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Neutral and negative donors in quantum dots

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Abstract. Using a method introduced earlier, the exact solutions of ground states of neutral donor (D^0) centres in different quantum dots (QDs) have been obtained, and, taking the Chandrasekhar-type function as a trial function, the ground states of negative donor (D^-) centres in QDs have been calculated. The dimensionality and potential-shape effects of QDs on the binding energies $E_B(D^0)$ and $E_B(D^-)$ of D^0 and D^- centres have been studied. The ratio σ of $E_B(D^0)$ to $E_B(D^-)$ has clearly demonstrated the so-called freeze-out effect in different QDs. According to the value of σ obtained, calculated results of different quantum-well structures can be checked and compared with others. It has been shown that the freeze-out effect and the electron-correlation effect are strongly dependent on the dimensionality of QDs and weakly dependent on the potential shape, and that the polarization term of the trial function not only brings in the important electron-correlation effect but also modifies the behaviour of the single-electron orbitals.

Recently, the electronic structures and properties of neutral and negative donor centres in various low-dimensional structures, such as two-dimensional quantum wells (2DQWs), quantum-well wires (QWWs) and quantum dots (QDs), have been the subject of considerable study [1–9]. The fundamental study is important in its own right as reducing the dimensionality often introduces unexpected physical phenomena.

In order to achieve a better understanding of the dimensional characteristics of multi-electron systems with less positive ion centres, a D^- centre in a spherical quantum dot has been studied [2]. However, one effect not addressed by the study is the effect that the dimensionality and potential shape of QDs have on the binding energy, the ratio of D^- to D^0 binding energy and the electron correlation. This effect should be considered to obtain the correct quantum levels of a D^- centre in a quantum dot because there are different neutral-donor wavefunctions in different QDs and they can cause quite different Coulomb and exchange potentials seen by the other electron. The dimensionality and potential-shape effect on D^- states is related to but different from that on D^0 states, and, therefore, it is interesting to investigate both of them.

This paper explores the effect on the binding energies of D^0 and D^- ground states in QDs. This is done by determining the ground states for 3D D^0 and D^- centres in QDs with a spherically rectangular potential well or a 3D isotropic parabolic potential and comparing the results with those for 2D D^0 and D^- centres in QDs with a circularly rectangular potential well or a 2D isotropic parabolic potential. For definiteness let us write down the potential forms for the QDs mentioned above. The forms of circularly and spherically rectangular potential wells are given by

$$V(\rho) = \begin{cases} \infty & \text{if } \rho \geqslant R_0 \\ 0 & \text{if } \rho < R_0 \end{cases} \tag{1}$$

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and

$$V(r) = \begin{cases} \infty & \text{if } r \geqslant R_0 \\ 0 & \text{if } r < R_0 \end{cases} \tag{2}$$

whereas the 2D and 3D isotropic parabolic potentials have the forms

$$V(\rho) = \tfrac{1}{4}\gamma^2\rho^2 \tag{3}$$

and

$$V(r) = \tfrac{1}{4}\gamma^2 r^2 \tag{4}$$

respectively. In the above equations R_0 is the radius of a QD and ρ and r are electron–QD (donor) centre distances in 2D and 3D conditions, respectively. In this paper, effective atomic units are used so that all energies are measured in units of the effective Rydberg Ry^* and all distances are measured in units of the effective Bohr radius a^*. It is interesting to point out that the parabolic potential introduced by a magnetic field perpendicular to the 2D plane is equal to that shown in (3) if the dimensionless magnetic-field strength in effective atomic units is equal to γ, which is related to the confinement energies. This is the reason why the forms of (3) and (4) are taken.

The Hamiltonian for an electron in a QD and a neutral donor D^0 at the centre of the QD is

$$H(W) = \begin{cases} -\Delta_3 - 2W/r + V(r) & \text{for 3D QDs} \\ -\Delta_2 - 2W/\rho + V(\rho) & \text{for 2D QDs} \end{cases} \tag{5}$$

