

FELLOWSHIP IN PHYSICS/MODERN OPTICS  
AND QUANTUM ELECTRONICS

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## STATEMENT OF PROBLEM

This research was directed toward femtosecond time-resolved spectroscopy of optical nonlinearities in semiconductors. The experimental setup utilizes ultrashort pulses in a variable-delay pump-probe configuration. The laser is a balanced colliding-pulse mode-locked ring-dye laser amplified by a copper-vapor laser. Its output is a train of pulses at 620 nm with an 8.5 kHz repetition rate; pulse duration is 60 fs. The pulse energy is a few microjoules and it generates a white light continuum that serves as a fast probe. A fiber-grating pulse-compression stage provides an alternate probe source at  $< 20$  fs near 620 nm. In addition, second-harmonic generation in a nonlinear crystal can produce a 310-nm pump for excitation of widegap semiconductors. With these tools we can measure the dynamics of optical nonlinearities, including the initial transients and the recovery.

## RESULTS OF THE FELLOWSHIP

During the first three years, this Army Science and Technology fellowship successfully met a large number of its goals. The proposed femtosecond laser system was built. This system was continually upgraded, remaining competitive with the best systems maintained at only a handful of institutions around the world. In addition, research into CuCl thin film etalons and  $\text{CdS}_x\text{Se}_{1-x}$ -doped glasses was initiated. This research grew to include several related areas of semiconductor physics that are particularly suited to study with the femtosecond laser system. These areas include coherent effects in semiconductors, MBE-grown ZnSe, Optical Stark Shift, and CdSe quantum dots. In the six-month extension of the fellowship, research into quantum dots continued, and included an investigation into a relatively new semiconductor-doped glass — CdTe. In addition, laser research, always an ongoing effort to keep ours a state-of-the-art system, continued. Results of our research have been presented at international conferences and published in well-known journals. Two Physical Review Letters were published, in addition to a large number of other papers. Our graduate student supported by this program has completed all preliminary requirements, passed both the written and oral section of the PhD exam, and accumulated a body of dissertation research.

Our accomplishments under this fellowship are discussed in the following sections.

### Femtosecond Laser System

A colliding-pulse modelocked laser was built. This instrument is a ring dye laser, cw pumped by an all-lines argon laser onto a gain jet of rhodamine 6G. A thin jet of

DODCI, a saturable absorber, passively mode-locks the cavity at 90 MHz. No tuning elements are used. The laser is allowed to operate at the DODCI's most efficient wavelength, 620 nm. The large number of modes lasing and the saturating effects of the two dyes allow production of subpicosecond pulses. The addition of a four prism intracavity dispersion compensator allowed us to fine tune the pulse width to  $< 50$  fs (see Fig. 1).

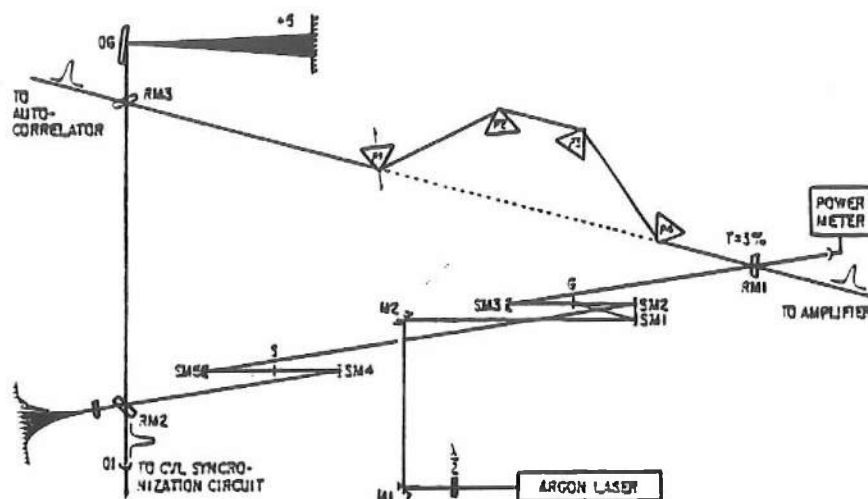


Figure 1. Schematic of the balanced colliding-pulse modelocked (CPM) ring dye laser. RM = ring mirror, SM = spherical mirror, G = gain jet, S = saturable absorber jet, DG = diffraction grating.

