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SEMICONDUCTORS INVESTIGATED BY TIME RESOLVED
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ULTRAFAST SPECTROSCOPY AND LASER LAB R R ALFANO

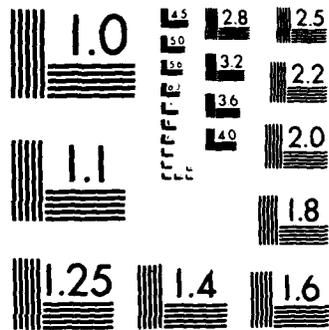
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| Four major accomplishments have been achieved to help further the development of faster photonic and electronic devices. | | | |
| 1. We have shown theoretically and experimentally how one can determine accurately one of the most important parameters in microstructures - the bandgap discontinuity in valence and conduction bands at the heterojunction from the photoluminescence measurements for ultrathin wells in the range of 15 to 80 Å for GaAs/AlGaAs and GaInAs/AlInAs structures. | | | |
| 2. A model was developed using the electron degeneracy to describe the much slower carrier - optical phonon loss relaxation rate measurements in 2D as compared to 3D. | | | |
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3. The valence and conduction band deformation potentials were separately determined for the first time in semi-magnetic semiconductor alloys of $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ from the shift in photoluminescence spectra versus x . The valence band deformation potential of wurtzite crystals is much larger compared to the one in zinc blende. This technique measures separately the values of valence and conduction band deformation potential instead of the difference between them.

4. The direct picosecond spin dephasing time and degree of spin alignment of photo-excited electrons in semi-magnetic semiconductor alloys of $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ was measured. The fast dephasing times arise from the spin exchange between free carriers and the localized Mn^+ ions.

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PROGRESS REPORT
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ULTRAFAST SPECTROSCOPY AND LASER
LABORATORY
OF
THE CITY COLLEGE OF NEW YORK
NEW YORK, NY 10031

DEPARTMENT OF PHYSICS

PROFESSOR R. R. ALFANO

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**SEMICONDUCTORS INVESTIGATED BY TIME RESOLVED
SPECTROSCOPY USING FEMTOSECOND AND
PICOSECOND LASER TECHNOLOGY**

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New York, NY 10031**

Program Manager: Dr. Gerald Witt

Report Date: March 8, 1986

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Introduction

This report summarizes the progress achieved during the 1984-1985 period in the research effort supported by AFOSR under contract #85 0013.

Four major accomplishments were made during this period in:

1. Microstructure semiconductor research.
2. Electron-degeneracy effects on electron optical phonon energy loss rate for two- and three-dimensional electron systems.
3. Measurement of deformation potential using photoluminescence in magnetic semiconductor alloys $Cd_{1-x}Mn_xSe$.
4. Spin polarization and dephasing times of carriers in alloy semiconductors $Cd_{1-x}Mn_xSe$.

The following describes the four areas in greater detail.

1. Microstructure Research

Band-engineering has produced spectacular photonic devices such as quantum lasers, photodetectors as well as high speed logic elements.¹ The intrinsic asymmetry associated with conduction-band-discontinuity (ΔE_C) and valence-band-discontinuity (ΔE_V) at the interface plays an important role in this "hot" field. For this reason, determining ΔE_C and ΔE_V accurately is of considerable interest and importance.

We have calculated the ΔE vs L_z for various quantum well structures using their ΔE_V as a parameter and various sets of masses for heavy-hole (hh) and light-hole (lh). It was found that there are always two distinct regions for the well widths L_z to find ΔE_V accurately from those calculated curves. One region is called sensitive range ($15\text{\AA} < L_z < 80\text{\AA}$) in which ΔE 's are very sensitive to the chosen value of ΔE_V in spite of various sets of masses for hh and lh. In this region, it is possible to determine ΔE_V very accurately by fitting ΔE 's to experimental data. The other region ($L_z > 80\text{\AA}$) is called insensitive range for ΔE 's to

the value of ΔE_V . By this feature, we can state that it is almost impossible to determine ΔE_V accurately using the optical transitions in thick quantum wells.

The calculated result² of ΔE vs L_z for GaAs/(Al,Ga)As structure in fig. 1 was compared with the limited data available in the sensitive range yielding a good agreement with dingle's 15-85 rule: $\Delta E_V=0.15\Delta E_g$, where ΔE_g is the difference of the bandgaps at the interface.

