Ultrafast Coherent Dynamics of Fano Resonances in Semiconductors

U. Siegner, M.-A. Mycek, S. Glutsch, and D. S. Chemla
Department of Physics, University of California at Berkeley and Materials Sciences Division, Lawrence Berkeley Laboratory, Berkeley, California 94720
(Received 27 June 1994)

We present the first investigation of the ultrafast dynamics of the coherent emission from Fano resonances in semiconductors. Time-resolved femtosecond four-wave mixing (FWM) in GaAs under high magnetic field shows that dephasing is dominated by the coupling between discrete states and continua responsible for the Fano interference. In striking contrast to atomic systems and Lorentzian excitations in semiconductors, the decay of the time-integrated FWM signal is not related to dephasing. This decay is due to quantum interference originating from the interplay between semiconductor many-body effects and Fano interference.

PACS numbers: 78.47.+p, 42.50.Md

Quantum mechanical coupling between a discrete state and an energetically degenerate continuum of states leads to new eigenstates, which in linear spectroscopy manifest themselves by an asymmetric line shape, known as a Fano resonance [1]. Fano resonances are characterized by a pronounced minimum in the absorption spectrum at an energy where the transition amplitudes of the discrete state and the continuum interfere destructively. Fano interference has been observed in a variety of atomic, molecular, and semiconductor systems in frequency domain experiments [2–4].

Recently, we have demonstrated how the application of a magnetic field to a bulk semiconductor results in Fano interference [5]. The field leads to the formation of magnetoexcitons corresponding to each pair of conduction band and valence band Landau levels with the same Landau quantum number n. Since the field quantizes only states with the wave vector in the plane perpendicular to the field, each Landau level is associated with a one-dimensional continuum. Coulomb interaction then couples higher-order magnetoexcitons to energetically degenerate continuum states corresponding to lower Landau levels, giving rise to pronounced Fano resonances in the linear absorption spectrum [5].

So far, the implications of Fano interference have not been studied experimentally in the time domain. Time domain experiments are facilitated if nonlinear optical techniques like, e.g., four-wave mixing (FWM) are applied. This experimental technique can easily be implemented to study Fano resonances in a semiconductor under magnetic field.

In semiconductor physics, FWM experiments have yielded valuable information on the dephasing of excitonic and continuum excitations. Moreover, it has been found that the nonlinear optical response of a semiconductor is strongly affected by Coulomb mediated many-body effects [6]. In a FWM experiment, the sample is excited by two short laser pulses, separated by a time delay Δt. The FWM signal pulse can be integrated in time and analyzed as a function of the time delay (time-integrated FWM: TI FWM) as well as, for a fixed time delay, analyzed in real time by means of up-conversion techniques (time-resolved FWM: TR FWM) or spectrally resolved (FWM power spectrum). For Lorentzian transitions, the decay of both the TR and the TI FWM as well as the linewidth of the FWM power spectrum are determined by the dephasing rate. Although in semiconductors Coulomb interaction considerably changes the shape of the TI and TR FWM signal, for Lorenztian-like resonances in semiconductors these two signals decay on the same time scale, which is determined by dephasing [6].

In this Letter, we investigate the dynamics of the coherent emission stimulated from a Fano resonance in a semiconductor directly in the time domain. Femtosecond FWM experiments have been performed on bulk GaAs in a high magnetic field. We experimentally demonstrate that in this system the decay of the TI FWM signal is quasi-instantaneous and is not related to dephasing, in striking contrast to atomic systems and uncoupled excitonic and continuum excitations in semiconductors. The decay of the TI FWM signal is due to quantum interference. The dephasing rate of Fano resonances in semiconductors can only be determined by TR FWM. The dephasing is dominated by the coupling between magnetoexcitons and continuum states, which gives rise to Fano interference. These experimental results cannot be understood within the model of an atomic Fano resonance, but imply that Fano interference and many-body effects must be treated on an equal footing.

