Electronic States in Semiconductor Quantum Dots

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Ottobre 1999
a Philip Marlowe, Ciccio Ingravallo,
Santamaria e tutti gli altri
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Introduction

The experimental and theoretical study of electronic systems was limited for a long time to naturally occurring atoms, molecules and crystals. Ideal crystals are three-dimensional systems, while an effective reduction of geometry to lower dimensions — by a strong spatial localization, namely confinement of an electron in at least one direction at the de Broglie wavelength — occurs only in the case of crystal imperfections.

The beginning of the 70s marked the new era of research on electronic structures of lower dimensions. The first to appear were quantum wells[1, 2]. A quantum well is a thin, flat layer of semiconductor sandwiched between two layers of another semiconductor with larger bandgap. Owing to this bandgap difference, the conduction and valence band edges of the two materials do not align with each other (band off-set). The band off-set produces the potential responsible for confining the carriers in the thin layer. In a semiconductor the de Broglie wavelength is relatively large, hence the motion of electrons bound in a layer as thin as several crystalline monolayers is quasi-two-dimensional, and the excitations in the perpendicular (growth) direction are strongly quantized. A material that is commonly used for creating quantum wells is GaAs, which in combination with a ternary solution Al$_x$Ga$_{1-x}$As serving as a barrier allows for the creation of epitaxial layers with high crystalline quality as a result of the almost equal lattice constants.

The new, unusual properties of quasi-two-dimensional systems, which promised applications in electronics and opto-electronics, attracted the attention of many research laboratories. This in turn resulted in a rapid development of production technology and extensive research, which culminated in the discovery of the Quantum Hall Effect[3] in 1979. At present, many properties of the quasi-two-dimensional systems are well investigated and understood, and quantum wells have been produced and implemented for years in numerous devices, such as laser diodes used in CD players or microwave receivers used in satellite television.

Since the beginning of the 80s, the rapid progress in technology, including very accurate lithographic techniques, possibly combined with self-organized growth techniques, made it possible to confine electrons in quantum wires, i.e. quasi-one-dimensional structures. Quantum wires are produced, for example, in the form of miniature strips, etched in a sample containing a quantum well, or in V- and T-shapes obtained by controlled overgrowth processes[4].

Complete quantization of the electron’s free motion is implemented by trapping it in a quasi-zero-dimensional quantum dot (QD). For example, Reed et al.[5] at Texas Inst. reported the creation of a square QD with a side length of 250 nm, etched by lithography. Subsequent publications reporting the creation of QDs in other laboratories soon appeared[6, 7, 8], with diameters already reduced to 30-45 nm.

The name “dot” suggests an exceedingly small region of space. A semiconductor QD, however, is made out of roughly a million atoms with all their electrons. Virtually all electrons are tightly bound to the nuclei of the material, however, and the number of free electrons in the dot can be very small: between one and a few hundreds. The de Broglie wavelength of these electrons is comparable to the size of the dot, and the electrons occupy discrete quantum levels (akin to atomic orbitals in atoms) and have a discrete excitation spectrum. If the number of free electrons $N_e$ is large, it is reasonable to expect that their electronic properties may be qualitatively described by appealing to known properties of the infinite ($N_e \rightarrow \infty$) electron gas. However for small numbers of electrons (e.g. $N_e < 20$) the electronic properties of the
system are likely to be strongly $N_e$-dependent, particularly if the dot is small. A characteristic quantity for QDs is the addition energy, which is analogous to the ionization energy of an atom. This is the energy required to add or remove a single electron from the dot. It is a finite, measurable quantity that depends on $N_e$ and arises from quantization of charge in units of $e$.

Because of the analogies to real atoms, QDs are frequently referred to as artificial atoms[9]. As in real atoms, electrons are attracted to a central location. In a natural atom, this central location is a positively charged nucleus; in an artificial atom, electrons are typically trapped in a bowl-like parabolic potential well in which they tend to fall in towards the bottom of the bowl. Moreover, QDs can be coupled one to each other, if the heterostructure barriers are sufficiently thin, thus one can extend the analogy with "natural" systems speaking of artificial molecules.

The basic technological motivation to study QDs is that smaller components should be faster and may also dissipate less heat. There is also the added feature that quantum-mechanical effects are so important in such systems that devices with fundamentally new properties could be obtained. In this perspective a relevant example are single electron transistor devices. Otherwise, since QDs absorb and emit light in a very narrow spectral range, they might find application in the construction of more efficient and more controllable semiconductor lasers. The strong quantization of electron energy, with parameters suitable for laser action, particularly in the so-called self-assembled QDs, will probably allow QD-based lasers to work at higher temperatures and at lower injection currents[10]. The small dimensions and the possibility of dense packing of QD-matrices could also permit them to be used for computer memory media of huge capacity. In addition, an intriguing challenge that is now being explored is the possibility to use QDs for implementing quantum information devices[11].

Apart from their potential use as novel devices, QDs are interesting from a fundamental point of view. First of all it is possible to control their shape, their dimensions, the structure of energy levels, and the number of confined electrons $N_e$. Applying external fields generated by standard laboratory sources transitions never observed in the spectra of "natural" atoms can be readily seen in artificial ones. Second, the energy spectrum of the few-electron QD is expected to be extremely rich since the single-electron confinement energy, the cyclotron energy for modest fields and the electron-electron interaction energy can all be of similar magnitude (typically a few meV), and they scale differently as far as one varies dot parameters. The energy spectrum is also likely to be strongly $N_e$-dependent. Of particular interest is the investigation of small-$N_e$ precursors of exotic many-body states such as fractional-quantum-Hall-effect states[12] and Wigner solids[13]. To summarize, QDs are an ideal laboratory to investigate electronic correlation, and this is the issue the present Thesis is mainly addressed to.

Current experiments concerned with QDs focus mainly on studying their optical properties (absorption and emission of light in the visible or far infrared range, and the Raman scattering of light) and electric properties (capacitance and transport studies). QDs are therefore artificial atoms with the additional intriguing possibility of connecting source and drain contacts to probe their atomic states.

In this Thesis we investigate the electronic structure of few-electron QDs. In such systems the strongly correlated electronic states show a rich phenomenology and require a many-body approach. We study the dependence of electronic configurations on the variation of external tunable parameters such as
an applied homogeneous magnetic field, the geometry of the dot, the coupling to another QD, and the number of electrons. Approximate theoretical schemes are also discussed and compared, and the role of electronic correlation is analyzed.

We focus on the so-called vertical QDs[14, 15], single and coupled. These structures, to be described later, have abrupt and thin tunnel barriers, present a relatively low rate of disorder (impurities, electrode potential fluctuations, etc), and are sufficiently well defined to allow for the formation of systematically degenerate sets of electronic energy levels. We will see that the presence of degeneracies (or, equivalently, of symmetries) has important consequences including: (i) The appearance of an electronic shell structure reminiscent of periodic table of elements of atomic physics. (ii) Ground state configurations with non-zero values of magnetization and spin.

As mentioned above, transport measurements (Single-Electron-Tunneling- and Capacitance-Spetrosopies) are a direct spectroscopic tool to probe ground state energies of few-particle, strongly correlated artificial atoms and molecules. The relevant quantity measured, that is the addition energy, will be easily extracted from our calculations and then compared with experimental findings.

The structure of the present Thesis is the following:

- In Chapter 1 we briefly review construction and measurement methods of QDs. After recalling the simplest phenomenological models for electron transport through dots (Coulomb blockade), basic single electron experiments are presented. The Chapter is a primer about the concept of addition spectrum which is essential throughout this work. It ends with a survey of existent theoretical work.

- In Chapters 2 and 3 we illustrate the theoretical framework for the computation of many-body electronic states. After an analysis of the model system, Chap. 2 is concerned with single-particle states, including mean-field solutions of the many-body problem. The Self-Consistent methods are presented and discussed. In Chap. 3 we introduce a many-body approximate Hamiltonian to correctly treat the electronic correlation. The Wigner-von Neumann theorem which rules over transitions between different electronic configurations is reviewed.

- Chap. 4 displays our results on the addition spectra of single QDs. The main points are the evidence of an electronic shell structure and spontaneous magnetization and spin-polarization brought about by electrostatic and exchange effects (Hund’s rules). Data agree with measurements in presence of an external magnetic field.

- Chap. 5 is a critique of the results of the previous Chapter. The influence of dot geometry on the addition energy is analyzed, different theoretical approaches such as many-body Hamiltonian, Hartree-Fock method and exact diagonalization technique are compared, and the importance of electronic correlation in the approximations introduced is discussed.

- Chap. 6 analyzes the energy spectrum of an artificial molecule. Here we vary also the coupling between dots: unexpected configurations are found in intermediate regimes, in addition to those pertaining to the two limiting cases of two well separated QDs or a unique, strongly connected system, respectively.

- Final conclusions stress the main findings of this Thesis.
The main reviews on the subject are Refs. [16, 17, 18, 19, 20]. Ref. [21] is an introduction to single charge tunneling.
Chapter 1

Quantum Dots: a Survey

QDs are the object of our investigation, atom-like systems whose electronic properties are probed by single-electron transport spectroscopies. This Chapter has two main objectives: first, it aims at reviewing basic experimental facts and at elucidating the concept of Coulomb blockade and single-electron spectroscopy. Second, it surveys existent electronic structure studies. We start reminding the conditions to observe energy and charge quantization, whose experimental evidences are the so-called Coulomb oscillations (Section 1.1). After, we describe some state-of-the-art QD devices together with the main transport measurement techniques (Sec. 1.2). A simple theory of electron transport through dots is outlined in Sec. 1.3. Section 1.4 is a survey of theoretical works on QD electronic properties.

1.1 Energy and Charge Quantization

In this Section we examine the circumstances under which Coulomb charging and energy quantization effects are important.

1.1.1 Charge quantization

Let us consider the electronic properties of the small conductor depicted in Fig. 1.1 (a), which is coupled to three terminals. Particle exchange can occur with only two of the terminals, as indicated by the arrows. These source and drain terminals connect the small conductor to macroscopic current and voltage meters. The third terminal provides an electrostatic or capacitive coupling and can be used as a gate
Figure 1.1: Schematic of a quantum dot, in the shape of a disk, connected to source and drain contacts by tunnel junctions and to a gate by a capacitor. (a) shows the lateral geometry and (b) the vertical geometry.

electrode. If we first assume that there is no coupling to the source and drain contacts, then the small conductor acts as an island for electrons. The number of electrons on this island is an integer $N_e$, i.e. the charge on the island is quantized and equal to $N_e e$. If we now allow tunneling to the source and drain electrodes, then the number of electrons $N_e$ adjusts itself until the energy of the whole circuit is minimized.

When tunneling occurs, the charge on the island suddenly changes by the quantized amount $e$. The associated change in the Coulomb energy is conveniently expressed in terms of the capacitance $C$ of the island. An extra charge $e$ changes the electrostatic potential by the addition energy $e^2 / C$. This addition energy becomes important when it exceeds the thermal energy $k_B T$. A second requirement is that the barriers are sufficiently opaque such that the electrons are located either in the source, in the drain, or on the island. This means that quantum fluctuations in the number $N_e$ due to tunneling through the barriers is much less than one over the time scale of the measurement (this time scale is roughly $e$ divided by the cur-
1.1 Energy and Charge Quantization

This requirement becomes a lower bound for the tunnel resistances $R_t$ of the barriers. To see this, consider the typical time to charge or discharge the island $\Delta t = R_tC$. The Heisenberg uncertainty relation: $\Delta E \Delta t = (e^2/C) R_tC \geq \hbar$ implies that $R_t$ should be much larger than the resistance quantum $\hbar/e^2 = 25.813$ kΩ in order for the energy uncertainty to be much smaller than the addition energy. To summarize, the two conditions for observing effects due to the discrete nature of charge are[19]:

$$R_t \gg \hbar/e^2, \quad (1.1)$$

$$e^2/C \gg k_B T. \quad (1.2)$$

The first criterion can be met by weakly coupling the dot to the source and drain leads. The second criterion can be met by making the QD small (the capacitance of an object scales roughly with its radius $R$).

1.1.2 Coulomb Oscillations and Coulomb Staircase

While the tunneling of a single charge changes the electrostatic energy of the island by a discrete value, a voltage $V_g$ applied to the gate (with capacitance $C_g$) can change the island’s electrostatic energy in a continuous manner. In terms of charge, tunneling changes the island’s charge by an integer while the gate voltage induces an effective continuous charge $q = C_g V_g$ that represents, in some sense, the charge that the dot would like to have. This charge is continuous even on the scale of the elementary charge $e$. If we sweep $V_g$ the build up of the induced charge will be compensated in periodic intervals by tunneling of discrete charges onto the dot. This competition between continuously induced charge and discrete compensation leads to so-called Coulomb Oscillations in a measurements of the current as a function of gate voltage at a fixed source-drain voltage. This is the essence of the Single Electron Tunneling Spectroscopy (SETS).

An example of a SETS measurement[22] is shown in Fig. 1.2 (a). In the valley of the oscillations, the number of electrons in the dot is fixed and necessarily equal to an integer $N_e$. In the next valley to the right the number of electrons is increased to $N_e + 1$. At the crossover between the two stable configurations $N_e$ and $N_e + 1$, a “charge degeneracy”[23] exists where the number can alternate between $N_e$ and
Figure 1.2: (a) An example of a measurement of Coulomb oscillations to illustrate the effect of single electron charges on the macroscopic conductance. The conductance is the ratio $I/V_{sd}$ and the period in gate voltage $V_g$ is about $e/C_g$[22]. (b) An example of a measurement of the Coulomb staircase in $I - V_{sd}$ characteristic. The different curves have an offset for clarity ($I = 0$ occurs at $V_{sd} = 0$) and are taken for five different gate voltages to illustrate periodicity in accordance with the oscillations shown in (a)[24].
\( N_e + 1 \). This allowed fluctuation in the number (i.e. according to the sequence \( N_e \rightarrow N_e + 1 \rightarrow N_e \rightarrow \ldots \)) leads to a current flow and results in the observed peaks.

An alternative measurement is performed by fixing the gate voltage, but varying the source-drain voltage \( V_{sd} \) \((\text{non-linear transport})\). As shown in Fig. 1.2 (b)[24] one observes in this case a non-linear current-voltage characteristic exhibiting a \textit{Coulomb Staircase}. A new current step occurs at a threshold voltage \( (\simeq e^2 / C) \) at which an extra electron is energetically allowed to enter the island. It is seen in Fig. 1.2 (b) that the threshold voltage is periodic in gate voltage, in accordance with the Coulomb oscillations of Fig. 1.2 (a).

### 1.1.3 Energy Quantization

Electrons residing on the dot occupy quantized energy levels. To be able to resolve these levels, the energy level spacing \( \Delta E \) must be greater than the thermal noise:

\[
\Delta E \gg K_B T.
\]

The level spacing at the Fermi energy for a box of size \( L \) depends on the dimensionality. Including spin degeneracy, we have:

\[
\Delta E = (N_e/4) \hbar^2 \pi^2 / mL^2 \quad \text{1D}, \quad (1.3)
\]

\[
\Delta E = (1/\pi) \hbar^2 \pi^2 / mL^2 \quad \text{2D}, \quad (1.4)
\]

\[
\Delta E = \left(1/3 \pi^2 N_e\right)^{1/3} \hbar^2 \pi^2 / mL^2 \quad \text{3D}. \quad (1.5)
\]

The characteristic energy scale is thus \( \hbar^2 \pi^2 / mL^2 \). The level spacing of a 100 nm 2D dot is \( \sim 0.03 \text{ meV} \), which is large enough to be observable at dilution refrigerator temperatures of \( \sim 100 \text{ mK} = \sim 0.009 \text{ meV} \). In addition, electrons in semiconductor heterostructures have a small effective mass which further increases the level spacing. As a results, semiconductor QDs are true artificial atoms, with both observable quantized charge states and quantized energy levels. Using 3D metals to form a dot, one needs to make dots as small as \( \sim 5 \text{ nm} \) in order to observe atom-like properties.

The fact that quantization of charge and energy can drastically influence transport through a QD is demonstrated by the Coulomb oscillations in Fig. 1.2 (a) and
the Coulomb staircase in Fig. 1.2 (b). One can obtain spectroscopic information about the charge state and energy levels of the interacting QD electrons by analyzing the precise shape of the Coulomb oscillations and the Coulomb staircase. In this way, single electron transport can be used as a spectroscopic tool.

1.2 Construction and Measurement

We focus here on results from two techniques[25, 26, 27, 28] which have particularly affected our understanding of QDs. These methods are unique in allowing extraordinarily high-resolution spectroscopy of QDs. The energy resolution of these spectroscopies is limited solely by the sample temperature.

1.2.1 Single-Electron Capacitance Spectroscopy

Fig. 1.3 (a) shows schematically the type of sample used in one of these experiments. The QD is located between two capacitor plates. It is close enough to one of the plates to allow single electrons to tunnel between the QD and the nearby plate. The artificial atom is far enough from the other capacitor plate to prohibit tunneling to this plate.

Fig. 1.3 (b) shows how the sample is actually realized. A crystal wafer is grown layer by layer, starting from the bottom of the diagram. The first layer is a silicon-doped GaAs layer. The silicon doping makes this layer metallic, and it acts as the bottom capacitor plate of Fig. 1.3 (a). Then a thin (10-nm) AlGaAs barrier layer is grown. This barrier is thin enough that electrons can leak through it. Above that, there is a GaAs quantum well, and grown on the top of the well is a thick (not leaky) AlGaAs barrier. On top of the crystal wafer, chromium is deposited to form the top capacitor plate, which is referred to as the “gate”. Additional sample processing on the sample surface is used to create a gate which laterally confines electrons in the quantum well below, creating a QD.

Electric fields can be created by applying a voltage between the plates of the capacitor. If the top plate (gate) is made positive compared to the bottom one, electrons from the bottom plate will be attracted in the direction of the top plate, towards the artificial atom. Single electrons can thus be coaxed to tunnel into the
Figure 1.3: (a) Schematic diagram of a quantum dot located between two capacitor plates. The artificial atom is actually two-dimensional; the bowl-like shape is to represent the force tending to move electrons to the centre of the dot. (b) Diagram of the sample used in Single-Electron Capacitance Spectroscopy (SECS) experiments in a crystal grown using molecular-beam epitaxy. The QD is the black disk in the quantum well. (c) Capacitance of the sample containing the QD as a function of the top plate (gate) voltage. The first peak on the left represents the first electron entering the QD[18].
dot or expelled from it through the application of voltages on the top plate. After each electron is added to the dot, additional gate voltage is usually needed to coax another electron onto the dot. There are therefore specific gate voltages at which an electron is added to the dot, and it is this spectrum of voltages which is known as *addition spectrum*.

The motion of the single electrons into or out of the dot can be detected using a simple physical principle. When a single electron tunnels into the QD, it moves closer to the top plate of the capacitor, and electrons in the top plate tend to be pushed away from the plate; that is, some charge is induced on the top plate. In these samples, the amount of charge induced is about half of an electron’s charge. A specialized transistor enables detection of this charge and thereby allows measurement of the gate voltages at which single electrons were added to the QD. A small a.c. voltage of frequency around 100 kHz is added to the d.c. gate voltage. When the gate voltage is adjusted to a voltage at which an electron can be added to the dot, the a.c. voltage causes the electron to tunnel back and forth between the dot and the bottom plate. At these gate voltages, charge appears synchronously with the a.c. voltage. A synchronous detector then registers a signal only at these voltages, yielding a peak in the response. The scheme measures the capacitance signal due to a single electron and is known as Single-Electron Capacitance Spectroscopy (SECS).

Results from this method, taken on a sample cooled to 0.3K, are shown in Fig. 1.3 (c). The first peak on the left of the graph corresponds to the first electron in the QD. Subsequent peaks correspond to additional electrons added to the QD. The peak structure is completely reproducible as one scans the gate voltage up and down, and the widths of the peaks directly reflect the temperature of the sample.

### 1.2.2 Single-Electron Tunneling Spectroscopy

One structure commonly studied by means of SETS, which will be referred to as a *planar* QD, is created by patterning several metal electrodes, or gates, on the surface of a two-dimensional electron gas heterostructure, in a similar manner to the construction of the SECS device we examined above. Fig. 1.4 depicts the structure of a representative planar QD. A negative voltage applied to a gate raises the
Figure 1.4: Quantum dot on the intersection of electrodes; four internal electrodes localize the electrons, and four external ones serve as contacts for the electron tunneling to and from the dot[29].

electrostatic potential in its neighborhood and, typically around -0.5 V, depletes the underlying two-dimensional electron gas in the vicinity of the gate. Consequently, under suitable biasing conditions, a small region of electron gas remains at the center of the structure, and is isolated from the remainder of the electron gas.

We have already outlined the SETS experiment in Subsection 1.1.2: like the SECS method, this experiment also measures the gate voltages at which electrons are added to the dot. The detection method is however completely different. Two current leads are in such proximity to the dot that electrons may tunnel between the leads and the dot. A small voltage difference is applied between the left and right leads, and the experiment consists of monitoring the current through the QD as the gate voltage is varied. Owing to the simple geometry of these structures, this energy change varies in linear proportion to the gate voltage. Multiplying $V_g$ scales in Fig. 1.2 (a) or Fig. 1.3 (c) by a geometric factor converts this scale into an energy scale for the QD.
However, SETS experiments in planar QDs have some serious shortcomings. The tunneling barriers which form a QD do not have a well defined shape, and are strongly influenced by the voltage applied to the gates. Consequently, it is not possible to investigate the regime where $N_e$ approaches zero. Furthermore, the zero-dimensional quantization energy is much smaller than the “classical” addition energy $e^2/C$. Therefore, the Coulomb oscillations are usually very periodic, and quantum mechanical many-body effects which lead to aperiodicity in the addition spectra cannot be observed. An alternative approach is to investigate SETS with a *vertical* geometry[14, 27, 30, 31, 32].

### Vertical Quantum Dots

In these structures current flows vertically with respect to the heterostructure layers. Like in their planar counterparts, in these vertical QDs the electrons are confined by the combination of the heterostructure layers providing vertical confinement and lithography to provide in-plane confinement; however, the details of fabrication are quite different. Here no in-plane tunneling is required, allowing strong lateral confinement which is achieved by eliminating all but a narrow pillar in a modified double-barrier (single QD) or triple-barrier (double dot) tunneling heterostructure.

Fig. 1.5 (a) shows a schematic diagram of a gated vertical QD made from a double-barrier heterostructure (DBH)[33]. The DBH consists of an undoped 12.0-nm In$_{0.05}$Ga$_{0.95}$As well and undoped Al$_{0.22}$Ga$_{0.78}$As barriers of thickness 9.0 and 7.5 nm. The source and drain contacts are made from *n*-GaAs and are lightly doped close to the DBH. The DBH is processed to form a mesa with a top contact geometrical diameter of 0.5 $\mu$m through the use of a combined dry and wet etch to a point just below the DBH region. The effective diameter for electron conduction is much smaller than the geometrical dot diameter, and is estimated to be approximately 0.1 $\mu$m when the dot contains a few electrons. A circular Schottky gate is placed on the side of the mesa close to the DBH. The cross-sectional view of the device appears in the scanning electron micrograph in Fig. 1.5 (b). The dot in this device is strongly confined by the heterostructures in the vertical direction and is parabolically confined by the depletion potential in the lateral direction.
Figure 1.5: (a) Schematic diagram of the gated QD device. (b) Scanning electron micrograph of the cross section of the device. The white bar indicates the position of the double-barrier heterostructure[33].
The Schottky gate is suitable for squeezing the lateral size of the dot or to change the number of electrons $N_e$ in the dot without breaking the rotational symmetry. The inclusion of In in the well reduces the conduction band edge below the Fermi level of the contacts. This allows to study linear transport through a vertical QD without doping the barriers.

Double-dot vertical structures[34] are alike, except that a triple barrier replaces the double barrier heterostructure. Fig. 1.6 schematically depicts a vertical double QD structure. The resulting conduction band profile at zero bias is shown pictorially in Fig. 1.7, along with the heterostructure parameters.

In this short review of QD devices we have completely neglected other important types of semiconductor QDs such as semiconductor microcrystals or the so-called “self-assembled” QDs. These devices can be characterized by strong spatial confinement and consequently high values of quantization energies ($\sim 50 – 100$ meV). The interested reader can refer to Refs. [20, 35].
Figure 1.7: Pictorial profile of the conduction band for the triple barrier heterostructure at zero bias. The material parameters are also given [34].
1.3 Single-Electron Transport

The electronic structure of QDs mimicks that of an isolated atom. It is possible to attach wires to this “atom” and study transport through its discrete electronic states. In this section we present the theory of Coulomb oscillations, following Ref. [36].

