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Laterally coupled jellium-like two-dimensional quantum dots

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Abstract

Many studies have been performed to describe quantum dots using a parabolic confining potential. However, infinite potentials are unphysical and lead to problems when describing laterally coupled quantum dots. We propose the use of the parabolic potential of a homogeneous density distribution within a 2D jellium, with the Coulombic potential outside that jellium. The electronic structure for such quantum dots is calculated using a wavelet-based electronic structure code, developed for this purpose. The potential is compared with its limits for infinitely small and infinitely large radii of the jellium, i.e. a 2D atom and a pure parabolic potential respectively. Finally, two such laterally coupled quantum dots are studied.

1. Introduction

Quantum dots have recently attracted much interest both experimentally and theoretically [1]. Quantum dots are human-made nanoscale structures in which electrons are confined in all three spatial directions. As they show typical atomic properties like discrete energy levels and shell structures, they are often referred to as artificial atoms. However, in contrast to natural atoms, in quantum dots the number of electrons is tunable.

Starting from quantum dots as a structure, more complex systems are conceivable and likely to have prospects in future applications. An example is the analogy of a two-atom molecule consisting of two coupled quantum dots, where the coupling can be both vertically and laterally.

Many studies have been performed to describe quantum dots using a parabolic confining potential, see for example [2–5]. However, such infinite potentials are unphysical. Furthermore, they give rise to practical problems when describing laterally coupled quantum dots. Various other potentials have been suggested that do not have this problem. A first example is a parabolic potential only from the nearest centre [6]. Such a potential enables

lateral coupling but still has the disadvantage that it goes to infinity. In order to get rid of this limit others have suggested potentials that are close to the harmonic potential at their bottom but have a smooth lateral boundary with a finite asymptotic value. Examples of such potentials are a Gaussian potential [7] and a smooth boundary potential [8].

In this paper we propose a new potential that is also harmonic at its centre and has a constant asymptote, but that is physically motivated as derived from the description of a quantum dot as a 2D jellium. The potential we propose is the parabolic potential of a homogeneous density distribution within a 2D jellium, with the Coulombic potential outside that jellium. This potential is described in section 2. Section 3 describes how the Kohn–Sham equations are used to calculate the electronic structure of such quantum dots using an in-house wavelet-based electronic structure code. This is used in section 4 to study our new potential for various radii of the jellium. These results are compared with its limit for infinitely small and infinitely large R , i.e. 2D atoms and a pure parabolic potential respectively. Finally, in section 5 a quantum dot molecule consisting of two laterally coupled quantum dots is studied.

Throughout the paper we use the donor Hartree $H_{\text{HD}} = m_e^* E_{\text{H}} / \epsilon^2$ as a unit of energy and the donor Bohr radius $a_{\text{D}} = \epsilon a_0 / m_e^*$ as a unit of length, where E_{H} is the atomic Hartree energy, a_0 is the atomic Bohr radius, m_e^* is the effective mass and ϵ is the effective dielectric constant of the host material. In case of GaAs these units are typically 10.96 meV and 101.89 Å, respectively. Further units are the electron charge e as the unit of charge and the effective electron mass m_e^* as the unit of mass.

2. Potential

The potential that we propose to study laterally coupled quantum dots is derived from the description of a quantum dot as a 2D jellium. This stems from the way quantum dots can be created experimentally. Because of the application of a gate potential, an area of electron depletion will appear near the gate, which can be modelled most simply by assuming a constant positive background (i.e. jellium). We propose the parabolic potential of a homogeneous density distribution within the 2D jellium, with the Coulombic potential outside that jellium, i.e.

$$V_e(r) = \begin{cases} -\frac{Q}{R} - \frac{Q}{4\pi R^2}(R^2 - r^2) & r \leq R \\ -\frac{Q}{r} & r > R, \end{cases} \quad (1)$$

where R is the radius of the circular jellium and Q is its total charge. The offset $-Q/R - Q/(4\pi)$ in the harmonic part is required to make the potential continuous and asymptotically go to zero at large distances. This potential has been plotted in figure 1 for $Q = 24$. The left part of the figure shows how the potential is constructed from its two parts, in this case for $R = 4$. The right part shows the potential for three different radii of the jellium.

