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Resonance broadening of the light-hole exciton in $GaAs/Al_xGa_{1-x}As$ quantum wells

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As a result of the mixing of the heavy- and light-hole subband states in undoped quantum wells, electron–heavy-hole pairs are coupled with electron–light-hole pairs. Consequently, when the light-hole exciton peak lies in the electron–heavy-hole continuum, it is broadened, an effect analogous to a Fano resonance. This phenomenon is examined both theoretically and experimentally, and good quantitative agreement between the two is found.

I. INTRODUCTION

There has been a great deal of interest recently in the effects of valence-band mixing (VBM) on the optical properties of $GaAs/Al_xGa_{1-x}As$ quantum well structures. Theoretical[1,2] and experimental[3-5] investigations have shown that one effect of VBM is to significantly modify exciton binding energies and oscillator strengths and permit "parity-forbidden" exciton transitions to occur. In addition, the effect of VBM on quasi-two-dimensional excitons in a magnetic field has recently been shown to be substantial.[6] In this paper, we report a different manifestation of VBM in quantum wells; namely, the broadening of an exciton peak arising from its interaction with a continuum.

VBM in $GaAs/Al_xGa_{1-x}As$ quantum well structures arises because of the fourfold degeneracy of the bulk heavy- and light-hole valence bands at the valence-band edge. The four Bloch states at this point are basis functions for angular momentum, $J=\frac{3}{2}$, with heavy-hole states (h) having $m_j=\pm\frac{3}{2}$ and light-hole states (l) having $m_J=\pm\frac{1}{2}$. Since these bands have different band-edge masses, confinement in a quantum well lifts the degeneracy creating one ladder of heavy-hole subbands and another of light-hole subbands. If the in-plane wave vector is zero, i.e., $k_{\parallel}=0$, these subbands are uncoupled so that the subband states are of strictly h or l character. If $k_{\parallel}$ is finite, however, the subbands are strongly coupled and of mixed h-l character; that is, they are composed of mixtures of the four Bloch states with different m_J. This broken degeneracy produces two sets of exciton transitions, one set derived from the electron and heavy-hole subbands (e-h) and the other derived from the electron and light-hole subbands (e-l). Since the hole subband states have mixed h-l character, the Coulomb interaction couples the (e-h) and the (e-l) states, an effect which we will refer to as the intersubband interaction. Consequently, when an exciton derived from one pair of subbands lies in the continuum of states associated with the other pair of subbands, it exhibits a Fano-type resonance[7] in which the exciton peak is broadened and shifted in energy.

Near the energy band edge of quantum wells, the absorption spectrum is dominated by two ground-state exciton peaks associated with the heavy-hole ($11h$) and light-hole ($11l$) excitons. In thin wells the energy separation between hole subbands is larger than exciton binding energies so that the $11l$ exciton lies in the $11h$ continuum. As the well thickness increases, this energy separation decreases until the $11l$ exciton drops below the continuum. In this paper, the linewidth of the ground-state $11l$ exciton peak, ΔE_L^{1s}, is studied as a function of quantum well width throughout this region, so that the effect of the $11h$ continuum on the $11l$ exciton can be examined. In Sec. II, the absorption spectrum is calculated in the absence and in the presence of VBM and ΔE_L^{1s} is extracted. In Sec. III, ΔE_L^{1s} is determined experimentally for a series of $GaAs/Al_xGa_{1-x}As$ quantum well samples with wells of various thicknesses, using photoluminescence excitation spectroscopy. Finally, a comparison is made between theoretically calculated and experimentally measured values of ΔE_L^{1s}.

II. THEORY

In our treatment, an exciton in an undoped quantum well is described by an electron and hole confined by their respective square-well barriers, and interacting through a Coulomb attraction. The electron subband states, which are assumed parabolic, and the hole subband states are calculated in the effective-mass approximation. VBM of the hole subbands is incorporated by employing the 4×4 Luttinger-Kohn Hamiltonian,[8] which is solved using the subband $\mathbf{k}\cdot\mathbf{p}$ method.[2] The exciton wave function is expanded in products of electron and hole subband states, and a set of coupled effective-mass equations is derived for the excitonic spectrum in the quantum well. In particular, $11h$ and $11l$ excitations are described by the pair of equations[2]

$$[\varepsilon_{1e}(k)-\varepsilon_m(k)-E]\Phi_m^{1e}(k)+\sum_{m'}\int V_{mm'}^{1e1e}(k,k')\Phi_{m'}^{1e}(k')dk'=0,\quad m,m'=1h,1l \tag{1}$$

