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A study of two confined electrons using the Woods–Saxon potential

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Abstract

In this paper, we studied two electrons confined in a quantum dot with the Woods–Saxon potential by using the method of numerical diagonalization of the Hamiltonian matrix within the effective-mass approximation. The great advantage of our methodology is that it enables confinement regimes by varying two parameters in the model potential. A ground-state behavior (singlet → triplet state transitions) as a function of the strength of a magnetic field has been investigated. We found that the confinement barrier size and the barrier inclination of a Woods–Saxon potential are important for the singlet–triplet oscillation of a two-electron quantum dot. Based on the computed energies and wavefunctions, the linear and nonlinear optical absorption coefficients have been examined between the ^{1}S state ($L = 0$) and the ^{1}P state ($L = 1$). The results are presented as a function of the incident photon energy for the different values of the barrier size and height. It is found that the optical properties of the two-electron system in a quantum dot are strongly affected by the barrier height and size.

1. Introduction

The progress in semiconductor technology has opened up a rich field of studies focused on the fundamental electron–electron interactions, quantum effects and optical properties in quantum dots (QDs) [1, 2]. The experimental study of semiconductor QDs is expanding rapidly [3–6] and electron–electron interaction and correlation effects are shown to be of great importance [7–9] in such systems. In the meantime, a large number of theoretical investigations [10–13] of electronic structures and related magnetic and optical properties in QDs have been performed to explain the experimental observations. Based on a numerical solution of the Coulomb interaction between electrons, a complex ground-state behavior (singlet → triplet state transitions) as a function of a magnetic field has been predicted [8, 14]. Remarkably, these ground-state transitions for $N = 2$ have been observed experimentally [5].

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 [15–19]. In the optical transition of quantum confined few-electron systems, the analysis of the two-electron states is inevitable because the confinement of quasiparticles in such a structure leads to the enhancement of the oscillator strength for electron–electron excitations. Meanwhile, the dependence of the optical transition energy on the 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 [20]. However, to our knowledge, there are only a few studies on the nonlinear optical property of few-electron QDs [21, 22].

The influence of spatial confinement on the properties of a QD system is one of the most interesting properties to be investigated in the study of confined systems. However, most of the previous studies are concerned with two-electron systems in a parabolic potential QD which possesses infinite depth and range or a box potential QD which is not completely solved because of the boxing effects. It is inappropriate for a description of the experimentally measured charging of the QD by the finite number of excess electrons [23]. Some experimental results suggest that the real confining potential is nonparabolic and possesses a well-like shape. More recently, Boyacioglu and co-workers investigated the problem of two electrons in a three-dimensional QD with Gaussian confinement by a variational method [24].

The behavior of electrons is very important in understanding few-electron QDs. Thus, a good description of the motions of these electrons is very useful in studying few-electron QDs. Since QDs are created mainly through producing a lateral confinement restricting the motion of the electrons, which are initially confined in a very narrow quantum well, they usually have the shape of flat discs, with transverse dimensions considerably exceeding their thickness. The energy of single-electron excitations across the disc exceeds other characteristic energies in the system, and the confined electrons can be considered as two-dimensional. The lateral potential of a QD differs significantly from the Coulomb potential binding electrons in an atom—the latter has a peculiarity in the center. Depending on the method used to create the dot, the lateral potential can be approximated by a model potential. The potential of an etched dot with a considerable radius is fairly close to a rectangular well with rounded edges. When a dot is small (i.e. when its radius is comparable to the characteristic length of the variation of the lateral potential near the edge), a good approximation offers simple smooth potentials, such as a Wood–Saxon potential $V_{\rm WS}(R_0, r) = V_0/(1 + \exp[(R_0 - r)/\gamma])$ or a Gaussian well $V(R_0, r) = -V_0 \exp(-r^2/R_0^2)$.

