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FINAL REPORT

CONTRACT NO.: ONR N00014-81-K-0430 (May 1, 1986- Oct. 30, 1988)

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RESEARCH PROGRESS

Work accomplished in the last funding period is summarized below:

(A) Electronic Properties of Semiconductor Superlattices

Semiconductor superlattices have drawn substantial interest in the last decade. In an effort to understand the electronic structures, we have developed some new computational methods, including the muti-band effective-mass method, the Wannier-orbital method[7] and the bond-orbital method[15]. We have used these methods to calculate the detailed band structures of many semiconductor superlattices, including GaAs-AlGaAs, GaSb-AlSb, and InGaAs-InP systems. The muti-band effective-mass method is for calculating quantum-well confined states derived from bulk states near the band extrema and can be used to study acceptors[4,10] and excitonic states[3,9,21] in quantum wells, the Wannier- orbital method is most suitable for understanding the inter-valley scattering in conduction-band states, and the bond-orbital method is ideal for studying the mixing of states in superlattices (or coupled quantum wells) derived from near zone-center bulk states.

The inter-valley scattering effect is important in short-period superlattices. For example, when the GaAs layer in a GaAs-AlAs superlattice is sufficiently thin ($< 30\AA$), the Γ-valley derived energy levels become close to the X-valley derived levels, causing substantial mixing (Γ-X mixing). Such an effect has been observed experimentally by applying an electric field to the superlattice. The effect may have useful applications in electro-optical devices. Mixing of Γ and L states can also exist in superlattices involing materials like GaSb, in which the L-valley is the lowest. Using the Wannier-orbital method, we have examined the the conduction band energies of (001) GaAs-$Al_xGa_{1-x}As$ and $Al_xGa_{1-x}As$-AlAs superlattices as functions of layer thicknesses, alloy composition, wave vectors, as well as external hydrostatic

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Re: N00014-81-K-0430 Final Report

Technical Reports Office:

Principal Investigator Yia C. Chang and the University of Illinois, Physics Department Business Office have received an inquiry from the University of Illinois, Grants and Contracts Office concerning whether we sent copies of the Final Report of N00014-81-K-0430 to the Defense Technical Information Center (DTIC). Our records indicate we sent the final report to ONR but are unclear concerning those copies that were to go to DTIC.

Thus, we are sending the required number of copies of the Final Report to you now. If you have not previously received these reports, I apologize for any inconvenience or problems this may have caused.

Sincerely,

Stephen F. Knell

Stephen F. Knell
Business Manager

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pressure.[7] We found that there can be substantial mixing between the Γ and (001) X-valley states. The amount of Γ-X mixing is extremely sensitive to the layer thicknesses. Due to symmetry considerations, the existence of such mixing depends critically on whether the barrier material (the material with higher Γ minimum and lower X-minimum) contains an even or odd number of atomic layers in each superlattice unit cell. We have also performed detailed theoretical studies of the L-valley derived states in (001) GaSb-AlSb quantum wells and superlattices.[19] We found that the intervalley mixing between the (1/2,1/2,1/2) and the (1/2,1/2,-1/2) L-valleys plays an important role in determining the electronic structures of GaSb-AlSb quantum wells.

The newly developed bond-orbital method was used to calculate the valence subband structures of GaAs-$Ga_{1-x}Al_xAs$ quantum wells grown in different directions ([001], [111] and [110])[20] and $In_{1-x}Ga_xAs$-InP strained-layer quatum wells.[23] High quality superlattices grown along the [111] and [110] directions are available only very recently, Our calculations demonstrate the differences in band structures for quantum wells grown along different directions. The $In_{1-x}Ga_xAs$-InP strained-layer quatum well has important applications in quantum well lasers for optical communication. Our theroretical studies provide some guidelines for the engineering of related laser structures.

Most superlattices grown to date consist of semiconductor alloys (e.g. $Ga_xAl_{1-x}As$). If one uses an alloy as the well material in the superlattice, it is expected that the disorder effect on the electronic states will be important, because of the reduced dimensionality. We have developed an approximation technique which allows us to efficiently implement the coherent potential approximation (CPA) for treating the effects of disorder in alloy superlattices.[14] Using this method we computed the density of states of superlattices within both VCA (virtual crystal approximation) and CPA. Due to the two-dimensional nature of the bound states in superlattices, the effect of disorder is found to be much more pronounced for alloy superlattices than for bulk alloys. We find that the energy shifts due to disorder effect for typical superlattice states are around 10-100 meV.

