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Optical Spectra and Exciton State in Vertically Stacked Self-Assembled Quantum Discs *

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We study the oscillator strengths of the optical transitions of the vertically stacked self-assembled InAs quantum discs. The oscillator strengths change evidently when the two quantum discs are far apart from each other. A vertically applied electric field affects the oscillator strengths severely, while the oscillator strengths change slowly as the radius of one disc increases. We also studied the excitonic energy of the system, including the Coulomb interaction. The excitonic energy increases with the increasing radius of one disc, but decreases as a vertically applied electric field increases.

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Semiconductor quantum dots have been studied experimentally and theoretically in recent years,[1−7] due to the fact that these zero-dimensional structures have many interesting properties such as delta-function-like density of state[8−10] and potential applications for quantum computing and light storage devices.[11−13] Self-assembled quantum dots were also predicted to be the candidate for realizing the storage and retrieval of exciton.[14]

Korkusiński and Hawrylak presented a simple method to calculate electronic levels of a coupled vertically stacked self-assembled quantum-disk (QD) system (CVSSQS).[15] We presented further work to calculate the quantum-confined Stark effect of the CVSSQS.[16,18] In this Letter, we study the oscillator strengths of the optical transitions of the InAs/GaAs CVSSQS and the excitonic energy of the CVSSQS including the Coulomb interaction.

The schematic picture of the CVSSQS can be found by Korkusiński and Hawrylak.[15] The system consists of two vertically stacked InAs QDs. Each disc is formed on a wetting layer (WL) of width W, and covered by the GaAs barrier material. In the system, H is the QD height and D is the thickness of the middle-tunnelling barrier. R_1 and R_2 are the lower- and upper-disk radii respectively. The difference between the conduction band offsets of the quantum discs and the surrounding material gives the confining potential V_0.

The CVSSQS radial symmetric axis was set as the z-axis which points from the lower disc to the upper one. We set the middle point of the tunnelling barrier between the QDs as the origin O of the radial coordinate system. The effective Rydberg $R_{ed} = m_d e^4/2\epsilon^2\hbar^2$ and the effective Bohr radius $a_{Bd} = \epsilon\hbar^2/m_d e^2$ were used as the units of energy and length, respectively. Here m_d, e, and ϵ are the electron effective-mass in the discs, the electron charge, and the dielectric constant, respectively.

Ignoring the exciton effect, the optical-transition matrix element can be calculated by

$$\begin{aligned}\langle \Phi_e|\boldsymbol{P}|\boldsymbol{\Phi}_h\rangle &= \iint r\mathrm{d}z\mathrm{d}r g^{\nu}_{re}(z) f^{\nu}_{me}(r) g^{\nu'}_{rh}(z) \\ &\quad \cdot f^{\nu'}_{m'h}(r)\langle m|m'\rangle\langle c|\boldsymbol{P}|\upsilon\rangle \\ &= \iint r\mathrm{d}z\mathrm{d}r g^{\nu}_{re}(z) f^{\nu}_{me}(r) g^{\nu'}_{rh}(z) \\ &\quad \cdot f^{\nu'}_{m'h}(r)\delta_{mm'}\langle c|\boldsymbol{P}|\upsilon\rangle \\ &= I_{eh}\boldsymbol{P}_{cv}\delta_{mm'}, \end{aligned} \tag{1}$$

where $g^{\nu}_{re}(z)$ and $f^{\nu}_{me}(r)$ ($g^{\nu'}_{rh}(z)$ and $f^{\nu'}_{m'h}(r)$) are the eigenfunctions of the electron (hole) in the z direction and the r direction, respectively. The ν (ν') is the subband index, and m (m') is the angular momentum channel, $|m\rangle$ ($|m'\rangle$) is the angular wavefunction, $|c\rangle$ and $|\upsilon\rangle$ are the Bloch wavefunctions at the conduction-band bottom and valence-band top, and I_{eh} is the overlap integral for the envelope functions of the electron and hole. Then the oscillator strength of the optical transition is defined as $K = |I_{eh}|^2$.

