Quantum interference in the system of Lorentzian and Fano magnetoexciton resonances in GaAs

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Using femtosecond four-wave mixing (FWM), we study the coherent dynamics of Lorentzian and Fano magnetoexciton resonances in GaAs. For unperturbed Lorentzian magnetoexcitons, we find that the time-integrated FWM signal decays due to dephasing processes as expected for Lorentzian resonances. The time-integrated FWM signal from a single Fano magnetoexciton resonance, however, decays quasi-instantaneously although the dephasing time of the Fano resonance is much longer than the time resolution of the experiment. This fast decay is the manifestation of destructive quantum interference. Although destructive quantum interference in our system is closely related to the dynamics of Fano resonances, for the simultaneous excitation of Lorentzian and Fano magnetoexciton resonances destructive quantum interference also strongly affects the dynamics of Lorentzian magnetoexcitons due to quantum-mechanical coupling between the two types of resonances.

I. INTRODUCTION

In semiconductor physics, four-wave-mixing (FWM) experiments have been extensively used to study the dynamics of electronic states. This time-domain optical technique employs the coherent nonlinear polarization generated by two short time-delayed laser pulses to study interactions between optical excitations and their environment. Due to these scattering processes, the polarization dephases. Since the nonlinear polarization is the source of the FWM signal pulse, the FWM signal contains information about the dephasing and scattering.

The analysis of a two-level system with homogeneous Lorentzian broadening demonstrates that the temporally integrated intensity of the FWM signal pulse (TI FWM signal) is proportional to the coherent polarization present in the sample at the time the second laser pulse is applied.[1] Thus, in this situation the decay of the TI FWM signal as a function of the time delay between the excitation pulses is a direct measure of the dephasing of the polarization and the underlying scattering processes.

The FWM signal from a system consisting of several discrete levels with Lorentzian broadening shows a modulated decay due to interference between different frequency components. The modulation gives further information on the energy spacing and coupling between levels,[2,3] whereas the decay of the TI FWM signal is still determined by dephasing processes.

To describe FWM experiments in semiconductors, the theoretical description has to be refined. The nonlinear optical response of a semiconductor is strongly affected by Coulomb correlation, which can be accounted for in the framework of the semiconductor Bloch equations.[4] The effect of Coulomb correlation on the FWM signal is very clearly seen if at a fixed time delay the shape of the FWM signal pulse is traced by upconversion techniques (time-resolved FWM: TR FWM). Coulomb correlation results in a delayed rise of the coherent emission from a homogeneously broadened transition in a semiconductor,[5,6] unlike the free polarization decay of a homogeneous two-level system. Coulomb effects also modify the shape of the TI FWM signal[7,8] but do not change the relation between the TI FWM decay and the dephasing rate as long as a Lorentzian resonance is considered.

In a recent paper,[9] we have studied the coherent dynamics of Fano resonances in semiconductors. Fano interference is the result of quantum-mechanical coupling Γ between a discrete state and an energetically degenerate continuum of states.[10] If both the discrete state and the continuum can be optically excited, Fano interference manifests itself in absorption by an asymmetric line shape with a pronounced minimum at the energy where the optical transition amplitudes of the discrete state and the continuum are out of phase. Fano resonances are observed in a bulk semiconductor under magnetic field, where Coulomb interaction couples higher-order magnetoexcitons to continua corresponding to lower-order Landau levels.[11]

For excitation of a single Fano resonance in bulk GaAs under magnetic field, we have reported a quasi-instantaneous decay of the time-integrated FWM signal under experimental conditions, where the dephasing time is much longer than the experimental time resolution.[9] Thus the decay of the TI FWM signal does not reflect dephasing, unlike the case of a Lorentzian resonance in a semiconductor. The fast decay of the TI FWM signal is the manifestation of destructive quantum interference.

Other types of quantum interference involving Lorentzian resonances in semiconductors have been reported in the literature, e.g., the well-known quantum beating between two Lorentzian exciton resonances,[12] which manifests itself by a periodic modulation of the FWM signal in the time domain but does not affect

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the decay. More complicated excitation schemes involve Lorentzian excitonic and free-carrier states at zero magnetic field, where quantum interference effects beyond the two-resonance quantum beating have been observed.[13–15]

In this paper, we present a comprehensive experimental study of the destructive quantum interference observed in FWM experiments on Fano resonances in GaAs under magnetic field. The linear optical absorption of the GaAs sample we have studied and the technical details of the FWM experiments are discussed in Sec. II of the paper. In Sec. III, we summarize the experimental results obtained exciting a single Fano resonance. In particular, we point out the relation between the TR FWM, the TI FWM, and the spectrum of the FWM signal and define the term "destructive quantum interference." We will show that destructive quantum interference cannot be understood in an atomic model, i.e., neglecting Coulomb correlation in a semiconductor. In order to further characterize the conditions under which destructive quantum interference occurs, we examine the transition from Lorentzian magnetoexciton dynamics to the dynamics of magnetoexcitons with a Fano absorption profile in Sec. IV. Experimentally, this transition can be realized varying the excitation energy and thereby the relative weight with which Lorentzian and Fano magnetoexciton resonances contribute to the FWM signal. While the dynamics of unperturbed Lorentzian magnetoexcitons is regular, we find that destructive quantum interference dominates the TI FWM response if Lorentzian and Fano magnetoexcitons are excited simultaneously. These experiments inherently involve the observation of quantum beating between magnetoexcitons corresponding to different Landau levels. Finally, we summarize our results in Sec. V.

II. EXPERIMENT

All experiments have been performed on a high-quality GaAs bulk sample, in which the inhomogeneous broadening of the optical transitions can be neglected. The 1.0-μm layer of GaAs was grown on a GaAs substrate by molecular beam epitaxy. To allow for transmission experiments, the substrate was removed by chemical etching and the GaAs layer was glued on a sapphire disk.

