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6. AUTHOR(S)

Duncan G. Steel

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

University of Michigan
Department of EECS
1106 EECS Bldg.
Ann Arbor, MI 48109-2122

8. PERFORMING ORGANIZATION REPORT NUMBER

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13. ABSTRACT (Maximum 200 words)

Work on this program focused on the development and application of advanced optical spectroscopy and methodology for the study of the quantum optical properties of semiconductor heterostructures. The work includes the first observation of the nonlinear optical polariton reflecting the complex nature of exciton-exciton interactions as well as the first observation of the light-hole-heavy-hole exciton Raman coherence. The program then moved toward the study of quantum dots, and reported the first observation of the coherent nonlinear optical response from a single quantum dot exciton as well as the first demonstration of the coherent optical control and exciton wave function engineering in a single quantum dot. The work also provides the first demonstration of optically induced Zeeman coherence and two-electron entanglement in a single dot. Finally, we developed a nonlinear optical near field scanning microscope for directly probing nano-optical structures. The first data from this system is remarkable and provides a mapping of the center-of-mass of the wave function of a localized exciton.

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Quantum Optical Studies of Semiconductors

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Principal Investigator: Duncan G. Steel
Department of Electrical Engineering and Computer Science
Department of Physics
Harrison M. Randall Laboratory of Physics
The University of Michigan
Ann Arbor, MI 48109
Phone: 734-764-4469
Email: dst@umich.edu

PUBLICATIONS

Journal Publications

1. K. B. Ferrio and D. G. Steel, "Observation of the Ultrafast Two-Photon Coherent Oscillation in a GaAs/AlGaAs Multiple-Quantum-Well: a Signature of Biexcitonic Optical Nonlinearity", , Phys. Rev. B, Rapid Comm. **54**, ppR5231-5234 (1996).
2. D. C. Kilper, D. G. Steel, R. Craig, D. Scifres, "Polarization dependent noise in photon-number squeezed light generated by quantum-well lasers," Optics Letters **21**, pp 1283-1285 (1996).
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A. C. Schaefer, J. Erland and D. G. Steel, "Nondiffusive Excitonic Transport in GaAs and the Effects of Momentum Scattering", Physical Review B Rapid Communications **54**, pp R11046-R11049 (1996).

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5. Michael Lewis, Peter Wolanin, Ari Gafni, Duncan Steel, "Near-Field Optical Microscopy of Single Molecules by Femtosecond Two-Photon Excitation," Optics Letters **23**, 1111-1113(1998).
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7. Nicolas H. Bonadeo, John Erland, D. Gammon, D. Park, D.S. Katzer, D.G. Steel, "Coherent Optical Control of the Quantum State of a Single Quantum Dot" Science **282**, 1473 (1998).
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9. N.H.Bonadeo, D.G. Steel, R. Merlin, "Anomalous selection rules and heavy-light hole beats: stress effects in GaAs," in press, Physical Review (1999).
10. N. H. Bonadeo, A. S. Lenihan, Gang Chen, J. R. Guest, D. G. Steel D. Gammon, D. S. Katzer and D. Park, "Single Quantum Dot States Measured by Optical Modulation Spectroscopy," in press, Appl. Phys. Lett. (1999).
11. J. Erland, J.C. Kim, N.H. Bonadeo, and D.G. Steel, D.S. Katzer, and D. Gammon, "Non-exponential Photon Echo Decays from Nanostructures: Strongly and Weakly Localized Degenerate Exciton States," in press, Phys. Rev. B, Rapid Communications (1999).

Invited Conference Papers

1. D.G. Steel, "Quantum Optics in Semiconductor and Heterostructures", Plenary Talk, APS May '97 Meeting.
2. Nicolas Bonadeo, Dan Gammon, D.G. Steel, "Quantum optics of a single exciton", ILS/OSA Annual Meeting, '97.
3. Duncan Steel, "Near Field Spectroscopy of Single Biomolecules," Microscopy and Microanalysis Society, 1997.
4. Duncan Steel, "Nano-Nonlinear Optics", Symposium on Single Molecule Spectroscopy, Washington, 1997.
5. N. H. Bonadeo, J. Erland, Gang Chen and D. G. Steel, "Coherent Optical Excitation of Single Excitons in Quantum Dots," invited paper IQEC'98, OSA Technical Digest 7 , p161-162 1998).
6. D. Gammon, N. Bonadeo, D.G. Steel, E.S. Snow "Optical Nano-Spectroscopy of Single Qdots", OSA Topical on Dephasing in Semiconductors, 1998.
7. Nicolas Bonadeo, D.G. Steel, D. Gammon "Nonlinear Nano-Optics: Probing one exciton at time", APS March Meeting, 1998.
8. D. Gammon, N.H. Bonadeo, J. Erland, D.G. Steel, S.W. Brown, T.A. Kennedy, E.S. Snow, B.V. Shanabrook, D.S. Katzer, D. Park. "Optically Probing and Controlling Single Quantum Dots" International Conf.. on the Physics of Semiconductors (ICPS'98)), Israel(1998).
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10. Gang Chen, N. H. Bonadeo, E. A. Tabak, D. Gammon, D. S. Katzer, D. Park and D. G. Steel, " Magneto-Optical Studies of Excitons in Single GaAs Quantum Dots," QELS'99 (1999).
11. D.G. Steel, N.H. Bonadeo, J. Erland, E.S. Snow, D.S. Katzer, D. Gammon, "Coherent Control, Wave Function Engineering, and Nonlinear Optics in a Single Quantum Dot," invited paper, Centennial Meeting, APS, March 1999.
12. Jeff Guest, D. Gammon, N.H. Bonadeo, J. Erland, D.G. Steel, E.S. Snow, D.S. Katzer and D. Park, "Localized Excitons: Probing One Quantum Dot at a Time" MRS-99.

