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13. ABSTRACT (Maximum 200 words)
Work on this program is aimed at developing and understanding nano-optical structures with the specific goal of developing and applying quantum otpical methods to characterize and manipulate the quantum states of these systems. The measurements have resulted in a number of publications which demonstrate key features of new quantum mechanical structures. These features include optically induced and detected quantum entanglement of two exciton states as well as a demonstration of a classical Bell state, a Rabi oscillations corresponding to full coherent control of the quantum state of a single quantum dot. The measurements were then extended to show optically induced and detected quantum entanglement in self assembled quantum dots through the detection of Raman quantum beats. Finally, we developed and demonstrated the first low temperature near field optical microscopy using coherent nonlinear optical spectroscopy techniques to directly probe the transition dipole and map out the center of mass motion of an excitonic wave function.

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THE AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

Nano-Optics: Coherent Nonlinear Optical Response and Control of Single Quantum Dots

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Abstract

Work on this program is aimed at developing and understanding nano-optical structures with the specific goal of developing and applying quantum optical methods to characterize and manipulate the quantum states of these systems. The measurements have resulted in a number of publications which demonstrate key features of new quantum mechanical structures. These features include optically induced and detected quantum entanglement of two exciton states as well as a demonstration of a classical Bell state, and Rabi oscillations corresponding to full coherent control of the quantum state of a single quantum dot. The measurements were then extended to show optically induced and detected quantum entanglement in self assembled quantum dots through the detection of Raman quantum beats. Finally, we developed and demonstrated the first low temperature near field optical microscope using coherent nonlinear optical spectroscopy techniques to directly probe the transition dipole and map out the center of mass motion of an excitonic wave function

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EDUCATIONAL ACTIVITY

A number of students participated in the program as evidenced in the above publications. Five of the students have since graduated with a Ph.D and gone on to postdocs or permanent positions including one student taking a permanent position at Lucent and another student joining NRL. Several new students have joined the group and will be involved in the new program.

COLLABORATIONS

The work in the program is the result of an intense collaboration with Dr. D. Gammon at The Naval Research Laboratory supported by DARPA to develop quantum dot structures and spintronic based devices. In addition, the manybody theory component of the analysis of our findings is supported through our collaboration with Professor L.J. Sham (UCSD), supported by ARO and NSF. Our work has also benefited by our collaboration with Professor P.K. Bhattacharya (U. Mich.) who is supported by several agencies to develop self-assembled quantum dot structures.

SUMMARY OF FINDINGS

Nearly all of the research findings presented in this report have been reported in the annual reports. However, for completeness, we list all the major developments that resulted in publications, and we review a few of the most important results.

Introduction

The objectives of this program focused on developing and applying quantum optical methods based on coherent nonlinear laser spectroscopy to the study of physics in nanoscopic semiconductors. The physics included the study of disorder in heterostructures and then shifted to what is now the primary focus of the next program (AFOSR Grant No. F49620-01-1-0502), namely the study of quantum dots (QD). The experimental approach is based primarily on various near field optical techniques, including both apertured and unapertured GaAs/AlGaAs single quantum well samples as well as exploiting our development and application of the first low temperature coherent nonlinear optical microscope based on scanning near field microscopy methodology. These results have just now been published in *Science*. The program emphasis has shifted with knowledge and developments in the research community to now include a major effort at developing nano-structures for application to *quantum based devices, including quantum computing and nano-structure based quantum optics such as photon-on-demand sources and optically controlled spintronic systems.* It is these latter areas which form the core of the current program

Summary of the most important achievements:

Demonstration of optical modulation spectroscopy for the measurement of single quantum exciton oscillator strengths (*Appl. Phys. Lett.* **75**, 2933 (1999)).

Demonstration of optically induced two exciton state entanglement and detection in a single quantum dot (*Science*, **289**, 1906 (2000)).

Development of the first low temperature near field optical microscope using a new coherent nonlinear optical spectroscopy technique to provide the first direct measurement of optical excitations (*Science*, **293**, 224 (2001)).

Demonstration of Rabi oscillations in a single quantum dot corresponding to one qubit rotations (*Physical Review Letters.* 87, 133603-1 (2001)).

