Excitons in novel quantum materials:

a Monte Carlo study

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ABSTRACT

The exact binding energies of excitons in novel type-II $In_{1-x}Ga_xAs/GaSb_{1-x}As_y$ quantum well materials in the effective mass model are evaluated by diffusion quantum Monte Carlo simulations. The results support the experimental interpretation that a stable excitonic ground state can exist in spatially separated electron-hole systems. The electron(hole) correlation effects and quantum confinement are shown to enhance the binding of the excitons. The calculated results for various magnetic fields provide a guide for further experiments in accurate determination of the binding energies, which is important in the optoelectronic application of novel quantum well materials.

1. INTRODUCTION

Research activities in excitons experienced a sharp rise after Esaki and Tsu's seminal work on quantum well structures in 1970.[1] These structures display several interesting features, such as the enhancement of the binding energy of excitons and optical absorption over their bulk values, which results in well-defined exciton-absorption lines at room temperture. The majority of the study so far has been focused on the $GaAs/Ga_{1-x}Al_xAs$ system.[2] With $x \leq 0.40$, this system is a type-I quantum well in which electrons and holes are essentially confined in the same spatial regions. Dignam and Sipe[3] produced excitons in the "confinement-induced" type-II GaAs/AlAs superlattice, where electrons and holes are separated in different layers. The drawback of this system is that it is a confinement-induced type-II structure and hence is only type-II for a certain range of layer thickness with the GaAs layers thinner than 35 $\AA$ and the AlAs layers thicker than 20 $\AA$.

In this paper, diffusion and variational quantum Monte Carlo simulations of excitons in the type-II semiconductor quantum well system $In_{1-x}Ga_xAs/GaSb_{1-y}As_y$ are performed for a wide range of composition $(0,0) \leq (x,y) \leq (0.62,0.64)$ and well width $25\AA \leq L \leq 200\AA$ under the applied magnetic field. The exciton binding energy is found to increase with the decrease of the layer thickness and with the increase of the composition. The calculated results suggest that a novel semiconductor-excitonic insulator (Bose condensate)-semimetal transition should be observable in this system.

2. DIFFUSION MONTE CARLO TECHNIQUE

The diffusion quantum Monte Carlo (DQMC) approach employed in this work is very similar to that of Reynolds *et al.*[4]. It starts with the Schrödinger equation in imaginary time, $\tau = \frac{it}{\hbar}$,

$$\begin{aligned} \frac{\partial \Psi(\vec{R},\tau)}{\partial \tau} &= D\nabla^2 \Psi(\vec{R},\tau) - (V - E_0)\Psi(\vec{R},\tau) \\ &= -H\Psi(\vec{R},\tau) \end{aligned} \tag{1}$$

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where $H = T + (V - E_0)$ and $D = \frac{\hbar^2}{2m}$.

By choosing a probability density function $f(\vec{R},\tau) = \Phi(\vec{R})\Psi(\vec{R},\tau)$, , where $\Phi(\vec{R})$ is a trail wave function, we have

$$\begin{aligned} \frac{\partial f(\vec{R},\tau)}{\partial \tau} &= D\Phi(\vec{R})\nabla^2\Psi(\vec{R},\tau) + E_0\Phi(\vec{R})\Psi(\vec{R},\tau) - V\Phi(\vec{R})\Psi(\vec{R},\tau) \\ &= D\Phi(\vec{R})\nabla^2\Psi(\vec{R},\tau) - D\nabla^2\Phi(\vec{R})\Psi(\vec{R},\tau) + (E_0 - E_L)\Phi(\vec{R})\Psi(\vec{R},\tau) \end{aligned} \tag{2}$$

Here $E_L = \Phi^{-1}(\vec{R})H\Phi(\vec{R})$ is the local energy. Determining $\vec{F}(\vec{R}) = 2\nabla \ln \Phi(\vec{R})$ as a drifting velocity imposed on the diffusion, one can get

$$D\nabla^2 f(\vec{R},\tau) - D\nabla\cdot[f\vec{F}(\vec{R})] = D[-\Psi(\vec{R},\tau)\nabla^2\Phi(\vec{R}) + \Phi(\vec{R})\nabla^2\Psi(\vec{R},\tau)] \tag{3}$$

Combining eqs.(2) and (3), therefore, we convert the imaginary-time Schrödinger equation(1) of $\Psi(\vec{R},\tau)$ into a diffusion equation of f:

$$\frac{\partial f}{\partial \tau} = D\nabla^2 f - \nabla\cdot[f\vec{F}(\vec{R})] + [E_0 - E_l(\vec{R})]f \tag{4}$$

This equation resembles a diffusion plus a branching process, but now there is a drift velocity imposed on the diffusion, The singularities in the branching factor are now almost gone, since E_L (unlike V) is almost a constant. In fact, as $\Phi \to \Psi$, E_L approaches a constant. Furthermore, the drift term now guides the walk preferentially into the region where the wave function is large.