Contributed Conference Papers

1. N.H. Bonadeo, R Merlin and D. G. Steel, "Ultrafast Electric Field Polarization Evolution in the Excitonic Free Polarization decay of GaAs," QELS'96.
2. K.B. Ferrio, J.R. Guest, and Duncan G. Steel, "Anomalous decay and polarization of electronic Raman coherence in GaAs," QELS'96,.

3. A.C. Schaefer and D. G. Steel, "The effects of momentum scattering on exciton motion: Observation of non-diffusive transport," QELS'96.
4. D C. Kilper, A. C. Schaefer, J. Erland, and D. G. Steel, "Coherent nonlinear optical spectroscopy using photon-number squeezed light" QELS'96.
5. A.C. Schaefer, D.C. Kilper, J. Erland, D.G. Steel, "Nonlinear optical spectroscopy using photon-number squeezed light and the investigation of excitation induced noise," APS-DAMOP (1996).
6. Kyle Ferrio, Duncan Steel, "State-specific scattering of electronic Raman coherence in GaAs," APS-DAMOP (1996).
7. D.C. Kilper, P.A. Roos, J.L. Carlsten, A.C. Schaefer and D.G. Steel, "Quantum polarization dynamics in semiconductor lasers", presented at "Polarization Effects in Laser and Spectroscopy" (PELS'97) to be held at the University of Toronto on 24-28th May 1997
8. J.C. Kim, G. Chen, A. C. Schaefer, N. Bonadeo, D. Gammon and D. G. Steel, "Strongly Localized Excitons in a Narrow Single Quantum Well: Ensemble Dynamics of Single-Quantum-Dot Excitons," QELS'97 OSA Tech. Dig. **12**, pp 148-149 (1997).
9. A.C. Schaefer, N. H. Bonadeo and D. G. Steel, "The Coherent Nonlinear Optical Response of the Exciton-Polariton," QELS'97 OSA Tech. Dig. **12**, pp 16-17 (1997).
10. H. Bonadeo, D. Gammon and D. G. Steel, "Resonant Nonlinear Optical Response of a Single Quantum Dot," QELS'97 OSA Tech. Dig. **12**, pp 64-65 (1997).
11. N.H. Bonadeo, A. S. Lenihan, D. Gammon and D. G. Steel, "Single quantum dot-like excitonic states measured by optical modulation spectroscopy," QELS'97 OSA Tech. Dig. **12**, pp 16-17 (1997).
12. J.C. Kim, G. Chen, A.C. Schaefer, N.H. Bonadeo, D.G. Steel, D. Gammon, "Dynamics of strongly localized excitons," Bulletin Am. Phys. Soc. **42**, p70 (1997).
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14. A.C. Schaefer, N.H. Bonadeo, D.G. Steel, "Nonlinear optical response of the exciton-polariton due to exciton-exciton interactions," Bulletin Am. Phys. Soc. **42**, p204 (1997).
15. Gang Chen, Anne Schaefer, Dan Gammon, Duncan Steel, "Disorder Induced Interference in Exciton Decay Dynamics," IQEC'98 OSA Technical Digest **7**, p225-226 (1998).
16. J. Erland, J.C. Kim, D. Gammon, D.G. Steel, "Biexponential decoherence in photon echoes: Spectral evidence for nearly degenerate localized excitons." , IQEC '98, OSA Technical Digest **7** p. 210-211 (1998).
17. M.K. Lewis, P. Wolanin, A. Gafni, D.G. Steel, "Two-Photon Induced Fluorescence Imaging of Single Molecules Using Near-Field Scanning Optical Microscopy" , IQEC '98, OSA Technical Digest **7** p227-228 (1998).
18. J. Erland, N.H. Bonadeo, D. Gammon, and D.G. Steel, "Coherent Control of a Quantum Dot Exciton Wave Function", EQEC 98, 14-18 September 1998, Glasgow, Scotland, Post Deadline Paper EPD2.5.
19. J.C. Kim, J. Erland, D.G. Steel, and D Gammon, "Exciton Dynamics in Disordered Energy Landscape," APS March Meeting (1998).
20. D. Gammon, S.W. Brown, T.A. Kennedy, E.S. Snow, Gang Chen, N.H. Bonadeo, D.G. Steel"Magneto-optical spectroscopy of single quantum dots: electron and nuclear spin," APS March Meeting (1999).

21. J. R. Guest, T. H. Stievater, A. S. Lenihan, Gang Chen, D. Gammon,, D. S. Katzer, D. Park, D. G. Steel, "Nano-Optics: Imaging the Resonant Nonlinear Response of Individual Localized Excitons," QELS'99 (1999).
22. N.H. Bonadeo, Gang Chen D. Gammon, D. Park, D. S. Katzer, and D. G. Steel, "Single Quantum Dot States: Energy Relaxation and Coupling," QELS'99 (1999).
23. A.C. Schaefer, D.G. Steel, "Non-Diffusive Transport of Excitons in GaAs and the Effects of Momentum Scattering" American Physical Society, March Meeting, 1996.

