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13. ABSTRACT *(Maximum 200 words)*

Research on this program has focused on the study and application of the nonlinear response of GaAs and GaAs heterostructures. The work has resulted in the demonstration that the primary nonlinear optical response in GaAs is in fact not due to static interactions in the Hamiltonian (e.g., Coulomb, exchange, etc.), but rather due to strong dynamic exciton-exciton interactions, similar to resonance collisional broadening in dense gases. This so-called excitation induced dephasing contribution has been shown to be a primary contributor to the observed polarization dependences of the nonlinear response which were not accounted for by the earlier theory. Other work on this project has focused on the detection and characterization of quantum coherences, notably the two-photon induced coherence associated with the biexciton and the Raman coherence associated with the heavy-hole-light-hole interaction. Measurements have shown the ultrafast two-photon oscillation of the two-photon coherence and the unexpectedly fast relaxation of the Raman coherence. The measurements provide insight into the nature of relaxation. Related work has shown new behavior regarding exciton transport including magnetic field induced freezeout and, in high quality material, ballistic transport.

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Nonlinear Optical Studies of Excitons in Semiconductor Heterostructures

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K. B. Ferrio and D. G. Steel, "Detection of Heavy-Hole-Light-Hole Raman Coherence in GaAs quantum wells," in preparation.

BOOK CHAPTER CONTRIBUTION

D. G. Steel, H. Wang, S. T. Cundiff, "Four-wave mixing in quantum-well systems," invited chapter in *Optics of Semiconductor Nanostructures*, pp 75-126, Fritz Henneberger, Stefan Schmitt-Rink, and Ernst Göbel, eds (Akademie Verlag GmbH, Berlin, 1993).

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INVITED PAPERS IN NATO PROCEEDINGS

D. G. Steel, S. T. Cundiff, H. Wang, "Coherent Nonlinear Laser Spectroscopy of Excitons in Quantum-wells," Optical Phenomena in Semiconductor Structures of Reduced Dimensions, David J. Lockwood and Aron Pinczuk, eds, NATO ASI, Series E **248**, pp 187-200, Kluwer Academic Publishers (Dordrecht, 1993).

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INVITED CONFERENCE PAPERS

D. G. Steel, H. Wang, S. T. Cundiff, M. Jiang, "Nonlinear Spectroscopy of Excitons: A Probe of Disorder," to be presented the symposium "Frontier in Laser-Condensed Matter Interactions," ILS'92, organized by M. V. Klein.

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D. G. Steel, Kyle Ferrio, H. Wang, Min Jiang, Anne Schaefer, "The role of exciton interactions in the GaAs nonlinear optical response," International Workshop on Lasers, 1994.

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Anne C. Schaefer, Min Jiang, Duncan G. Steel, "Investigation of the Optically Induced Increase of the Effective Excitonic Oscillator Strength in GaAs," IQEC '94.

K. B. Ferrio, Hailin Wang, Vinod Subramaniam, and Duncan G. Steel, "Distinct Higher-Order Nonlinear Optical Response for Spin-Dependent and Spin-Independent Contributions to Four Wave Mixing," IQEC '94.

Min Jiang, A. C. Schaefer, D. G. Steel, "Measurement of the Red Shift due to Spin-Dependent Nonlinear Interactions in GaAs," IQEC '94.

A. Schaefer, M. Jiang, D. Steel, "Nonlinear Induced Absorption in GaAs: Evidence of Excitation Induced Decay," APS Meeting, March, 1994.

N. H. Bonadeo, M. Jiang, Vinod Subramaniam and D. G. Steel, "Polarization studies of the free polarization decay on GaAs: Evidence for Excitonic Coherence Transfer," QELS'95 (1995).

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K. B. Ferrio, J. R. Guest, and Duncan G. Steel, "Electronic Raman Coherence in GaAs: State-Specific Scattering Processes," QELS '96, accepted.

A. C. Schaefer and D. G. Steel, "The effects of momentum scattering on exciton motion: Observation of non-diffusive transport," QELS '96, accepted.

A. C. Schaefer, D. G. Steel, "Non-Diffusivė Transport of Excitons in GaAs and the Effects of Momentum Scattering," American Physical Society, March Meeting, 1996.

Educational Activity

Several students have participated in the program as evidenced in the above publications. Three of the students have since graduated with a Ph.D. One former graduate student went to Bell Laboratories and is now an assistant tenure track professor. The other student recently finished a two-year postdoctoral position at Philipps University in Marburg, Germany and has accepted a position at Bell Laboratories. A third student has recently graduated and has accepted a postdoctoral position in fiber optics communications. Two other students will be graduating this year. Their work provides the bulk of the data in the following section on future work which includes preliminary results. Two to three new students have just joined the group and will be involved in the new program.

