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Optical properties in modulation-doped GaAs-$Ga_{1-x}Al_xAs$ quantum wells

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Optical properties of modulation-doped GaAs-$Ga_{1-x}Al_xAs$ semiconductor quantum wells are studied with the use of a multiband effective-mass theory. We consider the case where $Ga_{1-x}Al_xAs$ is doped with acceptors and the first GaAs valence band is populated with holes. Significant mixing of light- and heavy-hole states at points away from the zone center gives rise to interesting optical properties.

Semiconductor quantum-well heterostructures have played an important role in advances in semiconductor science and technology and a large number of optical measurements on quantum-well heterostructures have been made. Recent photoabsorption and excitation spectra of GaAs-$Ga_{1-x}Al_xAs$ superlattices have revealed that the selection rule $\Delta n = 0$ (where n is the principle quantum number) is not strictly obeyed.[1-4] Calculations of electronic properties of semiconductor quantum wells and superlattices have been performed using various methods.[5-13] Recently, optical properties of semiconductor superlattices have been calculated using a tight-binding method and the importance of heavy- and light-hole band mixing in determining optical properties was noted.[14]

In this paper, we report on calculations of detailed optical properties of modulation-doped quantum-well heterostructures based on a multiband effective-mass method which incorporates mixing of heavy- and light-hole states and the excitonic effect. It is found that light- and heavy-hole states with envelope functions of both parities are mixed and hence dipole-allowed transitions between all pairs of valence and conduction subbands occur. Thus the violation of the $\Delta n = 0$ selection rule in the observed spectra can be explained. It is further found that the valence-band structure is a complicated function of the width and depth of the well and the doping concentration. Our calculations show that for some values of the well parameters the second valence subband has a negative effective mass, in agreement with previous theoretical calculations.[13,14] Thus a singularity can occur in the joint density of states causing peaked structures to appear in the band-to-band absorption, and we find that the binding energies of excitons associated with these subbands are substantially enhanced.

For clarity, we consider a modulation-doped quantum-well structure in which the barrier material is doped with acceptors. At low temperatures the acceptors are ionized with the liberated hole carriers residing in the quantum well. We consider the low-doping case where only the first valence band is filled (with Fermi level E_F).

In our model, the effective-mass Hamiltonian for the electron is given by

$$H_e = \frac{P^2}{2m_e^*} + V_e(z) - V_d(z) \ ,$$

where m_e^* is the effective electron mass, $V_e(z)$ is a finite square well potential, and $V_d(z)$ is an additional parabolic potential of height V_0 which models the effect of modulation doping. We have chosen the z axis to be perpendicular to the quantum-well interfaces. The effective-mass Hamiltonian for the spin-$\frac{3}{2}$ hole operates on a four-component spinor wave function with column index $m = \frac{3}{2}, \frac{1}{2}, -\frac{1}{2}$, and $-\frac{3}{2}$. In the effective-mass approximation we have

$$(H_h)_{m,m'} = T_{m,m'} + [V_h(z) + V_d(z)]\delta_{m,m'} \ ,$$

where $V_h(z)$ is a finite square well potential. The kinetic energy matrix $T_{m,m'}$ is given in the limit of infinite spin-orbit splitting by the 4×4 $\mathbf{k} \cdot \mathbf{p}$ expression of Luttinger and Kohn.[15] Our treatment for the electronic structure is similar to that reported by Fasolino and Altarelli.[13]

Strictly speaking, the electrostatic modulation-doping potential should be obtained in a self-consistent Hartree-Foch calculation for holes in a confining well. The parabolic-potential approximation is equivalent to the assumption that there is a uniform density of holes in the well. Examination of the calculated hole ground-state wave function for low doping shows that the assumption of uniform hole density is fairly good. The relation between the potential V_0 and the two-dimensional hole concentration (hole column density) in the quantum well is given by $N = (1/4\pi e^2)(8\epsilon V_0/W)$, where ϵ is the static dielectric constant, W is the well width, and e is the electric charge of the hole.

