Disorder mediated biexcitonic beats in semiconductor quantum wells

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(Received 30 April 1996)

Beats in the transient four-wave-mixing signal from strongly inhomogeneously broadened semiconductor quantum wells are observed. Based on their phase and polarization properties, the beats are assigned to biexcitons in the mesoscopically disordered material. With increasing excitation intensity a halving of the beat period is observed and is accurately reproduced by calculations including fifth-order contributions. [S0163-1829(96)04731-5]

The coherent interaction between light and semiconductors is a subject of extensive current interest. Under low to moderate excitation conditions, the coherent response near the fundamental absorption edge in a direct-gap semiconductor is dominated by excitonic effects. Although early transient four-wave-mixing (TFWM) experiments on semiconductors[1] were interpreted in terms of the existing understanding for atomic and molecular systems based on the optical Bloch equations (OBE),[2] it quickly became apparent that this was not sufficient for semiconductors, since their coherent response is substantially more complex due to the many-body interactions of the electron-hole excitations.[3] At the level of an effective Hartree-Fock approximation these interaction effects are included in the semiconductor Bloch equations (SBE). In a more phenomenological way aspects of the interband exchange effects can be modeled as ''local field''[4,5] allowing a relatively simple numerical analysis of experiments.[5] Efforts to understand the polarization dependence of the TFWM signal[6–8] demonstrated the importance of interactions beyond the Hartree-Fock level such as biexcitonic contributions[9–16] and excitation-induced dephasing.[17,18]

The presence of inhomogeneous broadening due to well width fluctuations in quantum well (QW) structures has both complicated interpretation and presented an opportunity for the study of disorder.[19,20] The interplay of disorder and many-body interactions has recently been examined.[21] In this paper, we present results that show that the combined contributions of both biexcitons and strong inhomogeneous broadening produce new features in the time integrated TFWM (TI TFWM) signal. In particular, in this situation, biexcitonic beating becomes apparent in TI TFWM showing unique temporal and polarization dependences. Note that in a homogeneously broadened system *no beating due to biexcitons is present in the TI TFWM signal*, although it is present in the time resolved TFWM signal (TR TFWM).[13] Furthermore, we observe intensity-dependent half-period contributions to the beats[11] and show that these arise from fifth-order contributions to the signal.

Strong inhomogeneous broadening is typical of very thin QW's because an interface fluctuation of a single monolayer is a significant fraction of the total well width, and hence of the confinement energy. In such a system the excitons can be sufficiently strongly localized by the interface fluctuations so that they actually behave as if they were confined in a quantum dot[22–24] where the biexcitonic binding energy is greatly enhanced.[25,22] We have performed experiments on two sets of thin multiple QW's (MQW). One set is GaAs/AlAs grown by molecular-beam epitaxy[26] (well width of 4.1 nm) and the other lattice matched (InGa)As/InP grown by metallo-organic vapor phase epitaxy (well widths of 1.6 and 3.0 nm).[27] All samples exhibited inhomogeneously broadened absorption and luminescence lines of approximately 15–20 meV in width. For the (InGa)As/InP samples the alloy well material also contributes to the inhomogeneous broadening (estimated to be approximately 8 meV, with the remainder being due to well width fluctuations); the monolayer splitting is around 35 meV.

A Kerr-lens-mode-locked Ti:sapphire laser producing 110-fs pulses was used to perform the experiments on the GaAs/AlAs samples. For the (InGa)As/InP samples the Ti:sapphire laser was used to synchronously pump a femtosecond optical parametric oscillator that produced pulses in the 1.1–1.4 μm wavelength regime.[28] The sample was held at 5 K in helium vapor. All experiments were performed in the standard two pulse TFWM geometry where two pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are incident on the sample with a time delay τ between them (see inset Fig. 1). The signal is emitted in the direction $2\mathbf{k}_2-\mathbf{k}_1$. For TI TFWM the signal is time integrated with a slow detector and measured as a function of τ. The excitation spectra were centered on the low-energy wings of the respective absorption spectra, where the localization is expected to be the strongest.

