Investigation of Quantum Effects in Heterostructures

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This report summarizes the most important results obtained under grant #DAAL03-90-C-0008, namely:

- Growth and characterization of a new class of III-V based diluted magnetic semiconductors
- Observation of one-dimensional conduction on surface channels of INAS
- Demonstration of TAMM states in semiconductor superlattices
- Realization of electric-field tunable stimulated emission
- Demonstration of resonant interband magnetotunneling in type II heterostructures
- Measurement of dynamic spin polarization in diluted magnetic semiconductor quantum wells

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FINAL REPORT

EMILIO E. MENDEZ

August 20, 1993

U. S. ARMY RESEARCH OFFICE
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A) STATEMENT OF WORK

In the last twenty years, quantum structures prepared epitaxially with prescribed configurations of semiconductor materials have exhibited unprecedented transport, optical and magnetic properties, which have stimulated device inventions contributing to innovations for information and communication systems. Investigation in quantum wells and superlattices has resulted in a fertile field that now constitutes about one third of world-wide semiconductor research and whose impact goes beyond semiconductor materials to reach systems involving metals and insulators.

Under this grant we have focussed on the search in semiconductor heterostructures of novel properties that contribute to the fundamental understanding of these materials as well as to applications in information-handling technologies.

In our original proposal the work was divided in five categories that reflect the interdisciplinary character of the field and of our own research: molecular beam epitaxy, reduced-dimensionality structures, optoelectronics, resonant tunneling, and diluted magnetic semiconductors. In the following, we will summarize our most important results using the same five categories.

B) MOST IMPORTANT RESULTS

1. MOLECULAR BEAM EPITAXY (see also DILUTED MAGNETIC SEMICONDUCTORS)

The technique of molecular beam epitaxy underlies our basic and applied physics work and therefore is central to our effort. In addition, we have done materials science research in selected areas of special relevance for applications.

- Growth Mode and Dislocation Distribution in ZnSe/GaAs System
  With the goal of understanding and controlling the growth mechanisms of wide-gap II-VI materials for their optimization regarding lasing action, we have investigated the dependence of the dislocation characteristics on the initial growth mode of ZnSe epilayers on GaAs (001). For the case where the initial growth proceeds by the formation and coalescence of three-dimensional islands, the threading dislocation density has been found to be an order of magnitude higher and the misfit dislocation length much shorter than that for the case where the initial growth is by a two-dimensional layer-by-layer mode. The growth modes can be controlled by the initial conditions in terms of the Zn/Se fluxes and the substrate temperature. In addition, we have found that three-dimensional island growth of ZnSe is effectively suppressed by placing a sub-monolayer (a 1/4 monolayer) of Zn on the GaAs surface prior to the ZnSe growth, which results in a drastic reduction of threading dislo-
cations. The Zn sub-monolayer is believed to play an important role in changing the surface tension, which might be generalized in terms of modified surfactant effect. Measurements from photoluminescence and Raman scattering have confirmed the high quality of the ZnSe films with low dislocation density grown with the two-dimensional mode.

2. TRANSPORT IN REDUCED-DIMENSIONALITY STRUCTURES

The following two examples highlight our research in in-plane transport, using an original one-dimensional channel on an InAs surface or a two-dimensional interferometer provided by a double-gate structure.

- **One-Dimensional Electron Conduction on Surface Channels**

  The Fermi-level of an InAs surface is known to be pinned above the conduction band edge. On the other hand, GaAlSb has been found to form electron barriers with its surface Fermi-level pinned within the energy gap. Making use of these properties, we have fabricated GaAlSb/InAs/GaAlSb structures and achieved one-dimensional electron conduction on the cleaved surface channel of InAs. The width of the channel is given by the thickness of the InAs layer, and the one-dimensional structure is completed after cleaving without further processing. The resistance along the channel was measured as function of temperature and magnetic field. Both strong negative magnetoresistance and conductance fluctuations were observed. Analyses of the results led to a consistent evaluation of the phase coherent length, demonstrating the behavior of one-dimensional electron conduction.