By measuring the ΔE 's for two ultrathin 14.5-Å and 19.3-Å quantum wells from photoluminescence spectra³ (inset of Fig. 1), we determined band-discontinuities for (Ga,In)As/(Al,In)As structure without any ambiguity which agrees with Dingle's rule.

Our experimental and theoretical work support Dingle's rule and question the recent work of Miller at Bell Telephone Laboratories who gives the relationship of $\Delta E_V=0.4 \Delta E_g$.

The theoretical ΔE vs L_z curves can be used to determine the band-discontinuities accurately for other microstructures. The key is to use ultrathin wells ($L_z < 80 \text{Å}$).

Determination of the band-discontinuity of the heterojunctions

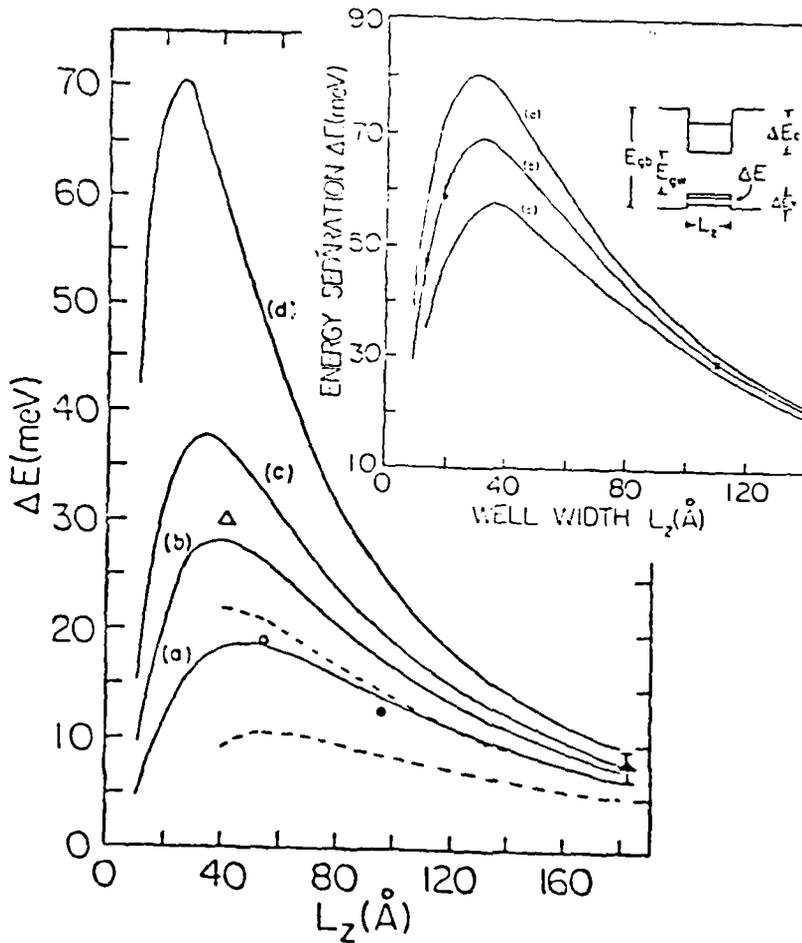


Fig. 1

ΔE - The calculated energy separation between heavy-hole and light-hole subbands. The width of the quantum well. (L_z)

2. Electron Degeneracy Effects on Electron-Energy-Loss Rate for two-and three-dimensional Electron Systems

Information about the hot carrier energy relaxation in semiconductors is important for the design and fabrication of high speed and high field ultrasmall devices. It is crucial to understand the underlying mechanisms which lead to a much smaller electron-energy-loss rate for two-dimensional (2D) electron system

