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Excitons associated with subband dispersion in GaAs/$Al_xGa_{1-x}As$ superlattices

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Excitonic effects associated with saddle points and subband dispersion were studied in a series of GaAs/$Al_{0.18}Ga_{0.82}As$ superlattices at 5 K using photoluminescence excitation spectroscopy. The well width was fixed at 75 Å and the barrier widths (L_b's) were 105, 60, and 35 Å. Distinct new peaks and structures were detected in addition to the major $n=1$ excitonic peaks (1HH, 1LH). The origin of these peaks in superlattices is attributed to the excitonic resonance occurring near the lower band edge and the upper saddle points of the subband. This excitonic resonance is brought about by the redistribution of the oscillator strengths of the continuum states due to the electron-hole Coulomb interaction. Excellent agreement was found between the data and theoretically generated curves which incorporate saddle-point excitonic effects and valence-band mixing. We also observed a decrease in exciton binding energies, approaching the bulk GaAs limit, with a decrease in L_b.

Excitons in quantum-well structures have been investigated extensively both experimentally and theoretically.[1,2] However, excitons in superlattice where the well-to-well coupling is significant, have not received as much attention.[3-5] In this Rapid Communication, we report on the experimental and theoretical studies of excitonic effects in GaAs/$Al_xGa_{1-x}As$ superlattices. By using photoluminescence excitation (PLE) spectroscopy, we have observed drastic changes in excitation spectra with variations in the barrier width L_b of the superlattice samples.[4] In the thin-barrier samples we examined, pronounced peaks and bumps appeared in addition to the major heavy-hole (HH) and light-hole (LH) exciton peaks. These additional structures arise from Coulomb interaction between electrons and holes associated with saddle points of the superlattice band structure.[3,4] Excellent agreement was found between experimental data and theoretically generated absorption spectra which incorporate the saddle-point excitonic effects.

A series of high-quality GaAs/$Al_xGa_{1-x}As$ superlattice (SL) samples were grown by molecular-beam epitaxy on (100) GaAs substrates. The well width, L_z, and the aluminum concentration in the barrier, x, were fixed at 75 Å and 0.18, respectively. The barrier widths L_b's were 105, 60, and 35 Å. The layer thicknesses were determined by transmission electron microscopy. The total thickness of the superlattices ranged from 0.6–1 μm. The aluminum concentrations were determined by the photoluminescence measurements of $Al_xGa_{1-x}As$ films which were grown under the same molecular-beam epitaxy conditions as the SL samples. Photoluminescence excitation spectroscopy experiments were carried out at 5 K as described in Ref. 6.

In superlattices, the zone-folding effects of the band structure along k_z, the wave vector in the sample growth direction, can lead to the energy subband dispersion and the formation of the M_1-type critical points (saddle points) at the Brillouin-zone boundary.[7,8] Recently, Chu and Chang theoretically investigated Coulomb interaction of electrons and holes associated with saddle-point states in superlattices.[3] They calculated photoabsorption line shape changes of 1HH excitons in GaAs/$Al_{0.25}Ga_{0.75}As$ superlattices with $L_z=80$ Å as a function of L_b ranging from 100 to 15 Å. For the thick-barrier case ($L_b=100$ Å), the band structure is dispersionless in the growth direction and the calculated absorption spectrum is similar to that of a quantum well. For the thin-barrier case, the subband dispersion in the growth direction is large and the resulting absorption spectrum below the saddle point is similar to that of bulk GaAs. For energies near the saddle point the absorption coefficient dips down and smoothly joins a curve appropriate for a two-dimensional system. In the intermediate regime ($L_b=28$–70 Å), prominent structures were found between the main exciton peak and the saddle point indicating a redistribution of the oscillator strengths of the continuum states by the Coulomb interaction. These structures may be interpreted as exciton resonances.

Figure 1 shows the PLE spectra taken from a series of GaAs/$Al_{0.18}Ga_{0.82}As$ SL samples with $L_z=75$ Å in the vicinity of $n=1$ LH exciton peaks. Figures 1(a), 1(b), and 1(c) correspond to the SL's with $L_b=105$, 60, and 35 Å, respectively. A small aluminum mole fraction ($x=0.18$) is chosen here, so that the conduction subband has substantial dispersion even for a decent barrier width. This reduces the fractional error in the barrier thickness of our