The low-temperature, σ^- circularly polarized absorption spectrum of the high-quality GaAs bulk sample we have studied is shown in Fig. 1 for a magnetic field $B = 10$ T. The linear optical properties of this sample have been described in detail in Ref. [5]. Because of mechanical strain at low temperature, the light hole (lh) and heavy hole (hh) exciton transitions are split [7] where the hh exciton is at the higher energy. The application of a magnetic field leads to two series of magnetoexciton transitions

 0031-9007/95/74(3)/470(4)$06.00

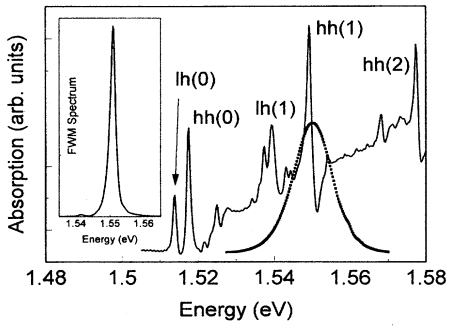

FIG. 1. Low-temperature (1.6 K) linear absorption spectrum of GaAs for a magnetic field of 10 T and σ^- circularly polarized excitation. The laser spectrum (dotted line) indicates the excitation conditions in the FWM experiments. Inset: Power spectrum of the FWM signal at a time delay $\Delta t = 0$ and a carrier density of about 10^{16} cm^{-3}.

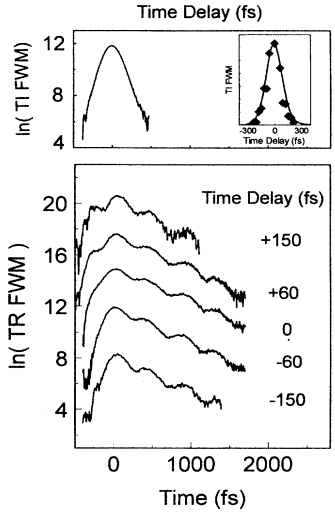

FIG. 2. Semilogarithmic plot of the time-integrated (top) and time-resolved (bottom) FWM signal from a Fano resonance in GaAs at a magnetic field of 10 T. The time-resolved FWM signals taken at different time delays have been shifted along the vertical axis for clarity. Inset: Numerically integrated intensity of the time-resolved FWM (diamonds) and experimental time-integrated FWM signal (solid line) vs time delay.

corresponding to lh and hh resonances, respectively. The lowest-order lh($n = 0$) and hh($n = 0$) magnetoexcitons are Lorentzian lines since they are not in resonance with continuum states. In contrast, the higher-order lh($n \geq 1$) and hh($n \geq 1$) magnetoexcitons form Fano resonances as demonstrated by the line shape and, in particular, by the pronounced absorption minimum at the high-energy side of the resonances.

The FWM experiments have been performed with σ^- cocircularly polarized 100 fs pulses from a self-mode-locked Ti:sapphire laser. The spectrum of the excitation pulses is centered at the hh(1) Fano resonance, as shown in Fig. 1. The magnetic field is 10 T and the temperature of the sample is 1.6 K in all experiments. The inset in Fig. 1 shows the FWM power spectrum obtained at $\Delta t = 0$. The excitation density in the FWM experiment is about 10^{16} cm^{-3}. The FWM spectrum is comprised of an intense narrow contribution from the hh(1) Fano resonance at 1.551 eV and a small contribution from the lh(1) Fano resonance at 1.541 eV, which has also been weakly excited. From the width of the FWM spectrum, a decay of the coherent emission on a time scale of several hundreds of femtoseconds can be expected.

The time domain behavior of the coherent emission stimulated from the Fano resonances is depicted in Fig. 2 for the same excitation density as above. The upper part shows the TI FWM intensity as a function of the time delay in a semilogarithmic plot. Surprisingly, the decay of this signal cannot be resolved with a 100 fs laser pulse, i.e., the TI FWM signal decays quasi-instantaneously and does not show the temporal behavior anticipated from the FWM spectrum.

The lower part of Fig. 2 is a semilogarithmic plot of the TR FWM signal for different time delays between -150 and $+150$ fs. The TR FWM signals are slightly modulated with a beat period which is in good agreement with the energy splitting between the hh and lh contribution in the FWM power spectrum. For all time delays Δt, the TR FWM curves decay much slower than the TI FWM signal. In fact, the decay of the TR FWM signals takes place on a time scale of several hundreds of femtoseconds, as expected from the FWM spectrum. The decay of the TR FWM signals depends only very weakly on the time delay Δt. The intensity of the TR FWM signal, however, changes dramatically if the time delay is changed. This follows clearly from the inset of Fig. 2, which depicts the numerically integrated intensity of the TR FWM signal for different time delays (diamonds) and the experimentally obtained TI FWM signal (solid line) in a linear plot. The different data are in excellent agreement, showing that the experimental results are consistent.