We consider a SETS experiment, in the limit of zero source-drain bias $V_{sd} \to 0$ (linear transport). Since transport through a QD proceeds by tunneling through its discrete electronic states, it will be clear that a net current can flow only for certain values of the gate voltage $V_g$. In the absence of charging effects (non-interacting system), a conductance peak due to resonant tunneling occurs when the chemical potential $\mu$ in the reservoirs lines up with one of the single-electron energy levels in the dot. This condition is modified by the charging energy (i.e. electrons interact via Coulomb repulsion). To determine the location of the conductance peaks as a function of $V_g$ requires only consideration of the equilibrium properties of the system, as we now discuss.

The probability $P(N_e)$ to find $N_e$ electrons in the QD in equilibrium with the reservoirs is given by the grand canonical distribution function

$$P(N_e) \propto \exp \left[ - \frac{(F(N_e) - N_e \mu)}{K_B T} \right],$$

where $F(N_e)$ is the free energy of the dot and $T$ the temperature. The reservoir chemical potential $\mu$ is measured relative to the conduction band bottom in the reservoirs. In general, $P(N_e)$ at $T = 0$ is non-zero for a single value of $N_e$ only (namely the integer which minimizes the thermodynamical potential $\Omega(N_e) = F(N_e) - N_e \mu$). In that case, the conductance $G \to 0$ in the limit $T \to 0$. As pointed out in Ref. [23], a non-zero $G$ is possible only if $P(N_e)$ and $P(N_e + 1)$ are both non-zero for some $N_e$. Then a small applied voltage is sufficient to induce a current through the dot, via intermediate states $N_e \to N_e + 1 \to N_e \to N_e + 1 \to \ldots$. To have $P(N_e)$ and $P(N_e + 1)$ both non-zero at $T = 0$ requires that both $N_e$ and $N_e + 1$ minimize $\Omega$. A necessary condition is $\Omega(N_e + 1) = \Omega(N_e)$, or

$$F(N_e + 1) - F(N_e) = \mu.$$  \hfill (1.6)

This condition is also sufficient, unless $\Omega$ has more than one minimum (which is usually not the case).
1.3 Single-Electron Transport

Despite its apparent simplicity, Eq. (1.6) is the most important concept of Chapter 1. It expresses the equality of the electrochemical potential of dot and leads. The usefulness of this result is that it maps the problem of determining the location of the conductance peaks onto the more familiar problem of calculating the electrochemical potential $F(N_e + 1) - F(N_e)$ of the QD, i.e. the energy cost associated with the addition of a single electron to the dot. At $T = 0$, the addition energy $A(N_e)$ takes the form

$$A(N_e) = E_0(N_e) - E_0(N_e - 1) = E_F,$$

(1.7)

where $E_0(N_e)$ is the ground state energy of the $N_e$-dependent interacting system, and $E_F$ is the Fermi energy of the reservoirs (we have relabeled $N_e$ by $N_e - 1$). This opens the way to a study of exchange and correlation effects on conductance oscillations in a QD (see e.g. Figs. 4.1 and 4.2).

1.3.1 “Classical” Theory of Coulomb Blockade

At $T = 0$ the free energy $F(N_e)$ equals the ground state energy of the dot, for which we take the simplified form $U(N_e) + \sum_p E_p$. Here $U(N_e)$ is the charging energy, and $E_p (p = 1, 2, \ldots)$ are single-electrons energy levels in ascending order, measured relative to the bottom of the potential well in the QD. The term $U(N_e)$ accounts for the charge imbalance between dot and reservoirs. The sum over energy levels accounts for the internal degrees of freedom of the QD. Each level contains either one or zero electrons. Spin degeneracy, if present, can be included by counting each level twice, and other degeneracies can be included similarly. The energy levels $E_p$ are assumed to be independent of $N_e$. We conclude from Eq. (1.7) that a peak in the low-temperature conductance occurs whenever

$$E_{N_e} + U(N_e) - U(N_e - 1) = E_F,$$

(1.8)

for some integer $N_e$.

We adopt the simple approximation of the “classical” model of taking the charging energy into account macroscopically. This is also known as “constant-interaction model”[37]. This simple model cannot explain addition spectra like the
ones appearing in Figs. 4.1 and 4.2 which show correlation and exchange effects. However, it is still useful to understand most SETS experiments. We write

$$U(N_e) = \int_0^{N_e} dQ' \phi(Q'),$$

(1.9)

where

$$\phi(Q) = Q/C + \phi_{\text{ext}}.$$  

(1.10)

is the potential difference between dot and reservoir, including also a contribution $\phi_{\text{ext}}$ from external charges (in particular those on a nearby gate electrode). We parameterize the Coulomb interactions among the electrons in the dot and between electrons in the dot and those somewhere else in the environment (as in the leads) by means of a capacitance $C$, which is assumed to be independent of $N_e$. This is a reasonable assumption as long as the dot is much larger than the screening length (i.e. no electric fields exist in the interior of the dot). The charging energy then takes the form

$$U(N_e) = \frac{(eN_e)^2}{2C} + eN_e\phi_{\text{ext}}.$$  

(1.11)

$Q_{\text{ext}} \equiv C\phi_{\text{ext}}$ plays the role of an “externally induced charge” on the dot, which can be varied continuously by means of an external gate voltage (in contrast to $Q$ which is restricted to integer multiples of $e$). In terms of $Q_{\text{ext}}$ one can write

$$U(N_e) = \frac{(eN_e + Q_{\text{ext}})^2}{2C} + \text{constant},$$  

(1.12)

which is equivalent to Eq. (1.11). We emphasize that $Q_{\text{ext}}$ is an externally controlled variable, via the gate voltage, regardless of the relative magnitude of the various capacitances in the system.

Substitution of Eq. (1.11) into Eq. (1.8) gives

$$E^*_N = E_n + \left(N_e - \frac{1}{2} \right) \frac{e^2}{C} = E_n + e\phi_{\text{ext}}$$  

(1.13)

as the condition for a conductance peak. The left-hand-side of Eq. (1.13) defines a renormalized energy level $E^*_N$. The renormalized level spacing $\Delta E^*_N = \Delta E + e^2/C$ is enhanced above the bare level spacing by the charging energy. In the limit $e^2/(C\Delta E) \to 0$, Eq. (1.13) is the usual condition for resonant tunneling.
1.3 Single-Electron Transport

Figure 1.8: Single-electron tunneling through a QD, under the conditions of Eq. (1.13), for the case where the charging energy is comparable to the level spacing. An infinitesimally small voltage difference is assumed between the left and right reservoirs. Note that in the picture the symbol $e$ is positive, contrary to the convention followed in the main text. After Ref. [39].

the limit $e^2/(C \Delta E) \to \infty$, Eq. (1.13) describes the periodicity of the classical Coulomb-blockade oscillations in the conductance versus electron density[38].

In Fig. 1.8 we have illustrated the tunneling of an electron through the dot under the conditions of Eq. (1.13). In panel (a) one has $E_{N_e} + e^2/2C = E_F - e \phi (N_e - 1)$, with $N_e$ referring to the lowest unoccupied level in the dot. In panel (b) an electron has tunneled into the dot. One now has $E_{N_e} - e^2/2C = E_F - e \phi (N_e)$, with $N_e$ referring to the highest occupied level. The potential difference $\phi$ between dot and reservoir has decreased by $|e|/C$ (becoming negative), because of the added electron. Finally, in panel (c) the added electron tunnels out of the dot, resetting the potentials to the initial state of panel (a).

Let us now determine the periodicity of the oscillations. Theoretically, it is convenient to consider the case of a variation of the Fermi energy of the reservoirs at constant $\phi_{\text{ext}}$. The periodicity $\Delta E_F$ follows from Eq. (1.13),

$$\Delta E_F = \Delta E^* = \Delta E + \frac{e^2}{C}.$$  \hspace{1cm} (1.14)

In the absence of charging effects, $\Delta E_F$ is determined by the irregular spacing $\Delta E$ of the single-electron levels in the QD. The charging energy $e^2/C$ regulates
Figure 1.9: Diagram of the bare energy levels (a) and the renormalized energy levels (b) (or additon spectrum) in a QD for the case $e^2/C \approx 2 \langle \Delta E \rangle$. The renormalized level spacing is much more regular than the average bare level spacing $\langle \Delta E \rangle$. Note that the spin degeneracy of the bare levels is lifted by the charging energy. After Ref. [40].

The spacing, once $e^2/C \geq \Delta E$. This is illustrated in Fig. 1.9, for the case that there is no degeneracy. The spin degeneracy of the levels is lifted by the charging energy. In a plot of $G$ versus $E_F$ this leads to a doublet structure of the oscillations, with a spacing alternating between $e^2/C$ and $\Delta E + e^2/C$.

Experimentally, one studies the Coulomb-blockade oscillations as a function of gate voltage. To determine the periodicity in that case, we first need to know how $E_F$ and the set of single-particle energy levels $E_p$ depend on $\phi_{\text{ext}}$. In a two-dimensional electron gas, the external charges are supplied by ionized donors and by a gate electrode (with an electrostatic potential difference $\phi_{\text{gate}}$ between gate and electron-gas reservoir). One has

$$\phi_{\text{ext}} = \phi_{\text{donors}} + \alpha \phi_{\text{gate}},$$  \hspace{1cm} (1.15)

where $\alpha$ (as well as $C$) is a rational function of the capacitance matrix elements of the system. The value of $\alpha$ depends on the geometry. It is reasonable to assume that the electron gas densities in the dot and in the leads increase, on average, equally fast with $\phi_{\text{gate}}$, in a planar QD. For equidistant energy levels in the dot we may then assume that $E_F - E_N$ has the same value at each conductance peak. The
1.4 Survey of the Theoretical Work

In this Section we survey the theoretical literature on the electronic structure of QDs. This short review does not mean to be complete in any sense: instead we just mention some fundamental papers in the field, with emphasis on electronic structure and transport properties in the low-magnetic-field regime. For completeness, we also include recent references that appeared after the publication of our own work.

1.4.1 Single Quantum Dot

True *ab initio* calculations for QDs, taking into account the detailed atomistic structure as well as all many-body interactions, are at the moment very difficult, because of the complexity of the system. It is a fact that almost all theoretical works rely on the envelope function approximation (see Subsection 2.1.1) and assume a simplified confining potential which is usually parabolic (see Subsection 2.1.4). Kumar et al. [41] solved a Schrödinger-Poisson Equation for a square QD and found that in the low-energy excitation spectrum the single-particle confinement potential is parabolic to a good approximation; Stopa [42] obtained similar results and the evidence of a spin-degenerate shell structure corresponding to the symmetries of the parabolic potential by means of Density Functional Theory (DFT) [see Subsec. 2.1.4 for more details].

**Exact Diagonalization**

Because of the competition between kinetic energy, Coulomb interaction and Zeeman energy, diagonalization of the exact many-body hamiltonian is the most desirable approach in many cases. Unfortunately, this is limited to very few electrons \( N_e \sim 6 \), and frequently special constraints are imposed to reduce the compu-
tational effort, e.g. the basis set is truncated to the first Landau level in strong magnetic fields, some degrees of freedom are frozen, etc.

The first who stressed the importance of the electronic correlation was Bryant[43], who performed numerical calculations for $N_e = 2$ electrons in a two-dimensional (2D) rectangular box with infinite-potential walls at $B = 0$ magnetic field. He investigated the interplay of the kinetic energy ($\sim L^{-2}$) and the potential energy ($\sim L^{-1}$) as a function of the the box size $L_x, L_y$. He found the energy spectrum to depend strongly on the ratio $L_x/L_y$. For sufficiently large $L$ the potential energy dominates and the possible precursor of a Wigner solid can be seen in both one- ($L_x \gg L_y$) and two-dimensional ($L_x \sim L_y$) limits.

In the context of the fractional quantum Hall effect (FQHE), Laughlin[44] studied the 2D system with $N_e = 3$ in a strong magnetic field. The effect of confinement on single-particle wavefunctions and energies was, however, neglected. Laughlin explicitly constructed spin-polarized correlated states in the lowest Landau level (LLL) and showed that they well approximate exact eigenstates. The ground state turned out to be incompressible over a range of applied pressure because only special values of the angular momentum $M = 3m$ ($m = 1, 2, 3$) of the ground state minimize Coulomb repulsion. The role of electron-electron interactions including the effect of confinement on single-particle spectra has been studied by Maksym and Chakraborty[45]. They calculated energy spectra for $N_e = 3, 4$ spin-polarized electrons in strong magnetic fields and pointed out the competition of kinetic and interaction energy. As a result of this competition, the angular momentum of the ground state of the electron system changes with increasing $B$ through a series of “magic numbers”. These strongly correlated states can be considered as precursors of FQHE ground states with different filling factors. Correlated electronic states, however, cannot be probed by infrared spectroscopic techniques because far-infrared radiation couples to the center-of-mass motion and hence it is insensitive to the interaction when the confinement is parabolic[46]. This explains why subsequent works focused on alternative physical probes of correlation.

Maksym and Chakraborty[47], Wagner et al.[48] investigated the changes in magnetization vs $B$ for $N_e = 2$. Increasing $B$ causes the ground-state angular momentum and spin to discontinuously jump. Pfannkuche and Gerhardts[49] consid-
1.4 Survey of the Theoretical Work

Considered the effects of deviations from a parabolic confinement potential on infrared spectroscopy (coupling of relative and center-of-mass motion) in the two-electron system. Merkt et al.[50] studied the two-electron energy spectrum vs the dot size, while Pfannkuche et al.[51] examined the role of correlation in the singlet ground state.

Another exploited excitation channel was the radiative recombination of electrons with valence band holes localized by acceptors. Hawrylak and Pfannkuche[52] analyzed the magnetoluminescence spectrum for a $N_e = 3$, polarized and unpolarized, electron system. From the same system Hawrylak[53] deduced a spin- and angular-momentum-resolved addition spectrum which he successfully compared with SECS data[27].

Eric Yang et al.[54] exactly diagonalized a system up to 6 electrons (over a LLL-only basis) and obtained the addition spectrum in the strong $B$ regime. In particular, the so-called “maximum-density-droplet” region was explored, and precursors of FQHE droplets were found. With the same approximation later Wójs and Hawrylak[55] computed one- and two-particle spectral functions by diagonalization up to $N_e = 10$.

An interesting possibility we only mentioned in Subsec. 1.1.2 it is to probe excited correlated states by means of non-linear transport measurements. Pfannkuche and Ulloa[56] found quasiselection rules as hallmark of a correlation effect[57], prohibiting tunneling processes otherwise allowed by energy and spin conservation. The topic of spin blockade[58, 59] is intriguing on its own: Imamura et al.[60] considered the possibility of realizing spin-blockade in single and double QDs in a four-electron low-$B$ system.

We finally mention exact diagonalization studies at low- or zero-field where exchange effects induce spontaneous spin polarization. Palacios et al.[57] performed exact diagonalizations up to $N_e = 5$ and unrestricted Hartree-Fock (H-F) calculations up to 15 electrons. Noticeably the $B$-range considered was large. Eto[61], under certain approximations (frozen occupancies of the lowest single-particle levels), obtained exact results for $N_e \leq 8$. Ezaki et al.[62] considered how modifications in the fully three-dimensional QD geometry affect the open-shell structure, with $N_e \leq 11$. Finally we cite the exact diagonalization study ($N_e \leq 6$) of a self-assembled QD by Wójs and Hawrylak[63].
Hartree, Hartree-Fock, Density Functional Theory

A discussion of Hartree and H-F method applied to QDs can be found in[51, 64]. Pfannkuche et al.[65] found new features evolving in the far-infrared $B$-dependent spectra with an increasing number of electrons: the appearance of magnetoplasmon excitations was predicted by H-F calculations.

With regards to addition spectra at zero- or low-field, Oaknin et al.[66] performed a Hartree calculation, Wang et al.[67] a restricted H-F computation, while Macucci et al.[68] employed (spin-independent) density functional theory (DFT). All these latter works were unable to resolve the spin-dependent structure of addition spectra. Unrestricted H-F calculations were instead performed by Fujito et al.[69] and by Tamura[70]. The first one ($N_e \leq 12$) considered different three-dimensional geometries ($N_e \leq 12$) at $B = 0$, while the latter one investigated the $B$-dependent addition spectra of single and coupled QDs.

Steffens et al.[71] made use of current-spin DFT (CSDFT)[72, 73] to study evolution of Hund’s rules with $B$ in addition spectra. They found that, at low $B$, one can still speak of a generalized Hund’s rule referred to single-particle levels different than those at $B = 0$.

Spin and Charge Density Waves, Wigner Crystallization, Edge States

Kramers and co-workers[74] have carried out a numerical calculation for $N_e = 3, 4$ in a rectangular box $L_x \gg L_y$ using a “pocket-state” basis approach. This approach exploits the potential-energy minima associated with the classical $N_e$-electron configurations (see e.g. Ref. [75]) to introduce a finite set of approximate, localized basis functions for the $N_e$-electron system in configuration space: the Hamiltonian is then diagonalized within this basis. For small $L_x$ the single-particle confinement energy dominates; the charge density of the ground state is essentially homogeneous. As $L_x$ increases the potential energy begins to dominate and the electrons form an inhomogeneous charge distribution resembling a blurred version of the classical limit of equispaced point charges (Wigner molecule). The idea that under certain circumstances the QD ground state is predicted to have some order (whose form is determined by the equilibrium between the confinement and the Coulomb interaction) has been exploited also to explain the origin
of “magic numbers”[76, 77]. Wigner crystallization in QDs has also been studied by means of H-F calculations[78], unrestricted spin-and-space H-F[79], Quantum Monte Carlo simulations[80].

Another intriguing question is the breakdown of Hund’s rules at $B = 0$. CSDFT calculations[81] predicted spin-density wave excitations associated to “normal” ground state instabilities as far as the electron density increases. Spin-and-space unrestricted H-F calculations[79] found also some evidence of charge density wave instabilities. Moreover, CSDFT[82] and partial diagonalization technique approach[83] studied edge reconstruction phenomena in QDs in the FQHE regime, maybe connected with the so-called Chamon-Wen edge reconstruction[84].

Analytic Solutions

We mention just a few results. Johnson and Payne[85] exactly solved the problem of $N_e$ electrons of a QD which repel each other via harmonic interaction. Taut[86] solved exactly the two-body problem for a discrete set of harmonic oscillator energies. Dineykhan and Nazmitdinov[87] extended these results to a three-dimensional oscillator with different in-plane and perpendicular oscillator frequencies. Gonzales et al.[88] found Padé approximants for the ground state energies of closed shells.

1.4.2 Coupled Quantum Dots

Until now, double layer two-dimensional electron systems (DL2DESs) have been extensively studied[89, 90, 91]. These systems, which can be considered the limiting case of a double quantum dot (DQD) when the oscillator confinement energy goes to zero, exhibit the rich phenomenology of FQHE ground states.

Palacios and Hawrylak[92] investigated the phase diagram of DQDs at high $B$ in connection with the physics of DL2DESs, introducing an isospin quantum number analogous to the spin degree of freedom. In the regimes we study in this Thesis (namely $B \simeq 0$ and the overall range of the inter-dot coupling), however, the analogy is not so striking: unlike the isospin, the spin is a good quantum number for every value of the inter-dot coupling.
Some studies at $B \simeq 0$ appear in the literature. Oh et al.[93] solved the problem exactly for $N_e=2$. Tamura[70] treated the problem using a Hartree-Fock approach. Tokura et al.[94] performed an exact diagonalization for $N_e \leq 5$. Asano[95] has studied the phase diagram for $13 \leq N_e \leq 24$ by means of a many-body Hamiltonian with “core” electrons frozen in the lower shells. This “core” approximation might fail when the coupling between dots is strong. Imamura et al.[96] studied changes in spin and orbital angular momentum vs. $B$ and inter-dot coupling for $N \leq 4$. They interestingly find that there is a crossover between two regimes (inter-dot and intra-dot correlation, respectively), and that the crossover critical point depends on $B$.

Hubbard-like Hamiltonians for QDs were used —with different levels of approximations— in Refs. [97, 98, 99], while Refs. [100, 101] are recent DFT studies. The DQD was also analyzed from a classical point of view[102].
Chapter 2

Theory I: Single-Particle States

In this Chapter and in the next we present and discuss the theoretical framework for the computation of correlated many-body electronic states in semiconductor vertical QDs. Our approach is general enough to be easily extended to QDs with different geometries and to different experimental settings coupling single QDs between each other. Once electronic states are known, it is straightforward to derive Addition Spectra and to compare them with transport measurements. Applications of the theory and results will be shown in Chapters 4, 5, and 6. Here we present some introductory theoretical material. First we introduce the Hamiltonian describing the electronic motion in QDs (Section 2.1), within the framework of the envelope function approximation, where bulk material parameters as electron effective mass $m^*$ and static dielectric constant $\varepsilon_r$ are assumed. Then we focus on the one-electron problem: we consider analytic solutions in Sec. 2.2, while in Sec. 2.3 we think of one electron as moving in a self-consistent average potential due to the presence of other electrons in the dot. The theory of the true interacting many-electron system is left to the next Chapter.

2.1 Model

2.1.1 The Envelope Function Approximation

To start the study of electronic states in semiconductor QDs we note that the characteristic size of QDs, like those we reviewed in the previous Chapter, is at least one-two order of magnitude bigger than the characteristic length of the semiconductor lattice (unit cell). Hence, the QD has a macroscopic size in comparison to the unit cell, but it is small on all macroscopic scales. One often calls QDs
mesoscopic structures. As a consequence, it is reasonable for mesoscopic structures to make the usual bulk-semiconductor ansatz for the spatial single-particle wavefunctions \( \phi(\mathbf{r}) \)[35],

\[
\phi(\mathbf{r}) = \psi(\mathbf{r}) u(\mathbf{r}),
\]

where \( \mathbf{r} \) is the position of the electron, \( u(\mathbf{r}) \) is the rapidly oscillating periodic part of the Bloch function and \( \psi(\mathbf{r}) \) is the envelope function, which varies on the scale of several unit cells[103] (see Fig. 2.1). Eq. (2.1) implies that we have a non-degenerate conduction band with edge at \( k = 0 \) in the reciprocal space, as it is the case in usual GaAs/GaAlAs heterostructures, and that the Bloch function \( u(\mathbf{r}) \) refers to the state \( k = 0 \).

In the spirit of the ansatz (2.1), it seems sensible to assume that the energy eigenvalues of the electron in the periodic lattice, i.e., the energy bands, are not appreciably modified by the quantum confinement. Besides, most experimental work has been carried out on QDs built from wide-band-gap semiconductors where the electron energies are small compared to the band gap, hence non-parabolicity effects (i.e. energy-dependent conduction-band effective mass and band-edge mixing) can be neglected[16]. Of course, if QDs are so small that the potential profile cannot be considered anymore slowly varying with respect
to the periodicity of the crystalline potential, considerable shortcomings of the envelope function approximation are expected [105]. It seems then reasonable to use the envelope function approximation for all QDs but the self-assembled ones (the smallest ones). Therefore, we use the envelope function and the effective mass approximation to describe the free motion of electrons in QDs. This means that the only effect of the periodic lattice is to renormalize the electronic mass, otherwise the motion of electrons in the bulk crystal is that of a free electron gas. Throughout this Thesis we will consider only electrons which would belong to the conduction band if they were in the bulk, and we will assume that valence bands are completely filled.

2.1.2 Hamiltonian

Within the envelope function approximation, we may write the exact Schrödinger equation for $N_e$ electrons in a QD or in a system of coupled QDs, given by

$$\mathcal{H}\Psi = E\Psi,$$  \hspace{1cm} (2.2)

with

$$\Psi = \Psi(r_1 s_1, r_2 s_2, \ldots, r_{N_e} s_{N_e})$$

total envelope wavefunction of electrons at position $r_i$ and with spin projection along $z$-axis $s_i = \pm 1/2 (i = 1, \ldots, N_e)$, and $E$ total eigenvalue. The total Hamiltonian $\mathcal{H}$ is the sum of a part acting only on spatial coordinates $H_{\text{space}}$ and a term $H_{\text{spin}}$ acting on spin coordinates only:

$$\mathcal{H} = H_{\text{space}} + H_{\text{spin}}.$$  \hspace{1cm} (2.3)

We ignore the effect of spin-orbit coupling which can be shown to be small for a single conduction band and for a small number of electrons[106].

The spatial part $H_{\text{space}}$ is the sum of a single particle term $\sum H_0(i)$ and of the two-body Coulomb interaction term:

$$H_{\text{space}} = \sum_{i=1}^{N_e} H_0(r_i) + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{\kappa r_i} |r_i - r_j|,$$  \hspace{1cm} (2.4)
with $e < 0$ electronic charge (we use Gauss units throughout this Thesis) and \( \kappa_r \) relative dielectric constant of the host semiconductor, which is assumed to be experimentally known ($\kappa_r \approx 10$ in semiconductors). We have written the electron-electron interaction as translationally invariant. This is not \textit{a priori} true due to the presence of image charges in the surrounding dielectric materials and gates[16]. For a treatment of QDs surrounded by materials with a dielectric constant different from that of the host material, see Ref. [107].