(B) Optical properties of semiconductor quantum wells and superlattices

We have developed a new line-shape theory for photoabsorption in superlattices. The new method allows one to accurately and efficiently calculate the photo-absorption of a realistic superlattice, including excitonic effect, valence band mixing and inter-wellcoupling. The theory predicts realistic absorption spectra (without adjustable parameters) for GaAs-$Ga_{1-x}Al_xAs$ superlattices which are in exceelent agreement with the measured spectra. It is the predictive value that makes the calculation significant. The pred-

itions include some important physical phenomena such as the line shapes for saddle-point excitons and Fano resonances of discrete exciton states interacting with other continuum transitions. We have applied this newly developed line-shape theory to calculate the magneto-absorption spectra of GaAs-$Ga_{1-x}Al_xAs$ superlattces.[26] Our theory is valid for any strength of magnetic field; thus, we can follow the change of absorption spectra as a function of the magnetic field. We found interesting change of line-shape of the exciton resonances related to band-structure saddle-points due to magnetic field. Our theoretical spectra are in very good agreement with recent magneto-absorption measurements[R1]. Our calculations are useful for identifying the origin of many complicated features observed in the magneto-absorption spectra.

Figure 1 shows a calculated magneto-absorption spectrum and the corresponding experimental spectrum. As seen in the figure, the agreement between theory (with no adjustable parameters) and experiment is quite good.

(C) Phonons in Semiconductor Superlattices

We have performed detailed theoretical studies of phonon modes in semiconductor superlattices in an realistic rigid-ion model. We have calculated the dispersion curves for phonons propagating in any directions, taking into account the long-range Coulomb interaction.[12,16,25] Our theoretical studies provide the understanding of two key features of optical phonons in polar superlattices, namely the long wave-length anisotropy and the macroscopic interface modes. The theoretically predicted frequency splitting due to anisotropy is in good agreement with the available data[R2]

We have also developed a phenomenonlogical microscopic theory based on the rigid-ion model with simplified Coulomb interaction.[17] The theory provides some integral equations which allow easy calculation of the dispersion curves and displacement eigenvectors for long wave-length phonons in superlattices. The results obtained by the simple model agree well with those calculated by the full-scale rigid-ion model. This simple theory allows one to gain a deeper insight into the intriguing behavior of long wave-length optical phonons and the interface modes in polar superlattices.

(D) Nonlinear Optical Properties in Superlattices

Free carriers in p-type semiconductors can absorb light via intervalence-band transitions. As the intensity of incident radiation increases, the optical pumping rates become comparable with the relaxation rates, and the carrier distribution will deviate substantially from the thermal equilibrium distribu-

tion. The absorption will be saturated at sufficiently high intensity. In semiconductor superlattices, inter-subband transitions have nonlinear optical effects similar to those from bulk intervalence-band transitions. Inter-subband transitions can happen in both n-type and p-type semiconductor superlattices. The major difference between the two is that for n-type superlattices, the inter-subband optical matrix elements are appreciable only for incident radiation polarized along the growth direction, whereas for p-type superlattices the optical matrix elements are appreciable for incident light polarized in any directions. We have performed calculations of the saturation behavior of the inter-subband transitions in p-type semiconductor superlattices[6,24]. The nonparabolicity of the valence bands due to band-mixing effect is taken into account. Finite carrier lifetimes due to the carrier-phonon scattering are considered within the deformation potential approximation. Deviation of hole distribution from thermal equilibrium due to optical pumping is calculated by solving the coupled rate equations. We find that the linear absorption spectrum due to inter-subband transitions has a number of structures corresponding to transitions from the first few subbands (predominately the first heavy-hole subband) to higher valence subbands. We have also studied the absorption spectra of a probe beam in the presence of an intense pump beam. The pump beam can cause a steady-state non-equilibrium carrier distribution, thus substantially modifing the absorption spectra for the probe beam. We have obtained several interesting pump-and-probe absorption spectra and they may have possible applications for light-by-light modulation.

Change of index of refraction due to the applied electric field (electo-optic effect) has an important application in providing adjustable phase modulation of a light beam. We have examined theoretically the electro-optic effect in semiconductor superlattices[11]. Excitonic effects are considered for wide-barrier superlattices and are found to substantially enhance the electro-optic effect near the onset of the excitonic absorption. We also find that the electo-optic effect in small band-gap superlattices (such as GaInAs-InP systems) tends to be larger than that in the large band-gap (GaAs-AlGaAs) superlattices.

