

A solvable model of hydrogenic impurities in quantum dots

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Abstract

A solvable model is developed for electronic structure calculations of shallow hydrogenic impurities in two-dimensional quantum dots. We replace the actual Coulomb interaction (local potential) between the electron and the hydrogenic impurity by a projective operator (non-local separable potential) to determine the resulting electronic states in closed form. It is shown that non-local separable potentials may be used to accurately calculate the energy shift of the electronic levels as a function of the size of the quantum dot and the impurity position.

1. Introduction

The binding energy of electrons in a hydrogenic impurity located in zero-dimensional semiconductor nanostructures, like quantum dots (QDs) and quantum rings, has been attracting much attention during the last few years [1–10]. Due to the spatial confinement within the zero-dimensional nanostructure, the binding energy is strongly enhanced, as compared to its magnitude in bulk semiconductors or even in quantum wells or quantum wires [11, 12]. Thus, spectroscopy tools provide information about the confining properties of electrons and holes bound to hydrogenic impurities in zero-dimensional nanostructures [13]. Within the effective-mass approximation, the interacting electron and impurity pair is usually described by a Hamiltonian including the Coulomb potential plus a confining parabolic potential. Since no analytical solutions are available for off-center impurities, the binding energy is often obtained by means of variational techniques [1–6, 9].

In this work we present an alternative approach based on the non-local (separable) potential (NLP) method, in which the actual potential is replaced by a projective operator [14, 15] (for a brief summary of applications of NLP in condensed matter physics see [16]). The NLP method has already been successfully used to determine in a closed form the binding energy of confined excitons in parabolic QDs [17]. This method yields an exactly solvable envelope function equation from which the electron states can be readily obtained with the desired accuracy. Consequently, the effects of the confining potential on the hydrogenic impurity levels can be studied as a

function of the system parameters (size of the QD and impurity position) with little computational effort.

2. Model Hamiltonian

Consider a two-dimensional (2D) gas of noninteracting electrons confined in a zero-dimensional nanostructure in the presence of a hydrogenic impurity. In the framework of the effective-mass approximation, the single-electron Hamiltonian can be written as $\mathcal{H} = \mathcal{H}_0 + V_d$, where

$$\mathcal{H}_0 = \frac{\mathbf{p}^2}{2m} + V_{\text{QD}}(r), \quad (1)$$

$$V_d(r) = -\frac{e^2}{\epsilon|\mathbf{r} - \mathbf{r}_d|}. \quad (2)$$

Here the pair \mathbf{p} and \mathbf{r} are the usual momentum and coordinate in the plane of the 2D electron gas, respectively. The effective mass of the electron is denoted by m and $V_{\text{QD}}(r)$ is the QD confining potential (to be defined below), assumed to be independent of the polar angle. The hydrogenic impurity is located at a position \mathbf{r}_d from the origin and the Coulomb potential is screened by the background dielectric constant ϵ .

Inspired in our previous treatment of Coulomb forces in parabolic QDs [17], we replace the Coulomb potential $V_d(\mathbf{r})$ in (2) by an NLP to obtain the envelope function $\chi(\mathbf{r})$ from the effective-mass equation

$$(\mathcal{H}_0 + V_{\text{NL}}) |\chi\rangle = E |\chi\rangle, \quad (3)$$

where the projective operator is defined as

$$V_{\text{NL}} \equiv -\frac{\lambda \hbar^2}{2m} |v\rangle\langle v|. \quad (4)$$

Here the *shape function* $v(\mathbf{r} - \mathbf{r}_d)$ will be specified later and λ is referred to as the coupling constant. It is important to stress that this replacement is *exact*, in the sense that it is always possible to find an NLP (or a sum of them) able to reproduce any set of given electronic states [18] and, consequently, there is no theoretical limitation to the numerical accuracy with which physical results can be obtained. In spite of its seemingly more complicated form, equation (3) is amenable to analytical solution for any arbitrary shape function v , provided the eigenstates of the zero-dimensional nanostructure without the impurity are known. Therefore, one can test different functions $v(\mathbf{r} - \mathbf{r}_d)$ until the desired accuracy of the results is obtained. Typically naive functions with very few adjustable parameters are good candidates [14, 17, 19, 20].