To obtain the energies and envelope wave functions, the Schrödinger equations for the electron and hole are solved. Because of the translational symmetry along the x and y directions, the parallel wave vector $\mathbf{k}_\parallel = k_x\hat{\mathbf{x}} + k_y\hat{\mathbf{y}}$ is a good quantum number. The envelope functions for the electron and hole are in the Bloch form (letting $\boldsymbol{\rho}_\parallel = x\hat{\mathbf{x}} + y\hat{\mathbf{y}}$):

$$\psi^{e,h}_{n,m}(\mathbf{k}_\parallel, \boldsymbol{\rho}_\parallel, z) = e^{i\mathbf{k}_\parallel \cdot \boldsymbol{\rho}_\parallel}\Phi^{e,h}_{n,m}(\mathbf{k}_\parallel, z) \ ,$$

where the index n labels the subband, m labels the z component of the spin, and the labels e and h refer to electron and hole, respectively. Upon substitution of these expressions into the Schrödinger equation we obtain a set of equations to be solved for Φ^e and Φ^h.

The potentials V_e, V_h, and V_d are even in z and so $\Phi^e_{n,m}(\mathbf{k}_\parallel, z)$ and $\Phi^h_{n,m}(\mathbf{k}_\parallel, z)$ have definite parity. At $\mathbf{k}_\parallel = \mathbf{0}$, the hole Hamiltonian ($H_h$) is diagonal in the spin indices and the associated wave functions are pure spin-$\frac{3}{2}$ ($-\frac{3}{2}$) heavy-hole states or pure spin-$\frac{1}{2}$ ($-\frac{1}{2}$) light-hole states, whereas away from the zone center the presence of the off-diagonal terms results in mixing between heavy- and light-hole states. It can be shown that at any $\mathbf{k}_\parallel$, the two sets of states (even $\frac{3}{2}$, odd $\frac{1}{2}$, even $-\frac{1}{2}$, odd $-\frac{3}{2}$) and (even $-\frac{3}{2}$, odd $-\frac{1}{2}$, even $\frac{1}{2}$, odd $\frac{3}{2}$) are completely decoupled;

therefore, all valence subbands are doubly degenerate.

The subband structure and envelope wave functions for bound electron and hole states can be obtained by variational method using 10 even and 10 odd Gaussian-type orbitals. Having obtained the subband energies and envelope wave functions, the optical absorption coefficient for vertical transitions can be obtained. Apart from a constant factor, the band-to-band absorption coefficient is given by[16]

$$\alpha_B(\hbar\omega)\sim\frac{1}{\hbar\omega}\sum_{n,n'}\sum_{\mathbf{k}_\parallel}|\hat{\boldsymbol{\epsilon}}\cdot\mathbf{P}_{n,n'}(\mathbf{k}_\parallel)|^2[1-f(E_n(\mathbf{k}_\parallel))]\Delta(E_{n'}(\mathbf{k}_\parallel)-E_n(\mathbf{k}_\parallel)-\hbar\omega)\ , \tag{1}$$

where $\hat{\boldsymbol{\epsilon}}$ is the polarization direction of the electric field, $\hbar\omega$ is the energy of the incident photons and $E_n(\mathbf{k}_\parallel)$, and $E_{n'}(\mathbf{k}_\parallel)$ are valence- and conduction-subband energies, respectively. $f(E)$ is the temperature-dependent Fermi distribution function for holes. $\Delta(E)\equiv(\Gamma/\pi)(E^2+\Gamma^2)^{-1}$ describes the effects of line broadening with a half-width of Γ. In the envelope function approximation we have

$$\mathbf{P}_{n,n}(\mathbf{k}_\parallel)=\sum_{mm'}\langle sm'|\mathbf{p}|pm\rangle\int_{-\infty}^{\infty}dz\,\Phi^{e*}_{n'm'}(\mathbf{k}_\parallel,z)\Phi^{h}_{nm}(\mathbf{k}_\parallel,z)\ ,$$

where $\langle sm'|\mathbf{p}|pm\rangle$ is the optical matrix element between the s-like spin-$\frac{1}{2}$ conduction Bloch state $|sm'\rangle$ and the p-like spin-$\frac{3}{2}$ hole Bloch state $|pm\rangle$, which is proportional to the matrix element $\langle s|P_x|x\rangle$ defined in Ref. 17. In our calculation, we average over all polarization directions of the incident light.