In equation(4),$\Psi(\vec{R},\tau)$ will approach the true ground-state wave function as long as $\Phi(\vec{R})$ is not orthogonal to the exact ground state of H, and the time-dependent expectation value

$$E(\tau) = \frac{\langle\Phi(\vec{R})|H|\Psi(\vec{R},\tau)\rangle}{\langle\Phi(\vec{R})|\Psi(\vec{R},\tau)\rangle} = \frac{\int d\vec{R} f(\vec{R},\tau)E_L(\vec{R})}{\int d\vec{R} f(\vec{R},\tau)} \tag{5}$$

will become the exact ground-state energy E_0 as $\tau \to \infty$. In general, the statistical error of the Monte Carlo data is given by the total simulation time. The time step interval is chosen so that the error introduced will be within the statistical error of the data.

In type-II $In_{1-x}Ga_xAs/GaSb_{1-y}As_y$ quantum wells shown in Fig.2, the exciton is formed by photoexciting electron-hole pairs, where the zero point of the energy is at the bottom (top) of the quantum well for the electron (hole). In the presence of an applied magnetic field along the growth direction, the Hamiltonian of an electron-hole pair can be written as

$$H(\vec{r}_e,\vec{r}_h) = H(\vec{r}_e) + H(\vec{r}_h) + V(\vec{r}_{eh}) + U_e(z_e) + U_h(z_h) \tag{6}$$

Where $\vec{r}_e$ and $\vec{r}_h$ are the electron and hole position, respectively. The kinetic and magnetic energies are

$$H(\vec{r}_\sigma) = \frac{1}{2m_\sigma}[-i\hbar\nabla_\sigma \pm \frac{e}{c}\vec{A}_\sigma]^2, \quad \sigma = \{e,h\} \tag{7}$$

where

$$\vec{A}_\sigma = \frac{1}{2}\vec{B}\times\vec{r}_\sigma \tag{8}$$

The Coulomb energy is

$$V(\vec{r}_{eh}) = -\frac{e^2}{4\pi\varepsilon_0\epsilon|\vec{r}_e - \vec{r}_h|}, \tag{9}$$

where $\epsilon = \sqrt{\epsilon_1\epsilon_2}$ is the average static dielectric constant of the two materials. The quantum well potential for the electron and the hole are respectively written as

$$U_e(Z_e) = V_e\Theta(|Z_e| - nd - \frac{L_e}{2}), \tag{10}$$

and

$$U_h(Z_h) = V_h\Theta(|Z_h - \frac{d}{2}| - nd - \frac{L_h}{2}) \quad (11)$$

Here $\Theta(x)$ is the step function ($\Theta(x) = 1$ for $x > 0$; $\Theta(x) = 0$ for $x \leq 0$), and V_e (V_h) is the magnitude of the conduction(valence) band discontinuy.

A better trail wave function will make the simulations converge faster. The trail state used in this work takes the form

$$\Phi(\vec{r}_e, \vec{r}_h) = f(\vec{r}_{eh})g(\vec{r}_{eh}) \prod_{\sigma=e,h} Q(z_\sigma)\omega(\rho_\sigma) \quad (12)$$

where $f(r) = \exp[-ar/(1+br)], g(r) = \exp[-a^{'}r^2/(1+b^{'}r)], \omega(\rho_\sigma) = \exp[-\mu_\sigma {\rho_\sigma}^2/(1+\lambda_\sigma\rho_\sigma)]$, and ${\rho_\sigma}^2 = {x^2}_\sigma + {y^2}_\sigma$. The quantum confinement functions are written as

$$Q(z_\sigma) = \begin{cases} \cos[k_\sigma(z_\sigma - nd\frac{z_\sigma}{|z_\sigma|})] & z_\sigma \text{in well} \\ \exp[-k^{'}{}_\sigma|z_\sigma - nd\frac{z_\sigma}{|z_\sigma|}|] & z_\sigma \text{in barrier} \end{cases} \quad (13)$$