Demonstration of coherent optically manipulation of the quantum dot biexciton and quantum entanglement (*Physical Review Letter* **88,** p117901 (2002).

Demonstration of quantum entanglement and Raman coherence between excitonic states in self-assembled quantum dots (in press, *Physical Review Letters*, 2002).

Direct measurement of the transition dipole in a single quantum dot (Submitted, *Physical Review Letters).*

Demonstration of optically induced and detected exciton entanglement in single quantum dots

Entanglement is one of the most spectacular peculiarities of quantum mechanics that contrast with classical physics. While it can occur between separate degrees of freedom of a single particle, only inter-particle entanglement bears the properties of nonlocality that is essential for quantum information manipulation. Entangled states involving photons or massive particles have been produced by various experimental means.

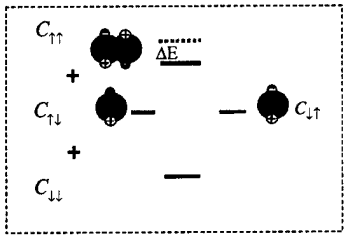

Figure 1. Exciton energy level excitation scheme representing two interacting exciton systems for studies of two-exciton state entanglement and Rabi oscillations.

However, for eventual application, it is ultimately highly desirable to be able to work with these systems in a solid state environment because of the great wealth of technology and relative ease of manipulation and fabrication of solid state devices which would potentially enable integration of both the quantum solid state component with the necessary optical sources and detectors.

In this program, we have succeeded in producing and detecting, for the first time, an entangled state of two exciton-states localized in a single gallium arsenide (GaAs) QD, demonstrating an important first step toward the goal of entangling excitons confined to two or more semiconductor QD s. We have demonstrated entanglement involving the optical Block vectors of the two spin polarized exciton states as well as entanglement between the ground state of the dot and the biexciton state.

The system we are working with can be represented in the energy excitation level scheme shown in Fig. 1. In this work, the different exciton transitions (the red and green, and two exciton red-green state) are in a single dot, but work is underway to demonstrate this behavior in coupled quantum dots. Working in a single dot, the experiments are somewhat easier, and serve as a model for studies on the more complex but interesting system of coupled dots. It is of interest to show that we can create nonfactorizable excited state wave functions, e.g., $|\psi\rangle = C_{\downarrow\downarrow}|\downarrow\downarrow\rangle + C_{\uparrow\uparrow}|\uparrow\uparrow\rangle$ or $|\psi\rangle = C_{\downarrow\uparrow}|\downarrow\uparrow\rangle + C_{\uparrow\downarrow}|\uparrow\downarrow\rangle$.

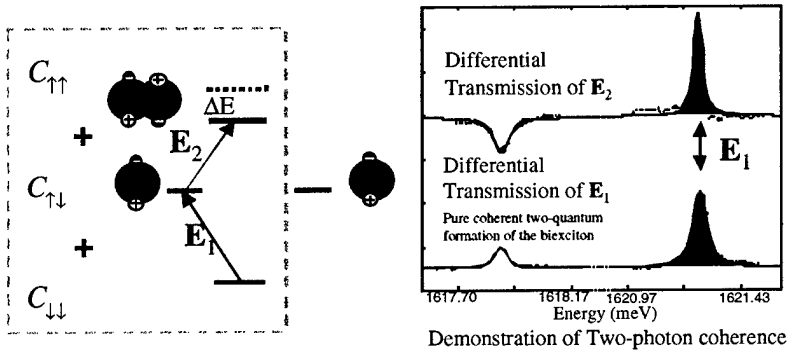

Figure 2. Experimental data showing first the optical excitation and manipulation of the resonantly excited biexciton state within a single dot (upper trace) and the data showing production of the entangled state (lower trace).

Single quantum dot states were studied using apertured samples of interface fluctuation dots provided by our collaborators at NRL. The key results are illustrated schematically with the corresponding data in Fig 2 and 3. In the first, we created the Bell state $|\psi\rangle = C_{\downarrow\downarrow}|\downarrow\downarrow\rangle + C_{\uparrow\uparrow}|\uparrow\uparrow\rangle$. This measurement comes about by exploiting the power of coherent nonlinear laser spectroscopy measurements where, in the density matrix picture, we measure $\rho_{\downarrow\downarrow\uparrow\uparrow}$. The data is obtained using two frequency stabilized narrow band lasers that are independently tunable but have a mutual coherence time longer than the dephasing time of the state. One laser field is used to excite the exciton transition and the second field is scanned across the biexciton resonance. Measurement of the entangled state are made by homodyne detecting the coherently emitted field with exciton excitation field, E_1. Measurements from the data of the decoherence rate show the absence of pure

dephasing at 5 K. The results are published in *Physical Review Letters* **88,** p117901 (2002).