BRIEF OUTLINE OF RESEARCH FINDINGS:

The objective of the current program has been the development and application of nonlinear laser spectroscopy to the study of excitation dynamics near the band edge of semiconductor heterostructures. Our early studies have underscored the major role of inherent growth and process disorder in determining the nonlinear response. Hence we have found that complimentary experiments in high-quality bulk GaAs were essential to facilitate our understanding of the *intrinsic* response and provide crucial insight into the effects of confinement and disorder in heterostructures.

Our progress has relied on a broadly based experimental program employing both high-resolution frequency-domain (developed by us on earlier ARO programs) and picosecond/femtosecond time-domain coherent and incoherent nonlinear spectroscopic techniques such as four-wave mixing (FWM) and variations such as the stimulated photon echo, time-resolved free polarization decay, and more traditional differential transmission (DT) measurements. We have obtained new understanding of the dynamics and energy level structure associated with excitons in GaAs/AlGaAs quantum-wells, particularly as these properties are affected by disorder and magnetic fields. Equally important, however, has been our work in bulk GaAs which shows for the first time that the nonlinear response in this system at moderate densities is due to dynamic

exciton-exciton interactions leading to a nonlinear response proportional to the effective exciton-exciton collision cross-section.

Some of the most important results are summarized below:

Studies of exciton-exciton interactions:

In a collaboration with the theoretical group directed by Stephan Koch (previously at the University of Arizona and now at the University in Marburg), we have performed a series of experiments on the heavy hole (hh1) exciton in GaAs to demonstrate that the nonlinear optical response is due to exciton-exciton scattering which leads to dephasing of the exciton coherence, a behavior identified as excitation-induced dephasing (EID) and similar to resonant collisional broadening in atomic vapors. We have shown that the nonlinear susceptibility is proportional to the exciton-exciton scattering cross-section, and this observation led to the inclusion of scattering terms previously ignored in the semiconductor Bloch equations. The results are critical in explaining the dependence of the nonlinear response on the polarization of the incident beams, behavior which could not be reconciled with the original SBE.

The effects of excitation-induced dephasing were also included in the ordinary optical Bloch equations. The solution of these modified optical Bloch equations then provided a simple physical picture for the interpretation of experimental behavior and comparison with other nonlinear optical effects in dense systems, such as local fields. The equations also permitted an analytic solution which gave excellent agreement with experiments in bulk GaAs.

The more detailed SBE were applied (by the Koch group) to the complex quantum-well system for insight into the polarization dependence of the signal measured in our experiments.

High-resolution FWM polarization-sensitive spectroscopy was performed in bulk GaAs and showed clear evidence of a red-shifted resonance at both the *lh*1 exciton and the *hh*1 exciton. Based on numerous measurements, this resonance is believed to arise from the biexciton. The *lh-lh* biexciton binding energy obtained is ~0.2 meV.

Progress in GaAs/AlGaAs quantum-wells – Direct measurement of the excitonic Zeeman splitting and lineshape in the presence of inhomogeneous broadening:

High-resolution frequency-domain nonlinear spectroscopic methods developed on this program using low-intensity cw lasers have been used to observe spin-flip-induced hole-burning in GaAs/AlGaAs multiple-quantum-wells. These measurements have provided the first direct observation of the excitonic Zeeman splitting within the inhomogeneous width and enable a measurement of the magnetic field dependent Landé g-factor, a determination which provides important information on the degree of band mixing.

Nonlinear magneto-optical properties of GaAs:

We have probed the nonlinear magneto-optical response of Wannier excitons in bulk GaAs at 4 K. The results show a large enhancement in the nonlinear optical response related to a field-induced decrease in exciton mobility due to magnetic freeze-out. High-resolution cw nonlinear spectroscopy was used to measure the relative strength of inter-Landau level coupling and showed that the nonlinearity is dominated by Coulomb effects.

Studies of the light hole in GaAs show the presence of an "extra resonance" in the nonlinear optical response in the presence of a magnetic field. The application of a magnetic field

induces a resonance which leads to the observation of a narrow dip in the primary resonance. The primary resonance has a width given by the exciton spin-flip rate; the narrow dip has a width determined by the recombination rate. The solution of the optical Bloch equations shows that this behavior reflects the dynamics of two observables represented by distinct spatial Fourier components of the total population and the net population transfer between the two oppositely spin-oriented excitonic states, similar to the normal decay modes of the system. The relative amplitude of these observables is determined by varying the relative polarizations of the incident fields and the strength of the applied magnetic field.

Studies of four-wave mixing lineshapes:

We have shown theoretically and experimentally that, contrary to the generally accepted assumption that cw hole-burning spectroscopy can study only slow dynamics, a specific cw four-wave mixing interaction can be used to *selectively* suppress slow processes contributing to dephasing in order to observe the fast contributions. These results were critical to observations of the spin-flip-induced hole-burning described above.