Figure 1 shows the low-temperature (1.6 K) linear absorption of this sample under circular (σ^-) excitation for different magnetic fields. The splitting between the heavy hole (hh) and the light hole (lh) exciton observed at zero field is due to mechanical strain at low temperatures, which shifts the lh valence band below the hh valence.[16]

A magnetic field applied to a semiconductor leads to the formation of magnetoexcitons corresponding to each pair of valence and conduction band Landau levels with the same Landau quantum number n.[17] In a bulk semiconductor, however, the quantization of the electronic states is not complete. The field does not quantize states with wave vector parallel to the direction of the field. These states form one-dimensional continua corresponding to each Landau level. Thus, discrete magnetoexcitons corresponding to the higher-order Landau level pairs are degenerate to continuum states of lower Landau levels. In Ref. 11 we have shown that magnetoexcitons and continuum states are coupled, giving rise to Fano resonances, and that the Fano coupling Γ is due to Coulomb interaction.

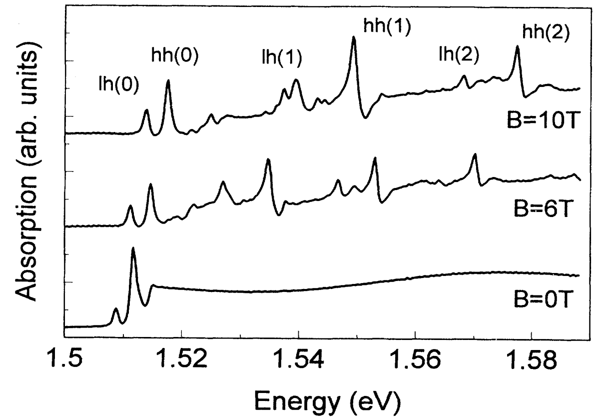

FIG. 1. Low-temperature (1.6 K) linear optical absorption of GaAs for different magnetic fields B under circularly polarized excitation (σ^-). The origin of the different resonances is explained in the text.

Pronounced Fano resonances are observed in the 6 T and 10 T spectrum in Fig. 1. In this respect, the important feature is the absorption minimum at the high-energy side of each resonance in the continuum part of the spectra. This minimum is below the continuum absorption demonstrating coupling and interference between magnetoexcitons and continua. In Ref. 11, we have assigned lh and hh magnetoexcitons to the different resonances: The lowest-order magnetoexcitons lh(0) and hh(0) at the band edge correspond to hh and lh Landau level pairs with Landau quantum number $n = 0$. The hh(0) and lh(0) magnetoexciton transitions are Lorentzian lines since these magnetoexcitons are not in resonance with continuum states. In contrast, the lh($n \geq 1$) and hh($n \geq 1$) magnetoexcitons form Fano resonances. Summarizing these results, the absorption spectrum at finite magnetic field consists of two series of resonances corresponding to lh and hh magnetoexciton transitions. As expected, the hh transitions have the larger oscillator strength. In both series, the lowest-order resonances are Lorentzian, whereas all higher-order resonances have Fano absorption profiles.

Four-wave-mixing experiments have been performed in the standard self-diffraction configuration with cocircularly polarized (σ^-) 100-fsec pulses from a self-mode-locked Ti:sapphire laser. Two pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ and intensities $I_{\mathbf{k}_1} = 2I_{\mathbf{k}_2}$ generate the FWM signal pulse emitted in the direction $2\mathbf{k}_2 - \mathbf{k}_1$. The excitation pulses are separated in time by a time delay Δt, where $\Delta t > 0$ refers to pulse 1 arriving before pulse 2. Importantly, we have analyzed the FWM signal pulse in three different ways. We measure the TI FWM signal, i.e., the temporally and spectrally integrated FWM signal pulse, as a function of the time delay Δt. At fixed time delays, upconversion of the FWM signal pulse with a reference pulse yields the TR FWM signal. Spectral information on the FWM signal is obtained measuring the power spectrum at fixed time delays Δt. In the case of a homogeneously broadened Lorentzian two-level system, the TI and the TR FWM signals decay with a time constant $T_2/2$, where T_2 is the dephasing time, which leads to the homogeneous broadening $\gamma = 2\hbar/T_2$ of the FWM spectrum. In turn, T_2 can also be determined from the FWM spectrum. All experimental data presented in Secs. III and IV have been taken at a temperature of 1.6 K.

III. SINGLE FANO RESONANCE

We now focus on the coherent response of a single Fano resonance in GaAs. The experiments discussed in this section have been performed at a magnetic field $B = 10\,\mathrm{T}$. At this relatively large field, due to the large energy splitting between adjacent resonances, we can avoid the excitation of resonances corresponding to Landau level pairs with different Landau quantum numbers n. With the excitation laser spectrum centered on the hh(1) Fano resonance, as shown in Fig. 2, we mainly excite this resonance and only very weakly the lh(1) Fano resonance.

The TI FWM signal from the hh(1) Fano resonance is depicted in the inset of Fig. 2 (carrier density $N \approx 1 \times 10^{16}$ cm^{-3}). This signal cannot be resolved with 100-fsec pulses, i.e., it decays quasi-instantaneously over almost three orders of magnitude. The TI FWM signal does not exhibit any modulation, but the shape of the signal only reflects the laser pulse.

Using a Lorentzian model, from the instantaneous decay of the TI FWM, one would expect the dephasing time to be much shorter than 100 fs, which is the time resolution of the experiment. In this case, also the TR FWM signal should decay quasi-instantaneously and the FWM spectrum should be as broad as the laser spectrum. Surprisingly, Fig. 3 shows that the TR FWM signals decays almost exponentially with a much longer time constant of about 350 fs, nearly independent of the time delay Δt. The data in Fig. 3 have been obtained at the same carrier density as the TI FWM signal. The FWM spectrum for $\Delta t = 0$ in the inset of the figure is dominated by the contribution from the hh(1) Fano resonance at 1.551 eV. A small contribution from the lh(1) Fano resonance at 1.541 eV is also visible. The energy splitting between the lh and the hh Fano resonance well agrees with the period of the slight beating modulation observed in the TR FWM experiment. The width δE of the dominating hh emission line is 2 meV, i.e., the FWM power spectrum is much narrower than the laser spectrum.