3. General solution

As mentioned above, equation (3) can be exactly solved for any arbitrary shape function $v(\mathbf{r} - \mathbf{r}_d)$. To proceed, we consider the resolvent of the Hamiltonian \mathcal{H}_0 as follows:

$$\begin{aligned} |\chi\rangle &= -(\mathcal{H}_0 - E)^{-1} V_{\text{NL}} |\chi\rangle \\ &= \frac{\lambda \hbar^2}{2m} \sum_v \frac{|\phi_v\rangle\langle\phi_v|}{E_v - E} |v\rangle\langle v| \chi, \end{aligned} \quad (5)$$

where $|\phi_v\rangle$ refers to the eigenstates of \mathcal{H}_0 , E_v being the corresponding eigenvalues. For simplicity we assumed that the energy spectrum of \mathcal{H}_0 is discrete, although the case of a continuous spectrum can also be easily handled (see the next section). Finally, projecting onto the ket $|v\rangle$ and assuming $\langle v|\chi\rangle \neq 0$ we arrive at

$$1 = \frac{\lambda \hbar^2}{2m} \sum_v \frac{|\langle v|\phi_v\rangle|^2}{E_v - E}. \quad (6)$$

This transcendental equation provides the electron energy E in the zero-dimensional nanostructure in the presence of the impurity potential, parameterized by the shape function $v(\mathbf{r} - \mathbf{r}_d)$ and the coupling constant λ . The calculation reduces to obtain the overlap integral between the shape function and the eigenfunctions ϕ_v . For a smooth shape function, this overlap will be a decreasing function of E_v since the number of nodes of ϕ_v increases with energy. In other words, the higher the level, the smaller the numerator of equation (6) since the overlap of a slow and a fast varying function (v and ϕ_v , respectively) is smaller. Therefore, the summation over eigenstates can be safely truncated after a few terms of increasing energy if a closed expression is not available.

4. Donor-bound electron in a 2D semiconductor

Besides the shape function $v(\mathbf{r} - \mathbf{r}_d)$, the NLP is characterized by the coupling constant λ . We will now demonstrate that there exists a simple relationship between λ and the bound

state (if any) energy of the electron bound to the donor in an infinite 2D semiconductor. Therefore, the knowledge of this energy level carries information about the coupling constant. The obtained value of the coupling constant will then be used when the donor is located within a QD (section 5). If the impurity is located in an infinite 2D semiconductor, the Hamiltonian (1) reduces to $\mathcal{H}_0 = \mathbf{p}^2/2m$, namely we drop $V_{\text{QD}}(r)$. Due to the translational invariance of \mathcal{H}_0 , we can set $\mathbf{r}_d = 0$ without loss of generality in this case. The energy spectrum of \mathcal{H}_0 is continuous and the summation appearing in equation (6) is replaced by an integration in momentum space. The eigenstates of \mathcal{H}_0 are plane waves $\phi_p(\mathbf{r}) = (2\pi\hbar)^{-1} \exp(i\mathbf{p} \cdot \mathbf{r}/\hbar)$ with energy $E_p = \mathbf{p}^2/2m$. Notice that the eigenfunctions are normalized since $\langle\phi_p|\phi_{p'}\rangle = \delta(\mathbf{p} - \mathbf{p}')$.

Let us assume now that $\mathcal{H}_0 + V_{\text{NL}}$ supports a bound state with energy $E_{2\text{D}} < 0$ and define $p_{2\text{D}}^2 \equiv 2m|E_{2\text{D}}|$. From equation (6) we get

$$\frac{1}{\lambda} = \hbar^2 \int d^2\mathbf{p} \frac{|\langle v|\phi_p\rangle|^2}{p^2 + p_{2\text{D}}^2} = \frac{1}{4\pi^2} \int d^2\mathbf{p} \frac{|v(\mathbf{p})|^2}{p^2 + p_{2\text{D}}^2}, \quad (7)$$

where

$$v(\mathbf{p}) = \int d^2\mathbf{r} v(\mathbf{r}) e^{i\mathbf{p} \cdot \mathbf{r}/\hbar} \quad (8)$$

is the Fourier transform of the shape function. From this expression we can draw several conclusions. Since the right-hand side of equation (7) is positive ($p_{2\text{D}}$ is real for bound states), the NLP supports a single bound state when $\lambda > 0$. Moreover, since the right-hand side of equation (7) is obtained as an integration in momentum space, convergence implies that $|v(\mathbf{p})|$ should decay fast enough at large momenta.

