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Electric field effects on the binding energy of hydrogen impurities in quantum dots with parabolic confinements

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Abstract

Using a variational approach, the binding energy of hydrogen impurities in (i) spherical quantum dots with parabolic confinements in an electric field and (ii) disc-like quantum dots with parabolic lateral confinements in a parallel electric field are calculated. Both the confinement and the electric field effects on the binding energy are investigated in detail.

1. Introduction

The important role of the physics of impurity states for the understanding of electronic properties of bulk semiconducting materials has been widely recognized since the early days of semiconductor science [1, 2]. In the last few decades, because of the great interest in the physics and technological applications of low-dimensional semiconductor structures such as quantum wells, quantum wires, and quantum dots [3–5], the problem of impurity states has received renewed attention. While for the quantum wells the impurity states have been studied in great detail [6, 7], the problem has been less thoroughly investigated for quasi-zero-dimensional systems of quantum dots (QDs). The main features to consider in relation to QDs are geometrical shape, size, and the confining potential. The shallow donor states in the simplest models of QDs with square (infinite or finite) potential wells were studied early on in a number of works [8, 9]. The effects of parabolic confining potentials on the binding energy of hydrogen impurities were recently calculated by Xiao *et al* [10], Bose and Sarkar [11, 12], and Varshni [13], using a variational method. It should be noted that virtually all of the studies of references [8–13] have been exclusively limited to the effect of confining potentials, and, moreover, all of these models are characterized by spherical symmetry which allows one to reduce the problem to that of solving a simpler equation in the radial variable. The spherical symmetry may be violated by different factors such as dot shapes, and confining or external

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field potentials. The effects of such 'symmetry-breaking' factors on the binding energy of hydrogen impurities in QDs, although fundamental and important for applications, have been even less studied [14, 15].

Concerning the confining potential, we would note that the results of a numerical self-consistent solution of the Poisson and Schrödinger equations in the Hartree approximation performed by Kumar *et al* [16] strongly support a parabolic form for the confining potentials for QDs fabricated from GaAs/AlGaAs heterostructures. Note also the good agreements between experimental data on various electronic characteristics and corresponding theoretical calculations, using the parabolic form for the confining potentials, for a large class of both three-dimensional (3D) [4, 5, 17] and two-dimensional (2D) [4, 5, 18] QDs.

The aim of this work is to calculate the binding energy of a hydrogen impurity, located in the centre of (1) spherical QDs with parabolic confinement in an external electric field and (2) 2D disc-like QDs with a lateral parabolic confinement in a parallel electric field. Unlike in the case of an external magnetic field [15], where two parabolic potentials, magnetic and confining, can partly be added together, an electric field—as was recently discussed by Murillo and Porras-Montenegro [14]—moving the parabolic confining potential along the field direction by a distance depending on both the field intensity and the confining potential strength results in an additional asymmetry of the problem. Taking this into account, two-parameter trial variational functions are suggested and the binding energies are calculated for the ranges of magnitudes of both the confining and the electric field potentials of practical interest; this gives, on one hand, the correct values of the binding energy in the limit case of zero field and, on the other hand, the electric field-induced corrections to the binding energy for both kinds of QD under study with different sizes of confining potential.

2. Theory

Our study starts with the standard dimensionless effective-mass Hamiltonian of a hydrogen impurity in a confining potential V_C and in an external electric field:

$$\mathcal{H} = -\nabla^2 - 2/r + V_C + \boldsymbol{F}\cdot\boldsymbol{r}. \tag{1}$$

Here, $\boldsymbol{F}$ is the dimensionless electric field; the impurity is assumed to be located at the centre of the QD, which is also chosen as the origin of the coordinate system. Throughout this work, effective atomic units are used, so all energies are measured in units of the effective Rydberg $R^* \equiv m^*e^4/2\hbar^2\varepsilon^2$ and all lengths are in units of the effective Bohr radius $a_B \equiv \hbar^2\varepsilon/m^*e^2$, where m^* is the electron effective mass, e is the elementary charge, ε is the dielectric constant of the dot materials. The confining potential V_C is assumed to have the parabolic form

$$V_C = \beta^2 r^2. \tag{2}$$

In the model, the polarization and image-charge effects are neglected. Such a model could be used for describing, for example, QDs fabricated from the GaAs/AlGaAs heterostructures [4, 5].