Exciton-Exciton interactions leading to quantum coherence:

Using a phase conjugate interferometer, we have detected the ultrafast nonradiative oscillation (~770 THz) of the two-photon induced quantum coherence associated with the excitation of the biexciton. This excitation pathway has a negligible contribution in long time scale measurements such as luminescence, but is clearly observable with short pulse excitation. The study of this pathway provides new insight regarding creation and decay of coherence.

Using spectrally narrow pulses we have observed the Raman coherence induced between the heavy hole and light hole. The results show a faster than expected decay of the coherence, pointing to strong state-specific reservoir interactions.

In the following, we describe in somewhat more detail some of the highlights of our progress during the past three years. Details of this work as well as progress in areas not highlighted below are presented in the publications identified above.

Studies of Exciton-Exciton Interactions:

Excitation Induced Dephasing as an Origin of the Nonlinear Optical Response

One of the most significant outcomes of the present work is the discovery that the intrinsic nonlinear optical response in bulk GaAs is due to a dynamical exciton-exciton interaction (in contrast to all the earlier work which assumed a static interaction) which leads to unique features of the nonlinear response. Specifically, it became clear at the end of the last funding period that the then current model for the nonlinear response was failing significantly to account for key experimental features including the polarization dependence of the nonlinear susceptibility. These discrepancies were observed in our laboratory by studying the time-resolved emission in a transient FWM experiment which showed that using linearly polarized light in a transient four-wave mixing experiment, the so-called co-polarized response was a photon echo while the cross-polarized

response was a free polarization decay. Under those experimental conditions, the model predicted no difference in the response. Other striking differences were also observed by our group and others.

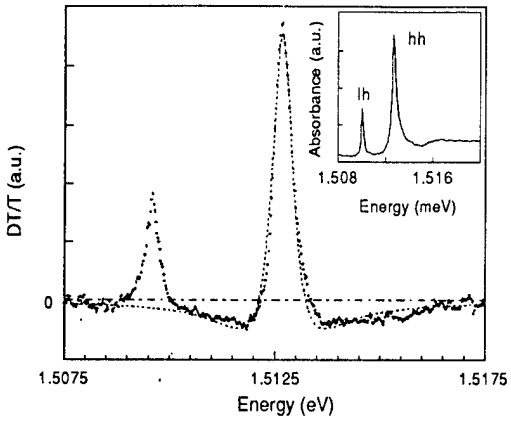

Figure 1 DT spectrum at an exciton density of $3x10^{15}/cm^3$. The dashed line is the fit discussed in the text. Height is the only free fitting parameter. Inset : Linear absorption of the sample.

To determine the origin of the polarization dependence, DT and FWM experiments were carried out at 6 K in a very high-quality homogeneously broadened 200-nm GaAs layer grown by molecular beam epitaxy. Uniaxial strain along the growth direction lifts the degeneracy between the *hh*1 and *lh*1 excitons as shown in the inset of Fig. 1. The absorption width is 0.2 meV for the *lh* exciton and 0.4 meV for the *hh* exciton, showing evidence for the high quality of this material.

We first considered the DT data which provide a basis for determining the dominant nonlinearities in the system. The laser pulses for the DT measurement had an autocorrelation width of 1.5 ps and a spectral width of 4 meV, centered 1 meV below the *hh* resonance. The probe beam was spectrally resolved with 0.1 meV resolution by an optical multichannel analyzer. The response of the *hh* is shown in Fig. 1. The DT spectrum of the *hh* is nearly symmetric and shows only a negligible shift in the exciton energy (less than 0.1 meV). The change in the oscillator strength is determined by the spectral integral of the *hh* DT response. Phase-space filling makes only a small contribution to the bleaching of the *hh* resonance as seen in the spectrum of Fig. 1, which has a net area less than 5% of the positive area. The dominant nonlinear optical response comes from changes in the linewidth (i.e., dephasing rate). As described more thoroughly below, this is a significant result: The change in linewidth not only shows that the dephasing rate is density-dependent, as is well-known in both bulk and quantum-well GaAs materials, it also implies that *the nonlinear response is proportional to the effective exciton-exciton collision rate*.

If the DT spectrum is represented by the difference of two Lorentzians, $f\left\{\gamma_2/(\delta^2+\gamma_2^2)-\gamma_1/(\delta^2+\gamma_1^2)\right\}$, where f is the oscillator strength, δ is the detuning from line-center, and $\gamma_{1,2}$ are the dephasing rates without and with the pump beam, respectively, then the zero-crossings in the DT spectra can be used to determine the density-dependent dephasing rate as well as the low-density line width. The obtained EID scattering rate is in general agreement with earlier FWM measurements. We fit the *hh* spectrum in Fig. 1 (dashed line) with no adjustable parameter except the peak height. As seen, the agreement is excellent.