The absorption coefficient including the excitonic effect in two-dimensional systems was previously calculated by Shinada and Sugano.[18] They find that (apart from a constant factor)

$$\alpha(E)\sim\frac{P_{cv}^2}{\hbar\omega}\left(\sum_{n=1}^{\infty}(n-1/2)^{-3}\delta(E+(2n-1)^{-2}E_0)+\frac{\theta(E)}{2(1+e^{-2\pi/\sqrt{E}})}\right)\ , \tag{2}$$

where $E=\hbar\omega-E_g$, E_0 is the binding energy of the exciton in the ground state, and P_{cv}^2 is the squared optical-matrix element. In our calculation, we ignore the absorption due to the exciton in bound excited states and replace the delta function term in (2) by a Lorentzian function $f_0\Delta(E+E_0)$. Here f_0 is the oscillator strength which is determined by

$$f_0=\left|\sum_{\mathbf{k}}\Phi_{nn'}(\mathbf{k}_\parallel)P_{nn'}(\mathbf{k}_\parallel)[1-f(E_n(\mathbf{k}_\parallel))]\right|^2\ ,$$

where $\Phi_{nn'}(\mathbf{k}_\parallel)$ is the ground-state exciton envelope function in $\mathbf{k}_\parallel$ space. We further replace the function $\theta(E)$ in (2) by $4\hbar\omega\alpha_B(\hbar\omega)$, where α_B is our band-to-band transition. Note that if we let $P_{nn'}(\mathbf{k}_\parallel)=P_{cv}\cdot\delta_{nn'}$, then our model will give an absorption spectrum identical to that described by (2) with broadening and ignoring excited excitonic bound states.

The exciton envelope function in $\mathbf{k}_\parallel$ space, $\Phi_{nn'}(\mathbf{k}_\parallel)$ for relative motions between the electron and hole was obtained by solving the two-dimensional effective-mass equation

$$\sum_{\mathbf{k}_\parallel}H_{nn'}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\Phi_{nn'}(\mathbf{k}_\parallel)=E_{nn'}\Phi_{nn'}(\mathbf{k}_\parallel)$$

with

$$H_{nn'}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)=[E_{n'}(\mathbf{k}_\parallel)-E_n(\mathbf{k}_\parallel)]\delta_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel}+V_{nn'}(\mathbf{k}_\parallel-\mathbf{k}'_\parallel)/\epsilon(\mathbf{k}_\parallel-\mathbf{k}'_\parallel)\ ,$$

where $\epsilon(\mathbf{q})$ is the two-dimensional wave-vector-dependent dielectric function of Stern[19] which takes into account the screening effect due to free holes. The bare Coulomb potential in real space is

$$V_{nn'}(\rho)=e^2\int\int dz_e\,dz_h|\Phi^{e}_{n'}(0,z_e)|^2|\Phi^{h}_{n}(0,z_h)|^2/[\rho^2+(z_e-z_h)^2]^{1/2}\ ,$$

which is computed numerically. The nonparabolicity of the valence-subband structure $[E_n(\mathbf{k}_\parallel)]$ is fully incorporated in our calculation and is found to have an important effect on the exciton binding energy. The widths of the exciton lines, which depend on their lifetimes, are difficult to calculate. For purposes of generating artificial absorption spectra, we selected a set of half-widths which allowed us to reproduce photoluminescence spectra reported by Gossard.[4] We found that reasonable choices of Γ are given by the simple relation $\Gamma=n_en_h$ meV, where n_e and n_h are principle quantum numbers of the conduction- and valence-band states.

The method just described was applied to the GaAs-$\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ system. The electron well depth is taken to account for 60% of the band-gap difference and the hole well depth accounts for the remainder.[20] For the Luttinger parameters we adopted the values of Lawaetz for GaAs.[17] The line broadening factor Γ for band-to-band transitions was taken to be 0.5 meV.