For the sake of concreteness, we now focus on the *Gaussian* NLP. Gaussian shape functions were formerly considered by Knight and Peterson for band-structure calculations in solids [14]. We then take the following axially symmetric shape function

$$v(r) = \frac{1}{\pi a^2} \exp\left(-\frac{r^2}{a^2}\right), \quad (9)$$

whose Fourier transform is $v(\mathbf{p}) = \exp(-p^2 a^2/4\hbar^2)$. After performing the integration in equation (7) we arrive at

$$\lambda = 4\pi \frac{\exp(-p_{2\text{D}}^2 a^2/2\hbar^2)}{\Gamma(0, p_{2\text{D}}^2 a^2/2\hbar^2)}, \quad (10)$$

where $\Gamma(b, z)$ is the incomplete Gamma function [21].

Two free parameters (λ and a) must be set properly to account for the ground state of a hydrogenic impurity in a 2D semiconductor. The energy level is $E_{2\text{D}} = -4 \text{ Ryd}^*$, Ryd^* being the effective Rydberg. Therefore, $p_{2\text{D}} = \hbar/a_{2\text{D}}$, where $a_{2\text{D}}$ is the 2D effective Bohr radius, being half the effective Bohr radius in the bulk semiconductor. However, the estimation of the potential range a is not obvious. Previously, we have demonstrated that Yamaguchi's NLP provides nearly exact results to describe Coulomb potentials in 2D geometries [17]. The shape function of Yamaguchi's NLP is simply the Coulomb local potential times the ground state envelope function for this local potential, namely

$v_Y(\mathbf{r}) = (1/r) \exp(-r/a_{2D})$. Therefore, Yamaguchi's NLP is singular at the origin while the Gaussian NLP is regular. This difference will be important in small QDs, when the envelope function is strongly localized around the center. Unfortunately, Yamaguchi's NLP does not allow us to deal easily with off-center impurities later and we will focus only on the Gaussian NLP hereafter. In order to minimize their differences, the parameter a in (9) should be as small as possible (ideally $a \rightarrow 0$) so that the Gaussian shape function approaches the δ -function limit. We then take $a^2 = \mu a_{2D}^2$ with μ an adjustable parameter as small as possible in the numerical calculation. Finally, the coupling constant is found to be

$$\lambda = 4\pi \frac{e^{-\mu/2}}{\Gamma(0, \mu/2)} \quad (11)$$

in terms of the adjustable parameter μ .

5. Donor-bound electron in a parabolic quantum dot

After having discussed the main features of the NLP in an unbound medium, we now turn to its application to donor-bound electrons embedded in zero-dimensional nanostructures. We consider a parabolic QD with confining frequency ω and take $V_{QD}(r) = (1/2)m\omega^2 r^2$. The eigenfunctions of \mathcal{H}_0 in this case are (see, e.g., [22])

$$\phi_{n\ell}(r, \theta) = R_{n\ell}(r) \frac{e^{i\ell\theta}}{\sqrt{2\pi}} \quad \ell = 0, \pm 1, \pm 2 \dots$$

$$n = 0, 1, 2, \dots \quad (12)$$

with

$$R_{n\ell}(r) = C_{n\ell} z^{|\ell|} e^{-z^2/2} L_n^{|\ell|}(z^2), \quad (13)$$

where $z = r/\mathcal{L}$, $\mathcal{L} = \sqrt{\hbar/m\omega}$ being the QD size. $L_n^{|\ell|}(z^2)$ denotes the generalized Laguerre polynomial [21]. The corresponding eigenenergies are $E_{n\ell} = \hbar\omega(2n + |\ell| + 1)$ and the normalization constant is given by $C_{n\ell} = (1/\mathcal{L}) \sqrt{2n!/(n + |\ell|)!}$.

5.1. On-center impurity

As an illustrative example, we consider an impurity located at the center of the QD since equations are simpler. Off-center impurities are discussed later. If the impurity is located at the center, the entire nanostructure is axially symmetric. As a consequence, only s states contribute to summation (6), namely $\langle v|\phi_{n\ell}\rangle$ vanishes for $\ell \neq 0$. A straightforward calculation yields

$$\langle v|\phi_{n\ell}\rangle = \frac{\mathcal{L}}{\sqrt{\pi}a^2} \gamma_n \delta_{\ell 0}, \quad (14)$$

with

$$\gamma_n = \int_0^\infty du \exp\left[-\frac{1}{2}\left(1 + \frac{1}{\mu\beta}\right)u\right] L_n^0(u)$$

$$= \frac{2\mu\beta}{1 + \mu\beta} \left(\frac{1 - \mu\beta}{1 + \mu\beta}\right)^n, \quad (15)$$

where the shape function is given by (9). For brevity we introduced the confining parameter $\beta \equiv a_{2D}^2/2\mathcal{L}^2$. This parameter determines the magnitude of the donor-bound