Woods and Saxon introduced a potential to study elastic scattering of 20 MeV protons by a heavy nuclei half a century ago [25]. The Woods–Saxon potential is a reasonable potential for nuclear shell models and hence attracts a lot of attention in nuclear physics and it is used to represent the distribution of nuclear densities [26]. The Woods–Saxon potential possesses finite depth and range. The Woods–Saxon potential is a continuous function that has two adjustable parameters. These parameters allow one to treat different systems and to simulate smooth model potentials and infinite or finite constant barriers with the same efficiency. Obviously, the Woods–Saxon potential becomes a rectangular potential well when $\gamma \to 0$. Recently, the Woods–Saxon potential has also been used in the study of an impurity in the center of a QD by variational methods [27]. Although the Woods–Saxon potential is not a new model potential, its use to simulate the confinement barrier appears to be novel. Hence, in the present work, we will focus on studying the transition of the ground state, and the linear and nonlinear optical properties of the two-electron systems in a two-dimensional QD with Woods–Saxon confinement. By using the matrix diagonalization method, we will find the energy spectrum of the low-lying states, and the linear and nonlinear third-order optical absorption coefficients of the two-electron QD. Furthermore, we will investigate the quantum size and the incident optical intensity for the optical properties of a two-electron QD.

2. Singlet–triplet oscillation

The Woods–Saxon potential is given by

$$V_{\rm WS}(R_0, r) = \frac{V_0}{1 + \exp[(R_0 - r)/\gamma]}, \tag{1}$$

where R_0 denotes the confinement barrier position, i.e. the average radius of the QD, V_0 is the height of the potential barrier, and $V_0 > 0$, and γ control the confinement barrier slope. These parameters guarantee the flexibility of the present methodology treating the two-electron systems in QDs. We emphasize that the barrier inclination increases as the parameter decreases.

The system we study is two interacting electrons moving in the (x, y) plane, confined by a Woods–Saxon potential and subject to a perpendicular magnetic field B which is assumed to be along the z direction. Therefore, the Hamiltonian of the system can be given by

$$H = \sum_{i=1,2}\left[\frac{1}{2m_{\rm e}}\left(\vec{p}_i - \frac{e}{c}\vec{A}\right)^2 + V_{\rm WS}(R_0, r_i)\right] + \frac{e^2}{\epsilon r_{12}} - g^*\mu_{\rm B}BS_z, \tag{2}$$

where $\vec{r}_i$ ($\vec{p}_i$) is the position vector (the momentum vector) of each electron originating from the center of the dot, $m_{\rm e}$ is the effective mass of an electron, $r_{12} = |\vec{r}_1 - \vec{r}_2|$ is the electron–electron separation, e is the electronic charge of an electron and $\epsilon = 4\pi\epsilon_0\epsilon_{\rm r}$ is the effective dielectric constant of the QD. g^* is the effective Landé factor, $\mu_{\rm B}$ is the Bohr magneton and S_z is the z component of the total spin. With symmetric gauge for the magnetic field $\vec{A} = B(-y, x, 0)/2$, the Hamiltonian is then

$$H = \sum_{i=1,2}\left[\frac{p_i^2}{2m_{\rm e}} + \frac{1}{2}m_{\rm e}\omega_{\rm c}^2 r_i^2 + V_{\rm WS}(R_0, r_i)\right] - \frac{1}{2}\omega_{\rm c}L_z + \frac{e^2}{\epsilon r_{12}} - g^*\mu_{\rm B}BS_z, \tag{3}$$

where $\omega_{\rm c} = eB/cm_{\rm e}$ is the cyclotron frequency and L_z is the total orbital angular momentum along the z direction.