Generally, the binding energy is defined as

$$\mathcal{E}_B = \mathcal{E}_0 - \mathcal{E}_C \tag{3}$$

where $\mathcal{E}_C$ and $\mathcal{E}_0$ are ground-state energies of the Hamiltonian of equation (1) with and without the Coulomb term, respectively.

2.1. Spherical QDs

The effect of an electric field on the binding energy of hydrogen impurities in bulk semiconductors is well known [2]. In addition, without external fields the effect of parabolic

confining potentials on the binding energy of hydrogen impurities in spherical QDs (S-QDs) has been investigated in detail [10–13]. To clarify how these two factors are combined in affecting the binding energy, we rewrite the Hamiltonian of equations (1), (2) for S-QDs in the form

$$\mathcal{H} = -\nabla^2 - \frac{2}{\sqrt{\rho^2 + z^2}} + \beta^2\left[\rho^2 + \left(z + \frac{F}{2\beta^2}\right)^2\right] - \frac{F^2}{4\beta^2} \tag{4}$$

where the z-axis is chosen along the electric field direction, and $\rho^2 = x^2 + y^2$. From this expression it is clear that the electric field moves the location of the harmonic confining potential along the field direction by a distance of $-F/2\beta^2$, and, at the same time, shifts the total energy by $-F^2/4\beta^2$, while the 'strength' of the parabolic confining potential is still the same β-value. Leaving aside the constant term $F^2/4\beta^2$, in the Hamiltonian of equation (4) the field F then appears only in combination with the confining parameter β in the distance expression for $F/2\beta^2$, which unambiguously implies a strong correlation between two effects, confining and electric field, on the impurity states. Furthermore, since the characterization length $L_c = 1/\sqrt{\beta}$ of the parabolic confining potential is often considered the effective radius of the QD, it is meaningful to define the ratio $\mathcal{F} = (F/2\beta^2)/L_c \equiv F/2\beta^{3/2}$ as the measure of the electric field F for a given value of β (i.e. given QD). The quantity $\mathcal{F}$, i.e. the 'effective electric field', will then be the main parameter in the problem under study, and, certainly, all calculations should be performed only for the range of $\mathcal{F} \leqslant 1$.

Without the Coulomb term, the ground-state energy of the Hamiltonian (4) is well known:

$$\mathcal{E}_0 = 3\beta - \beta\mathcal{F}^2. \tag{5}$$

Taking into account the Coulomb potential, the Hamiltonian (4) cannot be solved analytically. To calculate the ground-state energy $\mathcal{E}_C$, and then the binding energy $\mathcal{E}_B$ of equation (3), we will use a variational approach. In order to choose an adequate trial function, it is important to note that, as is well known, the matrix diagonal element of the electric field potential can only be finite for states of undefined parity. An even or odd function should not be used as the trial function. In view of this and of the fact that the total potential in the Hamiltonian of equation (4) consists of only two terms, corresponding to the hydrogen Coulomb centre at the coordinate origin and to the three-dimensional harmonic oscillator located on the z-axis at $z = -F/2\beta^2$, we can suggest a trial function of the form

$$\Psi = C\exp(-ar)\exp\{-(\beta/2)[\rho^2 + (z + b\beta^{-1/2}\mathcal{F})^2]\} \tag{6}$$

where C is a normalized constant, and a and b are variational parameters.

