Coupled-Exciton States in 2D-Semiconductor Systems*

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We review the coupling among tlie different exciton states in a semiconductor quantum well. Three intrinsic mechanisms are considered: tlie coupling among the heavy- and light-hole excitons due to the off-diagonal terms of tlie Luttinger Hamiltonian, the coupling induced by the strain present in lattice mismatched heterostructures and the coupling among the excitons originated from the same host-hole subband due to the three-dimensional character of the Coulomb interaction.

I. Introduction

The optical transitions in semiconductor quantum wells are dominated by tlie correlation between the electron and the hole, that is, the excitonic effects. The carricr confinement along the growth direction enhances the exciton binding energy and its oscillator strength. This gives a strong stability for the exciton states. They are not dissociated at room temperature or by the applicatioii of longitudinal electric fields (up to 100 kV/cm). These cliaracteristics have offered tlie possibility of interesting device applications[1]. The first theoretical approaches considered a decoupled exciton model, with parabolic valence subbands[2,3]. With the improvement of sample quality, fine structures in the exciton states became possible to be observed. Those are consequence of the coupling among the different. excitons and originate from tlie crystal and the heterostructure potentials. They raise an entire new aspect to the hydrogen-like states clisracteristics of tlie Wannier exciton.

Here, me review some of tlie couplings among the different excitons in semiconductor quantum wells (QWs). Iii Section II we cliscuss tlie simple uncoupled model. The coupling among the excitons due to intrinsic effects is included in Section III. Three mechanisms are considered: the off-diagonal terms of the Luttinger Hamiltonian which couples heavy- and light-hole excitons, the biaxial strain in lattice mismatched QWs and tlie three-dirnensional character of the Coulomb interaction. Finally, in Section IV we draw our final rernarks.

II. Uncoupled Exciton States

The yuantum-well corifinement breaks the valence band degeneracy at the center of the Brillouin zone. Choosing the direction of diagonalization of the total angular momentum along the growth direction, it is possible to separate the valence subbands into heavy- and light-hole subbands for the z-dependent motion[4]. When an electron is excited into the conduction subbands, the correlation between the electron and the hole left in the valence band gives origin to the exciton states. Taking a parabolic conduction band, the total Hamiltonian for the exciton state is written as[5]

$$\underline{H}_{\text{exc}} = \left[-\frac{\hbar^2}{2m_e^*} + \frac{\partial^2}{\partial e^2} + V_e Y(z_e^2 - L_z^2/4)\right]\underline{1} + \underline{H}_h(z_h,\rho) - \frac{e^2}{\epsilon\sqrt{\rho^2+(z_e-z_h)^2}}\underline{1}\,, \tag{1}$$

*Invited talk.

where

$$\underline{H}_h(z_h,\rho)=\begin{bmatrix} A_+ & C & B & 0 \\ C^* & A_- & 0 & -B \\ B^* & 0 & A_- & C \\ 0 & -B^* & C^* & A_+ \end{bmatrix}, \quad (2)$$

and

$$\begin{aligned} A_\pm &= \frac{\hbar^2}{2m_0}(-\gamma_1 \pm 2\gamma_2)\frac{\partial^2}{\partial z_h^2} + V_h Y(z_h^2 - L_z^2/4) \\ &- \frac{\hbar^2}{2m_0}\left(\frac{m_0}{m_e^*}+\gamma_1 \pm \gamma_2\right)\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right) \quad (3.a) \\ B &= -\frac{\sqrt{3}\hbar^2\gamma_3}{m_0}\frac{\partial}{\partial z_h}\left(\frac{\partial}{\partial x} - i\frac{\partial}{\partial y}\right) \quad (3.b) \\ C &= -\frac{\sqrt{3}\hbar^2}{2m_0}\left[\gamma_2\left(\frac{\partial^2}{\partial_x^2}-\frac{\partial^2}{\partial_x^2}\right) - 2i\gamma_3\frac{\partial^2}{\partial x\partial y}\right] \quad (3.c) \end{aligned}$$