The parameter k_σ and $k^{'}_\sigma (i = \{e, h\})$ can be obtained by the solution of the standard secular equation:

$$\cos(qd) = \cos(k_\sigma L_e)\cosh(k^{'}_\sigma L_h) - \frac{\xi - \xi^{-1}}{2}\sin(k_\sigma L_e)\sinh(k^{'}_\sigma L_h), \quad (14)$$

where $\xi = k_\sigma m^*_W / k^{'}_\sigma m^*_B$. Here we adopt the commonly used boundary conditions that Q and $Q^{'}/m^*_{W(B)}$ are continuous at the interface. m^*_W and m^*_B are effective masses in the well and the barrier, respectively. a is determined by the cusp condition. The other parameters b, $a^{'}$, and $b^{'}$ in Eq.(12) are optimized by the variational Monte Carelo simulation first before $\Phi(\vec{r}_e, \vec{r}_h)$ is used as a guide for the diffusion quantum Monte Carlo calculation.

Using equation (12), the drifting velocities in our problem are

$$\begin{aligned} \vec{F}_e(\vec{r}) = 2\{ & -[v(r)(x_e - x_h) + \mu_e u_e(\rho_e)x_e]\vec{i} \\ & -[v(r)(y_e - y_h) + \mu_e u_e(\rho_e)y_e]\vec{j} \\ & -[v(r)(z_e - z_h) + \tfrac{1}{Q(z_e)}\tfrac{\partial Q(z_e)}{\partial z_e}]\vec{k}\}, \end{aligned} \quad (15)$$

and

$$\begin{aligned} \vec{F}_h(\vec{r}) = 2\{ & [v(r)(x_e - x_h) - \mu_h u_h(\rho_h)x_h]\vec{i} \\ & -[v(r)(y_e - y_h) - \mu_h u_h(\rho_h)y_h]\vec{j} \\ & -[v(r)(z_e - z_h) + \tfrac{1}{Q(z_h)}\tfrac{\partial Q(z_h)}{\partial z_h}]\vec{k}\}, \end{aligned} \quad (16)$$

where

$$v(r) = \frac{a}{r(1+br)^2} + \frac{a^{'}(b^{'}r + 2)}{(1+b^{'}r)^2} \quad (17)$$

and

$$u_\sigma(\rho_\sigma) = \frac{2 + \lambda_\sigma\rho_\sigma}{(1+\lambda_\sigma\rho_\sigma)^2} \quad (18)$$

3. EXCITONS IN QWs WITH MAGNETIC FIELD

It is understood that the exciton binding energy in quantum well structures depends strongly on the strength of the applied magnetic field. In order to give a detailed study, six typical $In_{1-x}Ga_xAs/GaSb_{1-y}As_y$ quantum wells with various compositions (x, y): (0.00, 0.00)=A, (0.16, 0.15)=B, (0.25, 0.23)=C, (0.40, 0.35)=D, (0.55, 0.45)=E, and (0.62, 0.64)=F are used. The material parameters adopted in this work are taken from Ref.5. For example, Fig.1 shows the results as the composition (x, y)=(0.16,0.15) and the quantum well width L=90Å. In this work, the

confinement of holes are taken into account by using a realistic potential well with a finite depth. The resulting enlarged overlap of the wave functions of two particles increases the binding energy. Our calculations show clearly that E_B increases monotonically with the increasing of the magnetic field. We also got the binding energies without magnetic field. At $L = 30$Å, the calculated binding energies are 2.61 meV for InAs/GaSb and 2.98 meV for $In_{0.45}Ga_{0.55}As/GaSb_{0.55}As_{0.45}$. Our binding energy in InAs/GaSb is smaller than the two-dimensional limiting value(~5meV), but is about 3 times larger than the 0.9 meV given by Bastard *et al.*[6]. Based on the cyclotron resonance (CR)-X separation in a hydrogen-like model, an experimental estimation of the binding energy is about 4 meV[7].

