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Negative-donor centers and absorption spectra of quantum dots

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Abstract

Negative-donor centers in quantum dots with parabolic confinement potential have been studied for the case with the presence of a perpendicular magnetic field. Calculations are carried out by using the method of numerical diagonalization of the Hamiltonian matrix within the effective-mass approximation. The ground-state electron structures and angular momentum transitions are investigated. The binding energies of the ground and some bound excited states are obtained as a function of the applied magnetic field strength. As the magnetic field strength B is increased, there may appear more bound states. On the basis of the computed energies and wavefunctions, the linear optical absorption coefficients have been examined, for between the ^{1}P and ^{1}S states. The results are presented as a function of the incident photon energy for the different values of the confinement strength and the magnetic field strength. It is found that the optical properties of the negative-donor centers in quantum dots are strongly affected by the confinement strength and the magnetic field strength.

1. Introduction

A neutral shallow donor impurity confined in low-dimensional semiconductors can readily bind a second electron to form a negative-donor center (D^-), stable in a strong magnetic field up to room temperature [1]. A negative-donor center is analogous to the H^- ion, which offers an interesting example of a few-particle system where the electron–electron correlation plays a decisive role in trapping and keeping of a second electron [2]. Since the existence of D^- centers in center-doped GaAs/$Al_xGa_{1-x}As$ multiple quantum wells (QWs) was first reported by Huant *et al* [1] in 1990, many experimental [3–5] and theoretical [6–22] investigations for D^- centers in QWs, and quantum dots (QDs) with and without magnetic fields have been carried out. Much of this work has concentrated on GaAs/GaAlAs structures; particularly on isolated nanostructures subjected to an external magnetic field directed perpendicular to the heteroplanes. The reason for this is that the magnetic field significantly increases the stability of D^- centers.

From the point of view of quantum confinement, engineering the electronic structure of materials by means of shape and size control offers the possibility of tailoring the energy spectrum to produce desirable optical transitions. These features are useful for the development of optoelectronic devices with tunable emission or transmission properties and ultra-narrow spectral linewidths. Hence, optical properties of QDs have been investigated both experimentally and theoretically by many authors [23–28]. In the optical transition of quantum confined few-particle systems, the analysis of the negative-donor center states is unavoidable because the confinement of quasiparticles in such structure leads to the enhancement of the oscillator strength of electron-donor excitations. Meanwhile, the dependence of the optical transition energy on the confinement strength (or dot size) allows the tunability of the resonance frequency. Very recently, Sahin investigated the linear optical properties of a spherical QD containing one and two electrons with a hydrogenic impurity [29].

Since QDs are created mainly through producing a lateral confinement restricting the motion of the electrons, which are initially confined in a very narrow QW, they usually have the shape of flat disks, with transverse dimensions considerably exceeding their thickness. The energy of single-electron excitations across the disk exceeds other characteristic energies in the system, and the confined electrons can be considered as two-dimensional. In most studies, a harmonic oscillator potential were used to describe the lateral confinement of electrons. In the present work, we will focus on studying the electronic properties and optical absorption spectra of the negative-donor centers in QDs with a parabolic lateral confining potential in magnetic fields by using the method of numerical diagonalization of the Hamiltonian.

2. Theory

In the effective-mass approximation, the Hamiltonian for the negative-donor center in a parabolic QD when the magnetic field is applied perpendicular to the x–y plane is given by

$$H = \sum_{i=1,2}\left[\frac{1}{2m_e^*}(\vec{p}_i + \frac{e}{c}\vec{A}_i)^2 + \frac{1}{2}m_e^*\omega_0^2 r_i^2\right] + V_c - g^*\mu_B BS, \quad (1)$$

with

$$V_c = -\frac{e^2}{\epsilon}\sum_{i=1,2}\frac{1}{r_i} + \frac{e^2}{\epsilon r_{12}}, \quad (2)$$