where $\rho = (x,y)$ is tlic in-plane relative coordinate, γ_1's arc tlie Luttinger parameters and c is the static dielectric constaiit. Tlie simplest approximation consists in neglecting tlie contribution of the higher bands. Namely, we consider only tlic diagonal terms of the Luttinger Hamiltonian, neglecting the off-diagonal ones. A further approximation is to consider separable variables for the z-motion and the in-plane motion. This is a good approximation for tlic Coulomb states for QWs in the range $L < 200$ Å[6]. Within these approximations, the exciton states may be classified iii two different series, each associated to a different hole-host band. Each one of these series is composed of different familics of exciton states, associated to the different quantum well conheci states for tlic electrons and holes[7]. These familics of states are theii splitted into different in-plane symmetries, similar to tlie two-dimensional hydrogen problem[8].

The exciton wave-function is constructed following a separable variables approach. The z-part of the wavefunction is assumed to be dominated by the quantum well potential and is not affected by the Coulomb interaction in first approximation. The in-plane motion is determined variationally. The general form of tlie variational wave-functions are:

$$\Psi_{\rm exc}(z_e,z_h,\rho) = \chi_n(z_e)\varphi_m(z_h)f_{ih}(\theta,\rho)\ , \quad (4)$$

with

$$\begin{aligned} f_{1s}(\theta,\rho) &= \sqrt{2/\pi\lambda_{1s}^2}\exp(-\rho/\lambda_{1s}) \quad (5.a) \\ f_{2p\pm}(\theta,\rho) &= 2/\sqrt{3/\pi\lambda_{2p\pm}^4}\rho\exp(-\rho/\lambda_{2p\pm} \pm i\theta)\ , \quad (5.b) \\ f_{3d\pm}(\theta,\rho) &= 2/\sqrt{15/\pi\lambda_{3d\pm}^6}\rho^2\exp(-\rho/\lambda_{3d\pm} \pm 2i\theta)\ , \quad (5.c) \end{aligned}$$

where $\chi_n(\varphi_m)$ are the n^{th} (m^{th}) electron (hole) QW wave-function ancl tlie λ_i's are the variational parameters. Figure 1 shows (a.)the $1s-E_nHH_m(LH_m)$ exciton binding energy ancl (b) the oscillator strength as a function of tlie QW width, for GaAs-$Ga_{0.7}Al_{0.3}As$ QW's in the simplest parabolic approximation[7]. We observe a general trend for the binding energy of the ground electron and hole levels. The binding energy increases from its bulk value as tlie well width decreases, reflecting the increasing quasi-bidimensionality of the system. Tliis trend goes until at least one of the two particles loses its quasi-bidimensionality, when its wave-function penetrates largely into tlie barrier and the QW level approaches tlie height of the well. The exciton hinding energy then decreases as we further decrease the QW width. The same behavior happens for the excited QW electron aiid hole levels. In this case, the maximum in the exciton binding energy happens at larger values of QW widths as tlie particle level indices (n., m) increasc. The oscillator strength follows a similar behavior. Noticc that we only consider the exciton states with n+m = even number since, for a symmetric QW, they are the only ones to have a.non-zero oscillator strength. We should also observe that, for the equivalent Iiole level, tlie binding energy is larger for the light-hole excitons than tlie heavy-hole ones. Tliis is due to the mass-reversal effect for the liole subbands, a consequence of the diagonal approximation. The heavy-hole shows a lighter cffective mass in comparison to the light-hole effective masses. Excitons with other symmetries for the in-plane part of tlie wave-function (p, d, f, etc.) do not show any oscillator strength in this approximation[9]. However, when the coupling among the excitons takes place, these excited exciton states play an important role.