3. Approach

The electronic structure for the 2D quantum dots is calculated by solving the Kohn–Sham equations numerically within the effective mass approximation

$$\left(-\frac{1}{2}\nabla^2 + V_e(\mathbf{r}) + V_{\text{H}}(\mathbf{r}) + V_{\text{xc}}(\mathbf{r})\right)\Psi_i(\mathbf{r}) = \epsilon_i \Psi_i(\mathbf{r}). \quad (2)$$

To solve this 2D problem we have developed a wavelet-based electronic structure calculations program, analogous to our 3D electronic structure code for molecules [9]. Briefly, the program calculates the electronic structure self-consistently. Starting from an initial guess for the

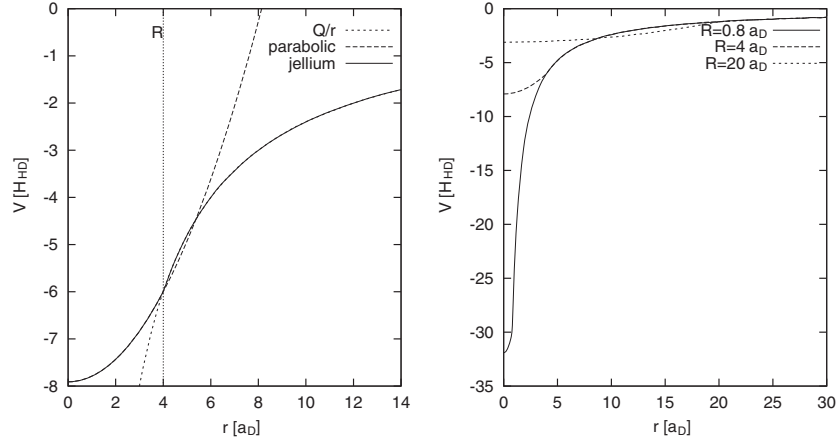


Figure 1. Left: the potential consists of two parts: quadratic within a radius R and Coulombic outside this region. Right: the potential for three different radii of the jellium, with constant charge Q .

wavefunctions, the parts of the Hamiltonian are calculated. Using this ‘fixed’ Hamiltonian, the wavefunction is improved by using a minimization scheme. Then the Hamiltonian is recalculated, the wavefunction updated, etc. This is repeated until a self-consistent solution has been reached. The choice for wavelets as the basis set allows for an accurate description over a range of length scales. This is because wavelets constitute a multiresolution approach, allowing us to use low resolution and to add extra resolution only in those regions where necessary, which is a nice property due to the well-known fact that electronic wavefunctions vary much more rapidly near the ‘atomic’ centres than in interatomic regions. For more details on the use of wavelets for electronic structure calculations see [9, 10].

The parts constituting the Hamiltonian are, however, different in 2D space from those in 3D space. The external potential describes the confinement of the quantum dot. We use the one proposed in equation (1) and the simple harmonic one for comparison. A 2D version of the exchange and correlation energy functionals is given by Tanatar and Ceperley [11]. They suggest the use of the local density approximation (LDA) for the exchange potential. In 2D form this potential is [12]

$$V_x(\mathbf{r}) = -\sqrt{\frac{8\rho_e}{\pi}}. \quad (3)$$

The correlation potential is the functional derivative of their correlation energy functional, yielding

$$V_c(\mathbf{r}) = \frac{a_0(6a_1t^2 + 7t^3)}{4(a_3 + a_2t + a_1t^2 + t^3)} - \frac{a_0t^2(a_2t + 2a_1t^2 + 3t^3)(a_1 + t)}{4(a_3 + a_2t + a_1t^2 + t^3)^2} \quad (4)$$