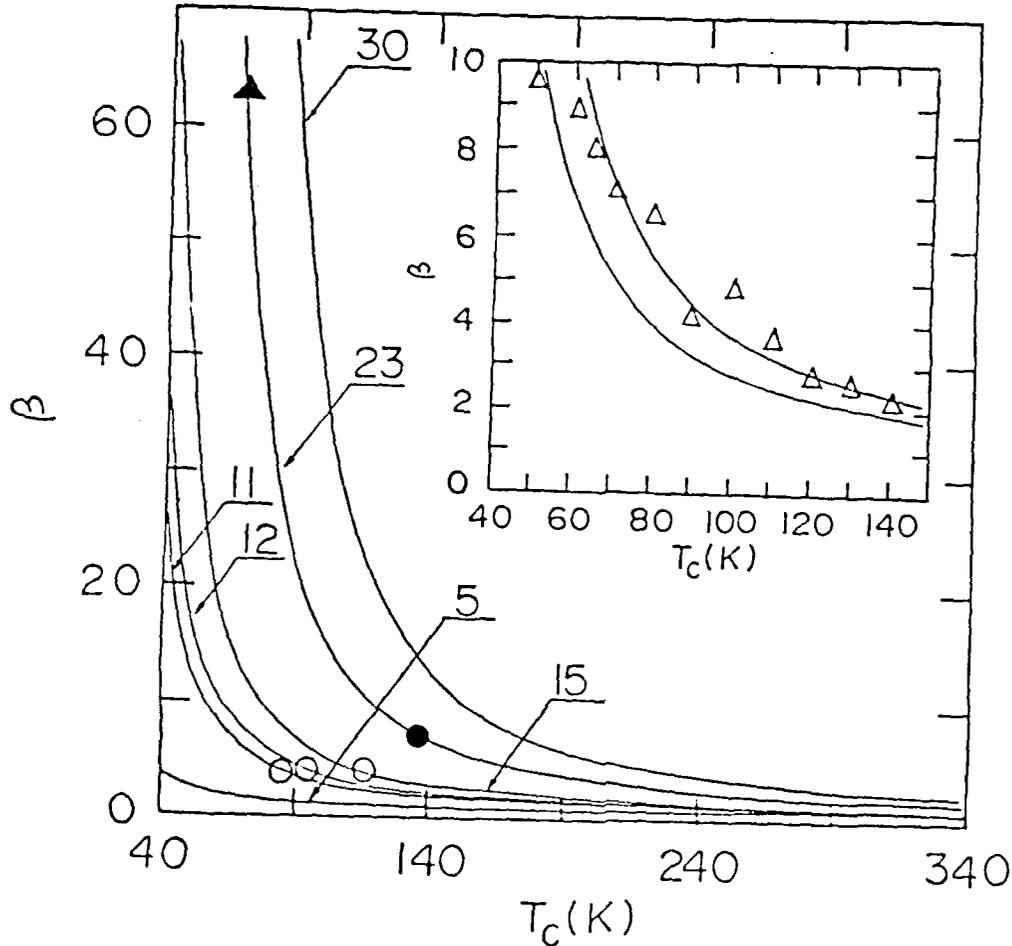
much smaller electron-energy-loss rate for two-dimensional (2D) electron system than for three-dimensional (3D) electron system.

Previous studies⁵⁻⁹ focused on how electron-phonon interaction is modified in 2D electron system in order to explain why electron-energy-loss rate for 2D case is much smaller than 3D case. These studies have generated conflicting interpretations regarding the influence of reduced dimensionality and plasma density on the electron-energy-loss rates. The conventional calculation of the rates from Fermi's Golden Rule as numerically calculated by Basu et al.⁸ with and without the effects of degeneracy does not clear up this controversial issue.

We¹⁰ primarily focused on the final available energy width for electrons to be scattered back for each electron system without involving intermediate states and the calculation of electron-phonon interaction matrix elements. The calculated ratio β of electron-energy-loss rates for 3D and 2D electron systems are shown in Fig. 2 based on difference of electron-degeneracy for 3D and 2D cases for different quasi-Fermi-energies (in meV).

The striking conclusions¹⁰ extracted from Fig. 2 enable us to uniquely explain various experimental results.⁴⁻⁷ The circles indicate our result⁴ that β is about 4 obtained from detailed power balance for a set of single quantum well. The solid triangle and solid circle are the experimental data from Ryan et al.⁶ as well as Xu and Tang⁷, respectively. Comparing our model to the most recent experimental data by Shah et al.⁵, not only the much lower electron-energy-rate for 2D than 3D can be explained, but also a quantitative agreement of the trend of β vs T_c can be achieved. This is shown in the inset of Fig. 2.

Fig. 2 Electron-degeneracy effects on electron-energy-loss rate for two- and three-dimensional electron systems



β - The calculated ratio of electron-energy-loss rates for 3D and 2D electron systems. T_c - The electron temperature.