For simplicity, higher-lying electron and hole levels in the summation have been omitted since only the interaction of the $11l$ exciton with the $11h$ continuum is considered here. $\varepsilon_{1e}(k)$ and $\varepsilon_m(k)$ are the electron and hole subband energies. The subscript, $1e$, designates the first electron subband, while m ranks the hole subbands which are either strictly h or strictly l in character at $k=0$; k is the magnitude of the relative wave vector of the electron-hole state in the plane of the well. The kernel, $V_{mm'}^{1e1e}(k,k')$, is the matrix element of the screened Coulomb potential between products of electron-hole states of different wave vectors, k and k', integrated over the polar angle of k'. The angular dependence in $\Phi_m^{1e}(k)$ has been found to be small for the ground s-like exciton states[9] and so it has been neglected.

VBM causes $\varepsilon_m(k)$ to be nonparabolic, and mixes the e-h and e-l states through the off-diagonal terms, $m\neq m'$, which represent the intersubband interaction. It should be emphasized that, in the absence of VBM, these terms vanish and the e-h and e-l states remain distinct.

The coupled equations, given above, are solved using the modified quadrature method.[10] Unlike a variational solution, this technique yields not only the bound exciton states but also a discrete set of continuum states. The oscillator strengths for each state are then calculated, and the absorption spectrum is determined in the dipole approximation following Ref. 2. This discrete spectrum is smoothed by broadening each spectral line (exciton and continuum) with Lorentzians of half width, Γ. The value of Γ is chosen so that the full width at half maximum (FWHM) of the $11h$ exciton roughly matches that determined by experiment.

The calculated absorption spectrum for a 10 nm well with $x=0.3$ and $\Gamma=0.5$ meV [a full width at half maximum (FWHM) of 1 meV] is shown in Fig. 1. In the figure, E_L^{1s} refers to the energy of the ground-state ($1s$) $11l$ exciton peak, while E_H^{1s} and E_H^{2s} refer to the energies of the ground ($1s$) and first excited ($2s$) $11h$ exciton peaks, respectively. E_H^{2s} approximately defines the beginning of the continuum of states associated with the heavy-hole exciton. The dashed spectrum was calculated neglecting the intersubband interaction. In this case, the FWHM of the light-hole exciton peak, ΔE_L^{1s} (measured from the $11h$ continuum), and that of the heavy-hole exciton peak, ΔE_H^{1s} (measured from the zero of absorption) are the same. They remain the same independent of quantum well width if the intersubband interaction is absent. The solid line spectrum was calculated including the intersubband interaction. The signatures of the Fano effect on E_L^{1s} are now evident; i.e., the peak is shifted in energy broadened, and slightly asymmetric. It will be convenient to define the quantity, $R=\Delta E_L^{1s}/\Delta E_H^{1s}$. In this case, $R\simeq1.7$.

Similar calculations were performed for quantum well thicknesses ranging from 7 to 20 nm. As the well thickness increased and E_L^{1s} emerged from the heavy-hole continuum, it passed throught E_H^{2s} and finally fell below it. For this latter case, the FWHM of E_L^{1s} (now measured from the zero of absorption) dropped dramatically and $R\simeq1$. In the region of well thicknesses where E_L^{1s} lies at the edge of the $11h$ continuum, the reference point, from which the peak height is to be measured, is no longer clear because the high-energy side of the peak lies on the $11h$ continuum while the low-energy side lies below it. Nevertheless, if the zero of absorption is chosen as the reference point, the resulting FWHM can be determined. Although the exact meaning of this quantity is unclear, it can be compared with the experimental value of R derived in a similar fashion.

