Exchange effects on excitons in quantum wells

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Exchange splitting of quasi-two-dimensional excitons confined in quantum wells is studied theoretically by performing a simplified calculation with a scaling argument. Both the exchange energy and the longitudinal-transverse splitting are found to be enhanced drastically when the confinement of the exciton increases. Taking into account the difference of the enchange interaction between the heavy-hole and the light-hole exciton, the intrinsic splittings of the exciton ground states are calculated for GaAs-$Ga_{1-x}Al_xAs$ quantum wells within the envelope-function approximation.

Among many interesting topics of quasi two-dimensional materials, the problem of excitons in quantum-well (QW) structures have recently attracted much attention. It is well known that the confinement of the wave function increases both the binding energy and the oscillator strength of the exciton.[1,2] Another effect, recently observed experimentally,[3] concerns the important enhancement of the electron-hole exchange interaction (EHEXI) with increasing the two-dimensional (2D) confinement. In this paper we report a theoretical investigation of the exchange effect and related intrinsic splitting of the exciton in QW's.

The electron-hole exchange interaction closely depends on the spatial extension of the exciton wave function.[4–7] In the case of 3D excitons, the exchange energy is given by $\varepsilon_{ex}^{3D}=\Omega J_0 |\varphi_{1s}^{3D}(0)|^2$ and $|\varphi_{1s}^{3D}(0)|^2=(\pi a_0^{*3})^{-1}$ for the $1s$ ground state, where Ω is the volume of the unit cell, J_0 is the exchange integral, and a_0^* is the effective Bohr radius of the exciton.[4] In 2D materials like QW's, the size of the exciton wave function decreases rapidly and the overlap of the electron and hole increases with the decrease of the well thickness. This leads to an enhancement of the exchange effect which can be much larger than in 3D cases. Within the effective-mass approximation, the wave function of a nondegenerate exciton in a QW can be expressed by the following ansatz (the spin multiplicity will be included in the next part):

$$\Phi=\frac{1}{\sqrt{A}}e^{i\mathbf{K}\cdot\mathbf{R}}\varphi_\xi^{2D}(\mathbf{r}_e-\mathbf{r}_h)\chi_e(z_e)\chi_h(z_h)\times U_{c,0}(\mathbf{r}_e)U_{v,0}(\mathbf{r}_h)\ , \tag{1}$$

where A is the area, and $\mathbf{K}$ and $\mathbf{R}$ the in-plane wave vector and coordinate of the center of mass. φ_ξ^{2D} represents a quasi-2D exciton envelope function, specified by the quantum number ξ; χ_e and χ_h are solutions of the finite-square-well problem for the electron and the hole, respectively. $U_{c,0}$ and $U_{v,0}$ are the zone-center Bloch functions of the conduction and the valence bands. The envelope functions φ_ξ^{2D}, χ_e, and χ_h are slowly varying in comparison with the modulation on the atomic scale of the Bloch functions. The dominant contribution to the EHEXI arises when the electron and the hole are on the same site. Taking into account the difference in the probability of finding the electron and the hole in different unit cells, a simple algebra gives the exchange energy (short range of the EHEXI) as follows:

$$\varepsilon_{ex}^{2D}=\Omega J_0 W_{eh}\,|\varphi_\xi^{2D}(0)|^2 \tag{2}$$

with

$$W_{eh}=\int_{-\infty}^{\infty}dz[\chi_e(z)\chi_h(z)]^2\ . \tag{3}$$

Making use of the values of the 3D exciton, Eq. (2) becomes

$$\varepsilon_{ex}^{2D}=(\pi a_0^{*3})\varepsilon_{ex}^{3D}W_{eh}\,|\varphi_\xi^{2D}(0)|^2\ . \tag{4}$$

Similarly, the longitudinal-transverse splitting (long-range part of the EHEXI) is given by

$$E_{LT}^{2D}=(\pi a_0^{*3})E_{LT}^{3D}W_{eh}\,|\varphi_\xi^{2D}(0)|^2\ , \tag{5}$$

where E_{LT}^{3D} denotes the corresponding value of the 3D exciton.