where $t = (\pi\rho_e)^{1/4}$ and the parameters a_0 , a_1 , a_2 and a_3 were taken from table 4 of [11]. The Hartree potential V_H is calculated using a 2D version of Chelikowsky’s direct integration method [13] that we have developed. Using this method, the Hartree potential V_H is evaluated on a grid by assuming the integrand does not change appreciably within a square of area h^2 around each grid point i, j . V_H is given by

$$V_H(x_i, y_j) = \sum_{i', j'} \rho_e(x_{i'}, y_{j'}) g(x_i - x_{i'}, y_j - y_{j'}), \quad (5)$$

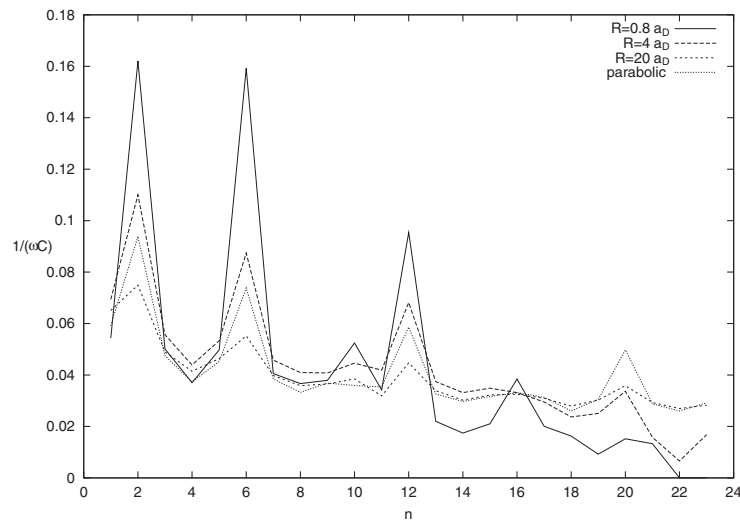


Figure 2. Scaled inverse of the capacitance for different confining potentials as a function of the number of electrons.

where, for $i, j \neq i', j'$,

$$g(x_i - x_{i'}, y_j - y_{j'}) = \frac{h^2}{\sqrt{(x_i - x_{i'})^2 + (y_j - y_{j'})^2}}. \quad (6)$$

For $i, j = i', j'$, i.e. near the square root singularity, an explicit integration over the square yields

$$g(0, 0) = 2h \ln\left(\frac{\sqrt{2} + 1}{\sqrt{2} - 1}\right). \quad (7)$$

4. Single quantum dot

The confining potential we proposed in equation (1) has two interesting limits. First, for $R \rightarrow \infty$ we get the harmonic potential

$$V_e(r) = -\frac{Q}{4\pi} + \frac{1}{2}\omega^2 r^2, \quad (8)$$

where ω is related to the jellium charge density $\rho = Q/\pi R^2$ via

$$\omega = \sqrt{\frac{\rho}{2}}. \quad (9)$$

In this limit we return to the parabolic confining potential that has been studied extensively. Second, for $R \rightarrow 0$ the potential reduces to

$$V_e(r) = -\frac{Q}{r}, \quad (10)$$

which is the potential for 2D atoms [14, 15].

These two limits have a different shell structure. For the parabolic potential the shell will be filled with 2, 6, 12, 20, ... electrons, whereas for the 2D atoms these numbers are 2, 8, 14, 24, ... This shell structure can be visualized using the capacitance C :

