrier-carrier scattering due to strongly screened Coulomb interaction with an effective screening length given by the average nearest-neighbor
distance between carriers [12, 49]. This model predicts $\tau_{\text{hd}} \propto N^{-1/3}$, in agreement with the high-density data in [12]. Carrier LO-phonon interaction was neglected in the high-density regime [12]. The solid line in Fig. 5.7 shows the relation $\tau_{\text{hd}}(\text{fs}) = 6.8[N(10^{13}\text{cm}^{-3})]^{-1/3}$ extrapolated to lower densities. Our data in Fig. 5.7 demonstrate that significantly shorter decay times of the continuum dephasing are obtained below $10^{17}\text{cm}^{-3}$ than predicted by the extrapolation of the high-density results. This is equivalent to a weaker density dependence of the decay below $10^{17}\text{cm}^{-3}$, i.e., to a weaker increase of the decay times for decreasing density. These results indicate that, at carrier densities below $10^{17}\text{cm}^{-3}$, another process contributes to the dephasing besides Coulomb scattering. This process should have a weak density dependence, such as LO phonon scattering [85] [86].

Electron-LO-phonon scattering times of 165 fs and 132 fs have been experimentally determined in [82] and [87], respectively. Theoretically, an electron-LO-phonon scattering time of 220 fs was predicted [88]. In the following, we will use an electron-LO-phonon scattering time $\tau_{\text{lo}}$ of 165 fs [82] independent of the density, which is justified for $N \leq 10^{17}\text{cm}^{-3}$ [85] [86]. Adding the electron-LO-phonon scattering rate $1/\tau_{\text{lo}}$ to the high-density FWM decay rate $1/\tau_{\text{hd}}$ [12], which neglects electron-LO-phonon scattering, we obtain for the FWM decay time at lower densities $\tau = (1/\tau_{\text{hd}} + 2/\tau_{\text{lo}})^{-1}$. Electron-LO-phonon scattering is accounted for as an energy relaxation process [39] and the semiconductor continuum is treated as an inhomogeneously broadened system [89] to derive this relation. $\tau$ is plotted as solid line in Fig. 5.7 and reasonable agreement with the experimental results is found.

To the best of our knowledge, the data of Fig. 5.7 represented the first experimental proof that the strong-screening model for carrier-carrier scattering of Ref. [12] together with the electron-LO-phonon scattering time
can approximately describe the dephasing time of the semiconductor continuum at lower densities.

This is somewhat surprising because a quantum kinetics description of the Coulomb scattering [76, 90-92] seems more appropriate during the built-up of screening on the 10-fs time scale than the strong-screening model [12]. Moreover, on these time scale, a quantum kinetics description of electron-LO-phonon scattering is required [93, 94]. A recently developed quantum kinetics theory accounts for both Coulomb and LO phonon scattering [76]. It describes the loss of coherence of semiconductor continuum transitions correctly, even at lower carrier densities [76], i.e., our data are in agreement with the calculated decay times of the FWM signal. In this approach the Coulomb potential is modeled by a time-dependent screened Coulomb interaction [95]. This shows that both LO phonon scattering and a screened Coulomb interaction have to be considered to describe the dephasing of semiconductor continuum transitions.

An older quantum kinetics theory predicts a decay of the spectrally integrated FWM signal \( \propto \exp\left(-\frac{A}{\Theta}\right) \) with \( \Theta \propto N^{-\eta} \) [90, 91], also obtained from a quasi-classical theory [96]. In these quantum kinetics theories a bare Coulomb potential is assumed because the time for the built up of screening is in the order of the inverse plasmon frequency, which is much longer than the observed dephasing times [90, 91]. LO phonon scattering is neglected. However, such a simplified quantum kinetics theory does not describe our data, especially at lower carrier densities. This again indicates that, for densities below 10⁻⁷ cm⁻³, both LO phonon and Coulomb interaction have to be considered to describe the dephasing of semiconductor continuum transitions.