In Fig. 1 we show the TI TFWM signal for three different polarizations of the two incident pulses: (a) cocircular, (b) colinear, and (c) crossed linear, all for the (InGa)As/InP (1.6-nm well, 20.2-nm barrier) sample. Clear beats, corresponding to an energetic splitting of 3.2 meV (in the GaAs/AlAs sample the splitting is even as high as 5.2 meV),[29] are present in (b) while they are absent from (a). These results strongly suggest that biexcitons are the origin of the beats as a biexciton is composed of excitons of opposite spins and, hence, should not be excited by cocircularly polarized beams. Further confirmation of this assignment is obtained

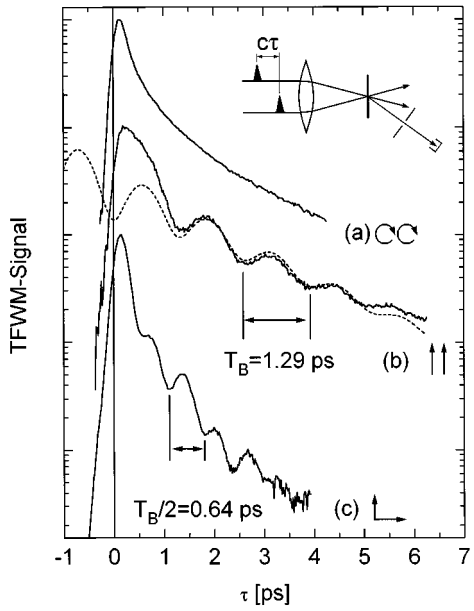

FIG. 1. TI TFWM traces for the (1.6 nm)/(20.2 nm) (InGa)As/InP MQW sample. (a) Circularly copolarized pulses, (b) linearly copolarized pulses, (c) linearly cross-polarized pulses. The dotted line in (b) is a phenomenological fit with a damped sinusoidal function. The experimental traces are normalized and shifted by one order of magnitude relative to each other. The inset illustrates the experimental geometry.

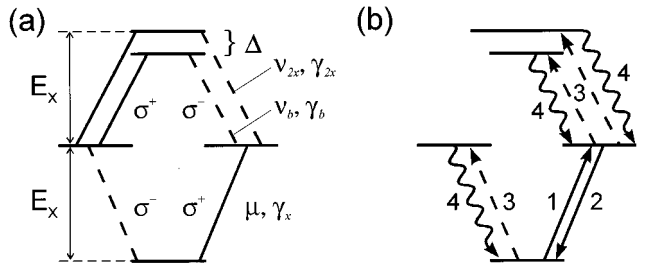

FIG. 2. (a) The five-level scheme used for the interpretation of the data. E_x is the $1s$ excitonic resonance energy and Δ the biexcitonic binding energy. The optical matrix elements and dephasing rates of the exciton, biexciton, and two-exciton scattering state are indicated as well as the circularly polarized selection rules for the transitions. (b) Schematic diagram showing the various contributions to the third-order polarization that result in beats. Each arrow is labeled by the corresponding order in perturbation. The wiggly arrows correspond to the radiating polarizations.

from examining the temporal dependence of the beats in Fig. 1(b) and noticing that *there is a beat minimum at* $\tau=0$. This is obscured in the data due to effects caused by the finite pulse duration around $\tau=0$, so to aid in realizing this we have fit the data for $\tau>1.5$ ps using a simple damped cosine function, superimposed on an exponential decay [broken curve in Fig. 1(b)].

To understand why both the phase of the beats and the very presence of the beats itself are surprising, the various contributions to the TFWM from a biexcitonic 5-level system (5LS) [Fig. 2(a)] must be more closely considered. As mentioned above, for a homogeneously broadened system there are no biexcitonic beats in the TI TFWM signal. There are beats in the TR TFWM signal that arise due to an interference between the fields of a given handedness radiated by the transition between the one-exciton state to the bound biexciton state with those radiated by the transitions from either the ground state to the opposite handed one-exciton state and/or the one-exciton state to unbound two-exciton state. The radiating transitions are shown in Fig. 2(b) as wiggly lines. At $t=0$ these two contributions destructively interfere for colinear polarization because the population gratings in the ground state and one-exciton states have opposite signs, and these population gratings contribute to the source terms for the third-order polarization. For homogeneous broadening, there will be no beats in the time integrated signal since the temporal shape of the TR TFWM is independent of τ; it just decreases in overall amplitude with increasing delay τ. In the presence of inhomogeneous broadening the TR TFWM becomes a photon echo, and, as the echo position is 2τ, it moves through the beats, causing them to become apparent in the TI TFWM signal. (For a similar discussion for the case of a three-level system see Ref. 31. The photon echo nature of the signal was confirmed for the GaAs/AlAs sample earlier.[26])