$$C(n) = \frac{1}{\mu(n+1) - \mu(n)}, \quad (11)$$

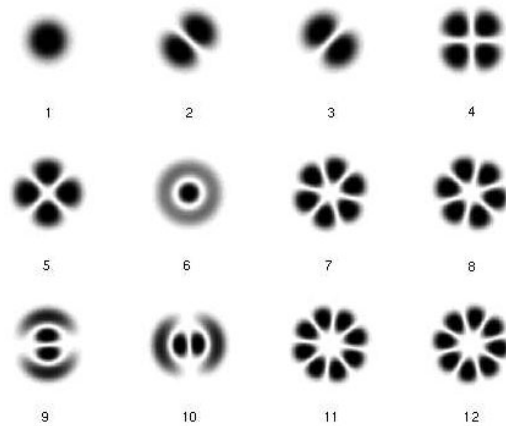


Figure 3. A contour plot of the first 12 orbitals $\Psi_i(r)$ for the parabolic confining potential.

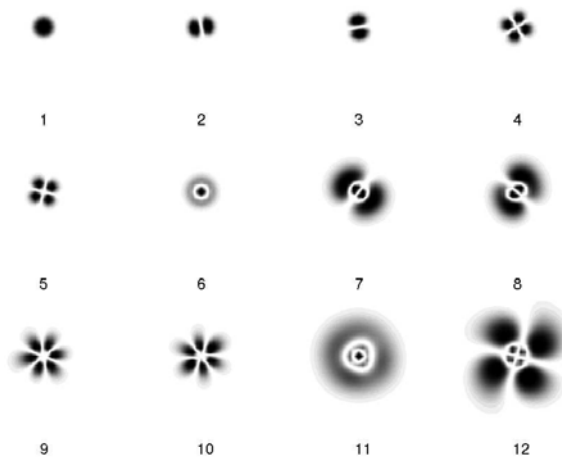


Figure 4. A contour plot of the 12 orbitals $\Psi_i(r)$ for the potential with $R = 0.8a_D$ and $Q = 24e$.

where n is the number of electrons in the quantum dot and μ is the chemical potential $\mu(n) = E(n) - E(n - 1)$. The filling of a shell will result in a peak in the inverse of this capacitance. This inverse capacitance has been plotted in figure 2 for the parabolic potential ($R \rightarrow \infty$) and for three different values of R , each a factor 5 apart, i.e. $20a_D$, $4a_D$ and $0.8a_D$. For the choice $Q = 24e$, these radii correspond, according to equation (9), to the strength ω of the parabolic potential near the centre of 1.07, 5.36 and 26.78 meV respectively, which are numbers consistent with experimental values. For the pure parabolic potential the value 1.07 meV was used. As can be seen in the figure, for $R = 20a_D$ the shell filling is similar to the case of the pure parabolic potential, for $R = 4a_D$ the difference between the higher orbitals is already larger and for $R = 0.8a_D$ the distinction can be seen by a movement of the peak in the inverse capacitance from $n = 20$ to 16.

The orbitals corresponding to the parabolic potential and to the potential with $R = 0.8a_D$ are plotted in figures 3 and 4 respectively. Because the orbitals corresponding to the parabolic potential are smaller, the scale of the latter figure is twice as small as that of the former. As expected, the innermost orbitals are very similar. The higher orbitals, however, are much wider for the potential with $R = 0.8a_D$, since these electrons are much less confined.

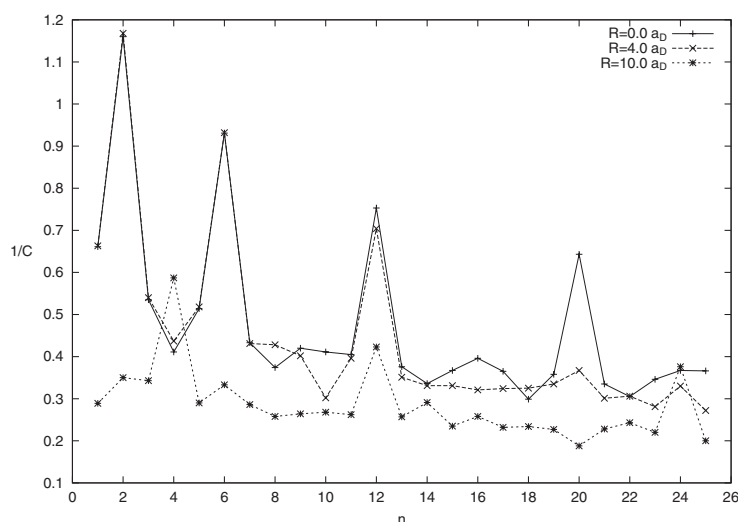


Figure 5. Inverse of the capacitance for different inter-centre distances as a function of the number of electrons.