3. Measurement of Deformation Potential using Photoluminescence Cd_{1-x}Mn_xSe Alloys

For the first time, we have pointed out an unique method of measuring optical deformation potential in semiconductors using laser induced fluorescence. In CdMnSe (semi-magnetic semiconductor), the substitution of Mn at cation site (Cd), introduces a local pressurized environment. The pressure is equivalent to exter-

nally applied hydrostatic pressure on pure CdSe. This pressure manifested itself as optical deformation potential, will cause the conduction band and valence band to shift in opposite directions with respect to vacuum level. The increased or decreased bandgap could be monitored using the above bandgap laser photon excitation photoluminescence. This is a first time separate determination of conduction and valence band deformation potential. In the past only the difference was measured. This also points out a large valence band deformation potential in wurtzite crystals compared to the one in zinc blende.

In figure 3, the increase in the bandgap is shown as a function of Mn concentration. Up to Mn concentration of 30%, the Mn localized states remain above the conduction band and the photoluminescence peak shifts are corresponding to band-band transition. The slope for $x < 0.3$ corresponds to joint (conduction and valence band ($1.2 \text{ eV}/x$)) optical deformation potential, for $x > 0.3$ the photoluminescence peak is due to Mn ($4G$)-valence band transition. In this case, the slope ($0.4 \text{ eV}/x$) corresponds only to the valence band as $4G$ Mn is almost stationary with respect to the vacuum level. Using the x-ray data of lattice change with Mn concentration we deduced the conduction and valence band optical determination potential as shown in the figure.