It is interesting to note that the Hamiltonian of equation (4) has the same form as that for the problem of an off-centre hydrogen impurity in a QD with parabolic confinement without an external electric field, where the separation between the impurity location and the parabolic dot centre is equal to $F/2\beta^2$. The binding energy in such a problem was recently calculated by Bose [11], using a trial function similar to that of equation (6), but with only one parameter, a (i.e. equation (6) with $b = 1$). In view of the symmetry of the Hamiltonian (4) there is clearly a question as regards the accuracy of such one-parameter trial functions as that of reference [11], where the asymmetry associated with the field is not accounted for adequately. The important role of a second parameter b was especially emphasized by Galiskii *et al* [19] in relation to the analogous problem of calculating the polarizability of a hydrogen atom. For even the simpler problem of an on-centre hydrogen impurity in spherical QDs without external fields, Varshni [13] also shows that inclusion of the second parameter associated with the confining potential gives more accurate results for the binding energy. Thus, we choose the trial function with two variational parameters of equation (6).

Substituting the trial function of equation (6) into the Hamiltonian $\mathcal{H}$ of equation (4), we obtain

$$\frac{\langle\Psi|\mathcal{H}|\Psi\rangle}{\langle\Psi|\Psi\rangle} = 3\beta - \beta b^2\mathcal{F}^2 - a^2 + \beta(1-b)/b + [(a-2)I_0 - 2\beta a I_2 + 2\beta^{1/2}abJ_1 + 2\beta^{3/2}(b-1)J_2]/I_1 \tag{7}$$

where

$$I_n = (1/\mathcal{F})\int_0^\infty r^n \exp(-\beta r^2 - 2ar)\sinh(2b\beta^{1/2}\mathcal{F}r)\,\mathrm{d}r$$

$$J_n = \int_0^\infty r^n \exp(-\beta r^2 - 2ar)\cosh(2b\beta^{1/2}\mathcal{F}r)\,\mathrm{d}r$$

and where the constant term $-F^2/4\beta^2 \equiv -\beta\mathcal{F}^2$ is not yet included.

2.2. Disc-like QDs

Let us consider a 2D disc-like QD (2D DL-QD) in the (x, y) plane in a parallel electric field. Choosing the x-axis along the field direction, the Hamiltonian of equations (1), (2) for 2D DL-QDs can then be written in a form similar to that for the S-QDs of equation (4):

$$\mathcal{H} = -\nabla^2 - \frac{2}{\sqrt{x^2+y^2}} + \beta^2\left[y^2 + \left(x + \frac{F}{2\beta^2}\right)^2\right] - \frac{F^2}{4\beta^2}. \tag{8}$$

The ground-state energy of this Hamiltonian without the Coulomb potential is

$$\mathcal{E}_0 = 2\beta - \beta\mathcal{F}^2 \tag{9}$$

where the effective electric field $\mathcal{F}$ has already been defined: $\mathcal{F} = F/\beta^{3/2}$. In order to calculate the ground-state energy $\mathcal{E}_C$, following the route discussed above for S-QDs, we use a variational approach with a trial function of the form

$$\Psi = C\exp(-a\rho)\exp\{-(\beta/2)[y^2 + (x + b\beta^{-1/2}\mathcal{F})^2]\}. \tag{10}$$

Here, the meanings of C, a, and b are the same as those explained below equation (6), and $\rho = \sqrt{x^2+y^2}$.

Using the trial function of equation (10), from the Hamiltonian of equation (8) we obtain the variational energy (without the constant term $-\beta\mathcal{F}^2$):

$$\frac{\langle\Psi|\mathcal{H}|\Psi\rangle}{\langle\Psi|\Psi\rangle} = 2\beta - a^2 - \beta b^2\mathcal{F}^2 + [(a-2)J_0^0 - 2a\beta J_2^0 + 2ab\beta^{1/2}\mathcal{F}J_1^1 + 2(b-1)\beta\mathcal{F}^2 J_2^1]/J_1^0 \tag{11}$$

where

$$J_m^n = \int_0^\infty x^n \exp(-\beta x^2 - 2ax) I_n(2b\beta^{1/2}\mathcal{F})\,\mathrm{d}x$$

with $I_n(x)$ being the nth-order hyperbolic Bessel function [20, 21].