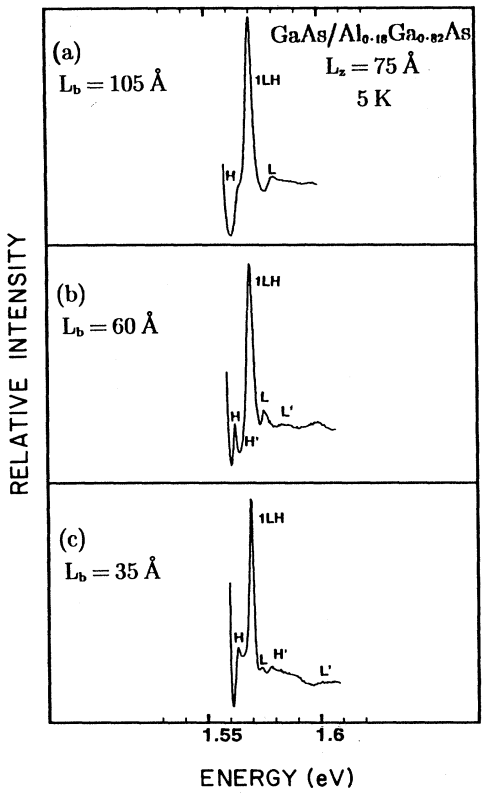

FIG. 1. The PLE spectra taken from a series of GaAs/$Al_{0.18}Ga_{0.82}As$ superlattice samples with a fixed well width. The barrier widths are shown next to each spectrum. The primary light-hole excitonic peak is indicated by 1LH. Secondary peaks H and L are found near the lower band edge, whereas peaks H' and L' appear near the upper band edge. The 1LH peaks for (a) and (b) were shifted downward to align with that of (c).

samples. The detector was set at the peak of the $n=1$ HH exciton of each sample. The spectra of Figs. 1(a) and 1(b) were shifted downward by 8.2 and 7.6 meV, respectively, so that the 1LH peaks align with one another. The spectrum in Fig. 1(a) is similar to those that have been observed in isolated wells,[9,10] whereas spectra 1(b) and 1(c) are significantly different.

For $L_b=105$ Å, a bump appears on the high-energy side of 1LH separated by ≈ 10 meV. No additional structures are noticed near this bump, which is indicated by L in Fig. 1(a). The corresponding bump for 1HH is indicated by H. These structures H and L develop into clear peaks for $L_b=60$ and 35 Å. A trend in spectral line-shape changes, similar to that shown in Figs. 1(a)–1(c), was observed with another series of $GaAs/Al_xGa_{1-x}As$ SL samples. The L_z and x values were fixed at 65 Å and 0.12, respectively, whereas the L_b's were 130, 80, and 60 Å. Peaks H and L were observed in all three samples. The line shape of the spectrum for $L_b=130$ Å is essentially the same as that in Fig. 1(a). The spectra taken from the rest showed more structures than those in Figs. 1(b) and 1(c).

The theoretical calculations performed here are based on the method described in Ref. 3. To get realistic absorption spectra, we have included the effect of valence-band mixing[11–13] on the saddle-point excitons. To make the computation feasible, the axial approximation introduced by Altarelli[12] was used. This allows one to reduce the number of valence subbands by a factor of 2 (each valence subband becomes doubly degenerate). Furthermore, the valence-subband structure becomes spherical in the x-y plane. A specific angular-dependent phase factor was introduced in the exciton envelope function in the same way as described in Ref. 14. Since the number of states are substantially larger in this situation, the brute force matrix diagonalization is impractical. Thus we adopted the recursion method[15] to calculate the density of states for each heavy- or light-hole component (labeled with $v=\frac{3}{2}$ or $\frac{1}{2}$), which in turn gives rise to the absorption spectrum when multiplied by the proper optical matrix element squared. The problem is therefore substantially simplified. We shall consider excitonic states with orbital angular momenta $l=0$ and 1 (i.e., s-like and p-like) only. The higher angular momentum states are much less important in the calculation of the absorption coefficient.[14] Due to symmetry the two sets of excitonic states described by [(s-like, $v=\frac{3}{2}$), (p-like, $v=\frac{1}{2}$)] and [(p-like, $v=\frac{3}{2}$), (s-like, $v=\frac{1}{2}$)] are decoupled. To obtain accurate results, many subband states of different principal quantum numbers are needed in the expansion of the exciton states. In the present calculation, we included both s-like and p-like excitonic states associated with 1HH, 2HH, and 1LH valence subbands only. We refer to this as a three-band model. The adequacy of this model was tested by including more subband states but with smaller number of mesh points in $\mathbf{k}$ space and we found qualitatively the same results. Details of the theoretical calculations will be presented elsewhere.[16]

Figure 2 shows the calculated absorption coefficients for excitonic states associated with the first conduction subband of a GaAs-$Al_{0.18}Ga_{0.82}As$ superlattice ($L_z=75$ Å, $L_b=100$ Å) and a GaAs-$Al_{0.18}Ga_{0.82}As$ superlattice ($L_z=75$ Å, $L_b=60$ Å) obtained in the three-band model (dashed curves). The corresponding experimental data (solid curves) are superposed for comparison.