To understand these key experimental observations along with other critical observations reported in these experiments, our collaborators at the University of Arizona analyzed the semiconductor Bloch equations which describe the optical response of a multi-band semiconductor to an external optical field. Within the statically-screened Hartree-Fock approximation these equations account for phase-space blocking (including band filling and exchange effects) and screening of the Coulomb potential. In this study EID is modeled by the density dependence of the imaginary part of the dynamically screened Hartree-Fock self-energies. Consideration was restricted to the two optical polarization equations for the $m_j=\pm 3/2$ *hh* excitons because, from the Luttinger-Kane theory, the only consequence of the *hh-lh* coupling is the mixing of *s*-like and *d*-like excitons combined with slight energy shifts, and the creation of *d*-excitons is forbidden in optical transitions. Excellent agreement with experiment was obtained.

Finally, we note that although it may seem unusual that incoherent processes which lead to additional dephasing should give rise to coherent optical interactions such as FWM, such behavior has long been known in atomic systems. Effects of collision-induced coherent phenomena were first predicted by Bloembergen and subsequently observed. Eventually it was even predicted and observed (on this program) that the vacuum radiation field (through spontaneous emission) also gives rise to terms in the FWM response.

Biexcitonic Contribution to the Collinear Optical Response in GaAs:

It has been well established theoretically that the binding energy of biexcitons in semiconductors decreases rapidly with a decreasing ratio of the effective hole mass to the effective electron mass. Hence, the relatively large binding energy of biexcitons in CuCl (with a binding energy E_B ~30 meV) or CdS and CdSe (E_B ~5 meV) facilitated the detection and study of these excitations. The existence of biexcitons in GaAs, however, has been much more difficult to confirm because of the small binding energy (theoretical estimation E_B~ 0.15 meV). Traditionally, investigations of biexcitons were conducted either by two-photon absorption at an energy equal to half of the biexciton energy or by analyzing the excitation density dependence of

photoluminescence associated with radiative decay from the biexcitonic state to the free exciton state. Recently, the effects of biexcitons in GaAs quantum-wells, where E_B is enhanced because of confinement, have been studied through the spectral and polarization properties of the nonlinear optical response of the material. The existence of biexcitons in GaAs/AlGaAs quantum-wells was suggested by the temporal oscillation behavior in DT for oppositely-handed circularly-polarized pump and probe beams, as well as in FWM for linearly cross-polarized excitation beams. In a strongly inhomogeneously broadened system, the photon echo was attributed to a two-exciton coherence. The transient FWM response measured at negative time delay, distinct dephasing rate, and alteration in the homogeneity of line broadening detected in transient FWM with different exciton spin characteristics were also attributed to biexcitonic effects. More directly, careful studies of the spectrally-resolved transient FWM experiments have also been able to show a clear contribution from the biexciton resonance. In bulk GaAs, a photoluminescence peak characterized by a superlinear dependence on excitation level was attributed to a biexciton resonance, though the reported binding energy (0.5 meV) was larger than the theoretical estimation.

Using cw polarization-sensitive FWM in strained bulk GaAs, we have observed polarization-dependent resonances in the nonlinear optical response that are red-shifted with respect to *lh* and *hh* excitonic resonances, respectively (the shift is ~0.2 meV at the *lh)*. The spin dependence of this red-shifted signal, the excitation-density-independent value of the red shift and the observation of an induced absorption at a corresponding energy supports the assignment of this behavior to bound exciton complexes, i. e. biexcitons. Measurements of the decay dynamics demonstrate that the response is associated with step-wise excitation, and the red shift corresponds to the binding energy of the biexcitons (~0.2 meV for *lh-lh* and ~1 meV for *lh-hh* complex). Furthermore, decay profiles measured with differential FWM (in the presence of a incoherent pump beam), clearly exhibit evidence of nonlinearities distinct from the excitons and consistent with the assignment of biexcitons.

The polarization dependence of the phase conjugate FWM response was first investigated as a function of the back beam frequency, ω_3, (denoted FWMω_3) with $\mathbf{E}_1^*$ and $\mathbf{E}_2$ tuned in resonance with the *lh* excitons. In these experiments, fields $\mathbf{E}_1$ and $\mathbf{E}_2$ intersect in the sample at small angle while field $\mathbf{E}_3$ is aligned to be counter-propagating with respect to $\mathbf{E}_2$. As shown in Fig. 2b, the FWM$\omega_{3\|}$ ($\mathbf{E}_1 \| \mathbf{E}_2 \| \mathbf{E}_3$)shows no frequency shift with respect to the linear absorption. Such results are consistent with earlier theoretical predictions and experimental reports [2] showing that the Coulomb screening and the exchange interaction give nearly equal but opposite shifts in bulk GaAs. On the other hand, the FWMω_3 spectrum obtained for ($\mathbf{E}_1 \perp \mathbf{E}_2 \| \mathbf{E}_3$) (Fig. 2c) clearly reveals a *red shift* and changes in line width with respect to the *lh* and *hh* exciton resonances. (The small resonance at 1.5033 eV saturates easily with excitation density and is believed to be

