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Transformation from Type-Ⅱ to Type-Ⅰ Excitons in Spherical Core-shell Quantum Dots

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Abstract: The excitons in the spherical semiconductor core-shell quantum dots embedded in an insulator matrix were studied theoretically in a simplified model within the framework of effective mass approximation by using the variational approach. The calculations for the values of the wave functions as functions of coordinate, and the critical potential as functions of the ratio of shell width to core radius were performed numerically, respectively. It was found that there exists a critical potential for a given ratio of the shell width to core radius, when the potential decreases cross this critical value the exciton transforms from a type-Ⅱ exciton into a type-Ⅰ exciton. For a given potential, there exists a critical ratio of the shell width to core radius, when this ratio decreases cross this critical value the exciton transforms from a type-Ⅱ exciton into a type-Ⅰ exciton. The essential features were clarified, which may serve as a starting point for the analysis of excitons in real materials.

Key words: exciton; quantum dot; variational approach

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1 Introduction

During the last decade, technology advances in colloid chemistry have led to fabrication of high-quality and size-controlled semiconductor nanocrystal core-shell structure quantum dots[1~5]. According to their band alignment, the heterostructures can be classified as type-Ⅰ (the minimum of conduction band and the maximum of valence band are in the same material) and type-Ⅱ (the minimum of conduction band is in the core and the maximum of valence band is in the shell material, or rice versa) core-shell structures. The excitons in the heterostructures therefore can be classified type-Ⅰ exciton and type-Ⅱ exciton. Theoretically, a few approaches have been used to study the core-shell quantum dots including the effective mass approximation[6~8], envelop function method[9~11], empirical tight-binding method[12], and atomistic calculation[13].

Type-Ⅰ exciton and type-Ⅱ exciton has dramatically different properties. For example, in general the exciton binding energy in a type-Ⅱ core-shell quantum dot is much smaller than the binding energy of a bulk hydrogenic impurity, but the binding energy of type-Ⅰ exciton is larger than the binding energy of a bulk hydrogenic impurity due to the effect of the overlapping electron and hole wave functions within the dot. Another instance is the Stark effect in the core-shell quantum dots. The transition energy for type-Ⅰ exciton generates a parabolic redshift under an electric field, but a linear blueshift for type-Ⅱ exciton[14]. However, for the lower barrier height case, with wave function leakage into the dot and the shell, the binding energy of a type-Ⅱ exciton can be larger than the binding energy of a bulk hydrogenic impurity, which displays a typical

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type-Ⅰ exciton behavior. More recently, Bartnik *et al.*[15] studied the electronic structure of PbSe/PbS core-shell quantum dots within a four-band envelop function formalism. It was found that even though PbSe/PbS is a type-Ⅱ heterostructure, but for typical quantum dot size, the type-Ⅱ properties can not be observed. Instead, the wave functions are predicted to extend throughout both materials. That means an exciton in a type-Ⅱ core-shell quantum dots may not be a type-Ⅱ exciton.

In the present work, we study theoretically the transformation of excitons in spherical semiconductor core-shell quantum dots embedded in an insulator matrix in a simplified model within the framework of effective mass approximation by using the variational approach. Some numerical results are presented and the essential features are clarified, which may serve as a starting point for the analysis of exciton in real materials.

2 Theory

Let us consider a spherical core-shell quantum dot consisting of a semiconductor core with radius R_1 surrounded by another semiconductor shell with width of ΔR (therefore the outer radius of the shell is $R_2 = R_1 + \Delta R$). The core-shell quantum dot is embedded in an insulator matrix. Assuming that the valence band offset is so large that the hole can be considered completely confined in the core, the wave function of the hole is

$$\psi_h = \sqrt{\frac{2}{R_1^3 j_1^2(x_{10})}} j_0(x_{10} r_h / R_1) Y_{00}(\theta, \varphi) \quad (1)$$

where j_0 is zero-order spherical Bessel function which has its first zero at $r_h = R_1$, $x_{10} = \pi$, and Y_{00} is the corresponding spherical harmonic function. We assume that the hole wave function is fixed in our problem, therefore considering the electronic structures only is enough.

With the assumption that the different dielectric constants of the materials can be accounted for as single effective-medium dielectric constant and the effective band mass of the electron has the same value in the two materials, the Hamiltonian of the electron can be written as

$$H = -\frac{\hbar^2}{2m}\nabla^2 - \frac{e^2}{4\pi\varepsilon |\boldsymbol{r} - \boldsymbol{r}_h|} + V_e(\boldsymbol{r}) \quad (2)$$

where $\boldsymbol{r}$ and $\boldsymbol{r}_h$ are the coordinate of the electron and the hole, $V_e(\boldsymbol{r})$ is the quantum confinement potential of the electron described by

$$V_e(\boldsymbol{r}) = \begin{cases} 0, & \text{for } r < R_1 \\ -V, & \text{for } R_1 < r < R_2 \\ \infty, & \text{for } r > R_2 \end{cases} \quad (3)$$

We choose the trial wave function has the following form

$$\psi = \begin{cases} A e^{\alpha r} \dfrac{\sin k(R_2 - r)}{r}, & \text{for } R_1 < r < R_2 \\ B e^{\pm \beta r}, & \text{for } 0 < r < R_1 \end{cases} \quad (4)$$

with

$$k = \sqrt{2mE/\hbar^2} \quad (5)$$

$$\beta = \sqrt{2m(V - E)/\hbar^2} \quad (6)$$

In Eq. (4), the sign "±" take "+" when the derivation of the wave function $\dfrac{\sin k(R_2 - r)}{r}$ at $r = R_1$ is positive, and "−" otherwise. A and B are normalization constants and α is the variational parameters, which will be determined by minimizing the expectation values of the Hamiltonian (2).