The data in Fig. 2 provide clear evidence that two different time scales are involved in the decay of the TI and the TR FWM signal from Fano resonances in semiconductors. This is a characteristic feature of Fano resonances. Furthermore, we have performed experiments where the laser is tuned to excite either only the hh(0) and lh(0) Lorentzian resonances or simultaneously both the Lorentzian and Fano resonances. We have found that, as expected, in the first case the TI FWM and the TR FWM decay on the same time scale (data not shown). In the other case, however, we found again a significant difference between the TI FWM and TR FWM decay [8].

In the following, we will demonstrate that only the decay of the TR FWM is related to dephasing and that these findings are independent of the carrier density in a wide density range. The TI FWM signals for the

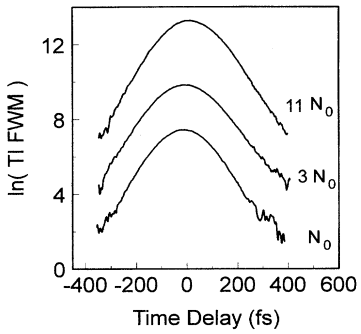

FIG. 3. Semilogarithmic plot of the time-integrated FWM signal vs time delay for the carrier densities $N_0 = 4 \times 10^{15}$ cm^{-3}, $3N_0$, and $11N_0$. The curves have been shifted along the vertical axis for clarity.

densities $N_0 = 4 \times 10^{15}$ cm^{-3}, $3N_0$, and $11N_0$ depicted in Fig. 3 show that the decay of the TI FWM is quasi-instantaneous over the whole density range, i.e., it takes place on a sub-100-fs time scale. In contrast, the decay of the corresponding TR FWM data can be resolved in time. In Fig. 4(a) the decay times obtained from the TR FWM data are plotted vs time delay for the different carrier densities. The decay time of the TR FWM signal is $\approx$250 fs for $N = 11N_0$, $\approx$350 fs for $N = 3N_0$, and $\approx$450 fs at N_0. The comparison of Fig. 3 and Fig. 4(a) demonstrates that the TR FWM signal and the TI FWM signal decay on completely different time scales over a wide range of carrier densities, showing that this behavior is intrinsic to Fano resonances in semiconductors.

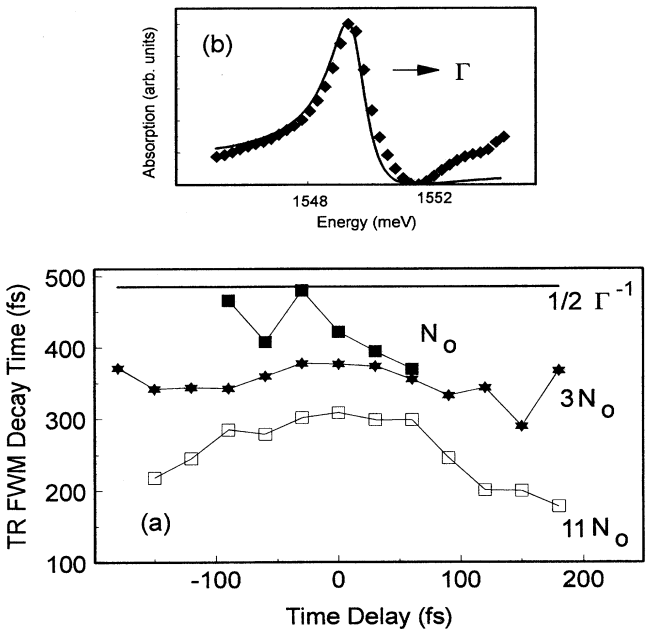

FIG. 4. (a) Decay times of the time-resolved FWM signal vs time delay for different carrier densities $N_0 = 4 \times 10^{15}$ cm^{-3}, $3N_0$, and $11N_0$. The solid line is the decay time due to intrinsic dephasing. (b) Experimental linear absorption profile of the Fano resonance studied in the FWM experiments (diamonds) and calculated Fano profile (solid line) using the parameters given in the text.