An equally important study is the demonstration to be able to optically entangle and detect the orthogonally polarized states represented by

$$|\psi\rangle = C_{\downarrow\uparrow}|\downarrow\uparrow\rangle + C_{\uparrow\downarrow}|\uparrow\downarrow\rangle$$

in Fig. 3. Again, these measurements exploited the power of coherent nonlinear optical spectroscopy using frequency locked high resolution lasers. This represented a major step in that it showed that we could optically entangle and detect two orthogonally polarized states. In these experiments, measurements were made in the presence of a magnetic field to further increase the fine structure splitting (using the Zeeman splitting) and restore the circularly polarized optical selection rules. Again, in the density matrix picture, the measurement corresponds to detecting the term $\rho_{\downarrow\uparrow\uparrow\downarrow}$. As in the case of the two-photon coherence and entanglement, there was no detectable pure dephasing. The results were published in *Science*, **289**, 1906 (2000).

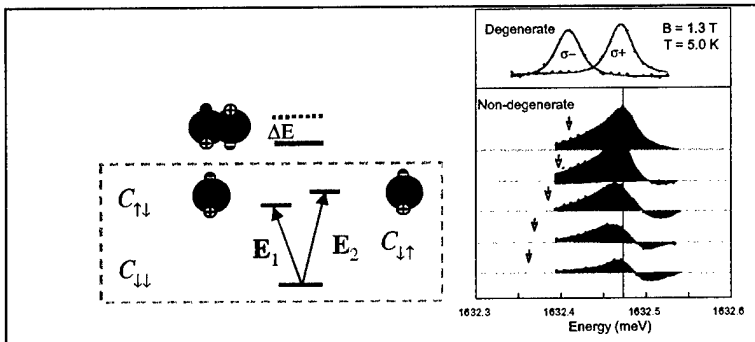

Figure 3. Experimental demonstration of the optically induced and detected quantum entanglement of two polarized exciton states, excited with orthogonally circularly polarized optical fields. The interference type line shape is the unique signature of the entanglement in this experiment.

Exciton and Biexciton Rabi oscillations.

In the atomic picture, it is well known that in two and three level systems, strong resonant excitation leads to Rabi oscillations, a phenomenon critical to coherent optical control and closely related to the observation of the Mollow Triplet. Experiments demonstrating this behavior have direct implications for qubit rotations in quantum computing and photon-on-demand sources for quantum communications. In this research period, we have successfully demonstrated Rabi oscillations on the ground state to exciton transition and the exciton to biexciton transition.

For these experiments, the simple theory shows that the state of the system oscillates as a function of the pulse area, defined as $\Theta(t) = (\mu_{eg} \bullet \hat{\varepsilon}_1) \int_{-\infty}^{t} E_1(t')dt'$. A weak probe pulse is then used to determine the state of the system after the strong pulse excitation. The pulse area can then be varied by either changing the pulse length or the pulse amplitude. In these experiments, it was easiest to vary the pulse amplitude.

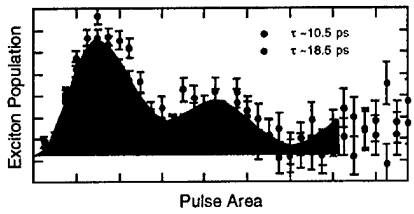

Figure 4. Experimental data with a theoretical fit of the Rabi oscillations observed on a single quantum dot ground state to exciton state transition as a function of pulse area.

Figure. 4 shows the result of exciting and probing the ground state to exciton transition. The data shows the distinctive oscillation predicted by the simple theory. An overall decay in amplitude is also seen in the data which is not accounted for the in the simple theory. A more complete theory takes in account the possibility of scattering by adjacent excitonic states spatially nearby in the crystal. These states are off resonance, their presence only becomes an issue at higher excitation levels. Our collaborators at NRL are working to develop quantum dot

architectures with stronger confinement and weaker uncontrolled interactions.