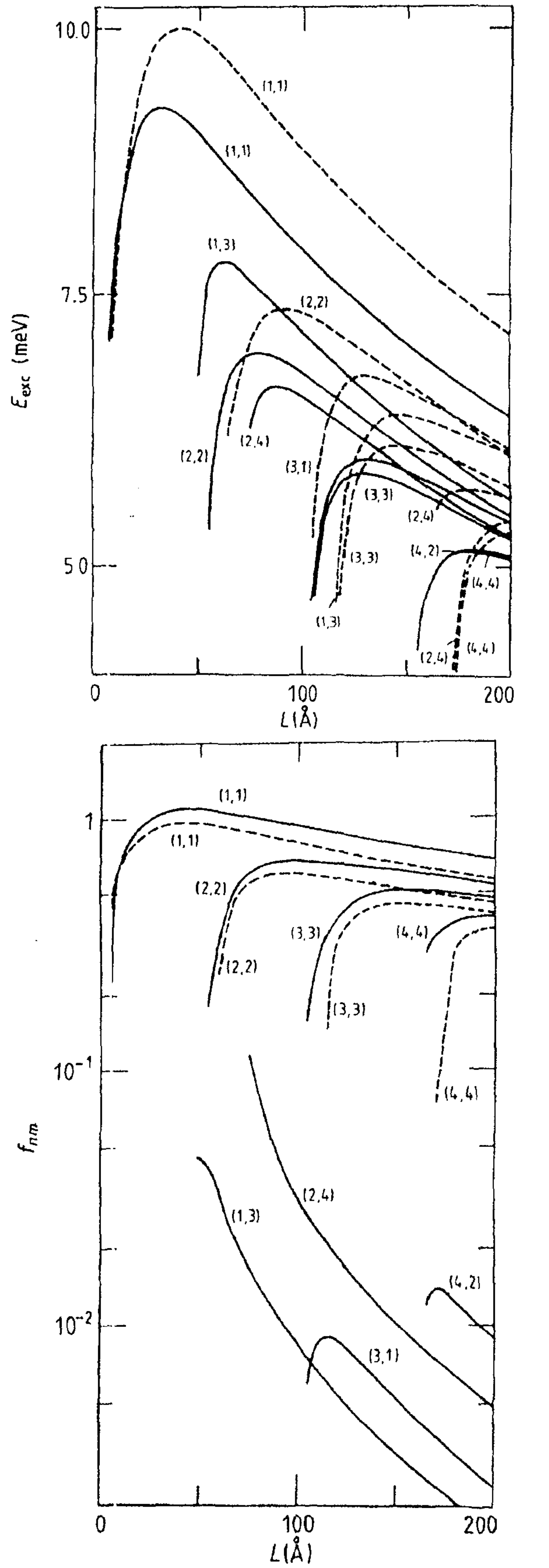

Figure 1: $1s - E_nHH_m$ (solid liiies) and E_nLH_m (dashed lines) (a) exciton binding energies and (b) oscillator strengths in GaAs-$Ga_{0.7}A_{0.3}As$ quantum wells as a function of the GaAs layer thickness. (after Ref. 7).

III. Coupled exciton states

The simple picture discussed so far is strongly modified once we consider the coupling among the different excitons. For the full exciton Hamiltonian, all the exciton states are coupled. This originates from three intrinsic mechanisms characteristic of the system: The off-diagonal terms of the Luttinger Hamiltonian which couple heavy-hole excitons to light- hole excitons. These terms are of second-order in k. As a consequence, they couple exciton states with different in-plane symmetries[10–15]. A second rnechanisrn is the coupling among the liglit- and split-off hole bands, which is enhanced by the QW[16]. A third mechanism is the three-dimensional character of the Coulomb interaction which couples exciton states belonging to different QW levels but tlie same hole-host band[17–19].

Further exciton coupling may be obtained when we consider the effect of external fields. If the field follows the QW cylindrical symmetry, only the intensity of the coiipling will change. On tlie contrary, for fields which break this symmetry (e.g., an in-plane uniaxial stress), additional coupling among the exciton states may be observed[16].