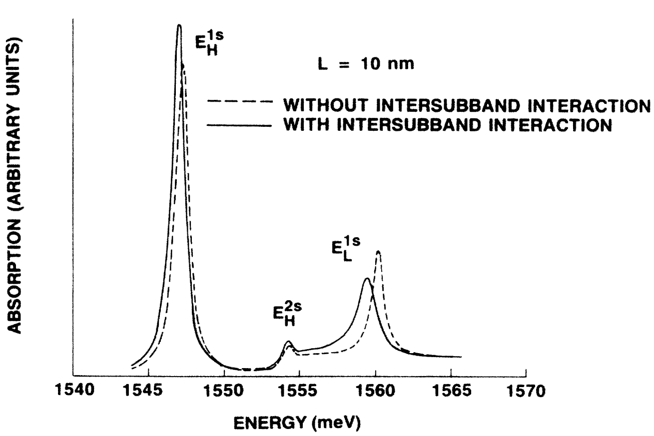

FIG. 1. Calculated absorption spectrum of a 10-nm $GaAs/Al_{0.3}Ga_{0.7}As$ quantum well with (solid line) and without (dashed line) the intersubband interaction.

III. EXPERIMENTS, RESULTS, AND DISCUSSION

Nominally undoped $GaAs/Al_{0.3}Ga_{0.7}As$ quantum well (QW) samples, grown using molecular-beam epitaxy, exhibited strong low-temperature photoluminescence (PL) and narrow exciton PL peaks, which are indications of superior crystal quality. For the photoluminescence excitation (PLE) spectroscopy measurements, the samples were mounted, strain free, in a flowthrough liquid-helium cryostat, and excited with a tunable-dye-laser source pumped with a Kr-ion laser. Details of PLE spectroscopy, which is analogous to absorption spectroscopy, especially at low temperatures, can be found elsewhere.[11]

Low-temperature PLE spectra of several single QW's of differing well thicknesses are presented in Fig. 2 (the peaks are labeled as in Fig. 1). At the temperatures at which these spectra were obtained, broadening of exciton peaks, in samples such as these, is thought to be primarily extrinsic, i.e., sample related. In particular, it is considered due to local differences in the quantum well width, L_Z, as a result of growth conditions. Since the energy of a quantum well exciton is a sensitive function of L_Z, variations in L_Z produced by monolayer fluctuations at the interface between different semiconductor layers leads to spatial variations in exciton energy across the sample. This is manifested by broadened PL, and PLE, peaks since the experiment probes an area of the sample ($\simeq 100$ μm diameter) much larger than an exciton diameter. This effect is especially important for narrow QW's (say, $L_Z < 10$ nm) and, since the energy of light-hole excitons is a stronger function of L_Z than the energy of heavy-hole excitons, light-hole excitons will be broadened more by this mechanism than heavy-hole excitons. In order to minimize the effect of this mechanism on our experimental results the ratio of the full width at half maximum (FWHM) of the light-hole exciton to that of the heavy-hole exciton (i.e., $R = \Delta E_L^{1s}/\Delta E_H^{1s}$) is determined. Thus, if only this mechanism were active in the broadening, R would be slightly larger than 1 and relatively constant over the QW width range studied in this work.

In Fig. 3, the measured values of R, for a series of QW's are presented together with theoretical predictions. These values are given as a function of the distance between the light-hole exciton ground state, E_L^{1s}, and the edge of the heavy-hole continuum, which is taken as E_H^{2s}, i.e., $\Delta E = E_l^{1s} - E_H^{2s}$. A negative ΔE indicates that E_L^{1s} does not overlap the continuum (i.e., L_Z is large) while positive ΔEs indicate that E_L^{1s} and the continuum are overlapping (i.e., small L_Z).

The excess broadening of the light-hole exciton PL peak over the heavy-hole PL peak due to variations in L_Z, calculated for a one monolayer fluctuation, is indicated by the solid line in Fig. 3. It is clear that the interface fluctuation mechanism cannot account for the observed variation in R as a function of QW width. However, the overall agreement between the calculated values of R based on VBM and the experimental values is very good. For $\Delta E \simeq 0$, R is very large (~ 3) but reduces to a smaller value (~ 2) for $\Delta E > 0$. This is partially, but no totally due to the method by which the FWHM of E_L^{1s} was deter-