To demonstrate the exciton to biexciton Rabi oscillation, we use a prepulse tuned and polarized to excite the specific ground state to exciton transition. The pulse area for that pulse is adjusted to be π, to create a complete inversion as seen in Fig. 4. The pump pulse is then tuned and polarized to excite the biexciton state, the pulse area of that pulse being adjustable by controlling the pulse amplitude again. The degree of population in the biexciton state is then probed by using an orthogonally polarized optical field (relative to the pump pulse and from the same laser) to probe the gain on the orthogonally polarized biexciton to exciton transition. This geometry would historically be referred to as the Raman gain configuration. The Rabi oscillation is seen in Fig. 5 as a function of pulse area.

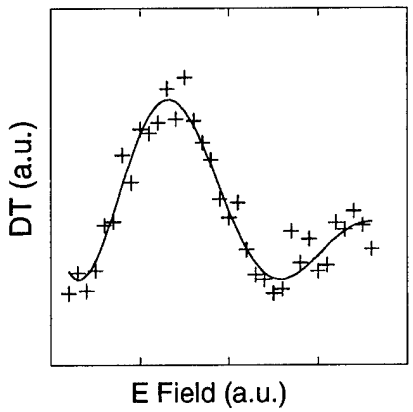

Figure 5. A full Rabi oscillation is demonstrated in the three-beam experiment where the pulse area of the pump pulse is varied and the biexciton population is probed at ~10ps delay.

Development and application of a low temperature near field scanning optical microscope (NSOM) for nonlinear optical spectroscopy

A major objective of the past several years has been to develop a methodology that allows us to probe individual semiconductor nano-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 rather than the indirect measurements of earlier NSOMS (based, for example, on luminescence). This microscope was the first of it s kind (published in *Science* **293**, pp224-227 (2001)). The results are quite profound and demonstrate the complexity of exciton localization leading to quantum dot formation 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 6 actually provides the first mapping 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 the previous final report. However, in practice, it is much more 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 the included reprints. This capability provides a means to map the optical dipole in space and energy with high resolution, revealing the optical local density of states (LDOS) of the system in analogy to previous scanning tunneling microscopy (STM) work.

The disordered layer was buried 130nm in the sample. This was essential to prevent surface broadening of the spectral lines but led necessarily to a spatial resolution that was degraded relative to that available within a few 10 s of nm of the tip. Combined with the aperture size, this set the resolution limit to approximately250nm. A solid-immersion lens has achieved resolution on similar length scales at low temperature, but those measurements have achieved their technological limit in contrast to the approach in this paper, where near-field technology has been demonstrated to $12\,nm$ resolution on surface structures. It is interesting to note that the nonlinearity of the signal mechanism enhances the spatial resolution afforded by the microscope; this advantage has previously been exploited by our laboratory to provide sub-$\lambda/2$ resolution with uncoated probes (*Optics Letters* **23**, 1111-1113(1998)).

The nonlinear mapping of the optical LDOS characterizes many isolated eigenstates and is separable in energy and space $S_{NL}(\boldsymbol{R},\omega) = \sum_n \zeta_n f_n(\omega) h_n(\boldsymbol{R})$ where, for a localized excitonic eigenstate $|n\rangle$, $f_n(\omega)$ is the lineshape of the nonlinear response, $h_n(\boldsymbol{R})$ is the

optical-field distribution convolved with the excitonic wavefunction and ζ_n is a constant.