The calculated valence-subband structure along the (100) and (110) directions for a 100-Å quantum well with an aluminum concentration $x=0.4$ in the absence of doping is shown in Fig. 1. We label the pure states of definite parity at $\mathbf{k}_\parallel=\mathbf{0}$ by HHn and LHn for the nth heavy- and light-hole levels, respectively. As can be seen in Fig. 1, the band structure is nearly independent of direction in $\mathbf{k}_\parallel$ space. The band structure is seen to be rather complicated since coupling between heavy- and light-hole states gives rise to significant level repulsion between the bands. At points in $\mathbf{k}_\parallel$ space where the noninteracting bands cross, an anticrossing phenomenon is clearly observed. Since the strength of the coupling terms increase with increasing $\mathbf{k}_\parallel$, effective masses for some of the valence-band states are observed to be negative. In particular, the increasing strength of the level repulsion interaction between the LH1 and HH2 state for increasing $\mathbf{k}_\parallel$ causes an upward shift in the LH1 level, hence the appearance of the negative LH1 mass.

The variation of the hole effective mass at Γ as a function of the well width W for the first three valence subbands

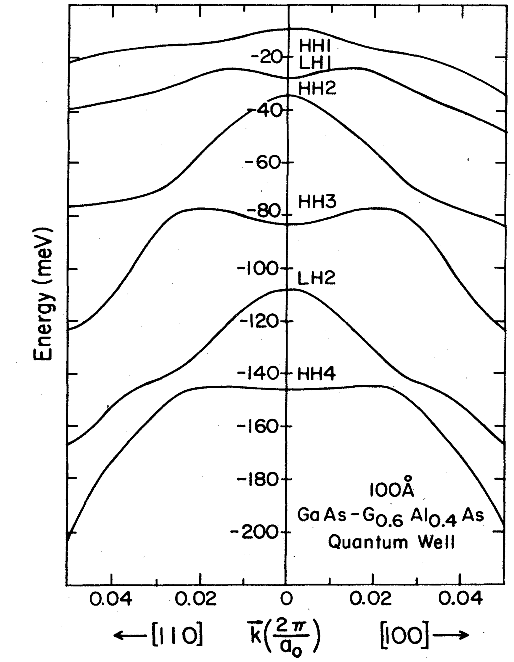

FIG. 1. Hole subband energies of a 100-Å GaAs-Al$_{0.4}$Ga$_{0.6}$As quantum well in the absence of modulation doping.

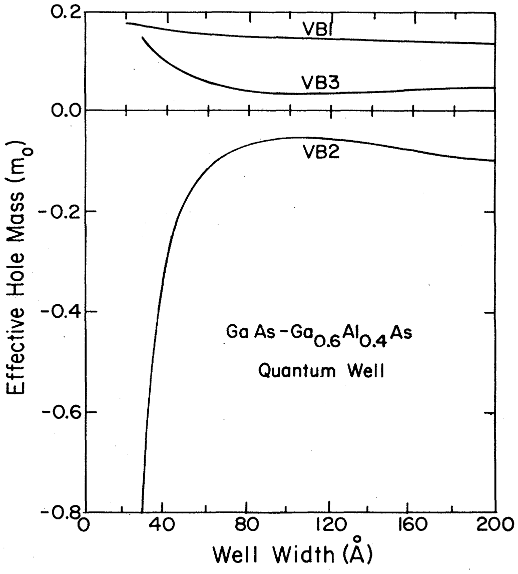

FIG. 2. Zone-center hole effective masses in a 100-Å GaAs-Al$_{0.4}$Ga$_{0.6}$As quantum well for the first three valence subbands as a function of well width in the absence of modulation doping.

(VB 1–3) is illustrated in Fig. 2 for $x=0.4$ in the absence of modulation doping. It is seen that the VB2 effective mass remains negative for all quantum wells with reasonable width. With a doping potential of height $V_0=15$ meV, these effective masses are found to be practically unchanged. Thus we take the density of states hole mass for HH1 in Stern's expression for the dielectric function to be $0.14m_0$.

We now turn to a discussion of the optical absorption. Because of the mixing of heavy- and light-hole states of both even and odd parity at points away from the zone center our theory predicts the existence of vertical transitions between all pairs of valence and conduction subbands. Transitions which are forbidden based on theories which