We will discuss the single particle term $H_0$ in a moment. The spin part $H_{\text{spin}}$ is simply a Zeeman coupling between the total-spin magnetic moment and an external magnetic field $\mathbf{B} = B\hat{z}$ which we possibly apply along the $z$-axis:

$$ H_{\text{spin}} = g^* \mu_B B \frac{S_z}{\hbar}, $$

where $\mu_B$ is the Bohr magneton ($\mu_B = 5.7884 \times 10^{-2}$ meV/T) and $g^*$ is the effective $g$-factor ($g^* \approx 1$). The total $z$-component of spin $S_z$ is given by

$$ S_z = \frac{\hbar}{2} \sum_{i=1}^{N_e} \sigma_{zi}, $$

where $\sigma_z$ is the $2 \times 2$ Pauli matrix such that

$$ \sigma_{zi} \chi_{\pm}(s_i) = \pm \chi_{\pm}(s_i), $$

with $\chi_{\pm}(s)$ single-particle spin eigenfunction, and $\hbar$ reduced Planck’s constant.

The total spin $\mathbf{S}$ is a vector operator,

$$ \mathbf{S} = (S_x, S_y, S_z), $$

whose components do not commute with each other and whose quantum number is $S$, as it is well known. Since typical single-particle energies computed from the spatial part of the Hamiltonian $H_0$ are of the order of meV, this Zeeman term is much smaller and it is usually neglected in finding out electronic ground states, while excited states are approximately considered doubly degenerate, at least in not-too-high magnetic field regimes. In other words, the way the spin term $H_{\text{spin}}$ works is just to lift the $(2S + 1)$-fold degeneracy of the total spin $\mathbf{S}$. Hence, the practical algorithm to find out the ground state electronic configuration we used
in our calculations was first to compute electronic terms as if $S$ were conserved, and then to choose the electronic term with maximum $S_z$ [51]. Incidentally, we observe that this modus operandi could be used in direct diagonalizations of $H_{\text{space}}$ to reduce Hilbert-space sector dimensions. This could be achieved by labelling subspaces with the additional quantum number $S$, in addition to $S_z$, even if $S$ is not a constant of motion when $B$ is present.

Now we examine the single-particle spatial Hamiltonian $H_0$. Its more general form is:

$$H_0(\mathbf{r}) = \frac{1}{2m^*} \left[ \mathbf{p} - \frac{e}{c} \mathbf{A}(\mathbf{r}) \right]^2 + V(\mathbf{r}),$$

(2.6)

where we have introduced the external constant magnetic field $\mathbf{B} = B\hat{z}$ by means of the vector potential $\mathbf{A}(\mathbf{r})$ [$\mathbf{B} = \nabla \times \mathbf{A}(\mathbf{r})$], and where $\mathbf{p}$ is the canonical momentum, $\mathbf{p} = -i\hbar \nabla$, and $m^*$ is the effective electron mass ($m^* \approx 0.1m_0$ in semiconductors, $m_0$ free electron mass). $V(\mathbf{r})$ is the single-particle confining potential of the QD (or the system of coupled QDs) and $c$ is the speed of light. To summarize, parameters we need are the effective mass $m^*$, the static dielectric constant $\kappa_r$ and the effective $g$-factor $g^*$. These quantities describe the effect of the crystalline periodic potential and the screening of all the other electrons with respect to the few electrons we look at in the dot.

### 2.1.3 Cylindrical Symmetry and Magnetic Field

Until now the Hamiltonian $H_0(\mathbf{r})$ is rather general and the choice of the $z$-axis and of $V(\mathbf{r})$ is completely arbitrary, so it could describe any type of dot geometry or confining potential. Now we specialize in QDs with a cylindrical symmetry around the $z$-axis. This is the case of disk-shaped QDs built by depletion of a two-dimensional electron gas by means of electrodes, or of the so called vertical QDs, single or coupled. This symmetry means that the projection along $z$ of the orbital canonical angular momentum $L_z$ and the confining potential $V(\mathbf{r})$ are commuting operators:

$$[L_z, V(\mathbf{r})] = 0,$$

(2.7)

with $L_z = xp_y - yp_x$ and $[A, B] = AB - BA$. Eq. (2.7) implies that $L_z$ is conserved, so the orbital angular momentum along $z$ is a good quantum number.
(in general this is false for the angular momentum vector \( \mathbf{L} \) except for spherical QDs at \( B = 0 \)).

To prove the previous statement, i.e.

\[
[H_0(\mathbf{r}) , L_z] = 0, \tag{2.8}
\]

we need to specify a gauge for the vector potential \( \mathbf{A}(\mathbf{r}) \): we choose the “symmetric” one, which is especially advantageous for our purposes, namely

\[
\mathbf{A}(\mathbf{r}) = \frac{1}{2} \mathbf{B} \times \mathbf{r}. \tag{2.9}
\]

Note that the symmetric gauge is also a Coulomb gauge, i.e.

\[
\nabla \cdot \mathbf{A}(\mathbf{r}) = 0, \tag{2.10}
\]

as it is easy to check, hence

\[
[\mathbf{A}(\mathbf{r}) , \mathbf{p}] = 0. \tag{2.11}
\]

We will immediately use this property, after having spent some words about gauge transformations.

**Canonical vs “True” Orbital Angular Momentum**

Of course one is free of choosing any vector potential \( \mathbf{A}(\mathbf{r}) \) whose curl gives the same magnetic field \( \mathbf{B} \), and all these different choices must be physically equivalent. However, for different forms of \( \mathbf{A}(\mathbf{r}) \) the Hamiltonian \( H_0(\mathbf{r}) \) changes and so the probability density current, the velocity and related quantities. In particular, if one defines, according to Heisenberg’s equations of motion, the velocity operator \( \mathbf{v} \) as \( \dot{\mathbf{r}} \),

\[
\dot{\mathbf{r}} = -\frac{\hbar}{i} [\mathbf{r}, H_0(\mathbf{r})],
\]

one obtains

\[
m^* \mathbf{v} = \mathbf{p} - \frac{e}{c} \mathbf{A}(\mathbf{r}),
\]

hence the “true” orbital angular momentum

\[
\mathbf{L}_{\text{true}} = \mathbf{r} \times m^* \mathbf{v}
\]
is different from the canonical one \( L \) and has the form
\[
L_{\text{true}} = L - \frac{e}{c} \mathbf{r} \times \mathbf{A}(\mathbf{r}).
\]

Eq. (2.12) is valid in every Coulomb gauge: in the symmetric gauge in addition we obtain the explicit expression
\[
L_{\text{true}} = L - \frac{e}{2c} \mathbf{r} \times \left[ \mathbf{B} \times \mathbf{r} \right].
\]

This difference is relevant when one looks at the electronic magnetization in the dot and wants to derive the correct giromagnetic ratio[108].

Let’s now go back to prove Eq. (2.8). We just expand the square of operators in the right member of Eq. (2.6) using the commutative property (2.11) and the explicit expression (2.9):
\[
\frac{1}{2m^*} \left[ \mathbf{p} - \frac{e}{c} \mathbf{A}(\mathbf{r}) \right]^2 = \frac{\mathbf{p}^2}{2m^*} + \frac{e}{2m^*c} \mathbf{p} \cdot \mathbf{r} \times \mathbf{B} + \frac{e^2B^2}{8m^*e^2} \left( x^2 + y^2 \right). \quad (2.13)
\]

By means of the triple product rule \( \mathbf{A} \cdot \mathbf{B} \times \mathbf{C} = \mathbf{A} \times \mathbf{B} \cdot \mathbf{C} \) and defining the cyclotron frequency \( \omega_c = \frac{|e|}{B/m^*c} \) we eventually obtain
\[
H_0(\mathbf{r}) = \frac{\mathbf{p}^2}{2m^*} + V(\mathbf{r}) + \frac{\omega_c}{2} L_z + \frac{m^*\omega_c^2}{8} \left( x^2 + y^2 \right). \quad (2.14)
\]

From Eq. (2.14), by inspection, keeping in mind Eq. (2.7) and the fact that \( \rho^2 = x^2 + y^2 \) is invariant under rotations around the \( z \)-axis, it turns out that \( L_z \) is conserved. This is true also for the total angular momentum along \( z \)
\[
L_{z}^{\text{tot}} = \sum_{i=1}^{N_e} L_{zi}
\]
for the many-electron system, because Coulomb interaction is invariant under any rotation.

### 2.1.4 Dot Confining Potential

We now describe the confining potential \( V(\mathbf{r}) \) and the kind of dot we are going to examine. First we assume, consistently with Eq. (2.7), that we can separate \( V(\mathbf{r}) \) into the sum of two parts,
\[
V(\mathbf{r}) = V(x, y) + V(z),
\]
one for the motion of electrons in the plane $xy$ perpendicular to the growth axis $V(x, y)$, the other referring to the motion along the $z$-axis $V(z)$. Systems we think of are: i) planar single QDs built by depleting a two-dimensional electron gas by suitable electrodes, ii) disc-shaped vertical QDs, single or coupled, like those we discussed in Chapter 1. In both cases: 1) the confinement along the $z$ direction is much stronger than the in-plane one, 2) the potential profile $V(z)$ can be well described by a quantum well (QW), single or double, in the case of a single or a double QD, respectively.

The in-plane confining potential $V(x, y)$ is more worth discussing. The lateral potential of a QD differs significantly from the Coulomb potential binding electrons in an atom. Depending on the method used to create the dot, the lateral potential can be approximated by a different model potential. The potential of a square 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 $L$ of the variation of the lateral potential near the edge), a good approximation offers simple smooth potentials, such as a Gaussian well[20],

$$V(\rho) \propto -e^{-\rho^2/L^2}.$$  

In the cases where the scattering states can be neglected, the Gaussian well can be replaced at the bottom with a parabolic well. Throughout this work, we will always assume a parabolic form:

$$V(x, y) = \frac{1}{2} m^* \omega_0^2 (x^2 + y^2) = \frac{1}{2} m^* \omega_0^2 \rho^2,$$  

Equation (2.15)

with $\omega_0$ characteristic frequency, and $\rho$ in-plane vector position. This approximation seems particularly fit for QDs of type i) or ii) we have mentioned above. A schematic picture of the model potential $V(x)$ we use is drawn in Fig. 2.2.

Theoretical and experimental arguments lead to the assumption (2.15):

- The lateral confining potential has mainly an electrostatic origin and represents the static potential inside the dot at the equilibrium with all the electric charge density inside and outside the dot. Charge neutrality is ensured by considering the whole surrounding environment, electrodes included. Reasoning in a classical way, we can think that the electric field works against
the motion of electrons perturbing the equilibrium position inside the dot. In a first approximation, we can model the restoring force of the field as linear in the displacement. Hence, the electrostatic confining potential must be parabolic.

- Whatever model of circularly symmetric lateral potential we consider, it will not introduce any substantial error if we are interested only in the very first excited single-particle states.

- Full quantum mechanical calculations have been reported which stress the validity of the harmonic approximation for a large variety of QDs and values of parameters. These theoretical schemes take account, in a self-consistent way, of the combined effect of the interaction of electrons in the dot with external gates and/or image charges at the heterostructure interfaces and of the electrostatic screening of the charge inside the dot.
Kumar et al. [41] described the results of a numeric, self-consistent solution of the Poisson and Schrödinger equations in the Hartree approximation for a QD produced by a voltage applied to a metal electrode covering a square GaAs layer with an area of 300×300 nm and a thickness of 30 nm, created on the surface of a GaAs/AlGaAs QW. A small number of electrons \( N_e \sim 10 \) was considered. It follows from these calculations that despite the square shape of the electrode, the confining potential is to a good approximation circularly symmetric, with a diameter of \( \approx 100 \) nm, which is considerably smaller than the size of the electrode. The separation between the energy levels is almost independent of \( N_e \), and the magnetic field evolution of energy levels is similar to that of levels of a parabolic well (see Sec. 2.2).

Macucci et al. [68] reported similar calculations using a local density functional approach. They found significant deviations from the harmonic confinement due to the self-consistent Coulomb interaction of electrons inside the dot: as the number of electrons \( N_e \) is increased, the potential is gradually flatten out and raised.

Stopa [42] performed accurate density-functional calculations for \( N_e \leq 100 \) electrons in a lateral square QD (500 nm × 560 nm approximately) patterned via surface gates on the two-dimensional electron gas formed at the interface of a GaAs-Al\(_x\)Ga\(_{1-x}\)As heterosturcture. He found that the lateral confining potential can be regarded as parabolic to lowest order with quartic and higher order corrections whose influence increases near the perimeter. A parabolic confining potential was also obtained as a result of calculations regarding \textit{n-i-p-i} doping superlattices [109], wires in Si[110] and GaAs[111].

• Maybe the strongest argument in favour of Eq. (2.15) is an experimental one. Kohn’s theorem [46] states that the cyclotron frequency in a translationally invariant electron system is independent of the electron density and of the form of the electron-electron interaction. The theorem follows from the fact that the electric dipole of the radiation only couples to the centre of mass of the electrons, leaving the relative motion unchanged. This result has been generalized to parabolic quantum wells[112] and quantum
2.2 Single-Particle States

In this Section we carefully inspect solutions of the single-particle Hamiltonian $H_0(\mathbf{r})$ of Eq. (2.14) with the lateral parabolic confinement potential (2.15), which was discussed in the previous Section. A complete understanding of the single-particle energy spectrum and related degeneracies is the basis for a further investigation of the many-electron energy shell structure. In this regard, the logical approach is fully analogous to the study of electronic states in Atomic and Molecular Physics.

In Subsection 2.2.1 we look at the general structure of the single-particle wavefunction and at the motion along the growth direction, while in Subsection 2.2.2 we explicitly solve the in-plane electronic motion problem.

2.2.1 General Considerations

We solve the spatial single-particle Hamiltonian

$$H_0(\mathbf{r}) = \frac{\mathbf{p}^2}{2m^*} + \frac{m^*}{2} \Omega^2 \mathbf{\rho}^2 + V(z) + \frac{\omega_c}{2} L_z. \tag{2.16}$$

Here

$$\Omega^2 = \omega_0^2 + \omega_c^2 / 4$$

is an effective frequency: if $B \to 0$ then $\omega_c \to 0$ and $\Omega \to \omega_0$, i.e. we recover the zero-magnetic-field case, while if $B \to \infty$ then $\Omega \to \omega_c / 2$ and (2.16) becomes the Hamiltonian of a free electron (in the plane) in a transverse magnetic field. The spatial wavefunction solution $\phi(\mathbf{r})$ of the Schrödinger equation associated with Eq. (2.16) is the product of the in-plane wavefunction $\psi(\rho, \varphi)$ times the dots where the confinement is parabolic[45, 113, 114]. Far-infrared optical measurements on artificially fabricated semiconductor QDs have indeed found the absorption frequencies to be essentially independent of the number of electrons[115]; this implies that the bare confining potential is nearly parabolic. Besides, this latter assumption is consistent with the behaviour of absorption lines in presence of an external magnetic field.
wavefunction $\chi(z)$ describing the motion along $z$:

$$\phi(r) = \psi(\rho, \varphi)\, \chi(z), \quad (2.17)$$

where $(\rho, \varphi)$ is the in-plane position vector $(x, y)$ in polar coordinates.

**Motion along the growth axis**

Let’s focus on the motion along $z$. We will always assume a potential invariant under reflection with respect to the plane at $z = 0$ perpendicular to the growth direction, namely

$$V(z) = V(-z). \quad (2.18)$$

From general considerations it follows that $\phi(z)$ must be even or odd under $z \to -z$ reflection. In particular, if the ground state is bound, then it has to be even without nodes.

In the case of a single QD, we take as $V(z)$ a symmetric quantum well:

$$V(z) = V_0 \theta(|z| - \frac{L}{2}). \quad (2.19)$$

Here $\theta(x)$ is the Heaviside function, $L$ is the well width and $V_0$ is the well height. We assume the same dielectric constant in both the well and the barrier, while the effective mass can be different ($m_{\text{out}}^*$ in the barrier and $m_{\text{dot}}^*$ in the dot, respectively). The solution is standard in elementary Quantum Mechanics, except that the condition of continuity of derivatives on both side of one interface must be replaced by the condition

$$\frac{1}{m_{\text{out}}^*} \frac{\partial}{\partial z} = \frac{1}{m_{\text{dot}}^*} \frac{\partial}{\partial z}, \quad (2.20)$$

which derives from probability current density conservation[116]. We just summarize well known results: The energy of a bound state $\epsilon_z$ is given by

$$\epsilon_z = \frac{\hbar^2 \kappa^2}{2m_{\text{dot}}^*}, \quad (2.21)$$

with $\kappa$ assuming discrete values obtained by the solution of the transcendental equation

$$\kappa \gamma \tan \left( \frac{\kappa L}{2} \right) = \chi. \quad (2.22)$$
Here $0 < \kappa < \pi / L$, $\gamma = m_{\text{out}}^* / m_{\text{dot}}^*$, and $\chi = \sqrt{(V_0 - \epsilon_z)2m_{\text{out}}^*/\hbar^2}$. The form of the wavefunction $\chi(z)$, up to a phase, is

$$\chi(z) = \begin{cases} 
  c_0 e^{\kappa z} & \text{if } z < -\frac{L}{2}, \\
  c_0 \cos (\kappa z) & \text{if } -\frac{L}{2} < z < \frac{L}{2}, \\
  c_0 e^{-\kappa z} & \text{if } z > \frac{L}{2},
\end{cases} \quad (2.23)$$

with normalization constants

$$C = e^{\frac{\kappa L}{2}} \cos \left( \frac{\kappa L}{2} \right),$$
$$c_0 = \left[ \frac{L}{2} + \frac{\sin (\kappa L)}{2\kappa} + \frac{C^2}{\chi} e^{-\kappa L} \right]^{-\frac{1}{2}}.$$

As we have already mentioned in Sec. 2.1.4, the confinement of the motion along $z$ is much stronger than the lateral one: E.g., with typical GaAs parameters, $V_0 = 200$ meV, and $L \approx 10$ nm, the separation between ground and first excited state is about 100 meV, at least one order of magnitude bigger than characteristic lateral confinement energies. Hence, for a single QD, we will always postulate the adiabatic condition according to which the electron is in its ground state with respect to the motion along $z$. While this condition has no effect on the single-particle energy spectrum, which is entirely determined by the in-plane motion, it can have important quantitative consequences on the many-electron spectrum, affecting two-particle Coulomb and exchange integrals, as we shall see.

The case of two coupled QDs is a straightforward generalization of the one above. The potential $V(z)$ is a double QW:

$$V(z) = \begin{cases} 
  V_0 & \text{if } z < -L - d/2, \\
  0 & \text{if } -L - d/2 < z < -d/2, \\
  V_0 & \text{if } -d/2 < z < d/2, \\
  0 & \text{if } d/2 < z < L + d/2, \\
  V_0 & \text{if } L + d/2 < z,
\end{cases} \quad (2.24)$$

where $L$ is the width of both dots and $d$ the inter-dot distance (see Fig. 2.3). Although we could write down explicitly a transcendental equation more complex than (2.22) to find eigenvalues and wavefunctions, we prefer to solve the one-dimensional Schrödinger equation numerically. In Fig. 2.3 we show typical
Figure 2.3: Confinement potential $V(z)$ along the growth direction $z$ for a system of two coupled QDs. The lower energy wavefunctions (even ground state $|s\rangle$ and odd first excited state $|a\rangle$) are also drawn.

wavefunctions together with a characteristic potential profile. From energy arguments similar to those of the single QD case, we are now forced to consider not only the even ground state $|s\rangle$ but also the odd first excited state $|a\rangle$. This has important qualitative implications for the energy spectrum of double QDs, of course. Note that, in the limit $d \to \infty$, the ground state $|s\rangle$ turns into the symmetric superposition of ground states of single QWs localized on each dot, while the first excited state $|a\rangle$ is the antisymmetric linear combination, and the two eigenvalues are degenerate. In the opposite limit, $d \to 0$, one regains the single QW picture and $|s\rangle$ and $|a\rangle$ become the ground and first excited states of the single QD, respectively.

We now discuss the in-plane motion. By inspection of Eq. (2.16), it is evident that in the case $B = 0$ the Hamiltonian $H_0(\mathbf{r})$ is invariant under reflection in a plane parallel to the $z$ axis and which contains the origin. This explains why the eigenfunctions of $H_0(\mathbf{r})$, which are also eigenfunctions of the orbital angular
momentum $L_z$ with quantum number $m$, are doubly degenerate, with opposite numbers $m$ and $-m$ corresponding to the same energy, except the case $m = 0$. This degeneracy can also be associated to the time-reversal symmetry of the Hamiltonian (2.16). The presence of a magnetic field $B$ breaks the time-reversal symmetry (or the reflection symmetry), hence states with opposite $m$ are no more degenerate, as we shall see in the next Subsection.

### 2.2.2 Fock-Darwin States

In this Subsection we solve analytically the in-plane motion problem. It is convenient to look first at the case $B = 0$[117]. We already know that solutions $\psi(\rho, \varphi)$ of the time-independent Schrödinger equation associated with the in-plane Hamiltonian

$$
\left[ -\frac{\hbar^2}{2m^*} \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) + \frac{m^*}{2} \omega_0^2 \rho^2 \right] \psi(\rho, \varphi) = \varepsilon \psi(\rho, \varphi)
$$

(2.25)

are simultaneously eigenfunctions of the $z$-component of the orbital angular momentum

$$
L_z = \hbar \frac{\partial}{\partial \varphi},
$$

so we write $\psi(\rho, \varphi)$ as the product of a radial part $v(\rho)$ by an angular part:

$$
\psi(\rho, \varphi) = v(\rho) e^{-im\varphi}.
$$

(2.26)

The angular part $e^{-im\varphi}$ is an eigenfunction of $L_z$ with integer quantum number $m$ ($m = 0, \pm 1, \pm 2, \ldots$).

At this point, we note that unfortunately in the QD literature there are different conventions about the sign of orbital and spin angular momentum. To be consistent with previous works, from now on we choose to change sign in the definition of angular momentum. Hence the angular wavefunction

$$
\frac{1}{\sqrt{2\pi}} e^{-im\varphi}
$$

(2.27)

is eigenfunction of

$$
L_z = -\hbar \frac{\partial}{i \partial \varphi}
$$

(2.28)

with eigenvalue $hm$, and analogously the Zeeman spin term favours configurations with maximum positive $S_z$, if $B > 0$. 
After the separation of variables of Eq. (2.26), we obtain a radial ordinary differential equation from Eq. (2.25):

\[
\left[ \frac{\partial^2 v(\rho)}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial v(\rho)}{\partial \rho} - \frac{m^2}{\rho^2} v(\rho) \right] + \left[ k^2 - \lambda^2 \rho^2 \right] v(\rho) = 0.
\]  

(2.29)

The first bracket is originated by the Laplacian. Here \( k^2 \) is connected to the eigenvalue \( \varepsilon \) which we will determine compatibly with boundary conditions,

\[
\varepsilon = \frac{\hbar^2 k^2}{2m^*},
\]

and

\[
\lambda = \frac{m^* \omega_0}{\hbar}
\]

is a characteristic oscillator quantity such that \( \lambda^{-1} \) is the mean square radius of the ground state, \( \langle \rho^2 \rangle_{\text{g.s.}} = 1/\lambda \). Eq. (2.29), together with boundary conditions at the frontier of the variable domain, constitutes a Stourm-Liouville problem to be solved with the standard procedure of Wave Mechanics. By means of boundary conditions we ask for a meaningful physical wavefunction, namely \( v(\rho) \) remains finite as \( \rho \to 0 \) and \( v(\rho) \to 0 \) as \( \rho \to \infty \) (bound state).