It is instructive to compare the decay times of the TR FWM signals to the parameters which can be obtained from the linear absorption data. The linear absorption line shape of a Fano resonance is $|\mu_{Fg}|^2 \propto (\epsilon - q)^2/(1 + \epsilon^2)$ [1], where μ_{Fg} is the dipole matrix element between the ground state and the Fano eigenstate and ϵ is a normalized energy given 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 is the strength of the coupling between the discrete state and the continuum. Γ is related to the coupling matrix element V by $\Gamma = \pi|V|^2$. The parameters q, Γ, and Ω, which determine the Fano profile, can be obtained from the experimental absorption data. We have extracted these parameters for the hh(1) Fano resonance which dominates the FWM response. A closeup of the linear absorption data (diamonds) and the Fano profile calculated from the parameters $q = -2.86$, $\Gamma = 0.668$ meV, and $\Omega = 1549.5$ meV (solid line) is shown in Fig. 4(b). The agreement is excellent.

In the context of time domain experiments, the coupling constant Γ is the most important parameter. It can be expected that the coupling between a discrete state and a continuum, which is at the origin of Fano interference, leads to a temporal decay of the coherent emission from a Fano resonance. This decay is intrinsic to Fano interference. In order to see how the coupling constant Γ is related to a decay in the time domain, we have analytically calculated the linear susceptibility $\chi(\omega) = \int |\mu_{Fg}|^2/(E - \omega)\, dE$ in frequency domain and by Fourier transform obtained the time domain $\chi(t) \propto [a\delta(t) + \Theta(t)]\exp(-\Gamma t - i\Omega t)$ [a = const, $\delta(t)$ Dirac function, and $\Theta(t)$ step function]. It decays exponentially with Γ^{-1} in the long-time limit. Thus it is reasonable to introduce an intrinsic dephasing time Γ^{-1} for a Fano resonance similar to the Lorentzian case where a dephasing time is introduced as the inverse of the half width at half maximum of the Lorentzian absorption profile. For the hh(1) Fano resonance which dominates the FWM response $\Gamma = 0.668$ meV corresponds to an intrinsic dephasing time $\Gamma^{-1} = 970$ fs. The intrinsic decay time $\Gamma^{-1}/2 = 485$ fs is plotted as a thick solid line in Fig. 4(a).

It is obvious from Fig. 4(a) that the decay time of the TR FWM signal approaches the intrinsic decay time if the density it lowered. In fact, at the lowest density N_0 the TR FWM decay time is already very close to the intrinsic dephasing limit imposed by the coupling to continuum states. Consequently, we conclude that the decay of the *time-resolved* FWM signal is due to dephasing, as in the case of Lorentzian resonances. Moreover, we find that the dephasing is governed by the intrinsic coupling

at low densities. The quasi-instantaneous decay of the *time-integrated* FWM signal, however, is *not* related to dephasing, unlike the case of Lorentzian excitons in semiconductors in which the TR FWM and the TI FWM decay on the same time scale determined by the dephasing time. Therefore, we conclude that the quasi-instantaneous decay of the TI FWM signal results from quantum interference.

In order to examine whether an atomic Fano model can account for the experimental results, we have calculated the third-order polarization $P^{(3)}(\Delta t, t)$ for the case where, as assumed by Fano [1], the coupling matrix element V is constant. The results of the calculation and the experimental FWM results can be directly compared since the TR FWM signal is $\propto |P^{(3)}(\Delta t, t)|^2$ and the TI FWM is $\propto \int |P^{(3)}(\Delta t, t)|^2 \, dt$. Within the model and using parameters corresponding to our experiment and to the linear absorption profile of Fig. 4(b), we have numerically calculated the density matrix from the Heisenberg equation for a two-pulse FWM experiment with finite pulses. The numerical calculations show that the TI FWM signal and the TR FWM signal decay on the same time scale determined by dephasing, in contradiction to the experimental results for a Fano resonance in a semiconductor. The same theoretical results have been obtained analytically [9]. Thus the atomic Fano model cannot account for the nonlinear optical response of Fano resonances in a semiconductor although the model well describes the Fano profiles observed in the linear absorption spectrum.