In the expressions for the variational energies of equations (7) and (11), a and b are variational parameters, while β is the parameter characterizing the QD under study. Given β, the quantity $\mathcal{F}$ measures the electric field. Minimizing the energies of equation (7) or equation (11), including the constant term $-\beta\mathcal{F}^2$, we will obtain the ground-state energies $\mathcal{E}_C \equiv \mathcal{E}_C(\beta, \mathcal{F})$, respectively for S-QDs or 2D DL-QDs. Further, from equation (3) with $\mathcal{E}_0$ of equation (5) for S-QDs or equation (9) for 2D DL-QDs, the binding energy will be determined. The field-induced corrections $\Delta\mathcal{E}_B(\beta, F) = \mathcal{E}_B(\beta, F) - \mathcal{E}_B(\beta, 0)$ then describe the effect of

the electric field on the binding energy of a hydrogen impurity in QDs of the given β, while the zero-field binding energy $\mathcal{E}_B(\beta, 0)$ describes the confinement effect alone, which for S-QDs should be compared with the results of Xiao *et al* [10] and Varshni [13]. For the 2D DL-QDs, we do not know of any work dealing with calculations of even zero-field binding energies $\mathcal{E}_B(\beta, 0)$ which could be referred to for comparison.

3. Numerical results and discussion

As mentioned above, all energies in this work are measured in units of R^*. Taking GaAs as a typical QD material, one has [22] $m^* = 0.063m_0$, $\varepsilon = 12.9$, $a_B = 10.19$ nm, and therefore $R^* = 5.478$ meV. For these values of the material parameters, the value of the effective field $\mathcal{F} = 1$ corresponds to a field of $\approx 5.4 \times 10^5$ V m^{-1} or $\approx 3 \times 10^4$ V m^{-1} for $\beta = 1$ or 0.1, respectively.

Figure 1 shows the binding energy $\mathcal{E}_B$ as a function of the effective field $\mathcal{F}$ for S-QDs with $\beta = 4$, 2, 1, 0.5, 0.25, and 0.1 (figure 1(a)) and for 2D DL-QDs with $\beta = 1$, 0.5, 0.25,