Fig. 3 Bandgap shifts

ACCOMPLISHMENT

MEASUREMENT

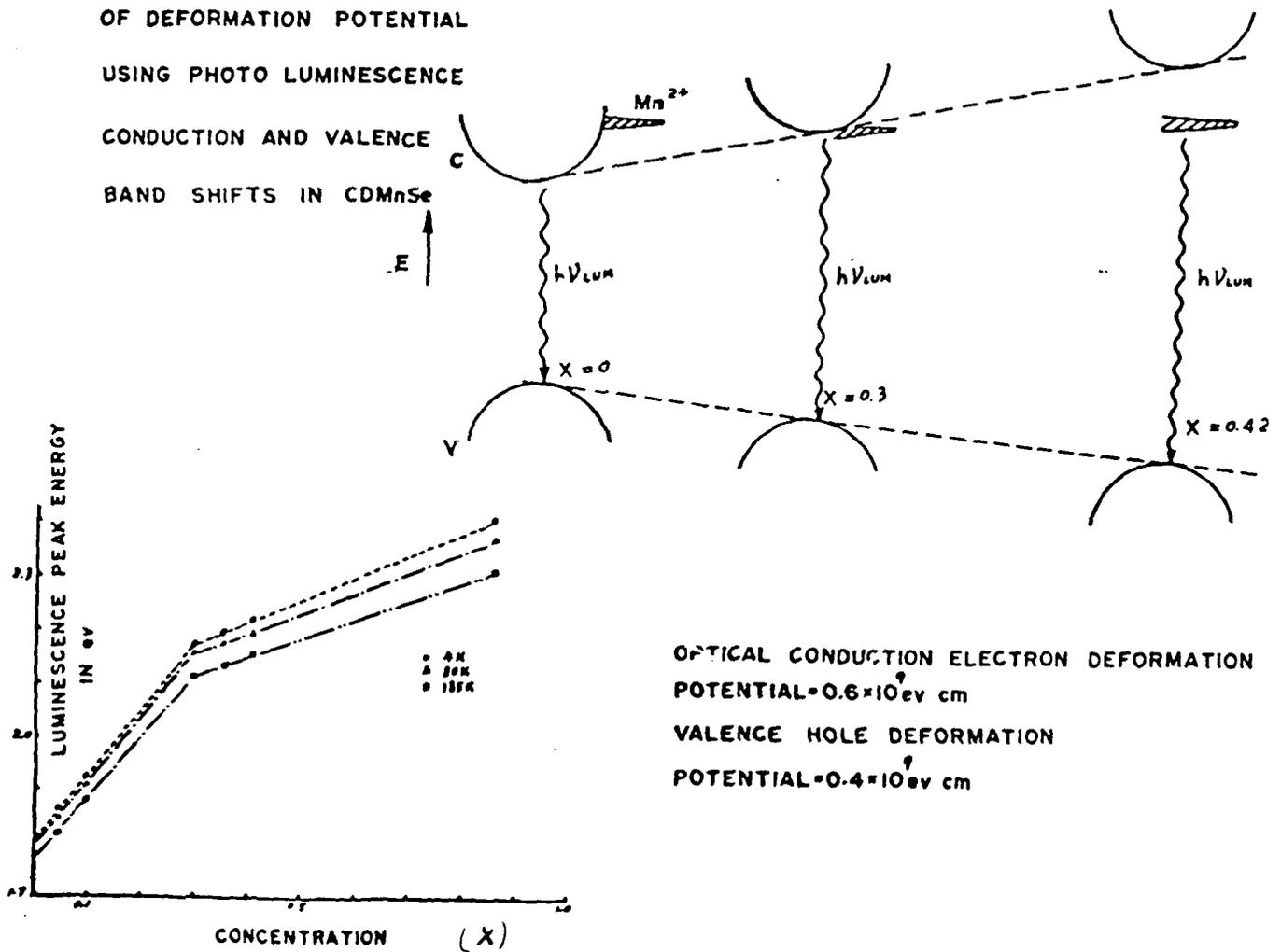
OF DEFORMATION POTENTIAL

USING PHOTO LUMINESCENCE

CONDUCTION AND VALENCE

BAND SHIFTS IN CdMnSe

Accomplishment-Measurment of deformation
using photoluminescence in $Cd_{1-x}Mn_xSe$



4. Spin Polarization and Dephasing Time of Carriers in Alloy Semiconductors
Cd_{1-x}Mn_xSe

For over 10 years, it was known that the dipole allowed circularly polarized above bandgap photon can induce a net spin alignment of photogenerated electrons. The observed ratio of spin up:spin down is reflected in the recombination photoluminescence. Most past work used steady state techniques to estimate spin alignment. One can now determine the percentage of spin alignment and spin dephasing time. The experiments we carried out were with single pulse excitation using Nd:glass laser having pulse width of 6 psec. The time resolved detection was achieved using high resolution streak camera. This is a major accomplishment.