This discrepancy points to the reason for the failure of the atomic model in the case of the nonlinear optical response of Fano resonances in semiconductors. Unlike in atomic systems, in the semiconductor system discussed here the nonlinearity and the quantum interference, which gives rise to the formation of Fano resonances, have the same origin. Both originate from Coulomb interaction. Coulomb interaction provides the coupling between discrete magnetoexcitons and continuum states which results in Fano interference [5]. Besides Pauli blocking, Coulomb interaction also considerably contributes to the nonlinearity which generates the FWM signal [6]. Thus Fano interference and optical nonlinearity are very closely related in a semiconductor. Therefore, only a model which treats Fano interference and Coulomb mediated semiconductor many-body effects on the same footing can fully describe the experimental results and account for the quantum interference observed in the TI FWM experiment. The atomic Fano model, which does not account for many-body effects, consequently can only describe the linear optical response of a Fano resonance in a semiconductor.

A full treatment of the nonlinear optical response of Fano resonances in GaAs requires the solution of the semiconductor Bloch equations in the presence of a magnetic field up to at least third order in the electric field of the laser. This theory is not yet available.

In conclusion, we have presented the first experimental study of the time domain response of Fano resonances. Using four-wave mixing spectroscopy, we have studied Fano resonances in GaAs under high magnetic field. Our results demonstrate that in the time domain the coupling, which gives rise to Fano interference, dominates the dephasing of the coherent emission from the Fano state. Thus dephasing experiments allow a direct investigation of the coupling between discrete and continuum states. This time domain technique should be applicable to a variety of atomic, molecular, and solid state systems. For Fano resonances in semiconductors, the dephasing information can only be obtained from time-resolved FWM experiments. The decay of the time-integrated FWM signal is instantaneous and not related to dephasing. This particular feature of the nonlinear optical response of Fano resonances in semiconductors is due to quantum interference related to the interplay between Fano coupling and optical nonlinearity, both of which in semiconductors originate from Coulomb interaction.

The authors are indebted to P. Thomas for many helpful discussions. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Division of Materials Sciences of the U.S. Department of Energy, under Contract No. DE-AC03-76SF00098. U. S. and S. G. wish to thank the Deutsche Forschungsgemeinschaft for financial support.

[1] U. Fano, Phys. Rev. **124**, 1866 (1961).
[2] *Electron and Photon Interaction with Atoms,* edited by H. Kleinpoppen and M. R. C. McDowell (Plenum, New York, 1976).
[3] J. J. Hopfield, P. J. Dean, and D. J. Thomas, Phys. Rev. **158**, 748 (1967).
[4] D. Y. Oberli, G. Böhm, G. Weimann, and J. A. Brum, Phys. Rev. B **49**, 5757 (1994).
[5] S. Glutsch, U. Siegner, M.-A. Mycek, and D. S. Chemla, Phys. Rev. B (to be published).
[6] See, for example, S. Schmitt-Rink and D. S. Chemla, Phys. Rev. Lett. **57**, 2752 (1986); S. Schmitt-Rink, D. S. Chemla, and H. Haug, Phys. Rev. B **37**, 941 (1988); 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); M. Wegener, D. S. Chemla, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. A **42**, 5675 (1990); S. Weiss, M.-A. Mycek, J.-Y. Bigot, S. Schmitt-Rink, and D. S. Chemla, Phys. Rev. Lett. **69**, 2685 (1992); D.-S. Kim, J. Shah, T. C. Damen, W. Schäfer, F. Jahnke, S. Schmitt-Rink, and K. Köhler, Phys. Rev. Lett. **69**, 2725 (1992); H. Wang, K. Ferrio, D. G. Steel, Y. Z. Hu, R. Binder, and S. W. Koch, Phys. Rev. Lett. **71**, 1261 (1993).
[7] F. H. Pollak and M. Cardona, Phys. Rev. **172**, 816 (1968).
[8] U. Siegner, M.-A. Mycek, S. Glutsch, and D. S. Chemla, Phys. Rev. B (to be published).
[9] P. Thomas (private communication).