If one looks at the differential equation (2.29) in the neighbourhood of \( \rho \approx 0 \) one retains, at the lowest order,

\[
\frac{\partial^2 v(\rho)}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial v(\rho)}{\partial \rho} - \frac{m^2}{\rho^2} v(\rho) = 0.
\]  

(2.30)

Because \( v(\rho) \) must be finite around the origin, we can write down \( v(\rho) \) as a Taylor expansion for \( \rho \approx 0 \), and the first term in the expansion has to be

\[
v(\rho) \approx \rho^s,
\]

with \( s \) integer [a constant is not a solution of (2.30)]. From the compatibility condition

\[
s^2 - m^2 = 0
\]

we choose \( s = |m| \) to keep \( v(\rho) \) finite at the origin. By means of a similar argument, we find the asymptotic form of \( v(\rho) \) as \( \rho \to \infty \):

\[
v(\rho) \approx e^{-\lambda \rho^2/2}.
\]
2.2 Single-Particle States

Now we combine the two results and postulate the form of the true wavefunction as

\[ \psi(\rho) = \rho^{|m|} e^{-\lambda \rho^2/2} F(\rho), \quad (2.31) \]

where \( F(\rho) \) must tend to a non-zero constant value as \( \rho \to 0 \) and cannot diverge faster than \( e^{\lambda \rho^2/2} \) as \( \rho \to \infty \). Inserting (2.31) into Eq. (2.29) the differential equation

\[
\frac{d^2 F(\rho)}{d\rho^2} + \left[ \frac{2|m| + 1}{\rho} - 2\lambda \rho \right] \frac{dF(\rho)}{d\rho} - \left[ 2\lambda (|m| + 1) - k^2 \right] F(\rho) = 0 \quad (2.32)
\]

is obtained for \( F(\rho) \) which, by using, instead of \( \rho \), the dimensionless variable

\[ x = \lambda \rho^2 \]

is transformed into the Kummer equation

\[
x \frac{d^2 F(x)}{dx^2} + \left[ (|m| + 1) - x \right] \frac{dF(x)}{dx} - \frac{1}{2} \left[ (|m| + 1) - \frac{k^2}{2\lambda} \right] F(x) = 0 \quad (2.33)
\]

whose solution regular at \( x = 0 \) is the Confluent Hypergeometric Series[118]

\[ F(x) = M(a, |m| + 1; x) \]

where

\[ a = \frac{1}{2} (|m| + 1) - \frac{k^2}{4\lambda}. \]

For large values of \( x \) this function would diverge as \( e^x \), thus preventing normalization. If, and only if

\[ a = -n \quad (2.34) \]

with \( n = 0, 1, 2, \ldots \) integer radial quantum number, the Hypergeometric Confluent Series becomes a polynomial and the wavefunction can be normalized. We then have the solutions

\[ \psi_{nm}(\rho, \varphi) = C_{nm}\rho^{|m|} e^{-\lambda \rho^2/2} M(-n, |m| + 1; \lambda \rho^2) e^{-im\varphi} \quad (2.35) \]

and

\[ \varepsilon_{nm} = \hbar \omega_0 (2n + |m| + 1). \quad (2.36) \]
Figure 2.4: Shell structure of the single-particle energy spectrum, and associated degeneracies. Each shell corresponds to the energy $\hbar \omega_0 (N + 1)$, where $N = 2n + |m|$ is fixed, and $(n, m)$ are the radial and azimuthal quantum numbers, respectively. The degeneracy of each shell (not taking account of spin) is $N + 1$.

Here $C_{nm}$ are normalization factors, $\varepsilon_{nm}$ is the eigenvalue depending on the couple of radial and azimuthal quantum numbers $(n, m)$, and $\psi_{nm}(\rho, \varphi)$ are called Fock-Darwin states, after [119] and [120].

Let’s focus now on the structure of the single-particle energy spectrum. In Fig. 2.4 we have schematically depicted its shell structure and the associated degeneracies. Each shell corresponds to the energy $\hbar \omega_0 (N + 1)$, where $N = 2n + |m|$ is fixed. The shell degeneracy (not taking account of spin) is then $N + 1$.

Even without analyzing the wavefunctions’ analytical structure, we can very easily compute the mean square radius $\langle \rho^2 \rangle_{nm}$ over any orbital, because the starting Hamiltonian is quadratic in coordinate operators. In such a fortunate case we can write down the Virial Theorem for bound states[121] (analogous to the one of Classical Mechanics) as

$$
\left\langle \left. -\frac{\hbar^2}{2m^*} \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \right\rangle = \left\langle \frac{m^*}{2} \omega_0^2 \rho^2 \right\rangle,
$$

where the symbol $\langle \ldots \rangle$ stands for quantum mechanical average over any state.
Figure 2.5: Plot of Fock-Darwin wavefunctions along the $x$-axis, at $y = 0$ ($\varphi = 0, \pi$). All $\psi_{nm}$'s are real along this direction, and states corresponding to $\pm m$ are the same. Functions corresponding to the three lowest energy shells are plotted.

Hence, one has for the state $(n, m)$:

$$\langle m^* \omega_0 \rho^2 \rangle_{nm} = \langle \varepsilon_{nm} \rangle_{nm} = \hbar \omega_0 (2n + |m| + 1),$$

(2.37)

which we can recast as

$$\langle \rho^2 \rangle_{nm} = \frac{1}{\lambda} (2n + |m| + 1).$$

(2.38)

We see that $\sqrt{\langle \rho^2 \rangle}$ scales as the square root of $N + 1$, and that it is the same for each orbital of a given shell, even for different values of $m$. This considerable spatial extension of probability density for high quantum numbers is consistent with the "softness" of the parabolic potential. In a real QD, i.e. in a system with a finite spatial extension, there are of course physical bounds to the accuracy of this description.

Now we consider wavefunctions. The Hypergeometric Confluent Series, for special values of parameters, reduces to simpler classes of Orthogonal Polynomials, in this case the Generalized Laguerre Polynomials $L^{[\lambda]}_n(x)$. The connection
functions corresponding to the three lowest energy shells are plotted.

The quantity \( P_{nm}(\rho) \) is given by

\[
P_{nm}(\rho) = 2\pi \rho |\psi_{nm}(\rho, \varphi)|^2.
\]

The quantity \( P_{nm}(\rho) \, d\rho \) is the amount of charge contained between \( \rho \) and \( \rho + d\rho \) associated to the state \((n, m)\). Note that \( P_{nm} \) does not depend on the sign of \( m \), and that the normalization is such that \( \int_0^\infty d\rho P_{nm}(\rho) = 1 \).

The formula is

\[
L_n^m(x) = \frac{(n + |m|)!}{n!|m|!} M(-n, |m| + 1; x),
\]

and an expression for \( L_n^m(x) \) is given by

\[
L_n^m(x) = \sum_{i=0}^{n} (-1)^i \frac{(n + |m|)!}{(n - i)!(|m| + i)!} x^i.
\]

We write down explicitly the first few polynomials:

\[
\begin{align*}
L_0^0(x) &= 1, \\
L_0^1(x) &= 1, \\
L_0^2(x) &= 1, \quad L_0^1(x) = -x + 1, \\
L_0^3(x) &= 1, \quad L_1^1(x) = -x + 2, \\
L_0^4(x) &= 1, \quad L_1^2(x) = -x + 3, \quad L_2^2(x) = 1 - 2x + x^2/2,
\end{align*}
\]
\[ L_3^0(x) = 1, \quad L_3^1(x) = -x + 4, \quad L_3^2(x) = 3 - 3x + x^2/2, \]

\[ \ldots \]

Besides, some useful relations are worth mentioning:

\[ L_n^{|m|}(0) = \frac{(n + |m|)!}{n! \cdot |m|!}, \quad L_n^0(x) = 1, \quad L_n^{|m|}(x) = -x + |m| + 1. \]

If \( m = 0 \), \( L_n^0(x) \) reduces to the Laguerre Polynomial \( L_n(x) \),

\[ L_n^0(x) = L_n(x). \]

The following orthogonality relations hold:

\[
\int_0^\infty dx \, x^{|m|} e^{-x} L_n^{|m|}(x) L_{n'}^{|m|}(x) = \delta_{n,n'} \frac{(n + |m|)!}{n!}. \quad (2.41)
\]

By means of Eq. (2.39) and (2.41) we are now able to calculate normalization factors \( C_{nm} \), so the expression (2.35) for the properly normalized Fock-Darwin states turns into (up to a phase)

\[
\psi_{nm}(\rho, \varphi) = \lambda^{|m|+1/2} \sqrt{\frac{n!}{\pi (n + |m|)!}} \rho^{|m|} e^{-\lambda \rho^2 / 2} I_{n-|m|}(\lambda \rho^2) e^{-i m \varphi}. \quad (2.42)
\]

A few explicit formulae, covering the four lowest shells, are given below:

\[
\psi_{00}(\rho, \varphi) = \sqrt{\frac{\lambda}{\pi}} e^{-\lambda \rho^2 / 2},
\]

\[
\psi_{0\pm 1}(\rho, \varphi) = \sqrt{\frac{\lambda}{2\pi}} \rho e^{-\lambda \rho^2 / 2} e^{\mp i \varphi},
\]

\[
\psi_{0\pm 2}(\rho, \varphi) = \sqrt{\frac{\lambda}{2\pi}} \rho^2 e^{-\lambda \rho^2 / 2} e^{\mp 2i \varphi},
\]

\[
\psi_{10}(\rho, \varphi) = \sqrt{\frac{\lambda}{\pi}} (1 - \lambda \rho^2) e^{-\lambda \rho^2 / 2},
\]

\[
\psi_{0\pm 3}(\rho, \varphi) = \sqrt{\frac{\lambda}{6\pi}} \rho^3 e^{-\lambda \rho^2 / 2} e^{\mp 3i \varphi},
\]

\[
\psi_{1\pm 1}(\rho, \varphi) = \sqrt{\frac{\lambda}{2\pi}} \rho^2 (2 - \lambda \rho^2) e^{-\lambda \rho^2 / 2} e^{\pm i \varphi}.
\]

In Fig. 2.5 we have plotted a few wavefunctions corresponding to the three lowest energy shells. The plot is along the \( x \)-axis, at \( y = 0 \), where all wavefunctions are real, and depend only on the absolute value of \( m \), while in general
wavefunctions are complex, and the phase depends both on the value and sign of $m$. One sees that the state $(0, \pm 1)$ changes sign under $x \to -x$ reflection (in general, under a reflection with respect to a plane containing the origin and the $z$-axis), while the other states are symmetric. Besides, states $(0, \pm 1)$ and $(0, \pm 2)$ have one node, while $(1, 0)$ has two nodes. It is interesting to plot not only the wavefunction itself, but also the radial charge density $P_{nm}(\rho) = 2\pi\rho |\psi_{nm}(\rho, \varphi)|^2$. Fig. 2.6 shows this latter quantity corresponding to the same states of Fig. 2.5. The distribution $P_{nm}(\rho)$ represents the amount of probability density of the orbital $(n, m)$ contained between $\rho$ and $\rho + d\rho$. The normalization prefactor $2\pi\rho$ ensures that

$$\int_0^\infty P_{nm}(\rho) \, d\rho = 1.$$ 

One clearly sees that, at fixed $n$, more and more probability weight is moved to larger values of $\rho$ as long as $m$ is increased. This is in agreement with the expression (2.38) for the mean square radius. Besides, it is easy to check that $n - 1$ represents the number of nodes of the radial part of the wavefunction. In this latter plot the fact that the outer maximum is more important than the inner one is exaggerated as compared to the plot of the wavefunction.

We can get a more useful picture of the distribution of the Fock-Darwin orbitals in angle by showing polar diagrams. The functions

$$\frac{1}{\sqrt{2\pi}} e^{-im\varphi}$$

as they stand are not very easy to visualize, since they represent complex functions. However, we can make simple real linear combinations of them. Thus, we can take the sum of functions for a positive and an equal negative $m$ and divide by $\sqrt{2}$, or we can take the difference and divide by $-i\sqrt{2}$. These functions will be real and will be normalized and orthogonal, so that this transformation represents a unitary transformation. This is done in Fig. 2.7, where we plot the square of these latter linear combinations as a function of angle $\varphi$, for $m = 0, \pm 1, \pm 2, \pm 3$ ($m = 0$ does not depend on angle). Finally, Fig. 2.8 shows two-dimensional contour plots of Fock-Darwin orbitals of the four lowest energy shells. Here the quantity plotted is the square modulus of the wavefunction (gray scale). As the shell number is increased, the probability density is split far apart, and the density spatial pattern becomes more and more complex.
Figure 2.7: Dependence of Fock-Darwin wavefunctions on angle. Polar diagrams of linear combinations of eigenfunctions of angular momentum $L_z$ (corresponding to a degenerate value of energy) are depicted. The square modulus of $(e^{-im\phi} \pm e^{im\phi})/\sqrt{4\pi}$ is plotted as a function of azimuthal angle $\phi$. 
Figure 2.8: Two-dimensional contour plot of Fock-Darwin states of the four lowest energy shells. The gray scale has the same normalization for all \((n,m)\) orbitals, with \(n\) radial and \(m\) angular quantum numbers, respectively. Each plot domain is a square of 5.6×5.6 dimensionless length units, where the dimensionless real space is defined by \((x, y) \lambda^{1/2}\), with \(\lambda = m^* \omega_0 / \hbar\). Within each shell, independent linear combinations of degenerate wave functions with different values of \(m\) are plotted.
Fock-Darwin States in Transverse Magnetic Field

It is straightforward to extend all the results obtained until now with $B = 0$ to the case $B \neq 0$. From the Hamiltonian (2.16) holding in the general case and from the fact that the solutions (2.42) are eigenfunctions of the angular momentum $L_z$ we can immediately write down the new energy eigenvalues as

$$
\varepsilon_{nm} = \hbar \Omega \left( 2n + |m| + 1 \right) - \frac{\hbar \omega_c}{2} m.
$$

The wavefunctions now have the same form of Fock-Darwin states (2.42) except that the quantity $\lambda$ occurring in (2.42) must be replaced with

$$
\lambda^* = \frac{m^* \Omega}{\hbar}.
$$

Fig. 2.9 is a plot of a few lowest eigenvalues $\varepsilon_{nm}$ (in units of $\hbar \omega_0$) vs the magnetic field $B$ (expressed as $\omega_c/\omega_0$). As $B$ is increased, one sees that levels labeled by the maximum value of $m$ go first downwards and then slightly upwards crossing all levels of lower shells but the ones corresponding to the maximum value of $m$ within one shell. It is worth to note that, in the limit $B \to \infty$, these lowest eigenvalues (i.e. those labeled by $(0, m)$, with $m > 0$) tend to an asymptotic form corresponding to the lowest Landau level. A second asymptote, with a steeper slope, corresponding to the first excited Landau level, accumulates a second bunch of Fock-Darwin states, as long as $B$ rises, and so on. Landau levels are solutions of the free in-plane electronic motion in the presence of an external static and homogeneous magnetic field. As $B \to \infty$, the parabolic confinement potential becomes negligible with respect to the magnetic confinement and one regains the free electron picture. In our approach, Landau levels are obtained as a special case of Eq. (2.42) and (2.43) putting $\omega_0 = 0$. Some care is required to take account of the degeneracy, which turns into a continuum. For more details on this point, see Ref. [122].

2.3 The Hartree-Fock Method

Until now we have considered solutions of the single-particle Hamiltonian (2.16). This is only a starting point for the study of the strongly correlated system of
Figure 2.9: Plot of a few lowest eigenvalues $\varepsilon_{nm}$ (in units of $\hbar \omega_0$) for a in-plane single-particle parabolic confinement potential vs magnetic field (in units of $\omega_c/\omega_0$).

$N_e$ electrons, which we will face in Chapter 3. In particular, we will solve a many-body model Hamiltonian, written on the basis of some suitable complete and orthogonal set of single-particle wavefunctions. In this perspective, it is not fundamental that this set actually represents solutions of the one-body Hamiltonian (2.16). Instead, this one-particle basis should embody the essential physics of the system. In this regard, we can think that electrons move independently in an average potential produced by all of the electrons. Our strategy then is to use single-particle wavefunctions which are solutions of a suitable effective Hamiltonian describing many-body effects as a one-particle mean-field effective potential. Hence, one has an effective Schrödinger equation where the motion of other electrons is described by an average electrostatic potential (Hartree equation[123])
2.3 The Hartree-Fock Method

which has to be determined self-consistently, together with the wavefunction itself. If one, in addition, takes account of Pauli’s Exclusion Principle, then obtains the Hartree-Fock (H-F) equation[124, 125]. In this Section we will briefly review the main points of this method. To summarize, we look at H-F method because:

- One-particle wavefunction bases computed by means of H-F method can constitute the optimal choice in writing down a many-body model Hamiltonian. This procedural scheme, borrowed by Quantum Chemistry, is an ambitious program and could be a natural development of the work reported in this Thesis.

- The H-F method is a classical paradigm for a mean-field approach to the many-body problem. Hence it is sensible to compare its results with those obtained by means of other many-body techniques, as we will do in Chapter 4.

To keep the discussion simple, in this Section we consider only H-F solutions of the many-body Hamiltonian $\mathcal{H} \approx H_{\text{space}}$ (2.4) [we neglect the Zeeman spin-term, as discussed in subsection 2.1.2] with the single-particle Hamiltonian $H_0(\mathbf{r})$ of Eq. (2.16) referred to a single QD, the generalization to the case of couple QDs being straightforward.

2.3.1 The Variational Principle

To derive the H-F Equation, we have first to introduce the Variational Principle. In this subsection we use Dirac’s notation of “bra” and “ket” for state vectors. We first want to show that the exact many-body Schrödinger equation

$$\mathcal{H} |\Psi\rangle = E |\Psi\rangle \quad (2.45)$$

is equivalent to the variational equation

$$\delta E |\Psi\rangle = 0, \quad (2.46)$$

with

$$E |\Psi\rangle = \frac{\langle \Psi | \mathcal{H} |\Psi\rangle}{\langle \Psi | \Psi \rangle}. \quad (2.47)$$
The variation (2.46) can be obtained from (2.47):

$$\langle \delta \Psi | \mathcal{H} - E | \Psi \rangle + \langle \Psi | \mathcal{H} - E | \delta \Psi \rangle = 0. \tag{2.48}$$

Since $|\Psi\rangle$ is, in general, a complex function, we can carry out the variation over the real and imaginary part independently, which is equivalent to carrying out the variation over $|\delta \Psi\rangle$ and $\langle \delta \Psi |$ independently. To see this we use the fact that Eq. (2.48) is valid for arbitrary infinitesimal $|\delta \Psi\rangle$. We can replace $|\delta \Psi\rangle$ by $i |\delta \Psi\rangle$ and get

$$-i \langle \delta \Psi | \mathcal{H} - E | \Psi \rangle + i \langle \Psi | \mathcal{H} - E | \delta \Psi \rangle. \tag{2.49}$$

Together with Eq. (2.48), we find

$$\langle \delta \Psi | \mathcal{H} - E | \Psi \rangle = 0 \tag{2.50}$$

and the complex conjugate equation. Since $|\delta \Psi\rangle$ is arbitrary, Eq. (2.50) is equivalent to the eigenvalue problem (2.45).

The approximation of such variational methods like H-F consists of the fact that $|\Psi\rangle$ is usually restricted to a set of mathematically simple trial wave functions. As soon as the true function is not in this set, the minimal solution is no longer the exact eigenfunction, but only an approximation. The variational method is especially well suited for determining the ground state, since for any trial wavefunction $|\Psi\rangle$ it can be shown[126] that

$$E \langle \Psi | \mathcal{H} | \Psi \rangle \geq E_0, \tag{2.51}$$

and thus the true ground state energy $E_0$ will always be the lower bound of a variational calculation. In cases where the ground state energy is not degenerate, the equality sign in (2.51) is valid, if and only if $|\Psi\rangle$ is proportional to the true ground state $|\Psi_0\rangle$. If we are interested in the first excited state, we then have to carry out the variation within the subspace entirely orthogonal to $|\Psi_0\rangle$. Within this subspace the first excited state $|\Psi_1\rangle$ has the minimal expectation value of $\mathcal{H}$. To find $|\Psi_1\rangle$, we must carry out the variation with the subsidiary condition

$$\langle \Psi_1 | \Psi_0 \rangle = 0. \tag{2.52}$$

In principle we can continue and calculate the whole spectrum using this method.

In practice, however, we do not know $|\Psi_0\rangle$ exactly. From a variation in a restricted subset of the Hilbert space, we find only an approximation $|\Phi_0\rangle$. For
the calculation of an approximation $|\Phi_1\rangle$ to the first excited state $|\Psi_1\rangle$, we have to solve the variational equation (2.46) with the supplementary condition that $|\Phi_1\rangle$ is orthogonal to $|\Phi_0\rangle$:

$$\langle \Phi_1 | \Phi_0 \rangle = 0. \quad (2.52)$$

For the second excited state, we must have two supplementary conditions, namely: $\langle \Phi_2 | \Phi_1 \rangle = 0$ and $\langle \Phi_2 | \Phi_0 \rangle = 0$. These supplementary conditions are coupled to the problem via Lagrange parameters. We thus see that for higher excited states this method quickly gets rather complicated, therefore it has been applied mainly for the calculation of the ground state. Sometimes, however, these conditions are simply fulfilled because of symmetry properties, as, for example, is the case for states with different angular momentum quantum numbers.

**Hartree-Fock Equations**

We shall assume now that there is an average single-particle potential (later to be called the *Hartree-Fock potential*)

$$\mathcal{H}_{HF} = \sum_{i=1}^{N_e} h(r_i, s_i) \quad (2.53)$$

whose eigenfunction having the lowest eigenvalue $E_{0, HF}$ is an approximation to the exact ground state function. This eigenfunction $\Psi_{HF}(r_1, s_1; r_2, s_2; \ldots; r_{N_e}, s_{N_e})$ is a *Slater determinant*

$$|\Psi_{HF}\rangle = \prod_{i=1}^{N_e} c^\dagger_{\alpha_i \sigma_i} |0\rangle \quad (2.54)$$

in which the Fermion operators (see Appendix A for Second Quantization formulae and notations we need) $c^\dagger_{\alpha \sigma}$, $c_{\alpha \sigma}$ correspond to the single-particle spin-orbitals $\phi_{\alpha \sigma}(r, s)$, which are themselves eigenfunctions of the single-particle Hamiltonian $h(r, s)$, namely:

$$h(r_i, s_i) \phi_{\alpha \sigma}(r_i, s_i) = \varepsilon_{\alpha \sigma} \phi_{\alpha \sigma}(r_i, s_i), \quad i = 1, \ldots, N_e. \quad (2.55)$$

Here $\alpha$ labels a set of single-particle quantum numbers. In the case of an isolated QD, assuming that the one-electron Hamiltonian $h(r, s)$ has axial symmetry, $\alpha$ stands for $(n, m)$, with $m$ usual azimuthal quantum number and $n$ radial quantum
number. In the case of a double QD, an index $i = s, a$ is added, to label the wavefunction for the motion along $z$. We prefer to keep the more general and compact $\alpha$. We obtain the lowest eigenvalue of $\mathcal{H}^{\text{HF}}$ if one occupies the $N_e$ lowest levels in the state $|\Psi^{\text{HF}}\rangle$. Note that, in general, spin-orbitals with the same set of quantum numbers $\alpha$ but with opposite spin $\sigma$ have a different functional form and a different eigenvalue $\varepsilon_{\alpha\sigma}$.