5. Laterally coupled quantum dots

Because of the asymptotic behaviour of the potential, where the potential goes to zero for large distances, arrays of quantum dots can be modelled straightforwardly. For example, two laterally coupled quantum dots form a quantum dot molecule.

Here, we will consider two quantum dots with charge $Q = 28e$ and radius $R = 4a_D$, corresponding to a parabolic potential of 5.78 meV. These quantum dots are studied as a function of the distance D between their centres, where the two quantum dots contain 12 electrons each.

For $D = 0$ the two quantum dots are centred at the same position and will therefore form just one quantum dot with the same radius but twice as high a charge density and thus twice as strong a confinement. For such a quantum dot, the first 12 orbitals will have the same form as with the pure parabolic potential; thus the orbitals are filled at 2, 6, 12 and 20 electrons.

This can be seen again using the inverse of the capacitance, which is shown in figure 5. In this figure it can also be seen that if the inter-centre distance D increases, quantum dot molecular orbitals come into existence with different shell structure. For $D = 10.0a_D$ the first three peaks in the inverse of the capacitance are at 4, 12 and 24 respectively. The orbitals for this case are shown in figure 6. In between, for $D = 4.0a_D$, both peaks at 20 and 24 can just be discerned. If the distance between the quantum dots increases further, the two quantum dots will be separated, resulting in two single quantum dots, which were described in the previous section.

In figure 7 the energy of this system of two quantum dots, with a total of 24 electrons, has been plotted as a function of the distance D between the two centres of the dots. The two limits of the energy as a function of the inter-centre distance of the quantum dots, i.e. for $D = 0$ and $D \rightarrow \infty$, can be understood using the 2D pure harmonic oscillator, i.e. without the electron–electron interaction. Because in both limits only the lower orbitals are filled, these orbitals are very similar to the solution of a harmonic oscillator up to an offset $-Q/R - Q/(4\pi)$, see equation (1). Such an offset does not change the shape of the eigenfunctions, it just causes a

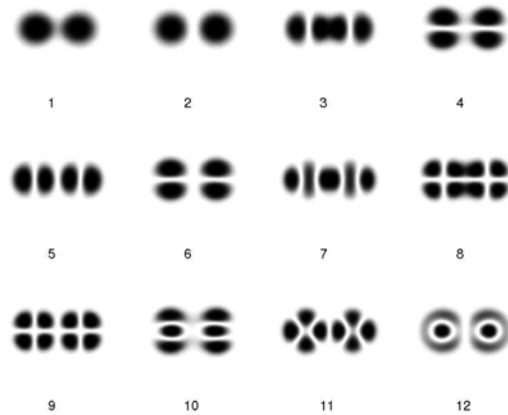


Figure 6. First 12 orbitals for two quantum dots at distance $D = 10.0a_D$.