The different couplings among the exciton states have been extensively studied by a number of authors. We will not review here all these effects but will concentrate in those couplings which are intrinsic to the system to be considered.

i) Heavy- and light-hole coupling

The off-diagonal terms of the Luttinger Hamiltonian coiiple tlie heavy- and light-hole states giving origin to highly-non-parabolic valence subband dispersions[20,21]. This coupling is responsible for two effects in the exciton states. First, we observe an increase in the excitoii binding energy[10–14]. The strong anticrossing pattern has the effect of increasing the in-plane effective mass for the ground-hole subband. The effect is even stronger for the light-hole when the ir-plane mass can assume an electron-like behavior.

This results in an increase of the exciton binding energy of tlie order of 10% - 20% for the excitons associated to tlie ground electron and hole subbands.

A more spectacular effect is the coupling of the heavy- and light-liole excitons having different in-plane symmetries. The s-like heavy (light)-hole excitons couple with the p-like and d-like light (heavy)-hole excitons. Bauer and Ando[13] and Ekemberg and Altarelli[14], following different approaches, showed the importance of including the continuum exciton states whenever these coupling is considered. Here, we follow an approach similar to tlie one developed by Bauer and Ando[13]. We project the full cxciton Hamiltonian into a non- ortliogonal basis. This basis is built based on wave-functions of tlie type described by Eqs. 2-3. The total wave-function is a. spinor with components formed by linear combinations of wave-functions having different values for λ. Tlie non-orthogonality of the basis assures tlie presence of a.set of states which mimics tlie continuum states. The eigenvalues are obtained by diagonalizing the generalized eigenvalue problem[5].

Figure 2 shows (a) tlie exciton energy relative to tlie bottom of tlie ground-electron and light-hole band-to-band transition and (b) the respective oscillator strength as a function of tlie QW width[5]. Here, only the coupling among the $1s - E_1LH_1$ and the d-like E_1HH_1 exciton states is shown. Tlie exciton binding energy in the diagonal approximation is also plotted. Far from tlie anticrossing region we observe a clear increase in the cxciton binding energy. As the $1s-E_1LH_1$ and tlie $3d-E_1HH_1$ excitons approach each other, the two states interact and an anticrossing is ohserved. Notice that the $1s - E_1LH_1$ couples also with tlie excited $d-E_1HH_1$ excitons states. Here, these states are represented by some discrete states. They actually represent the discrete and the continuum d-like E_1LH_1 exciton states. As a consequence of the anticrossing, there is an exchange in their oscillator strengths. In this region it is possible to observe optically both exciton states. These anti-crossing effects may be enhanced by the presence of external fields, like an electric field or a magnetic field applied along the growth direction. It has been possible to optically observe several excited exciton states which, in the absence of the coupling, are forbidden transitions[22,23].