Educational Activity

A number of students participated in the program as evidenced in the above publications. Four of the students have since graduated with a Ph.D and two are now faculty members in tenure track positions. The others have taken postdoctoral positions including one at AT&T. Two other students will be graduating within the next 18 months, including the student who has developed the low temperature nonlinear optical NSOM. Three new students have just joined the group and will be involved in the new program.

Summary of Findings

All of the research finding presented in this report have been reported in the annual reports. However, for completeness, we summarize the most important results.

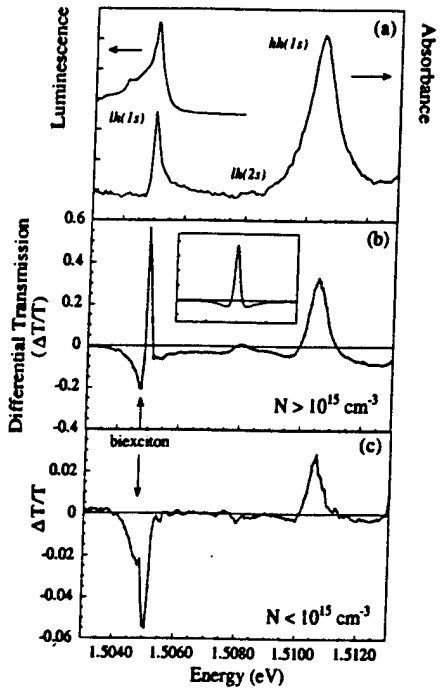
Introduction

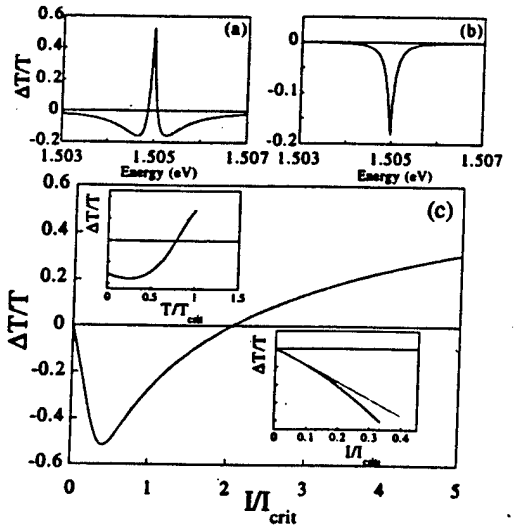
The objectives of the present program focus on developing and applying quantum optical methods based on coherent nonlinear laser spectroscopy to the study of nanoscopic physics in semiconductors, focusing explicitly on the optical behavior. The physics includes the study of disorder in heterostructures and the study of quantum dots (QD). The experimental approach is presently based in large part on the use of far field optical techniques, on both apertured and unapertured GaAs/AlGaAs single quantum well samples. However, a major effort exists now in the laboratory to develop a *low temperature* near field spectroscopy approach to allow us to study disorder and quantum dots without the complications that accompany sample aperturing. The success in this area has been very exciting.

The First Observation and Theory for the Nonlinear Exciton Polariton

As part of a carry over of the previous work and because the understanding of optical interactions in even bulk semiconductors remains limited, we completed a set of experiments to study the lowest order nonlinear optical effects in semiconductors. The work was motivated by our observation of what appears to be highly anomalous behavior in the nonlinear transmission. Specifically, the data (Fig 1 and 2) shows in Fig 1c that at the lowest intensity, the absorption increases at the light hole exciton (and even the heavy hole exciton at lower intensities). The lower energy shoulder is in fact due to the biexciton and is expected, but the optical induced increase in absorption was not predicted by any theory at that point. Following the hint however,

that the linear absorption line shape of the lh-exciton in Fig. 1a was itself indicative of polaritonic effects, we returned to a fully self consistent solution of the modified optical Block equations (including effects of dynamic exciton-exciton interaction leading to excitation induced dephasing reported earlier) and Maxwell equations, thus insuring polaritonic effects would be included. The results of the theory are shown in Fig. 2. The agreement between theory and experiment was remarkable and accounted for every aspect of the experimental data. The data now clearly shows the first observation of the nonlinear exciton polariton.





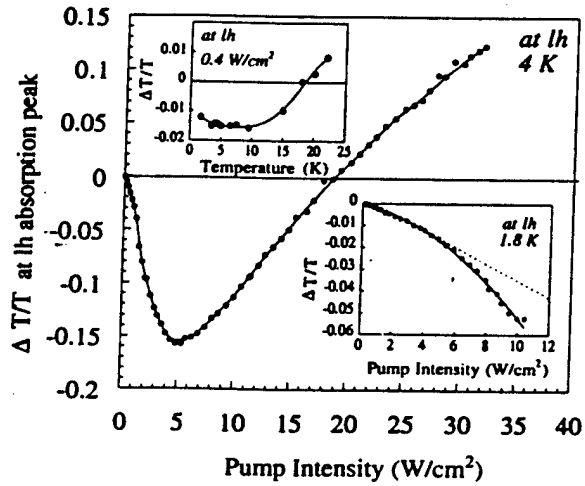


Figure 1 (Upper left) (a) Absorption and luminescence spectrum of 0.2 μm bulk GaAs. Full width of lh(1s) resonance is 0.2 meV. (b) and (c) Differential transimssion (DT) at high and low intensity Pump was resonant with lh(1s).

Figure 2 (Left) Behavior of DT at peak of lh resonance at 4 K as a function of pump intensity. Lower inset: Superlinear behavior of lh(1s) DT with increasing pump intensity. Upper inset: Dependence of induced absorption signal with temperature.

