Optical Coherence in Semiconductors: Strong Emission Mediated by Nondegenerate Interactions

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The presence of interactions between carriers in differing states, nondegenerate interactions, is experimentally demonstrated to result in strong, new contributions to the optical coherent response of semiconductors. These interactions are manifest in a new, state-selective, two-pulse four-wave-mixing technique as emission from a state that is excited by only one pulse. The emission arises due to interactions with another state that is excited by both pulses. These interactions are observed between the 1s exciton and continuum states and also between distinct magnetoexciton states. The resulting contributions must be included to properly understand the coherent response. [S0031-9007(96)00804-6]

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The coherent optical response of semiconductors due to intrinsic excitations is very sensitive to interactions among the excitations. This has clearly been demonstrated for interactions between energetically degenerate excitonic states [1–3], where the contributions of polarization scattering [1,2] and excitation-induced dephasing (EID) [3] have been distinguished. These experiments have been modeled in terms of the semiconductor Bloch equations (SBE) [4–6], which include the Coulomb interaction within a Hartree-Fock approximation.

In this Letter we demonstrate that nondegenerate interactions, i.e., interactions between carriers with very different energies, have a dramatic effect on the coherent response of semiconductors. While the nondegenerate interactions have often been observed in the incoherent response of semiconductors [7,8], it has not previously been realized that the interactions result in completely new contributions to the semiconductor coherent response. These new contributions lead to essential modification of the coherent response that completely dominates the dynamics observed in experimental techniques such as transient-four-wave-mixing (TFWM) for broad band excitation where multiple states are simultaneously excited. Furthermore, these interactions result in an effective coherent coupling between the continuum states. Interference between excitonic and continuum states has been observed in the context of excitonic wave packets [9], but the crucial role of interactions was not realized.

We observe the new contributions in a modified TFWM experiment where the first pulse has a narrower spectrum than the second, while maintaining complete coherence with the second pulse in the region of spectral overlap. We designate this partially nondegenerate TFWM (PND-TFWM). *This experiment shows the presence of strong coherent emission at energies that have no spectral overlap with the first pulse.* We observe such emission at the 1s exciton when the first pulse only overlaps the continuum states, or, with the application of a magnetic field, at the 1s magnetoexciton when the first pulse only overlaps the 2s and 3s magnetoexcitons. This is shown schematically in Fig. 1. This coherent emission cannot be explained if the relevant states are treated independently. By combining the states into a single system with a common ground state, we are able to readily include the interactions and show how they lead to the new contributions to the coherent response responsible for our observations. Additionally, this demonstrates how interference between multiple quantum mechanical pathways modifies the TFWM dynamics.

The standard two-pulse TFWM geometry is used [see Fig. 2(a)] [1–3,9–12], where two cocircularly polarized pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are separated by a time delay τ. The signal is emitted in the direction $2\mathbf{k}_2 - \mathbf{k}_1$ and either time integrated with a slow detector or spectrally resolved with a multichannel detector. The incident 40 fs pulses are produced by a Kerr lens mode-locked Ti:sapphire laser. They are nearly transform limited, with

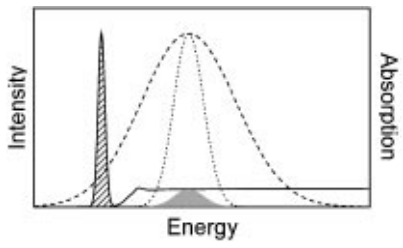

FIG. 1. Schematic illustration of the coherent emission. The spectrally narrow first pulse (dotted line) and spectrally broad second pulse (dashed line) only have mutual coherence in the shaded region. However, coherent emission is observed in regions that only overlap the second pulse (hatched region). The solid line is the absorption in a semiconductor to illustrate that, although the first pulse only overlaps the continuum, emission is observed at the exciton.