- **Interference and Dephasing of Ballistic Electrons**

  Electronic interference phenomena in the diffusive regime, where the elastic mean free path is the shortest relevant length scale, have been extensively studied. Examples include Ahronov-Bohm oscillations and universal conductance fluctuations. We have investigated the opposite limit in the ballistic regime by use of high-mobility, two-dimensional electron systems. A modified Young’s double-slit experiment was realized by a double-gate structure between an emitter and a probe electrode on GaAs-GaAlAs. Quantum interference was observed by applying gate voltages to modulate the wavelength and hence the accumulated phase change of the electrons. The measured dephasing length agrees well with the calculated electron-electron scattering length in an ideal two-dimensional gas. In contrast with the diffusive regime, electron dephasing here occurs via a single scattering event with an energy exchange on the order of the carrier excess energy.
3. **OPTOELECTRONIC PROPERTIES AND APPLICATIONS**

The emphasis has been on the effects of an electric field on the optical properties of quantum wells and superlattices, and their use as wavelength-tunable light sources.

- **Electron Coherence and Localization in Superlattices**

  We have shown previously that the application of an electric field breaks the superlattice minibands into Stark ladders and leads to electron localization. The number of observed Stark states is a good measure of the coherence of the electron states. Using optical spectroscopies on a variety of GaAs-GaAlAs superlattices, we have found that the coherence length increases drastically as the superlattice period and the electric fields are decreased, reaching a value well beyond 500 Å. The effect of localization, meanwhile, results in a sharp increase of the exciton binding energy, as the minibands narrow to discrete states, giving rise to a two-dimensional character of the excitons.

- **Tamm States in Superlattices**

  Intrinsic surface states, arising as a result of termination of periodicity at a crystal surface was first proposed by Tamm in 1932. They were never clearly observed because extrinsic effects usually dominate at the surface. In this work, we created such states in a controllable fashion by a terminating layer of AlAs in a GaAs-GaAlAs superlattice. The formation of the Tamm states was manifested by excitonic interband transitions in photoluminescence excitation spectra. Critical confirmation was provided by photocurrent experiments under an electric field, which showed additional transitions and anti-crossing interactions between the Tamm states and the Stark-ladder states associated with the superlattice. The work demonstrated the ability to use semiconductors to study a prescribed model surface in a controllable way.

- **Landau and Stark Quantization in Superlattices**

  The effects of a magnetic field on the Stark-ladder transitions in GaAs-GaAlAs superlattices were studied using photocurrent spectroscopy. For the field perpendicular to the interface, the formation of Landau ladders associated with each Stark transition was observed, thus allowing the determination of the binding energies. The interplay between Landau and Stark quantizations is clear from anticrossings among the different ladder states when the magnetic and electric energies are equal. The coupling is ascribed to the electron hole Coulomb interaction and the valence-band mixing. For parallel magnetic fields, the excitonic Stark transitions broaden and their intensities decrease until they eventually vanish. At fields above 17 T, new interband transitions appear, associated with magnetic states in both the valence and conduction bands. This behavior can be understood in terms of the
competition between electric-field and magnetic-field induced localization, and the dominance of the latter at the highest magnetic fields.

2. Fermi-Edge Singularity in GaAs-GaAlAs Quantum Wells
Pronounced many-body exciton effects have been observed through photoluminescence in modulation-doped n-type GaAS-GaAlAs single quantum wells. The structure was designed to achieve a near-resonance coincidence between a transition involving two-dimensional electrons at the Fermi level in the first conduction subband and an exciton transition from the second conduction subband. Under these conditions, strong and distinct Fermi-edge singularities are observed. A magnetic field induces further enhancement of the effect and leads to strong oscillations in the luminescence. The large amplitude variations reflect the stability of the exciton in terms of the position of the Fermi level relative to the Landau levels. Time-resolved spectra are consistent with these results.

3. Electric-Field-Tunable Coupled-Quantum-Well Laser
By optically pumping a laser heterostructure whose active region consists of coupled quantum wells, we have succeeded in tuning the stimulated emission by external electric fields. This was made possible by the relatively large energy shifts of the quantum states with the field in such coupled structures. Using typical GaAs-GaAlAs (Al=0.23) with two 50Å wells and a 20Å barrier, a range of tuning of 70Å has been achieved at low temperatures. Effects due to carrier screening and tunneling ultimately limit the tuning range. With optimized structural design to ensure the dominance of spatially indirect transitions between the wells and to minimize carrier leakage, operation at higher temperature and with wider tunable ranges should be possible.