(E) Indirect Excitons and Electron-hole Liquids in InAs quantum wells

Under intense photon field, the two-component electron-hole plasma is expected to exist in quantum wells in various phases: electron-hole plasma, electron-hole liquid (EHL), exciton gas, biexciton (exciton molecule) gas, or Wigner crystal. In order to observe the various phases of the electron-hole plasma, the lifetime for the recombination of electron-hole pairs must be

longer than the relaxation time for reaching thermal equilibrium. This could be satisfied in semiconductor quantum wells with an indirect conduction band, such as Ge-$Si_{1-x}Ge_x$ systems. We have proposed a rather unique quantum well system with an indirect valence band which may also be a good candidate for observing the electron-hole liquid[1].

With the presence of a compressive uniaxial stress in a small-gap semiconductor quantum well (e.g. GaInAs-InP systems), the first light hole subband tends to move upward toward the first heavy hole subband. At a critical strain the energies of the two subbands coincide with each other, and the level repulsion between them can cause the first valence subband to curve upward, resulting in an indirect valence band. Thus, the lifetime for the electron-hole recombination become long enough for possible realization of the electron-hole liquid. The binding energy per electron-hole pair in the liquid phase will be enhanced due to the large hole density of states at the maximum. We have studied the lifetime and energy dispersion of the indirect exciton which consists of an electron localized at the zone center and a hole localized at the maximum of the indirect valence band[13]. We found that the lifetime of excitons can be lengthened by three to five orders of magnitude when the valence band goes from direct to indirect under the uniaxial stress. We have also investigated the possible formation of the electron-hole liquid in InAs quantum wells under compressive uniaxial stress. We found that the electron-hole liquid phase is stable against the exciton-gas phase within the random-phase approximation[1].

(F) Qunatum Monte Carlo Quantum Monte Carlo Study of Multiexciton Complexes

There has been considerable experimental research in recent years [R3] on bound multiexcitonic complexes, systems of several excitons bound to shallow impurities in Si, the stability of which is enhanced by the large degeneracy of low lying conduction and valence band states. Recently, evidence has also been found of the existence of free complexes of three and four excitons, or polyexcitons and it has been suggested that these complexes can account for a recently observed unusual condensed plasma photoluminescence band in Si[R4]. Theoretical studies of bound multiexciton complexes include configuration interaction [R5] and density functional[R6] approaches, which can predict transition energies but are less accurate in determining ground state properties of these systems, for which many-body correlations is important.

We have used the Variational Monte Carlo method, employing a Monte Carlo technique to evaluate variational integrals for many-body problems, to calculate the binding energies and ground state wavefunctions of two, three, and four multiexciton complexes, in the spherical effective mass ap-

proximation. (see Figure 2) We found the optimal form of the variational trial wavefunction to be a linear combination of Jastrow many-body wavefunctions, providing a description of a system intermediate between the high density electron-hole liquid and a complex of loosely associated excitons. The results for smaller complexes have been compared to those of Green's Function Monte Carlo calculations which produce exact ground state energies within the limits of the model used; the variational results recover about 90 percent of the binding energy of these systems. Our binding energies compare well with experimental results for Si[1,2], within the limits of our model, although central cell corrections are necessary to account for some qualitative trends.

FUTURE WORK

(i)We will study the atomic geometries and electronic structures of semiconductor surfaces. The theoretical studies can help understanding the properties of growth surfaces and providing guidelines for better crystal growth.
(ii)We will study the electron-phonon interaction in short-period superlattices. We will calculate the carrier lifetimes due to carrier-phonon scatterings. We will consider both the Frölich mechanism and the deformation-potential mechanism.
(iii)We will study the transient behavior of the photoluminescence excitation and resonant Raman scattering process in superlattices, taking into account the carrier dynamics.
(iv)We will combine the envelope-function technique with the bond-orbital method[15] to study the acceptor states in quantum dots, wires, and superlattices.
(v)We will continue our Quantum Monte Carlo calculations on multiple-exciton complexes, including the central-cell correction.

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5. **Theory of Nonlinear Optical Properties of Semiconductor Superlattices**, Y. C. Chang (invited paper), Proceedings of the SPIE Conference on Quantum Well and Superlattice Physics, Vol. 792, p. 134 (1987).