The pulse energy from this laser is on the order of 100 pJ. While the laser has significant peak power, 100pJ is insufficient for most nonlinear experiments. The pulse energy is therefore amplified in a multipass dye amplifier pumped by a high-repetition-rate (8.5 kHz) copper-vapor laser. The scheme utilized is the popular "bow-tie" design, with six passes through a jet of rhodamine 640 and a saturable absorber after the fourth pass. This scheme gives a total gain of  $10^5$ , up to the microjoule level, and lowers the repetition rate to 8.5 kHz. A prism dispersion compensation stage brings the the pulse width to 60 fs.

At this point a portion of the pulse is split off for use as an intense pump source. The remainder of the pulse is focused onto a jet of transparent solvent, generating a continuum of wavelengths over the entire visible spectrum and up to 900 nm. This method provides an ideal probe source, because it has the same pulse width as the source (although complicated by the presence of chirp). An alternate probe source is a fiber-grating pulse compression stage. This source generates a narrower continuum in an optical fiber, and uses a pair of gratings to compress the pulse to  $< 20$  fs. For

experiments around 620 nm, this provides a chirp-free pulse. The pump wavelength can be at 620 nm, or at its second harmonic, 310 nm. As discussed below, we also made progress on a tunable pump source.

### CuCl Thin Film Etalons

Our group has fabricated etalons by growing thin films of CuCl and sandwiching them between highly reflective mirrors. The highly broadband continuum generated in the ethylene glycol jet enables us to probe these etalons in the transparent region below the 380-nm exciton, while generating carriers with the 310-nm pump. We measured the dynamics of the nonlinear excitonic effects in CuCl, screening of the excitons, and their recovery (see Fig. 2). The shift of the Fabry-Perot peak attributed to exciton screening was also measured.