Figure 3 (Upper right) Calculated DT based on the classical polariton model with spatial dispersion for increased damping due to increased exciton density. Spectra of DT for (a) $\gamma/\gamma_{crit} = 4$, (b) $\gamma/\gamma_{crit} = 0.4$. (c) Behavior of DT at absorption peak as a function of pump intensity. Lower inset: Superlinear behavior of DT for pump and probe at *same* resonance. Upper inset: Theoretical dependence on temperature, T. In above, I_{crit} and T_{crit} are constants defined as the values at which $\gamma=\gamma_{crit}$, calculated with the zero density and temperature absorption coefficien.

Detection of the Raman Coherence and Exciton-Exciton Correlations

While the simple atomic picture of the coherent nonlinear optical properties easily accounts for physical phenomena such as two-photon exciton and Raman and Zeeman coherences, such behavior in semiconductor structures is more complex because the effective anharmonicity in this system arises from exciton-exciton interactions or correlations. Clear observation of the latter two phenomena have not been made, while the former feature is of course associated with the biexciton. In some systems, the biexciton may not exist as a sharply defined bound state, and hence the presence of exciton-exciton correlations is not readily observable in the multiphoton absorption spectrum. More important, however, is the fact that Raman and Zeeman coherences are interesting from the standpoint of quantum information devices since these states represent two-electron entanglement.

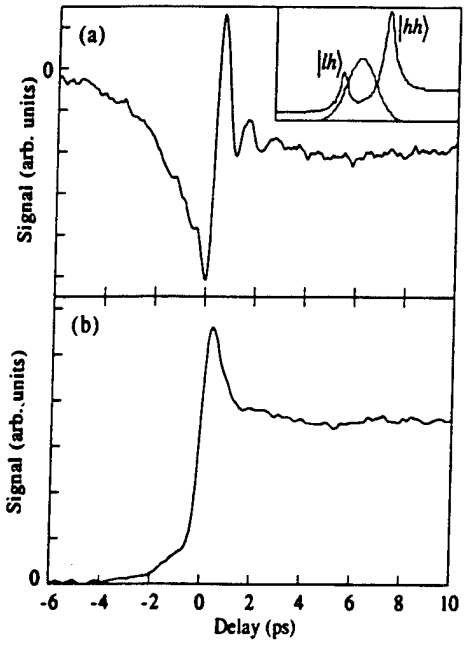


Figure 4. (a). Raman coherence oscillations appear in homodyne-detected FWM for the tuning shown in the inset. The pulse bandwidth is 2 meV, and the total excitation density is 9×10^{15} cm^{-3}. (b). For a slightly different tuning, the beats are suppressed. (The overall sign of the DT response changes from (a) to (b), reflecting the sign change in EID as the laser is tuned closer to the lh exciton resonance.)

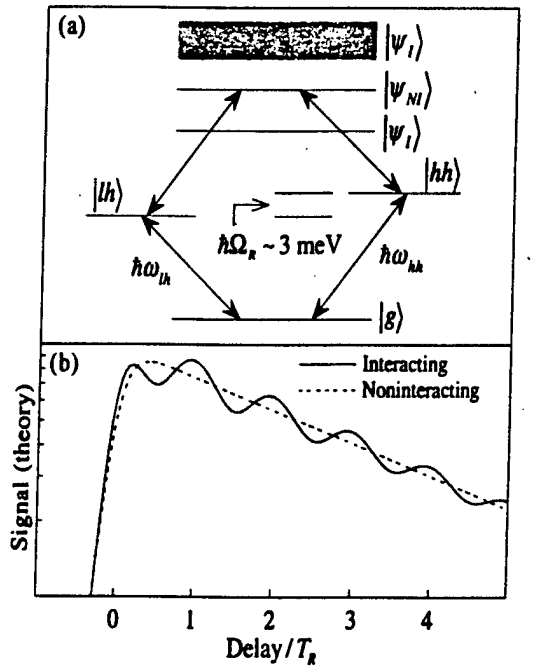


Figure 5. The *lh-hh* interaction produces bound or scattering states $|\psi_I\rangle$ (a) and produces Raman quantum beats (b: solid), which would be absent for noninteracting excitons (b: dashed). Energy differences have been greatly exaggerated for clarity. The Raman period and frequency are related by $T_R\Omega_R = 2\pi$.

As in the previous section, these experiments were performed in high purity bulk GaAs to avoid complications due to disorder. These experiments were designed to directly detect the lh-hh exciton Raman coherence. While the experiments resemble "traditional quantum beat"

experiments, the experiments are in fact profoundly different. This experimental configuration was designed to measure the time evolution of the phase optically induced at second order which is where the Raman coherence is to be seen. This is in contrast to traditional FWM experiments which report on time evolution of the system at odd order of optical interactions and report on dipole coherences. The data (Figure 4) shows clear evidence of the presence of exciton-exciton interactions leading to correlations as seen by the homodyne detected time evolution of the quantum phase of the system. In Fig. 5a, a simplified 2-electron model is provided which shows the result of the perturbation solution of the density matrix equations. Only in the presence of exciton-exciton interactions are the beats observed in the measurement as seen in the model prediction of Fig. 5b.

These results were crucial to our understanding and development of experiments in quantum dots discussed below.

The Coherent Nonlinear Optical Response of a Single Quantum Dot Exciton

In this part of the program, we changed the direction of our work to initiate experiments designed to push the state-of-the-art in the area of nano-optics. It is clear that as device scale lengths continue to decrease, the quantum size regime is becoming the domain of modern devices.