The Hamiltonian of the exciton can be written as

$$H = H_e + H_h + V_{e-h}, \tag{2}$$

where H_e and H_h are the effective-mass Hamiltonian of the electron and hole respectively, V_{e-h} is the Coulomb interaction term between the electron and hole. H_e can be written as

$$H_e = -\frac{1}{K_e}\Big[\frac{1}{r_e^2}\Big(r_e\frac{\partial}{\partial r_e}r_e\frac{\partial}{\partial r_e} + \frac{\partial^2}{\partial\theta_e^2}\Big)$$

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$$-\frac{\partial^2}{\partial z_e^2}\Big] - eFz_e + V_e(r_e, z_e), \tag{3}$$

$$H_h = -\frac{1}{K_r}\Big[\frac{1}{r_h^2}\Big(r_h\frac{\partial}{\partial r_h}r_h\frac{\partial}{\partial r_h} + \frac{\partial^2}{\partial\theta_h^2}\Big)\Big] + \frac{1}{K_z}\frac{\partial^2}{\partial z_h^2} + eFz_h + V_h(r_h, z_h), \tag{4}$$

$K_e = 1$ in discs (including WL), and $K_e = m_b/m_d$ in potential barrier regions. $K_r = m_r/m_d$ and $K_z = m_z/m_d$, m_r (m_z) is the effective-masses of the hole in r (z) direction. $V_e(r_e, z_e) = -V_{0e}$ ($V_h(r_h, z_h) = -V_{0h}$) inside the quantum discs, and in the barrier regions $V_e(r, z)$ ($V_h(r_h, z_h)$) is zero. The effective-mass Hamiltonians of the electron and hole were given in our previous work, together with corresponding eigenvalues.[18]

$$V_{e-h} = -\frac{2}{r_{eh}}.$$

The exciton wavefunction can be expanded in terms of electron and hole wavefunctions

$$\Psi_{ex} = \sum_{\nu,m,\nu',m'} C_{\nu m\nu' m'}\,\Phi_{\nu m}\Phi_{\nu' m'} \tag{5}$$

Then by solving the following secular equation, we can obtain the exciton energy,

$$|(E^e_{\nu m} + E^h_{\nu' m'} - E)\delta_{mn}\delta_{m'n'} + V_{\nu m\upsilon n,\nu' m'\upsilon' n'}| = 0, \tag{6}$$

where $E^e_{\nu m}$ and $E^h_{\nu' m'}$ are the eigenenergies of the electron and hole respectively, $V_{\nu m\upsilon n,\nu' m'\upsilon' n'}$ is the matrix element of the Coulomb interaction.

V_{0e} and ϵ were taken to be 0.6 eV[15] and $15.15\epsilon_0$,[17] respectively. The effective masses of the electron $m_b = 0.0665m_0$ and $m_d = 0.04m_0$.[17] Our calculations were limited to the heavy hole. The effective-masses of heavy hole in the z direction and in the r direction are: $m_z = m_0/(\gamma_1 - 2\gamma_2)$, $m_r = m_0/(\gamma_1 + \gamma_2)$ respectively, $\gamma_1 = 19.67$ and $\gamma_2 = 8.37$ for InAs, $\gamma_1 = 6.85$ and $\gamma_2 = 2.1$ for GaAs.[17] V_{0h} and the forbidden gap energy of GaAs were taken as 0.295 eV[19] and 1.42 eV, respectively. $R_1 = R_2 = 8.5$ nm, $H = 2$ nm and $W = 0.54$ nm.