By analytically solving the OBE for the phenomenological 5LS depicted in Fig. 2(a), this interpretation can be substantiated. While there exists a theoretical approach for biexcitonic effects, based on extending the SBE to include terms beyond the usual Hartree-Fock approximation,[32,33] it does not permit analytic solution. Recent work elucidated the correspondences between the two approaches and showed that the phenomenological approach is a good approximation of the more complete theory.[34] The phenomenological approach used here was able to successfully model several experimental results for homogeneously broadened samples.[13,15,35]

To obtain an analytic result that contains the basic features, we solve the OBE to third order in the laser field for the idealized case of δ-function pulses. Additionally we assume that the inhomogeneous broadening is sufficiently strong that the TI TFWM signal is given by $I(\tau)=\Sigma_{i=x,y}|P_i^{(3)}(t=2\tau,\tau\geqslant 0)|^2$, where

$$P_x^{(3)}(t=2\tau,\tau\geqslant 0)=4i\ \cos\phi\{\nu^2(e^{i\Delta\tau/h}+1)\times e^{-(\gamma_x+\gamma_b)\tau}-e^{-2\gamma_x\tau}\},$$

$$P_y^{(3)}(t=2\tau,\tau\geqslant 0)=4i\ \sin\phi\{\nu^2(e^{i\Delta\tau/h}+1)e^{-(\gamma_x+\gamma_b)\tau}\}.$$

Here we have assumed that the second pulse is polarized parallel to the x axis, and that the polarization of the first is rotated by an angle ϕ. The dephasing rates of the exciton and two-exciton states are denoted by γ_x and γ_b, respectively. Δ is the biexciton binding energy and ν_b the optical matrix element for the one-exciton to two-exciton transition (see Fig. 2). For parallel polarization ($\phi=0$) the signal indeed starts with a minimum, while for perpendicular polarization it starts with a maximum, in agreement with Figs. 1(b) and 1(c) and our analysis.

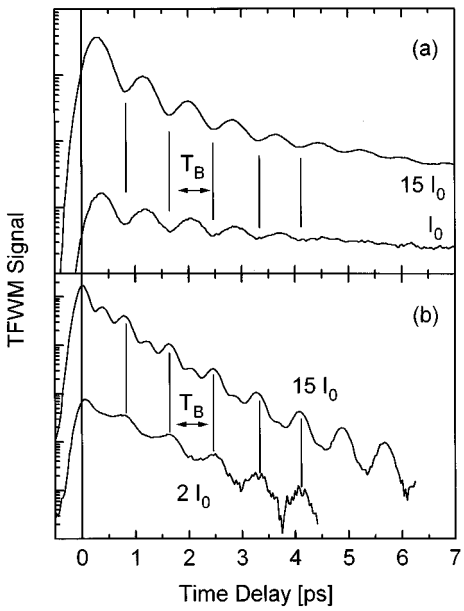

FIG. 3. TI TFWM traces for the (4.0 nm)/(3.0 nm) GaAs/AlAs MQW sample showing the intensity dependence. (a) Linearly copolarized pulses, (b) linearly cross-polarized pulses. The vertical lines spaced by the beating period T_B are guides to the eye. Note the faster decay in the perpendicular situation.