In this initial study, we worked to obtain the coherent nonlinear optical response of a single exciton in a quantum dot in order to combine the power of coherent nonlinear optical spectroscopy with the new technology for probing individual QD's. The first dots studied have been so-called natural dots formed due to strong localization, however, future dots are likely to be much higher quality with improved control over size and location.

Narrow quantum wells of the type described above result in strong localization of excitons and leads to energy level structure because of lateral confinement expected from quantum dot states. To study these systems at the single exciton level, we first used samples over coated with a layer of aluminum and then created small apertures in the aluminum ranging in size from 25 to 0.2 microns. (See further progress below using near field scanning microscopy.) The work has been done in collaboration with researchers at NRL. The smallest apertures provide spatial resolution so that under one aperture, only a few localized exciton states exist. Using the high energy resolution of coherent nonlinear spectroscopy, we were then able to differentiate between the different dot states. Based on this, we have now refined our measurements and have achieved some relatively spectacular results as shown in Fig. 6. In particular, *we have achieved the first coherent nonlinear optical study of single excitons localized in GaAs QD.* The measurements are based on homodyne detection of the coherent emission arising from the third order nonlinear optical response and show both an incoherent and coherent contribution, the latter arising from population pulsations at frequencies above 10 GHz and leads to the onset of two-beam coupling. The data shows the decoherence time of the excitation is of order 30 psec with a comparable energy relaxation time.

The fully degenerate (i.e., $\omega_1 = \omega_2$) nonlinear response as a function of frequency is shown in Fig. 6 obtained using frequency locked high resolution scanning cw lasers. This data was taken through a 0.5 μm aperture at 5 K with low spectral resolution (~50 μeV). The sharp

narrow lines of the PL, correspond to isolated QD excitons. In many cases, resonances seen in the PL are also seen in the nonlinear spectrum. However we note that, as in the case with PL and PLE, there are differences between the spectra which reflect important details of the electronic excitation. Many of the resonances in the degenerate nonlinear spectra can be well fit to a Lorentzian squared (right inset) as expected from the analysis for isolated homogeneously-broadened excitons. We note, however, that some of the spectral features appear more complex than that observed in the inset. At this point, it is unclear whether these features are intrinsic or the result of interference effects from the background nonlinear response, however, they remain under investigation. The signal strength of the response goes linearly with intensity I_1 because the homodyne-detected lowest order contribution to the nonlinear response corresponds to the third order susceptibility. The left inset of Fig. 6 shows this behavior with saturation at high powers which also leads to power dependent spectral linewidths

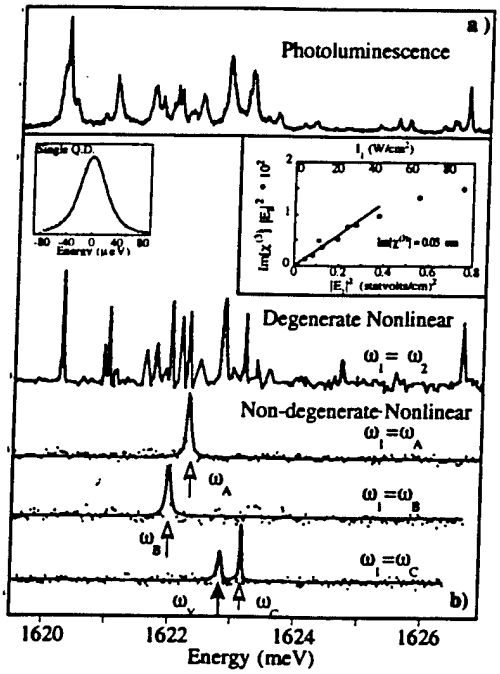


Figure 6. A comparison of the PL spectra with the fully degenerate and non-degenerate homodyne detected four-wave mixing spectra. The degenerate nonlinear spectra shows a strong similarity with the PL data, as expected. However, the non-degenerate spectrum shows the fully resonant nature of the nonlinear response. In this case, a strong resonance is observed at the pump frequency and the remaining resonances are suppressed except one lower energy resonance. The lower energy resonance appears to arise from a nearby dot that is excited by energy transfer. The left inset shows the linear dependence on pump power, expected of a third order nonlinear response in this geometry. The right inset shows a nonlinear least squares fit of the they to an isolated resonance.

Additional data (not shown) of the high resolution non-degenerate response shows the system has very little pure dephasing contributing to the linewidth indicating that these dots are extremely robust against pure dephasing processes, a result that may prove important in future applications.

The Magneto-Nonlinear Optical Response of a Single Quantum Dot Exciton: Detection of the Zeeman Coherence and 2-Electron Entanglement

Following our work on generation and detection of Raman coherence in bulk GaAs, we believed we could generate and detect the Zeeman coherence in a single quantum dot. Unlike the measurements above, we used high resolution cw methodology to achieve this result and demonstrate two-electron entanglement.

