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This final technical report summarizes fourteen research accomplishments under grant AFOSR-86-0031 to understand the physics underlying ultrafast transient phenomena that occurs in semiconductor microstructures. This type of research is necessary to help advance and develop the future generation of ultrafast microelectronic devices.

A list of 25 publications is attached to the report.

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from

Robert R. Alfano, Distinguished Professor

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The City College of New York

June 29, 1990



This final technical report summarizes the accomplishments of our research under grant AFOSR-86-0031 to understand the physics underlying ultrafast transient phenomena that occur in the semiconductor microstructures. This type of research is necessary to help advance and develop the future generation of ultrafast microelectronic devices.

Microstructure samples were obtained primarily from H. Morkoc of University of Illinois, M. Niigaki of Hamamatsu Photonic KK, Rick Bertaska of McDonnell Douglas, Emil Koteles of GTE, and M. G. Spencer of Howard University.

In the following final report, we review our key accomplishments achieved during the funding period of 12/01/85 to 11/30/89 which were reported annually, followed by a list of total of 25 publications.

Fourteen accomplishments were made during this grant:

➤ **1. Nonequilibrium phonon effects on the energy relaxation and lifetime of photogenerated carriers in GaAs MQW;**

A large population of nonequilibrium longitudinal optical phonons produced in the energy relaxation process of hot carriers manifests itself by nonequilibrium phonon-stimulated phonon replica which is located at about 30 meV below  $n=1$  electron-hole transition. The energy relaxation is substantially suppressed due to the existence of nonequilibrium phonons after an initial rapid cooling. The number of photoexcited carriers decreases anomalously fast within the first 30 ps after the photoexcitation. An effective carrier depletion time is determined to be as short as 10 ps. A mechanism which leads to such a short carrier depletion time is associated with the nonequilibrium phonon-stimulated phonon replica.

## **2. Dependence of electron temperature on well width in AlInAs/GaInAs single quantum well**

From steady state electron temperature variation with well thickness single GaInAs/AlInAs quantum wells determined from photoluminescence spectra it is shown that the electron and longitudinal optical phonon scattering dominates the excess energy loss of the thermalized two dimensional electrons in the single quantum wells and the scattering rate is independent of well thickness within the experimental accuracy. The average energy loss rate per hot-electron was determined to be much smaller than expected.

## **3. Determination of band offsets in semiconductor heterolayer via optical transitions in ultrathin quantum wells**

The energy separation ( $\Delta E$ ) between the heavy and light hole subbands as function of well width for ultrathin quantum well structures was calculated using various boundary conditions. It was found that the most sensitive range for the well width to determine accurately the band discontinuities is between 15 to 80 Å, where it is insensitive for well width greater than 80 Å using optical transition energies in quantum wells.

## **4. Photogenerated of high-density electron-hole plasma energy relaxation and rapid expansion in CdSe**

Based on the fact that the polar optical phonon emission rate is reduced due to screening by high density of e-h plasma, the remaining dominant mechanism for hot carrier cooling is the nonpolar optical phonon emission even though CdSe is a highly polar semiconductor. It has been observed that the photogenerated carrier density is much lower than the estimated density using experimental conditions. Rapid plasma expansion has been proposed as a possible explanation on the grounds

of the observed larger spatial width of the photoluminescence than the laser spatial width, moderate change of Auger recombination rate with the excitation density, and earlier formation of excitons at low temperatures.

#### **5. Ultrafast photoluminescence kinetics from the magnetic semiconductor CdCrSe excited by femtosecond laser pulse**

The relaxation kinetics of carriers has been observed to change substantially above and below the Curie temperature of 130 K. A deviation from a monotonic increase in the recombination rate is measured as temperatures below the Curie temperature. This complex behavior is attributed to the interaction between the short range spin critical fluctuation of magnetic ions and the spin of the hot carriers. The fastest recombination time was measured to be 3.8 ps.

#### **6. Physics in semiconductor GaAs and GaSe under picosecond laser-driven shock-wave compression**

Using a pump-probe technique, the photoluminescence peak of bulk GaAs was observed to blue-shift and split into two under the laser-induced shock wave excitation. In GaSe, we observed a broadening of the spontaneous emission which is proportional to the shock pressure.