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7. **Γ-X Mixing in GaAs/$Al_xGa_{1-x}As$ and $Al_xGa_{1-x}As$/AlAs Superlattices**, D. Z.-Y. Ting and Y. C. Chang, Phys. Rev. B**36**, 4357-4374 (1987).

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Figure Captions

Fig. 1. Theoretical absorption spectra (bottom) and experimental photoluminescence excitation spectrum taken by Deveaud et. al.[R1] (top) for a ($50\mathring{A}$/ $50\mathring{A}$) GaAs-$Al_{0.24}Ga_{0.76}As$ superlattice. Dashed: heavy-hole contribution, dotted:light-hole contribution, solid: total. All theoretical spectra are broadened by $1.5meV$.

Fig. 2 (a)Energy of N-exciton complex minus energy of N free excitons, in units of exciton binding energy E(X), as function of the electron-hole mass ratio σ. Circles: N = 2; X's: N = 3; Triangles: N = 4. (b) Energy of N-exciton complex bound to a donor impurity, minus the energy of a donor and N free excitons, in units of donor binding energy E(D), as function of the electron-hole mass ratio σ. Circles: N = 1; X's: N = 2; Triangles: N = 3.

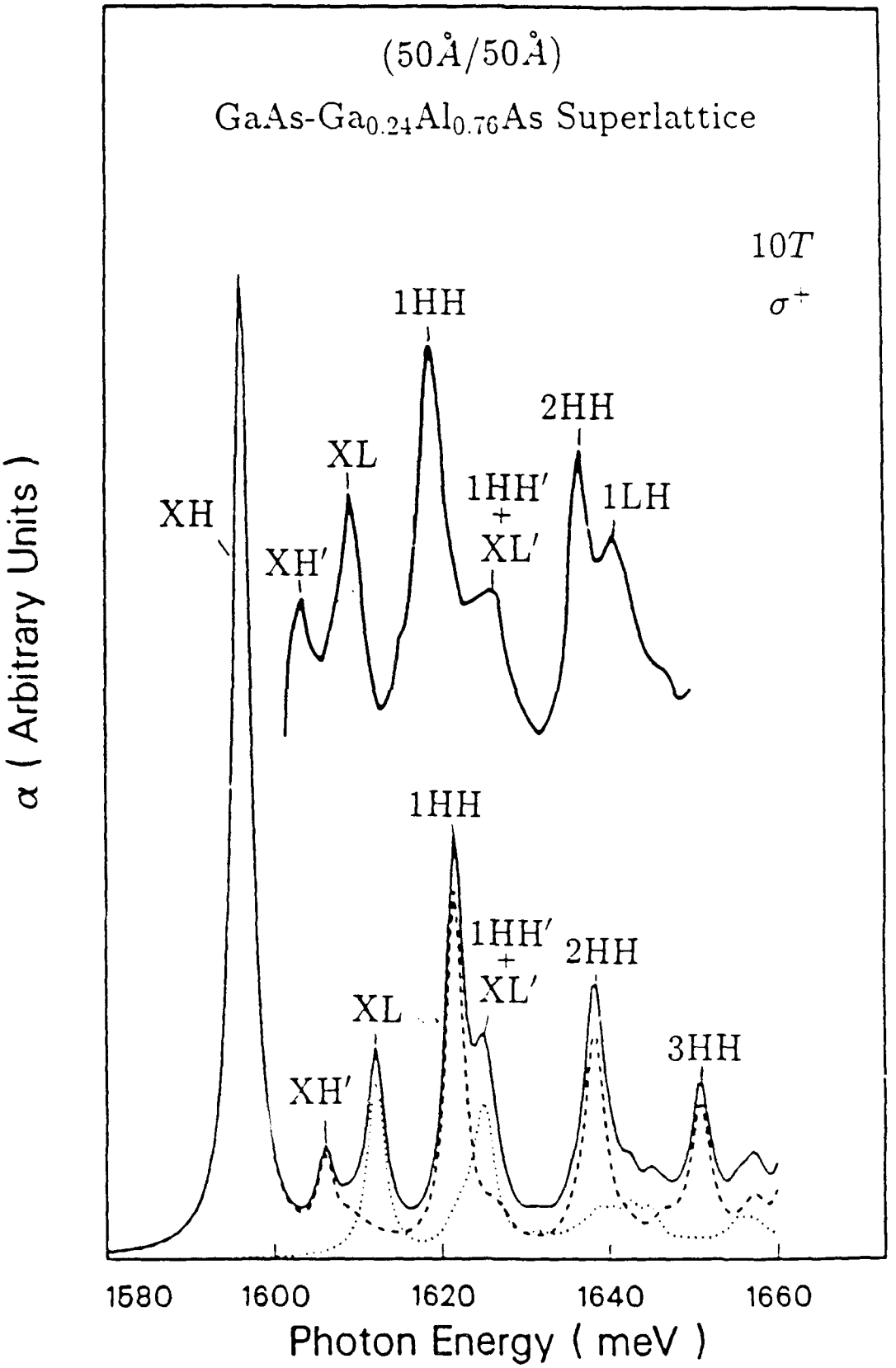
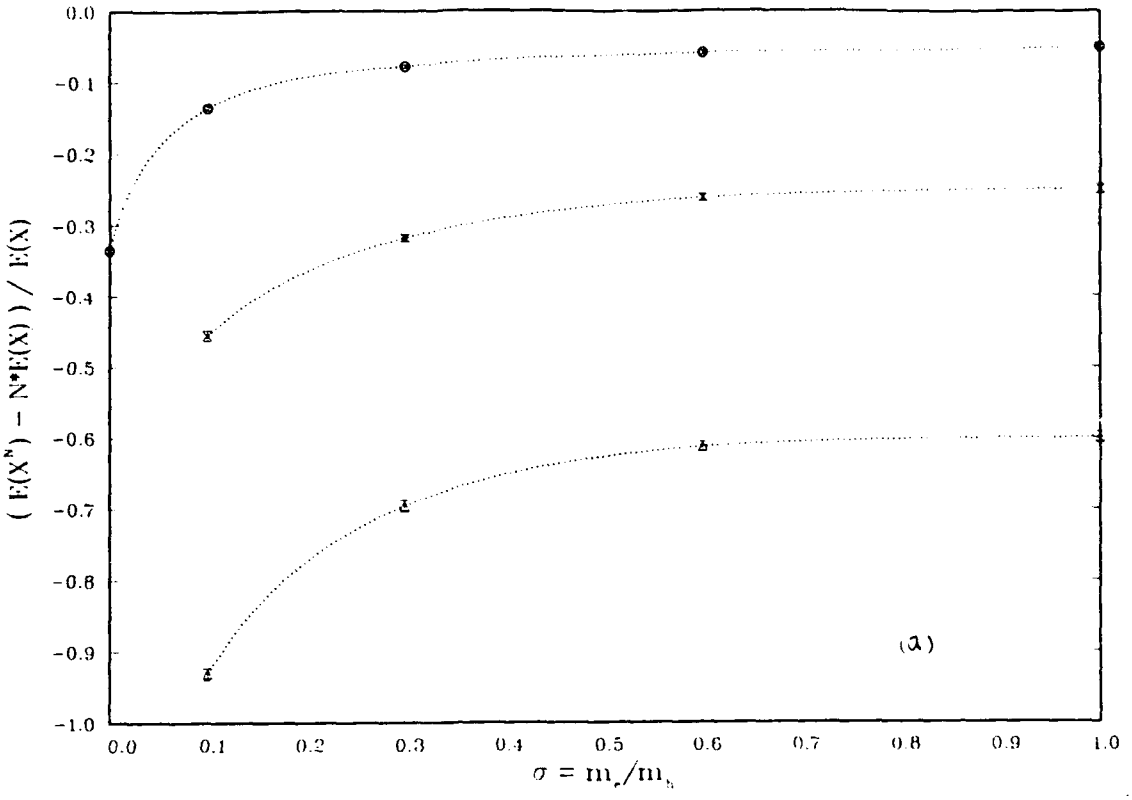

(50Å/50Å)
GaAs-Ga0.24Al0.76As Superlattice
10T
σ+
1HH
2HH
XL
XH
1HH′
+
XL′
1LH
XH′
1HH
1HH′
+
XL′
2HH
XL
3HH
XH′
α (Arbitrary Units)
1580
1600
1620
1640
1660
Photon Energy (meV)

Fig 1

Binding Energy of Multiexcitonic Complexes



Binding Energy of Donor Bound Multiexciton Complexes