It should be noted that the FWM power spectrum $|S(\omega, \Delta t)|^2$ and the TR FWM signal $|S(t, \Delta t)|^2$ are related: The power spectrum $|S(\omega, \Delta t)|^2$ is the Fourier transform of the autocorrelation of $S(t, \Delta t)$. This relation implies that a narrow power spectrum corresponds to a slowly decaying autocorrelation of $S(t, \Delta t)$. If the autocorrelation of $S(t, \Delta t)$ decays slowly, $S(t, \Delta t)$ itself and also the TR FWM signal $|S(t, \Delta t)|^2$ are slowly decaying functions. To find the quantitative relation between the width δE of the FWM power spectrum and the decay time τ of the TR FWM signal, we have used the fundamental relation $\tau = \hbar/\delta E$ from which a decay time $\tau = 330$ fs is obtained for the observed width $\delta E = 2\,\mathrm{meV}$. The decay time of the TR FWM signal at $\Delta t = 0$ is 375 fs, in reasonable agreement with this estimate. Thus, the long decay time of the TR FWM signal and the narrow width of the FWM spectrum correspond

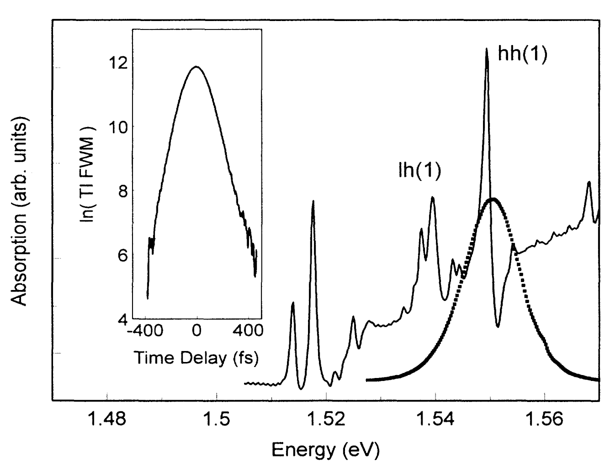

FIG. 2. Absorption spectrum of GaAs at $B = 10\,\mathrm{T}$ and laser spectrum indicating the excitation conditions in the four-wave-mixing (FWM) experiment. Inset: Time-integrated FWM signal (carrier density $N \approx 1 \times 10^{16}\,\mathrm{cm}^{-3}$).

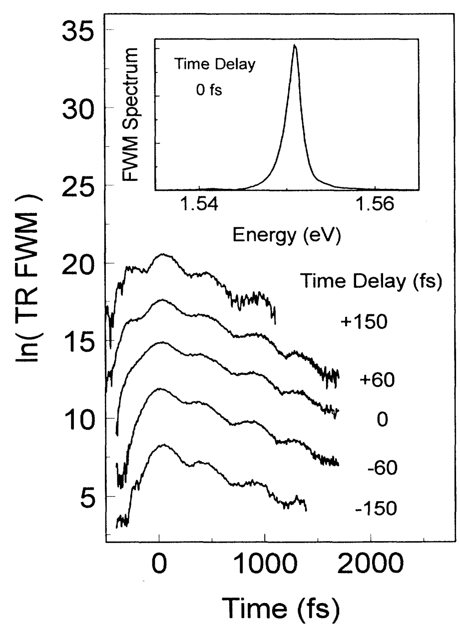

FIG. 3. Time-resolved FWM signal from a single Fano magnetoexciton resonance for time delays Δt between -150 fs and $+150$ fs. Inset: Spectrum of the FWM signal at $\Delta t = 0$ fs. Carrier density $N \approx 1 \times 10^{16}\,\mathrm{cm}^{-3}$, $B = 10\,\mathrm{T}$.

to each other even quantitatively, whereas the decay of the TI FWM signal takes place on a completely different, i.e., much faster, time scale. In Ref. 9, we have discussed in detail that the discrepancy between the quasi-instantaneous TI FWM decay and the slow TR FWM decay is found over a wide range of carrier densities, demonstrating that these findings are intrinsic to Fano resonances in semiconductors.

Moreover, in this paper we have shown that within the framework of the atomic Fano model worked out in Fano's original paper,[10] an intrinsic dephasing time Γ^{-1} of a Fano resonance can be defined, which is given by the inverse of the Fano coupling Γ between the discrete state and the continuum. The absorption profile of a Fano resonance is given by $|\mu_{Fg}|^2 \propto (\epsilon - q)^2/(1+\epsilon^2)$,[10] where μ_{Fg} is the dipole matrix element between the ground state and the Fano eigenstate and ϵ is a normalized energy defined by $\epsilon = (\Omega - E)/\Gamma$. Here, q is the ratio between the optical matrix elements of the transitions to the discrete state and the continuum, Ω is the energy of the discrete state, and Γ the Fano coupling. From the expression for $|\mu_{Fg}|^2$, the linear susceptibility $\chi(\omega) = \int |\mu_{Fg}|^2/(E-\omega^+)dE$ can be calculated analytically in frequency domain. Fourier-transform yields the time domain $\chi(t) \propto [a\delta(t) + \Theta(t)]\exp(-\Gamma t - i\Omega t)$ [a = const, $\delta(t)$ Dirac function, $\Theta(t)$ step function]. Thus, the definition of the intrinsic dephasing time is appropriate since the $\chi(t)$ of a Fano resonance decays exponentially with Γ^{-1} in the long time limit. Deducing the Fano coupling Γ and the other Fano parameters from the linear absorption data and comparing the intrinsic dephasing time Γ^{-1} to the decay times of the TR FWM signal, we have demonstrated that the decay of the TR FWM signal is due to dephasing of the Fano eigenstate.[9]

Thus the TR FWM and the FWM spectrum, whose width corresponds to the TR FWM decay, gives information on the dephasing of a Fano resonance, like in the case of Lorentzian resonances. The quasi-instantaneous decay of the TI FWM signal, however, is not the result of a dephasing process, in striking contrast to Lorentzian resonances. Consequently, we conclude that the fast decay of the TI FWM signal is the manifestation of quantum interference. Obviously, this quantum interference effect is destructive in the sense that it suppresses coherent emission at time delays Δt larger than the width of the excitation pulses in the FWM experiment.