The Hartree-Fock method is now defined in the following way. We set up the determinantal function (2.54), compute the average value of the Hamiltonian $\mathcal{H}$ on this state, and apply the variational principle (2.46), demanding the spin-orbitals to be chosen to make the average energy stationary. In this process, the variations of the spin-orbitals must be such to keep them orthonormal. To save space in notations, we introduce the symbols

$$x_i = (\mathbf{r}_i, s_i), \quad \xi_i = (\alpha_i, \sigma_i).$$

Within this convention, we assume that the symbol of integration over the differential quantity $d\mathbf{x}$ implies a true integration over the space vector and a summation over the spin variable:

$$\int d\mathbf{x} \rightarrow \sum_{s = \pm 1/2} \int d\mathbf{r}.$$ 

From the Second Quantization expression of the Hamiltonian $\mathcal{H}$ given in Appendix A is easy to compute the average energy $\langle \Psi^{\text{HF}} | \mathcal{H} | \Psi^{\text{HF}} \rangle$:

$$\langle \Psi^{\text{HF}} | \mathcal{H} | \Psi^{\text{HF}} \rangle = \sum_{i=1}^{N_e} \tilde{\varepsilon} [i|i] + \frac{1}{2} \sum_{i,j=1}^{N_e} \left( \tilde{V} [i,j|i,j] - \tilde{V} [i,j|i,j] \right)$$ (2.56)

where the single-particle diagonal element is

$$\tilde{\varepsilon} [i|i] = \int d\mathbf{x} \, \phi_{\xi_i}^*(x) \, H_0(\mathbf{r}) \, \phi_{\xi_i}(x)$$ (2.57)

and

$$\tilde{V} [i,j|k,l] = \int d\mathbf{x} \int d\mathbf{x}' \, \phi_{\xi_i}^*(x) \, \phi_{\xi_j}^*(x') \, \frac{e^2}{|\mathbf{r} - \mathbf{r}'|} \phi_{\xi_k}(x') \, \phi_{\xi_l}(x).$$ (2.58)

Because the integration over $d\mathbf{x}$ includes summation over spin, the exchange integral $\tilde{V} [i,j|i,j]$ is zero unless $\phi_{\xi_i}$ and $\phi_{\xi_j}$ are spin-orbitals associated with the
same spin. To derive the H-F equations, we now wish to vary the \( \phi_{\xi_i} \)'s in Eq. (2.56) maintaining the subsidiary conditions, namely the normalization of all \( \phi_{\xi_i} \)'s, and the orthogonality of any two \( \phi_{\xi_i} \)'s. We shall handle these subsidiary conditions by the method of undetermined multipliers:

\[
\delta \left[ \langle \Psi^\text{HF} | \hat{H} | \Psi^\text{HF} \rangle + \sum_{i,j=1}^{Ne} \lambda_{ij} \int dx \phi_{\xi_i}^*(x) \phi_{\xi_j}(x) \right] = 0. \tag{2.59}
\]

We choose to have \( \lambda_{ji} = \lambda_{ij} \), which gives us the correct number of independent multipliers \( \lambda_{ij} \), one for each subsidiary condition. We shall now vary a particular \( \phi_{\xi_i} \); if \( \langle \Psi^\text{HF} | \hat{H} | \Psi^\text{HF} \rangle \) is really a minimum, it will be a minimum as far as the variation of each \( \phi_{\xi_i} \) is concerned. We then find that Eq. (2.59) equals

\[
\int dx \delta \phi_{\xi_i}^*(x) \left[ H_0(r) \phi_{\xi_i}(x) + \sum_{j=1}^{Ne} \left( \int dx' \phi_{\xi_j}^*(x') \phi_{\xi_j}(x') \frac{e^2}{|x-x'|} \phi_{\xi_i}(x) 
- \int dx' \phi_{\xi_j}^*(x') \phi_{\xi_i}(x') \frac{e^2}{|x-x'|} \phi_{\xi_j}(x) + \lambda_{ij} \phi_{\xi_j}(x) \right) \right] = 0, \tag{2.60}
\]

where we have interchanged the names of the variables of integration in the two-body integrals. In Eq. (2.60) we have the statement that the integral of a certain function multiplied by \( \delta \phi_{\xi_i}^*(x) \), where this quantity is arbitrary, must be zero. That is, we must have the remaining factor of the integrand equal to zero, which leads to the equation

\[
H_0(r) \phi_{\xi_i}(x) + \sum_{j=1}^{Ne} \left( \int dx' \phi_{\xi_j}^*(x') \phi_{\xi_j}(x') \frac{e^2}{|x-x'|} \phi_{\xi_i}(x) 
- \int dx' \phi_{\xi_j}^*(x') \phi_{\xi_i}(x') \frac{e^2}{|x-x'|} \phi_{\xi_j}(x) \right) 
= - \sum_{j=1}^{Ne} \lambda_{ij} \phi_{\xi_j}(x). \tag{2.61}
\]

This equation is identical with the H-F equation, except for the occurrence of the nondiagonal terms, for \( j \neq i \), on the right side. It is possible to introduce linear combinations of the spin-orbitals \( \phi_{\xi_i}(x) \), determined by Eq. (2.61), by a unitary transformation, making no change neither in the form of the Slater determinant (up to a factor) nor in the form of the equations, except that the matrix \( \lambda_{ij} \) is transformed like the matrix of an ordinary operator under the action of a unitary transformation. In particular, we can make such a unitary transformation that we
diagonalize the matrix, so that we have only a term in \( \phi_{i}(x) \) on the right side of Eq. (2.61)[127]. In this case we let the matrix element \( \lambda_{ii} \) be called \(-\varepsilon_{HF}^{i}\), and we obtain the HF equations:

\[
H_{0}(r) \phi_{i}(x) + \sum_{j=1}^{N_{e}} \left[ \int d\,x' \phi^{*}_{j}(x') \phi_{i}(x') \frac{e^{2}}{|r - r'|} \right] \phi_{i}(x) - \sum_{j=1}^{N_{e}} \left[ \int d\,x' \phi^{*}_{j}(x') \phi_{i}(x') \frac{e^{2}}{|r - r'|} \right] \phi_{i}(x) = \varepsilon_{HF}^{i} \phi_{i}(x). \quad (2.62)
\]

This is a set of \(N_{e}\) integro-differential equations: each of them constitutes an eigenvalue problem, which is not linear, because the Hermitian operator acting on \( \phi_{i}(x) \) depends on the solution \( \phi_{i}(x) \) itself. Hence Eqs. (2.62) are solved self-consistently. The first sum (Hartree term) is made of direct or Coulomb integrals and represents the electrostatic interaction with the electronic charge, the \(i\)-th electron included; the second sum (Fock term) is a non local operator and is made of exchange terms. Note that the self interaction of the \(i\)-th electron on itself is not contained in the H-F operator, because when \(i = j\) the direct and exchange terms in the two sums are equal and cancel one each other. The interested reader can find a discussion on the physical meaning of Eq. (2.62) in the reference cited in note [127].

We shall now consider the relation between the “one-electron energies” \(\varepsilon_{HF}^{i}\), which do not have a true and precise physical interpretation in a many-body problem, and the quantity we are really interested in, i.e. the total energy of the system. First we multiply (2.62) on the left by \(\phi^{*}_{i}(x)\) and integrate over \(d\,x\) to express \(\varepsilon_{HF}^{i}\) in terms of Coulomb and exchange integrals, and then, using Eq. (2.56), we can write:

\[
\langle \Psi^{HF} | \mathcal{H} | \Psi^{HF} \rangle = \sum_{i=1}^{N_{e}} \varepsilon_{HF}^{i} - \frac{1}{2} \sum_{i,j=1}^{N_{e}} \left( V_{i;i;i,j;i,j} - V_{i;i,j;i,j;i,j} \right). \quad (2.63)
\]

Here we have used the notation of Appendix A for the Coulomb matrix elements \(V\), which are computed with respect to the H-F orbitals \(\phi_{i}(x)\) determined from Eq. (2.62).
2.3 The Hartree-Fock Method

2.3.2 Roothaan’s Equations

In Chapter 4 we will show H-F results for the ground state (and first excited state) of a single vertical QD. In this Subsection we shall describe the algorithm we numerically implemented. Instead of trying to solve directly the integro-differential equations (2.62), it is much more practical to build up approximate spin-orbitals \( \phi_{\xi}(x) \) as linear combinations of the single-particle solutions of \( H_0(r) \). We can set up such linear combinations, with adjustable coefficients; introduce them into the determinantal function; and vary the resulting orbitals to make the energy stationary. This variation procedure results in algebraic equations for the coefficients, rather than in differential equations as in (2.62). If we had enough atomic orbitals so that linear combinations of them could represent the true orbitals with complete accuracy, the resulting linear combinations would be exact solutions of Eq. (2.62). In actual cases, however, we use a truncated basis set. This scheme was suggested by Roothaan[128].

We start by assuming a finite number \( N_b \) of unperturbed orbitals \( \chi_{\alpha \sigma}(r) \), eigenfunctions of the single-particle Hamiltonian \( H_0(r) \) of Eq. (2.16). In terms of them, we expand the spin-orbitals of the H-F method according to the equation

\[
\phi_{\alpha \sigma}(r) = \sum_{p=1}^{N_b} c_p(\alpha \sigma) \chi_{\alpha_p}(r).
\]  

(2.64)

Note that here we consider only the spatial part of the spin-orbitals. \( \chi_{\alpha \sigma}(r) \) does not depend on the spin, instead of \( \phi_{\alpha \sigma}(r) \). Remember that \( \alpha \) stands for a suitable set of spatial quantum numbers. Now we substitute the expansion (2.64) into the H-F equations (2.62) (after having performed the sum over spin variables), multiply by \( \chi_{\alpha \sigma}(r) \) and integrate over \( d \mathbf{r} \). Finally we obtain[129]

\[
\sum_{p=1}^{N_b} c_p(\alpha \sigma) [F_{qp}(\alpha \sigma) - \delta_{q,p} \varepsilon_{\alpha \sigma}^{\text{H-F}}] = 0 \quad q = 1, \ldots, N_b
\]  

(2.65)

where

\[
F_{qp}(\alpha \sigma) = \delta_{q,p} \varepsilon_{\alpha q} + \sum_{\beta \sigma'} \sum_{l, \tilde{r} = 1}^{N_b} c_{l}^{*} (\beta \sigma') c_{\tilde{l}} (\beta \sigma') \left[ V_{\alpha \sigma, \beta \sigma', \beta \sigma, \alpha \sigma} \right] - V_{\alpha \sigma, \beta \sigma', \alpha \sigma, \beta \sigma'} \right]
\]  

(2.66)
and
\[ \tilde{\varepsilon}_{\alpha q} = \int d\mathbf{r} \chi_{\alpha q}^* (\mathbf{r}) H_0 (\mathbf{r}) \chi_{\alpha q} (\mathbf{r}) \] (2.67)
and the Coulomb matrix elements \( V \) are computed with respect to the basis functions \( \chi \). Note that the exchange integrals \( V_{\alpha \sigma, \beta \sigma'} \) on the second line of Eq. (2.66) are zero unless \( \sigma = \sigma' \).

The equations (2.65) form a set of \( N_b \) simultaneous linear homogeneous equations in the \( N_b \) unknowns \( c_p (\alpha \sigma) \), \( p = 1, \ldots, N_b \), which describe the spin-orbital \( \phi_{\alpha \sigma} (\mathbf{r}) \) in Eq. (2.64). For a non trivial solution, we must have
\[ \det \left[ F_{qp} (\alpha \sigma) - \delta_{q,p} \varepsilon_{\alpha \sigma}^{\text{HF}} \right] = 0. \] (2.68)
This is a secular equation whose \( N_b \) roots give the spin-orbital energies \( \varepsilon_{\alpha \sigma}^{\text{HF}} \). We number these in order of energy, from \( (\alpha \sigma)_1 \) to \( (\alpha \sigma)_{N_b} \), and it is this index which we have indicated as \( (\alpha \sigma) \) in Eq. (2.65). If \( N_b \) is greater than the number of occupied states \( (\alpha \sigma) \), as it usually is, we must then make an assignment of electrons to spin-orbitals, and when we are dealing with the ground state, we naturally assign them to the spin-orbitals of lowest energy.

Equations (2.65) must be solved by an iterative process, since the \( F_{qp} (\alpha \sigma) \) matrix depend on the spin-orbitals themselves, which in turn depend on the unknown coefficients \( c_p (\alpha \sigma) \). In our numerical calculations, we found good convergence with \( N_b = 4 \). For the actual computation of the Coulomb integrals, which is done once and for all before initializing the self-consistent procedure, see Appendix B.

2.3.3 Discussion

We do not mean here to discuss in a general way the H-F method: the subject is old and well known. However, there are some points which are worth stressing in this context, with relation to the study of QDs’ ground state electronic configurations by means of any mean-field theory.

- Let’s focus on the case of a single vertical QD at zero or low magnetic field. We anticipate that a shell model, analogous to the Periodic Table of Elements of Atomic Physics, is presently accepted to explain the main features of electronic structure, the strongest argument being the experimental evidence[28]. For closed shells, it can be shown[127] that the H-F
potential is perfectly central. On the other hand, the phenomenon called *self-consistent symmetry* is known to occur in the iterative procedure for solving the H-F equations[126], namely the symmetry of the initial trial wavefunction is preserved in all the successive improvements of the wavefunction during the recursive steps of the computation. Thus, if one chooses as starting wavefunction an orbital solution of the “bare” potential which has central symmetry, there are no inconsistencies in the theory.

- However, in the case of an open shell, where “valence” electrons do not occupy all possible degenerate spin-orbitals, the H-F potential in general has not central symmetry[127]. To impose a central-field type self-consistent symmetry is then a further approximation within the H-F method itself. The aim is to build global electronic densities with the correct symmetry properties expected *a priori* from the solution of the true Hamiltonian $\mathcal{H}$[130].

- Moreover, in these cases, a single Slater determinant is not an irreducible representation of the symmetry group of the true Hamiltonian $\mathcal{H}$. One is forced to set up linear combinations of Slater determinants with the correct symmetry properties, according to the Theory of Multiplets of Atomic Physics. Actually, this is not a big trouble, since it just corresponds to transform the Slater determinant basis set into another one by a unitary transformation. At the end of this “restoring-symmetry” process, the energy configuration is given by the average of the initial Hamiltonian $\mathcal{H}$ on a single vector of the Hilbert space.

- As we have just seen, the H-F potential in general has not the same symmetry of the true Hamiltonian $\mathcal{H}$. On the basis of this argument, one could introduce a *symmetry-violating mean field approach* to describe *phase transitions* to a symmetry-violating state. This could be done, by example, relaxing the condition that the H-F potential must be of central-field type, which is actually just an approximation in most cases. In the present context of finite systems, by spontaneous symmetry breaking we mean the formation of ground states of lower symmetry than that of the confining potential[126]. This approach has already been employed in the nuclear many-body prob-
lem, and there were recent claims of spontaneous symmetry breaking in QDs at low $B$ on the basis of mean-field calculations[78, 81, 79]. Caution is needed, however, when considering the latter results, because:

- According to general principles of Quantum Mechanics, the ground state of the system must be a representation of the symmetry group of the true Hamiltonian $\mathcal{H}$: one should try then to restore in some way the symmetries broken in the mean-field approach.[78]

- The H-F method is based on a variational principle, and because of the self-consistent symmetry of iterative solutions, if we start with a certain symmetry, we will always stay within this symmetry, and the minimum of energy can only be found among the wavefunctions that have this symmetry. Indeed, there is no guarantee that one has found the lowest energy configuration or that he has found the trial wavefunction with the correct physical behaviour, which is even more important.[131]
Chapter 3

Theory II: Many-Electron States

Continuing the exposition of the theoretical model, in this Chapter we face the true many-body problem. In Section 3.1 we derive a model Hamiltonian which describes the interacting electronic system, and which we use to compute $N_e$-dependent ground state energies and addition spectra. In particular, after a general introduction to the problem, in Subsection 3.1.1 we discuss the single-particle basis used, then we derive the Generalized Hubbard Hamiltonian (Subsec. 3.1.2), and eventually we show how to diagonalize the Hamiltonian exploiting the symmetries of the problem (Subsec. 3.1.3). In view of applications in Chapters 4, 5, and 6, we then review (Sec. 3.2) the Wigner-von Neumann theorem, which is a very general statement about how crossing of electronic terms can occur, varying some parameters of the system.

3.1 Generalized Hubbard Hamiltonian

Solving the problem of $N_e$ electrons in QDs exactly is an impossible task, since already with $N_e = 3$ the analogous problem in classical mechanics has only approximate solutions. One should find eigenvalues and eigenvectors of the total many-body Hamiltonian $\mathcal{H}$ (see Appendix A)

$$
\mathcal{H} = \sum_{\alpha \sigma} \varepsilon_{\alpha \sigma} n_{\alpha \sigma} + \sum_{\alpha \beta} t_{\alpha \beta}^0 c_{\alpha \sigma}^\dagger c_{\beta \sigma}^\dagger c_{\beta \sigma} c_{\alpha \sigma}
$$

$$
+ \frac{1}{2} \sum_{\alpha \sigma} \sum_{\beta \sigma'} \sum_{\gamma \sigma''} \sum_{\delta} V_{\alpha \sigma, \beta \sigma', \gamma \sigma''} \delta_{\sigma \delta} c_{\alpha \sigma}^\dagger c_{\beta \sigma'}^\dagger c_{\gamma \sigma''}^\dagger c_{\delta \sigma}, \quad (3.1)
$$
Figure 3.1: Two vertically coupled quantum dots. Dots 1 and 2 correspond to GaAs quantum wells confined by $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barriers. The $z$-axis is the growth direction, and $d$ is the width of the inter-dot barrier. Varying $d$ means to tune the coupling between the two dots. If the value of $d$ is large, the coupling is just of electrostatic nature, while if the inter-dot barrier is thin quantum-mechanical tunneling effects occur.

where the basis set of arbitrary single-particle states $\{ \varphi_{\alpha\sigma}(r,s) \}_{\alpha\sigma}$, over which $\mathcal{H}$ is written, is infinite and complete. If the number of electrons is very small, however, we could directly make $\mathcal{H}$ diagonal using only a reduced truncated basis set. The accuracy of solutions is controlled by enlarging the set and checking if the further corrections introduced are smaller than the error tolerated. Still, as the number of electrons $N_e$ increases, matrix dimensions become rapidly so large that the method of exact diagonalization can be implemented in QDs only for very few electrons[45, 52].

We choose then to build a many-body model Hamiltonian, replacing a computational problem with a physical one. The obvious requirement is that this model Hamiltonian should embody the essential physics of QDs. We are then forced to accurately consider the specific electronic problem under study.
As already stressed in Chap. 2, we consider vertical QDs like those experimentally studied in Refs. [28, 34]. These devices have shown a very rich phenomenology of correlated electronic states. As paradigmatic system we take the pillar of two vertically coupled QDs depicted in Fig. 3.1, the single-dot system being a special case. To describe the motion of correlated electrons in this system, we can write down an approximate Hamiltonian exploiting the analogy of QDs with natural molecules. If dots are far apart, the interaction between them is purely electrostatic and electrons are localized on each isolated QD: a “molecular” wavefunction of Heitler-London type should be a very good approximation to the true many-body state. On the other hand, if dots are very close, electrons can tunnel into the inter-dot barrier and the whole system should behave as a coherent entity in which electrons are completely delocalized. Intermediate regimes are controlled by the competition between these two opposite effects, i.e. localization vs delocalization: the approximate scheme better describing this balance is the Hubbard model.

The Hubbard model was derived starting from a set of single particle states (Wannier functions) localized on different sites of a lattice[132]. To do the same with an array of QDs, we need a complete set of suitable single-particle orbitals localized on each dot. This task is non-trivial at all.

### 3.1.1 The Choice of the Basis

We have already solved the single-particle problem for the double-dot system in Sec. 2.2. The spatial part of the wavefunction, eigenvector of the one-body Hamiltonian $H_0(\mathbf{r})$ of Eq. (2.16), is

$$
\phi_{nm\ell}(\mathbf{r}) = \psi_{nm}(\rho, \varphi) \chi_{\ell}(z),
$$

(3.2)

where $\psi_{nm}(\rho, \varphi)$ is a Fock-Darwin state and $\chi_{\ell}(z)$ is the wavefunction for the motion along $z$. The index $\ell$ assumes two values ($\ell = s, a$ for symmetric and antisymmetric states, respectively) for the double dot and only one ($\ell = s$) for the single dot. Even if $\phi_{nm\ell}(\mathbf{r})$ is a solution of $H_0(\mathbf{r})$, it is not a suitable basis for our purposes, since it represents an electron delocalized on both dots (see upper panel of Fig. 3.1).
of Fig. 2.3). However, performing the unitary transformation

$$
|1\rangle = (|s\rangle + |a\rangle) / \sqrt{2},
$$

$$
|2\rangle = (|s\rangle - |a\rangle) / \sqrt{2},
$$

(3.3)

where $|l\rangle$ stands for the orbital $\phi_{nm\ell}(r)$, one obtains a new basis set $\phi_{nm\ell}(r)$ [with $i = 1, 2$], which is still complete, orthonormal, as it is easy to check, and, in addition, localized on one dot or the other. The lower panel of Fig. 2.3 shows orbitals $\phi_{nm1}(r)$ and $\phi_{nm2}(r)$ localized on dot 1 and 2, respectively. If we now write Eq. (3.1) for the total Hamiltonian $H$ in terms of the new localized basis $\phi_{nm\ell}(r)$, making the index $\ell = (n, m, i)$ explicit, we obtain that many parameters $\varepsilon_{nm\ell}^0$, $t_{nm\ell,m'n'i'}^0$ are zero, because of the cylindrical symmetry of the system [see Eqs. (A.13) and (A.14), respectively]:

$$
\varepsilon_{nm\ell}^0 = \varepsilon_{nm} + (\varepsilon_s + \varepsilon_a) / 2,
$$

$$
t_{nm\ell,m'n'i'}^0 = \delta_{n,m'}\delta_{m,m'}(1 - \delta_{i,i'})(\varepsilon_a - \varepsilon_s) / 2.
$$

(3.4)

Here $\varepsilon_{nm}$ is the energy of a Fock-Darwin state and $\varepsilon_s, \varepsilon_a$ are the energies of symmetric and antisymmetric states along $z$, respectively. Thus, on this new localized basis, the total Hamiltonian $H$ becomes:

$$
H = \sum_{nm\ell} \left[ \varepsilon_{nm} + (\varepsilon_s + \varepsilon_a) / 2 \right] n_{nm\ell} - t \sum_{nm\ell} \sum_{i,i'} c_{nm\ell}^\dagger c_{nm'i'}
$$

$$
+ \frac{1}{2} \sum_{\sigma\sigma'} \sum_{n11} \sum_{n22} \sum_{m11} \sum_{m22} \sum_{i1} \sum_{i2} V_{n1m11\sigma,n2m22\sigma';n3m3i\sigma,n4m4i\sigma}
$$

$$
\times c_{n1m11\sigma}^\dagger c_{n2m22\sigma'}^\dagger c_{n3m3i\sigma'} c_{n4m4i\sigma},
$$

(3.5)

where we have defined the hopping coefficient $t$ as

$$
t = (\varepsilon_a - \varepsilon_s) / 2,
$$

and the prime on the sum means that $i \neq i'$. Until now, there is no approximation in (3.5).

### 3.1.2 Single and Coupled Vertical Quantum Dots

To obtain the model Hamiltonian we must simplify the two-body Coulomb term. Let’s consider [see Eq. (A.15)]

$$
V_{n1m11\sigma,n2m22\sigma';n3m3i\sigma,n4m4i'\sigma} =
$$
\[ e^2 \int \int \frac{\phi_{n_1 i_1}^\sigma (\mathbf{r}) \phi_{n_2 i_2}^\sigma (\mathbf{r}') \phi_{n_3 i_3}^{\sigma'} (\mathbf{r}') \phi_{n_4 i_4}^{\sigma'} (\mathbf{r})}{\kappa_\mathbf{r} |\mathbf{r} - \mathbf{r}'|} \, d\mathbf{r} d\mathbf{r}'. \] (3.6)

First we note that, because in a two-body scattering process the total orbital angular momentum is kept constant by Coulomb interaction, the only non-zero matrix elements are those with
\[ m_1 + m_2 = m_3 + m_4. \]

Then, we focus on states localized all on the same dot: i.e.
\[ i_1 = i_2 = i_3 = i_4 = i. \]

In this latter case the Coulomb matrix elements do not depend on the \( i \) index. Furthermore, among all the other terms, we isolate the “semi-diagonal” ones, namely those with
\[ (n_1, m_1) = (n_4, m_4) \quad \text{and} \quad (n_2, m_2) = (n_3, m_3), \] (3.7)

which we adopt as Coulomb (or “direct”) integrals \( U_{n_1 i_1; n_2 i_2} \)
\[ U_{n_1 i_1; n_2 i_2} = V_{n_1 i_1; i_2; n_2 i_2; n_1 i_1}, \]

and those with
\[ (n_1, m_1) = (n_3, m_3) \quad \text{and} \quad (n_2, m_2) = (n_4, m_4), \] (3.8)

which we call Exchange integrals \( J_{n_1 i_1; n_2 i_2} \)
\[ J_{n_1 i_1; n_2 i_2} = V_{n_1 i_1; n_2 i_2; n_1 i_1; n_2 i_2}. \]

Note that “intra-dot” direct and exchange integrals do not depend on the dot where electrons are localized. Following a similar procedure, we isolate also “inter-dot” terms with
\[ i_1 = i_4 \quad \text{and} \quad i_2 = i_3, \]
\[ (n_1, m_1) = (n_4, m_4) \quad \text{and} \quad (n_2, m_2) = (n_3, m_3), \] (3.9)

which we call inter-dot Coulomb terms \( \tilde{U}_{n_1 i_1; n_2 i_2} \): 
\[ \tilde{U}_{n_1 i_1; n_2 i_2} = V_{n_1 i_1; i_2; n_2 i_2; n_1 i_1}. \] (3.10)
Also inter-dot Coulomb terms $\tilde{U}_{n_1m_1;n_2m_2}$ do not depend on dot-site. We do not take into account inter-dot Exchange terms, which are usually very small. All direct and exchange terms are positive definite quantities.