The $In_{1-x}Ga_xAs/GaSb_{1-y}As_y$ system undergoes a transition from semiconductor state to semimetal state at $(x, y) \leq 0.25$[8], *where the conduction band edge of* $In_{1-x}Ga_xAs$ is lower than the valence band edge of $GaSb_{1-y}As_y$ due to size quantization effect. If the well width is less than or comparable to the effective Bohr radius of the exciton, the reduction in the binding energy of excitons caused by the separation is not very large. With an appropriate choice of materials, it should be possible to make $|E_G|$ less than the exciton binding energy E_B, so that one can observe the novel transitions among the semiconductor state, the excitonic insulator state with Bose condensate, and the semimetal state in the system.

The band gap energy E_G can be written as

$$E_G = \Delta E + \varepsilon_e^0 + \varepsilon_h^0 \tag{19}$$

where ΔE is defined as the energy difference between the bottom of the conduction band for $In_{1-x}Ga_xAs$ and the top of the valence band for $GaSb_{1-y}As_y$. ε_l^0 and ε_h^0 are the ground subband energies of electron and hole in the quantum wells respectively. Taking into account the exciton binding energy, the criterion for the phase transition can be written as

$$\begin{aligned} |E_G| - E_B < 0, &\quad \text{excitonic phase,} \\ > 0, &\quad \text{semiconductor phase,} \end{aligned} \tag{20}$$

In Fig.2 we give the phase diagram of a $In_{1-x}Ga_xAs/GaSb_{1-y}As_y$ quantum well with $(x, y) = (0.16, 0.15)$ as a function of L and v. Here $v = \frac{\hbar eB}{2\mu c R_y^*}$ and B is the applied magnetic field. It is clear that with an appropriate choice of materials one can observe a novel transition between the semiconductor state and the exciton state with Bose condensate in the present system.

In summary, we have presented the quantum Monte Carlo simulation results of the binding energy of the ground-state exciton in type-II $In_{1-x}Ga_xAs/GaSb_{1-y}As_y$ quantum wells under the applied magnetic field. For a wide range of composition $(0, 0) \leq (x, y) \leq (0.62, 0.64)$ and well width $25\text{Å} \leq L \leq 200\text{Å}$, we have studied the possibility of the novel semiconductor-excitonic insulator (Bose condensate)-semimetal transition in the system. The accurately calculated exciton ground state energies provide a reliable evaluation for a new type of excitonic matter, which is not only of fundamental interest but also important for practical device applications.

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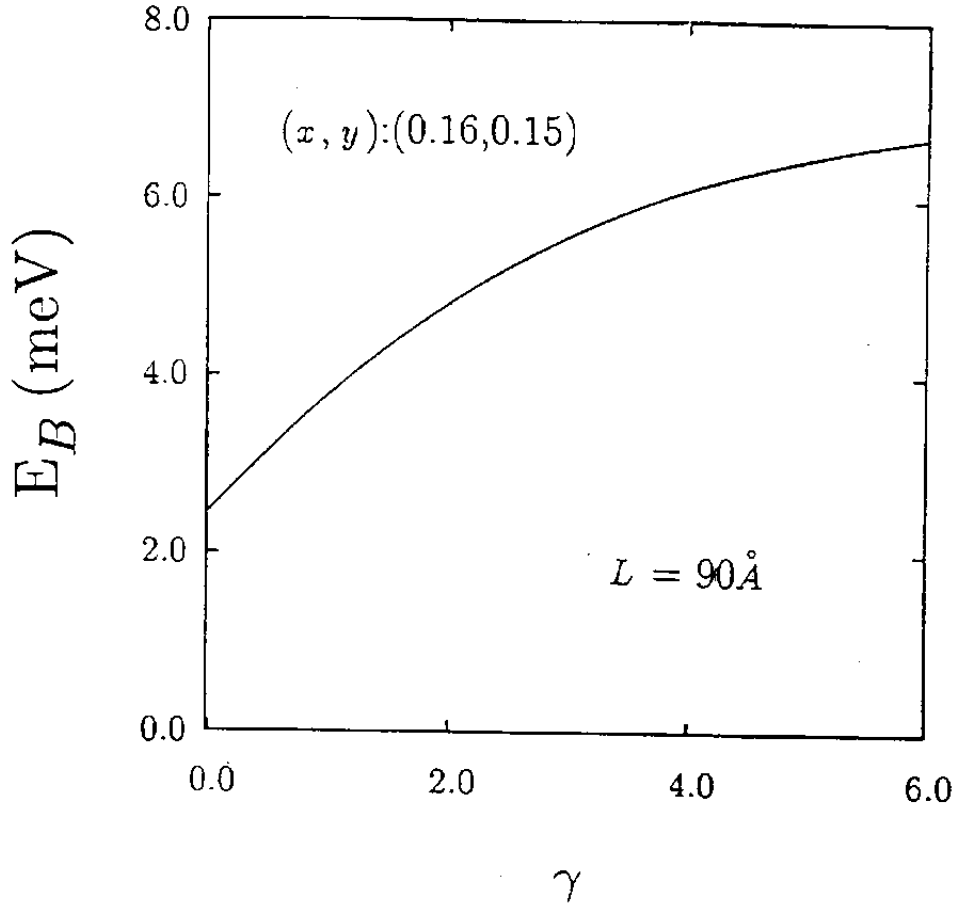