To characterize the conditions under which destructive quantum interference is observed, we have numerically calculated the FWM response within the atomic Fano model.[10] The input parameters for the calculation were chosen to reproduce the linear absorption profile of the hh(1) Fano resonance, which dominates the FWM signal. No additional homogeneous broadening has been introduced in the calculation. The results are depicted in Fig. 4. Both the TI FWM signal [Fig. 4(a)] and the TR FWM signal at $\Delta t = 0$ [Fig. 4(b)] exhibit pronounced slowly decaying contributions with decay times, which are determined by the Fano coupling Γ. Thus, the atomic Fano model qualitatively reflects the important feature of the TR FWM signal from a Fano resonance in a semiconductor: the pronounced contribution with a decay time, which is due to the Fano coupling, i.e., given by the intrinsic dephasing time Γ^{-1}, in agreement with the experiment.

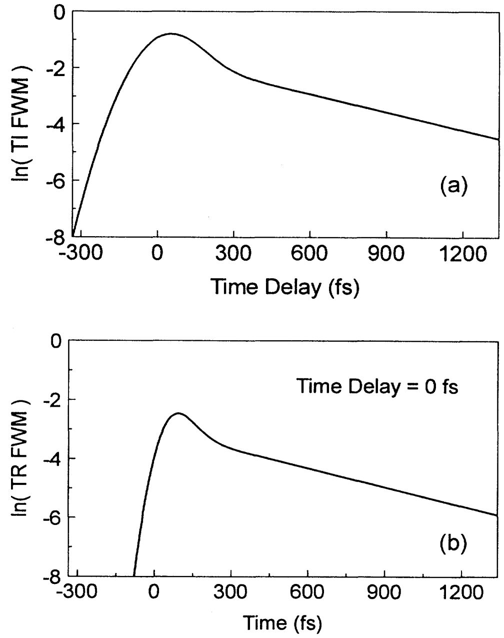

FIG. 4. Numerical calculation based on the atomic Fano model: (a) time-integrated FWM signal, (b) time-resolved FWM signal at $\Delta t = 0$.

The atomic Fano model, however, completely fails to describe the TI FWM signal observed experimentally. While the experimental TI FWM signal decays quasi-instantaneously over a dynamic range of 600, the atomic Fano model predicts a slowly decaying contribution, which at its onset is only a factor 5 smaller than the maximum of the signal.

We have also looked into the effect of the continuum background on which the experimental Fano profile is superimposed (compare to Fig. 1). In an atomic model, excitation of a continuum gives rise to an instantaneously decaying contribution to the TI FWM signal whose strength, relative to other contributions, scales as the ratio between excited continuum and the other resonances excited by the laser. For the conditions of our experiment, we find that the excited continuum background is much too small to account for a quasi-instantaneous decay over almost three orders of magnitude as observed experimentally. Thus, the atomic model cannot provide a complete and consistent description of the nonlinear optical response of a Fano resonance in a semiconductor, although it even quantitatively describes the Fano profiles in the linear absorption spectrum of GaAs.[11]

The reason for the failure of the atomic model in nonlinear optics is that this model does not account for the interplay between Fano interference and nonlinearity in a semiconductor. Unlike in atoms, in a semiconductor, Fano interference and nonlinearity are closely related. They both stem from Coulomb interaction. Coulomb interaction induces the coupling between magnetoexcitons and continuum states,[11] which gives rise to Fano interference, and contributes considerably to the nonlinearity in a semiconductor exploited in a FWM experiment.[4] To account for the quasi-instantaneous decay of the TI FWM signal from a Fano resonance in a semiconductor, Fano interference and nonlinearity have to be treated on an equal footing. Unfortunately, a rigorous theoretical approach, which requires the formulation and solution of the semiconductor Bloch equations in the presence of the magnetic field, is not yet available.

Although the above discussion gives only a preliminary understanding of the destructive quantum interference observed in TI FWM experiments on a Fano resonance in a semiconductor, it shows the characteristic features of this phenomenon. Destructive quantum interference is defined by two criteria: (i) Experimentally, it manifests itself by a quasi-instantaneous decay of the TI FWM signal under conditions where the dephasing time is much longer than the time resolution of the experiment. Of course, the difference between the decay time of the TI FWM and the dephasing time has to be verified experimentally comparing the TI FWM decay to the TR FWM decay or to the width of the FWM spectrum, which both give information on dephasing. The instantaneously decaying signal extends over several orders of magnitude, i.e., the fast decay dominates the shape of the TI FWM signal and a contribution with a dephasing-induced decay is strongly suppressed if present at all. (ii) Many-body Coulomb effects are a necessary prerequisite for the occurrence of destructive quantum interference. Consequently, the relation between the TR FWM signal, the TI FWM signal, and the FWM spectrum cannot even qualitatively be described by a theoretical model which neglects Coulomb effects. These criteria are not fulfilled, e.g., for quantum beats between Lorentzian excitons, showing in which respect destructive quantum interference is different from other quantum interference phenomena in semiconductors.

IV. COUPLED FANO AND LORENTZIAN MAGNETOEXCITON RESONANCES

In order to pinpoint the conditions under which destructive quantum interference is observed, we have experimentally examined the transition from Lorentzian magnetoexciton dynamics to magnetoexciton dynamics involving Fano resonances. This transition can be realized exciting the sample at different energies thereby varying the relative contribution of Lorentzian and Fano magnetoexcitons to the FWM signal. The experiments discussed in this section have been carried out at a magnetic field $B = 6\,\mathrm{T}$. Besides a change of the absolute resonance energies, the main effect of a smaller field is that the energy splitting between the different resonances is smaller, as can be seen from the comparison of the 6 and 10 T absorption spectra in Fig. 1. The smaller energy splitting enables us to excite magnetoexcitons corresponding to Landau level pairs with different Landau quantum number n with 100-fsec pulses.