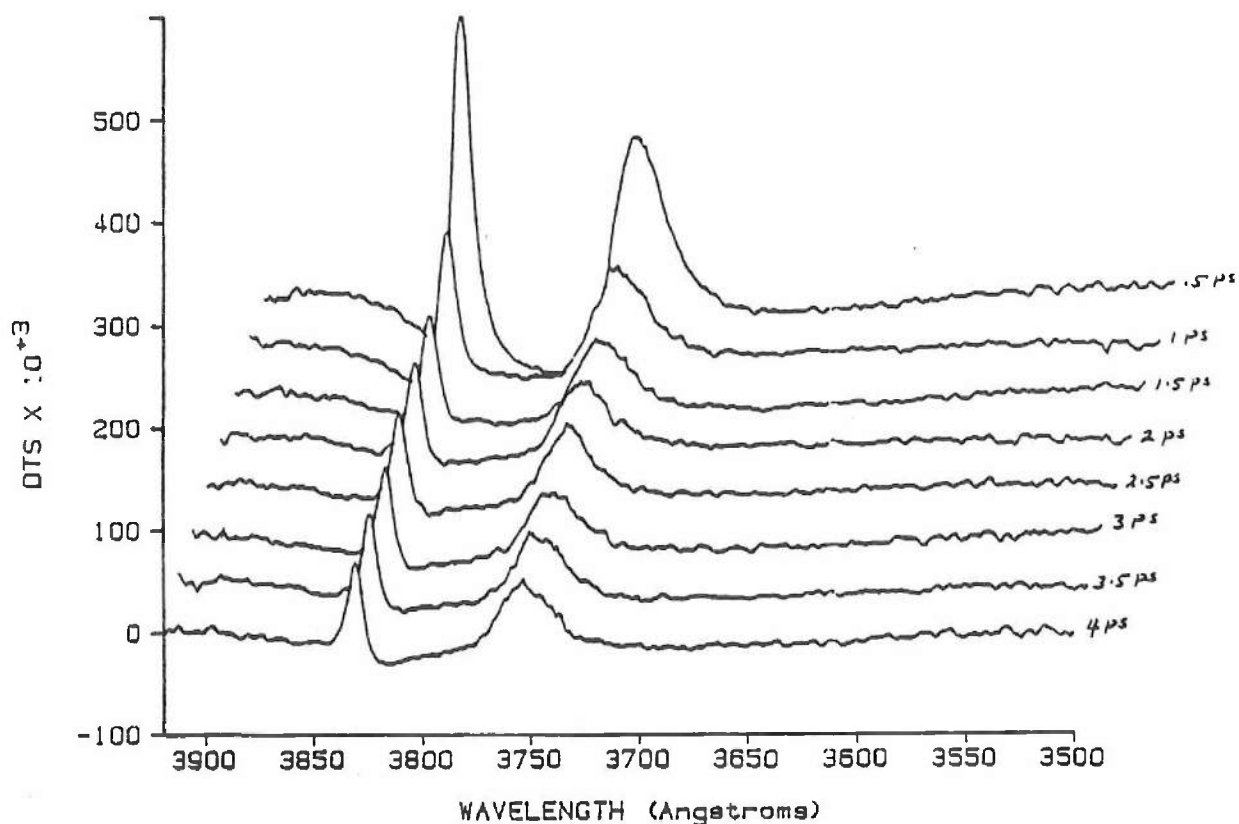


Figure 2: The dynamics of the nonlinear excitonic effects in CuCl. The screening of excitons and their recovery is shown.

### Commercial Semiconductor-Doped Glasses<sup>1-4</sup>

Using the femtosecond laser system, we studied the dynamics of the bandfilling nonlinearity in  $\text{CdS}_x\text{Se}_{1-x}$ -doped glasses. Using broadband, discrete-time measurements, we measured the thermalization of carriers from the initially nonthermal distribution by carrier-carrier and carrier-phonon scattering (see Fig. 3). This thermalization is followed by relaxation of the hot distribution down to the temperature of the lattice, at which point the bandfilling (blue-shift) nonlinearity occurs. Single-wavelength continuous-time measurements were used to determine the recovery time of this nonlinearity and its dependence on sample temperature and excitation intensity. In addition, the near-bandedge luminescence was time resolved to give an independent measure of carrier lifetime.

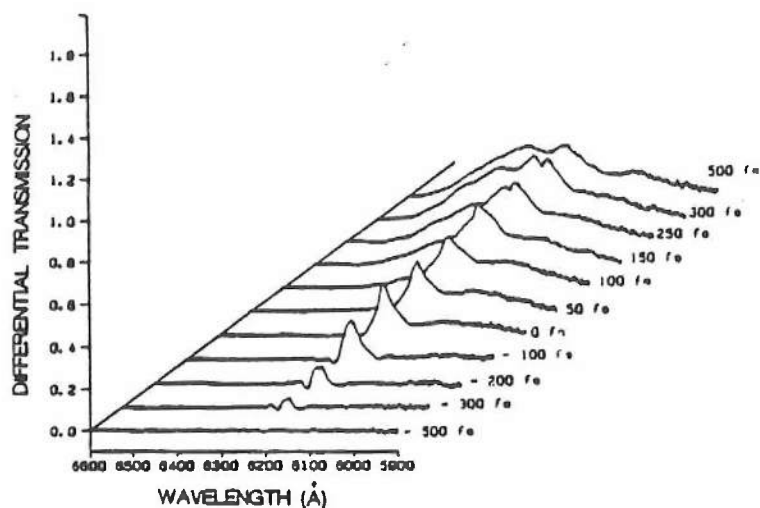


Figure 3. Differential transmission spectra of a  $\text{CdSe}_{0.2}\text{S}_{0.8}$  glass at 300 K for various delays between the pump and probe pulses.

### Coherent Effects in Semiconductors<sup>5-8</sup>

We studied in detail a family of pump-probe phenomena we call "coherent oscillations." These phenomena are oscillatory structures in the spectral differential transmission of semiconductors that occur prior to the many-bodied coherent effects such as those described above for doped glasses. We investigated three distinct examples of coherent oscillations. These examples arise from, and have been associated through experiment with, spectral hole burning, exciton Optical Stark Shift, and exciton bleaching by free carriers. The phenomena were observed in a variety of systems: in bulk CdS

and CdSe, in doped glasses, and in GaAs and GaAs-AlGaAs multiple quantum wells (see Fig. 4). Collaborating with a theoretical group, we described the results completely in terms of a strong femtosecond pulse interacting with the medium polarization.