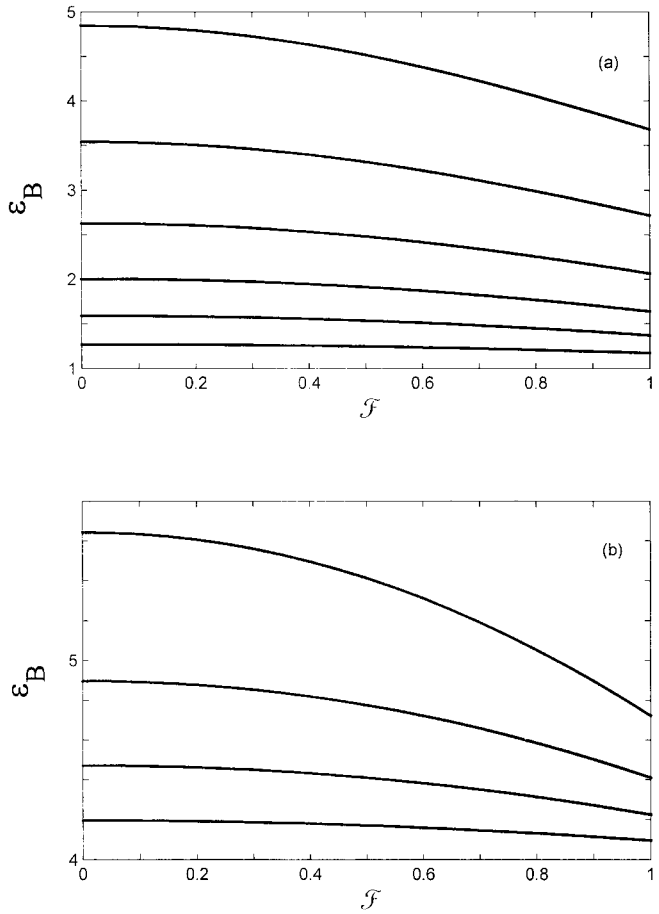

Figure 1. The binding energy $\mathcal{E}_B$ as a function of the effective electric field $\mathcal{F}$: (a) for S-QDs with $\beta = 4$, 2, 1, 0.5, 0.25, and 0.1 (from top); (b) for 2D DL-QDs with $\beta = 1$, 0.5, 0.25, and 0.1 (from top).

and 0.1 (figure 1(b)). The chosen range of β for S-QDs in figure 1(a) implies that we are only interested in the S-QDs with intermediate or strong confinements—most relevant experimental measurements are also on such objects. For the 2D DL-QD case, since, in reality, any DL-QD has of course a finite thickness, the structure cannot be considered two-dimensional unless its thickness is much less than its longitudinal size. Since the dot thickness is often not less than $\approx$3–7 nm (i.e. $\approx$0.3–0.7 a_B for GaAs) [4, 5, 16, 22], the dot longitudinal size L_C should be not less than some tens of nm (i.e. β should be not larger than 1).

For both the S-QDs and the 2D DL-QDs, as is clear in figure 1, the binding energy decreases as the field increases and the relative reduction

$$\delta\mathcal{E}_B \equiv (\mathcal{E}_B(\mathcal{F}=0) - \mathcal{E}_B(\mathcal{F}=1))/\mathcal{E}_B(\mathcal{F}=0)$$

slightly goes down with decreasing β: $\delta\mathcal{E}_B \approx 22\%$ for $\beta = 4$, and $\approx$8% for $\beta = 0.1$ in figure 1(a) for S-QDs and $\delta\mathcal{E}_B \approx 16\%$ for $\beta = 1$, and $\approx$2% for $\beta = 0.1$ in figure 1(b) for 2D DL-QDs. (Noting again that the problem is meaningless when $\mathcal{F} > 1$, and the difference in range of real electric fields for various curves associated with various values of β in the figure, it is clear that one should be careful when comparing the field dependence behaviours of different curves.)

In particular, the zero-field binding energies $\mathcal{E}_B(\beta) \equiv \mathcal{E}_B(\beta, \mathcal{F}=0)$, describing the effect of the confining potential alone, were calculated for many points in the chosen ranges of β-values to investigate how $\mathcal{E}_B$ depends on β. The numerical results obtained seem to fit well to the simple approximate expressions

$$\mathcal{E}_B(\beta) \approx \begin{cases} -0.194\beta^2 + 1.691\beta + 1.000 & \text{(S-QDs)} \quad (12a) \\ -0.284\beta^2 + 1.915\beta + 4.009 & \text{(DL-QDs).} \quad (12b) \end{cases}$$

It is important to note that these fitting expressions, deduced from the data for the limited ranges of β (from 0.1 to 4 for S-QDs and from 0.1 to 1 for 2D DL-QDs), could not be considered valid for all β. In the limit of very strong confinements (large β), the accuracy of the effective-mass approximation might be questioned, and, certainly, the central-cell corrections should be taken into account in calculating the binding energy [5, 9]. The ranges of β chosen in this study simply correspond to most QDs investigated experimentally [4, 5, 17, 18, 23].