Figure 5 depicts the absorption spectrum for $B = 6\,\mathrm{T}$ together with spectra of the excitation laser pulses used in the FWM experiments. We have excited either (i) at the Lorentzian hh(0) and lh(0) magnetoexciton resonances, (ii) between the Lorentzian magnetoexciton transitions and the hh(1) Fano magnetoexciton resonance, or (iii) close to the hh(1) Fano resonance. Under excitation condition (iii) the FWM spectrum depends very sensitively on the exact position and width of the pulse spectrum. We will present data taken with two slightly different excitation laser spectra close to the hh(1) Fano resonance, although we have plotted only one of these spectra in Fig. 5. The carrier density generated in the experiments discussed in this section is $\approx 1 \times 10^{16}\,\mathrm{cm}^{-3}$.

We first concentrate on the nonlinear optical response of Lorentzian magnetoexcitons. Under the excitation condition (i) indicated in Fig. 5 by the pulse spectrum at the lowest energy, the spectrum of the FWM signal does not exhibit any contributions from Fano resonances (data not shown), demonstrating that we are monitoring unperturbed Lorentzian magnetoexciton dynamics. The TI FWM signal is plotted as a function of the time delay in Fig. 6. The signal shows a complicated beating modulation on a time scale of roughly 1 ps, which originates from the beating between the hh(0) and the lh(0) magnetoexcitons and a two-photon resonance. This will be the subject of a forthcoming paper. For the present discussion of destructive quantum interference, the important feature of the TI FWM signal is the decay. We observe slowly decaying contributions to the signal both at positive and negative time delays. Averaging over the beating modulation, we find a decay time of 0.8 ps at positive time delays, from which a dephasing time of 1.6 ps is obtained assuming purely homogeneous broadening.[18] The FWM spectrum exhibits narrow emission lines (data not shown) in agreement with the long TI FWM decay. Consequently, we conclude that destructive quantum interference does *not* occur if only Lorentzian magnetoexcitons are excited. In GaAs under magnetic field, Fano resonances have to be involved in the FWM process to observe destructive quantum interference.

Moving the excitation laser spectrum to higher ener-

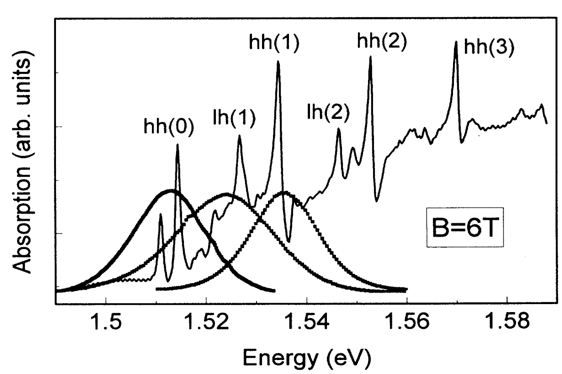

FIG. 5. Linear absorption spectrum for $B = 6\,\mathrm{T}$ and laser spectra indicating the excitation conditions in the FWM experiments.

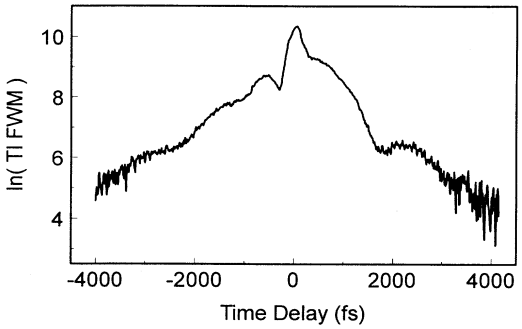

FIG. 6. Time-integrated FWM signal vs time delay exciting only Lorentzian magnetoexciton resonances (carrier density $N \approx 1 \times 10^{16}\,\mathrm{cm}^{-3}$, $B = 6\,\mathrm{T}$).

gies, i.e., exciting between the Lorentzian magnetoexcitons and the hh(1) Fano resonance, both Lorentzian and Fano magnetoexcitons contribute to the FWM signal. The FWM spectrum depicted for $\Delta t = 0$ in Fig. 7(b) shows that the coherent emission from the Lorentzian resonances around 1.515 eV is still the dominating contribution to the total intensity of the FWM signal as compared to the emission from the hh(1) Fano resonance at 1.537 eV. The width δE of the dominating emission line is 3.3 meV, corresponding to a decay τ on a time scale of 200 fs using $\tau = \hbar/\delta E$.

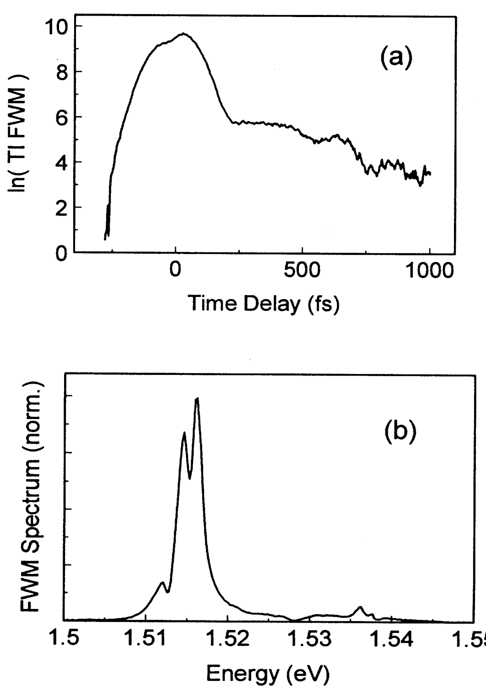

FIG. 7. Time-integrated FWM signal (a) and FWM spectrum at time delay $\Delta t = 0$ (b) for excitation between the hh(0) and the hh(1) resonance (carrier density $N \approx 1 \times 10^{16}\,\mathrm{cm}^{-3}$, $B = 6\,\mathrm{T}$).