We will now comment on the energy dependence of the continuum dephasing. Figure 5.7 shows that almost identical decay times of the continuum dephasing are obtained in the GaAs and the Al₀.₆₆Ga₀.₃₄As sample for all densities. Recalling that the excess energy varies by 80 meV between the samples, this demonstrates that the dephasing of the
continuum is almost independent of the excess energy for densities below $10^{17}\text{cm}^{-3}$. This is plausible given that Coulomb scattering in the quantum kinetics regime is only weakly wave vector and energy dependent [52, 90]. We note that the negligible energy dependence justifies the use of a single time constant to describe the decay of the coherent emission from a broadband continuum.

In conclusion, we have presented an experimental four-wave-mixing study with 16-fs pulses which has allowed us to unambiguously extract information about the decay of the coherent emission from continuum transitions in bulk semiconductors for densities below $10^{17}\text{cm}^{-3}$, i.e., in a density regime not accessed before. In this density regime destructive quantum interference is not effective. The FWM-PS show that the decay of TI-FWM signal at positive time delays is due to dephasing of continuum transitions. Our results demonstrate a weak density dependence and negligible energy dependence of the continuum dephasing in this density regime. We showed that both Coulomb and LO phonon scattering need to be considered to describe the dephasing of continuum transitions in bulk semiconductors. A simple relaxation rate approach approximately models the experimental results.
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6. Summary

In this thesis, coherent dynamics of exciton and continuum transitions in bulk semiconductors and semiconductor quantum wells has been investigated by means of four-wave-mixing spectroscopy with ultrabroadband pulses. It has been shown that exciton continuum interaction strongly affects the coherent emission.

In semiconductor quantum wells, excitons of higher-order subbands form exciton Fano continua. The coupling between the exciton and the continuum transitions is mediated by the bare Coulomb potential. The formation of exciton Fano continua has been experimentally verified in quantum wells with well widths of 160 Å and 500 Å by means of linear absorption measurements. The absorption profiles can be described by the atomic physics Fano model.

The nonlinear optics of exciton Fano continua in the coherent regime have been investigated by means of four-wave mixing with 100-fs and 20-fs laser pulses. This is the first study of coherent dynamics of exciton Fano continua in quantum wells without magnetic field. The dephasing of exciton Fano continua has been studied by spectrally resolved four-wave mixing. The dephasing times can be described by the atomic physics Fano model.

However, the decay of the spectrally and temporally integrated four-wave mixing emission from exciton Fano continua at lower carrier densities in the time-delay domain is not due to dephasing but rather to destructive quantum interference between exciton and continuum transitions. This effect results from the many-body Coulomb coupling between exciton and continuum transitions. The destructive quantum interference effect can be described by the semiconductor Bloch equations including density dependent dephasing terms. The introduction of density dependent dephasing terms is also known as excitation induced dephasing.
We have shown that, with partially non-degenerate four-wave mixing, coherent properties of one set of interband transitions can be detected in the coherent emission from other, spectrally separated, transitions. This leads to a fast decay of the four-wave mixing emission from discrete exciton transitions in the time-delay domain if coherence properties of continuum transitions are transferred to discrete exciton transitions. The other way around, the four-wave mixing emission from continuum transitions in the time-delay domain shows an oscillating behavior if coherence properties of discrete exciton transitions are transferred to continuum transitions. This means that coherent emission from the continuum can be generated which is absent otherwise. These results can be understood in a common ground state picture of strongly interacting transitions. The underlying concept is applicable to other multi-level systems with a common ground state.

We have shown that the destructive quantum interference is less important at higher carrier densities because the many-body Coulomb coupling between exciton and continuum transitions is weakened. Normal dephasing dominates the decay of the four-wave-mixing emission in the time delay domain at higher carrier densities. We have studied the dephasing of continuum transitions in bulk GaAs and Al$_x$Ga$_{1-x}$As with 16-fs pulses. The dephasing can be described by a relaxation rate approach accounting for electron-electron and electron LO-phonon scattering in a first approximation.
References


[80] In Ahn et al., *Phys. Rev. Lett.* vol. 82, pp. 3879-3882 (1999) frequency shifted FWM emission was also observed but the generation process is different since a time
resolved decay of the TI-FWM was not observed although discrete excitonic transitions at the bandedge were excited.


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