where $\vec{r}_i$ ($\vec{p}_i$) is the position vector (the momentum vector) of the ith electron originating from the center of the dot; m_e^* is the effective mass of an electron; $r_{12} = |\vec{r}_1 - \vec{r}_2|$ is the electron–electron separation; ω_0 is the strength of the confinement, ϵ is the effective dielectric constant, g^* is the effective Landé factor, μ_B is the Bohr magneton and S is the total spin of two electrons. With the symmetric gauge for magnetic field $\vec{A} = (B/2)(-y, x, 0)$, the Hamiltonian then reads

$$H = \sum_{i=1,2}\left(\frac{p_i^2}{2m_e^*} + \frac{1}{2}m_e^*\omega^2 r_i^2\right) + \frac{1}{2}\omega_c L + V_c - g^*\mu_B BS, \quad (3)$$

where $\omega^2 = \omega_0^2 + \omega_c^2/4$, $\omega_c = eB/cm_e^*$ is the cyclotron frequency, and L is the total angular momentum.

Introducing the coordinates

$$\vec{\xi}_1 = \vec{r}_1 - \vec{r}_2, \qquad \vec{\xi}_2 = (\vec{r}_1 + \vec{r}_2)/2, \quad (4)$$

then equation (3) can be rewritten as

$$H = H_0 + V_c, \quad (5)$$

with

$$H_0 = \frac{p_{\xi_1}^2}{2\mu} + \frac{1}{2}\mu\omega^2\xi_1^2 + \frac{p_{\xi_2}^2}{2M} + \frac{1}{2}M\omega^2\xi_2^2 - \tfrac{1}{2}\omega_c L - g^*\mu_B BS, \quad (6)$$

where $\mu = m_e^*/2$ is the reduced mass associated with $\vec{\xi}_1$, and $M = 2m_e^*$ is the total mass.

The Hamiltonian has cylindrical symmetry with respect to the QD axis, i.e., z-axis, which implies that the total orbital angular momentum L is a conserved quantity, i.e., a good quantum number. The total spin of two electrons, i.e., S, is also a conserved quantity. To obtain the eigenfunction and the eigenenergy associated with the D^- centers in QDs under magnetic fields, we diagonalize the Hamiltonian. As we know, the two electrons obey Fermi–Dirac statistics, which means that the electronic part of the total wavefunction must be antisymmetric, i.e., when $S = 0$ the spatial part of the electronic wavefunction must be symmetric and when $S = 1$ the spatial part of the electronic wavefunction must be antisymmetric. Thus, S can be used as a quantum number which indicates the parity of the state. Hence, the eigenstates of the D^- centers in QDs can be indicated by a series of energy levels with quantum numbers (L, S).

In order to diagonalize the Hamiltonian, a set of the harmonic oscillation product states were chosen as basis functions so that the eigenstates can be expanded in terms of them. It is obvious that all the associated matrix elements have analytical forms; thus the Hamiltonian matrix can be calculated very accurately and very fast. Let $\varphi_{n\ell}^{\omega'}(\vec{\xi})$ be a two-dimensional harmonic oscillator state with an eigenenergy $(2n+|\ell|+1)\hbar\omega'$, where ω' is an adjustable parameter and in general not equal to ω. Let

$$\phi_k(\vec{\xi}_1, \vec{\xi}_2) = [\varphi_{n_1\ell_1}^{\omega'}(\vec{\xi}_1)\varphi_{n_2\ell_2}^{\omega'}(\vec{\xi}_2)]_L\chi_S \quad (7)$$

where the orbital angular momenta ℓ_1 and ℓ_2 are coupled to L, and k denotes the set of quantum numbers n_1, ℓ_1, n_2, and ℓ_2. $\chi_S = [\eta(1)\eta(2)]_S$, where $\eta(i)$ is the spin state of the ith electron and the spins of two electrons are coupled to S. On the basis of the harmonic oscillator states a set of basis functions totally antisymmetric with respect to particle interchanges has been chosen as

$$\Psi_k = (1 - P_{12})\phi_k \quad (8)$$

where P_{12} denotes an interchange of the indices 1 and 2.

