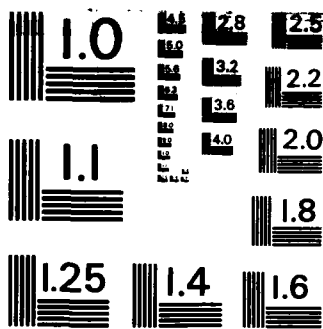


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Progress Report

December 1, 1982 - January 1, 1984

Semiconductors Investigated by Time Resolved Raman  
Absorption and Photoluminescence Spectroscopy  
Using Femtosecond and Picosecond Laser Techniques

Professor Robert R. Alfano  
Dr. A.G. Doukas

Department of Physics  
The City College  
New York, N.Y. 10031

Contract F49620-83-C-0027  
Air Force Office of Scientific Research  
Bolling AFB, DC 20332

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| 20. ABSTRACT (Continue on reverse side if necessary and identify by block number)<br>We report on the research performed during the period 1982-1983 under the auspices of AFOSR. The research effort follows two directions:<br>1) Laser development: subpicosecond laser, application of anti-resonant cavity to Nd:glass, study of the emerald laser, and study of a new mode-locking dye for shorter pulses.<br>2) Time-resolved fluorescence and absorption studies of CdCr <sub>2</sub> Se <sub>4</sub> , GaAs and Ga <sub>0.5</sub> In <sub>0.5</sub> P with the goal to understand the interaction and kinetics of photogenerated carriers and basic assignments of the valence-conduction |                       |   |

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20. band transitions ( $CdCr_2Se_4$ ). We have also investigated the dynamics of semi-insulating CdSe. <sup>2</sup> Finally we have continued the research on radiation damage ( neutrons and protons) in CdSe and GaAs.

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This progress report covers research supported by AFOSR 80-0079 performed during the period 1982-1983. The research effort has been concentrated in two directions:

- A. Laser development and
- B. Study of ultrafast processes in semiconductors.

A. Laser development program.

We have accomplished the following tasks:

1) The subpicosecond laser system (oscillator and amplifier) are operational.

2) A small ring mode-locked dye laser for testing purposes has been constructed and is also operational.

3) We have investigated an anti-resonant cavity for Nd:glass silicate rod. This cavity reduces the pulse duration to 2-3 psec from 8 psec normally produced in a conventional standing wave cavity. We are going to test phosphate rods as well as other mode-locking dyes (Kodak #5). We believe we can produce subpicosecond pulses from glass lasers.

4) We have observed lasing action from emerald and have investigated the underlying lasing processes associated with the vibronic transitions.

B. Semiconductor research program.

1) We have conducted a systematic investigation on  $\text{CdCr}_2\text{Se}_4$ , GaAs and GaSe using absorption and luminescence kinetic spectroscopy. In the magnetic semiconductor  $\text{CdCr}_2\text{Se}_4$  our measurements have resolved a long standing controversy over the assignments of the valence band to conduction band transitions, the electronic band structure, and the size of the fundamental gap.

2) We have observed that the hot photogenerated carrier distribution in both GaAs and GaSe cools at a slower rate at high electron-hole plasma densities. The slow kinetics arise from the screening of the electron-optical-phonon (GaAs) and hole-optical-phonon (GaSe) interactions. The deformation potential was calculated for carrier-phonon interaction in GaSe. The photoexcited carrier recombination in p-type GaAs was measured using the population mixing technique and the subpicosecond laser. The recombination of  $\text{CdCr}_2\text{Se}_4$  was measured to be less than 2 psec. The ultrafast recombination time may allow for the development of fastest optical switches at room temperature.

3) We have continued our research on radiation damage in semiconductors. Our work shows that  $\gamma$ -rays (600 KeV) and  $\beta$ -rays (14 MeV) bombardment at room temperature has not shown significant changes up to  $10^5$  rads. However, protons at 2 MeV and  $1.4 \times 10^{22} \text{ cm}^{-2}$  broadened the acceptor band, indicating the generation of new acceptors. The streak camera indicated an increase of free carrier lifetime from 50 psec to 70 psec, perhaps indicating a change in Fermi level (from semi-insulating to low resistivity n- or p-type). Further work is needed to find out whether these acceptors capture electrons like the native and chemical defects. GaAs luminescence was quenched by the same dose of protons, which defects capture the free carriers and how fast is still unknown.

4) The dynamics of semi-insulating CdSe was partly resolved by nonlinear luminescence and absorption. Two mechanisms that resist p-doping in CdSe are the chemical formation of deep donors from shallow acceptors, and the trapping of conduction electrons by

shallow acceptors by means of intermediate midgap states. The two main bands in CdSe are the free carrier band, whose luminescence is super linear in pump intensity, and the acceptor band, which is sub-linear in pump intensity in semi-insulating CdSe. It has been established that the shallow acceptor captures the conduction band electrons in semi-insulating CdSe, indicating that the second mechanism of resisting p-doping predominates. Subgap absorption was done to find deep states of the shallow acceptor. Time resolved work is needed to find which state is capturing electrons instead of emitting holes.