We would like to mention here that our results for the zero-field binding energies $\mathcal{E}_B(\beta)$ for the S-QDs are in good agreement with those of Xiao *et al* [10] and of Varshni [13]. For example, for the case of the largest hard-boundary radius $R = 7$, the binding energies presented in reference [13] are 1.490 63, 1.680 22, and 1.849 63 for $\beta = 0.2$, 0.3, and 0.4, respectively, while our calculations give corresponding values of 1.489 46, 1.680 20, and 1.849 63. The fact that the two results for $\beta = 0.4$ exactly coincide shows unambiguously that for such strong confinement the distance of $R = 7$ (in units of a_B) can really be considered infinite, and, consequently, the effect of the hard boundary located there on the binding energy is not yet visible.

To see more clearly the electric field effect, the field-induced reductions of the binding energies, $\Delta\mathcal{E}_B(\beta, \mathcal{F}) = \mathcal{E}_B(\beta, \mathcal{F}) - \mathcal{E}_B(\beta, 0)$, are presented in figure 2(a) and figure 2(b) for the S-QDs and 2D DL-QDs, respectively. Expressing the parameter $\mathcal{F}$ in the real electric field F, with the aim of achieving practical applicability, it seems that for both kinds of QD the data follow well the square-power law for F:

$$\Delta\mathcal{E}_B(\beta, F) = -Q(\beta)F^2 \quad (13)$$

where $Q(\beta)$ is approximately estimated as

$$Q(\beta) \approx \begin{cases} 0.256\beta^{-2} - 0.139\beta^{-1} & \text{for S-QDs} \quad (14a) \\ 0.251\beta^{-2} - 0.013\beta^{-1} & \text{for DL-QDs.} \quad (14b) \end{cases}$$

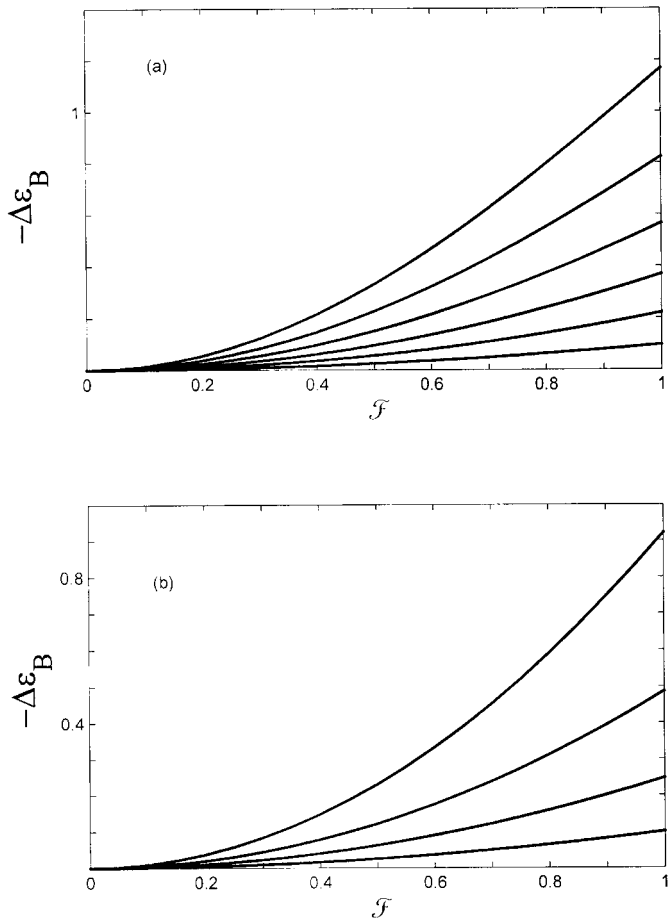

Figure 2. The negative of the electric field-induced corrections, $-\Delta\mathcal{E}_B$, to the binding energy as a function of the effective electric field $\mathcal{F}$: (a) for S-QDs with $\beta = 4$, 2, 1, 0.5, 0.25, and 0.1 (from top); (b) for 2D DL-QDs with $\beta = 1$, 0.5, 0.25, and 0.1 (from top).