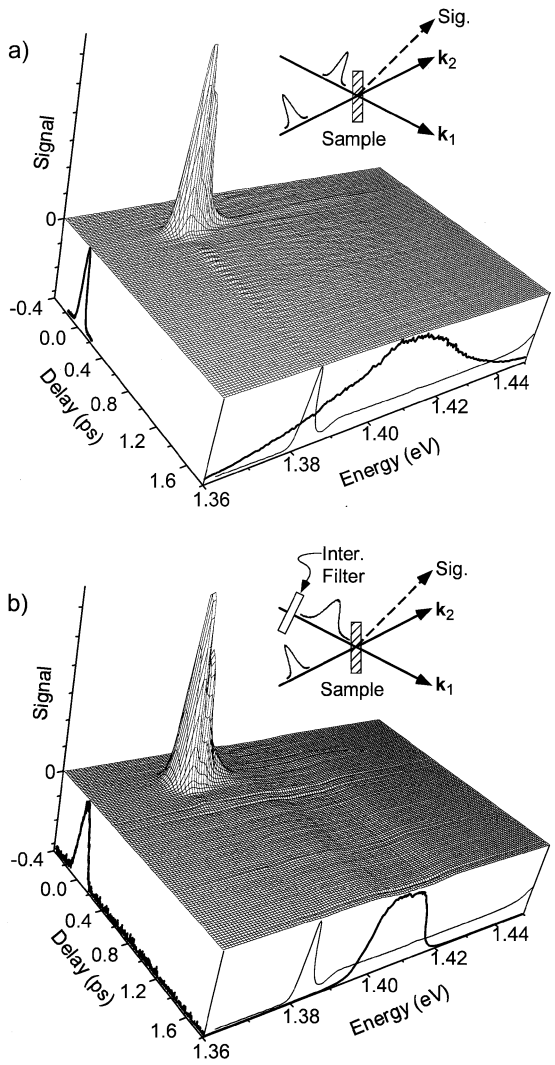

FIG. 2. TFWM signal as a function of photon energy and delay for (a) TFWM and (b) PND-TFWM. The laser spectrum (thick line) and linear absorption spectrum (thin line) are plotted on the front panel; the spectrally integrated TFWM signal is plotted on the side panel. Insets are the experimental geometry.

approximately a 30 meV bandwidth, the center of which is tuned approximately 22 meV above the $1s$ heavy hole (hh) exciton at 1.381 eV (see Fig. 2). This produces maximum excitation of the continuum, strong excitation of the $1s$ hh exciton and negligible excitation of the light hole (lh) exciton at 1.452 eV. For the PND-TFWM experiments [Fig. 2(b)], the $\mathbf{k}_1$ pulse is passed through an interference filter, narrowing its bandwidth to approximately 13 meV, stretching the pulse to approximately 90 fs but maintaining its mutual coherence with $\mathbf{k}_2$, which is still broad band. The spectrum center is the same as the broad band pulse. The narrow band pulse has negligible energy at the $1s$ hh exciton. Note that the broad band coherence and mutual coherence between pulses (obtained by deriving the pulses from a common laser oscillator) distinguish these experiments from those using continuum generation as a spectrally broad source [7,8].

The experiments were performed on a $In_{0.14}Ga_{0.86}As/GaAs_{0.71}P_{0.29}$ 50 period multiple quantum well with 8.3 nm wells and 8.0 nm barriers [13]. The layers are symmetrically strained, which increases the hh-lh splitting to 70 meV, allowing broad band excitation of the hh exciton and continuum states without lh-hh band mixing. The GaAs substrate is not removed as it is transparent at the energies of interest. The sample was held at 8 K in a split coil superconducting magnet.

In Fig. 2 we compare the spectrally resolved (a) TFWM and (b) PND-TFWM signals as a function of delay. The linear absorption spectrum, spectrum of $\mathbf{k}_1$, and spectrally integrated TFWM signal are also plotted in each. In Fig. 2(a) the TFWM spectrum is clearly emitted at the energy of the $1s$ exciton. The fact that the TFWM is dominated by the exciton for broad band excitation overlapping both the exciton and continuum has been observed earlier and attributed to the large exciton oscillator strength combined with its slow dephasing compared to the continuum states [14,15]. In Fig. 2(b) it is clear that the TFWM spectrum is still dominated by the exciton, even though the spectrum of $\mathbf{k}_1$ does not overlap it. *The dominance of the excitonic resonance in the case where the first pulse does not excite it is quite surprising* and has not previously been reported. The signal in Fig. 2(b) is comparable in strength to that in Fig. 2(a); indeed it is essentially the same when the difference in power for $\mathbf{k}_2$ is taken into account; i.e., this is not a small effect. The signal strength has a cubic dependence on the incident power, which verifies that the data correspond to the $\chi^{(3)}$ limit.