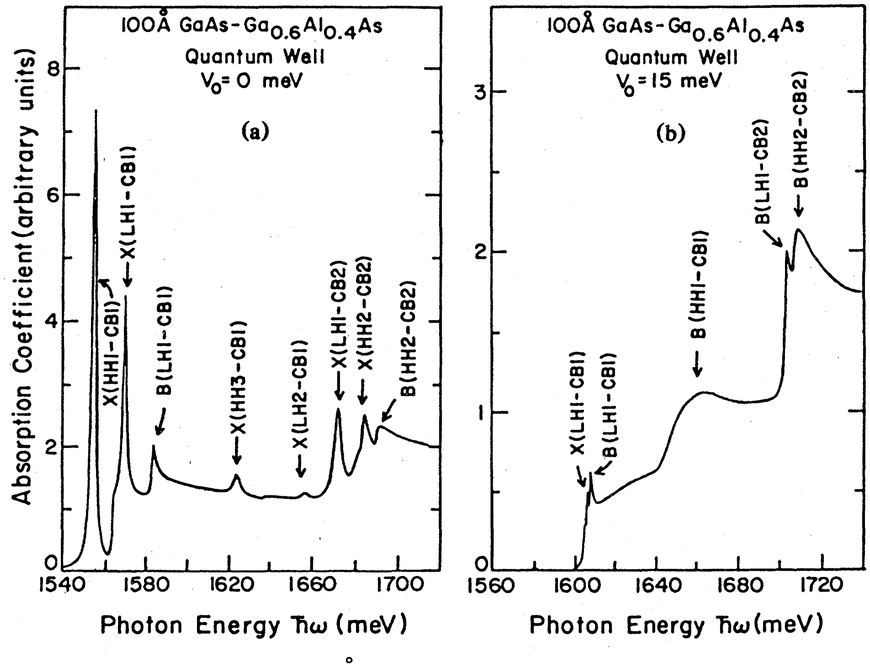

FIG. 3. Computed optical-absorption spectra of a 100-Å thick GaAs-Al$_{0.4}$Ga$_{0.6}$As quantum well for incident radiation polarized in the x-y plane (a) in the absence of modulation doping and (b) with a modulation doping potential with $V_0=15$ meV.

neglect heavy- and light-hole coupling can be explained by the existence of exciton peaks near the thresholds of transitions which violate the $\Delta n = 0$ selection rule. The presence of the negative effective masses results in the creation of sharp peaks in the joint density of states near the threshold of transitions between the negative mass valence bands and all conduction subbands; hence peaked structures in the band-to-band transitions are also expected.

The calculated absorption spectra for $x = 0.4$ and $W = 100$ Å in the absence of doping is shown in Fig. 3(a) for unpolarized light propagating along the z direction. The valence-band VB2 state is LH1 at the zone center and hence overlaps strongly with the CB1 ground-state envelope function. This strong overlap in conjunction with a sharp peak in the joint density of states then gives rise to a small peak in the band-to-band absorption spectrum labeled B(LH1-CB1). In the absorption spectrum, the major features are due to excitons. The strong peaks labeled X(HH1-CB1) and X(LH1-CB1) are the heavy- and light-hole excitons corresponding to $\Delta n = 0$ allowed transitions. Weaker exciton structures corresponding to $\Delta n \neq 0$ transitions [especially X(HH3-CB1)] are also evident. We find that the binding energies for X(LH1-CB1) and X(HH3-CB1) are 13.6 and 12.6 meV, respectively, which are substantially larger than the binding energies of other excitons (6–9 meV) due to the effect of large joint density of states.

If we dope the sample so that the doping potential is 15 meV, we get the absorption spectrum shown in Fig. 3(b). The average hole concentration in the well for this case is 8.18×10^{17} cm^{-3} and the Fermi level E_F lies 6.23 meV below the HH1 band edge; hence the HH1 valence band is partly filled by holes and the remaining valence bands are unfilled. At this level of doping the holes screen the Coulomb interaction so strongly that only the excitons associated with LH1 and HH3 subbands (with negative effective mass at Γ) are bound. The binding energies of these two excitons are 2.1 and 0.4 meV, respectively. The parabolic-doping potential shifts the energies of the valence- and conduction-band levels with the net result that the band-to-band transitions are shifted to higher energies. This partial filling of the HH1 band results in the suppression of all exciton transitions and sharp cutoff of the HH1-CB1 band-to-band transition below 1650 meV. The absorption edge in this case corresponds to transitions between VB2 and CB1. As can be seen in the figure, the B(LH1-CB1) transition due to negative LH1 effective mass becomes more pronounced and may even be observable.

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