where Δ_3 and Δ_2 are respectively 3D and 2D Laplace operators, $V(r)$ and $V(\rho)$ are 3D and 2D QD potentials as shown above and W is respectively equal to zero and one for the electron and the donor in the QDs. The orbital (l) and magnetic (m) quantum numbers can be well defined for the 3D QDs, whereas the magnetic (m) quantum number can be well defined for the 2D QDs. The radial equations can be solved exactly by using the method introduced by us [10], and, then, the nth eigenenergies $E_n(l, W = 0)$ and $E_n(l, W = 1)$ and the corresponding eigenfunctions $\psi_n^{(l)}(r, W = 0)$ and $\psi_n^{(l)}(r, W = 1)$, which are degenerate with respect to m, can be obtained for the 3D electron and donor in 3D QDs. This is similar to the method of obtaining the nth eigenenergies $E_n(m, W = 0)$ and $E_n(m, W = 1)$ and $\psi_n^{(m)}(\rho, W = 0)$ and $\psi_n^{(m)}(\rho, W = 1)$, which are degenerate with respect to m and $-m$, for the 2D electron and donor in 2D QDs. Compared with the binding energy of a D^0 centre in a 3D, 2D or 1D system, the binding energies of the D^0 ground and excited states in QDs can be defined as follows:

$$E_{n\mathrm{B}}(l, \mathrm{D}^0) = E_n(l, W = 0) - E_n(l, W = 1) \tag{6}$$

and

$$E_{n\mathrm{B}}(m, \mathrm{D}^0) = E_n(m, W = 0) - E_n(m, W = 1) \tag{7}$$

for 3D and 2D QDs, respectively. Therefore, the binding energies $E_{\mathrm{B}}(\mathrm{D}^0)$ of the ground states are given by

$$\begin{cases} E_{\mathrm{B}}(\mathrm{D}^0) = \frac{3}{2}\gamma - E_0(l = 0, W = 1) \\ E_{\mathrm{B}}(\mathrm{D}^0) = \gamma - E_0(m = 0, W = 1) \end{cases} \tag{8}$$

for the 3D and 2D parabolic potentials, and

$$\begin{cases} E_B(D^0) = 9.870/R_0^2 - E_0(l = 0, W = 1) \\ E_B(D^0) = 5.783/R_0^2 - E_0(m = 0, W = 1) \end{cases} \tag{9}$$

for the 3D and 2D rectangular potentials where the first terms on the right-hand side (RHS) in the equations are ground-state energies of an electron in 3D and 2D parabolic potentials and 3D and 2D rectangular ones, respectively.

The Hamiltonian for a D^- in a QD is given by

$$H = \begin{cases} H(1, W) + H(2, W) + 2/r_{12} & \text{for 3D QDs} \\ H(1, W) + H(2, W) + 2/\rho_{12} & \text{for 2D QDs} \end{cases} \tag{10}$$

where $H(i, W)$ is the Hamiltonian of the ith electron and the positive donor ion in the QD as given in (5) and $2/r_{12}$ and $2/\rho_{12}$ are interaction terms of the two 3D and two 2D electrons, respectively.

To determine the ground states (spin singlet states) in the QDs, a trial function that includes the electron-correlation effect and approaches the Chandrasekhar-type trial function at $\gamma = 0$ or $R_0 \to \infty$ is used [11]. It is as follows:

$$\Psi = \begin{cases} A(1 + Cr_{12})\{\psi(\lambda_1, r_1)\psi(\lambda_2, r_2) + \psi(\lambda_2, r_1)\psi(\lambda_1, r_2)\} & \text{for 3D QDs} \\ B(1 + C\rho_{12})\{\psi(\lambda_1, \rho_1)\psi(\lambda_2, \rho_2) + \psi(\lambda_2, \rho_1)\psi(\lambda_1, \rho_2)\} & \text{for 2D QDs} \end{cases} \tag{11}$$

where C, λ_1 and λ_2 are variational parameters and A and B are the normalization constants. $\psi(\lambda_i, r_i)$ and $\psi(\lambda_i, \rho_i)$ are exactly the lowest eigenfunctions of hydrogenic donors with the ion charge λ_i in the 3D and 2D QDs, respectively. λ_1, λ_2 and their difference $\Delta\lambda = \lambda_1 - \lambda_2$ describe one part of the two-electron correlation while the factor $(1 + Cr_{12})$ or $(1 + C\rho_{12})$ describes the other. In the limiting case of $\gamma = 0$ or $R_0 \to \infty$, it is clearly seen that the electrons are in ground-state hydrogenic orbitals; they tend to stay apart, keeping their repulsive interaciton energy down as indicated by the correlation factor $(1 + Cr_{12})$ or $(1 + C\rho_{12})$, which becomes relatively small as $r_{12} \to 0$ or $\rho_{12} \to 0$. It is interesting to point out that for D^- centres in different QDs, using the same kind of trial function as (11) is quite good for comparison, and, therefore, it is possible to obtain a reasonable conclusion based on variational calculations with the exact results of ground states of the electron and the D^0 centre in the QDs. Compared with the binding energy of a D^0 centre in QDs as mentioned above, the binding energy of the D^- ground state in the QDs is defined as follows:

$$E_B(D^-) = E(D^0) + E_0 - E(D^-) = 2E_0 - E_B(D^0) - E(D^-) \tag{12}$$

where $E(D^-)$ is the lowest level of the Hamiltonian of (10), i.e., the D^- ground-state energy in QDs, E_0 and $E(D^0)$ are, respectively, the lowest levels of an electron in the QDs without and with the Coulomb potential and $E_B(D^0)$ is the binding energy of the neutral donor as defined in (8) and (9).

In order to study the γ and R_0 dependence of the binding energies $E_B(D^0)$ and $E_B(D^-)$ of D^0 and D^- centres in QDs and the dimensionality and potential-shape effects of QDs on them, numerical calculations have been performed for 2D and 3D rectangular-potential QDs (2D and 3D RQDs) and 2D and 3D parabolic-potential QDs (2D and 3D PQDs). As shown in table 1, it is readily seen that as γ increases, both the $E_B(D^0)$ and $E_B(D^-)$ in 2D and 3D PQDs increase monotonically from their 2D and 3D values to quasi-zero-dimensional ones, and the

values are always larger for the 2D case than for the 3D case. To study the potential-shape effect of QDs on the binding energies of D^0 and D^- centres, we have also given $E_B(D^0)$ and $E_B(D^-)$ of 3D and 2D RQDs as functions of the effective γ^*, which is, respectively, defined as $6.580/R_0^2$ and $5.783/R_0^2$. The effective γ^* means that the ground-state energies of an electron in 3D and 2D PQDs are respectively equal to those in 3D and 2D RQDs if γ^* is taken to be equated to γ. As shown in the table, the binding energies of both D^0 and D^- centres in 3D and 2D RQDs increase slightly faster than those in 3D and 2D PQDs as both γ and γ^* increase. What we have mentioned above means that the binding energies are *mainly dependent on the dimensionality (3D or 2D) of the QDs and the confinement energies*, and that for the same confinement energy and dimensionality, they are slightly dependent on the potential shape.

Table 1. Binding energies $E_B(D^0)$ and $E_B(D^-)$ of D^0 and D^- ground states in 3D PQDs, 3D RQDs, 2D QWs, 2D PQDs and 2D RQDs with γ (γ^*) = 0, 0.1, 1, 3, 10, 100. The corresponding radius R_0 of 3D and 2D RQDs is also presented (see the text). The values of 2D QWs and those in brackets are taken from [12] and [8], respectively. Effective atomic units, i.e., the effective Rydberg Ry^* and Bohr radius a^*, are used.

		γ (γ^*)					
		0	0.1	1	3	10	100
3D PQDs	$E_B(D^0)$	1.00	1.143	2.01	3.14	5.40	16.31
	$E_B(D^-)$	0.0518 (0.056)	0.165	0.54	0.90	1.59	4.79
3D RQDs	$E_B(D^0)$	1.00	1.143	2.16	3.57	6.14	18.70
	$E_B(D^-)$	0.0518	0.165	0.60	1.00	1.72	5.05
	R_0	∞	8.112	2.565	1.481	0.811	0.257
2D QWs	$E_B(D^0)$	1.69 (1.74)	—	2.44 (2.52)	3.24 (3.36)	4.73	—
	$E_B(D^-)$	0.212 (0.23)	—	0.639 (0.65)	0.906 (0.94)	1.33	—
2D PQDs	$E_B(D^0)$	4.00	4.101	4.91	6.33	9.63	26.55
	$E_B(D^-)$	0.454 (0.511)	0.549	1.11	1.76	2.91	7.98
2D RQDs	$E_B(D^0)$	4.00	4.101	4.99	6.69	10.60	30.40
	$E_B(D^-)$	0.454	0.554	1.22	1.95	3.20	8.53
	R_0	∞	7.605	2.405	1.388	0.760	0.241