These results cannot be understood if all of the relevant states are independent, as is often assumed. The independent transition picture [Fig. 3(a)] results from ignoring the Coulomb interactions and predicts that the TFWM signal from the continuum is a photon echo. This can also be obtained from the SBE in the low excitation coherent limit for large detuning of the optical field [6], where only the Coulomb terms responsible for the formation of excitons are kept. The independent transition picture cannot reproduce our results; in particular, it predicts that the PND-TFWM spectrum should be determined by that of $\mathbf{k}_1$, clearly in disagreement with Fig. 2(b). We have verified this using simple calculations in which the TFWM response for δ-function pulses [16] is weighted by the real pulse spectrum [17]. In a more complete treatment based on the SBE, the internal fields are renormalized due to the strong semiconductor-photon coupling. Although the renormalization contains nondegenerate terms [4–6], the renormalization can be ruled out as an explanation for our observations because we observe no signal for completely nondegenerate incident fields [17].

The PND-TFWM results clearly show that the continuum states are interacting with the exciton to produce this coherent emission. While it is possible to see that new terms will arise if population dependences are phenomenologically included in the material constants (dephasing rate, oscillator strength, transition energy) of the optical

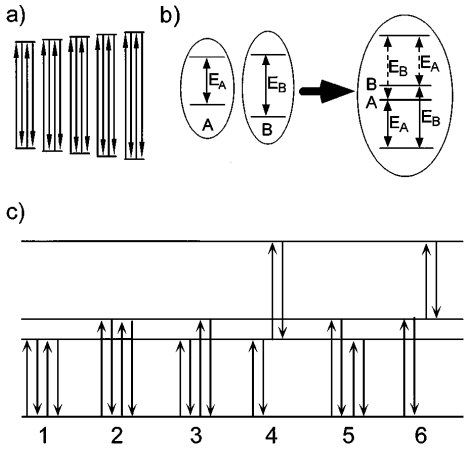

FIG. 3. (a) Independent transition picture. (b) Combination of a pair of two-level systems into a single four-level system. In the combined picture the dashed transitions correspond to excitation of that transition in the presence of a population of the other. The contributions to the TFWM signal are schematically illustrated in (c). If there are no interactions then terms 3 and 4 cancel each other, as do 5 and 6.

Bloch equations or SBE, as has been done for EID [3,18], a common ground state picture provides direct insight. Restricting the discussion to a pair of two-level systems for the moment, it is easy to transform from the independent level scheme to a four-level scheme with a common ground state [Fig. 3(b)]. This transformation results in new contributions to the coherent response of the four-level scheme as compared to the pair of two-level systems; these are numbers 3–6 in Fig. 3(c). However, these new contributions cancel each other, as long as the upper [dashed line in Fig. 3(b)] transitions are completely degenerate with the lower (solid) ones. Specifically in Fig. 3(c) terms 3 and 4 cancel each other, as do 5 and 6. If the systems (A and B) interact this degeneracy (any one of the dephasing rate, oscillator strength, or transition energy is sufficient) will be lifted and new contributions to the nonlinear response will appear as the cancellations no longer occur because the upper transitions represent excitation of a given transition in the presence of the other. This discussion can be generalized to apply to a semiconductor by replacing one of the states with the continuum. This picture has also been used in the context of quantum beats between excitons localized on separate interface islands [19].

Although this transformation into a multiple level system may not be the only possible approach to understanding our results, it does reveal several essential aspects of the coherent response. It allows insight into how interactions can give rise to a new contribution to the coherent response, without requiring a detailed understanding of the microscopic mechanisms. Additionally, perturbation calculations for the nonlinear response can be done starting from the appropriate level scheme, yielding ready connection to a significant body of previous work. Finally, a multiple level scheme does apply directly to magnetoexcitons (below), giving it wider applicability.

There are several interesting aspects of the optical response that are readily apparent in this scheme. First is the appearance of terms which couple the two transitions, allowing the excitation of one to result in coherent emission at the other. In the case of our PND-TFWM experiments the two pulses create a population in the continuum, which is converted to a signal by the broad band second pulse. We have verified this sequence by doing a three-pulse experiment [17]. Note that these terms do not correspond to the creation of a population in the state that radiates, i.e., there is no phase space filling, but rather correspond to a modification of the "environment" for that state due to the presence of other carriers [8]. For example, free carriers can increase the scattering, thereby enhancing the dephasing, or, through screening, can decrease the oscillator strength of the exciton. A second point is that the combined picture results in multiple quantum mechanical pathways, which can interfere with each other. The resulting interference is quantum mechanical, and can result in beating, but these beats do not correspond to a population oscillation of the radiating transition, but rather an oscillation of the environment.