#### **7. Optical transition and recombination lifetime in quasi-zero dimensional electron system in $\text{CdS}_{\alpha}\text{Se}_{1-\alpha}$**

Optical transitions and their recombination lifetimes between quantized levels (1S, 1P) in the conduction and valence bands of quasi-zero dimensional electron systems were measured. We have also observed stimulated excitonic emissions in CdSSe quantum dots under picosecond UV excitations at room temperature.

8. Picosecond dynamics of exciton dissociation by neutral carbon acceptors in GaAs quantum wells

We have experimentally studied 2D exciton capture process by neutral carbon acceptors in unintentionally doped GaAs MQWs using picosecond time resolved photoluminescence spectroscopy. It is demonstrated that excitons can be captured by neutral carbon acceptors in a 55-Å GaAs MQW structure. A capture time is found to be about 250 ps and independent of temperature below 80 K.

9. Determination of  $\Gamma$ -X mixing effects on the escape time of electrons in resonant states of GaAs/AlGaAs double-barrier tunneling structures

We have carried out a theoretical study on the electron escape time in double barrier GaAs/AlAs structures by taking  $\Gamma$ -X mixing into account. When  $n=1$   $\Gamma$ -state in GaAs well coincides with  $n=1$  X-state in AlAs barrier they couple together and split into  $\Gamma$ -like and X-like states. Our results show that the escape time of  $\Gamma$ -like electrons can be several orders longer than that of a pure  $\Gamma$  system.

10. Determination of intervalley  $X_6$ - $\Gamma_6$  scattering time in GaAs by picosecond pump-probe infrared absorption spectroscopy

We have investigated for the first time the direct dynamics of electrons in the  $X_6$  valley for a GaAs crystal by time resolved absorption spectroscopy. IR picosecond probe pulses were used to monitor the growth and decay of electron population in the  $X_6$  valley subsequent to the excitation by a 527 nm pump pulse. The intervalley  $X_6 \rightarrow \Gamma_6$  scattering time of  $0.50 \pm 0.35$  ps was determined. The scattering cross section for the  $X_6 \rightarrow X_7$  transition was estimated to be  $4.5 \times 10^{-17} \text{ cm}^2$ .

### **11. Determination of the effective mass and energy minimum of the X7 satellite conduction band of GaAs**

For the first time, a X6-X7 absorption spectrum of GaAs was directly measured using picosecond pump-probe absorption spectroscopy. From this spectrum, the energy minimum and the density of state effective mass of the X7 band were determined to be 0.345 eV and  $0.48 m_0$ , respectively.

### **12. $\Gamma$ -L Intervalley scattering rates in GaAs measured by femtosecond time-resolved four-wave mixing spectroscopy**

To complement our work under #10 above, we have employed a three-pulse transient grating technique to study intervalley transfer dynamics in highly photoexcited GaAs. The femtosecond four-wave mixing signal exhibits a two component relaxation of different magnitudes for various probe energies. The fast relaxation mechanism is due to electron in the L valleys scattering back to  $\Gamma$  valley. The effective transfer time for  $L \rightarrow \Gamma$  was found to be  $\sim 2$  ps.

During the past year of grant AFOSR-86 0031, special attentions was paid to understanding the ultrafast hole dynamics in the GaAs epilayer and electron dynamics in multiple quantum well structures grown on silicon substrates. This research is discussed in the next two sections #13 and #14.

### **13. Hole dynamics in GaAs epilayer grown on Si substrate**

Based on the time-resolved photoluminescence from transitions of electron in donor (Si) state to split valence bands, "heavy hole" band with spin momenta  $m_j = \pm \frac{1}{2}$  and "light hole" band with spin momenta  $m_j = \pm \frac{3}{2}$  in GaAs/Si under

photoexcitation density below the donor density, the hole dynamics was investigated with the following result:

- (a) The intraband hole thermalization time for heavy hole is found to be shorter than that for light holes. An upper limit of 6 ps for light hole thermalization was estimated.
- (b) The time needed for two types of holes to be in equilibrium is found to be about 11 ps.
- (c) We are able to determine the hot hole cooling curve by theoretically modeling the measured spectra at different times. The cooling curve provides useful information about hole-phonon interactions. We found hole cooling rate for the sample was six times smaller than expected based on hole scatterings with equilibrium longitudinal optical phonons. It was found that nonequilibrium phonon population with a LO-phonon lifetime of 8 ps could account this apparent slow hot hole cooling.