Figure 1(a) shows the dependence of oscillator strengths of the InAs/GaAs CVSSQS optical transitions on the thickness of the middle-tunnelling barrier D. The symbol E_mH_n ($m, n = 0, 1$) is used to denote that the transition is between the states of the electron and the heavy hole, m (n) $= 0$ means the bound state of the electron (heavy hole) and m (n) $= 1$ means the first excited state of the electron (heavy hole). We can see that the transition E_0H_0 is very strong (E_0H_1 and E_1H_0 are very weak) when the two discs are close to each other. When D is in the range 7 nm–9.5 nm, the oscillator strengths change evidently. Then the three oscillator strengths are very close.

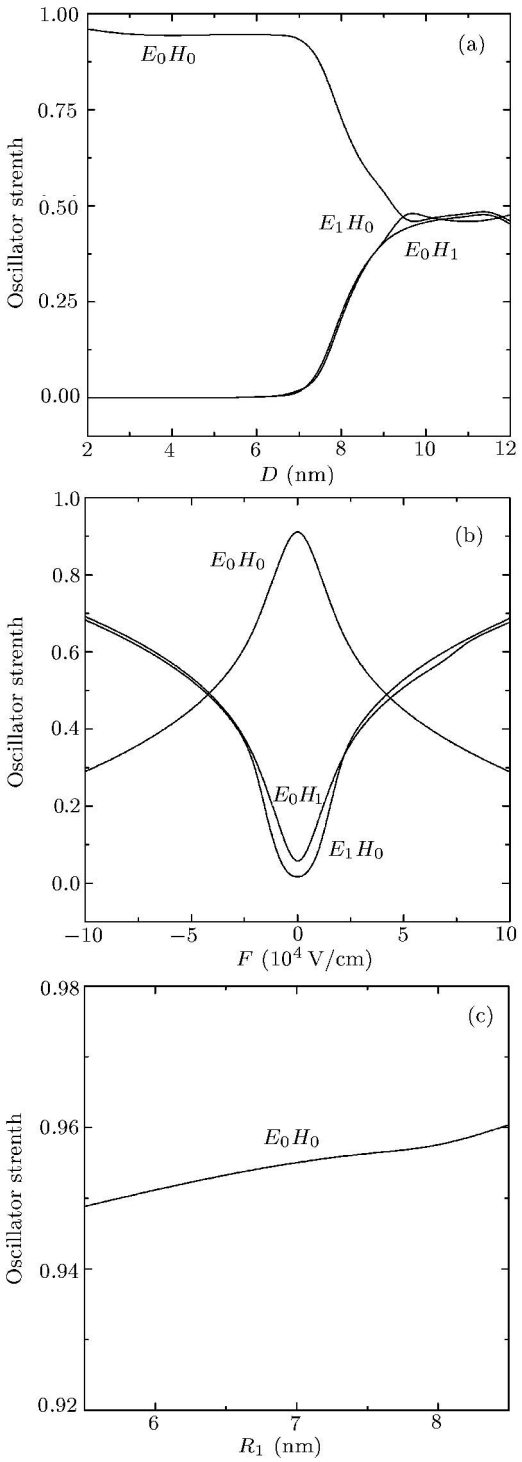

Fig. 1. (a) Oscillator strengths versus the thickness of the middle-tunnelling barrier D. (b) Oscillator strengths versus a vertically applied electric field $\boldsymbol{F}$, $D = 2$ nm. (c) Oscillator strength E_0H_0 as a function of the lower-disk radius R_1, $D = 2$ nm.

Let us analyse the phenomena qualitatively as follow. The motion can be separated in the growth (z) and radial directions. In the z direction, we deal with the two coupled quantum wells, the wavefunction can be written in forms of symmetric and antisymmetric