When a continuum coupled to a common ground state is involved, rather than inhomogeneously broadened discrete transitions, the quantum interference leads to a rapid decay of the TFWM signal as a function of delay [9]. The rapid decay due to such a quantum interference will mask the real dephasing rates of all participating states and always result in a signal that is pulse-width limited if the continuum bandwidth is larger than the pulse bandwidth. This can be seen in Fig. 2 by comparing the spectrally integrated signals; the decay is clearly slower for the PND-TFWM case, due to the smaller spectral width of continuum states contributing to the signal. To test that the excitonic emission does display interference due to the presence of multiple quantum mechanical pathways, we applied a 5 T magnetic field to the sample (Fig. 4). This breaks up the continuum into magnetoexciton states. In this case the TFWM signal displays quantum beats due to interference between the magnetoexcitons. The PND-TFWM from the *1s exciton displays quantum beats, the period of which is determined by the 2s-3s splitting*, as they are the only magnetoexcitons excited by $\mathbf{k}_1$. This proves that the evolution of the TFWM at the $1s$ exciton is determined by interference between terms corresponding to excitation of higher lying states by $\mathbf{k}_1$. It is this interference that leads to the rapid decay of the TFWM signal, anomalously rapid as compared to the spectral width (or time resolved decay), as has previously been observed [20]. Additionally, the fact that the ratio of the $1s$ emission signal strength to that of the higher lying excitons is similar for TFWM and PND-TFWM clearly shows the dominance of the contributions from the interaction induced cross terms.

Because the interparticle interactions are strongly screened at high excitation densities or in doped material, we expect that in these cases the discrete transition picture is more applicable. This is consistent with our observation

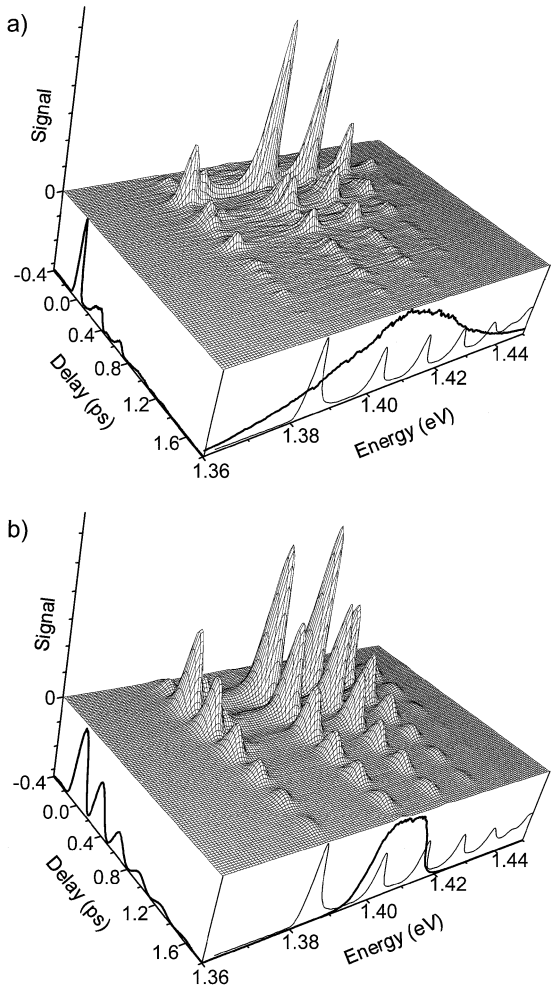

FIG. 4. Same as Fig. 2, but for an applied magnetic field of 5 T. The quantum beats at the 1*s* exciton result from simultaneous excitation of the 2*s* and 3*s* excitons.

of a stronger continuum contribution at high density [17]. It is also consistent with the fact that a photon echo from the continuum states [12], which only occurs in the discrete transition picture, not in the common ground state picture, has only been verified at high density [15] or in doped material [21].

In summary, these results provide new insight into the coherent response of semiconductors. By using a new experimental technique (PND-TFWM), the critical role of interactions between excitations of different energy is clearly demonstrated. The effects of these nondegenerate interactions can be understood in terms of a common ground state picture. This new technique suggests a new class of experiments, e.g., different polarization configurations, three-pulse experiments, etc., to elucidate the characteristics of the microscopic mechanisms and whether or not they are included within the formalism used to derive the SBE.

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