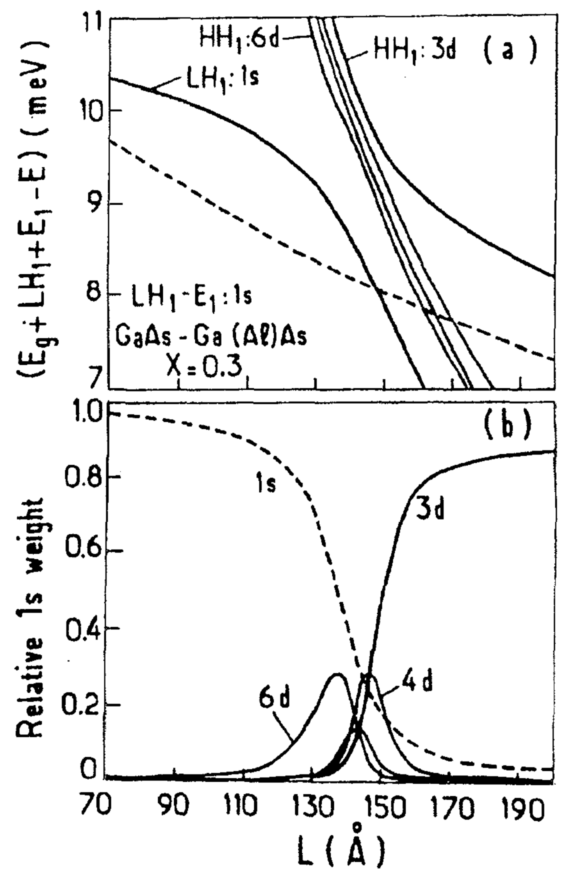

Figure 2: $1s - E_1LH_1$ (a) exciton binding energy and (b) relative oscillator strength of the transition as a function of the GaAs layer thickness in GaAs-$Ga_{0.7}Al_{0.3}As$ quantum wells using a nonorthogonal basis. Dashed line: diagonal approximation. Solid lines: full Hamiltonian. The anti-crossing with tlie $nd - E_1HH_1$ states is also shown. (after Rcf. 5).

ii) Lattice mismatched QW's

An interesting situation appears when the two host materials present a significant difference in their lattice parameter[24]. Here, we consider the case of a $In_xGa_{1-x}As$ QW with GaAs acting as barriers. The structure is assumed to be grown on a GaAs substrate. The lattice parameter that dominates the structure is the one of the GaAs. The InAs and the GaAs have a difference of 7% in their lattice parameters. The $In_xGa_{1-x}As$ layer is, therefore, undcr in-plane biaxial compression. Tliis does not altere tlie cylindrical symmetry of the structure. However, quantitative effects modify significantly tlie electronic properties. First, the heavy- and liglit-hole host bands in the $In_xGa_{1-x}As$ are splitted. This splitting is so stroiig that tlie light-hole may present a type-II confinement. More precisely, the electrons aiid the heavy-hole are confined in tlie $In_xGa_{1-x}As$ layer. With tlie strain effects, tlie liglit-hole band profile is practically flat and, depending of the In concentration, they may be slightly localized in tlie GaAs layer. A second effect is the strong coupling among the light-hole and the split-off-hole states. This coupling is already present in the QW in the absence of tlie strain but it reaclies stroiig values as a consequence of tlie biaxia coinpression.

These strain-induced effects alter significantly the exciton states. Due to the strong separation in energy, an in-plane pârabolic dispersion is quite accurate to describe the heavy-hole exciton states associated to tlie ground-hole subband. The situation is far more complicated for the light-hole exciton. The quantum well potential, the Coulomb interaction aiid tlie light-holes and split-off-lioles coupling rnay all be of the same order and must be considered in the same foot in tlie calculation. To describe tliis situation we allow tlie z-part of tlie light-hole exciton wave-function to depend on the Coulomb interaction. For that, tlie light-hole wave-functions are described by a set of gaussian wave-functions. The z-part of tlie split-off liole wave-function is simply described by the QW wave-functions in a similar way as it was discussed in the previous Section. The diagonalizaticn of the Hamiltonian projected onto these extended basis gives the exciton states[16].

Figure 3 shows the ration of the $1s - E_1LH_1$ to the $1s - E_1HH_1$ exciton oscillator strength as a function of (a) tlie QW width aiid the (b) In concentration. The factor 1/3 due to the periodic part of the Bloch function is already included. We observe that, for practically all the situations, the $1s - E_1LH_1$ oscillator strengtli assumes significant values, typical of a type-I exciton. This is a combined effect of the Coulomb interaction aiid the coupling among the light-hole and split-off hole excitons. The electron, which is strongly localized in tlie $In_xGa_{1-x}As$ layer, acts as a center of attractive potential for the hole. The ground and first excited states of the split-off QW are also localized in the $In_xGa_{1-x}As$ layer, further contributing to the localization of the light-hole into this layer. This effect is illustrated in Figure 4 where it is plotted the projection of the hole z-part of the exciton wave-function. We see that, although in the absence of electron and hole correlation the system is type-II, the exciton state rather shows a type-I character. This explains some of the controversy about the light-hole band offset for this system[25,26]. Although the systern may be type-II in what tlie electron and light-hole particles confinement is conceriied, tlie exciton transitions are rather type-I. Therefore, the presence of a sharp peak at the light-hole exciton transition can not be a criteria to characterize the band offset.