4. RESONANT TUNNELING
We have exploited the uniqueness of the Γ-X band alignment in GaAs-AlAs heterostructures and the band ordering in the InAs-GaSb system to study new mechanisms that shed light on the physics of resonant tunneling and on quantum energies of two-dimensional states.

4.1 Electron Tunneling Between Low Dimensional Systems
The studies of electron tunneling usually involve three-dimensional systems. Two-dimensional states appear in the quantum well in a double-barrier structure or at the emitter electrode adjacent to the barrier under proper bias conditions. The tunneling between two regions of two-dimensional systems shows a rather peculiar behavior: electrons can flow only at a singular voltage to satisfy the conservation of parallel momentum. This voltage corresponds to the energy difference between the two
states, and it becomes zero for a symmetrical structure. With a double barrier of GaAs-AlAs under hydrostatic pressure, a sharp feature near zero bias was observed, which was interpreted as tunneling between two-dimensional states in AlAs through GaAs as defined by the X-valley potential profile. In general, singularities in the current-voltage characteristics will be present for electron systems with dimensionality lower than three.

- **Resonant Interband Magnetotunneling In Type-II Heterostructures**

The unique situation of energy gap separation in InAs-GaSb, known as type-II structure, has led recently to extensive studies of resonant tunneling in this system using AlSb as a suitable barrier to form a polytype heterostructure. By applying a magnetic field parallel to the tunneling current we have demonstrated how resonant tunneling can spectroscopically resolve the Landau levels of a two-dimensional gas in a strong magnetic field. Besides allowing the determination of cyclotron energies and non-parabolicity effects, the spectroscopic analysis of Landau levels reveals for the first time the charge-transfer process between InAs and GaSb induced by the magnetic field. In addition, the experiments have connected in-plane and vertical transport by the observation that the tunneling conductivity shows zero-conductance regions at certain magnetic fields, in analogy to the zero-resistance regions of the in-plane quantum Hall effect, thus opening the door to the demonstration by tunneling of the fractional quantum Hall effect.

- **Oscillatory Landé g factor of Two-Dimensional Electrons**

Using resonant-tunneling spectroscopy in magnetic fields up to 30T, we have shown that the Landé factor, g, of two-dimensional electrons oscillates with field between its three-dimensional value and a value strongly enhanced by many-body effects. The experiments were carried out at 1.4K in GaSb-AlSb-InAs-AlSb-GaSb type II heterolayers, in which holes tunnel resonantly between two GaSb electrodes through magnetic states in the conduction band of an InAs quantum well. The density of these two-dimensional electrons was controlled by the application of external hydrostatic pressure. Under a field perpendicular to the layers, the current-voltage characteristics of a structure with an InAs well 150Å wide showed negative-differential-resistance features associated with Landau levels in the well. Above 16T the peak corresponding to the N=0 level split into its two spin components, from which the g factor was determined. For the largest electron density (N_s = 1.2 x 10^{12} cm^{-2}) the g factor oscillated with field between a minimum value of 8 and a maximum of 15. Variations of the same order were observed for lower densities, down to N_s = 5.2 x 10^{10} cm^{-2}. This large enhancement of the g factor confirms a twenty-year old theoretical prediction and shows quantitatively the importance of
many-body effects in 2D systems, which are at the core of the field-induced oscillations.

5. DILUTED MAGNETIC SEMICONDUCTORS

Our effort in this area has concentrated on the electronic properties of CdTe-CdMnTe quantum wells and on the preparation and characterization of a new family of III-V based magnetic semiconductor compounds exemplified by InMnAs.

- **Dynamic Spin Polarization In Diluted Magnetic Semiconductor Quantum Wells**
  Charge-carrier spin scattering in CdTe-CdMnTe multiple quantum wells was directly observed through circularly polarized luminescence in time-resolved experiments in the femtosecond regime. Separate observations of electron and hole relaxation were also accomplished by varying an external magnetic field. Surprisingly, the spin-flip scattering in these systems appears to be independent of the extremely strong spin-spin exchange interaction between their charged carriers and the magnetic ions, which we have calculated theoretically. Instead, the scattering time seems to be predominantly determined by the dimensionality of the electron system.