Now let us consider realistic structures such as GaAs-$Ga_{1-x}Al_xAs$ QW's. For the sake of simplicity, we first suppose that in the range of our investigation, the exchange interaction has a small influence on the confinement of the exciton wave functions and the *perturbation treatment* is still valid. Next, we adopt the approximation of *decoupled heavy- and light-hole subbands.*[1] Since the heavy-hole and the light-hole subbands are energetically split, two types of Wannier excitons can be referred to: the heavy-hole exciton (HHE) and the light-hole exciton (LHE). Consequently, and different from the 3D exciton, two values are expected for $W_{eh}\,|\varphi_\xi^{2D}(0)|^2$; each one with its own well-thickness dependence. The excitonic transition energies without the EHEXI contribution are $E_h=\varepsilon_g+E_e+E_{HH}-R_{HH}$ and $E_l=\varepsilon_g+E_e+E_{LH}-R_{LH}$, where ε_g is the bulk GaAs band gap; E_e, E_{HH}, and E_{LH} are the first subband energies for electrons, heavy holes, and light holes, respectively. R_{HH} and R_{LH} are, respectively, the binding energy of the heavy-hole and the light-hole excitons. In the exciton basis $\{|m_h,m_s\rangle\}$ where m_h and m_s are the z components of the hole $J=\frac{3}{2}$ and the electron $S=\frac{1}{2}$ angular momentum, and in a spherical approximation, the exchange Hamiltonian of 3D excitons is written as[7] $H=\Delta_0+\Delta_1\mathbf{J}\cdot\boldsymbol{\sigma}$, where $\Delta_0=-\frac{3}{2}\Delta_1=\frac{3}{8}\Delta$ and $\Delta=\frac{4}{3}\varepsilon_{ex}^{3D}$. In the case of a QW, we have to take into account the difference between the envelope functions of the HHE and the LHE. Let $\Delta_h=\frac{4}{3}\varepsilon_{ex}$(HHE), $\Delta_l=\frac{4}{3}\varepsilon_{ex}^{2D}$(LHE) and $\Delta_{hl}=W_{eh}^{hl}(\Delta_h\Delta_l/W_{eh}^hW_{eh}^l)^{1/2}$, where W_{eh}^h and W_{eh}^l are given by (3), respectively, for the HHE and the LHE; $W_{eh}^{hl}=\int_{-\infty}^{\infty}dz\,\chi_e^2(z)\chi_h^h(z)\chi_h^l(z)$, where $\chi_h^h(z)$ and $\chi_h^l(z)$ are the solutions for the heavy hole and the light hole, respectively. Since the long-range part of the EHEXI only gives rise to an additional splitting of the exciton states which involve the spin singlet and we deal with a delocalized Wannier exciton, the longitudinal-transverse splitting corresponds simply to a shift toward high energy of the longitudinal exciton.[8,9] The total Hamiltonian including the EHEXI contributions takes the following form:

$$
\begin{array}{ccccc}
|\pm\frac{3}{2},\pm\frac{1}{2}\rangle & |\pm\frac{3}{2},\mp\frac{1}{2}\rangle & |\pm\frac{1}{2},\pm\frac{1}{2}\rangle & |+\frac{1}{2},-\frac{1}{2}\rangle & |-\frac{1}{2},+\frac{1}{2}\rangle
\end{array}
$$

$$
\begin{bmatrix}
E_h & 0 & 0 & 0 & 0\\
0 & E_h+\frac{3}{4}\Delta_h & \mp\frac{\sqrt{3}}{4}\Delta_{hl} & 0 & 0\\
0 & \mp\frac{\sqrt{3}}{4}\Delta_{hl} & E_l+\frac{1}{4}\Delta_l & 0 & 0\\
0 & 0 & 0 & E_l+\frac{1}{2}(\Delta_l+E_{LT}^{2D}) & -\frac{1}{2}(\Delta_l+E_{LT}^{2D})\\
0 & 0 & 0 & -\frac{1}{2}(\Delta_l+E_{LT}^{2D}) & E_l+\frac{1}{2}(\Delta_l+E_{LT}^{2D})
\end{bmatrix} .
$$