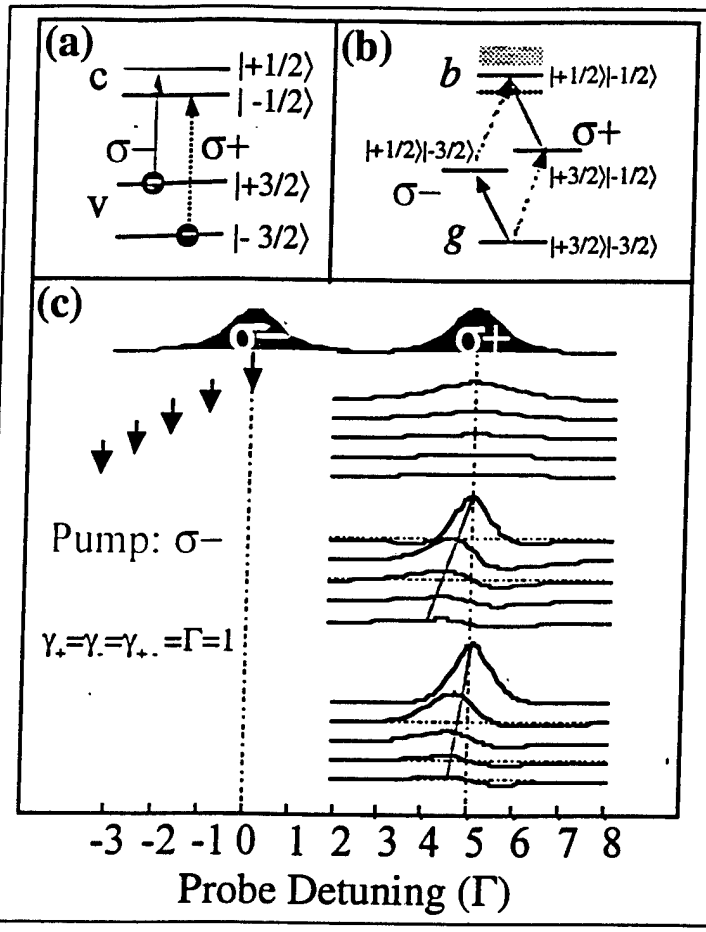


FIG. 7. (a) Band diagram for heavy-hole exciton transitions. (b) Four-level model for incorporating the four-particle Coulomb correlation into the problem. (c) Theoretical prediction for the non-degenerate experiment, assuming that the contribution from the levels beyond single-exciton levels can be neglected due to the Coulomb interaction. Upper curves show the incoherent contribution from the ground state depletion. Middle curves show the coherent contribution from the second order Zeeman coherence. Lower curves are the superposition of both contributions. In the absence of correlation, no signal at all would be observed.

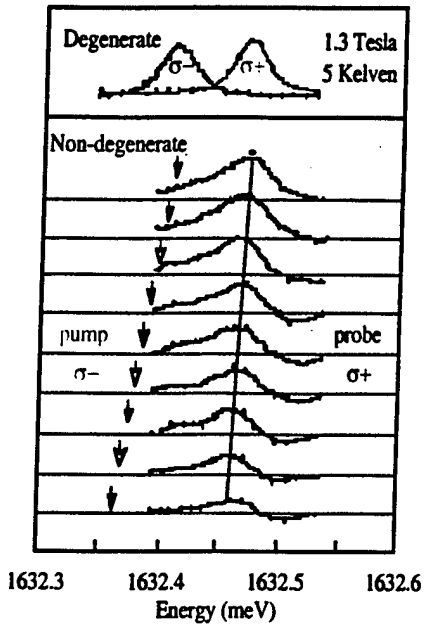


FIG. 8. Non-degenerate coherent nonlinear response. The pump is placed at the σ- (lower) state with σ- polarization. The probe is scanned across the σ+ state (upper) with σ+ polarization. The result shows interference lineshape identifying the Coulomb correlation and the contribution from Zeeman coherence (see text for details).

In this system, the correct minimal description is that of two electrons described by a basis state formed by correctly antisymmetrized eigenfunctions. Figure 7a shows the representation based on zone center conduction and valence band electrons. In the presence of a strong magnetic field, symmetry is restored as well as the usual optical selection rules (see further discussion below). Figure 7b shows the equivalent exciton representation formed by accounting for the antisymmetrization of the wave function in the two-electron basis. The results of a simple analysis of the coherent nonlinear response based on the density matrix master equations is given in Fig. 7c where the pump field at $E_1(\omega_1)$ is tuned to the σ- transition and probe field $E_2(\omega_2)$ is tuned to the σ+ transition. In the case of no interaction between the two electrons, there is no signal in the region of the σ+ transition. In the presence of just ground state depletion effects (saturation), the upper curve shows a simple resonance located at the center of the σ+ transition which does not move with tuning of ω_1. However, the second curve shows the effect of including the Zeeman coherence between the two levels. The Zeeman coherence represents a coherent-superposition of the two 2-electron states excited by the σ+ and σ- optical fields. This

superposition represents an optically induced entanglement of the two states. The final curve represents the expected coherent nonlinear optical response in the presence of both contributions. The experimental result is quite profound and is shown in Fig. 8. The data clearly shows the unmistakable signature of optically induced entanglement and Zeeman coherence. The results show that our understanding of this system is apparently correct and that we have now established a means to demonstrate entanglement. Our future work will focus on entanglement of two dots.

Coherent Control and Wave Function Engineering of a Single Quantum Dot Exciton

A part of the new thinking in coherent optical interactions is the idea that coherent optical radiation can be used to control and engineer the state of excitation of a quantum system. Using the results of our understanding based on the nonlinear optical spectroscopy discussed above, we have shown that we can extend the concepts of coherent control and wave function engineering developed in atomic/molecular systems and higher dimensional semiconductor structures to the limit of a single quantum system in a zero-dimensional quantum dot. Such proposals have been envisioned for implementation of various schemes for quantum computation and coherent information processing and transfer in which it is important to address and coherently control individual quantum units.