related to impurities.) The ratio of the amplitudes between $FWM\omega_{3\parallel}$ and $FWM\omega_{3\perp}$ is ~500:1, this large ratio, however, does not imply an intrinsically weaker SD response. In the cw FWM described here, the signal strength varies as the square of the excitation grating relaxation time. As was discussed earlier, the appropriate time in the $FWM_{\perp}$ geometry is twice the exciton spin-flip time (~125 ps); while in the $FWM_{\parallel}$ geometry, the time is mainly given by exciton recombination (~ .85 ns) for large grating spacing. Hence, if measurements were made in the transient regime, the strengths of the signals would be comparable.

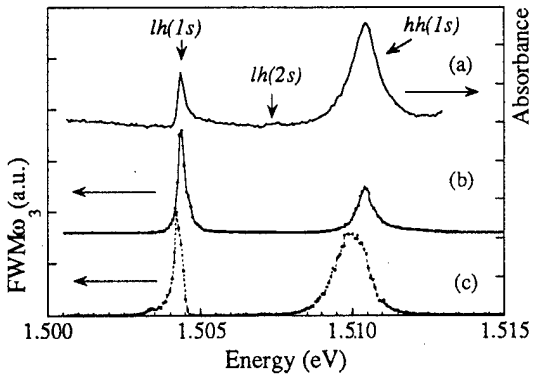

Figure 2. (a). Linear absorption spectrum. (b) $FWM\omega_{3\parallel}$ spectrum. (c) $FWM\omega_{3\perp}$ spectrum. Excitation energy determined by the frequencies of the fields giving rise to $\mathbf{E}_1^* \cdot \mathbf{E}_2$ is fixed at lh(1s) resonance.

Recalling that only spin-dependent interactions may generate a $FWM_{\perp}$ signal, a red-shifted $FWM\omega_{3\perp}$ response could, in principle, be due to several effects. For example, exchange effects are well known to lead to a shift in the resonance frequency. This explanation is unlikely, however, since the exchange interaction depends strongly on excitation density while experimentally we find that for excitation densities varying over more than an order of magnitude, the $FWM_{\perp}$ signal strength is nearly cubic while the value of the red shift remains *constant for the whole range of applied excitation densities*. Alternatively, a Raman-type contribution to the nonlinear response is also a possibility, however, such a resonance is determined by the detuning between $\mathbf{E}_1$ and $\mathbf{E}_2$, which is not the case for the present result. We also note that impurity effects could also lead to a shifted resonance (e.g., the FWM peak at 1.5033 eV) though contributions from impurity bound excitations saturate at relatively low excitation density, unlike the primary resonance in Fig. 1c. Hence, the density-independent value of the red shift, the selection rules, and the lack of saturation are all consistent with assigning the red-shifted $FWM_{\perp}$ resonance to biexcitons.

In the $FWM\omega_{3\perp}$ spectrum (Fig. 2c), the absence of a distinct excitonic spin-dependent component can be attributed to two effects: First, the dipole moment associated with the transition from exciton to biexciton is predicted to be "giant" due to the large biexcitonic Bohr radius, hence

the biexcitonic nonlinearity is expected to be larger than the spin-dependent component of the excitonic nonlinearity. Second, the binding energy of the biexciton has a value comparable with the line width of the exciton resonance. Hence, the two peaks cannot be well resolved but the whole peak appears at a somewhat bluer position and is asymmetric on the high energy side.

Assuming the $FWM_{\perp}$ response is due to biexcitons, we consider two excitation pathways: the two-photon coherence and the step-wise excitation. Clarification of the dominant pathway is possible by studying the decay dynamics of the $FWM_{\perp}$ response. For the two-photon-coherence, the perturbation sequence is: $N_G \xrightarrow{\mathbf{E}_2(\mathbf{E}_3)} p_{GX} \xrightarrow{\mathbf{E}_3(\mathbf{E}_2)} p_{GB} \xrightarrow{\mathbf{E}_1^*} p_{BX}, p_{GX}$ (Where G, X, B represent the ground, exciton and biexciton states respectively, and p and N describe the optical polarization and population respectively). The FWM signal arises from the optically-induced quantum coherence, p_{GB}, oscillating in time with frequency $2\Omega - \Omega_B = \omega_2 + \omega_3$ (with resonant excitation) where $\hbar\Omega$ is the exciton transition energy and $\hbar\Omega_B$ is the biexciton binding energy, as seen directly in CuCl [100]. For the step-wise excitation, the perturbation sequence is: $N_G \xrightarrow{\mathbf{E}_2(\mathbf{E}_1^*)} p_{GX} \xrightarrow{\mathbf{E}_1^*(\mathbf{E}_2)} N_X \xrightarrow{\mathbf{E}_3} p_{XB}$; The FWM signal is diffracted from the population gratings excited by $\mathbf{E}_1^*$ and $\mathbf{E}_2$. The contribution due to the back grating ($\mathbf{E}_3\mathbf{E}_1^*$) is experimentally verified to be negligible because the excitons quickly diffuse out of the tightly spaced grating. We have also shown that the contribution of the two-photon coherence is negligible for cw experiments, as expected, given the relative contributions of the different relaxation rates.