FIG.1 The exction binding energy (E_B) versus the magnetic field v with compositions (x, y)=(0.16,0.15) and quantum well width L=90Å, where $v = \hbar eB/(2\mu cR^*_y)$ and B is the applied magnetic field.

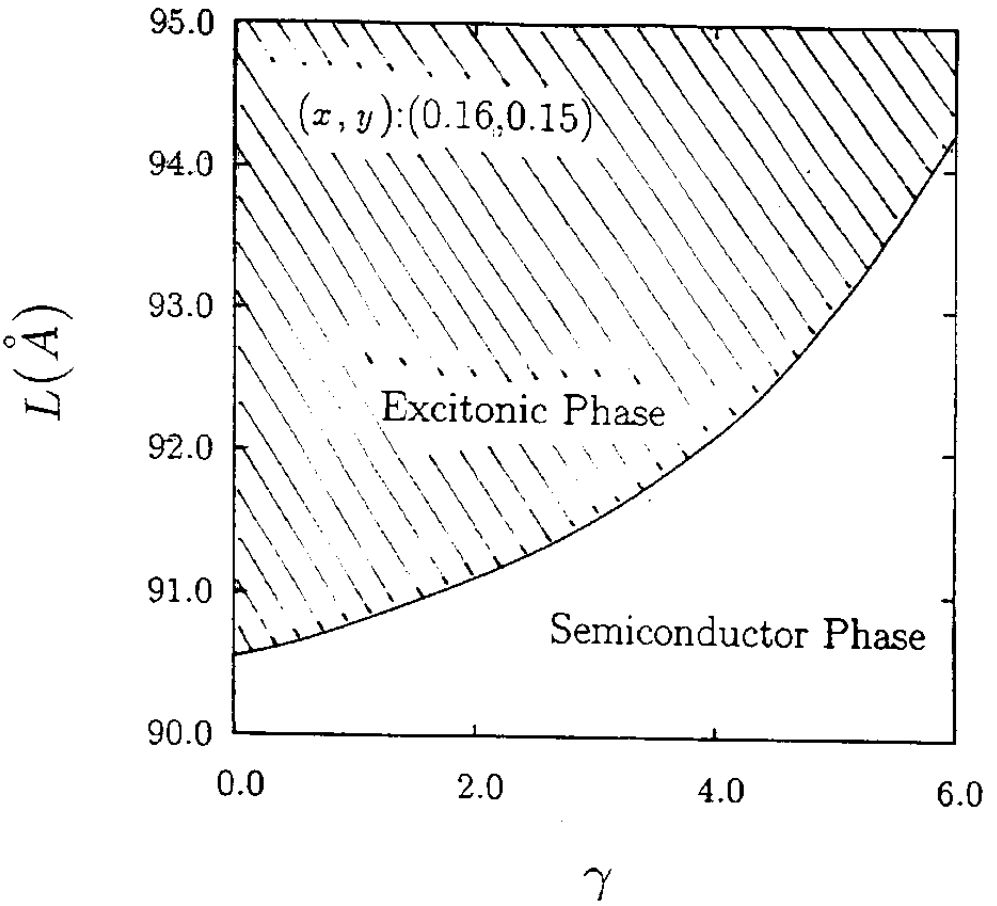

FIG.2 Phase diagram $L - \upsilon$ of a type-II $In_{1-x}Ga_xAs/GaSb_{1-y}As_y$ quantum well structure with $(x, y) = (0.16, 0.15)$, where $\upsilon = \hbar eB/(2\mu cR^*_y)$ and B is the applied magnetic field.