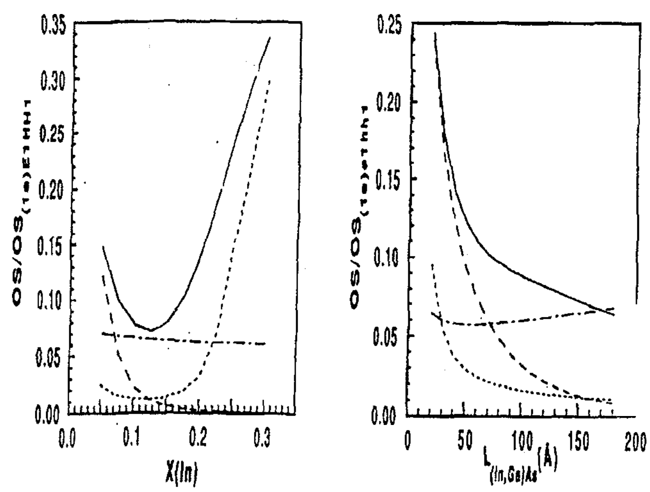

Figure 3: Relative oscillator strength for the transitions in an $In_xGa_{1-x}As$-GaAs quantum well (a) as a function of the In concentration for a $In_xGa_{1-x}As$ layer of 150Å and (b) as a function of tlie $In_xGa_{1-x}As$ layer for an In concentration of 12%. Full lines are tlie $1s - E_1LH_1$ exciton state including the split-off coupling. Dashed lines are the $1s - E_1LH_1$. exciton state without the split-off coupling. Dotted lines are the E_1LH_1 band-to-band transition including the split-off coupling and the dash-dotted lines are the $2s - E_1HH_1$ exciton state.

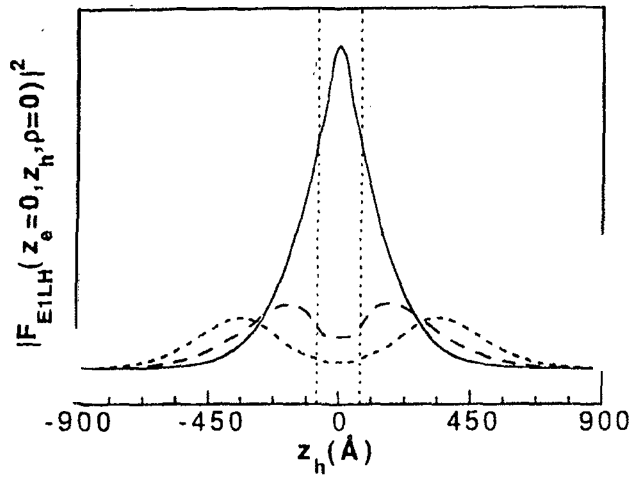

Figure 4: z-part of tlie Iiolc wave-function for an $In_{0.12}Ga_{0.88}As$-GaAs quantum well with an $In_{0.12}Ga_{0.88}As$ layer of 150 Å. Tlie GaAs layer is of 2000 Å. Full liiic is tlie $1s - E_1LH_1$ exciton state including the split-off coupling. Dashed line is tlie $1s - E_1LH_1$ exciton state without the split-off coupling. Dotted lines is the E_1LH_1 band-to-band traiisitioii including the split-off coupling.

iii) Resonant states

Let's concentrate now on the excitons associated to tlic same host-valence band aiid having the same in-plane symmetry. Within the separable variables approximation, each pair of electron and hole subbands gives a set of bound states followed by a continuum, for their in-plane relative motion. However, the three-dimensional character of the Coulomb interaction couples all these states. Actually, tlie bound states associated to the excited electron (liole) subbands are resonances of the ground electron and hole subband exciton[17-19]. Three main effects are observed due to the coupling of the bound states with tlie continuum states of lower subbands. The peak in the absorption associated to the bound exciton state is shifted in energy. A broadening is observed, due to the sharing of the oscillator strength of the bound state with the continuum states. A more spectacular effect is the supression of the absorption close to tlie exciton peak. This originates from a perfect destructive interference between the bound exciton and the continuum exciton wave-functions. This effect is well known from atomic and molecular physics and is known as the Fano resonance[27].