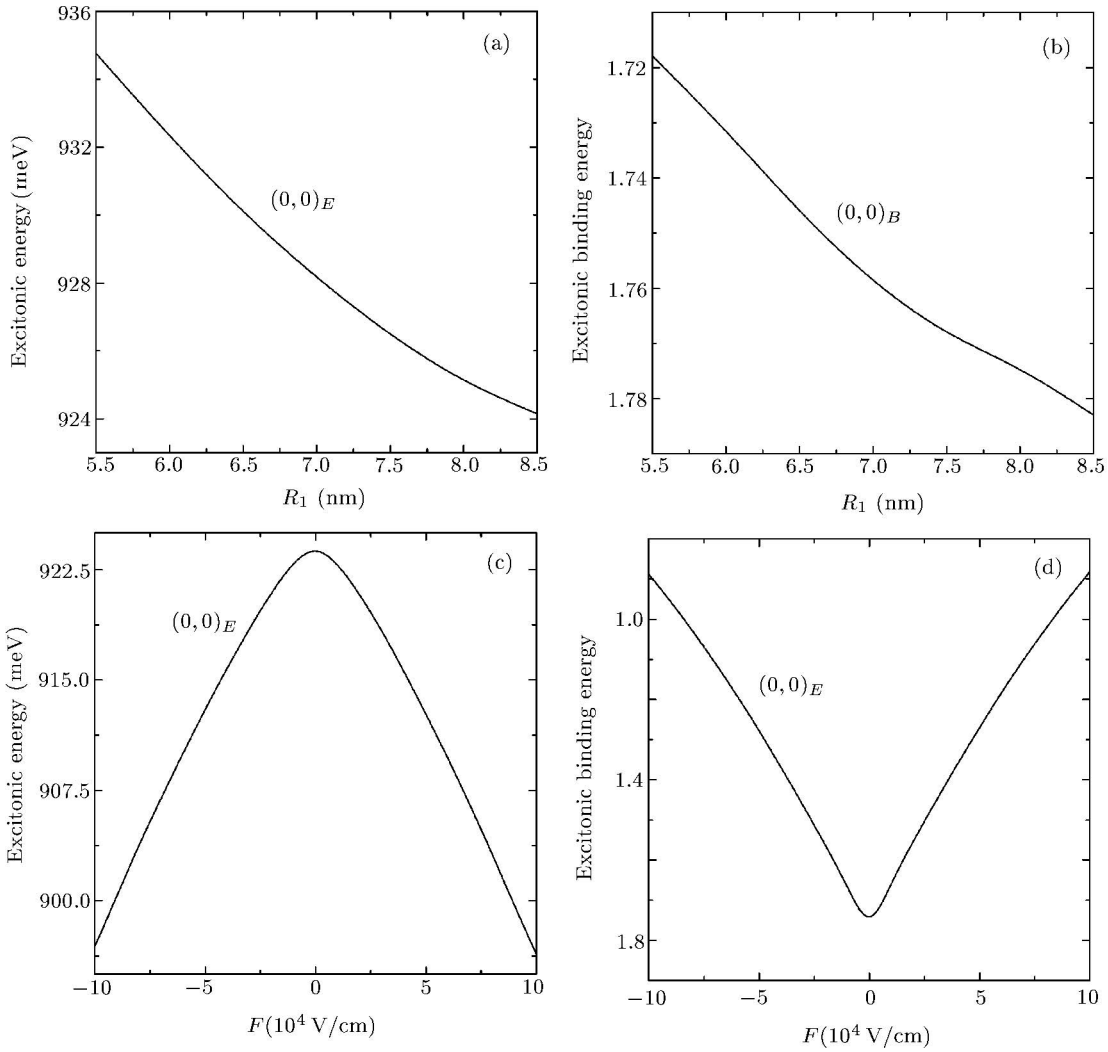

Fig. 2. (a) Excitonic energy versus the lower-disk radius R_1. (b) Excitonic binding energy versus the lower-disk radius R_1. (c) Excitonic energy as a function of a vertically applied electric field $\boldsymbol{F}$. (d) Excitonic binding energy as a function of vertically applied electric field $\boldsymbol{F}$, $D = 2\,\mathrm{nm}$. Here the excitonic binding energy is in units of the effective Rydberg $R_{ed} = m_d e^4/2\epsilon^2\hbar^2$ with m_d, e, and ϵ being the electron effective-mass in the discs, the electron charge, and the dielectric constant, respectively.