The TI FWM signal in Fig. 7(a) exhibits an initial fast decay followed by a slowly decaying contribution with a decay time of 250 fs. The slowly decaying part is about a factor 55 smaller at its onset than the maximum of the signal. The decay time of the slow contribution can be accounted for by the width of the dominating emission line in the FWM spectrum, i.e., the slow decay is due to dephasing. The remarkable feature of the TI FWM signal in the context of destructive quantum interference is the initial fast decay, which extends over one and a half orders of magnitude. Thus, the contribution decaying due to dephasing is rather weak. Such a behavior is not observed if only Lorentzian magnetoexciton transitions are excited (compare Fig. 6). The comparison with the dynamics of unperturbed Lorentzian magnetoexcitons suggests that the initial decrease of the TI FWM signal is the result of the additional excitation of the hh(1) Fano resonance. In view of the small contribution of the hh(1) Fano resonance to the total intensity of the FWM signal, the effect of the Fano contribution is surprisingly strong.

Since destructive quantum interference is intimately related to the excitation of a Fano resonance in our system, the data presented in Fig. 7 indicate that destructive quantum interference starts to affect the shape of the TI FWM signal leading to the initial fast decay under excitation conditions, where the contribution from Fano resonances is much smaller than the Lorentzian contribution to the FWM signal. Destructive quantum interference, however, does not dominate the TI FWM response under these excitation conditions, i.e., the part of the signal decaying due to dephasing is still clearly visible. These observations suggest that destructive quantum interference suppresses the slowly decaying dephasing part of the TI FWM signal rather than decreasing gradually the decay time. This behavior is fundamentally different from the changes observed in a TI FWM experiment if the dephasing time is decreased.

Besides the initial fast decay, the simultaneous excitation of Lorentzian and Fano magnetoexcitons manifests itself by the modulation of the FWM signal. The slowly decaying part of the signal exhibits a nonperiodic modulation, where the temporal spacing between adjacent minima decreases from 330 fs to 220 fs and 180 fs with increasing time delay. A temporal modulation on the time scale of 200 fs requires an energy splitting of about 20 meV. Consequently, the observed quantum beating cannot originate from the lh(0) and hh(0) magnetoexcitons only, but requires a contribution from the hh(1) Fano resonance. The modulation is the result of quantum beating between magnetoexcitons corresponding to Landau level pairs with different Landau quantum number, i.e., Lorentzian and Fano resonances. The nonperiodicity stems from the fact that more than two frequency components contribute to the FWM signal. Nonperiodic beating has been reported by other authors[19,20] and explained by polariton effects. We can rule out this explanation for the case discussed here on the grounds of the FWM spectrum and the time scale of the beating.

In the following, we show that the destructive quantum interference observed for a single Fano resonance at $B = 10\,\mathrm{T}$ completely dominates the TI FWM response at $B =$

6 T if the contribution from the Fano resonance to the FWM signal is large compared to the contribution from the Lorentzian magnetoexcitons.

The data presented in Fig. 8 have been obtained exciting slightly above the hh(1) Fano resonance (pulse spectrum not shown in Fig. 5). The FWM spectra for time delays $\Delta t = -80$ fs, 0 fs, and +80 fs are plotted in Figs. 8(a), 8(b), and 8(c), respectively. For all time delays, the FWM spectrum is dominated by the coherent emission from the hh(1) Fano resonance at 1.537 eV. Smaller contributions from the Lorentzian magnetoexcitons (around 1.515 eV) and from the lh(1) Fano resonance (at 1.529 eV) are also clearly visible. The width of the Lorentzian magnetoexciton contribution is 3.4 meV for all time delays, corresponding to a decay time of 190 fs using the relation $\tau = \hbar/\delta E$. The dominating emission line from the hh(1) Fano resonance has a width of 1.9 meV at $\Delta t = -80$ fs and $\Delta t = 0$ fs slightly increasing to 2.5 meV for $\Delta t = +80$ fs. These widths δE correspond to decay times τ of 340 fs and 260 fs, respectively, based on the relation $\tau = \hbar/\delta E$ between the width of the FWM spectrum and the decay time.

It is important to recall that we have shown in Sec. III that the width of the FWM spectrum is determined by dephasing. Thus, if the decay of the TI FWM signal was solely due to dephasing, the temporal decay of the TI FWM should occur on a time scale determined by the decay times calculated above and should be easily resolvable with 100-fsec pulses. The TI FWM signal in the inset of Fig. 8, however, decays quasi-instantaneously over two and a half orders of magnitude. Consequently, the TI FWM decay is not due to dephasing but due to destructive quantum interference. Considering the coherent dynamic response of a single Fano resonance discussed in Sec. III, this result has to be expected for excitation conditions under which the Fano resonance dominates the FWM signal.

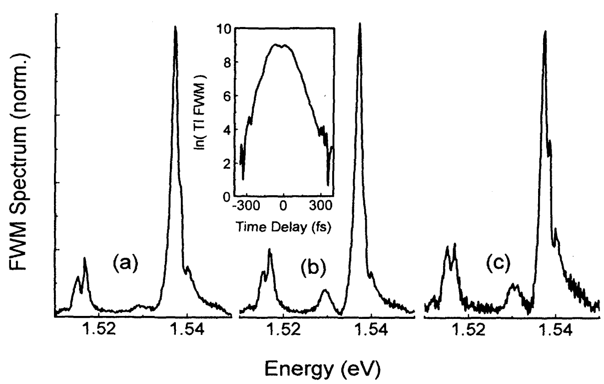

FIG. 8. Four-wave-mixing spectrum at time delays $\Delta t = -80$ fs (a), $\Delta t = 0$ fs (b), and $\Delta t = +80$ fs (c), for excitation slightly above the hh(1) resonance. Inset: Time-integrated FWM signal. Carrier density $N \approx 1 \times 10^{16}$ cm^{-3}, $B = 6$ T.