The Hamiltonian has cylindrical symmetry 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, S, also is a conserved quantity. Hence, the eigenstates of the two electrons in two-dimensional cylindrical symmetry QDs can be classified according to the total orbital angular momentum and the total spin momentum of the electrons, i.e. after solving our Hamiltonian, a series of energy levels which we indicate by the quantum numbers (L, S). Hence, the states of two electrons will be denoted by ^{2S+1}L. To obtain the eigenfunction and the eigenenergy associated with the two electrons in a two-dimensional Woods–Saxon confining potential QD, we diagonalized H. The exact diagonalization method consists in spanning the total Hamiltonian for a given basis and extracting the lowest eigenvalues (energies) of the matrix generated. The better the basis describes the Hamiltonian, the faster will be the convergence. The most common basis chosen is the one that describes the Hamiltonian at zero order.

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. To obtain the eigenenergies and eigenstates, H is diagonalized in the model space spanned by translational

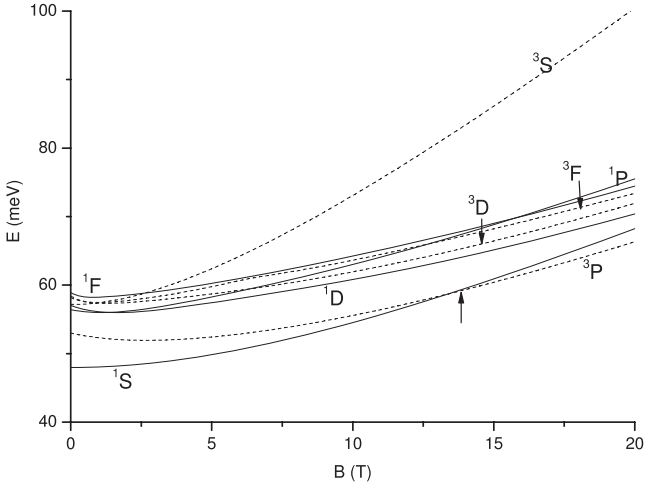

Figure 1. The energies of the low-lying states with $L \leqslant 3$ of a two-electron QD confined by the Woods–Saxon potential versus external magnetic field. The solid and dashed lines represent, respectively, the spin-singlet and spin-triplet states. The levels are labeled by quantum numbers ^{2S+1}L. Parameters are taken appropriately for GaAs, $V_0 = 30.0$ meV, $R_0 = 10.0$ nm and $\gamma = 3.0$.

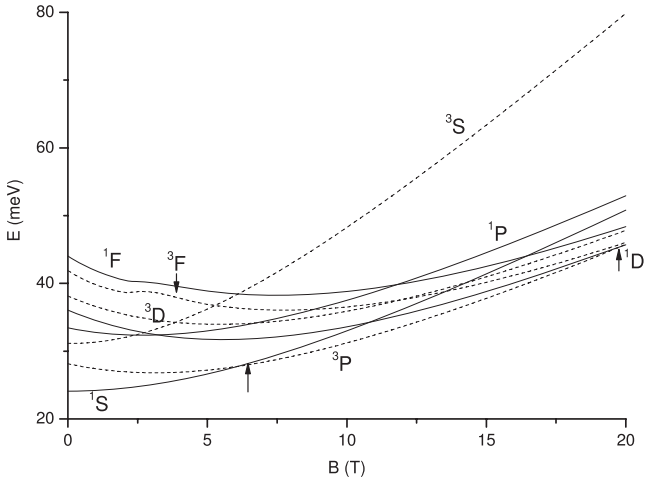

Figure 2. The same as figure 1, but for $R_0 = 20.0$ nm.

invariant harmonic product states:

$$\Phi^{LS}_{[K]} = \sum_{[K]} \tilde{A}\{[\varphi^{\omega}_{n_1\ell_1}(\vec{r}_1)\varphi^{\omega}_{n_2\ell_2}(\vec{r}_2)]_L \chi_S\}, \tag{4}$$

where $\varphi^{\omega}_{n\ell}$ is a two-dimensional harmonic oscillator wavefunction with a frequency ω (ω is an adjustable parameter) and an energy $(2n + |\ell| + 1)\hbar\omega$, and $[K]$ denotes the set quantum numbers $(n_1, \ell_1, n_2, \ell_2)$ in brief, $\chi_S = [\eta(1)\eta(2)]_S$. In a practice calculation, ω serves as a variational parameter to minimize the eigenvalues, while $\tilde{A}$ is a antisymmetrizer. The calculation of matrix elements is realized by using two-dimensional Talmi–Moshinsky coefficients [28].