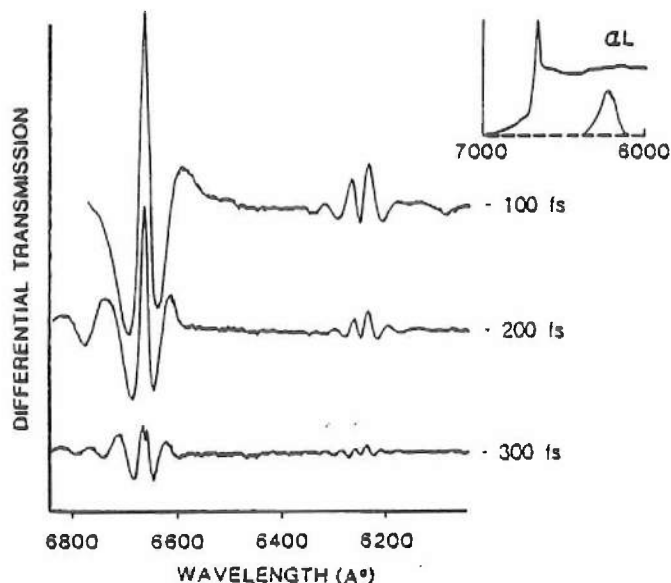


Figure 4. Differential-transmission spectrum for a pump-probe delay of -300, -200, and -100 fs measured for a thin CdSe platelet at 10 K. Inset: Absorbance  $\alpha L$  for the sample, and the pump spectrum.

#### MBE-Grown ZnSe<sup>9,10</sup>

We studied the recovery times of the excitonic nonlinearity in molecular-beam epitaxially-grown ZnSe. These single-crystal thin films of ZnSe were grown on GaAs substrates by 3M Corporation. The GaAs then is chemically etched away, leaving a small ZnSe window. This material has been shown to exhibit an optical nonlinearity that is attributed to Coulomb screening of the exciton. By monitoring a single probe wavelength on a continuous-time-delay scan, we measured the decay of the nonlinearity through carrier recombination over a 600 ps range. The effects of variations in sample thickness, temperature and excitation energy were also studied (see Fig. 5).

#### Optical Stark Effect<sup>11-13</sup>

We studied the Optical Stark Shift of the low-temperature exciton in bulk, wide-gap semiconductors. Using large detunings and low intensities, we demonstrated some interesting results. First, we minimized the generation of real carriers to allow the observation of an almost pure Stark Shift. In addition, working at low temperature in CdS, we observed a shift of the continuum (bandedge) states (see Fig. 6). We developed