Let $N = 2(n_1 + n_2) + |\ell_1| + |\ell_2|$. Let $\{\Phi_{LS}\}$ denote the set of basis functions with the total orbital angular momentum L and the spin angular momentum S of electrons 1 and 2 including all the Ψ_k having their N smaller or equal to an upper limit N_{max}. It is obvious that the total number of basis functions of the set is determined by N_{max}. In what follows N_{max} is in general given as 40. With $\{\Phi_{LS}\}$, the Hamiltonian matrix can be calculated. The accuracy of the solutions depends on how large the model space is. Since we are interested only in the low-lying states and in the qualitative aspects, the model space adopted is neither very large, to facilitate numerical calculation, nor very small, to ensure qualitative accuracy. This is achieved by extending the dimension of the model space step by step; in each step the new results are compared with previous results from a smaller space, until satisfactory convergence is achieved. In this work, the dimension of the model space is constrained by $0 \leqslant N \leqslant 40$. If N is increased by 2, the ratio of the difference in energy is less than 0.1%. After the diagonalization we obtain the eigenvalues and eigenvectors. Evidently, the eigenvalues depend on the adjustable parameter ω'. In practical calculation, ω' serves as a variational parameter to minimize the low-lying state energies.

We define the binding energy of the νth quantum state of D^- centers as

$$E_B(D^-) = E(D^0) + E_0 - E(D^-) \quad (9)$$

where $E(D^-)$ is the energy of the νth state of the D^- centers in the QDs, E_0 and $E(D^0)$ are, respectively, the lowest energy levels of an electron in QDs, without and with the Coulomb potential. The binding energy defined by (8) possesses the following physical interpretation: this is the minimum energy, which is required to liberate one electron from the bound state of D^- centers. After this dissociation process, the second electron is bound in the ground state of the D^0 center. The condition of stability against dissociation into a neutral donor and a free electron reads $E_B(D^-) \geqslant 0$.

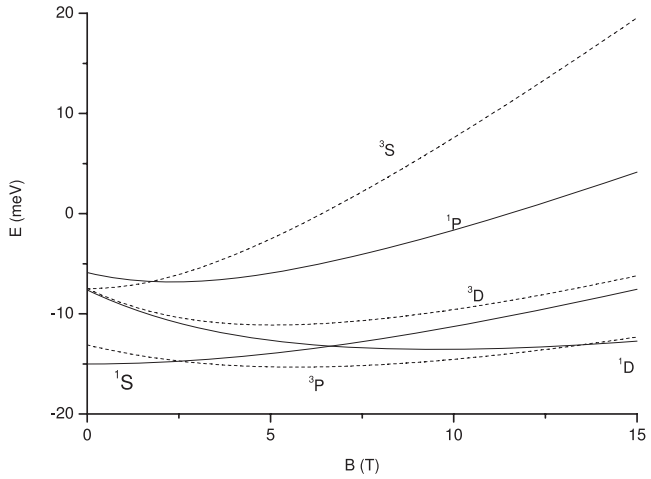

Figure 1. The lowest-lying energy levels for a negative-donor center in a parabolic QD with $\hbar\omega_0 = 3.0$ meV as a function of the magnetic field strength B. The solid and dashed lines represent, respectively, the singlet and triplet states.

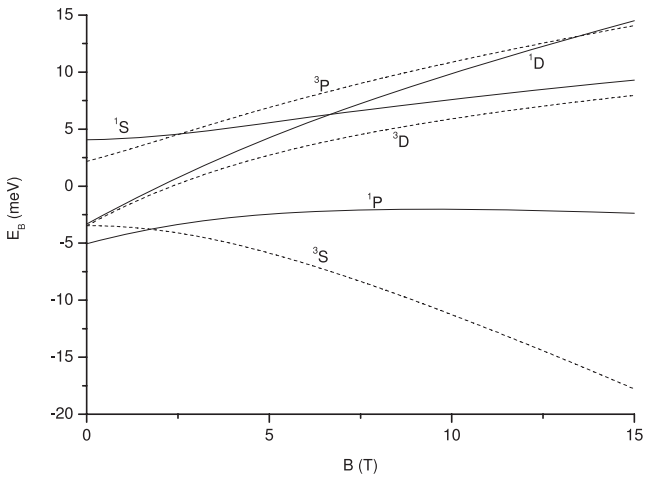

Figure 2. Binding energy of the low-lying energy states of a negative-donor center in a parabolic QD with $\hbar\omega_0 = 3.0$ meV as a function of the magnetic field strength B.