Based on the above analysis, we tentatively assign the $FWM_{\perp}$ resonance to a step-wise excitation; the red shift measures the binding energy of the two-exciton state, which is ~0.2 meV for *lh-lh*, which is very close to the theoretical estimation of the binding energy of biexcitons. With excitation beams creating *lh*-excitons, the red shift of $FWM\omega_{3_{\perp}}$ at the *hh* gives the binding energy for *lh-hh* two-exciton state. The larger binding energy for the *lh-hh* two-exciton state (~ 1 meV) compared to that for *lh-lh* state is believed to be due mainly to the difference in the effective masses of the *lh* and *hh*.

Detection of Biexciton Two-Photon Coherence in GaAs multiple quantum well:

The two-photon coherent excitation of the biexciton and the contribution to the FWM response is identified above by a perturbation sequence leading, at second order, to a term identified as the two-photon coherence, p_{GB}. This term reflects a direct two-photon excitation of the biexciton. This excitation pathway is *independent* of the exciton population as seen in the above perturbation

sequence. Moreover, this term is unique in that it oscillates in time at a frequency given above as $2\Omega - \Omega_B$. This oscillation does not lead to observable radiation but does lead to a detectable time dependent phase in the nonlinear susceptibility that can be detected in a phase conjugate interferometer. In transient FWM, this contribution occurs for the so-called negative time ordering of the pulses. Detection of the ultrafast oscillation, however, is significant in that it demonstrated the important nature of the coherent excitation, the presence of a two-photon quantum induced coherence, and because studies of the relaxation of this coherence provide new information on the nature of the complex interactions.

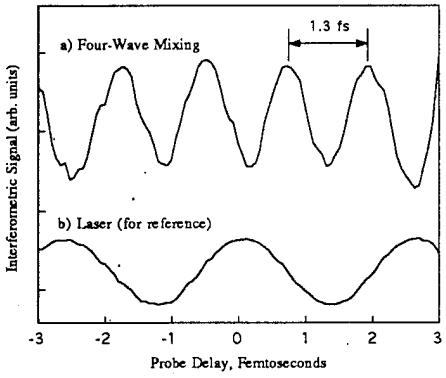

Figure 3. Measurement of the two-photon induced coherence due to the presence of the biexciton. (a) shows the oscillation of the two-photon coherent phase detected in a phase conjugate interferometer. (b) shows the response of an ordinary interferometer for comparison. As expected, the oscillation in (a) is approximately twice that of (b).

The first experimental observation of the ultrafast oscillation is shown in Fig. 3. The upper trace shows the nonradiative oscillation at a frequency ~770 THz, corresponding to the twice the exciton energy less the biexciton binding energy. Given the limited number of oscillations, there is not sufficient accuracy to determine the biexciton binding energy. It is important to note that in a phase conjugate interferometer, only the two-photon oscillation is detectable: no oscillation at or near the laser frequency is present in this geometry. Indeed, replacement of the sample with an ordinary mirror leads only to a DC offset, with no oscillation. For reference, a separate interferometer was configured to show the ordinary oscillation period in Fig. 3b. The faster oscillation of the biexciton coherence is immediately clear in Fig. 3a.

We note that further evidence in support of this interpretation is had by examining the polarization dependence of the amplitude of this oscillation as a function of time delay. For co-polarized exciton, non-two-photon-coherent related contributions dominate the coherent nonlinear response for positive time delays and also lead to a stronger response. The experiments show that indeed, the copolarized response favors the positive time ordering.

Progress in Studies of Quantum Wells

Direct Measurement of the Excitonic Zeeman Splitting and Lineshape in the Presence of Inhomogeneous Broadening:

Studies of the fundamental nonlinear optical response in quantum wells were made in the presence of a moderate magnetic field. Of these studies, the most significant result was our observation of spin flip induced hole burning leading to the first high resolution spectra resolving the exciton Zeeman doublet within the inhomogeneous linewidth.

Work by numerous groups as well as our own have clearly demonstrated the difficulty presented by interface disorder in the interpretation of the optical response of quasi-two-dimensional systems. Because of this, a significant portion of our effort during this research period was devoted to improving our physical understanding of bulk semiconductors. Nevertheless, we continued our experimental work on quantum-wells to provide further insight into the role of disorder in these systems. Measurements in these systems include the first experiments on the cw FWM response of excitons in a magnetic field. The cw measurements are unique in that the ultranarrow bandwidth of excitation in these systems enables spectral hole-burning (SHB) and measurements within the inhomogeneous width, where we have shown in earlier work on this program that parameters such as energy relaxation and dephasing rates can vary as a function of frequency.