Since the whole set of eigenstates of the harmonic product basis forms a complete basis in the Hilbert space, the procedure of increasing the number of linearly independent eigenstates is converging to the exact result. The limits are set only by the capacity of the computer to diagonalize $N \times N$ Hermitian matrices. On the other hand, 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 paper, the dimension of the model space is constrained by $0 \leqslant N = 2(n_1 + n_2) + |\ell_1| + |\ell_2| \leqslant 30$. 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 eigenstates. Evidently, the eigenvalues depend on the adjustable parameter ω.

Our numerical computation is carried out for GaAs QDs with the material parameters shown in the following: $\epsilon_r = 12.4$ and $m_e = 0.067m_0$, where m_0 is the single-electron bare mass and $V_0 = 30.0$ meV. In what follows the energy unit is meV and the length unit is nm. For simplicity, in this section, we restrict our study to $L \leqslant 3$ with the spin-singlet and the spin-triplet states of two electrons, which are denoted by ^{1}S, ^{3}S, ^{1}P, ^{3}P, ^{1}D, ^{3}D, ^{1}F and ^{3}F.

To see intuitively the feature of the ground state of a two-electron system confined in a QD with the Woods–Saxon confining potential, we set $R_0 = 10.0$ nm and $\gamma = 3.0$, and plotted in figure 1 the energy spectrum of the low-lying states as a function of the external magnetic field B. In figure 1 we note the singlet–triplet oscillation of the ground state with the increase of the external magnetic field. As we know, it is the competition between the single particle energy and the interacting energy that finally determines the total energy. The slope of the rising curve depends on L. A smaller L would lead to a larger slope because the negative term $\omega_c L_z/2$ is weaker. Therefore, when the magnetic field B increases, the curve with a small L crosses the curve with a larger L because the former is rising faster. Obviously, the crossing would lead to a transition of L and S of the ground state from one to another. In figure 1 we also note the singlet–triplet oscillation of the ground state. However, there occurs only one singlet–triplet transition of the ground state (i.e. from the ^{1}S state to the ^{3}P state) with increasing magnetic field strength. For a weak magnetic field, the ground state of the system is the spin-singlet $S = 0$. The ground-state spin is flipped to $S = 1$ at the crossing point of $B \simeq 13.4$ T as the strength of the magnetic field increases. In a two-electron parabolic QD, there are also singlet–triplet transitions of the ground state with increasing magnetic field strength. The two-electron parabolic QD has been studied in great detail by Merkt *et al* [29].

Obviously, the crossing points of the singlet–triplet oscillation depend on R_0 and γ. In order to show better the confinement barrier size effect, we take $R_0 = 20.0$ nm and $\gamma = 3.0$ and plotted in figure 2 the energy spectrum of the low-lying states of a two-electron system confined in a QD with the Woods–Saxon confining potential as a function of the external magnetic field B. We see the familiar singlet–triplet oscillations of the ground state and the crossing point shifts to