5) Semiconductor alloys like GaAsP, GaAlAs, GaInP are becoming increasingly important in the field of electron transport and electro-optical devices because one can adjust parameters such as bandgap and mobility for fabrication of multilayer heterostructures, quantum wells and lasers. Time resolved emission from electron-hole plasma in  $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$  were measured with a streak camera system at room temperature. The rise time of the emission is less than response time of detection system, approximately 10 ps. The decay time of emission is about 40 ps and does not change with excitation power. Time resolved emission spectra from electron-hole plasma were obtained at  $t=0$ ,  $t=8$ ,  $t=16$ ,  $t=24$  ps after excitation pulse. Analysis of time resolved emission spectra shows that high energy tail of emission reduces and emission shifts slightly to low energy. By fitting the emission spectra to theoretical expression for spontaneous recombination of electron-hole plasma the carrier temperature was determined to be  $380^\circ\text{K}$  at  $t=0$  and  $300$  at  $t=16$  ps. This gives an average value of  $1 \times 10^9$  eV/sec for carrier



energy relaxation rate due to scattering by LO phonons. Time integrated spectra of electron-hole plasma for different excitation power at room temperature and  $T=78$  K were measured. The peak of emission at RT for excitation power of  $26 \text{ MW/cm}^2$  is about 647 nm which arises from the recombination of electrons with heavy hole. As the excitation power increased the peak of emission shifted to higher energy. This is unusual because many body effects tend to renormalize the band gap to lower value. At  $180 \text{ MW/cm}^2$  the emission spectra were broad and showed an overlapping of two emission bands. In order to identify the nature of these two bands low temperature experiments were performed. Time integrated photoluminescence spectra of electron-hole plasma were measured at different excitation power. At low excitation power  $\sim 20 \text{ MW/cm}^2$  the peak of emission is at 640 nm. As excitation power increased another peak appears on the high energy side which is much broader than main emission band and grows in intensity with excitation power. These two bands can be explained qualitatively by recombination of high density screened holes photogenerated in split off valence band. At low excitation power the holes distributions are nondegenerate and emission mainly arises from recombination of electrons with holes in heavy band. The absence of high energy band at low excitation power is due to rapid thermalization of holes between valence bands where the probability of occupation for holes in heavy band will be higher than split off band by the Boltzman factor. The hole thermalization time is estimated to be in the order of a few ps. At high excitation power where hole distributions are degenerate and screening effect becomes important, two emission

bands were observed. At high carrier densities the screening of photogenerated carrier reduces the hole energy relaxation rates which give rise to the observation of high energy band. The intervalley transfer of holes from split off valence band to heavy band would slow down significantly so that a fraction of hole population remains long enough to give rise to the high energy band. In this case the emission spectra would consist of a superposition of two bands corresponding to the recombination of electrons with holes in heavy and split off valence bands.

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1. Photoluminescence Spectra of the Layered Semiconductor Gallium Selenide under Intense Picosecond Laser-Pulse Excitations, S. S. Yao and R. R. Alfano, Phys. Rev. B27, 2439 (1983).
2. Changes in the Photoluminescence Spectra of the Magnetic Semiconductor  $\text{CdCr}_2\text{Se}_4$  under High-power Picosecond-laser Excitation which determine that the Fundamental Gap is Direct, S. S. Yao and R. R. Alfano, Phys. Rev. B27, 1180 (1983).
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6. Phonon Enhanced Anti-Stokes Emission via Laser Action in Emerald, J. Buchert, A. Katz and R. R. Alfano (submitted).
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9. Optical Investigations of Chromium Chalcogenide Spinel Magnetic Semiconductors, W. J. Miniscalco, A. Lempicki, S. S. Yao, R. R. Alfano, N. G. Stoffel and G. Margaritondo, in Semiconductor Processes Probed by Ultrafast Laser Spectroscopy, R. R. Alfano ed., Academic Press (in press).
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11. Subpicosecond Dye Laser Technology, D. L. Rosen and B. Green, in Semiconductor Processes Probed by Ultrafast Laser Spectroscopy, R. R. Alfano, ed., Academic Press (in press).
12. Photoluminescence Dynamics of High Density Electron-Hole Plasma in  $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$  under High Power Picosecond Laser Pulse Excitation, H. Zarrabi and R. R. Alfano, SPIE (1983) (in press).
13. Native Acceptors in CdSe Studied through Nonlinear Luminescence and Mid-gap Absorption, D. L. Rosen, X. Li, and R. R. Alfano. Draft.

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4. Emerald Laser, J. Buchert, A. Katz, R. R. Alfano, Conference on Lasers and Electro-Optics, Baltimore, May 17-20 (1983).
5. Laser Action in Emerald, J. Buchert, A. Katz, R. R. Alfano, International Conference on Lasers, New Orleans, Dec. 13-17 (1982).
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8. Photoluminescence Dynamics of High Density E-H Plasma in  $\text{Ga}_{0.5}\text{In}_{0.5}\text{P}$ , H. Zarrabi and R. R. Alfano Proceedings of Society of Photo-optical Engineers, San Diego, Ca., August 24, 1983.
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