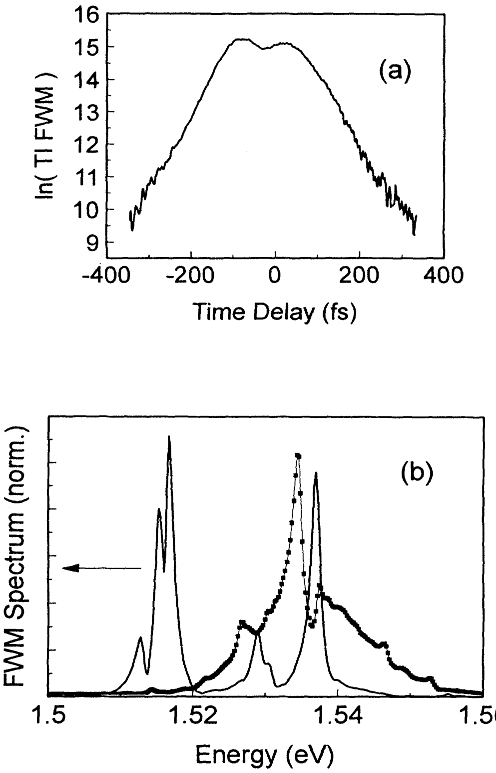

FIG. 9. Time-integrated FWM signal (a) and FWM spectrum at time delay $\Delta t = 0$ [solid line in (b)], for excitation at the hh(1) Fano resonance. Carrier density $N \approx 1 \times 10^{16}$ cm^{-3}, $B = 6$ T. The dotted line in (b) is the product of the linear absorption spectrum and the excitation laser spectrum.

Exciting at the hh(1) Fano resonance as indicated in Fig. 5 by the pulse spectrum at the highest energy, the FWM spectrum in Fig. 9(b) (solid line) shows that now Fano and Lorentzian contributions to the FWM signal have approximately equal intensity. The FWM spectrum consists of pronounced emission lines from the hh(1) (1.537 eV) and lh(1) (1.529 eV) Fano resonances and from the Lorentzian hh(0) and lh(0) magnetoexcitons (around 1.515 eV). The width of the dominating hh(1) Fano and the Lorentzian magnetoexciton emission lines is 1.6 meV and 2.8 meV, respectively, from which decay times of 400 fs and 240 fs are found. Thus, a dephasing-induced decay would have a time constant between 200 fs and 400 fs and could be resolved in this experiment. The decay of the TI FWM signal in Fig. 9(a), however, is instantaneous over the dynamic range of the measurement, which is about 200. It should be noted that a slow contribution of the same relative intensity as the one observed in the data depicted in Fig. 7 could easily be detected if it was present. The absence of a contribution with a decay time determined by dephasing gives clear evidence that destructive quantum interference is dominating the TI FWM response under experimental conditions, where Lorentzian and Fano contributions to the FWM signal have roughly equal weight in the spectrum.

In other words, the coherent emission at time delays larger than the width of the excitation pulses is strongly suppressed if Lorentzian and Fano magnetoexciton resonances are simultaneously excited with equal weight. This is a remarkable result in view of the fact that Lorentzian magnetoexcitons emit a pronounced coherent signal at time delays as large as 1 ps if the simultaneous excitation of Fano resonances is avoided (compare Fig. 6). Since a single Fano resonance does not produce a detectable FWM signal at large time delays, the perturbation of the coherent emission from the Lorentzian excitons cannot be due to interference between electric fields in the detector. From our experimental results, we can immediately conclude that Fano and Lorentzian magnetoexcitons are quantum mechanically coupled. This conclusion is in agreement with experimental results from cw FWM in GaAs under magnetic field[21] and theoretical predictions for magnetoexcitons in ideal two-dimensional semiconductors.[22]

The quantum-mechanical coupling between Lorentzian and Fano magnetoexcitons manifests itself in the TI FWM signal in Fig. 9(a) by two features. Of course, it is the reason for the beating modulation, which is not observed if only a single Fano resonance is excited (compare to the unmodulated TI FWM signal in the inset of Fig. 2). More importantly, as a result of the quantum-mechanical coupling between Lorentzian and Fano magnetoexcitons the destructive quantum interference, which is intimately related to the occurrence of Fano interference in our system, strongly affects the *Lorentzian* magnetoexciton dynamics and suppresses coherent emission from the *Lorentzian* resonances at time delays larger than the pulse width. In this respect, the FWM experiments involving Fano and Lorentzian magnetoexciton resonances show a different feature of the effect of destructive quantum interference.

To illustrate the interaction between Fano and Lorentzian magnetoexciton resonances, we have plotted the product of the linear absorption spectrum and the excitation laser spectrum as a dotted line in Fig. 9(b).[23] The absorption resonances of the Lorentzian magnetoexcitons are hardly visible compared to the absorption resonances of the lh(1) and, in particular, the hh(1) Fano resonances, which are redshifted with respect to the FWM emission (solid line). Although the Lorentzian magnetoexcitons are only very weakly excited by the laser pulse, they contribute considerably to the FWM spectrum. This result is reminiscent of earlier work in semiconductor quantum wells at zero magnetic field, where it was found that the FWM efficiency exciting the exciton is much larger than exciting free carriers in the continuum.[24] This result can be accounted for by the large difference between the dephasing times of excitons and free carriers at zero field.[24] In the case of Lorentzian and Fano magnetoexciton resonances, however, the difference between the dephasing times is rather small. The ratio between the dephasing times is about 2, since the dephasing times of unperturbed Lorentzian magnetoexciton and Fano resonances are of the order of 1 ps and 2 ps, respectively. This raises the question about the microscopic mechanism responsible for the largely different FWM efficiencies observed for Lorentzian and Fano magnetoexcitons. A possible explanation is that the Lorentzian magnetoexcitons are driven by the resonantly excited Fano resonances. Such a process may impose the dynamics of the Fano resonances, which is dominated by destructive quantum interference, on the Lorentzian magnetoexcitons, in agreement with the TI FWM data depicted in Fig. 9(a).

V. CONCLUSION

Using the FWM technique, we have experimentally studied the coherent nonlinear optical response of Fano and Lorentzian magnetoexciton resonances in GaAs under magnetic field. For the excitation of a single Fano resonance, the comparison between the TR FWM signal, the FWM spectrum, and the linear absorption profile of the Fano resonance demonstrates that the decay of the TR FWM signal and the width of the FWM spectrum are determined by dephasing, which takes place on a time scale of several hundreds of femtoseconds under our experimental conditions. In contrast, the decay of the TI FWM signal is quasi-instantaneous and not related to the dephasing time. This quasi-instantaneous TI FWM decay is the manifestation of destructive quantum interference. We have discussed that this experimental result cannot be explained by an atomic Fano model neglecting Coulomb correlation.