The excitonic wave function was manipulated and monitored on a time scale short compared to the loss of quantum coherence by controlling the optical phase of two picosecond pulses through timing and polarization. An exciton in an isolated QD was probed by exciting through a 500-nm diameter Al aperture. The photoluminescence (PL) and photoluminescence excitation (PLE) spectra exhibit atomic-like spectra (Fig. 9a) similar to that discussed above. For these experiments, we accounted for the fact that symmetry breaking due to the geometry of the quantum dot results in a change of the dipole selection rules and a fine structure splitting of the excitonic state into x and y states of polarization. The experiments concentrated on the $|E_1\rangle$ state that shows a linewidth of 17 μeV and a fine structure splitting of 60 μeV (Fig 1a, inset). We probed the state of the system by monitoring the luminescence from $|E_0\rangle$. Figure 9b shows the luminescence intensity as a function of the delay time between the phase-locked pulses (Y-polarized) and represents the autocorrelation function of the excitonic wavefunction corresponding to state $|E_{1Y}\rangle$. The exponential decay arises from the loss of coherence. Even more interesting is the behavior shown in Fig. 1c when a non-stationary wavefunction composed of a coherent superposition of $|E_{1X}\rangle$ and $|E_{1Y}\rangle$ states was created and measured by a pulse sequence polarized at 45°. The autocorrelation shows the wavefunction oscillating between two orthogonal states, $|E_{1X}\rangle+|E_{1Y}\rangle$ and $|E_{1X}\rangle-|E_{1Y}\rangle$. The oscillation period corresponds to the inverse of the difference frequency between the two optical transitions. In the third experiment we created a superposition of the states that had a π shift in the quantum phase with respect to the reference wavefunction (see Fig 9d). This was accomplished by polarizing the first beam -45° and the second beam at +45° and represents a cross correlation between the different excited states produced by the two pulses.

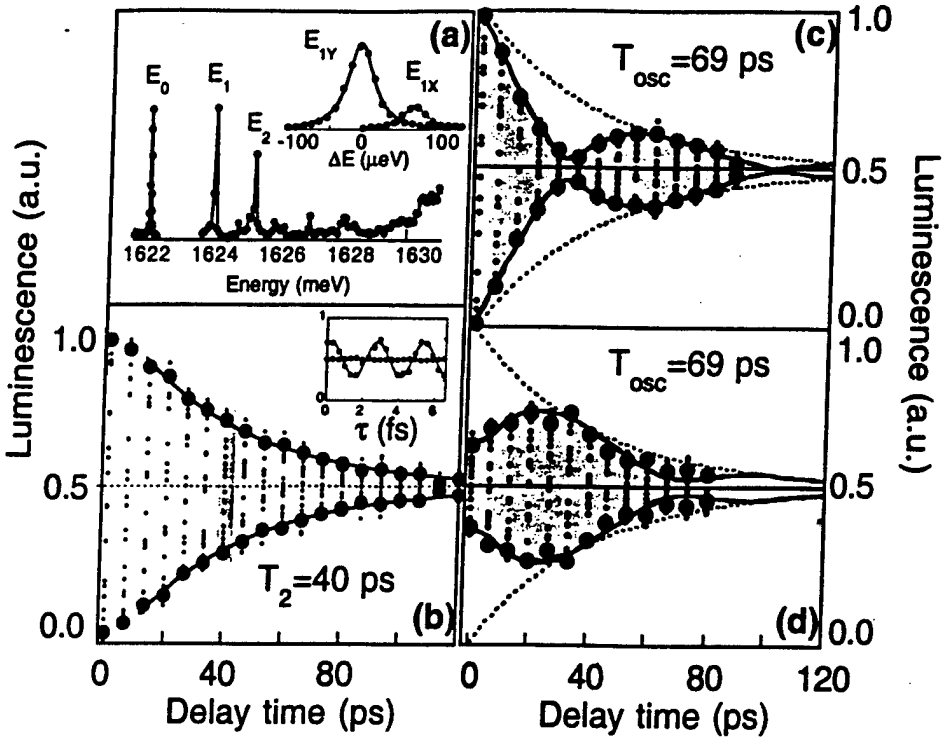


Fig. 9 (a) PL (filled circles) and PLE (open circles) spectra of the $|E_0\rangle$ state. Inset: $|E_1\rangle$ fine structure shows the splitting of the ($|E_{1X}\rangle$) and ($|E_{1Y}\rangle$) states. (b) The amplitude of the oscillation in PL as a function of delay (large filled circles), for Y-polarized pulses. The quantum interferogram measures the autocorrelation function of the excited state wavefunction. The inset shows an expanded view around τ=40 ps (corresponding to the shadowed region) showing the oscillations in PL on a femtosecond time scale. The large filled circles in the main figure are determined from a fit of the amplitude of oscillations. (c) The auto-correlation function of the excited state wave function for both pulses co-polarized and rotated to equally excite both the $|E_{1X}\rangle$ and $|E_{1Y}\rangle$ states. (d) The cross-correlation function between two excited state wave functions generated by orthogonally polarized optical pulses. The relative phase of the two superposition states produced by each pulse differs by π.

The measurements show coherent optical control of the quantum state of a single dot and, in the case of Fig. 9d, the feasibility of generating a simple target wave function. This work establishes the basic tools for developing more sophisticated control and creating a more complex wave function as achieved in atomic systems.

