Diffusion Monte Carlo study on excitonic complexes in type-II core-multishell nanowires

Takuma Tsuchiya
Division of Applied Physics, Graduate School of Engineering
Hokkaido University (Hokudai)
Kita 13 Nishi 8, Kita-ku, Sapporo 060-8628, Japan
E-mail: t.t@physics.org

Abstract. Binding energies of excitons and excitonic complexes in BeTe/ZnSe type-II core-multishell cylindrical nanowires are calculated by quantum Monte Carlo method. Binding energies are enhanced for negatively charged excitons, and reduced for positively charged excitons and biexcitons. These results are ascribed to excitonic Coulomb potential comes from different spatial distribution of electrons and holes.

1. Introduction
Spatial confinement of individual particles in nanostructures has strong influence on properties of excitons and excitonic complexes, i.e. biexcitons and charged excitons (trions). In type-I structures, in which electrons and holes are in the same spatial region, their binding energies are usually enhanced, because of the enhancement of the Coulomb energy between electrons and holes. However, in some structures, spatial confinement for electrons and holes is different, and binding energies are reduced and negative sometimes. [1-3] This feature is further pronounced in type-II structures, in which electrons and holes are spatially separated. Moreover, in type-II superlattices, for example, excitonic macro-molecules are predicted, as a result of an interaction between persistent electric dipoles of excitons. [4, 5]

Recent development of crystal growth technique has enabled us to grow core-multishell nanowires, [6, 7] and now it is possible to make type-II nanowires with good quality. In this paper, we investigate binding energies of excitons and excitonic complexes in type-II BeTe/ZnSe core-multishell nanowires, using quantum Monte Carlo method.

2. Calculation method
In the quantum Monte Carlo method, we use the imaginary time $\tau = it$ instead of time $t$, in the time-dependent Schrödinger equation. [8] As a result, the Schrödinger equation becomes a classical time-evolution equation with diffusion- and rate-equation term. This type of equation can be solved numerically by Monte Carlo method, and we can obtain exact ground-state energy within a small statistical error.

To improve the calculation efficiency, we usually employ trial wavefunction $\Psi$, in which important characteristics of actual wavefunction is taken into account. Then the equation to be solved is
Figure 1. Structure of BeTe/ZnSe type-II core-multishell nanowire.

where \( \Phi \) is the wavefunction, \( D = \hbar^2/2m_e F^2 = 2V/\Psi \), and \( E_e = H/\Psi \). For the trial wavefunction for negatively charged excitons, for example, we employ

where \( \phi_e(\vec{r}) \) and \( \phi_h(\vec{r}) \) are one particle wavefunctions for electrons and holes, respectively. For the present structure, shown in Fig. 1, we can obtain the accurate analytical forms of \( \phi_e(\vec{r}) \) and \( \phi_h(\vec{r}) \), using the Bessel and Neumann functions. For the inter-particle correlation function \( \Phi \), we employ

where \( r_{12} = |\vec{r}_1 - \vec{r}_2| \), \( a_{12}^* = -e\hbar^2/\mu_{12}q_1q_2 \) is the effective Bohr radius with the reduced mass \( \mu_{12} \), the charge \( q \), and the dielectric constant \( \varepsilon \), and \( b \) an appropriate coefficient. This function describes correlation wavefunction for \( r_{12} \to 0 \) accurately, and the resulting kinetic energy cancels the divergence of the Coulomb potential. The form of the trial function affects the accuracy of the results.

Though cross section of actual core-multishell nanowires is hexagonal, we assume cylindrical symmetry for the first step. The parameters used in the present calculation, are shown in Table 1. Binding energies for excitons and negatively charged excitons, for example, are defined by

\[
\text{Binding energy} = (E_e + E_h) - (E_e + E_h) - E_X
\]

where \( E_e \), \( E_h \), \( E_e \), and \( E_h \) are the ground state energies for an electron, a hole, an exciton, and a negatively charged exciton, respectively.

Table 1. Parameters used in the numerical calculation.

| Parameter       | Effective mass \( m_0 \) | BeTe | ZnSe | ZnBeMgTe | Dielectric constant |
|-----------------|--------------------------|------|------|----------|---------------------|
| Electrons       | 0.15                     | 2,300| 0    | 250      | 8.0                 |
| Holes           | 0.34                     | 0    | 900  | 1,000    |                     |
3. Results

Figure 2 shows the binding energies of excitons. Because of the spatial separation between an electron and a hole, binding energies are smaller than the binding energy of 22.1 meV in the bulk for the same parameters. However, for some wires thinner than the effective Bohr radius, ~4 nm, the binding energy exceeds the bulk value.

The binding energy of negatively charged excitons, shown in figure 3, is larger than the bulk value, 1.0 meV, and reaches 5 meV for thinner wires. On the other hand, those for positively charged excitons and biexcitons (not shown) are quite small for thinner wires and vanish for thicker wires, in spite that their binding energies are larger than negatively charged excitons in the bulk: 1.4 and 1.7 meV for positively charged excitons and biexcitons, respectively. These results are explained qualitatively through the Coulomb potential due to a cylindrical electric-double-layer caused by the type-II exciton. We assume, for simplicity, that the charge density is uniform along the wire and independent of angular coordinate. Then the electric potential $V(r)$ is given by

$$V(r) = -\int_0^r E_r(r) dr,$$

$$E_r(r) = \frac{1}{2\pi \varepsilon \varepsilon_0 r} \int_0^r Q_m(r') dr'$$

where $E_r$ is the electric field along radial direction, $Q_m$ the electric charge inside of the radius $r$, $\rho_z$ the charge density per unit length, $\psi_e$ and $\psi_h$ are normalized radial wavefunctions for electrons and holes. In the present structure, the hole and the electron of an exciton are in the core and in the inner shell, respectively. As a result, the electric field caused by the exciton points outside of the wire, and the resulting potential is attractive for an extra electron and repulsive for an extra hole, as shown in figure 4. For an extra exciton, the potential is repulsive, because the repulsive potential for a hole is larger than that for an electron. Note that for reversed structures in which electrons are in the core and
holes in the shell, binding energies for negatively and positively charged excitons should be reduced and enhanced, respectively.

Enhanced binding energy for negatively charged excitons for thinner wires, however, is not a result of this excitonic potential, because its depth is independent of the wire radius. Actually, the factor $1/r$ in eq. (5) and the factor $r$ in eq. (6) cancel each other out. In figure 5, the ratio of binding energy of a negatively charged exciton to that of an exciton is shown. In spite of large binding energy for thinner wires, the ratio decreases with decreasing $d_2$. This indicates that the increasing binding energy of negatively charged excitons for thinner wires is due to the enhancement of the electron-hole Coulomb energy, i.e. the exciton binding energy, and not due to enhancement of inter-particle correlation.

4. Summary

In summary, we have investigated binding energies of excitons and excitonic complexes in type-II core-multishell nanowires. Binding energy for negatively charged excitons is strongly enhanced, and those for positively charged excitons and biexcitons are reduced much. These results are explained by the Coulomb potential comes from the charge distribution of an exciton.

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