This gives, for the eigenvalues,

$$E_{1,2}=E_h \ ,$$

$$E_{3,5(4,6)}=\frac{1}{2}\left[E_h+E_l+\frac{3\Delta_h+\Delta_l}{4}\right]\pm\frac{1}{2}\left[\left[E_h+E_l+\frac{3\Delta_h-\Delta_l}{4}\right]^2+\frac{3}{4}\Delta_{hl}^2\right]^{1/2} ,$$

$$E_7=E_l+\Delta_l+E_{LT}^{2D} \ ,$$

$$E_8=E_l \ .$$

The fine structures of the HHE and the LHE consist, respectively, of two and three levels: the doublets $|3\rangle,|5\rangle$ and $|4\rangle,|6\rangle$ correspond to dipole-allowed transitions in σ polarization ($\mathbf{E}\perp[001]$). The singlet $|7\rangle$ (antisymmetric combination of $|+\frac{1}{2},-\frac{1}{2}\rangle$ and $|-\frac{1}{2},+\frac{1}{2}\rangle$) is dipole allowed in π polarization ($\mathbf{E}\parallel[001]$). The doublet $|1\rangle,|2\rangle$ and the singlet are forbidden. All the E_h, E_l, Δ_h, Δ_l, Δ_{hl}, and E_{LT}^{2D} are well-thickness dependent. The transition energies can be determined after finding all these contributions.

Our numerical investigation has been done for GaAs-$Ga_{1-x}Al_xAs$ quantum wells. First, we have calculated the first subband energies, E_e, E_{HH}, and E_{LH}, respectively, for electrons, heavy holes, and light-holes. Next we have evaluated the binding energies R_{HH} and R_{LH}, of the heavy-hole and the light-hole excitons as a function of the well width L. This has been done by a variational approach, similar to the one used by Greene and Bajaj[1] but including the effect of effective-mass mismatches between GaAs and $Ga_{1-x}Al_xAs$ and the variation of the effective masses with the subband gaps. Using the wave function obtained for the two types of excitons, the exchange energy and the longitudinal-transverse splitting have been obtained simultaneously. The values of the various physical parameters are $m_e=0.067$, $\gamma_1=6.85$, $\gamma_2=2.1$, and $\varepsilon=12.5$ for GaAs; $\gamma_1=3.45$ and $\gamma_2=0.68$ for AlAs. The corresponding parameters for $Ga_{1-x}Al_xAs$ were obtained by a linear interpolation. The energy-gap difference $\Delta\varepsilon_g$ between $Ga_{1-x}Al_xAs$ and GaAs is fairly well known:[10] $\Delta\varepsilon_g=1.115x+0.37x^2$ (eV). Concerning the band offset, we have taken $\Delta E_c=0.65\Delta\varepsilon_g$ and $\Delta E_v=0.35\Delta\varepsilon_g$ for the conduction band and the valence band, respectively. Figure 1 displays the well-width

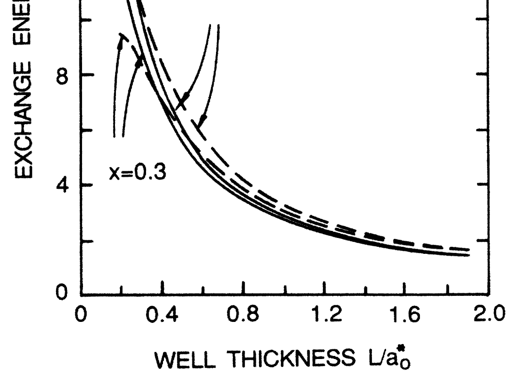

FIG. 1. Well-width dependence of the exchange energy for GaAs-$Ga_{1-x}Al_xAs$ quantum wells with aluminum contents of $x=0.3$ and 0.5. The solid lines correspond to the heavy-hole excitons, the dashed lines correspond to the light-hole excitons.

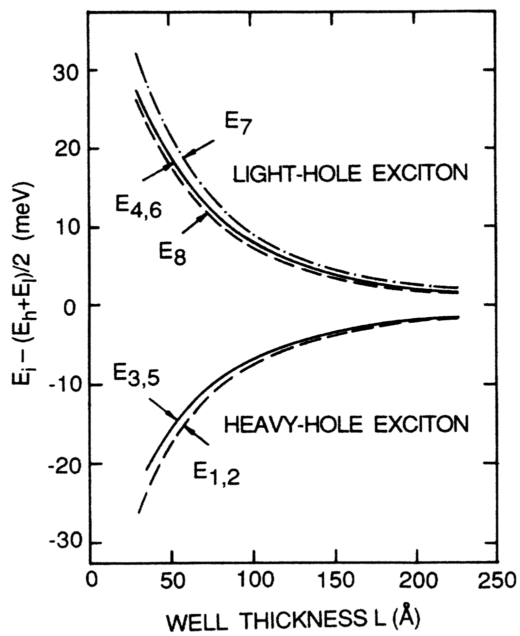

FIG. 2. Well-width dependence of the splitting between the transition energies for GaAs-$Ga_{0.5}Al_{0.5}As$ quantum wells. The $E_{3,5}$ and $E_{4,6}$ eigenstates are dipole allowed in σ polarization. The E_7 state is dipole allowed in π polarization and the others are forbidden.