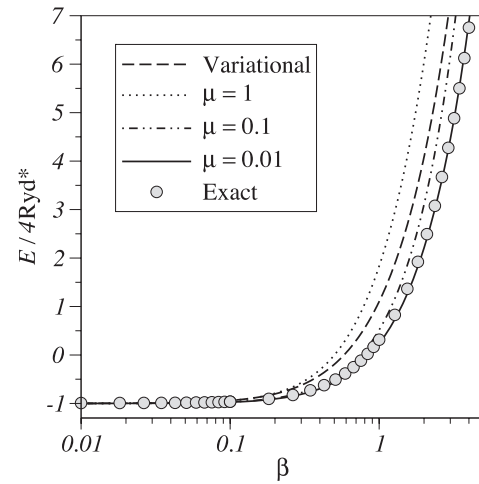


Figure 1. Energy of the ground state of a hydrogenic donor, in units of the 2D effective Rydberg (4 Ryd^*), as a function of the confining parameter $\beta = a_{2D}^2/2\mathcal{L}^2$. Different values of μ were used in (16). Circles correspond to the exact ground state energy.

electron confinement in the QD since the larger β , the higher the confinement. From (6) and (14) we finally get the following transcendental equation for the eigenenergies:

$$1 = \frac{e^{-\mu/2}}{\Gamma(0, \mu/2)} \frac{1}{(1 + \mu\beta)^2} \Phi \left[\left(\frac{1 - \mu\beta}{1 + \mu\beta} \right)^2, 1, \frac{1}{2} - \frac{E}{2\hbar\omega} \right], \quad (16)$$

where Φ is the Lerch function [23] and we have used (11).

Figure 1 shows the energy of the ground state of a hydrogenic donor, in units of the 2D effective Rydberg ($|E_{2D}| = 4 \text{ Ryd}^*$), as a function of the confining parameter β . Several values of the adjustable parameter μ were used in the calculation. The ground state energy is unchanged in large QDs ($\beta \rightarrow 0$), as compared to the donor-bound electron in a 2D semiconductor. By decreasing the QD size the ground state energy increases, as expected. The results are compared to the exact ground state energy, obtained by direct numerical diagonalization of the Hamiltonian \mathcal{H} given in (1) and (2) with $r_d = 0$. As we argued in the previous section, the smaller the parameter μ , the better the results. Large values of the parameter μ overestimate the ground state energy. It is worth mentioning that similar overestimations of the energy levels are found with more conventional approaches like the $1/N$ shifted expansion [24]. Our approach is actually free of this failure since it is always possible to select a small parameter μ for which the energy obtained from (16) matches the exact value in an arbitrary range of the confining parameter β . For instance, in figure 1 the exact results are very well reproduced by the NLP approach with $\mu = 0.01$. Notice that the largest confining parameter in this figure is $\beta = 4$, namely $\mathcal{L} \simeq 0.35a_{2D}$ (strong confinement regime, $\mathcal{L} < a_{2D}$). In GaAs $a_{2D} = 5 \text{ nm}$ and we conclude that $\mu = 0.01$ gives an excellent estimation of the ground state energy in small QDs with $\mathcal{L} = 1.75 \text{ nm}$.

We also carried out a variational calculation in order to compare it with the NLP approach. To perform the calculation, we used the following trial function, formerly introduced in [6] for donor-bound electrons in spherical QDs,

$R(r) = N(\eta) \exp(-r^2/2\mathcal{L}^2) \exp(-\eta r)$, where $N(\eta)$ is the normalization constant, λ being the variational parameter. The results are shown in figure 1. We notice that the variational approach fails in the strong confinement limit (β large), as compared to the exact results obtained by direct diagonalization. It is remarkable that the NLP approach gives more accurate results even if μ is not too small.

5.2. Off-center impurity

In the preceding paragraphs we have shown that the NLP provides accurately the energy of an electron bound to an on-center impurity. In what follows we use the same approach for a hydrogenic impurity at a position r_d from the origin of the QD. We set the X axis along the direction of r_d without loss of generality. Therefore, the shape function is now

$$v(r, \theta) = \frac{1}{\pi a^2} \exp \left[-\frac{1}{a^2} (r^2 + r_d^2 - 2rr_d \cos \theta) \right]. \quad (17)$$