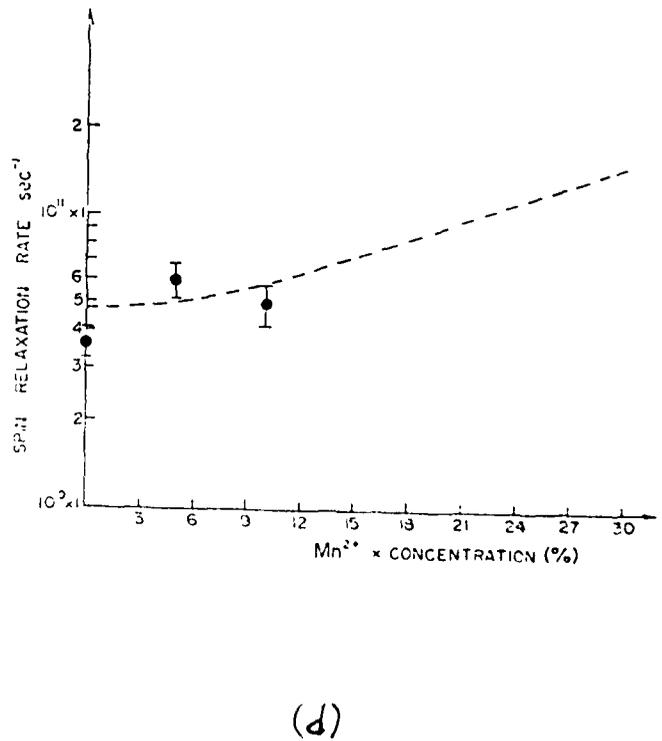
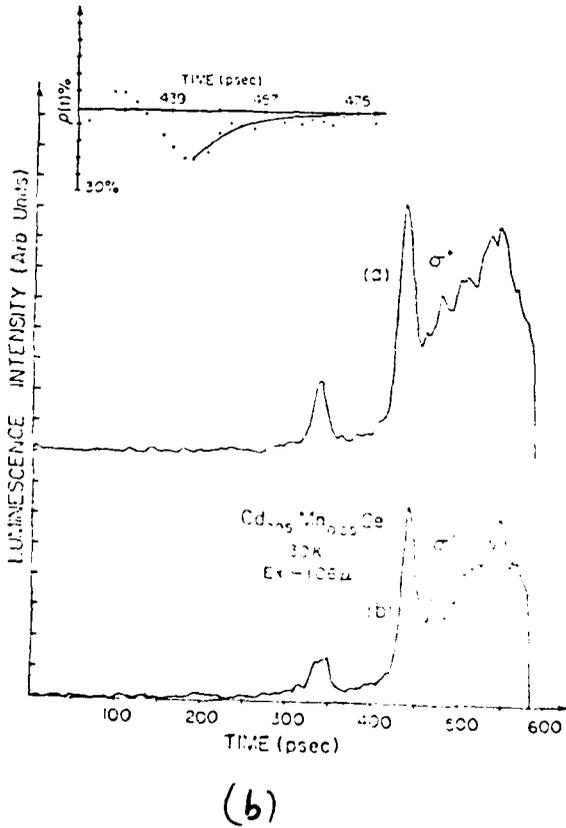
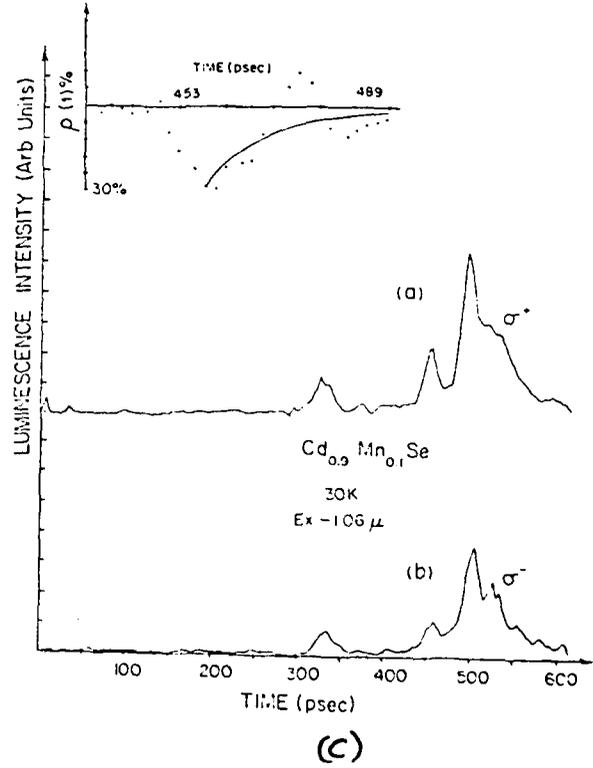
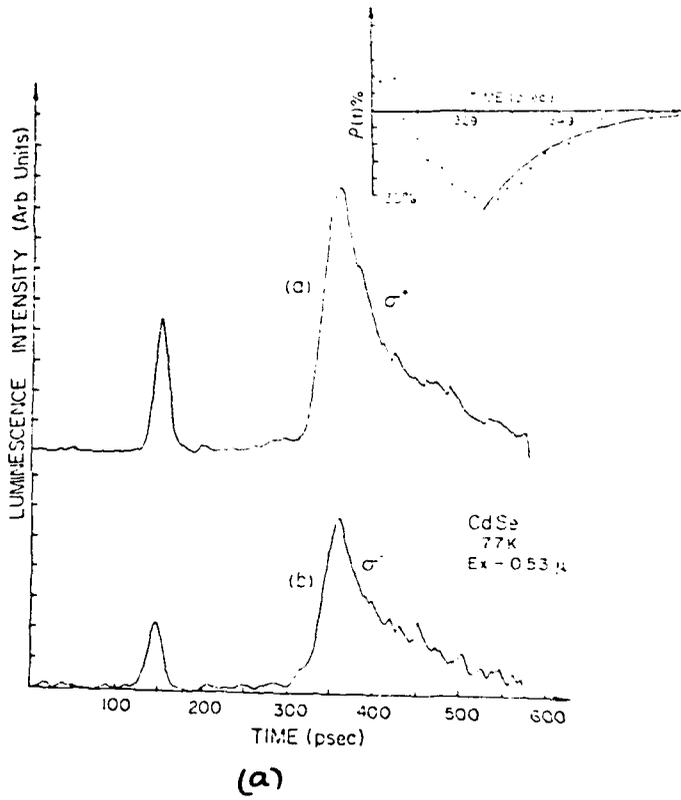
Here we review the first direct measurements of spin relaxation times in wurtzite CdSe and semi-magnetic semiconductor CdMnSe. Our past work was on spin relaxation and alignment in GaAs which was previously reported to you.