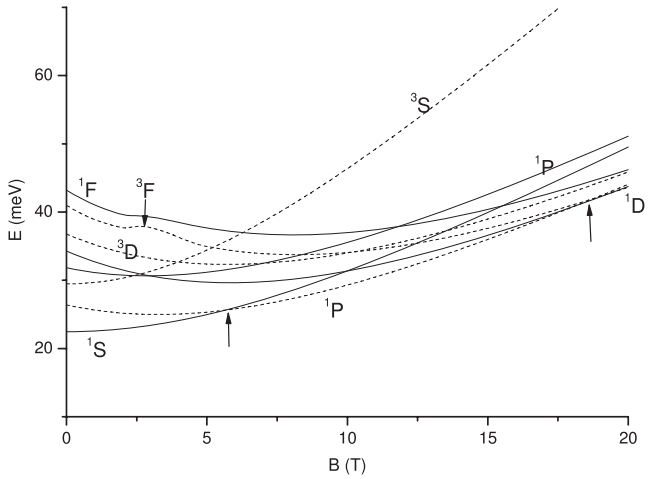

Figure 3. The same as figure 2, but for $\gamma = 2.0$.

the lower magnetic field strength, e.g. the first transition shifts to $B \simeq 6.2$ T. There occurs the second singlet–triplet transition of the ground state at $B \simeq 19.9$ T. This result can be interpreted as follows. When R_0 increases, the geometric confinement of electrons becomes weak. The lowest potential energy of QDs shifts to the larger radius and the energy of the higher orbit decreases. Hence, the singlet–triplet transition points shift to the lower magnetic fields. On the other hand, we find that the energies decrease obviously with increasing confinement barrier size. This is because, when the confinement barrier size increases, the confinement in QDs will decrease and the energies of the low-lying states will decrease.

On the other hand, in order to investigate the influence of the barrier inclination of a Woods–Saxon potential, i.e. a parameter γ, we take $\gamma = 2.0$ and plotted in figure 3 the energy spectrum of the low-lying states of a two-electron system confined in a QD with the Woods–Saxon confining potential as a function of the external magnetic field B. We find that the energy spectrum in figure 3 is qualitatively the same as that in figure 2. The difference is that the singlet–triplet transitions slightly shift to the lower magnetic field strengths, e.g. the first transition shifts to $B \simeq 5.7$ T and the second transition shifts to $B \simeq 18.0$ T. This is because the confinement energy decreases with decreasing γ. Hence, the confinement barrier size is more important than the barrier inclination for the singlet–triplet transition of the ground state.

3. Linear and nonlinear optics absorption coefficients

The optical absorption calculation is based on Fermi's golden rule, for which the total optical absorption coefficient is given by [30]

$$\alpha(\omega, I) = \alpha^{(1)}(\omega) + \alpha^{(3)}(\omega, I), \tag{5}$$

where

$$\alpha^{(1)}(\omega) = \frac{4\pi\beta_{\rm FS}\sigma_{\rm s}}{n_{\rm r}e^2} h\nu |M_{if}|^2 \delta(E_f - E_i - h\nu), \tag{6}$$

and

$$\alpha^{(3)}(\nu, I) = -\frac{32\pi^2\beta_{\rm FS}^2\sigma_{\rm s} I}{n_{\rm r}^2 e^4 \hbar\Gamma_{ff}} h\nu |M_{if}|^4 \delta^2(E_{fi} - h\nu) \times \left\{1 - \frac{|M_{ff} - M_{ii}|^2}{4|M_{fi}|^2} \times \frac{[(h\nu - E_{fi})^2 - (\hbar\Gamma_{fi})^2 + 2E_{fi}(E_{fi} - h\nu)]}{E_{fi}^2 + (\hbar\Gamma_{fi})^2}\right\}, \tag{7}$$

are the linear and the third-order nonlinear optical absorption coefficients, respectively. $n_{\rm r}$ is the refractive index of the semiconductor and it is taken as 3.2, e is the electronic charge of an electron and $\sigma_{\rm s}$ is the electron density in the QD. $\beta_{\rm FS} = e^2/(4\pi\epsilon_0\hbar c)$ is the fine structure constant. $h\nu$ denotes the photon energy. E_f and E_i are the final- and initial-state energy eigenvalues, respectively. $M_{fi} = 2q\langle f|\vec{R}|i\rangle$ is the electric dipole moment of the transition from the ith state to the fth state in the QD, where q is the electronic charge. Unlike a parabolic QD, the variables of a QD with Woods–Saxon potential is not independent. Hence, the electric dipole moment of the transition M_{fi} is not independent of the relative motion. This is not satisfied with the Kohn theorem [31]. On the other hand, the dipole operator is independent of the electron spin. The dipole-allowed optical transitions are always from the same spin states, but the angular momenta must differ by unity [32] Hence, we restrict our study to the transition of the ^{1}S state ($L = 0$) to the ^{1}P state ($L = 1$).