Development and Application of a Low Temperature Near Field Scanning Optical Microscope (NSOM) for Nonlinear Optical Spectroscopy: A Direct Measurement of the Center of Mass of the Localized Excitonic Wave Function

A major objective of the past several years has been to develop a methodology that allows us to probe individual quantum dot structures at high density without resorting to apertures. To this

end, we developed a low temperature near field scanning microscope (NSOM) designed to work based on detecting the coherent nonlinear optical response. Our first high resolution measurements are shown below. The results are quite profound and demonstrate the complexity of the localization of the dots in highly disordered systems. Because the measurements are based on the nonlinear optical response, we excite and probe the same optical dipole and do not rely on either energy or spatial relaxation as occurs in all other previous NSOM measurements. *The data in Fig 10 actually provides the first mapping out of the wave function of such a system.* The experimental setup for the optics is similar in concept to that needed to obtain the degenerate FWM response in Fig. 6. However, in practice, it is much complicated because the optical arrangement is in a fiber network rather than in free space on the table. A more detailed discussion will be presented in a preprint of a manuscript that is nearly completed.

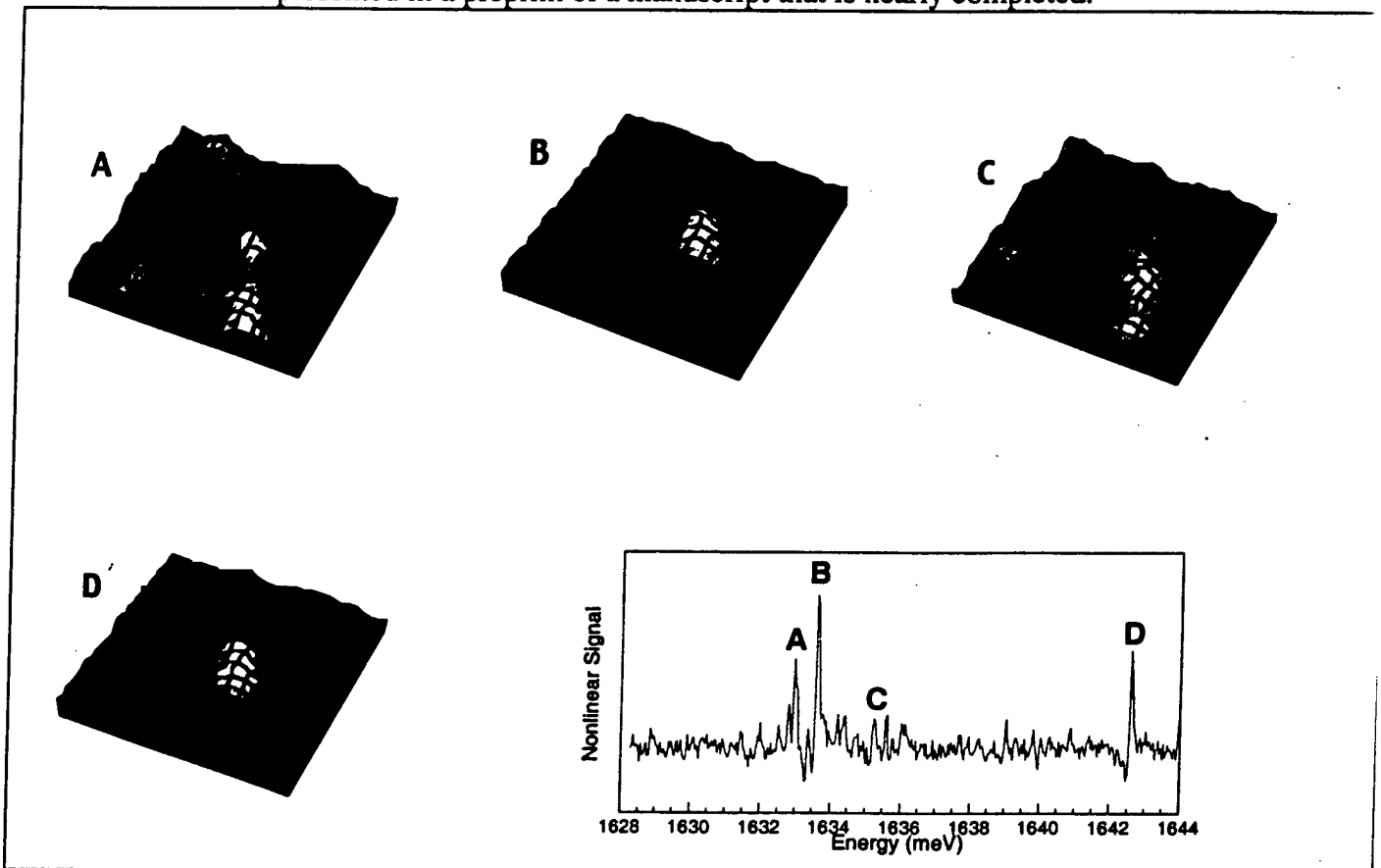


Figure 10: Degenerate nonlinear optical response images taken over 2 μm x 2 μm area at energies indicated in the spectrum (which was taken near the center of the images). Red-yellow represents positive nonlinear signal (or induced transmission), and blue represents negative nonlinear signal. Images are scaled independently. A) 1633.0 meV w/ relative signal of 2.14 B) 1633.6 meV w/ relative signal of 6.37 C) 1635.3 meV w/ relative signal of 1.55 D) 1642.7 meV w/ relative signal of 3.72. These images can be considered mappings (within the resolution of the probe) of the center-of-mass wavefunction $|\psi(x,y)|^2$ of the exciton in the disordered 2-D potential of the quantum well