From the continuity condition of ψ and $\partial\psi/\partial r$ at $r = R_1$, we obtain a secular equation

$$\begin{vmatrix} \dfrac{\sin k\Delta R}{R_1} & 1 \\ \alpha \dfrac{\sin k\Delta R}{R_1} - k \dfrac{\cos k\Delta R}{R_1} - \dfrac{\sin k\Delta R}{R_1^2} & \pm\beta \end{vmatrix} = 0 \quad (7)$$

Using Eq. (7) one can obtain the energy E, and the wave function ψ, respectively.

3 Numerical Results and Discussion

The calculations for the values of the wave functions are performed numerically. The results are displayed in the atomic units (the effective Rydberg radius $R_y^* = 1$ and the effective Bohr radius $a_B = 1$) and shown in Fig. 1 and Fig. 2.

In Fig. 1 we plot the values of the wave functions as a function of coordinate r for $R_1 = 1$ and $\Delta R = 1$. The curves 1, 2, 3 and 4 are corresponding

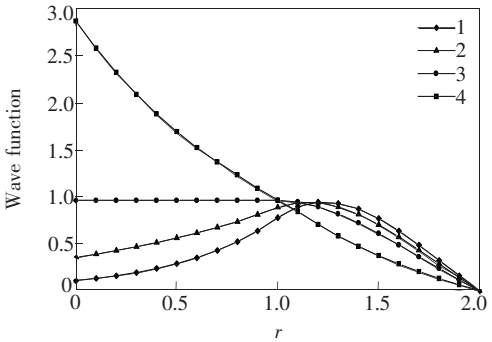

Fig. 1　The values of the wave functions as a function of coordinate r for $R_1 = 1$ and $\Delta R = 1$

to $V = 10$, $V = 6$, $V = 4.1$ and $V = 2$, respectively. It can be seen that there exists a critical potential $V_c = 4.1$. If $V > V_c$ the value of ψ has a peak at a distance from the origin. The exciton is a typical type-Ⅱ exciton. When $V < V_c$, the value of ψ has a maximum at the origin, that is, the center of electron charge and the center of hole charge coincide at the origin, the exciton must be considered as the type-Ⅰ exciton.

Fig. 2 plots the values of wave functions as a function of coordinate r for $V = 10$. The curves 1, 2, 3 and 4 describe the values of wave functions corresponding to the ratio $\gamma = \Delta R/R_1$ is 1.0, 0.8, 0.6 and 0.4, respectively. It can be found that there exists a critical radio $\gamma = 0.6$, above which the value of ψ has a maximum at a distance from the origin,

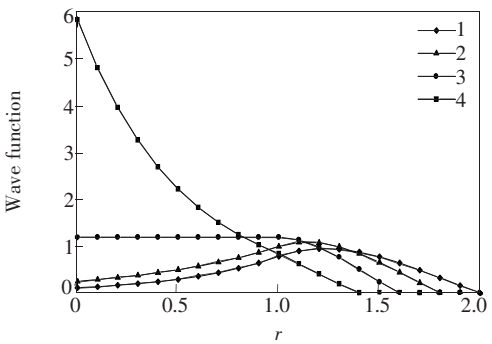

Fig. 2　The values of the wave functions as a function of coordinate r for $V = 10$

and the exciton is the type-Ⅱ one. When the radio decreases cross this critical value, the exciton transforms into a type-Ⅰ exciton.

Based on the above discussion, we can conclude that, for a given potential, with the shell width decreasing a type-Ⅱ exciton can transform into a type-Ⅰ exciton. For a given ratio of the shell width to core radius, with the potential decreasing a type-Ⅱ exciton may become a type-Ⅰ exciton. To clearly see the relationship of the critical potential with the ratio γ, we also calculate the critical potential as functions of the ratio of shell width to core radius, as listed in Table 1.

Table 1　The relationship of the critical potential V_c with the ratio γ of the shell width to core radius

γ	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1.0
V_c	43.45	19.57	12.27	9.00	8.45	6.39	5.03	4.18	4.10

4　Conclusion

In summery, we studied theoretically the excitons in the spherical semiconductor core-shell quantum dots embedded in an insulator matrix in a simplified model within the framework of effective mass approximation by using the variational approach. The calculations for the values of the wave functions as functions of coordinate and the critical potential as functions of the radio of shell width to core radius were performed numerically, respectively. It was found that for a given ratio of the shell width to core radius, there exists a critical potential, when the potential decreases cross this critical value the exciton transforms from a type-Ⅱ exciton into a type-Ⅰ exciton. For a given potential, there exists a critical ratio of shell width to core radius, when this radio decreases cross this critical value the exciton transforms from a type-Ⅱ exciton into a type-Ⅰ exciton.

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球型核壳量子点结构中二类激子向一类激子的转化

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摘要: 在有效质量近似下,采用变分方法研究了球型核壳量子点中激子的性质。数值计算了电子波函数随位置的变化关系以及临界势随球壳宽度和核半径的比值的变化关系。结果表明,当球壳宽度和核半径的比值为给定值时,存在一临界势,当局域势减小到临界势时,激子从二类激子转化为一类激子;当局域势为给定值时,存在一临界比值,当球壳宽度和核半径的比值减小到临界比值时,激子亦从二类激子转化为一类激子。该结果对研究球型核壳量子点中的电子结构有参考价值。

关 键 词: 激子; 量子点; 变分法

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