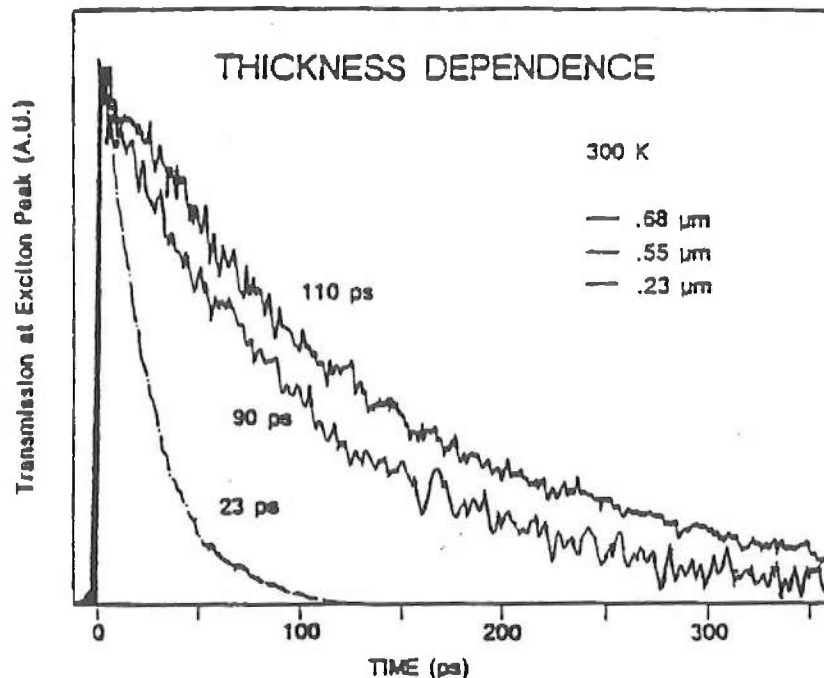


Figure 5. The measured response time of MBE-grown ZnSe as a function of thickness.

a theory to account for both the shifts and the coherent oscillations that occur before them. The oscillations are expected whenever the pulsewidths are comparable to the material-coherence decay time. This theory is in agreement with our experimental observations.

#### Quantum Dots<sup>14</sup>

In addition to the commercial semiconductor-doped glasses discussed earlier, we studied experimentally grown doped glasses that were heat treated to produce smaller microcrystallite sizes (on the order of the bulk semiconductor Bohr radius). The optical properties of these glasses depart from those of the bulk material. The bandedge is replaced by a discrete series of broadened transitions. Using a tunable femtosecond laser system at Ecole Polytechnique, ENSTA, we measured the differential transmission spectra while pumping into the transitions. We observed spectral hole burning of the transitions, from which we determined the polarization decay time (see Fig. 7). In addition, an induced absorption feature was observed on the high-energy side of the hole. Both results were explained in terms of bleaching of one-pair states and generation of two electron-hole pair states from pump and probe.

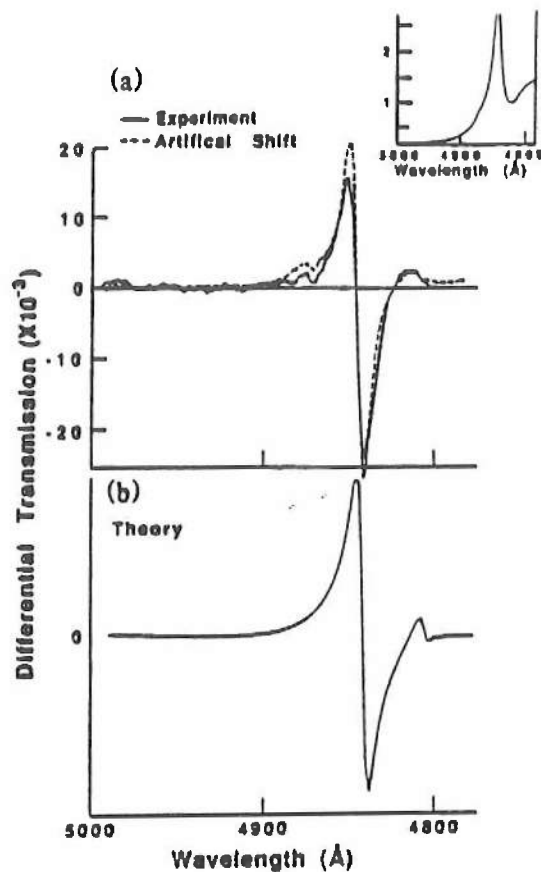


Figure 6. (a) The measured DTS in the vicinity of the B exciton (full line) and its comparison with a pure shift of the absorption spectrum (dashed line). See text for description. (b) The calculated DTS in the vicinity of the B exciton for  $t_p = -25$  fs and a pump pulse duration of 60 fs.

### Generation of Tunable Femtosecond Pulses

Even though our 620-nm pump/tunable probe femtosecond laser system has many applications, there are a number of experiments that require a tunable, high-power pump. In our experiments with quantum-dot glasses, for example, we would like to pump at various detunings from the resonances. For this reason, we are developing a continuum reamplification system. The system is a multipass dye amplifier pumped by a copper-vapor laser. It amplifies a 10-nm portion of the continuum. At present we can produce 0.5-microjoule, 115-fs pulses at 580 nm (in addition to the 620-nm pulses), with somewhat less power over the range 570-600 nm. With additional dyes, we expect to be able to produce all the pump wavelengths needed for our quantum dot experiments.