The electronic energy spectrum of quantum-well structures is fully quantized under a magnetic field parallel to the growth axis. Optical absorption reveals a ladder of magnetoexcitons corresponding to transitions between electron and hole Landau levels The magnetic fields also lift the Kramers degeneracy with the resultant Zeeman splitting depending on details of the band structure. The removal of this degeneracy is also expected to lead to a substantial increase of the spin-relaxation time between Zeeman-split Landau levels, since spin-relaxation can only take place via inelastic processes in the presence of a magnetic field. While the electron g-factor in GaAs heterostructures has been extensively studied, determination of the excitonic Zeeman splitting has proved to be more elusive. A precise determination of the excitonic Zeeman splitting in a quantum-well using linear optical spectroscopy is difficult, since interface disorder leads to exciton localization and an associated inhomogeneous broadening of the absorption profile. However, using frequency-domain nonlinear optical spectroscopy, we are able to directly resolve the excitonic Zeeman splitting and also provide further information on the complexities of spin relaxation in GaAs quantum-wells. The measurements reveal a *hh* splitting much smaller than that reported for bulk GaAs at low magnetic field and show a nonlinear dependence of the splitting on

magnetic field strength. The results reflect effects of the complex band structure of a quantum-well.

As we demonstrated earlier, nonlinear optical methods such as FWM have the advantage of being able to eliminate inhomogeneous broadening and accurately measure small energy separations. For GaAs/AlGaAs quantum-wells, a nearly monochromatic optical beam with σ_- circular polarization can be used to excite a narrow spectral hole (e.g., at energy E_- within the inhomogeneous absorption profile) at the *hh*(1) exciton associated with the 3/2 to 1/2 transition (see inset of Fig. 4 for the energy level diagram in a magnetic field). The width of the spectral hole is determined by the homogeneous line width. Spin-flips of electrons and holes associated with these excitons generate a spectral hole at the energy of the -3/2 to -1/2 exciton transition, designated E_+. The *spin-flip-induced* SHB at E_+ results from the reduced absorption due to the presence of carriers that have flipped their spins from E_- to E_+. This SHB can be probed using an optical beam with σ_+ circular polarization. The Zeeman splitting can then be obtained by measuring the energy difference between the spin-flip-induced SHB and the original SHB resonance.

Unfortunately such measurements are complicated by strong spectral diffusion of the localized excitons . Once created, localized excitons migrate rapidly among localization sites with different energies leading to a broad quasi-equilibrium distribution in energy, as we demonstrated earlier using FWM on this program. In the measurement described above, the nonlinear optical signal may be dominated by this distribution. In the limit that the spin relaxation time is long compared with the spectral diffusion time, nearly all spin-flipped excitons have diffused in energy. Hence, *the spin-flip-induced SHB resonance will be overwhelmed by the spectral diffusion process.* To avoid this complication, we have used a new method of FWM based on nearly degenerate four wave mixing (FWM). This method can significantly reduce the contribution of the quasi-equilibrium exciton distribution to the nonlinear optical response and allow us to resolve the spin-flip-induced SHB resonance.

The experimental configuration is again based on phase-conjugate FWM and the spectral response is measured by tuning $\mathbf{E}_3$. In the absence of spectral diffusion, the *spectral* width of the exciton grating created by $\mathbf{E}_2$ and $\mathbf{E}_1^*$ is given by the homogeneous line width, as expected from SHB, and the width of the FWM response is twice the homogeneous line width. In the presence of spectral diffusion, the spectral hole excited by $\mathbf{E}_2$ and $\mathbf{E}_1^*$ diffuses in energy, resulting in a spectral redistribution of the excitation. In this case, the FWM response arises from both the spectral hole and the quasi-equilibrium distribution of the exciton population as discussed above. The decay of the SHB population is determined by the sum of the excitonic spectral diffusion, spin-relaxation and recombination rates. However, the lifetime of the quasi-equilibrium

distribution is determined by the recombination time of the exciton. Setting $\delta = |\omega_1 - \omega_2|$ large compared with the recombination rate (but still smaller than or comparable with the spectral diffusion rate) significantly decreases the relative amplitude of the grating associated with the quasi-equilibrium distribution. Hence, the FWM response will be dominated by the SHB resonance.

In the first set of measurements, we used three circularly polarized optical beams rotating in the same direction in the laboratory frame. The nonlinear optical response, shown as squares in Fig. 4, involves only the σ_- excitons associated with the 3/2 to 1/2 transition. The width of the response corresponds to a homogeneous linewidth of 0.03 meV. The small line width confirms that the magnetoexcitons are localized and inhomogeneously broadened. As predicted, there is no contribution to the resonance from spectral diffusion as observed in the limit $\delta \rightarrow 0$.