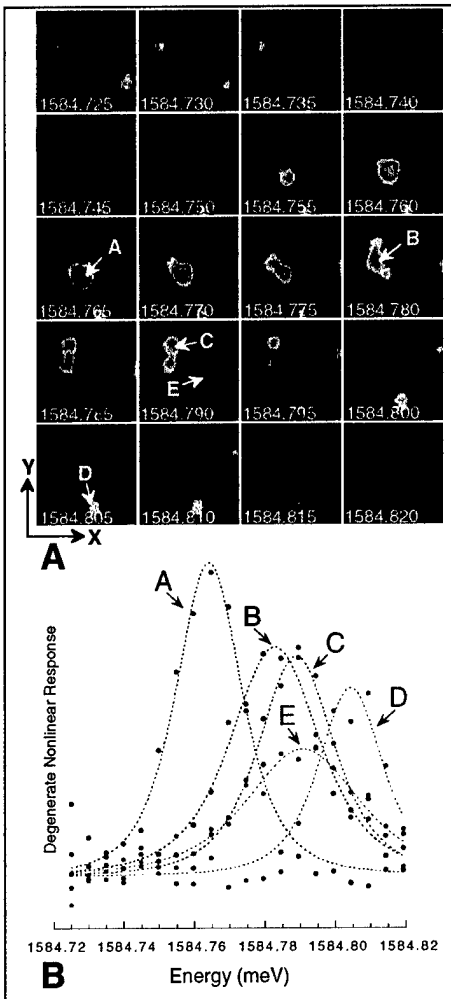

Figure 6. High spectral resolution three-dimensional CW nonlinear response. (A) Three-dimensional CW nonlinear response spanning $2\,\mu m \cdot 2\,\mu m \cdot 100\,\mu eV$ represented by spatial images recorded at different energies (annotated in meV). (B) The spectral lineshapes for the five eigenstates labeled in (A) are fit by Lorentzians squared

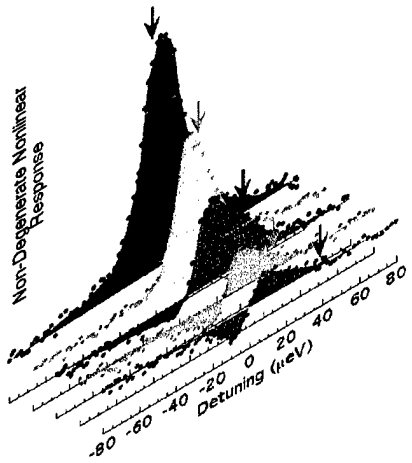

Figure 7. Non-degenerate (W_1 , W_2) spectral mapping of a single eigenstate. The spectral position of the pump (W_1) is indicated by the arrow.

This concept is clearly exemplified in Fig. 6a, where a three-dimensional data set spanning $2\,\mu m \cdot 2\,\mu m$ but only $100\,\mu eV$ in energy is represented by a series of images taken at different energies. For these isolated homogeneously broadened resonances, the spectral lineshapes take the form of a Lorentzian squared where the Lorentzian linewidth (FWHM) is $2\hbar\gamma_n$. As seen in Fig. 6b, the data are well fit by this form and reveal an $\hbar\gamma_n$ that ranges from 17 to 29 μeV (T_2 " $1/\gamma \approx 22 - 38\ ps$) in strong agreement with the far-field values obtained from hole-burning and with those obtained from studies through apertures. Unlike PL spectra that frequently have instrument limited linewidths for these systems, the high resolution of the frequency-stabilized lasers provides an unambiguous result. Detailed spatial and statistical analysis of the optical LDOS, which will be discussed in a future report, can provide a mapping of the excitonic center-of-mass wavefunction and shed light on phenomena such as level repulsion.

The various time scales associated with decoherence and energy relaxation of the optically induced quantum coherence can be extracted by utilizing the full power of coherent nonlinear spectroscopy. The nonlinear optical response of strongly localized excitons is comprised of an

incoherent and a coherent contribution. The details of the nearly degenerate nonlinear optical response is fully described in an early paper during the previous grant period (*Physical Review Letters* **81**, 2759, (1998)). The incoherent contribution is the result of simple saturation of the optical resonance by one optical field that is then probed by the second optical field; the relative phase between the fields does not matter (incoherent sources would suffice). The coherent contribution, on the contrary, arises from the mixing of the two fields through the excitation and is highly sensitive to their relative phase. In the above degenerate nonlinear data sets, these contributions are indistinguishable and, as a result, only the overall dephasing rate is accessible. In order to differentiate between these contributions (and between the various time scales, as discussed below), we employ two non-degenerate optical fields which have a mutual coherence time that is much longer than the time scale associated with the evolution of the quantum dot excitation (i.e., inverse energy relaxation rate and the exciton dephasing rate). For these experiments, the mutual coherence time of the two frequency-locked lasers is of order microseconds and easily satisfies this requirement. The influence of the coherent contribution is seen in the non-degenerate (W_1 „ W_2) nonlinear optical response shown in Fig. 7.