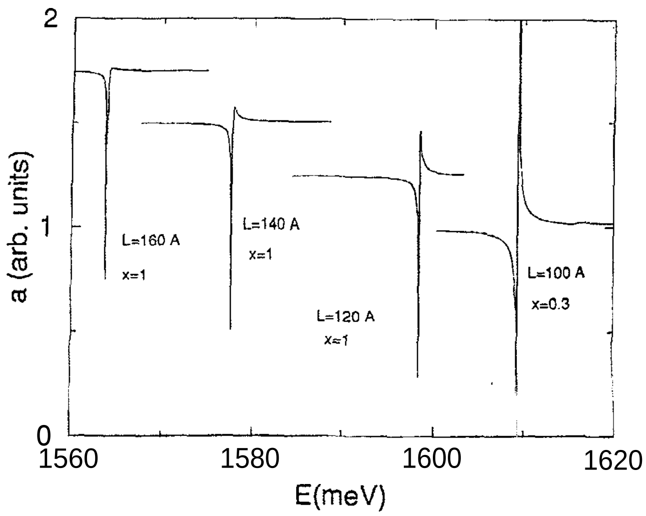

Figure 5: Normalized ahsorption spectra around the $1s - E_1HH_3$ exciton for $GaAs_xA_{1-x}As$ quantum wells with diffcrent well widths, L, aiid Al concentrations, x. The spectra intensities are shifted bv 0.2 for tlie sake of clearity. (after Ref. 18).

To investigate this cffect, we employed the method developed by Ekemberg and Altarelli[14]. Essentially, the QW exciton problem is clescribed within a.n effective-two- dimensional Coulomb interaction. The three-dimensional part of the interaction is then treated in perturbation following Fano's theory[18,27]. Figure 5 shows the absorption spectra of tlie $1s - E_1HH_3$ and the s-like continuum-E_1HH_1 exciton states for a series of single QW's. For all ttie QW's, the dip in the absorption appears on tlie low-energy side of the spectrum. This is a favorable situation for its observation. If it was localized on the high-energy side it would be difficult to resolve the dip from the 1s and 2s exciton states.

Homogeneous and inhomogeneous broadening may hamper tlie observation of this effect. Recently, however, a. suppression in the photoluminescence of excitation spectrum was observed at low temperatrue in high-quality double QW structures. It was succesfully identified to the Fano resonance formed by the coupling among tlie $1s - E_1HH_3$ aiid the s-continuum-E_1HH_1 exciton states[19].

IV. Final remarks

Tlie couplings among the exciton states reviewed here originate from intrinsic cliaracteristics of the semiconductor heterostructure system. Tliese effects add in richness to the simple hydrogen picture of the Wannier exciton. The couplings may be enhanced and monitored by tlie application of external fields along the growtl direction. The effects of an electric-field[22] and a magnetic-field[23] along tlie growth direction have been studied. They allowed the observation of the interaction among the exciton states. A more complex situation arises when we consider the presence of external fields applied perpendicular to the growth direction. In these caies, the external field breaks the cylindrical symmetry of the QW and additional coupling may be present[11].

Tlie observation of tliese fine structures is linked to the quality of the samples. We inay expect similar effects by reducing the dimensionality of the system. However, this has beeii shown to be quite a. difficult task. Some attempts have already shown some exciton effects in quantum wires[28] and quantum dot[29]. However, the quality of the samples does not allow yet the observation of fine structures. We hope that, with the improvement of tlie techniques for the of lateral patterning in the near future, it will be possible to observe exciton fine structures in even lower dimensional excitons.

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