In order to achieve a better understanding of the dimensionality and potential-shape effects of QDs on D^0 and D^- centres, we have also shown the values of $E_B(D^0)$ and $E_B(D^-)$ of 2D QWs of $Ga_{0.75}Al_{0.25}As$ with $L = 200$ Å (about $2a^*$) [8, 12] for the same γ and calculated the ratio σ of D^- to D^0 binding energy in different QDs. As shown in figure 1, with increasing γ (γ^*), σ values increase from the 2D and 3D values up to certain approximately constant values. This is the so-called freeze-out effect. This feature can be explained as follows. The extension of the outer orbital in a D^- centre sharply decreases with increasing confinement in a small-γ (γ^*) regime, compared with the extension of a neutral donor orbital, which decreases rather slowly, and the decreases in extension of both D^0 and D^- orbitals with increasing confinement in a large-γ (γ^*) regime are in much the

same ratio. In fact, both wavefunctions are mainly dependent on γ (γ^*) and the Coulomb potentials can be taken to be perturbation terms if γ (γ^*) is large enough. The perturbation calculation can also give the limit value of σ, which is very close to what we show here. It is clearly shown that the limit value of σ is about 0.3 for 3D and 2D PQDs while it is about 0.21 and 0.12 for the corresponding QWWs [9] and 2D QWs [8, 12], and that the limits of the other 3D and 2D QDs are slightly different from those of 3D and 2D PQDs, respectively. This means that the freeze-out effect (the limit value) is strongly dependent on the confined dimensionality and weakly dependent on the potential shape.

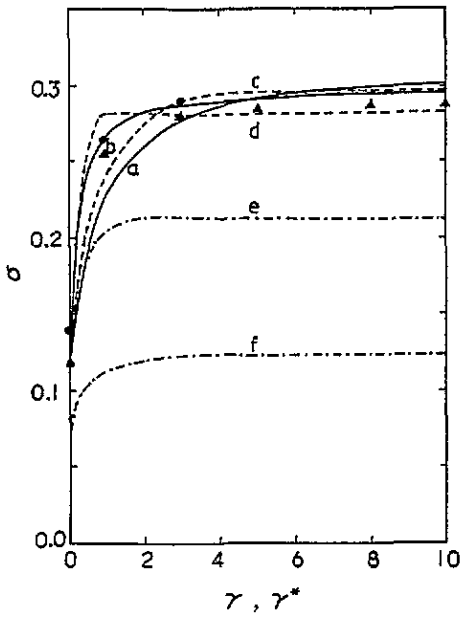

Figure 1. The ratio σ of $E_B(D^-)$ to $E_B(D^0)$ versus γ (γ^*) for 2D (solid line a) and 3D (solid line b) PQDs and 2D (dashed line c) and 3D (dashed line d) RQDs. The solid circles and triangles represent the ratios for 2D QWs with magnetic field γ from [8] and [12]. The dashed–dotted lines e and f represent those for QWWs from [9] and 2D QWs from [12], respectively.

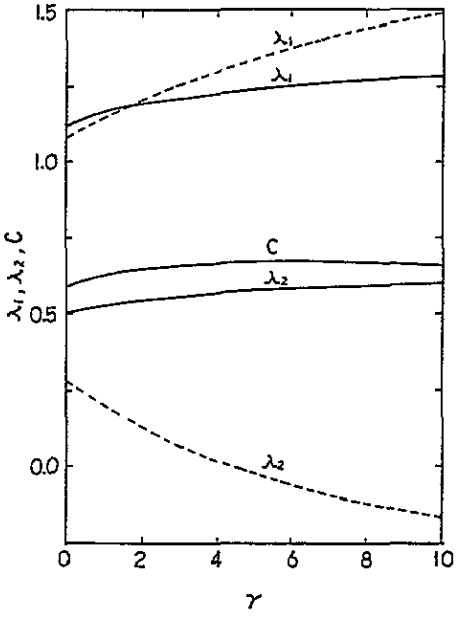

Figure 2. Variational parameters λ_1 and λ_2 with $C \neq 0$ (solid lines) and λ_1 and λ_2 with $C = 0$ (dashed lines) versus γ for D^- ground states in 2D PQDs.