Performing FWM experiments in which Lorentzian and Fano magnetoexciton resonances are simultaneously excited, we have characterized the conditions under which destructive quantum interference occurs and investigated the nature of this effect. If only Lorentzian magnetoexciton resonances contribute to the FWM response, destructive quantum interference is not observed and the decay of the TI FWM signal is determined by dephasing. Experiments involving a small contribution from a Fano resonance to the FWM signal indicate that destructive quantum interference should be understood as the suppression of the part of the TI FWM signal, which decays due to dephasing. As soon as the contributions from Fano and Lorentzian magnetoexciton resonances to the FWM signal are approximately equal, destructive quantum interference is observed. Our results show that Lorentzian and Fano magnetoexciton resonances are quantum-mechanically coupled. In the coupled system, destructive quantum interference also suppresses coherent emission from Lorentzian magnetoexcitons.

In summary, we have observed a different type of Fano resonance in GaAs under magnetic field. The TI FWM response of this resonance is dominated by destructive quantum interference but not by dephasing processes. This behavior cannot be understood on the basis of the existing theory for Fano interference and nonlinear optics in semiconductors. We hope that our experimental results will prompt the development of a rigorous theory for the nonlinear optical response of a three-dimensional semiconductor under magnetic field.

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[1] T. Yajima and Y. Taira, J. Phys. Soc. Jpn. **47**, 1620 (1979).

[2] L. Q. Lambert, A. Compaan, and I. D. Abella, Phys. Rev. A **4**, 2022 (1971).

[3] M. Koch, J. Feldmann, G. von Plessen, E. O. Göbel, P. Thomas, and K. Köhler, Phys. Rev. Lett. **69**, 3631 (1992).

[4] H. Haug and S. W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors*, 2nd ed. (World Scientific, Singapore, 1993), and references therein.

[5] S. Weiss, M.-A. Mycek, J.-Y. Bigot, S. Schmitt-Rink, and D. S. Chemla, Phys. Rev. Lett. **69**, 2685 (1992).

[6] D.-S. Kim, J. Shah, T. C. Damen, W. Schäfer, F. Jahnke, S. Schmitt-Rink, and K. Köhler, Phys. Rev. Lett. **69**, 2725 (1992).

[7] K. Leo, M. Wegener, J. Shah, D. S. Chemla, E. O. Göbel, T. C. Damen, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. Lett. **65**, 1340 (1990).

[8] M. Wegener, D. S. Chemla, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. A **42**, 5675 (1990).

[9] U. Siegner, M.-A. Mycek, S. Glutsch, and D. S. Chemla, Phys. Rev. Lett. **74**, 470 (1995).

[10] U. Fano, Phys. Rev. **124**, 1866 (1961).

[11] S. Glutsch, U. Siegner, M.-A. Mycek, and D. S. Chemla, Phys. Rev. B **50**, 17 009 (1994).

[12] See, e.g., E. O. Göbel, K. Leo, T. C. Damen, J. Shah, S. Schmitt-Rink, W. Schäfer, J. F. Müller, and K. Köhler, Phys. Rev. Lett. **64**, 1801 (1990); B. F. Feuerbacher, J. Kuhl, R. Eccleston, and K. Ploog, Solid State Commun. **74**, 1279 (1990); K. Leo, E. O. Göbel, T. C. Damen, J. Shah, S. Schmitt-Rink, W. Schäfer, J. F. Müller, K. Köhler, and P. Ganser, Phys. Rev. B **44**, 5726 (1991).

[13] J. Feldmann, T. Meier, G. von Plessen, M. Koch, E. O. Göbel, P. Thomas, G. Bacher, C. Hartmann, H. Schweitzer, W. Schäfer, and H. Nickel, Phys. Rev. Lett. **70**, 3027 (1993).

[14] T. Rappen, U. Peter, M. Wegener, and W. Schäfer, Phys. Rev. B **48**, 4879 (1993).

[15] A. Lohner, K. Rick, P. Leisching, A. Leitenstorfer, T. Elsaesser, T. Kuhn, F. Rossi, and W. Stolz, Phys. Rev. Lett. **71**, 77 (1993); A. Leitenstorfer, A. Lohner, K. Rick, P. Leisching, T. Elsaesser, T. Kuhn, F. Rossi, and W. Stolz, K. Ploog, Phys. Rev. B **49**, 16 372 (1994).

[16] F. H. Pollak and M. Cardona, Phys. Rev. **172**, 816 (1968).

[17] F. Bassani and A. Baldereschi, Surf. Sci. **37**, 304 (1973).

[18] We have checked the time-resolved FWM signal from the Lorentzian excitons at zero field and found that this signal does not show the signatures of a photon echo. Thus inhomogeneous broadening is negligible in our sample.

[19] K.-H. Pantke, P. Schillak, B. S. Razbirin, V. G. Lyssenko, and J. M. Hvam, Phys. Rev. Lett. **70**, 327 (1993).

[20] T. Rappen, G. Mohs, and M. Wegener, Phys. Status Solidi B **173**, 77 (1992).

[21] M. Jiang, H. Wang, R. Merlin, D. G. Steel, and M. Cardona, Phys. Rev. B **48**, 15 476 (1993).

[22] C. Stafford, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. B **41**, 10 000 (1990).

[23] Taking the product of linear absorption and excitation spectrum, we have checked that the excited continuum states cannot account for the quasi-instantaneously decaying contribution to the TI FWM signal, as discussed in Sec. III for the experiments on Fano resonances at $B = 10\,\mathrm{T}$.

[24] D.-S. Kim, J. Shah, J. E. Cunnigham, T. C. Damen, W. Schäfer, M. Hartmann, and S. Schmitt-Rink, Phys. Rev. Lett. **68**, 1006 (1992).