The δ function in equations (6) and (7) is replaced by a narrow Lorentzian by means of

$$\delta(E_f - E_i - h\nu) = \frac{\hbar\Gamma_{fi}}{\pi\{[h\nu - (E_f - E_i)]^2 + (\hbar\Gamma_{fi})^2\}}. \tag{8}$$

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

In order to study the optical properties of a two-electron QD, a numerical calculation has been performed for the linear absorption coefficient $\alpha^{(1)}$ as a function of the incident photon energy $h\nu$ in the range from 0 to 80 meV. In figure 4 the barrier height V_0 is set to be 112.23 meV, the barrier inclination γ is set to be 3.0 and the dot radii R_0 are set to be 10.0, 15.0 and 20.0 nm, respectively. From this figure we can find that the size effect of the QD is obvious. It is readily seen that the linear absorption coefficient of small-radius QDs is stronger than that of large-radius QDs. We also find that the smaller the dot radius R_0 is, the sharper the absorption peak will be and the bigger will be the absorption peak intensity. The physical origin of these results is that the linear absorption spectrum depends on the electron density in QDs, i.e. depends on the QD radius. Thus, this reason leads to the linear absorption coefficient increase. On the other hand, we find that the linear absorption peak values appear at $h\nu = 16.0$, 22.6 and 30.8 meV, respectively. Hence, as the dot radius decreases, the linear absorption peak position shifts to the high

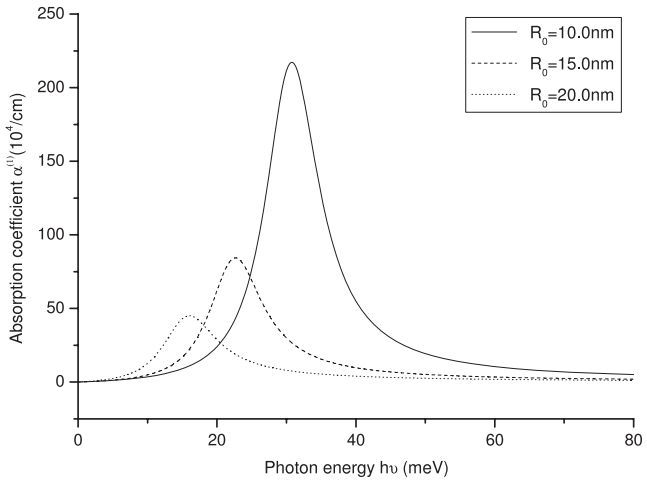

Figure 4. The linear absorption coefficient of a two-electron QD confined by the Woods–Saxon potential as a function of the incident photon energy $h\nu$ for three different values of the dot radius R_0. Parameters are taken appropriate for GaAs: $V_0 = 112.23$ meV and $\gamma = 3.0$.