Now we are ready to write down the model Hamiltonian. We achieve this goal separating the two-body part of $\mathcal{H}$ into a contribution where the four-operator product $c_\uparrow_{\ell_1} c_\downarrow_{\ell_2} c_{\xi_1} c_{\xi_2}$ is just given by the product of two number operators $n_{\xi_1} n_{\xi_2}$ and into another one which contains all other terms:

$$
\mathcal{H} = \frac{\varepsilon_s + \varepsilon_a}{2} \sum_{nm\sigma} n_{nm\sigma} + \sum_{nm\sigma} \varepsilon_{nm} n_{nm\sigma} - t \sum_{nm\sigma} c_{nm\sigma}^\dagger c_{nm2\sigma} + \text{h.c.} + \frac{1}{2} \sum_{n_1m_1n_2m_2} U_{n_1m_1;n_2m_2} \sum_{i\sigma} n_{n_1m_1i\sigma} n_{n_2m_2i-\sigma} + \frac{1}{2} \sum_{n_1m_1n_2m_2} \left[ U_{n_1m_1;n_2m_2} - J_{n_1m_1;n_2m_2} \right] \sum_{i\sigma} n_{n_1m_1i\sigma} n_{n_2m_2i\sigma} + \sum_{n_1m_1n_2m_2} \tilde{U}_{n_1m_1;n_2m_2} \sum_{\sigma\sigma'} n_{n_1m_11\sigma} n_{n_2m_22\sigma'} + \frac{1}{2} \sum_{\text{other} \; \text{terms}} V_{n_1m_1i_1\sigma,n_2m_2i_2\sigma',n_3m_3i_3\sigma',n_4m_4i_4\sigma} \times c_{n_1m_1i_1\sigma}^\dagger c_{n_2m_2i_2\sigma'}^\dagger c_{n_3m_3i_3\sigma'} c_{n_4m_4i_4\sigma} \cdot (3.11)
$$

The important point here is that the Coulomb matrix elements appearing in the last sum of (3.11) are always smaller than the direct and exchange terms $U, U - J$ and $\tilde{U}$ appearing in the previous lines. Indeed, we checked that the biggest “off-diagonal” terms are of the order of the exchange term $J$, while $J$ is always significantly smaller (something less than one order of magnitude, on the average) than the corresponding direct term. The reader can check this fact on his own directly in Appendix B for the case of a single two-dimensional QD. The approximation we choose consists in neglecting all these “off-diagonal” terms. We define then a Generalized Hubbard Hamiltonian (GHH) $\mathcal{H}^{\text{GHH}}$ such that

$$
\mathcal{H} \approx \mathcal{H}^{\text{GHH}}, \quad (3.12)
$$

where

$$
\mathcal{H}^{\text{GHH}} = \frac{\varepsilon_s + \varepsilon_a}{2} \sum_{nm\sigma} n_{nm\sigma} + \sum_{nm\sigma} \varepsilon_{nm} n_{nm\sigma} - t \sum_{nm\sigma} c_{nm\sigma}^\dagger c_{nm2\sigma} + \text{h.c.}
$$
This is the Hamiltonian we actually diagonalize for a system of two coupled QDs. The first two addenda are single-particle on-site terms, the third is the hopping term, the fourth one accounts for the intra-dot Coulomb repulsion between electrons with antiparallel spin, the fifth one refers to electrons with parallel spin (an exchange contribution must be subtracted to the direct one, in this latter case), while the last one is the inter-dot electrostatic interaction.

In the case of a single QD, we can suppress the site index \( i \), and the GHH Hamiltonian reduces to

\[
\mathcal{H}^{GHH} = \sum_{nm\sigma} \varepsilon_{nm} n_{nm\sigma} + \frac{1}{2} \sum_{n_1 m_1 n_2 m_2} U_{n_1 m_1; n_2 m_2} \sum_{\sigma} n_{n_1 m_1 \sigma} n_{n_2 m_2 - \sigma} \\
+ \frac{1}{2} \sum_{n_1 m_1 n_2 m_2} [U_{n_1 m_1; n_2 m_2} - J_{n_1 m_1; n_2 m_2}] \sum_{\sigma, \sigma'} n_{n_1 m_1 \sigma} n_{n_2 m_2 \sigma'},
\]

(3.14)

where we have suppressed a \( N_e \)-dependent constant term. In the single QD case, of course, there are no hopping term and inter-dot electrostatic coupling. Note that the single-dot Hamiltonian (3.14) commutes with the number operator \( n_{nm\sigma} \), while the double-dot Hamiltonian (3.13) does not. This means that, while in the first case single Slater determinants are eigenstates of \( \mathcal{H}^{GHH} \), in the second case we have to diagonalize the Hamiltonian matrix in order to find its eigenvalues.

### 3.1.3 Diagonalization and Addition Spectra

It is very convenient to exploit the symmetries of Hamiltonian (3.13), to reduce the dimensions of subspaces over which \( \mathcal{H}^{GHH} \) must be diagonalized. In this regard, it is evident by inspection of Eq. (3.13) that the number of electrons \( N_e \), the total orbital angular momentum (projection along the \( z \)-axis) \( L_z \), and the projection of the total spin along the \( z \)-axis \( S_z \) commute with the Hamiltonian (3.13).
Thus, the practical algorithm we used to extract eigenvalues and eigenvectors from $\mathcal{H}^\text{GHH}$ is the following:

- For every set of external parameters, such as an homogeneous external static magnetic field $B$ or a given value $t$ for the inter-dot coupling, we compute direct and exchange integrals appearing in Eq. (3.13). While for a two-dimensional disk these latter are analytically known (see Appendix B), in general we are forced to compute them numerically.

- On each subspace labeled by a different value of $N_e$, $S_z$, and $L_z$, the Hamiltonian $\mathcal{H}^\text{GHH}$ is diagonalized and the minimum energy state of that given subspace is found: the true ground state of the system is the absolute minimum over the entire set of quantum numbers allowed.

- From the knowledge of the $N_e$-dependent ground state of the system $E_0(N_e)$ one obtains in a straightforward way the addition energy $A(N_e)$, i.e. the quantity to compare with experimental measures:

$$A(N_e) = E_0(N_e) - E_0(N_e - 1).$$ (3.15)

Actually we considered QDs so weakly coupled to the leads that it is a very good approximation to treat dots as isolated systems (as we implicitly did): the addition energy $A(N_e)$ is then the resonance energy at which an electron can tunnel between this “closed” system and the leads, idealized as electron reservoirs [see Eq. (1.7)].

### Double Dot Symmetries

Here we spend some words on symmetry notations. Ground state (or excited state) configurations, frequently called “electron terms”, are labeled in the molecular notation: $\Sigma$, $\Pi$, $\Delta$, $\Phi$, $\Gamma$, $I$ correspond to the total orbital angular momentum $M = 0, \ldots, 5$, and the left superscript is $2S_z + 1$:

$$^{2S_z+1}M.$$ 

Actually, we forgot other two symmetry operations for the double dot: (i) inversion with respect to the center of the molecule $r = 0$, (ii) reflection with respect to
3.2 Symmetries and Wigner-von Neumann Theorem

the plane $z = 0$. Under both transformation (i) and (ii) the many-body states have a definite parity. States are labeled with the right subscript $g$ (even) or $u$ (odd) according to transformation (i), and by the right superscript $+$ (even) or $-$ (odd) according to (ii). To summarize the notation, we use the symbol $^{2S_z+1}M_{g/u}$.

It is worth mentioning that the above notation is different from the standard molecular one because of the presence of the magnetic field. In fact, in the standard notation one has as left superscript $2S + 1$ instead of $2S_z + 1$, where $S$ is the total spin and $2S + 1$ is the molteplicity, and as right superscript an index $\pm$ which refers to the reflection with respect to a plane containing the symmetry axis of the molecule instead of the plane orthogonal to it. If $B = 0$, the two reflection symmetries are not independent, but one can be obtained from the other through the inversion symmetry $g/u$. Moreover, the total spin vector is conserved (in the total Hamiltonian $H$, not in $H^{GHH}$), not only its projection along $z$, and the degeneracy is actually $2S + 1$. $B$ removes the spin degeneracy, by means of the Zeeman coupling term, and a reflection with respect to a plane containing the symmetry axis of the molecule is no more a symmetry operation. In this regime, the superscript $2S_z + 1$ does not mean any spin degeneracy.

3.2 Symmetries and Wigner-von Neumann Theorem

We mentioned that ground states configurations depend on parameters such as $B$ or $t$. The question which arises is whether we are allowed to observe transitions, i.e. crossing of electron terms, as far as we vary such parameters. The relevance of this issue is twofold: (i) The “phase space” of artificial molecules is much larger than that of “natural” molecules. In particular, in natural molecules the internuclear distance is almost fixed, while we can tune as much as we like the inter-dot coupling. Another example is the relevance of magnetic field on electronic motion in QDs, which is much bigger than in the “natural case” (this is due to the fact that, at the present time, maximum values of $B$ reachable in laboratories are comparable to typical QDs’ energy scales but are order of magnitude smaller than typical chemical energies). (ii) These transitions can be experimentally observed.
The answer to the question whether electron-term crossings are allowed is positive, according to the so-called Wigner-von Neumann theorem (proved in Appendix C).

*If electron terms depend on one parameter, only terms of different symmetry can intersect, while the intersection of terms of like symmetry is impossible.*

By *symmetry* we here understand all possible forms of symmetry: with respect to rotations about an axis, reflections in planes, inversion, and also with respect to interchanges of electrons. In coupled QDs this means that we may be dealing with terms of different $L_z$, different $S_z$, different parity. We emphasise that this result not only is true for the coupled QD system, but is a general theorem of Quantum Mechanics; it holds for any case where the Hamiltonian contains some parameter and its eigenvalues are consequently functions of that parameter. In particular, in Chapter 6 we analyze electron terms of coupled dots and find that, as the inter-dot distance varies, terms with different values of spin and angular momentum cross.
Chapter 4

Addition Spectra in Single Vertical Quantum Dots

Our work was inspired by experimental findings of Tarucha and coworkers[28], hence Figs. 1 and 2 of the paper which display our theoretical results for addition spectra should be compared directly with data of Ref. [28]. For the convenience of the reader, we have reproduced these data below.
Figure 4.1: (a) Coulomb oscillations in the current vs gate voltage at $B = 0$ T observed for a 0.5 $\mu$m-diameter dot. (b) Addition energy variation (labeled “addition energy” in the figure) vs electron number for two different dots with diameter 0.5 $\mu$m and 0.44 $\mu$m, respectively. The inset shows a schematic diagram of the device. The dot is located between the two heterostructure barriers. After Ref. [28].

Figure 4.2: Measured evolution of the third, fourth, fifth, and sixth addition spectrum current peaks with $B$ field from 0 to 2 T observed for the 0.5 $\mu$m-diameter dot of Fig. 4.1. After Ref. [28].
Addition energies in semiconductor quantum dots: Role of electron–electron interaction

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(Received 18 November 1997; accepted for publication 24 December 1997)

We show that the addition spectra of semiconductor quantum dots in the presence of magnetic field can be studied through a theoretical scheme that allows an accurate and practical treatment of the single-particle states and electron–electron interaction up to large numbers of electrons. The calculated addition spectra exhibit the typical structures of Hund-like shell filling, and account for recent experimental findings. A full three-dimensional description of Coulomb interaction is found to be essential for predicting the conductance characteristics of few-electron semiconductor structures. © 1998 American Institute of Physics. [S0003-6951(98)03608-0]

In semiconductor quantum dots (QDs) Coulomb-correlation effects are expected to become more and more pronounced as their spatial confinement is increased. This has been so far one of the reasons of interest for such systems: they display the rich shell structure typical of atomic physics, but in addition they offer the possibility to tailor the confining potential and to vary the electron occupation by adjusting external parameters, thus allowing direct investigation of fundamental properties of charge correlation.

At the same time, however, these results imply that many-body effects cannot be neglected in the theoretical description of QDs, and may be very relevant for predictions on their possible applications, for example, in semiconductor-based few-electron devices. While capacitance and tunneling spectroscopy experiments are becoming available for different dot structures, it is still an open and difficult problem to devise theoretical approaches allowing to include many-body effects in calculations for realistic systems. Exact calculations are necessarily limited to very few electrons; state-of-the-art QD structures instead involve several electrons (of the order of ten or hundred), therefore in general out of reach for these methods.

In this letter we propose an accurate and effective theoretical scheme that includes electron–electron interaction and can be applied to systems up to a large number of electrons. This approach can treat arbitrary three-dimensional (3D) confinement potentials, and hence deal with realistic QD structures also in the presence of external magnetic field. We apply this scheme to structures that were investigated in a recent experiment. The results are in good quantitative agreement with the observed spectra, and provide a transparent interpretation of their physical origin.

A key quantity that characterizes transport into a QD is the addition energy, i.e., the energy $A(N)$ required in order to place an extra electron into a dot that is initially occupied by $N-1$ particles. Such quantity, analogous to electron affinity in atomic physics, can be measured experimentally as a function of $N$. A systematic experiment on disk-shaped dots has shown that the voltage increment $\Delta A$ between successive single-electron tunneling processes—i.e., between two successive maxima in the conductance—peaks at “magic” values of $N$ ($N=2,6,12$), consistent with the filling of complete shells calculated for a dot of the same symmetry. Moreover, the experimental analysis showed that unusually large values of $\Delta A$ also occur for values of $N$ corresponding to half-shell filling ($N=4,9$). The origin of these features, reminiscent of Hund’s rule in atomic physics, is intimately related to electron–electron interaction; therefore their quantitative evaluation is a challenge for any theoretical analysis of addition spectra in QDs.

Our description is based on an accurate treatment of the single-particle Hamiltonian and on the inclusion of Coulomb correlation according to first-order perturbation theory: assuming the many-particle ground state to be described by a single Slater determinant, the total energy of the full interacting Hamiltonian is

$$E(N) = \sum_{a\sigma} \epsilon_a(\hat{n}_{a\sigma}) + \frac{1}{2} \sum_{a\beta} \left[ U_{a\beta} (\hat{n}_{\beta-a}) + (U_{a\beta} - J_{a\beta}) (\hat{n}_{\beta-a}) \right],$$

(1)

where $(\hat{n}_{a\sigma})$ is the orbital occupation number, $a$ denoting the set of radial and angular quantum numbers $(n,m)$, and $\sigma$ the spin value. According to Eq. (1), the evaluation of the energies for the various many-electron states in the dot requires only the knowledge of the Coulomb and exchange matrix elements

$$U_{a\beta} = e^2 \int \frac{|\phi_a(r)|^2 |\phi_{a'}(r')|^2}{\kappa |r-r'|} \, dr dr',$$

(2)

$$J_{a\beta} = e^2 \int \frac{\phi_a^*(r) \phi_{a'}^*(r') \phi_{a'}(r) \phi_{a'}(r')}{\kappa |r-r'|} \, dr dr'.$$

(3)

Here $\kappa$ is the dielectric constant and $\phi_a$ are single-particle real-space eigenstates of the dot in the presence of the applied magnetic field.

The important feature of the present scheme is that $U_{a\beta}$, $J_{a\beta}$ are calculated directly, contrary to other common approaches where Coulomb and exchange integrals are taken as input parameters. Thus, many-body effects are taken into...
account through (1), and quantum-confinement effects through the single-particle ingredients, energies $\epsilon_a$ and wave functions $\psi_a$; the latter enter only in the calculation of Coulomb and exchange matrix elements. This has the advantage that realistic confining potentials (e.g., extracted from high-resolution transmission electron microscope (TEM) micrographs) and an external magnetic field can be included quite easily by choosing the appropriate single-particle Hamiltonian, as is now commonly done for the calculation of optical properties.\(^{12}\)

Note that previous approaches have modeled the QD confinement in terms of a purely two-dimensional (2D) coordinate space.\(^7\) As a matter of fact, in most state-of-the-art QD structures the confinement is far from being 2D: for example, in the gated dots of Ref. 5 the typical confinement width in the growth direction $z$ (of the order of 10 nm) is comparable with the typical extension of the carrier ground state in the lateral-confinement ($xy$) plane. Therefore, in our calculations we fully retain the 3D nature of the problem and mimic the dots of Ref. 5 by assuming a finite-barrier quantum-well potential along $z$ and a parabolic potential in ($xy$). Note that the assumption of parabolic potentials reflects the experimental indications due to the specific sample structure and is not required by our model. Within the usual envelope-function approximation, the only input parameters are the electron effective masses ($m^* = 0.065m_0$ in the dot and $m^* = 0.079m_0$ in the barrier), the dielectric constant in the dot ($\kappa = 12.98$), and the quantum-well height and width ($V_0 = 200$ meV and $L = 12$ nm).

Our results for the addition-energy variations, $\Delta A(N) = A(N+1) - A(N)$, are displayed in Fig. 1 as a function of the electron number $N$ for different dots, characterized by different in-plane confinement energies $\hbar\omega_0$. Here, $A(N)$ is obtained as $E(N) - E(N-1)$, where $E(N)$ is the ground-state energy in Eq. (1). As we can see, $\Delta A(N)$ exhibits peaks corresponding both to complete and half-shell filling, and is in excellent agreement with experiments in Ref. 5.

The origin of this result is in the delicate interplay between single-particle contributions and electron–electron repulsion, which according to Eq. (1) defines the ground-state configuration: the single-particle term favors complete shell filling, while the repulsion among parallel-spin electrons, smaller than the repulsion among opposite-spin ones, makes the configurations with maximum total spin energetically favored. Hund’s rule is therefore already contained in Eq. (1) and clearly explains the physical origin of the half-shell-filling structures. In fact, adding an electron to a half-filled shell forces the double occupancy of a level; consequently, $\Delta A$ is raised by the dominant Coulomb repulsion $U_{\alpha\alpha}$ between opposite-spin electrons on the same level.

Ground-state configurations and filling rules change when a magnetic field is applied. It affects both single-particle energies and $U$ and $J$ integrals through the induced changes in the wave function localization. For sufficiently large values, the magnetic field energetically favors configurations with higher total angular quantum number $m$. This is the physical origin of the wiggles in the $A(N)$ vs $B$ plot shown in Fig. 2 and observed in the experiments of Ref. 5.

Figure 3 shows the Coulomb and exchange integrals vs $B$ for the first states, obtained for $\hbar\omega_0 = 7.5$ meV. For comparison, we also show the corresponding quantities calculated within the simplified 2D model. We clearly see that $U$ integrals describing the interaction between opposite-spin electrons are smaller by a few meV in the case of 3D confinement, while the differences in the interaction between parallel-spin electrons are much smaller. This affects dramatically the energy balance which determines ground-state configurations, thus clearly showing the inadequacy of a pure 2D description of state-of-the-art QD structures.

The proposed approach shares with the Hartree–Fock (HF) method the ansatz for total energy, expressed as the average of the exact Hamiltonian over a single Slater determinant; the variational prescription—allowing the construction of optimal single-particle orbitals through self-consistency—is not contained in our approach. We notice, however, that in the present case the results of perturbation theory are already much closer to the exact results than in previous 2D calculations.\(^{12}\) This is due to the fact that Cou-
lomb integrals are artificially enhanced in 2D with respect to the realistic 3D description. Hence the role of self-consistency and the approximations of HF theory are much less relevant in 3D. Finally, we point out that the present formalism can be reduced to a Hubbard model description: expression (1) is in fact formally equivalent to the exact solution of a single-site Hubbard Hamiltonian. This is important in view of a direct extension of this approach to coupled dots.

In summary, we have proposed an effective theoretical approach for the analysis of addition spectra of quantum dots by combining a careful 3D description of electron–electron interaction with the realistic treatment of single-particle properties. By construction, the proposed scheme is ideally suited for applications to dots with confinement potentials of arbitrary shapes and symmetries. A full 3D description of Coulomb interaction is found to be crucial in determining single-electron conductance properties of realistic nanostructures.

The authors are grateful to C. Calandra, P. Hawrylak, and D. Pfannkuche for useful discussions. This work was supported in part by the EC through the TMR-Ultrafast and HCM-Psik networks.

13A direct comparison with exact results for a 3D vs 2D two-electron dot is given in M. Rontani, F. Rossi, F. Manghi, and E. Molinari (unpublished).
Chapter 5

The Importance of Electronic Correlation
The Importance of Electronic Correlation
Coulomb correlation effects in semiconductor quantum dots: The role of dimensionality

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We study the energy spectra of small three-dimensional (3D) and two-dimensional (2D) semiconductor quantum dots through different theoretical approaches (single-site Hubbard and Hartree-Fock Hamiltonians); in the smallest dots we also compare with exact results. We find that purely 2D models often lead to an inadequate description of the Coulomb interaction existing in realistic structures, as a consequence of the overestimated carrier localization. We show that the dimensionality of the dots has a crucial impact on (i) the accuracy of the predicted addition spectra, and (ii) the range of validity of approximate theoretical schemes. When applied to realistic 3D geometries, the latter are found to be much more accurate than in the corresponding 2D cases for a large class of quantum dots; the single-site Hubbard Hamiltonian is shown to provide a very effective and accurate scheme to describe quantum dot spectra, leading to good agreement with experiments.

[S0163-1829(99)10211-X]

I. INTRODUCTION

Adding an electron into a semiconductor quantum dot (QD) produces a variation in the energy of the system that depends on single-particle quantum confinement as well as on the Coulomb interaction between carriers.1 Understanding such addition-energy is a key step toward controlling the physics of single-electron devices. At the same time, the addition spectra of quantum dots offer a unique probe of few-electron interactions in regimes that are not experimentally accessible in atomic physics. The experimental effort in this direction developed very rapidly after the recent fabrication of controlled small-QD devices based on gated vertical heterostructures2 or self-assembled dots.3 The resulting addition spectra show a clear shell structure, corresponding to the symmetries of the confining potential, with a filling sequence analogous to Hund’s rule in atomic physics.

From the theoretical point of view, a general interpretation of these features was obtained by calculating the energy spectrum for a strictly two-dimensional (2D) quantum dot, and using either exact methods (for very few electrons), or approximate—usually Hartree-Fock—methods.4 The assumption of a purely 2D model was initially motivated by the typical disklike shape of the QD potential, whose extension along z is (slightly) smaller than the lateral extension of the carrier ground state in the xy plane. If one adopts a separable picture for the QD confining potential, \( V = V(z) + V(x,y) \), the relevant (i.e., lowest) single-electron states can be all associated to the ground state of \( V(z) \). From the point of view of single-particle states the 2D assumption is therefore justified.

In view of the three-dimensional (3D) nature of the Coulomb interaction, however, the 2D model introduces additional approximations in the calculation of the Coulomb integrals, which are sensitive to the spatial extension—2D vs 3D—of the single-particle wave functions.5,6 In turn, Coulomb integrals control electron-electron correlation, and influence the quantitative determination of addition spectra and their dependence on magnetic field. At the same time, the strength of the Coulomb interaction is also the key parameter determining the accuracy and range of validity of the approximations which must be introduced for dots with many electrons.

In this paper we investigate theoretically the addition spectra of realistic QD structures, with special emphasis on the effects of electron-electron repulsion and their dependence on the geometry and dimensionality of the confining potential. In Sec. II, we compare different approximate solutions of the general Hamiltonian for \( N \) interacting electrons confined in a QD structure; in particular we consider the single-site Hubbard (SSH) scheme introduced in Ref. 5, and the standard Hartree-Fock (HF) method.

In Sec. III, we focus on the simplest case, i.e., a two-electron system within a parabolic confining potential, and calculate the exact energy eigenvalues and pair-correlation functions for the 2D and 3D cases. As in Ref. 7, we use this prototypical system—called artificial or QD helium—as a reference to evaluate the accuracy of the different approximation schemes; We find that both the importance of corrections beyond the HF scheme, and the differences between HF and SSH schemes, are drastically reduced for a realistic 3D description of the dot with respect to its 2D modelization, mainly as a consequence of the reduced Coulomb integrals. This suggests the reliability of a fully 3D mean-field treatment of semiconductor QD’s.

Section IV is then devoted to the application of HF and SSH methods to 3D and 2D quantum dots with a larger number of electrons. We compare both methods for QD structures of different geometries, and demonstrate that SSH is an accurate and efficient scheme for realistic, i.e., 3D-like, dots. Finally, we discuss the implications of our results for the interpretation of recent experimental data vs magnetic field in QD structures, and draw some conclusions.

II. THEORETICAL APPROACH: EXACT FORMULATION AND APPROXIMATION SCHEMES

Our aim is to describe \( N \) electrons, confined in a QD structure (with harmonic in-plane confining potential) and
interacting via Coulomb law, possibly in the presence of an external magnetic field perpendicular to the plane. The general $N$-particle Hamiltonian is

$$\hat{H} = \sum_{i=1}^{N} \hat{H}_i(i) + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{\kappa |\mathbf{r}_i - \mathbf{r}_j|},$$

(1)

where the single-particle Hamiltonian, within the effective-mass approximation, is

$$\hat{H}_i(i) = \frac{1}{2m^*} \left( \mathbf{p} + e \mathbf{A}(r_i) \right)^2 + \frac{1}{2} m^* \omega_0^2 (x_i^2 + y_i^2) + V(z_i).$$

(2)

Here $\mathbf{A}$ is the vector potential, $\kappa$ and $m^*$ are the scalar dielectric constant and the effective electron mass in the semiconductor, $\omega_0$ is the characteristic oscillator frequency of the in-plane confining potential, and $V(z)$ is the confining potential along $z$; $V(z)$ can be chosen either as a harmonic potential $V(z) = \frac{1}{2} m^* \omega_0^2 z^2$, a square well, or a zero-width infinite barrier to describe spherical, cylindrical, or disk-shaped QD structures, respectively. Here Zeeman coupling between spin and magnetic field has been neglected.