Using the analysis presented in (*Physical Review Letters* **81**, 2759, (1998)), the data in Fig. 7 show an unambiguous interference lineshape as the pump field is tuned away from line center. The coherent contribution dominates the nonlinear response and a dispersion-like lineshape emerges. From curve fitting, we get find no significant pure dephasing and $T_2 \approx 25\ ps$ and $T_1 = 1/\Gamma_{\parallel} \approx 16 - 3\ ps$.

Quantum coherence and entanglement in self-assembled quantum dots

Self assembled quantum dots are distinct from interface fluctuation dots in that they form through stress and are typically much smaller; they are in the so-called strong confinement regime. Efforts to date to carry out coherent nonlinear spectroscopy on these systems have not had much success except very recently in ensemble measurements. This appears to be due to the much weaker coupling to the electromagnetic field, for reasons that remain unclear. Our own group has been working on these structures since it is possible to imagine scalable architectures based on this growth technique much more easily than with interface dots.

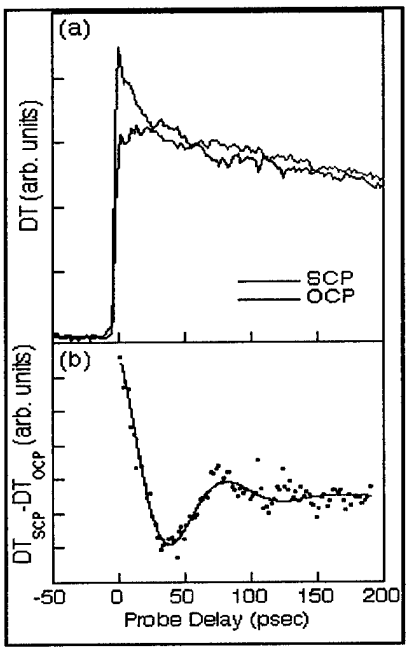

Figure 8. The homodyne detected four-wave mixing response of an ensemble of self-assembled quantum dots. The measurements are performed with a choice of excitation polarization to create a coherent superposition of the two fine structure split states. The result is that we see a coherent oscillation that reflects the existence of a coherent superposition of these two states.

Our measurements have focused on simple polarization sensitive differential transmission in ensemble samples grown by P.K. Bhattacharya s group here at Michigan. We obtained weak but detectable signals which reported on state and spin relaxation. However, more importantly by using excitation polarizations which are a combination of vertical and horizontal polarizations, such as circular, we can create a superposition of the two fine structure split exciton states. This is shown in Figure 8(a), for a probe which has the same (blue) or opposite (red) helicity to the pump pulse. At early delays, an oscillation is superimposed on the overall decay signal. In Figure 8(b), the subtraction of the two signals (DT_{SCP} —DT_{OCP}) shows this more clearly. These oscillations are true quantum beats resulting from the coherence between the two polarization states of the excitons. The details of the analysis of this kind of experiment was presented in an earlier paper during the previous program (*Physical Review Letters* **80**, 786 (1998)). The solid line is a fit to an exponentially damped cosine function. The oscillation period corresponds to the energy splitting between the states, which in this case

is found to be ~50 μeV. The experimentally determined decay constant is 37 psec. This decay is caused by the loss of coherence between the two polarization states; however, the influence of inhomogeneous broadening on the measured decay time is still being investigated theoretically. These beats are similar to those discussed earlier by our group and represent evidence of an entangle state between the pseudo spin states associated with the optical Block vector. (This work is in press in *Physical Review Letters.)*

Summary

This program has resulted in new understanding of importance to nonlinear optical nano-science, and has shown that nano-structures may be potentially viable structures for optically controlled quantum devices. Five students received their Ph.D. due in part to support from this program, and a number of other students are in the Ph.D. pipe-line . Their success has been outstanding, at least two went on Bell Labs, and the remaining three are in academic postdoctoral positions. This work has laid the foundation for our future studies of spin based quantum dots and more complex structures based on interacting quantum systems.