linear combinations of the single quantum well orbit: $f_s(z) = [f_1(z) + f_2(z)]/\sqrt{2}$, $f_{as}(z) = [f_1(z) - f_2(z)]/\sqrt{2}$, $f_1(z)$ and $f_2(z)$ are the corresponding orbit of each isolated well. Considering the electron only, the bound state (first excited state) wavefunction is symmetric (antisymmetric). The probability distributions of the electron in the two discs are the same. It is the same case to the heavy hole when $D < 7\,\mathrm{nm}$. If the distance between the two discs is so far ($D > 9.5\,\mathrm{nm}$) that D is a macro quantity to heavy hole, the heavy hole is mainly in one disc. Then the bound state (first excited state) wavefunction of the heavy hole in the z direction $f_{sh}(z) = [f_{1h}(z) + f_{2h}(z)]/\sqrt{2} \to f_{1h}(z)$ or $f_{2h}(z)$ ($f_{ash}(z) = [f_{1h}(z) - f_{2h}(z)]/\sqrt{2} \to f_{1h}(z)$ or $-f_{2h}(z)$). This is why the oscillator strength E_0H_0 is almost half of that when D is little, and the transitions E_0H_1 and E_1H_0 are close to the transition E_0H_0 when $D > 9.5\,\mathrm{nm}$. To heavy hole, D changes from a micro quantity to a macro quantity in the region $7\,\mathrm{nm} < D < 9.5\,\mathrm{nm}$, so the oscillator strengths change quickly.

Figure 1(b) shows the oscillator strengths of the optical transitions as functions of a vertically applied electric field, when $D = 2\,\mathrm{nm}$. We can see that the oscillator strength E_0H_0 decreases with field increases, but it is on the contrary to oscillator strengths E_0H_1 and E_1H_0. The stronger the field, the more evident the changes. Positive and negative fields affect the oscillator strengths in the same way.

Figure 1(c) shows the oscillator strength of the optical transition E_0H_0 as a function of the lower-disk radius R_1, when $D = 2\,\mathrm{nm}$. The oscillator strength increases slowly as R_1 increases. Because the distribution probabilities of the electron and the heavy hole

are mainly in the discs, and the shorter the radius, the larger the probabilities. The distribution probabilities of the electron and the heavy hole are very small in the region $R_1 < r < R_2$. This makes the E_0H_0 increases slowly with R_1. E_0H_1 and E_1H_0 are close to zero in our calculation.

Then we calculate the excitonic energy of the InAs/GaAs CVSSQS including the Coulomb interaction. Our calculation is limited to the bound state of the exciton (the electron and the heavy hole are both in the bound state). Here $(0,0)_E$ and $(0,0)_B$ are used to denote the excitonic energy and the excitonic binding energy, respectively. Figures 2(a) and 2(b) show the dependence of the excitonic energy and the excitonic binding energy of the CVSSQS on the lower-disk radius R_1 respectively, when $D = 2\,\mathrm{nm}$. The excitonic energy $(0,0)_E$ decreases quickly as R_1 increases, while the excitonic binding energy decreases slowly as R_1 increases.

Figures 2(c) and 2(d) show the excitonic energy and the excitonic binding energy of the CVSSQS as functions of a vertically applied electric field, when $D = 2\,\mathrm{nm}$. The field lowers the excitonic energy, but lifts the excitonic binding energy. The stronger the field, the lower (higher) the excitonic energy (the excitonic binding energy).

In conclusion, the oscillator strengths of the optical transitions change evidently when the two discs are far apart from each other, or the system is under a vertical electric field. The excitonic energy and the excitonic binding energy decrease with increasing the lower-disk radius. Vertical electric field lowers the excitonic energy, but lifts the excitonic binding energy.

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