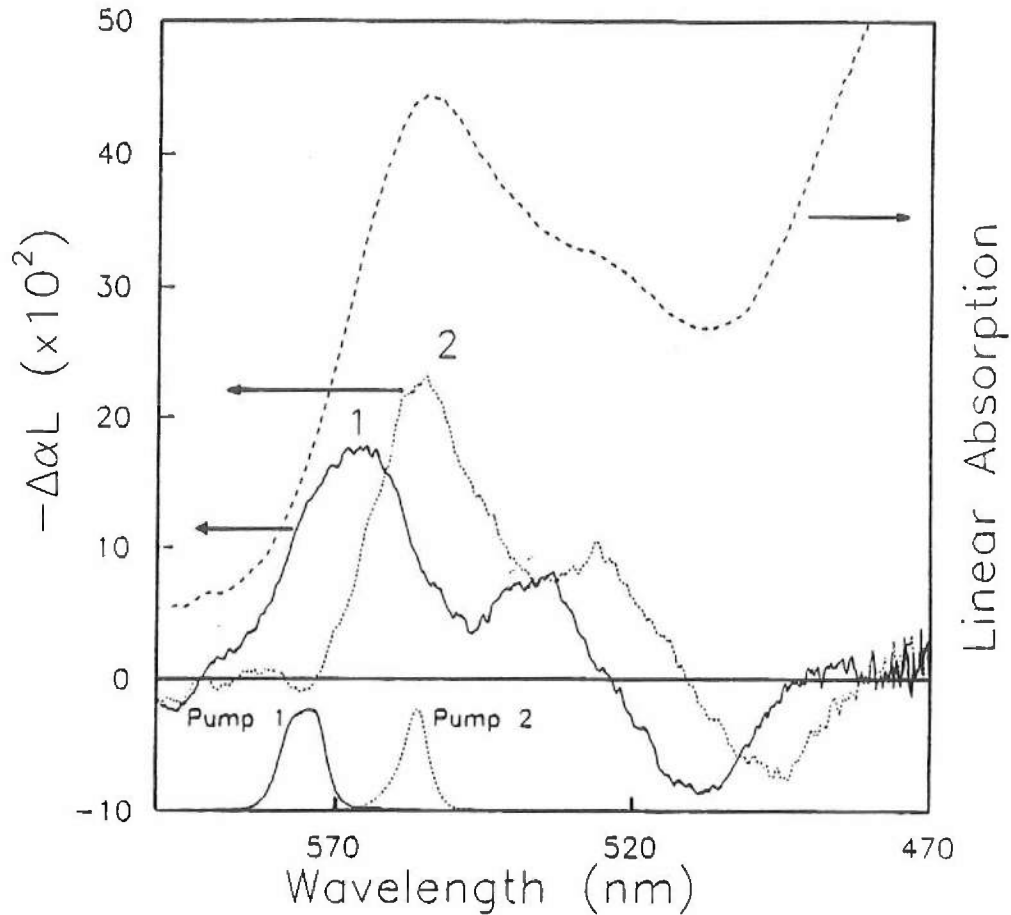


Figure 7. The differential transmission of a quantum dot sample for two pump wavelengths inside the A-transition at 10 K. The energetic positions of the pumps are indicated and the linear absorption spectrum is plotted. The linear absorption coefficient at the peak of the lowest quantum confined transition, A, is  $\alpha l \sim 2.0$ .

### CdTe Quantum Dots

Quantum-confined CdTe-doped glasses were grown at the University of Arizona Materials Sciences Department. As usual, when the temperature and duration of the growth phase are varied, microcrystallites of various sizes are produced. Because the Bohr radius of CdTe is  $\cong 70 \text{ \AA}$ , these samples exhibit a higher degree of quantum confinement than do CdS and CdSe. Several higher-level transitions can be seen in the linear absorption spectra for CdTe. We measured the inhomogeneous line width through a hole-burning experiment. We also measured the lifetime at 77 K, for comparison with that of CdSe quantum dots. In addition, we studied bleaching and induced absorption around the higher-level transitions for comparison with the theory used for CdSe.

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