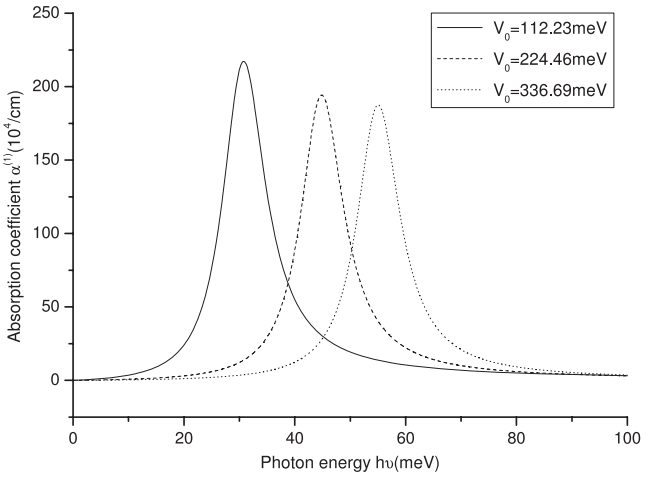

Figure 5. The linear optical absorption coefficient of a two-electron QD confined by the Woods–Saxon potential as a function of the incident photon energy $h\nu$ for three different values of the confinement barrier heights V_0. Parameters are taken appropriate for GaAs: $R_0 = 10.0$ nm and $\gamma = 3.0$.

photon energies, which shows a dot-radius-induced blueshift of the resonance in QDs. The physical origin is that, with decreasing dot radius, the Coulomb interaction energy in a QD is increased, leading to the increase of the energy difference between the ^{1}S and ^{1}P states. The linear absorption spectrum peak values are decreasing with increasing dot radius, because the energy levels come close to each other.

Figure 5 shows the linear absorption spectrum of the two electrons in a Woods–Saxon QD with $R_0 = 10$ nm as a function of the incident photon energy $h\nu$ in the range from 0 to 100 meV for three different barrier heights, i.e. $V_0 = 112.23$, 224.46 and 336.69 meV, respectively, corresponding to three Al concentrations [33]. The confinement effect of the barrier height (i.e. Al concentration) seems clear. It can been seen that the linear absorption peak positions shift to higher energies (blueshift) with increasing V_0. This is because the energy difference between the ^{1}S and ^{1}P states will increase with increasing V_0. On the other hand, figure 5 also shows that the linear absorption peak intensity decreases with increasing V_0, i.e. the smaller the barrier height is, the sharper the linear absorption peak will be and the bigger will be the absorption peak intensity. This is because the electron dipolar transition matrix element decreases with increasing V_0.

In figure 6, in order to see clearly the influence of nonlinear optical absorption, we set $V_0 = 112.23$ meV, $R_0 = 10.0$ nm and $\gamma = 3.0$ and plot the linear and third-order nonlinear absorption coefficients ($\alpha^{(1)}$ and $\alpha^{(3)}$), as well as the total absorption coefficient ($\alpha = \alpha^{(1)} + \alpha^{(3)}$), as a function of the incident photon energy $h\nu$ for two different incident optical intensities 2.5×10^7 W m^{-2} (solid line) and 7.5×10^7 W m^{-2} (dashed line), respectively. It is readily seen that the large $\alpha^{(1)}$, which comes from the linear susceptibility term, is positive, whereas $\alpha^{(3)}$, which is generated by the nonlinear third-order susceptibility term, is negative. So the coefficient α is significantly reduced by the $\alpha^{(3)}$ contribution. Hence, since $\alpha^{(3)}$ can induce nonlinear absorption, $\alpha^{(3)}$ should be considered when the incident optical intensity I is strong.

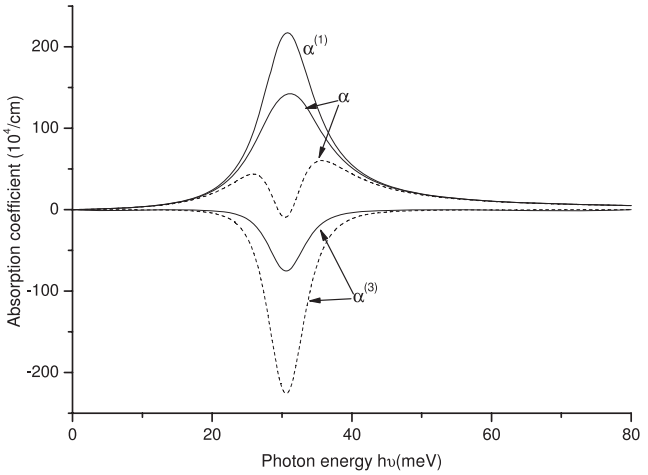

Figure 6. The linear, third-order nonlinear and total absorption coefficients of a two-electron QD confined by the Woods–Saxon potential as a function of the incident photon energy $h\nu$ for two different values of the incident optical intensity I. Parameters are taken the same as in figure 4.