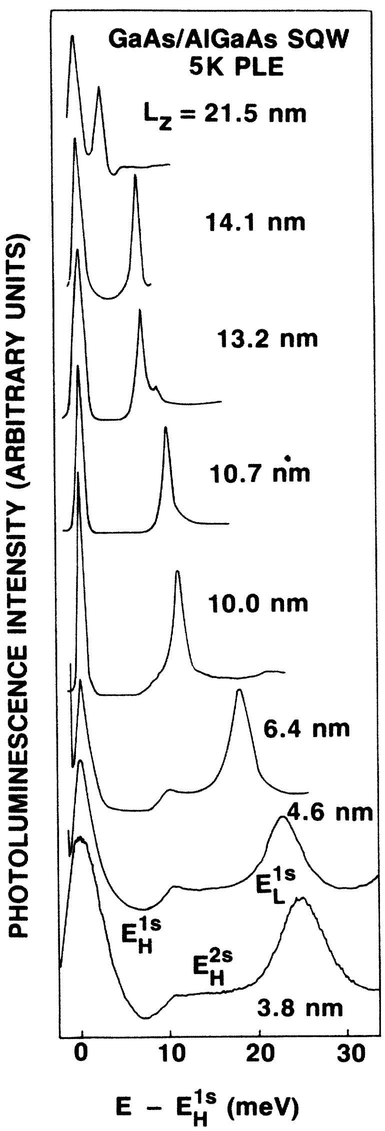

FIG. 2. Experimental low-temperature photoluminescence excitation spectra of several single GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells with different well thicknesses. The horizontal energy scale for each spectrum was shifted so as to line up the E_H^{1s} peaks for easier comparison.

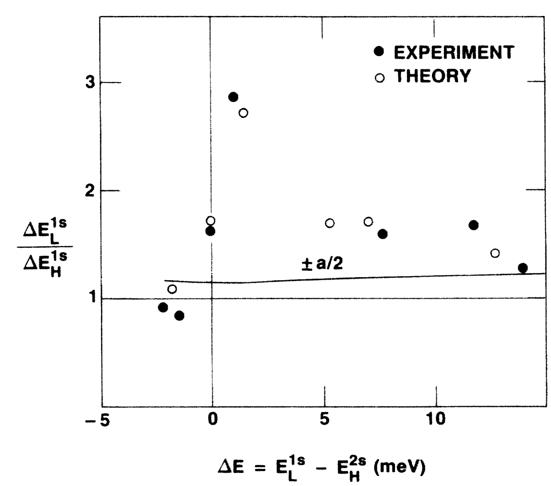

FIG. 3. The ratio R of the FWHM's of the $11l$ to the $11h$ excitons as a function of the energy difference, $\Delta E = E_L^{1s} - E_H^{2s}$. The solid line is the calculated excess broadening of the $11l$ exciton with respect to the $11h$ exciton due to quantum-well width fluctuations.

mined when E_L^{1s} was coincident with E_H^{2s} (see Sec. II). Some additional broadening is expected at this point due to the interaction between E_L^{1s} and the excited states of the heavy-hole exciton which occur near the continuum edge. For $\Delta E > 0$, E_L^{1s} is away from these states and only the continuum states contribute to the interaction. For very large ΔE (very thin wells), R decreases further since the separation between the ground-state heavy- and light-hole subbands increases and the VBM correspondingly decreases. For $\Delta E < 0$, the theory predicts $R \simeq 1$, while experimentally a smaller R is observed. It is not completely clear whether this is a real effect or some artifact of the experiment. However, it is noteworthy that the experimental R is consistently smaller than that predicted by the model of interface roughness (solid line).

It should be noted that, in principle, it is unimportant how the overlap between the light-hole (LH) ground exciton state and the continuum of heavy-hole (HH) states is achieved. The broadening mechanism is the same. In particular, it is possible to bring this interaction about using uniaxial stress to split the ground-state exciton in bulk GaAs and to move the LH exciton into coincidence with the HH continuum. When this occurs, an increase in the linewidth of the LH exciton by a factor of about 3 is observed.[12] This is in excellent agreement with the results presented in Fig. 3.

IV. SUMMARY

It has been argued that the experimental observation of excess broadening of the ground-state light-hole exciton in a semiconductor quantum well when its energy overlaps that of the heavy-hole exciton continuum of states is a consequence of the mixing of the light- and heavy-hole subband states. Good agreement between the experimentally observed broadening as a function of quantum well width (and thus light-hole exciton and heavy-hole continuum overlap) and that calculated using a model incorporating valence subband mixing is strong evidence in favor of this interpretation.

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