The optical absorption calculation is based on Fermi's golden rule, for which the optical absorption coefficient is given by [29]

$$\alpha = \frac{4\pi\beta_{\rm FS}\sigma_s}{n_{\rm r}e^2}h\upsilon|M_{\rm fi}|^2\delta(E_{\rm f} - E_{\rm i} - h\upsilon), \qquad (10)$$

where $n_{\rm r}$ is the refractive index of the semiconductor and it is taken as 3.2. e is the electronic charge of an electron, σ_s is the electron density in the QD, $\beta_{\rm FS} = e^2/(4\pi\epsilon_0\hbar c)$ is the fine structure constant, and $h\upsilon$ is the photon energy. $E_{\rm f}$ and $E_{\rm i}$ are the final-state and initial-state energy eigenvalues, respectively. $M_{\rm fi} = e\langle f|\vec{r}|i\rangle$ is the electric dipole moment of the transition from the i state to the f state. The δ function in equation (9) is replaced by a narrow Lorentzian by means of

$$\delta(E_{\rm f} - E_{\rm i} - h\upsilon) = \frac{\hbar\Gamma_{\rm fi}}{\pi\{[h\upsilon - (E_{\rm f} - E_{\rm i})]^2 + (\hbar\Gamma_{\rm fi})^2\}}. \qquad (11)$$

Here Γ is the phenomenological operator. The nondiagonal matrix element $\Gamma_{\rm fi}(f \neq i)$ of operator Γ, which is called the relaxation rate of the fth state and the ith state, is the inverse of the relaxation time $T_{\rm fi}$ for the states $|f\rangle$ and $|i\rangle$, namely $\Gamma_{\rm fi} = 1/T_{\rm fi}$, and $T_{\rm fi}$ is taken as 0.14 ps.

3. Numerical results and discussion

Our numerical computation is carried out for one of the typical semiconducting materials, GaAs, as an example with the material parameters as follows: $\epsilon = 12.4$, $g^* = -0.44$, and $m_{\rm e}^* = 0.067m_{\rm e}$, where $m_{\rm e}$ is the mass of a free electron. The material parameters used in the calculations correspond to the QDs of Maksym *et al* [30]. For simplicity, we restrict our study to the $L \leqslant 2$ states, i.e., the ground-state (^{1}S) and the following low excited states: ^{3}S, ^{1}P, ^{3}P, ^{1}D, and ^{3}D.

Figure 1 shows the lowest-lying energy levels for a negative-donor center as a function of the magnetic field strength B. The confining energy $\hbar\omega_0$ is set to be 3.0 meV [31], which corresponds to a QD with characteristic radius $R = (\hbar/m_{\rm e}^*\omega_0)^{1/2} = 19.47$ nm. In what follows the energy unit is meV and the length unit is nm. The solid and dashed lines represent the results for the singlet and triplet states, respectively. We can see that an important aspect of the D^- centers under the magnetic field is that the ground-state transition can appear as B increases. From figure 1, we find that the first ground-state transition of the D^- center occurs at $B = 2.6$ T (from the ^{1}S state to ^{3}P state, i.e., the spin of the ground-state changes from $S = 0$ to 1) and the second one occurs at $B = 13.5$ T (from ^{3}P to ^{1}D, the triplet $\rightarrow$ singlet-state transition). It is the competition between the single-particle energy and the interacting energy that finally determines the energy. We know that the slope of the rising curve depends on L. A smaller L would lead to a larger slope because the negative term $-\frac{1}{2}\omega_{\rm c}L$ is weaker. Therefore, when B increases, the curve with a small L might cross the curve with a larger L because the former is rising faster. Obviously, the crossing would lead to a transition of L of the true ground state from one to another. However, the transition is strictly limited to between two magic numbers of L [32]. The origin of the magic numbers is in the quantum constraint arising from the Pauli principle [32].