By comparing with theoretically generated absorption spectra, we attribute peaks H and L to a mixture of $2s$ exciton states and the band-edge resonance arising from a redistribution of the oscillator strengths caused by the Coulomb interaction. In a quantum well with no band dispersion such as the sample in Fig. 1(a), H and L are most likely $2s$ states merged to a steplike band continuum. In superlattices with subband dispersion, a significant portion of H and L can come from the lower band-edge resonance as discussed in Ref. 3. The relative intensity contributions between the $2s$ states and the band-edge resonance depend on the sample parameters. The distinction between the two separate contributions is difficult because of their line-shape changes with L_b.

In contrast to the nearly symmetric line shape of the fundamental exciton peaks, H and L exhibit asymmetric line shapes. The degree of asymmetry increases with an increase in the subband dispersion. It is most clearly evidenced in Fig. 1(b), where peak L has a longer tail toward the high-energy side than peak H. It appears that the

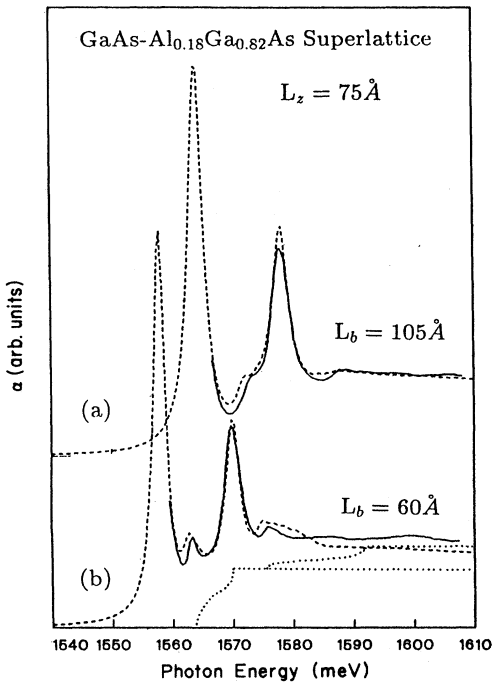

FIG. 2. Theoretical absorption spectra (dashed curves) and experimental PLE data (solid curves) for GaAs/$Al_{0.18}Ga_{0.82}As$ superlattices with well width L_z =75 Å and barrier widths L_b =100 Å and 60 Å. The continuum states of 1HH and 1LH for L_b =60 Å are shown by the dotted lines.

sharp rise of these asymmetric peaks originates from 2s states, and the slowly decreasing tail from the band-edge resonance. Such asymmetric line shapes can be generated theoretically as shown in Fig. 2, and results are in good agreement with the data.

In addition to aforementioned peaks H and L, we detect other peaks and structures in the spectra, which cannot be assigned to any optical transitions between the states with $n \neq 1$ such as the forbidden transition between $n=2$ heavy-hole and $n=1$ conduction states. According to the theoretical calculations, exciton resonance can also occur near the saddle points. The degree of resonance enhancement depends on L_b. These resonance peaks are less pronounced than those secondary peaks near M_0 points. Therefore, they should be harder to detect than H and L. In the spectra we observed, peaks marked as H' and L' are attributed to arising from such saddle-point resonance enhancement. Similar structures were also observed in GaAs/$Al_{0.12}Ga_{0.88}As$ samples with L_b =80 and 60 Å. Our attribution is based on the considerations of transition energies. Further verifications, however, are needed for a conclusive identification.

Peaks H and L have asymmetric line shapes with 2s states located on their rising edge. Therefore, the separation between the primary exciton peaks and the onset of the secondary peaks can be used in estimating the 1s-2s energy separations. By comparing the theoretical values of the ratios of 1s-2s energy separations to the exciton binding energies obtained in a variational calculation,[17] one can obtain a good estimate of the exciton binding energies. In cases of isolated wells with thick barriers, the binding energy E_b is known to be about 10 meV for 1HH.[18] As L_b gets smaller, the SL samples will become more like bulk GaAs. The E_b's of these superlattices should also approach that of GaAs, i.e., 4.2 meV.[19] Hence, the binding energies of the superlattice samples we examined should lie between these two extrema. What we measured are indeed within this range. The details of our theoretical and experimental studies on the subject of exciton binding energies will be reported elsewhere.[20]

In summary, we have observed pronounced peaks on the high-energy side of the fundamental 1HH and 1LH peaks in the PLE spectra taken from a series of GaAs/$Al_{0.18}Ga_{0.82}As$ superlattice samples with a fixed well width of 75 Å. The barrier widths L_b's were 105, 60, and 35 Å. These new peaks are formed as a consequence of the Coulomb interaction associated with the subband dispersion.[3] The lower-energy peaks (H and L) contain 2s exciton contributions and the low band-edge excitonic resonance, whereas the higher ones (H' and L') are excitonic resonance peaks formed near the saddle points. These peaks which have asymmetric line shapes were most prominent in intermediate region of L_b (60 Å). Excellent agreement was found between the observed PLE spectra and theoretically generated absorption curves which incorporate saddle-point excitonic effects and valence-band mixing. We also found that the exciton binding energies decreased toward that of bulk GaAs with a decrease with L_b, consistent with calculations.

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