#### 14. Electron dynamics in GaAs multiple quantum wells on Si

We have carried out time and energy resolved photoluminescence measurements on two p-modulation doped GaAs multiple quantum well (MQW) structures grown on Si substrates with 40 and 188 Å well thicknesses and fixed barrier width of 250 Å at different lattice temperatures and photoexcitation densities. Making use of the built-in strain in GaAs MQW due to thermal coefficient difference between GaAs MQW and Si substrate and the freedom to select well thickness and doping density, we are able to study electron dynamics with heavy holes and light holes in 188 Å and the 40 Å wells, respectively. Two main results are described as follows:

- (a) It was found that energy relaxation of photogenerated hot electrons in 188 Å-wells with doped-in heavy holes was much slower than in the 40 Å-wells with doped-in light holes under the condition of similar electron and hole densities. The reason for slow energy relaxation in 188 Å-wells was found to be the built-up of nonequilibrium longitudinal optical (LO) phonons that hot electrons interact with. This observation demonstrates a low electron energy relaxation process normally occur in modulation p-doped GaAs MQWs grown on GaAs substrate is due to weak energy exchange between hot electrons and heavy holes resulting in an enhancement of nonequilibrium LO phonon population. With freedom to choose the mass of doped hole, we are able to switch off the nonequilibrium LO phonon effect on hot electron energy relaxation process in the 40 Å-wells.
- (b) Since the Fermi-level for the doped-in light holes is very close but slightly below the heavy hole subband edge in the 40 Å-wells, this allow us to determine both electron and hole temperatures with an accuracy 0.5 K. We have measured the electron and hole temperature cooling curves in ranges of 32 to 4 K and 8 to 4 K, respectively. By fitting these cooling curves we have determined the electron-hole energy exchange rate to be  $0.05(T_e - T_h) \text{ K} / \text{ps}$  in the measured temperature regions. It was also found that the light hole and acoustic phonon interaction was two orders of magnitude stronger than expected.

List of publications under AFOSR-86-0031 support (1986-1990)

- (1) "Determination of valence-band discontinuity via optical transitions in ultrathin quantum wells", Kai Shum, P. P. Ho, and R. R. Alfano, Phys. Rev. B 33, 7259(1986)
- (2) "Dependence of electron temperature on well width in the  $\text{Al}_{0.48}\text{In}_{0.52}\text{As}/\text{Ga}_{0.47}\text{In}_{0.53}\text{As}$  single quantum well", Kai Shum, P. P. Ho, Alfano, D. F. Welch, and L. F. Eastman, IEEE Journal of Quantum Electronics, QE-22, 1811(1986).
- (3) "Photogenerated high-density electron-hole plasma energy relaxation and experimental evidence fro rapid expansion of the electron-hole plasma in CdSe", M. R. Junnarkar and R. R. Alfano, Phys. Rev. B 34, 7045 (1986).
- (4) "Temperature dependence of the ultrafast photoluminescence kinetic from the magnetic semiconductors CdCrSe excited by femtosecond laser pulse", P. P. Ho, W. Lam, A. Katz, S. S. Yao, R. R. Alfano, IEEE J. Quant. Elec. QE-22, 205 (1986).
- (5) "Spin dephasing kinetics of free carriers in alloy semimagnetic semiconductor CdMnSe by one and two photon excitation", M. R. Junnarkar and R. R. Alfano, *Ultrafast Phenomena V*, ed. A. E. Siegman, pp. 251, Springer Verlag (1986).
- (6) "Ultrashort spontaneous lifetimes for transitions in quasi-zero dimensional electron systems in  $\text{CdS}_x\text{Se}_{1-x}$ ", Kai Shum, G. C. Tang, M. R. Junnarkar, and R. R. Alfano, Appl. Phys. Lett. 51, 1839 (1987).
- (7) "Energy relaxation and ballistic diffusion of photoexcited carriers in symmetric and asymmetric quantum wells", Kai Shum, M. R. Junnarkar, H. S. Chao, R. R. Alfano, and H. Morkoc, SPIE 793, 6 (1987).