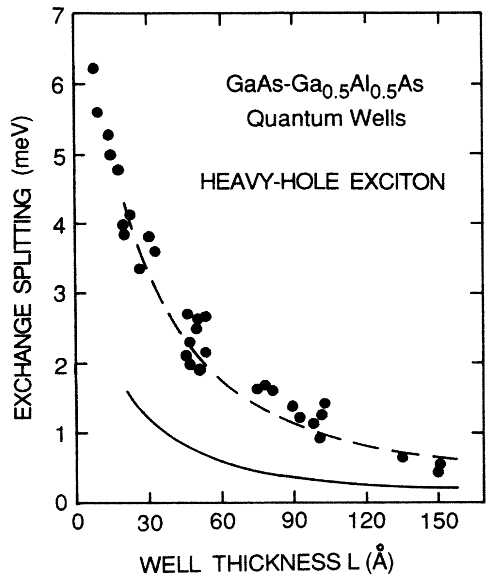

FIG. 3. Well-width dependence of the exchange splitting of the heavy-hole exciton for GaAs-$Ga_{0.5}Al_{0.5}As$ quantum wells. The solid and dashed lines are obtained from the theoretical calculation, using the bulk values of the exchange energy in GaAs $\Delta=0.1$ and 0.3 meV, respectively. The experimental points are taken from Ref. 3.

dependence of the exchange energy of the heavy-hole exciton Δ_h (solid lines) and the light-hole exciton Δ_l (dashed lines) for two different values of aluminum content (the energy is expressed in unit of ε_{ex}^{3D} and the 3D exciton Bohr radius is used to measure the well width). For $x=0.5$ QW's, the first excitonic transition in the barriers is indirect, but this seems likely not to significantly influence the resulting EHEXI. It is worth noting that the behavior of the EHEXI in QW's is similar to those of the exciton binding energy;[1] the relative increase in the exchange effect, however, is much more rapid than the effective Rydberg one when the well width decreases.

The well-thickness dependence of the splitting between transition energies, calculated for GaAs-$Ga_{0.5}Al_{0.5}As$ QW's, is given in Fig. 2. The bulk values of GaAs have been used as $\Delta=0.3$ meV (see below) and $E_{LT}^{3D}=0.08$ meV.[11] All energies are given with respect to the average value $(E_h+E_l)/2$. As mentioned above, the fine structure of the HHE exhibits two doublets from which one is dipole allowed (solid line) and the other one dipole forbidden. The fine structure of the LHE exhibits three states from which only one (the doublet) is dipole allowed (solid line) in σ polarization.

Finally, we try to compare our theoretical results with published experimental data. Recently, from photoluminescence experiments on a series of high-quality GaAs-$Ga_{0.5}Al_{0.5}As$ QW's, Bauer *et al.*[3] have evidenced a double substructure on the exciton spectra. This doublet concerns the dipole-allowed and dipole-forbidden transitions of the heavy-hole exciton. The observation of the forbidden one in low-temperature luminescence is due to thermalization effects. In that work,[3] the dependence of the splitting versus the well thickness was also studied. The authors found an increase of the exchange splitting with reduction of the well width. Figure 3 shows the variation of the heavy-hole-exciton exchange splitting as a function of the well width. The solid and dashed lines are obtained from the present calculation, using for the bulk value of GaAs $\Delta=0.1$ and 0.3 meV, respectively. The experimental points are taken from Ref. 3. Since the experimental value of the exchange energy Δ in 3D GaAs is not well known[12] and the absolute values of the experimental data of the splitting ΔE need further conformation, our theoretical fits should be only qualitative.

In conclusion, we have studied the electron-hole exchange interaction of Wannier exciton in quantum-well structures by performing a simplified calculation. It has been shown that both the short- and the long-range parts of the exchange interaction exhibit an important enhancement. The intrinsic splittings of the excitons due to the exchange effect are closely related to the 2D confinement of the exciton wave functions. Although our comparison between the theoretical fits and the experimental data are qualitative, a correct behavior of the relative increase of the heavy-hole-exciton exchange splitting has been obtained in the case of GaAs-$Ga_{0.5}Al_{0.5}As$ quantum wells.

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