While the field dependence $\propto -F^2$ of the field-induced corrections $\Delta\mathcal{E}_B$ is perhaps easy to guess like that for the bulk problem, our calculations suggest the expressions of equation (14) for the factors $Q(\beta)$ for the ranges of β studied. In reality, since the QD size L_C is often not less than several a_B, i.e. β is much less than 1, these factors $Q(\beta)$ could then even be approximated in the simpler form $Q(\beta) \approx 0.25\beta^{-2}$ for both the kinds of QD.

Finally, in the case of DL-QDs, though the present study is devoted to the two-dimensional model, we have performed some calculations, taking into account a finite thickness of dot. To this end, we simply added to the parabolic lateral confining potential $V_C = \beta^2\rho^2$ an infinite square potential along the z-axis: $V_z = 0$ for $|z| < d/2$ and $=\infty$ for $z \geqslant d/2$, where d is the dot thickness. The field direction is still parallel to the (x, y) plane. The ground-state energy $\mathcal{E}_0$ is just given by the well-known expression: $\mathcal{E}_0 = 2\beta - \beta\mathcal{F}^2 + \pi/d^2$. The trial function can be chosen in the form defined by the product of the function of equation (9) with the well-known eigenfunction of the infinite square potential V_z. For dots with the same value of β of 0.25 (i.e. $L_C = 2$), but with different thicknesses, $d = 0.3$, 0.5, and 0.7, the zero-field binding energies obtained are 3.573, 3.241, and 3.001, respectively, which should be

compared with the value of 4.472 for the 2D DL-QD with $d = 0$. Thus, our calculations show a considerable decrease of $\mathcal{E}_B(0)$ with increasing dot thickness even at $d \approx 0.5$. However, the calculations also show that for each thickness the β-dependence of $\mathcal{E}_B$ still seems to follow well the relation of equation (12*b*) with a shift associated with the last constant term only. And, more important, for all cases for which calculations were performed ($d \leqslant 0.7$, $L_C \geqslant 2$), the field-induced reductions of the binding energy $\Delta\mathcal{E}_B(\beta, d, \mathcal{F}) = \mathcal{E}_B(\beta, d, \mathcal{F}) - \mathcal{E}_B(\beta, d, 0)$ are almost independent of d and still well described by the same expression, equation (14*b*), as was suggested for the 2D model. In reality, besides the hard boundary there should also be a confining potential along the z-direction, which would certainly result in a smaller effective thickness of dots and therefore favour further applying the 2D DL-QDs model, even quantitatively.

4. Conclusions

We calculated the binding energies of hydrogen impurities in the centres of S-QDs with parabolic confinement in an external electric field and of 2D DL-QDs with parabolic lateral confinement in a parallel electric field, using a variational method. Simple two-parameter trial functions were suggested that, on one hand, give correct values for the binding energies in the limit case of zero field and, on the other hand, allow us to calculate the field-induced corrections to the binding energy for the ranges of electric field and dot size corresponding to experiments. The main results are, for easy use, summarized in the empirical expressions of equations (12), (14), describing the effects of the confinement and of the electric field on the binding energy. As regards the finite-thickness effect in DL-QDs, our calculations show that, for the ranges of parameters studied, though the binding energy itself considerably decreases with increasing thickness, the field-induced corrections are almost insensitive to it and are still well described by equation (14*b*). The suggested expressions would be very simple to compare with experiments. However, regrettably, we cannot find any data suitable for this.

We note that, although, in reality, the 3D etched GaAs/AlGaAs QDs, for example, are cubic, since the size of the cubes is always much greater than the characteristic length L_C of the confining potentials, the effect of the cubical hard boundaries on the binding energy is relatively small, and therefore the present model should be applicable. As regards the DL-QDs, as discussed above, experimental data support 2D electronic properties of such the structures as measured in references [18, 23]. Thus, it is hoped that this work might provide some useful insight into the physics of donor states in QDs and stimulate experimental interest in the problem.

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