Figure 2 shows the variation of the binding energy of the low-lying energy states of a negative-donor center in a parabolic QD with $\hbar\omega_0 = 3.0$ meV as a function of the magnetic field strength B. From this figure we find the following results: (1) the binding energies $E_{\rm B}(D^-)$ increase as the magnetic field strength B increases except for the ^{3}S state. A qualitative explanation is as follows: the Hamiltonian H contains positive terms (the first four terms in H, i.e., the single-particle energy) and negative terms (the Coulomb attractive energy $V_{\rm c}$, $-\frac{1}{2}\omega_{\rm c}L$ and $-g^*\mu_{\rm B}BS$), which compete. Obviously, when the magnetic field strength B increases, the confinement strength will increase. We note that as the confinement strength is increased (i.e., the confined potential radius is reduced), the Coulomb attractive energy increases, and the single-particle energy also increases. On the other

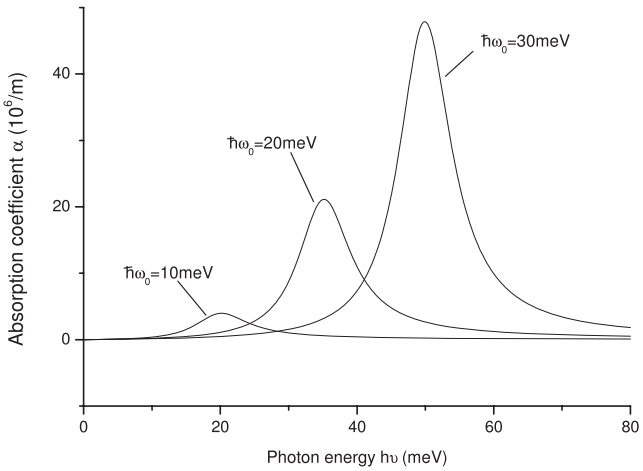

Figure 3. The linear optical absorption coefficient of a negative-donor QD as a function of the incident photon energy $h\upsilon$ for three different values of the confinement strength $\hbar\omega_0$ in the absence of magnetic field.

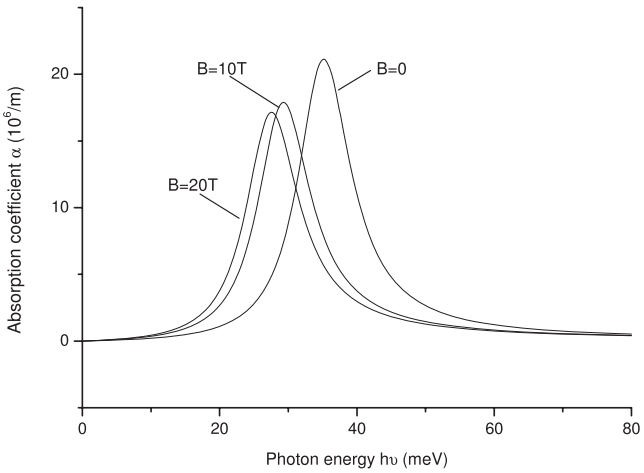

Figure 4. The linear optical absorption coefficient of the negative-donor QD with $\hbar\omega_0 = 20.0$ meV as a function of the incident photon energy $h\upsilon$ for three different values of the magnetic field strength.