The variation of σ with γ (γ^*) can be quite different between 3D and 2D QDs as shown in figure 1. It is readily seen that σ increases from 0.0518 to the limit value for 3D QDs much faster than from 0.113 to the limit value for 2D QDs and σ of RQDs increases faster than the corresponding one of PQDs as γ (γ^*) increases from zero to three. It is worthwhile to note that in the regime of γ (γ^*) between zero and three, σ of 3D QDs can be larger than that of 2D QDs even though it is much smaller in the pure 3D case than in the pure 2D case, and both $E_B(D^0)$ and $E_B(D^-)$ of 3D QDs are smaller than the corresponding ones of 2D QDs. Comparing 2D QWs in the absence of a magnetic field with the pure 3D and 2D results, we find that σ of 2D QWs is larger than those of both pure 3D and 2D cases. It is similar to those of 3D and 2D RQDs. σ (0.302 and 0.280) for $\gamma^* = 10$ of 2D and 3D RQDs are larger than the corresponding σ (0.281 and 0.270) of $\gamma^* = 100$. This means that for 2D QW and RQD systems there is a maximum ratio reached before the limit value is obtained. This is an interesting problem to study further.

It is well known that electron correlation effects play an important role in determining the electronic structures and the binding energies of D^- ground states, so a reasonable trial function should include these effects. As shown in (11), the correlation term of two electrons is included in the Chandrasekhar-type trial function. Using the trial function with and without the polarization term in (11) and making a comparison between the two cases, the correction effect can be studied.

Using the trial function with the polarization term, we have obtained that the binding energy $E_B(D^-)$ of 2D PQDs is equal to 0.454, 1.110, 1.760, 2.914 and 7.975 Ry^* for $\gamma = 0$, 1, 3, 10 and 100, respectively. The corresponding ratio R of the binding energy difference due to omission of the polarization term to the binding energy is given as 0.3255, 0.165, 0.134, 0.125 and 0.086. It is shown that the correlation effect on binding energy decreases with increasing γ.

Our calculated results have also shown that the correlation effects in 2D RQDs are about the same as those in 2D PQDs when $\gamma = \gamma^*$. Furthermore, the effects decrease with increasing γ (γ^*) for 3D QDs much faster than those for 2D QDs, and, for example, the ratio R is respectively equal to 0.479, 0.111, 0.072, 0.045 and 0.018 for 3D QDs with $\gamma = 0$, 1, 3, 10 and 100. This is because of the dimensionality effect and the fast rate of decrease of the confinement region of the electron in 3D QDs.

In addition to the binding energies of D^- centres in QDs, the variational parameters λ_1, λ_2 and C are useful quantities to ascertain electron correlation, as they provide some insight into the spatial extension of the trial function. At γ (γ^*) = 0, the best values of λ_1, λ_2 and C are respectively 1.075, 0.478 and 0.312 for 3D D^- and 1.12, 0.505 and 0.587 for 2D D^- while the best values of λ_1 and λ_2 with $C = 0$ are respectively 1.039 and 0.283 for 3D D^- and 1.08 and 0.28 for 2D D^-. For $C \neq 0$, both the λ_1 and λ_2 increase with γ (γ^*) and the difference $\Delta\lambda = \lambda_1 - \lambda_2$ changes with γ (γ^*) very slowly. For $C = 0$, however, λ_1 increases and the λ_2 decreases, so $\Delta\lambda$ increases with increasing γ (γ^*). For example, the values of λ_1 and λ_2 with $C \neq 0$ and $C = 0$ of D^- centres in 2D PQDs are shown in figure 2. What we have mentioned above clearly demonstrates that the finite value of C of (11) not only introduces an important correlation effect but also modifies the behaviour of the single-electron orbitals.

In conclusion, we have used the exact solution and the Chandrasekhar-type trial function, which consists of the exact eigenfunction of the Hamiltonian $H(i, \lambda_i)$, and obtained the binding energies of D^0 and D^- centres in QDs. The dimensionality and potential-shape effects of QDs on $E_B(D^0)$ and $E_B(D^-)$ and the ratio σ have been studied. The study of the binding energy and electron-correlation and freeze-out effects of D^- centres in different QDs is important not only to understand electronic structures in low dimensions but also to explain the experiments on D^- centres in 2D QWs [3,4] in a magnetic field and in other kinds of QW structure. Since 2D QWs in a magnetic field form a kind of QD between 3D and 2D PQDs and D^- centres could be located anywhere in QDs, it should be interesting to study the positional dependence of D^- states in RQDs, PQDs and other kinds of quantum dot. Finally, it is worthwhile to point out that the D^- excited states can be quite different between different QDs. This is an interesting subject to study further.

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