- **Heterostructure Type Transition Induced by a Magnetic Field**
  The application of a magnetic field changes the band structure of a semiconductor, in particular, the relative band edge positions of the two component materials in a heterostructure. The effect is especially pronounced when the band edges are close in energy and the spin splittings are large. Both are satisfied in the CdTe-CdMnTe system. With a rather modest field, this ordinarily type I structure becomes type II, in that, although electron states remain in CdTe, ground hole states are confined in CdMnTe. The emission energies and intensities from luminescence experiments clearly reflect this kind of type transition in heterostructures. Of more interest is the simultaneous change in the nature of the excitons. The spatial separation of carriers in the type-II system drastically reduces the binding energy, which must be taken into account to understand the highly nonlinear behavior of the emission energy versus the magnetic field.

- **Superlattice in CdTe-CdMnTe Heterostructures**
  In a quantum heterostructure, the discrete states in isolated wells become coupled to form superlattice minibands as the barrier thicknesses are reduced. The realization of this situation has been demonstrated in CdTe-CdMnTe from both optical and electro-optical experiments. In the former case, Raman scattering profiles showed broadened resonances with peak energies shifting to lower values with decreasing CdMnTe layers, as a consequence of the finite miniband width. Under applied electric field, the miniband narrows in energy and breaks into a series of Stark ladder
states. The luminescence spectra observed under these conditions exhibited intra-well transitions at increasingly higher energies with fields, and also inter-well transitions corresponding to the first ladder. That no other ladders were detected, in contrast to the GaAs-GaAlAs system, was a reflection of the relatively short coherent length in the present case.

- **Diluted Magnetic Semiconductors**
  A new class of III-V based diluted magnetic semiconductors has been synthesized by molecular beam epitaxy. As exemplified by InMnAs, the incorporation of Mn can be as large as 20%, and both paramagnetic alloys and ferromagnetic clusters can be prepared, depending on the growth temperature. The paramagnetic alloy is typically n-type and its energy gap decreases with Mn composition. Diluted magnetic semiconductor materials have long been of great interest, for they combine two large branches of condensed-matter physics. This work will allow to pursue many interesting electronic and magnetic properties, up to now limited largely to II-VI compounds, in the much better controlled and far more important III-V semiconductors.

- **Magnetotransport Properties of p-type InMnAs**
  P-type InMnAs, grown by molecular beam epitaxy with substrate temperatures above 275°C and Mn compositions between 0.1 and 0.3%, exhibited striking magneto-transport properties, namely, hysteretic behavior of the resistance at low temperatures and a dominant anomalous Hall resistance throughout the 0.4K-300K temperature range. The former suggests the occurrence of ferromagnetic order caused by a cooperative phenomenon accompanied by hole localization, in contrast to the conventional Ruderman-Kittel-Kasuya-Yoshida mechanism. The anomalous Hall effect originates from the interaction between the holes and the 3d spins of the Mn ions. Remnant magnetization, unsaturated spins and large negative magnetoresistance were also observed at low temperatures, results all explained by the formation of large bound magnetic polarons.

- **Diluted Magnetic Semiconductor Heterostructures**
  As a next step to exploit the novel properties of the new III-V based diluted magnetic semiconductors, p-(In,Mn)As/(Ga,Al)Sb heterostructures were successfully grown by molecular beam epitaxy on GaAs(100) substrates. We found that the structures, especially with thin (In,Mn)As layers (5-30nm), exhibit strong galvanomagnetic and magneto-optical effects due to perpendicular ferromagnetic order in InMnAs. Polar Kerr rotation characteristics indicate that the rotation is relatively large (0.2°) and has complex wavelength and thickness dependence. The origin of ferromagnetic order is probably a combination of carrier-induced and magnetoelastic effects in biaxially-strained (In,Mn)As layers.
C) LIST OF PUBLICATIONS

• Magnetic Field Induced Type-I to Type-II Transition in a Semi Magnetic CdTe/CdMnTe Superlattice, Phys. Rev. B42, 5891 (1990).
• Competition Between Magnetic-Field- and Electric-Field-Induced Localizations in GaAs/Ga0.65Al0.35 As Superlattices, Phys. Rev. B 44, 13124 (1991).


Effect of Surface Tension on the Growth Mode of Highly Strained InGaAs on GaAs(100), Appl. Phys. Lett. 62, 46 (1993).


Structural Quality and the Growth Mode in Epitaxial ZnSe/GaAs(100), J. Appl. Phys. 73, 2294 (1993).


D) SCIENTIFIC PERSONNEL SUPPORTED BY THE PROJECT

E) INVENTIONS