hand, the Hamilton contains two negative terms $-\frac{1}{2}\omega_c L$ and $-g^*\mu_B BS$ which are proportional to B and therefore would cause a linear decrease in the energy. The binding energy of the low-lying states in a QD is determined by a competition of these results. It is obvious that, for the ground state, these two terms are zero because $L = 0$ and $S = 0$; hence, the binding energy of the ground state slowly increases with increasing B. (2) When the magnetic field strength is less than $B = 2.1$ T, the D^- center confined in a QD possesses two bound states, i.e., the ^1S and ^3P states, and for $2.1 \leqslant B < 2.4$ T, there exist three bound states in the range of our study. When $B \geqslant 2.4$ T, as the magnetic field strength B is increased further, the fourth bound state (^3D) appears. Hence, as B is increased further, there are more bound excited states appearing. However, the ^3S and ^1P states are always unbound. This is because the single-particle energies of the ^3S and ^1P states are always larger than those of negative terms with increasing B. (3) We find that the binding energies of the states with $L+S$ even, i.e., the magic L states, increase more quickly than those of the adjacent states with $L + S$ odd, non-magic L states with increasing B. This feature is a consequence of the mechanics symmetry. It has been discussed in previous publications and this will not be repeated here [32–34].

Furthermore, in order to study the optical properties of a negative-donor center QD, a numerical calculation has been performed for the linear optical absorption coefficient α as a function of the incident photon energy $h\upsilon$ in the range from 0 to 80 meV in the absence of magnetic field. We restrict our study to the transition of the ^1S state to the ^1P state. In figure 3 the confining energies $\hbar\omega_0$ are set to be 10.0, 20.0, and 30.0 meV. From this figure we can find that the quantum effect of the QD is obvious. It is readily seen that the linear optical absorption coefficient of small-radius QDs (the stronger confinement strength) is much stronger than that of large-radius QDs. We also find that the stronger the confinement strength $\hbar\omega_0$ is, the sharper the linear absorption peak will be and the bigger the absorption peak intensity will be. One physical origin of these results is that the optical absorption spectrum depends on the electron density in QDs, i.e., depends on the QD size. Another reason is that the electronic dipolar transition matrix element increases with the confinement strength of the parabolic potential. Thus, these lead to the optical absorption coefficient increase. On the other hand, we also find that the linear absorption peak values appear at $h\upsilon = 20.2$, 35.2, and 50.0 meV, respectively. Hence, as the confinement strength $\hbar\omega_0$ increases, the absorption peak will move to the right, which shows a confinement-strength-induced blue shift of the resonance in QDs. The physical origin is that, with the dot radius decreasing, the Coulomb binding energy, between the electron and the donor, is increased, leading to the increase of the energy difference between the ^1S state and the ^1P state. The absorption spectrum peak values are decreasing with increasing dot radius, because the energy levels come close to each other [29, 35].

Figure 4 shows the linear absorption spectrum, for between the ^1P state and the ^1S state of the negative-donor QD with $\hbar\omega_0 = 20.0$ meV, as a function of the incident photon energy $h\upsilon$ in the range from 0 to 80 meV for three different magnetic field strengths, i.e., $B = 0$, 10 and 20 T, respectively. The effect of the magnetic field also seems clear. It can been seen that the linear absorption is smaller for stronger magnetic field and the absorption peaks shift to lower energies (red shift) with increasing B. This is because the energy difference between the ^1S and ^1P states will decrease with increasing B. Thus, it leads to the optical absorption coefficient decrease.

In conclusion, we have investigated the feature of the low-lying states for $L \leqslant 2$ of the negative-donor centers confined by a parabolic QD as a function of magnetic field strength. The ground-state transition of the D^- centers occurring as a function of B was found. For our D^- center QDs, it was found that there are two bound states in weak magnetic field and there appear to be more bound states with increasing B. The linear optical absorption coefficient has been examined on the basis of the computed energies and wavefunctions. The

results are presented as a function of the incident photon energy for the different values of the confinement strength and the magnetic field strength. It is found that the optical properties of the negative-donor center in QDs are strongly affected by the confinement strength and the magnetic field strength. This may be important in the quantitative understanding the optical and magnetic properties of the negative-donor center QDs. These features make the parabolic QDs very promising candidates for optical material and device use.

Acknowledgment

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