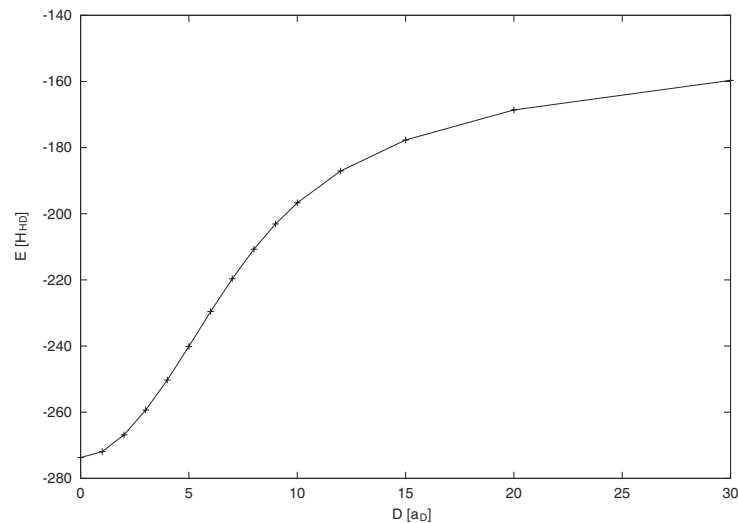


Figure 7. The energy E of the two quantum dot system versus the inter-centre distance D .

change with the same value in the energy eigenvalues, which are well known for the 2D pure harmonic oscillator to be $m\omega$ with degeneracy m , for $m = 1, 2, 3, \dots$, where each orbital can occupy two electrons. Thus, in terms of ω the lowest energy eigenvalues of one quantum dot are $m\omega - \frac{1}{2}(4\pi R + R^2)\omega^2$ with degeneracy m .

One single quantum dot thus makes two contributions to the total energy: first, the contribution of the harmonic oscillator $2 \cdot (1 \cdot 1 + 2 \cdot 2 + 3 \cdot 3)\omega = 28\omega$; and second, the offset, which for our Q and R is equal to $12 \cdot [-8(\pi + 1)\omega^2]$. Because $Q = 28$, and $R = 4$, $\omega = \sqrt{7/(8\pi)}$. Thus the total energy is equal to $28\sqrt{7/(8\pi)} - 84(\pi + 1)/\pi$, which is approximately equal to $-96H_{HD}$. Two such quantum dots infinitely far apart therefore yield $E \approx -192H_{HD}$, when the electron–electron interaction is neglected.

The other limit is where the two quantum dots are centred at the same place. This yields one quantum dot with the same radius but with a charge twice as high, and twice as many electrons, i.e. 24. Thus, again, there are two contributions to the total energy, namely

$2 \cdot (1 \cdot 1 + 2 \cdot 2 + 3 \cdot 3 + 4 \cdot 4 + 2 \cdot 5)\omega = 80\omega$, and an offset $24 \cdot [-8(\pi + 1)\omega^2]$, where $\omega = \sqrt{7/(4\pi)}$. The total energy is therefore equal to $80\sqrt{7/(4\pi)} - 336(\pi + 1)/\pi$, which is approximately equal to $-383H_{\text{HD}}$.

The discrepancy between these limits and the results in figure 7 can be explained by the omission of electron–electron interactions in the calculation of the limits. Because these electron–electron interactions are dominated by repulsion and make a (quadratically) larger contribution to the total energy for higher electron densities, the discrepancy is much larger for the ‘fused’ dot ($D = 0$) than for the infinitely far, separated dots ($D \rightarrow \infty$).

The limit for the electronic energy for infinite dot separation can be calculated in a second way as well, i.e. as twice the energy of one such single dot. For our dots this results in $-142H_{\text{HD}}$. It has been confirmed that the approach towards this limit in figure 7 is, as expected, $(-2nQ + nn)/D$.

6. Conclusion

In conclusion, we have studied the electronic states of quantum dots, solving the Kohn–Sham equations in the effective mass approximation using a model potential for a 2D jellium. For this purpose a 2D electronic structure program has been developed. Specific to this program are the use of wavelets as a basis and the evaluation of the Hartree potential using a 2D version of Chelikowsky’s direct integration method which we developed. The orbitals have been shown for single quantum dots and the filling of the shells has been compared with the case of a parabolic model potential. By laterally coupling quantum dots, which can be done straightforwardly using our potential, quantum dot molecules have been modelled.

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