- (8) "Confinement effects on the scattering of electrons by polar optical phonons in semiconductor quantum wells", Kai Shum and R. R. Alfano, SPIE 793, 70 (1987).
- (9) "Optical transitions and recombination lifetimes in quasi-zero dimensional electron system in  $\text{CdS}_x\text{Se}_{1-x}$ ", Kai Shum, G. C. Tang, M. R. Junnarkar, and R. R. Alfano, SPIE 793, 150 (1987).
- (10) "Shock-wave-induced collision broadening of the photoluminescence spectra in GaSe", X. Z. Lu, S. Lee, R. Garuthara, and R. R. Alfano, Appl. Phys. Lett. 51, 1789 (1987).
- (11) "GaAs photoluminescence under picosecond-laser-driven shock compression", X. Z. Lu, R. Garthara, S. Lee, and R. R. Alfano, Appl. Phys. Lett. 52, 93 (1987).
- (12) "Reply to [Comment on 'Determination of valence band discontinuity via optical transitions in ultrathin quantum wells']", Kai Shum, C. Zhang, P. P. Ho, and R. R. Alfano, Phys. Rev. B 37, 1408 (1988).
- (13) "Nonequilibrium phonon effects on the time-dependent relaxation of hot carriers in GaAs MQW", Kai Shum, M. R. Junnarkar, H. S. Chao, R. R. Alfano, and H. Morkoc, Solid-state Electronics 31, 451 (1988).
- (14) "Effects of nonequilibrium phonons on the energy relaxation and recombination lifetime of photogenerated carriers in undoped GaAs quantum wells", Kai Shum, M. R. Junnarkar, H. S. Chao, R. R. Alfano, and H. Morkoc, Phys. Rev. B 37, 8923 (1988).
- (15) "Ultrafast processes in quasi-zero-dimensional semiconductor particles in glasses", Kai Shum, R. R. Alfano, SPIE 794, (1988).
- (16) "Intervally scattering rates in GaAs measured by time resolved four-wave mixing spectroscopy", A. Katz and R. R. Alfano, Appl. Phys. Lett. 53, 1065 (1988).

- (17) "Resonant level lifetime in GaAs/AlGaAs double-barrier structures in the consideration of  $\Gamma$ -X mixing", K. Shum, T. F. Zheng, M. Lax, R. R. Alfano (submitted to Appl. Phys. Lett., 1990).
- (18) "Picosecond dynamics of exciton capture by neutral carbon acceptors in GaAs quantum wells" K. Shum and R. R. Alfano (Submitted to Phys. Rev. B).
- (19) "Observation of stimulated photon emission in CdSeS/glass spherical quantum well structures under picosecond UV excitation", K. Shum and R. R. Alfano (in preparation).
- (20) "Hydrogen-molecule-like complex in undoped GaAs quantum wells", K. Shum and R. R. Alfano, (submitted to Phys. Rev. Lett.).
- (21) "Picosecond hole dynamics in GaAs grown on silicon", Kai Shum, Y. Takiguchi, J. M. Mohaidat, F. Liu, and R. R. Alfano Appl. Phys. Lett. 56, 2328 (1989).
- (22) "The intervalley X6- $\Gamma$ 6, L6 scattering times in GaAs measured by ultrafast pump-probe infrared absorption spectroscopy", W. B. Wang, N. Ockman, M. Yan, and R. R. Alfano, Solid State Electron., 32, 1337 (1989).
- (23) "Hot hole energy relaxation dynamics in GaAs grown on Si", SPIE, (1990).
- (24) "Determination of the intervalley X6- $\Gamma$ 6, L6 scattering times and the density of states effective mass of X7 band in GaAs by picosecond time resolved absorption spectroscopy", W. B. Wang, N. Ockman, M. A. Cavicchia, M. Yan, and R. R. Alfano, SPIE, (1990).
- (25) "Determination of the density of states effective mass and the energy minimum of the X7 satellite conduction band in GaAs from the X6-X7 absorption spectrum", W. B. Wang, N. Ockman, M. A. Cavicchia, and R. R. Alfano, Appl. Phys. Lett. 57, (1990).

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4. H. S. Chao (Graduate Student)
5. A. Katz (Research associate)
6. S. Lee (Graduate student)
7. W. B. Wang (Graduate Student)
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2. Kai Shum (08/87)
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