The spin-flip-induced spectral hole is probed by reversing the polarization direction of $\mathbf{E}_3$. The resulting resonance is shown as circles in Fig. 4; the energy difference between the two peaks is the excitonic Zeeman splitting (0.19 meV at 4 T). The small signal at the original SHB position is most likely due to the residual ellipticity of the circularly polarized beams. Nonlinear signals due to the spectrally diffused excitons overwhelm the spin-flip-induced SHB for $\delta=0$. Note that spin-relaxation of σ_+ excitons requires absorption of acoustic phonons and is slower compared with spin-relaxation of σ_- excitons. The observed spin-flip-induced spectral hole is considerably weaker at 4 T when $\mathbf{E}_2$ and $\mathbf{E}_1^*$ excites σ_+ excitons.

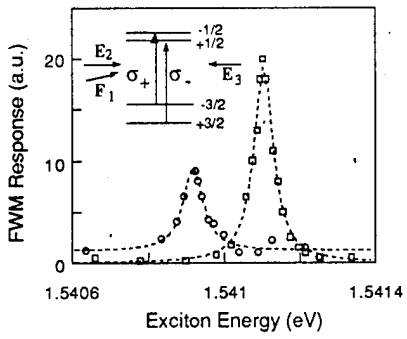

Figure 4. Spectral hole-burning FWM response at 4T. All three beams are circularly polarized with $\mathbf{E}_1$ and $\mathbf{E}_2$ exciting $\sigma-$ excitons. Squares represent when $\mathbf{E}_3$ interacts with $\sigma-$ excitons and circles represent when $\mathbf{E}_3$ interacts with $\sigma+$ excitons. Dashed lines are Lorentzian fits to the response. Inset: Conduction and heavy-hole valence band energy levels in a GaAs quantum-well.

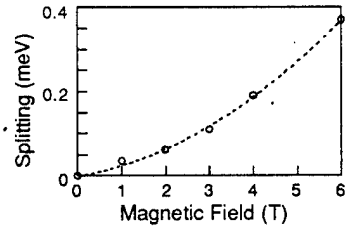

Figure 5. Magnetic field dependence of the excitonic Zeeman splitting. The dashed line is a least-square fit with a quadratic dependence.

Recent measurements have shown that at low and intermediate magnetic fields, the electronic g-factor at the lowest Landau level in GaAs quantum-wells is close to the value for bulk GaAs. In

contrast, the Zeeman splitting of the *hh*1 exciton obtained above is very small in comparison with that reported for bulk GaAs. Our results seem to be in agreement with the earlier magneto-reflectance measurements where the heavy hole Zeeman doublet was not resolved. The small Zeeman splitting attributed to the strong valence band mixing in quantum-well structures has been recently predicted by numerical calculations of magnetoexcitons using the Luttinger Hamiltonian. In particular, the mixing of σ_- excitons with excitons at higher energy pushes the σ_- exciton to lower energy. The above calculation also predicts an eventual sign change of the Zeeman splitting at higher magnetic fields where the band mixing effects overcome those of the Zeeman interaction. However, theoretical determination of the magnetic field at which the zero crossing occurs is difficult since the cancellation of the two competing contributions depends strongly on parameters of the model. The sign change of the splitting has not been observed in our measurements up to 6 T. It is interesting to note that disorder-induced localization is expected to enhance the band mixing effects if the localization scale is smaller than the exciton Bohr radius. Using the recombination rate of the localized exciton, we have estimated the localization scale to be comparable to the exciton Bohr radius.

Figure 5 displays the magnetic field dependence of the Zeeman splitting for the *hh*1 exciton. The dashed line in the figure represents a quadratic dependence. Clearly the quadratic behavior may not extend into the high-field region. The observed field dependence is somewhat surprising since the calculations predict a field dependence slower than linear. The observed dependence may be due in part to the nonparabolicity of the conduction band, which was not included in the calculations. Note that our results differ considerably from those obtained from nonlinear quantum beats in a 30-Å stepwise GaAs quantum-well. Zeeman splittings reported in the quantum beat measurement (0.5 meV at 4 T) are proportional to magnetic fields with a field strength ranging from 1 to 5 T, and are very close to those measured for impurity bound excitons in bulk GaAs . The sign of the Zeeman splitting can not be determined in quantum beat measurements. We note also that using the relative amplitudes of the two SHB resonances, we have been able to determine a spin relaxation time for the exciton of 100 psec at 4 K.

Summary

Efforts on this program have not only provided excellent educational opportunities for a number of students, but their research has lead to new understanding of the fundamental nature of optical interactions in semiconductors and semiconductor heterostructures. Their studies have provided the basis for work during the next funding period which will focus on the study of nanostructures and disorder.