The overlap between v and $\phi_{n\ell}$ appearing in (6) is given by

$$\langle v | \phi_{n\ell} \rangle = \frac{\sqrt{2\pi}}{\pi a^2} e^{-r_d^2/a^2} \times \int_0^\infty dr r e^{-r^2/a^2} R_{n\ell}(r) I_{|\ell|} \left(\frac{r_d r}{\mu \beta \mathcal{L}^2} \right), \quad (18)$$

where $I_{|\ell|}(z)$ is the modified Bessel function [21]. After some algebra we obtain

$$\langle v | \phi_{n\ell} \rangle = \frac{\mathcal{L}}{\sqrt{\pi} a^2} \gamma_n \exp \left(-\frac{r_d^2/\mathcal{L}^2}{2(1 + \mu\beta)} \right) \times \sqrt{\frac{n!}{(n + |\ell|)!}} \left(\frac{r_d/\mathcal{L}}{1 + \mu\beta} \right)^{|\ell|} L_n^{|\ell|} \left[\frac{r_d^2/\mathcal{L}^2}{1 - \mu^2\beta^2} \right], \quad (19)$$

with γ_n given by (15). Notice that this expression reduces to (14) in the limiting case $r_d \rightarrow 0$, as expected.

To get an estimation of the binding energy of the electron, we focus on the magnitude

$$E_c = E - \frac{1}{2} m \omega^2 r_d^2, \quad (20)$$

where E is obtained by solving (6). The last term in the above equation is nothing but the potential energy at the position of the donor due to the presence of the QD confining potential. Figure 2 shows the results for three different donor positions when $\mu = 0.01$. The curve corresponding to $r_d = 10^{-3}\mathcal{L}$ is the same as that given in figure 1 for $\mu = 0.01$, within the numerical uncertainty. The energy level does not change much even if the donor is located at a distance half the QD radius. Remarkably, the binding energy is found to decrease as the impurity moves apart from the center. This effect is more pronounced for smaller QDs, as seen by comparing the upper and lower curves in figure 2. Similar conclusions were drawn in spherical QDs by using a variational approach to estimate the impurity binding energy [22].

6. Conclusions

We have introduced a solvable model to obtain the energy levels of an electron bound to a hydrogenic impurity in QDs.

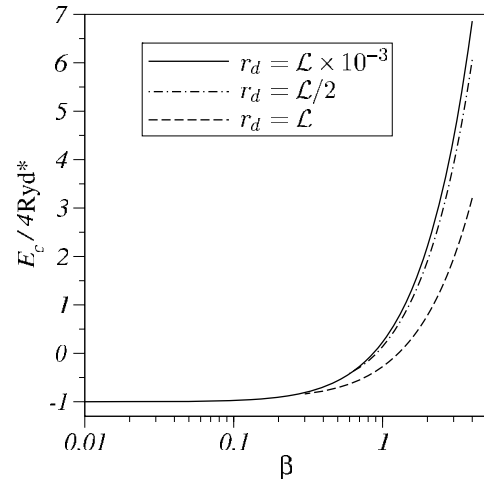


Figure 2. Energy E_c , in units of the 2D effective Rydberg (4 Ryd^*), as a function of the confining parameter $\beta = a_{2D}^2/2\mathcal{L}^2$, for different positions of the donor. $\mu = 0.01$ for all curves.

In the framework of the effective-mass approximation, the confining potential arising from the QD is assumed to be parabolic with confining frequency ω . The Coulomb potential due to the donor is replaced by an NLP. We stress again that this substitution is exact provided the appropriate shape function $v(r)$ is used. This function is nothing but the local potential times the actual envelope function [15]. Thus, the *exact* shape function $v(r)$ is known only after solving the effective-mass equation for the local potential. Therefore, the way to proceed is to solve the effective-mass equation for an *arbitrary* shape function, as we have shown in section 3. Once the solution is obtained, one can choose a potential shape function that reproduces the observed energy values of the physical system being considered. We have proven that a Gaussian NLP is suitable for describing off-center hydrogenic impurities in QDs of all sizes for which the effective-mass approach holds, provided a small value of the parameter μ is selected (ideally $\mu \rightarrow 0$). As a major result, we have shown that the binding energy is found to decrease as the impurity moves apart from the center. The decrease of the binding energy is more pronounced at large values of the confining parameter β .

Finally, we stress that the NLP approach introduced above is not restricted to parabolic QDs but other low-dimensional nanostructures could also be studied as well. Notice that the only major requirement of the NLP approach is the knowledge of the electronic states in the absence of the impurity potential, namely the eigenstates of the Hamiltonian (2). Fortunately, the electronic states of several low-dimensional nanostructures like quantum rings [25, 26] are now well known. These states are the starting point to study impurity states with little computational effort within the NLP framework.

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