This general Hamiltonian can be written in second quantized form on the complete and orthonormalized basis of single-particle states

$$\hat{H} = \sum_{a \alpha} c_{a \alpha}^\dagger c^a_{a \alpha} + \frac{1}{2} \sum_{a \alpha \beta \beta'} \sum_{a \beta'} V_{a \sigma \beta \alpha^* \gamma \sigma^* \delta \beta^* \alpha} c_{a \sigma \beta}^\dagger c_{a \sigma^* \beta^*} c_{a \gamma \sigma}^\dagger c_{a \gamma^* \beta^*}.$$  

(3)

Here $c_{a \alpha}$ are the eigenenergies of the one-particle Hamiltonian $\hat{H}_0$ and $c_{a \alpha}^\dagger$ the creation and destruction operators for an electron with orbital index $a$ and spin $\sigma$; and $V_{a \sigma \beta \alpha^* \gamma \sigma^* \delta \beta^* \alpha}$ are the two-body matrix elements of the electron-electron interaction

$$V_{a \sigma \beta \alpha^* \gamma \sigma^* \delta \beta^* \alpha} = \sum_{s s'} \int \phi_{a \sigma}(r, s) \phi_{a \sigma^*}(r', s') \phi_{a \beta}(r') \phi_{a \beta^*}(r', s') e^2 |r-r'| \phi_{a \gamma}(r', s') \phi_{a \gamma^*}(r', s') \phi_{a \delta}(r) \phi_{a \delta^*}(r) dr dr'$$

where $\phi_{a \sigma}(r, s) = \phi_\sigma(r) c_{a \sigma}(s)$ are the single-particle eigenfunctions.

It is useful to isolate, among the Coulomb matrix elements, the "semidiagonal" ones, namely,

$$V_{a \sigma \beta \alpha^* \gamma \sigma^* \delta \beta^* \alpha} = V_{a \sigma \beta \alpha^* \gamma \sigma^* \delta \beta^* \alpha} = U_{a \beta},$$

$$V_{a \sigma \beta \alpha^* \gamma \sigma^* \delta \beta^* \alpha} = J_{a \beta}.$$

These are the usual direct and exchange integrals which can be written more explicitly as

$$U_{a \beta} = e^2 \int \frac{\phi_\alpha^* (r) \phi_\beta^* (r') |\phi_\beta (r')|^2}{\kappa |r-r'|} dr dr'.$$

(4)

\[ J_{a \beta} = e^2 \int \frac{\phi_\alpha^* (r) \phi_\beta^* (r') |\phi_\beta (r')|^2}{\kappa |r-r'|} dr dr'. \]

(5)

In this way, Eq. (2) becomes

$$\hat{H} = \hat{H}^\text{SSH} + \frac{1}{2} \sum_{a \beta} \sum_{a \beta'} \sum_{\alpha \alpha'} V_{a \sigma \beta \alpha^* \gamma \sigma^* \delta \beta^* \alpha} \phi_{a \sigma}(r) \phi_{a \sigma^*}(r') \phi_{a \beta}(r) \phi_{a \beta^*}(r') dr dr' + \frac{1}{2} \sum_{a \beta} \sum_{\alpha \alpha'} U_{a \beta} \phi_{a \beta}(r) \phi_{a \beta^*}(r') dr dr'$$

(6)

where the prime on the first summation is to omit the terms with $\alpha = \delta, \beta = \gamma$ and $\alpha = \gamma, \beta = \delta$ and

$$\hat{H}^\text{SSH} = \sum_{a \alpha} \phi_\alpha r_{a \alpha}^\dagger r_{a \alpha} + \sum_{a \beta} (U_{a \beta} - J_{a \beta}) \hat{r}_{a \beta}^\dagger \hat{r}_{a \beta} + U_{a \beta} \hat{r}_{a \beta}^\dagger \hat{r}_{a \beta} - J_{a \beta} \hat{n}_{a \beta}^\dagger \hat{n}_{a \beta}.$$

(7)

The relevance of this formal partition is twofold: (i) it naturally leads to a perturbation expansion in the off-diagonal interactions which are in general smaller than the semidiagonal ones; (ii) moreover, the unperturbed term $\hat{H}^\text{SSH}$ is one-body-like, with single Slater determinants as exact eigenstates. The SSH approach defined in Ref. 5 consists of assuming that $\hat{H} = \hat{H}^\text{SSH}$, which amounts to neglecting the second- and higher-order contributions in the off-diagonal interactions, the first-order one being exactly zero.

The assumption that the off-diagonal $V_{a \sigma \beta \gamma \delta \alpha}$ are negligible with respect to the semidiagonal ones is implicit in all the methods which describe electron correlation in terms of the Hubbard model, either in its original form, including only on-site interaction between opposite spin electrons, proportional to $U_{a \beta}$, or adding the interaction between parallel spin electrons as well, proportional to $(U_{a \beta} - J_{a \beta})$. The important point here is that when the Hubbard model is applied to an isolated QD, i.e., to a single site, the Hubbard Hamiltonian turns out to be one-particle-like: this is so because the intersite hoppings of the traditional Hubbard Hamiltonian is absent in this case and the commutator $[\hat{H}^\text{SSH}, \hat{n}_{a \alpha}]$ is zero. As a consequence, the Slater determinants, eigenstates of the single-particle Hamiltonian $\hat{H}_0$, are exact eigenstates of $\hat{H}^\text{SSH}$ as well.

Within the SSH approach the total energy of $N$ electrons in a QD structure is given by

$$E^\text{SSH}(N) = \langle \Phi^N | \hat{H}^\text{SSH} | \Phi^N \rangle = \sum_{a \sigma} \phi_\sigma r_{a \sigma}^\dagger r_{a \sigma} + \sum_{a \beta} \left( U_{a \beta} \hat{n}_{a \beta}^\dagger \hat{n}_{a \beta} - J_{a \beta} \right) \langle \hat{n}_{a \beta}^\dagger \hat{n}_{a \beta} \rangle + \langle U_{a \beta} - J_{a \beta} \rangle \langle \hat{n}_{a \beta}^\dagger \hat{n}_{a \beta} \rangle,$$

(8)

where $\langle \Phi^N \rangle$ is a Slater determinant eigenvector of $\hat{H}_0$, and $\langle \cdot \rangle$ denotes the average over the many-particle eigenstate, which in our case simply reduces to the orbital occupation number.

The proposed SSH approach shares in common with Hartree-Fock methods the form of the total energy, which in both schemes is expressed as the average of the exact Hamiltonian over a single Slater determinant; the variational
prescription—allowing for the construction of optimal single-particle orbitals through the self-consistent solution of a single-particle eigenvalue problem—is not present in the SSH approach. We notice, however, that the importance of self-consistency is strongly related to the relative weight of Coulomb matrix elements: the HF potential entering the self-consistent HF one-particle Hamiltonian is in fact related to the direct and exchange Coulomb integrals; similarly, the SSH approximation is exact—without any need of self-consistency—whenever the higher-order contributions from the off-diagonal Coulomb matrix elements are negligible. For this reason we expect that a lower localization of the confined single-particle states in three dimensions with respect to two dimensions, giving rise to smaller non-diagonal Coulomb integrals, will reduce the difference between HF and SSH results. To check this point in detail, we have explicitly performed HF calculations; we have used in particular the matrix form of the unrestricted HF equation. Whenever possible, it is obviously useful to compare the outcomes of different approximate schemes with exact results. This is done in Sec. III, where we consider the exactly solvable two-electron QD (artificial helium) in different confinement regimes; we will show that the differences between HF and SSH results will be always comparable with those between HF and exact results, and that they scale with the dimensionality of the confining potential.

III. TWO-ELECTRON PROBLEM

In this section we will study the motion of two electrons within a QD structure in two and three dimensions. In this case, the exact Hamiltonian (1) reduces to

$$\hat{H} = \hat{H}_d(1) + \hat{H}_d(2) + \frac{e^2}{\kappa |r_1 - r_2|}. \tag{9}$$

Here $r_i$ is the position of the electron, $r_i = (x_i, y_i)$ in two dimensions or $r_i = (x_i, y_i, z_i)$ in three dimensions, and $p_i$ the corresponding momentum.

To solve this equation, we perform the standard transformation to center of mass (CM) coordinates, $\mathbf{R} = (r_1 + r_2)/2$, $\mathbf{P} = \hat{p}_1 + \hat{p}_2$, and relative-motion (rm) coordinates, $\mathbf{r} = r_1 - r_2$, $\mathbf{p} = (\hat{p}_1 - \hat{p}_2)/2$. The two-body Hamiltonian thus splits into CM and rm parts:

$$\hat{H} = \hat{H}_{CM} + \hat{H}_{rm}, \tag{10}$$

where

$$\hat{H}_{CM} = \frac{\hat{\mathbf{P}}^2}{2M} + \frac{1}{2} M \omega^2 \mathbf{R}^2, \tag{11}$$

$$\hat{H}_{rm} = \frac{\hat{\mathbf{P}}^2}{2\mu} + \frac{1}{2} \mu \omega^2 \mathbf{r}^2 + \frac{e^2}{\kappa \mathbf{r}}. \tag{12}$$

with $M = 2m^*$, and $\mu = m^*/2$. The CM Hamiltonian $\hat{H}_{CM}$ has the form of a simple harmonic oscillator. For the rm Hamiltonian $\hat{H}_{rm}$, it is easy to separate variables and obtain a radial differential equation, which gives solutions with the same set of quantum numbers as for the harmonic oscillator.

Solutions and notations for the 2D and 3D cases are summarized in Appendix A for both $\hat{H}_{CM}$ and $\hat{H}_{rm}$.

By denoting the CM and rm quantum numbers with capital and small letters, respectively, the eigenvalues for the two-particle system can be written as

$$E_{NM, nm} = \hbar \omega (2N + |M| + 1) + \epsilon_{nm} \tag{13}$$

in the 2D case, and

$$E_{NL, nl} = \hbar \omega \left( 2N + \frac{L + 3}{2} \right) + \epsilon_{nl} \tag{14}$$

in the 3D spherical case, the cylindrical 3D helium QD reducing to an effective 2D one (see Appendix A). Here $\epsilon_{nm}$ and $\epsilon_{nl}$ are the rm eigenvalues in two and three dimensions, respectively. Note that degeneracy is strongly reduced by Coulomb interaction with respect to the non-interacting case.

The corresponding two-particle total eigenfunctions are

$$\Psi_{NM, nm, SS}(r_1, s_1; r_2, s_2) = \Phi_{NM}(\mathbf{R}) \psi_{nm}(r) \chi(S, S_\tau) \tag{15}$$

for 2D and 3D cylinders, and

$$\Psi_{NM, nm, SS}(r_1, s_1; r_2, s_2) = \Phi_{NM}(\mathbf{R}) \psi_{nl}(r) \chi(S, S_\tau) \tag{16}$$

for a 3D sphere. Here $\Phi(\mathbf{R})$ and $\psi(r)$ are the spatial CM and rm eigenfunctions, respectively, and $\chi(S, S_\tau)$ is the spin function of a state with total spin $S(S+1)$ and $z$ projection $S_\tau$. Note that the parity of the rm spatial eigenfunction is defined (total orbital angular momentum and spin are conserved) and connected with the value of total spin by the antisymmetry of the two-particle total wave function $\Psi(r_1, s_1; r_2, s_2)$. For both the disk and the cylinder, this implies that if $m$ is even, the state is a singlet ($S = 0$), and if $m$ is odd, the state is a triplet ($S = 1$). Similarly for the sphere case, if $l$ is even, the state is a singlet ($S = 0$), and if $l$ is odd, the state is a triplet ($S = 1$).

In the above eigenvalues and eigenfunctions of the two-electron dot, the ingredients related to the CM Hamiltonian are known analytically (see Appendix A), while the rm energies and wave functions must be determined numerically. This is done by exact diagonalization of the rm eigenvalue problem (Appendix A), thereby yielding the full 2D and 3D spectrum of the QD helium.

Before comparing these exact results with the SSH approach, we point out that Hamiltonian (9) can be translated into a second-quantized form; this is done in terms of the same quantum numbers using CM and rm variables. The two-particle Hilbert space is the Kronecker product of the CM and rm single-particle spaces, generated, respectively, by the basis $\{|N\rangle\}_N$ (with eigenvalues $E_N$ and creation operators $\hat{a}_N^\dagger$) and $\{|n\rangle\}_n$ (with eigenvalues $\epsilon_n$ and creation operators $\hat{a}_n^\dagger$). Here, for simplicity, $N$ and $n$ label the whole set of CM and rm quantum numbers, respectively. The second-quantized form of the two-particle Hamiltonian $\hat{H}$, in this variable, is then given by

$$\hat{H} = \sum_N E_N \hat{a}_N^\dagger \hat{a}_N + \sum_n \epsilon_n \hat{a}_n^\dagger \hat{a}_n + \sum_{nm} V_{nm} \hat{a}_n^\dagger \hat{a}_n^\dagger + \sum_{nm} V_{nm} \hat{a}_n \hat{a}_n^\dagger. \tag{17}$$
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FIG. 1. Ground-state spatial pair-correlation function $g(x)$ for 3D (spherical) and 2D two-electron QD's: exact and SSH results are reported. Here $x = (2m^*a_r^r \hbar^2)$ is the dimensionless radial coordinate and $g(x)$ is normalized in such a way that $f_{0,0}(x)dx = 1$. The in-plane confinement energy is $h \omega_0 = 5$ meV.

This formulation allows us to obtain the result of the previously discussed Hubbard model, by simply neglecting all off-diagonal matrix elements in Eq. (17):

$$\mathbf{H}_{\text{SSH}}^{\text{2D}} = \sum_{i,j} E_i a_i^\dagger a_j + \sum_\mathbf{k} \left( \epsilon_{\mathbf{k}} + V_{\mathbf{k},0} \right) a_i^\dagger a_{\mathbf{k}},$$  \hspace{1cm} (18)

i.e., manifestly the noninteracting Hamiltonian "renormalized" by Coulomb interaction.

In order to check the reliability of the approximations and the role of dimensionality of the confining potential, we have calculated ground-state properties for QD’s with different confinement energies, i.e., different values of $h \omega_0$, assuming either a 2D or 3D confining potential. The quality of the ground-state eigenfunctions can be probed by the spatial pair correlation function $f(r)$:

$$f(r) = K \left( \sum_{\mathbf{r}_j} \delta(r-r_1) \right).$$  \hspace{1cm} (19)

Because of the circular symmetry, $f(r)$ depends only on the modulus of the relative distance $r$. Here, the factor $K$ is chosen in such a way that, if we define the dimensionless relative distance $s = r \sqrt{2m^*a_r^r \hbar^2}$, the quantity $g(x) = x^2 f(x)$ for the 2D and 3D cylinder cases, and $g(x) = x^2 f(x)$ for a 3D sphere is normalized:

$$\int_0^\infty g(x)dx = 1.$$

We have calculated this quantity both exactly and according to the SSH scheme for an in-plane confining energy $h \omega_0 = 5$ meV (throughout the paper we use $m^* = 0.065m_e$ inside the dot and $m^* = 0.079m_e$ outside; $\kappa = 12.98$, as in the QD of Ref. 2; and $m_e$ is the electronic mass); the results are shown in Fig. 1. The deviations between SSH and exact results clearly depend on the dimensionality of the confining potential: in disk-shaped 2D QD’s the SSH approximation is found to overestimate the probability of finding the two electrons close together, in analogy with HF results; the differences between exact and SSH results are significantly reduced assuming a 3D confining potential. This result is coherent with what is found for other ground-state properties: Fig. 2(a) shows the ground-state energies calculated for dots with different confinement energies, $h \omega_0$, in the range between 4.5 and 10 meV. We compare the exact results with the outcomes of HF and SSH calculations assuming 2D and 3D confinement potentials. Notice that the differences between HF and SSH are always smaller—by approximately 50%—than the corresponding differences with respect to the exact results; moreover the 3D confinement reduces the overall deviation of both HF and SSH by about 60%.

Since the SSH scheme is exact at the first perturbative order in the off-diagonal matrix elements of the e-e interaction, it is interesting to check the importance of the next perturbative corrections. Details of how the perturbative expansion is actually performed for the helium QD are reported in Appendix B. Figure 3 reports exact and SSH ground-state energies compared with the results of second-order perturbation theory, showing that second-order corrections become much smaller if a 3D confinement is assumed.

The situation becomes more complicated when considering dots with smaller confinement energies: in this case the HF and SSH differ from the exact result not only quantitatively but also qualitatively, predicting the two-particle ground state to be a triplet instead of a singlet, as it should be. This is shown in Fig. 2(b), where again the exact HF and SSH results are shown for dots of different confinement energies. The difference between triplet- and singlet-state energies decreases with increasing confinement energy both for SSH and HF approximations until a crossover occurs; assuming a 3D confining potential, the confinement energy of this crossover is reduced, and this again is true both for SSH and HF approximations.

We may summarize this analysis on helium QD by concluding that the assumed 3D confinement potential reduces the differences between approximate (SSH and HF) solutions
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LUDENIC QUANTUM DOTS. Here, A function of the electron number ing an electron to a half-shell, the physical origin of the half-shell-Coulomb repulsion $U'/h\omega_0$ opposing opposite-spin ones, makes the con total spin energetically favored. This behavior is the result of the interplay between single-particle contributions and electron-electron repulsion: the single-particle term favoring complete shell filling, while the repulsion among parallel-spin electrons, smaller than the repulsion among opposite-spin ones, makes the configurations with maximum total spin energetically favored (Hund’s rule). This is the physical origin of the half-shell-filling structure: indeed, adding an electron to a half-filled shell forces the double occupancy of a level; consequently, $\Delta A$ is raised by the dominant Coulomb repulsion $U_{\alpha\beta}$ between opposite-spin electrons on the same level.

For some nonclosed shell configurations the total spin turns out to be not determined by Hund’s rule: in particular, for $N=16$ we find a ground state with total spin $S=0$. Similar deviations from Hund’s rule have been found for large electron numbers ($N>20$) and associated with spin-density-wave instabilities; for smaller numbers ($N=16$ and 18) the same $S=0$ spin-density-wave state has been found to be a low-energy “spin isomer,” slightly higher in energy than the ground-state configuration.

From our calculations we may say that the $S=0$ configuration in dots with large $N$ may be favored by the reduced repulsion between electrons in high shells: in the fourth shell, for instance, the Coulomb integrals $U_{\alpha\beta}$ relative to orbitals with higher values of the orbital momentum may be smaller than the corresponding terms relating two levels with smaller angular momentum; the double occupation of an orbital with high orbital momentum $m$ (i.e., the level with $n=0$, $m=3$; see Appendix A for the notation) with antiparallel-spin electrons may therefore cost less than having parallel-spin electrons on different degenerate orbitals, but with smaller $m$ (i.e., the levels $n=1$ and $m=1$). The same interplay also explains the peaks in $\Delta A(N)$ for $N=14$ and 18.

We want to stress that also in the case of many electrons the reliability of the results of SSH approach is comparable with HF ones. The explicit comparison between the addition energy variation calculated according to SSH and HF schemes and for 2D and 3D confinements is reported in Fig. 5, showing that $\Delta A$ always peaks at the same electron numbers, and that the agreement between SSH and HF results improves on going from the 2D to the 3D confinement model.

Ground-state configurations and filling rules change when a magnetic field is applied. It affects both single-particle energies and Coulomb and exchange integrals through the induced changes in the wave-function localization. Figure 6 shows the $U$ and $J$ integrals vs $B$ for the first states, obtained for $h\omega_0=7.5$ meV. For comparison, we also show the corresponding quantities calculated within a strictly 2D confinement model. We can see that $U$ integrals describing the interaction between opposite-spin electrons are a few meV.

IV. MANY-ELECTRON PROBLEM

The key quantity that characterizes single-electron transport into a QD is the addition energy, i.e., the energy $A(N)$ required to place an extra electron into a dot that is initially occupied by $N-1$ electrons. Such a quantity, analogous to electron affinity in atomic physics, can be measured experimentally as a function of $N$. It has been shown\textsuperscript{2} that the measured voltage increment $\Delta A$ between successive single-electron tunneling processes—i.e., between two successive maxima in the conductance—peaks at “magic” values of $N$ corresponding to the filling of complete shells ($N=2, 6, 12$), as well as to half-shell filling (e.g., $N=4$). The existence of these half-shell filling features is reminiscent of Hund’s rule in atomic physics,\textsuperscript{2,11,12} and is intimately related to electron-electron interaction.

The results of SSH theory for the addition-energy variations, $\Delta A(N)=A(N+1)-A(N)$, are displayed in Fig. 4 as a function of the electron number $N$ for two different 3D cylindrical quantum dots. Here $A(N)$ is obtained as $E^{SSH}(N) - E^{SSH}(N-1)$, where $E^{SSH}(N)$ is the ground-state energy in Eq. (8). As we can see, $\Delta A(N)$ exhibits peaks corresponding both to complete and half-shell filling, thus well reproducing the experimental evidence in Ref. 2. This behavior is the result of the interplay between single-particle contributions and electron-electron repulsion: the single-particle term favors complete shell filling, while the repulsion among parallel-spin electrons, smaller than the repulsion among opposite-spin ones, makes the configurations with maximum total spin energetically favored (Hund’s rule). This is the physical origin of the half-shell-filling structure: indeed, adding an electron to a half-filled shell forces the double occupancy of a level; consequently, $\Delta A$ is raised by the dominant Coulomb repulsion $U_{\alpha\beta}$ between opposite-spin electrons on the same level.

For some nonclosed shell configurations the total spin turns out to be not determined by Hund’s rule: in particular,
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smaller in the case of 3D confinement, while the differences in the interaction between parallel-spin ones are much smaller. This is going to affect dramatically the energy balance which determines ground-state configurations, thus clearly showing the failure of a pure 2D description of state-of-the-art QD structures.

As already mentioned, according to the SSH approach the off-diagonal matrix elements of the electron-electron interaction are assumed to be negligible. In Fig. 6 the values of two of them are reported as functions of the applied magnetic field. As expected, we clearly see that for any \( B \) value they are negligible compared to all the other semidiagonal contributions, and even more so in three dimensions with respect to the 2D case.

Figure 7 shows the total energy \( E^{SSH} \) as a function of the applied magnetic field \( B \) for different values of the electron number \( N \) in a dot with confinement energy \( \hbar \omega_0 = 7.5 \) meV. For any given value of \( N \), all the possible configurations, denoted by the usual atomic physics terms \( ^nS^L \), have been considered.

Other authors have explicitly considered the question of dimensionality in theoretical modelization of semiconductor QD’s. Kumar, Laux, and Stern\textsuperscript{15} self-consistently computed the one-particle confining potential in a square QD. Accord-
ing to their results, our assumption of an in-plane parabolic
confining potential plus a well in the perpendicular direction
is seen to be quite reliable and general, as well as the ansatz
of considering only the ground-state motion along \( z \), at least
for few electron dots. Steinbeck et al.\(^{16}\) pointed out the im-
portance of a full 3D model to treat spin-density excitations
(SDE’s) in semiconductor wires and dots. Specifically, they
used the analogous of Eq. (A4) as an effective 3D Coulomb
interaction, and they found that a 2D description artificially
enhances the interaction strength and is unable to predict
experimental Raman spectra. The necessity of a 3D model-
ization is then seen to emerge not only in the description
of ground-state and single-particle processes, like addition
spectra, but also in two-particle processes, like SDE’s.

V. SUMMARY AND CONCLUSIONS
We have presented a theoretical investigation of Coulomb-correlation effects in semiconductor quantum dots.
In particular, we have performed a detailed analysis of the addition-spectrum problem for few-electron quantum-dot
structures (macroatoms), pointing out possible analogies
with more conventional Coulomb-correlation effects in
atomic physics.

Our primary goal was to understand to which extent the various approximation schemes, such as Hartree-Fock or
Hubbard models, are able to properly describe Coulomb cor-
relation in realistic, state-of-the-art QD’s. To this end, we
have first compared approximate results to the exact solution
for the prototypical case of a two-electron system, the so-
called quantum-dot helium; we have repeated such analysis
for different dimensionalities, considering 3D (spherical and
cylindrical geometry) and pure 2D structures. The main re-

sult is that the degree of accuracy of any approximation
scheme depends strongly on the dimensionality of the prob-
lem. More specifically, the pure 2D model—often used for a
description of quantum dots—is found to give approximate
results which differ significantly from the exact solution. We
have demonstrated that this is not a general failure of the approximation scheme, but that it rather reflects a pathologi-
ocal behavior originating from the unphysical nature of the
pure 2D model. Indeed, for the case of a 3D cylindrical model—which provides a much better description of realistic
QD structures—the difference between an exact solution and
approximate results is found to be much smaller, thus con-
firming the validity of the various approximation schemes
considered.