The figures 4a, b, and c show time resolved photoluminescence from CdSe and magnetic semiconductor Cd_{0.95}Mn_{0.05}Se and Cd_{0.9}Mn_{0.1}Se. The photoluminescence has the degree of polarization on the order of 30% at t=0, which also corresponds to spin polarization factor of ~ 30%. Spin polarization factor is defined as $p(t) = \frac{n_{\uparrow} - n_{\downarrow}}{n_{\uparrow} + n_{\downarrow}}$ and decays in time with a single exponential. Each curve shows circular polarized luminescence for 6+ and 6- (right and left handed, respectively). The measured values of p(0) and spin relaxation time T_S in CdSe are ~ 45 and 30 psec, respectively. The measured values of p(0) and spin relaxation time T_S in CdMnSe with 5% Mn are 20% and 16 psec, respectively. The measured value of p(0) and spin relaxation time in CdMnSe with 10% Mn are 28% and 20 psec, respectively. The spin relaxation in pure CdSe is assigned to a mechanism due to non-centrosymmetric nature of crystal lattice. In CdMnSe the spin relaxation occurs due to two

equally important mechanisms - one due to non-centrosymmetry of lattice and the other due to spin exchange interaction between free electrons and localized Mn^{2+} ions.

The spin relaxation rates are plotted in the figure 4(d) and compared to a theoretical fit based on spin exchange interaction. More work is needed with higher concentration of Mn in CdMnSe in order to check the validity of the theoretical model.

Figure 4 Spin Dephasing time and polarization



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Ph.D. Thesis at the Institute for Ultrafast Spectroscopy
and Lasers

The City College of New York

| <u>TOPIC/DATE/FUNDING AGENCY</u> | <u>PRESENT POSITION</u> |
|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------|
| 1. "Picosecond and Steady State Spectroscopy of the Wurtzite Semimagnetic Semiconductor $Cd_{1-x}Mn_xSe$ " by Mahesh Junnarkar, 1986, AFOSR, NSF | IUSL |
| 2. "Time Resolved Spectroscopy of Ternary Semiconductors $GaAs_{1-x}P_x$ and $Ga_xIn_{1-x}P_x$ under Picosecond Laser Pulse Excitation" by Hassan J. Zarrabi, 1985, AFOSR | General Optronics |
| 3. "Picosecond and Steady State Spectroscopy of Defects in Semi-Insulating CdSe" by David L. Rosen, 1985, AFOSR | NRL |
| 4. "A Study of Energy Transfer in the Photosynthetic Blue-Green Algae Nostoc Sp. probed by Picosecond Spectroscopy" by Aaron Dagen, 1985, NSF | Perkin Elmer |
| 5. "Energy Transfer between Dye Molecules Investigated by Steady State and Time Resolved Spectroscopy" by Poyang Lu, 1982, AFOSR, NSF | IBM |
| 6. "The Emission and Absorption Characteristics of Chalcogenide Semiconductors using Steady State and Time Resolved Picosecond Spectroscopy" by Shingshwang Yao, 1982, AFOSR | IBM |
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| 8. "Carrier Transport in Amorphous Silicon utilizing Picosecond Photoconductivity" by Anthony M. Johnson, 1981, BTL | Bell Labs |
| 9. "Energy Transfer in the Primary Stages of the Photosynthetic Process investigated by Picosecond Time Resolved Fluorescence Spectroscopy" by Francesco Pellegrino, 1981, NSF | Sperry |
| 10. "Reorientational Relaxation Kinetics of Polyatomic Molecules in Different States of Condensed Media investigated by Picosecond Laser Pulse Induced Kerr Effect" by Ping-Pei Ho, 1978, NSF | CCNY Prof. EE |

Ph.D. Thesis in Progress

1. "Ultrafast Transient Diffraction Gratings of Photoexcited Carriers in GaAs Structures" by A. Katz, AFOSR
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