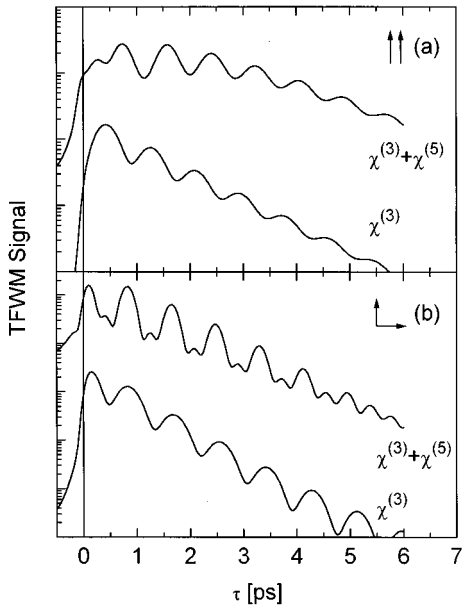

FIG. 4. Theoretical TI TFWM traces. Notation is the same as in Fig. 3. Parameters: $\mu=1$, $\nu_b=0.85$, $\nu_{2x}=0.7$, $\gamma_x^{-1}=4.0$ ps, $\gamma_b^{-1}=2.0$ ps, and $\gamma_{2x}^{-1}=1.5$ ps.

This behavior should be contrasted with beats that arise from two excited states coupled to a common ground state by the light field (e.g., light hole–heavy hole, bound free, etc.).[36] In this case the beats always start with a maximum for colinearly polarized pulses. This analysis shows that the quantum beat phase yields important information as to the underlying level system. Previously obtained experimental data might thus contain more and different information than hitherto realized. Note that because terms due to the biexciton contribute to the third-order response, *there will always be biexcitonic contributions to the linearly polarized response* and they will be density independent.

An additional intriguing feature in Fig. 1(c) is the appearance of half-period beats (see also Ref. 11). Our analysis provides a natural explanation for this feature. In Fig. 3 we present data for the GaAs/AlAs MQW sample (4.1-nm wells, 3.9-nm barriers), showing an additional beating with half the period of the biexcitonic beat period for cross-polarized high-intensity excitation, which is absent for all other conditions. We observe similar behavior in six different samples (see also Ref. 28). The intensity dependence suggests that half-period beats arise from higher than third-order contributions. We numerically solve the OBE to fifth order in the laser fields, including finite pulses, finite inhomogeneous broadening, and local field effects. The inhomogeneous broadening is assumed to be completely correlated; uncorrelated broadening would lead to a rapid decay of the beats.[37] The results (Fig. 4) do reproduce all of the features observed in the experiment: beating at the biexciton frequency for low intensity; beats start with a minimum for copolarized and a maximum for cross; half-period contributions for elevated intensities; no beats for cocircular excitation (not shown). The calculations clearly show that the half-period features are due to the fifth-order terms. The local fields are essential for the correct reproduction of the phase of the half-period oscillation. Additionally different decay times[7,8] for copolarized and cross-polarized excitation can be qualitatively explained by our model, assuming a faster dephasing for the upper (two-particle) transitions than for the lower (one-particle) transitions.

In summary, we have presented the results of TI TFWM experiments on strongly inhomogeneously broadened semiconductor MQW and their theoretical interpretation on the basis of the OBE. We observe beats that, based on their polarization properties and phase, we can assign to the combined presence of inhomogeneous broadening and biexcitons. We show that the inhomogeneous broadening is essential for biexcitonic beating to be present in the time integrated signal, which has not previously been realized. Half-period beats are also observed and are attributed to fifth-order effects. The TI TFWM technique allows one to precisely measure binding energies for inhomogeneously broadened excitonic absorption lines and complements the ''classic'' photoluminescence techniques, which work best when the linewidth is considerably smaller than the binding energy.

The authors would like to acknowledge fruitful discussion with K. Victor, V. M. Axt, and J. Kuhl. We thank M. Preis for expert technical assistance. S.T.C. received funding from the Alexander von Humboldt Foundation. T.F.A. and M.K. received funding from the Alexander von Humboldt Foundation. T.M. received funding from the scientific branch of the NATO through the Deutscher Akademischer Austauschdienst (DAAD). This work has been financially supported by the Deutsche Forschungsgemeinschaft through the Leibniz- and Gerhard-Hess-Förderpreis and through the Sonderforschungsbereich 383.

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