Further, in order to show better the influence of the incident optical intensity I for the total absorption coefficient α, in figure 7, we set $V_0 = 112.23$ meV and $R_0 = 10.0$ nm and plot α as a function of the incident photon energy $h\nu$ for eight different values of $I = 0$, 1.5×10^7 W m^{-2}, 2.5×10^7 W m^{-2}, 3.5×10^7 W m^{-2}, 4.5×10^7 W m^{-2}, 5.5×10^7 W m^{-2}, 6.5×10^7 W m^{-2} and 7.5×10^7 W m^{-2}, respectively. The maximum absorption coefficient corresponding to the threshold photon energy decreases with increasing I. The absorption will be strongly bleached at sufficiently high incident optical intensities. Figure 7 shows that the strong absorption saturation begins to occur at around $I = 3.5 \times 10^7$ W m^{-2}. When the incident optical intensity exceeds this value, the absorption peak will be significantly split into two peaks, which will be strongly bleached as a consequence of the absorption.

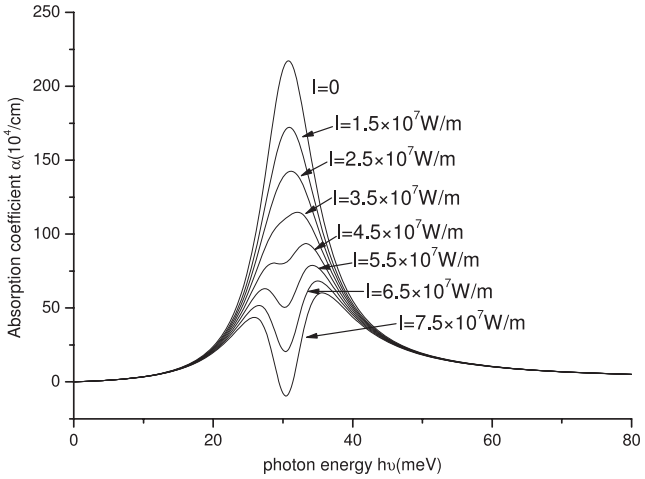

Figure 7. The total absorption coefficient of a two-electron QD confined by the Woods–Saxon potential as a function of the incident photon energy $h\nu$ for eight different values of the incident optical intensity I. Parameters are taken the same as in figure 4.

4. Summary

In conclusion, we have proposed a novel procedure to study the confinement effects of a two-electron system, i.e. the use of the Woods–Saxon confining potential function to simulate the spatial confinement of QDs. The great advantage of our methodology is that it enables confinement regimes by varying two parameters in the model potential. We have calculated the energy levels of the four spin-singlet and spin-triplet states as functions of the magnetic field by exact diagonalization within the framework of effective-mass theory. A ground-state behavior (singlet → triplet state transitions) as a function of the strength of a magnetic field has been found. We found that the confinement barrier position and the barrier inclination of a Woods–Saxon potential are important for the ground-state transition and the feature of low-lying states for a two-dimensional two-electron quantum dot. Based on the computed energies and wavefunctions, the linear and nonlinear optical absorption coefficients have been examined between the ^{1}S state ($L = 0$) and the ^{1}P state ($L = 1$). The results are presented as a function of the incident photon energy for the different values of the barrier height and size. It is found that the optical properties of the two-electron system in a QD are strongly affected by the barrier height and size. The present results are useful in understanding the electronic and optical properties of a two-electron QD.

Acknowledgment

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