The same analysis has been then extended to many-
electron systems for which addition-spectra measurements
are available. Using different approximation schemes, we
find that the deviations between the full 3D model and the
simplified 2D quantum-dot model are very significant. The
full 3D model is found to reproduce the experimental data
for a large class of QD structures where simplified 2D mod-
el fails. We conclude that this is due to the unphysical char-
acter of the pure 2D confinement, for which the various ap-
proximation schemes often yield unreliable results. A proper
description of the QD structure in terms of fully 3D single-
particle wave functions is therefore required; we have shown
that in this case approximate approaches can give an accurate
description of correlation effects in the macroatoms made
available by present semiconductor technology.

ACKNOWLEDGMENTS
We are grateful to D. Pfannkuche and D. Vanossi for useful
discussions. This work was supported in part by the EC through the TMR Network “Ultrafast quantum optoelec-
nronics,” and by MURST–Italy.

APPENDIX A: CM AND RM SOLUTIONS
FOR THE TWO-ELECTRON DOT
The present appendix is organized as follows: In Sec. A.1
we shall show how to reduce the 3D cylindrical helium prob-
lem to an effective two-dimensional one; in Sec. A.2 we shall
summarize the 2D and 3D solutions of the one-particle
Schrodinger equations for the center-of-mass and the
relative-motion Hamiltonians, \( \hat{H}_{CM} \) and \( \hat{H}_{rm} \), as defined in
Eqs. (11) and (12).

1. 3D eigenvalue equation for the cylindrical QD
If only the lowest single-particle state \( \phi_0(z) \) of the quan-
tum well is relevant to the two-electron motion, we can write
the spatial-part \( \Psi(r_1,r_2) \) of our helium wave function as
\[
\Psi(r_1,r_2) = E \Phi(r_1,r_2); \quad (A1)
\]
This approximation is well justified for most cases of inter-
est. Indeed, for the typical QD structure used in the experi-
mental investigation of addition spectra\(^9\) (the quantum-well
width \( L = 12 \) nm, the barrier height \( V_0 = 200 \) meV), and the
energy separation between the ground and first-excited states
along \( z \) is 56 meV, about one order of magnitude larger than
typical in-plane single-particle confinement energies.

Let us now consider the global Schrodinger equation cor-
responding to the exact helium Hamiltonian of Eq. (9):
\[
\hat{H} \Psi(r_1,r_2) = E \Psi(r_1,r_2); \quad (A2)
\]
by substituting Eq. (A1), multiplying both sides by \( \phi_0^{*}(z_1) \phi_0^{*}(z_2) \), and integrating over \( z_1 \), and \( z_2 \), we obtain
\[
\left[ 2 \epsilon_0^2 + \sum_{i=1}^{2} \hat{h}_0(i) + \frac{e^2}{\kappa} \right] \Phi(r_1,r_2) = E \Phi(r_1,r_2). \quad (A3)
\]
The eigenvalue equation is then reduced to a 2D one, since
\( r_i=(x_i,y_i) \) and \( c(r)=c(|r_1-r_2|) \) is an effective Coulomb
potential, accounting for the geometry of the system:
\[
c(r) = \int_{-\infty}^{\infty} dz_1 \int_{-\infty}^{\infty} dz_2 \frac{|\phi_0^{*}(z_1)|^2 |\phi_0^{*}(z_2)|^2}{\sqrt{r^2+(z_1-z_2)^2}}. \quad (A4)
\]
From now on we will drop the constant ground-state energy
along \( z (\epsilon_0^2) \). As a first step, we evaluate \( c(r) \) by solving the
quantum well eigenvalue problem (allowing for different
values of the effective mass in the well and in the barrier).
Then we numerically integrate Eq. (A4). It is easy to show
analytically some important properties of \( c(r) \), namely, that
\[
0 \leq c(r) \leq 1, \quad \forall r, \quad (A5)
\]
The Importance of Electronic Correlation

\[ \lim_{r \to 0} r c(r) = 0, \quad \lim_{r \to \infty} r c(r) = 1. \quad (A6, A7) \]

These properties tell us that for large distances \( r c(r) \) tends to the bare Coulomb potential, and that it is strongly reduced in the neighborhood of the origin, i.e., the more relevant space region in the computation of Coulomb and exchange integrals. Figure 9 shows such an effective Coulomb potential \( C \) multiplied by the dimensionless variable \( x \) (introduced below) as a function of \( x \) for different values of the quantum-well width: A monotonic behavior is apparent, going from the bare Coulomb-potential value in the zero-width limit (the function is constant and equal to 1), into progressively smaller values, toward the infinite-width case.

2. Exact solutions

Let us first consider the CM equation, which has the form of a standard harmonic oscillator and can thus be solved analytically. For the 2D case (3D cylindrical), its eigenvalues are

\[ e^{2D}_{NM} = \hbar \omega_0 (2N + |M| + 1), \quad (A8) \]

\[ \Phi^{2D}_{NM,\sigma}(r,s) = (s,r|NM\sigma) = \lambda^{[M]+1/2} \sqrt{\frac{N!}{\pi (|M|)!}} e^{-\lambda r^2/2} L^{|M|}_N(\lambda r^2) \chi_\sigma(s). \quad (A9) \]

In the 3D spherical case,\(^1\) the eigenvalues are

\[ e^{3D}_{NL} = \hbar \omega_0 \left( 2N + L + \frac{3}{2} \right), \quad N=0,1,2, \ldots, \quad L=0,1,2, \ldots, \quad (A10) \]

and the orthonormalized eigenfunctions \( \Phi^{3D}_{NL,\sigma}(r,s) \) are

\[ \Phi^{3D}_{NL,\sigma}(r,s) = (s,r|NL\sigma) = \sqrt{\frac{2\lambda^{L+1/2}N!}{\Gamma(N+L+3/2)}} e^{-\lambda r^2/2} L^L_N(\lambda r^2) Y_{LM}(\theta,\varphi) \chi_\sigma(s). \quad (A11) \]

Here, \( \lambda = m^2 \omega_0 / \hbar, L_N^L \) are generalized Laguerre polynomials,\(^9\) \( \Gamma \) is the usual gamma function, \( \chi_\sigma \) denotes the spin function, and \( Y_{LM} \) are the spherical harmonics. We have used polar coordinates throughout: \( r=(r,\varphi) \) in the 2D (3D cylindrical) case and \( r=(r,\theta,\varphi) \) in the 3D spherical case. For the 2D (3D cylindrical) case the quantum numbers are \((N,M,\sigma)\): \( N \) is the radial quantum number, \( M \) the angular momentum quantum number (in this case the total angular momentum coincides with the component along \( z \), \( L_z = -\hbar M \)), and \( \sigma \) the spin component along \( z \). In the 3D spherical case, on the other hand, the quantum numbers are given by \((N,L,M_z,\sigma)\): here \( L \) is the total angular momentum quantum number, and \( M_z \) is the magnetic quantum number, \( M_z = -L, -L+1, \ldots, L \).

Let us now come to the single-particle Schrödinger equation for the \( \text{cm} \) Hamiltonian of Eq. (12). In this equation the variables are easily separable, and the problem is reduced to the solution of a radial differential equation. For the 2D (3D cylindrical) case, the \( \text{cm} \) eigenfunction in coordinate space is

\[ \varphi_{nm}(r) = R_{nm}(r) e^{-i m \varphi} \frac{1}{\sqrt{2 \pi}}, \quad (A12) \]

where \( R_{nm}(r) \) is the solution of the radial Schrödinger equation.

FIG. 9. Plot of the effective Coulomb potential \( C(x) \) multiplied by the dimensionless coordinate \( x \) \([= x \ C(x) \] as a function of \( x \) for different values of the quantum well width \( L \) and for a confinement energy \( \hbar \omega_0 = 5 \text{ meV} \). Notice that in the limit \( L \to 0 \) (2D case) \( C(x) \to 1/x \) and, therefore, \( x \ C(x) \to 1 \).

\[ N=0,1,2, \ldots, \quad M=0, \pm 1, \pm 2, \ldots, \]

and the corresponding orthonormalized eigenfunctions (the so-called “Fock-Darwin” states\(^9\)) are

\[ \Phi^{2D}_{NM,\sigma}(r,s) = (s,r|NM\sigma) = \lambda^{[M]+1/2} \sqrt{\frac{N!}{\pi (|M|)!}} e^{-\lambda r^2/2} L^{|M|}_N(\lambda r^2) \chi_\sigma(s). \quad (A9) \]
we have employed the notations $\bar{\kappa} = \mu \omega_0/h, \alpha = 2 \mu e^2/k^2$, and $k_{nl}^2 = 2 \mu \epsilon_{nm}/h^2$, where $\epsilon_{nm}$ is the rm eigenvalue. The effective Coulomb potential $c(r)$ is simply $1/r$ in the 2D case, and it is defined in Sec. A1 for the 3D cylindrical case.

For the 3D spherical case, the rm eigenfunction in coordinate space is

$$\varphi_{nlm}(r) = R_{nl}(r) Y_{lm}(\theta, \phi),$$

with $R_{nl}(r)$ satisfying the radial eigenvalue equation

$$\frac{\partial^2 R_{nl}(r)}{\partial r^2} + 2 \frac{\partial R_{nl}(r)}{\partial r} + \left[ k_{nl}^2 - \bar{\kappa}^2 r^2 - \frac{\alpha}{r} \right] R_{nl}(r) = 0;$$

where, again, we put $k_{nl}^2 = 2 \mu \epsilon_{nl}/h^2$, and $\epsilon_{nl}$ is the rm eigenvalue.

In order to obtain an exact solution for the rm eigenvalue problems, we rewrite Eqs. (A13) and (A15) in terms of the dimensionless variable $x = \sqrt{\bar{\kappa}} r$. For the 2D (3D cylindrical) case, Eq. (A13) becomes

$$\frac{d}{dx} \left( \frac{dR_{nm}(x)}{dx} \right) + \left[ - \frac{m^2}{x} - \bar{\alpha} C(x) + \bar{k}_{nm}^2 x - x^2 \right] R_{nm}(x) = 0,$$

$$\bar{R}_{nm}(x) = R_{nm}(r),$$

$$C(x) = \bar{\kappa}^{-1/2} c(\bar{\kappa}^{-1/2} x)$$

[for the 2D case it is simply $C(x) = 1/x$], while for the 3D case Eq. (A15) transforms into

$$\frac{d^2 \tilde{\chi}_{nl}(x)}{dx^2} + \left[ \frac{l(l+1)}{x^2} - \bar{\alpha} x + \bar{k}_{nl}^2 - x^2 \right] \tilde{\chi}_{nl}(x) = 0,$$

$$\tilde{\chi}_{nl}(x) = \chi_{nl}(r), \quad \chi_{nl}(r) = \frac{R_{nl}(r)}{r}.$$

The dimensionless parameters are $\bar{\alpha} = \bar{\kappa}^{-1/2} \alpha = 2 \sqrt{\bar{R}^*} / \hbar \omega_0$ and $\bar{k}_{nl}^2 = k_{nl}^2 / \bar{\kappa} = 2 \epsilon_{nl} / \hbar \omega_0; \bar{R}^* = e^4 m^* / 2 \kappa^2 h^2$ is the effective Rydberg energy. Actually, exact analytic solutions exist, but they are limited to 2D and 3D spherical cases only$^{20,21}$; thus we have chosen to solve Eqs. (A16) and (A17) by standard numerical methods. We stress that the numerical accuracy depends on the accurate specification of the boundary conditions that we impose through analytical asymptotic formulas for eigenfunctions near to the singular points 0 and $+\infty$, following the general methods of Ref. 22. In this way the numerical solution is very stable and efficient, thus overcoming possible difficulties related to the singlet ground state$^2$; in our calculations energy values are obtained with a nominal relative error of the order of $10^{-5}$.

APPENDIX B: HELIUM PERTURBATION THEORY

We employ the standard Rayleigh-Schrödinger perturbation theory to correct the SSH eigenvalues in Eq. (18) at the second order in the off-diagonal Coulomb matrix elements entering the total Hamiltonian (17). In the remaining part of this section we shall consider the 2D and 3D spherical cases, by neglecting the center-of-mass motion.

For the 2D case, the rm SSH eigenvalues $\epsilon_{nm}^{\text{SSH}}$ are given by$^{10}$

$$\epsilon_{nm}^{\text{SSH}} = \hbar \omega_0 (2n + |m| + 1) + \sqrt{R^*} \hbar \omega_0 S^{2D}(n, m), \quad (B1)$$

$$S^{2D}(n, m) = \frac{1}{|m|!} \left[ \frac{m + 1}{2} \right] \left[ 1 + \sum_{s=0}^{n-1} \frac{n! (-1)^{s+1} [(2s+1)!!]^2 |m|!}{(n-s-1)! 2^{2s+1} (s+1)! |(m+s+1)!|^2} \right]$$

while for the 3D case we have

$$\epsilon_{nl}^{\text{SSH}} = \hbar \omega_0 (2n + l + \frac{3}{2}) + \sqrt{R^*} \hbar \omega_0 S^{3D}(n, l),$$

$$S^{3D}(n, l) = \frac{1}{|l|!} \left[ \frac{l + 1}{2} \right] \left[ 1 + \sum_{s=0}^{n-1} \frac{n! (-1)^{s+1} [(2s+1)!!]^2 |l|!}{(n-s-1)! 2^{2s+1} (s+1)! |(l+s+1)!|^2} \right].$$
The Importance of Electronic Correlation

The first-order correction due to nondiagonal Coulomb matrix elements is equal to zero. The second-order correction \( \Delta e^{(2)}_0 \) to the ground-state energy is given by the well-known expression

\[
\Delta e^{(2)}_0 = \sum_n \frac{|V_{0n}|^2}{\epsilon_0 - \epsilon_n} \tag{B5}
\]

[see the notation in Eq. (17)]. The main idea is now to look for analytic expressions for the off-diagonal integrals \( V_{0n} \) and then to perform a numerical summation. However, expressions like those obtained in Eqs. (B2)–(B4) (Ref. 23) are not useful, since each integral is given by an alternated-sign summation, and numerical errors become rapidly critical as the quantum number \( n \) increases. In contrast, the solution can be obtained using an integration trick suggested in Ref. 24, so that all the terms in the summation are obtained with the same sign. For the 2D case one obtains

\[
V_{0n} = \sqrt{\frac{\Gamma^2(n+\frac{1}{2})}{\pi \Gamma(n+1) \Gamma(n+\frac{3}{2}) \Gamma(\frac{3}{2})}}, \quad n \to \infty. \tag{B6}
\]

For the 3D case one obtains

\[
V_{0n} = \sqrt{\frac{\Gamma^2(n+\frac{1}{2})}{\pi \Gamma(n+1) \Gamma(n+\frac{3}{2}) \Gamma(\frac{3}{2})}}, \quad n \to \infty. \tag{B7}
\]

As already pointed out, now the generic terms (B6) and (B7) in the sum (B5) have the same sign and the summation can be easily performed. The result for the 2D case is

\[
\Delta e^{(2)}_0 = -\mathcal{R}^* \ (0.691), \tag{B8}
\]

and for the 3D case it is

\[
\Delta e^{(2)}_0 = -\mathcal{R}^* \ (0.156). \tag{B9}
\]

Note that the 3D term is significantly smaller than the corresponding 2D one, and that in the 3D case the series converges more quickly.


13. Since in these cases the energy balance defining the ground-state energy according to Eq. (8) becomes very delicate, the dependence of Coulomb and exchange integrals on the dot dimensionality may give rise to different ground-state spin polarizations in 2D and 3D QD's.


Chapter 6

The Double-Dot Molecule
Multiple quantum phases in artificial double-dot molecules

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Received 16 June 1999; accepted 18 June 1999 by M. Cardona

Abstract

We study coupled semiconductor quantum dots theoretically through a generalized Hubbard approach, where intra- and inter-dot Coulomb correlation, as well as tunneling effects are described on the basis of realistic electron wavefunctions. We find that the ground-state configuration of vertically coupled double dots undergoes non-trivial quantum transitions as a function of the inter-dot distance \( d \); at intermediate values of \( d \) we predict a new phase that should be observable in the addition spectra and in the magnetization changes.

Keywords: A. Nanostructures; D. Electron–electron interactions; D. Quantum localization

Semiconductor quantum dots (QDs) are nano- or mesoscopic structures that can be regarded as ‘artificial atoms’ because of the three-dimensional carrier confinement and the resulting discrete energy spectrum. They are currently receiving great attention because they can be designed to study and exploit new physical phenomena: on the one hand, the nature and scale of electronic confinement allows the exploration of regimes that are not accessible in conventional atomic physics; on the other, they lead to novel devices dominated by single- or few-electron effects [1–3].

One of the important challenges at this point is to understand the fundamental properties of coupled QDs, the simplest structures that display the interactions controlling potential quantum-computing devices. Here also the inter-dot coupling can be tuned through external parameters, far out of the regimes known in ‘natural molecules’ where the ground-state interatomic distance is dictated by the nature of bonding. We expect that new phenomena will occur in these ‘artificial molecules’ (AMs) when the relative importance of Coulomb interaction and single-particle tunneling is varied. Through the study of such phenomena, coupled dots may become a unique laboratory to explore electronic correlations.

In this paper we analyze ground and excited few-particle states of realistic double quantum dots (DQDs) using a theoretical scheme that fully takes into account intra- and inter-dot many-body interactions. We focus on strongly confined vertically coupled DQDs, and show that, for a given number of electrons, \( N \), the ground-state configuration undergoes non-trivial quantum transitions as a function of the inter-dot distance \( d \); we identify specific ranges of the DQD parameters characterizing a ‘coherent’ molecular phase, where tunneling effects dominate, and a phase where the inter-dot interaction is purely electrostatic. At intermediate values of \( d \) we predict a new phase that should be observable by means of transport experiments in the addition spectra and in the magnetization changes.

From the theoretical point of view, a major difficulty is that we cannot make use of perturbative schemes [4,5] in the calculation of the DQD many-body ground state, since we want to investigate all inter-dot coupling regimes. We thus write the many-body hamiltonian \( \hat{H} \) within a generalized Hubbard (GHH) approach [6–11], 1 and choose a basis set formed by suitable single-particle wavefunctions localized on either dot (\( i = 1,2 \)), and characterized by orbital quantum numbers, \( \alpha \) (to be specified later), and by the spin quantum number, \( \sigma \). On this basis the many-body

1 Hubbard-like hamiltonians for QDs are also used—-with different levels of approximations—-in Refs. [6–11].
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200 meV) and the corresponding symmetric and antisymmetric configurations contributing to the many-body ground state are shown in the insets. Note that in the intermediate phase a quantum dot with dot distance \( d \) is shown together with two; the bottom panel displays \( s \) and \( e \) approximation), \( \epsilon \) the electronic charge and \( \kappa \), the relative dielectric constant.

Here we shall consider a specific type of nanostructures, namely vertically coupled cylindrical DQDs [12]; the ingredients entering the hamiltonian can therefore be defined explicitly. For simplicity, \( V(r) \) can be assumed to be separable in the \( xy \) and \( z \) components; the profile is taken to be parabolic in the \( xy \)-plane, with confinement energy \( \hbar \omega \), and a symmetric double quantum well (QW) along \( z \) [12,13]. The eigenstates of the \( xy \) harmonic potential are the usual Fock–Darwin states \( | \alpha \rangle = (|n,m \rangle \langle n,m| \) \( n, m \) being the radial and angular quantum numbers) [3]. Along \( z \), the QW thickness is such that only the lowest eigenstates (symmetric, \( |s\rangle \), and antisymmetric \( |a\rangle \)) are relevant for the low-energy spectrum. From these, we construct a complete set of states that are localized on either dot (see inset of Fig. 1): \( |1\rangle = (|s\rangle + |a\rangle)/\sqrt{2} \) and \( |2\rangle = (|s\rangle - |a\rangle)/\sqrt{2} \). The basis we use is therefore the direct product \( |\alpha \alpha \rangle = |\alpha \rangle \otimes |\alpha \rangle \otimes |\alpha \rangle \).

The single-particle energies are \( \varepsilon_\alpha = \varepsilon_\alpha + (\epsilon_s + \epsilon_a)/2 \), and the tunneling parameter is \( t = (\epsilon_s - \epsilon_a)/2 \), with \( \epsilon_s \), \( \epsilon_a \) double-well eigenergies, and \( \epsilon_\alpha \) oscillator energies.

Using the above expressions, the three-dimensional Coulomb integrals and the tunneling parameter are computed directly from the single-particle states for each sample. Fig. 1 shows the result for \( t, U_{\alpha \beta} \) and \( U_{\alpha \beta} \) \( | \alpha = \beta = (0,0) \rangle \) calculated for a \( \text{In}_{0.05}\text{Ga}_{0.95}\text{As} \) DQD (\( \kappa_\ell = 12.98 \),

\[ H = \sum_{i\sigma} \varepsilon_i \hat{n}_{i\alpha} - t \sum_{\alpha\beta(ij)} c_{i\alpha}^\dagger c_{j\beta} + \frac{1}{2} \sum_{\alpha\beta\gamma} U_{\alpha\beta\gamma} \hat{n}_{i\alpha} \hat{n}_{i\beta} \hat{n}_{i\gamma} \]

\[ + \frac{1}{2} \sum_{\alpha\beta\gamma\delta} U_{\alpha\beta\gamma\delta} \hat{n}_{i\alpha} \hat{n}_{i\beta} \hat{n}_{i\gamma} \hat{n}_{i\delta} \}

The first two addenda are the single-particle on-site and hopping term, respectively: \( \varepsilon_i \) are the single-particle energies, \( t \) the tunneling parameter; \( \hat{n}_i \) the occupation numbers and \( \hat{c}^\dagger (\hat{c}) \) are the creation (destruction) operators. The third and fourth terms account for intra-dot Coulomb interaction between electrons with antiparallel and parallel spins, respectively; \( U \) and \( J \) are the intra-dot Coulomb and exchange integrals. Finally, the last term represents the inter-dot Coulomb coupling, \( \hat{U} \) being the inter-dot Coulomb integrals. These integrals are in turn expressed in terms of single-particle states:

\[ U_{\alpha\beta} = \int \frac{\varepsilon^2 \phi_\alpha(r) \phi_\beta(r')}{\kappa \varepsilon \sqrt{|r - r'|}} \, dr \, dr', \]

\[ U_{\alpha\beta} = \int \frac{\varepsilon^2 \phi_\alpha(r) \phi_\beta(r')}{\kappa \varepsilon \sqrt{|r - r'|}} \, dr \, dr', \]

\[ J_{\alpha\beta} = \int \frac{\varepsilon^2 \phi_\alpha(r) \phi_\beta(r')}{\kappa \varepsilon \sqrt{|r - r'|}} \, dr \, dr', \]

with \( \phi_\alpha(r) \) the full three-dimensional single-particle wavefunctions (obtained within the usual envelope-function approximation), \( E \) the electronic charge and \( \kappa \), the relative dielectric constant.

Fig. 1. Parameters entering the many-body hamiltonian of a double quantum dot with \( \hbar \omega = 10 \text{ meV} \), plotted as a function of the inter-dot distance \( d \). The hopping coefficient \( t \) is shown together with two of the inter-dot and inter-dot Coulomb integrals. In the inset, the top panel shows the confinement potential \( V(z) \) (the barrier height is 200 meV) and the corresponding symmetric and antisymmetric single-particle wavefunctions, \( |s\rangle \) and \( |a\rangle \); the bottom panel displays the localized states, \( |1\rangle \) and \( |2\rangle \), obtained as combinations of \( |s\rangle \) and \( |a\rangle \), that are used as basis set for our calculation.

Fig. 2. Energies of the ground state (thick line) and three lowest excited states (thin lines) as a function of the inter-dot distance, \( d \), for a double QD occupied by \( N \) electrons. For \( N = 4 \), the prominent single particle configurations contributing to the many-body ground state are shown in the insets. Note that in the intermediate phase a significant contribution comes also from other Slater determinants (see text). The \( \pi \) molecular states are doubly degenerate because of the two-fold degeneracy of the second shell in the single-dots (\( \pi, \pi' \)).
effective mass $0.065m_e$ with $\hbar\omega = 10$ meV, as a function of the inter-dot distance (barrier width) $d$. Note that already around $d = 5$ nm the inter-dot Coulomb integral exceeds the single-particle hopping parameter.

To obtain the many-body energies and eigenstates, the Hamiltonian $H$ is then diagonalized exactly for each value of $N$ (total number of electrons) on each configuration subspace labeled by the quantum numbers $S$ (z-component of the total spin) and $M$ (z-component of the total orbital angular momentum).

This approach has two main advantages: first, it allows us to solve the many-body problem consistently in the different coupling regimes, from the limit where $t$ dominates over the Coulomb integrals to the opposite limit where $t$ is negligible. Second, it can provide quantitative predictions for given DQD structures, since it contains no free parameters and uses realistic ingredients ($t$, $U_{ab}$, $U_{af}$, $J_{gfi}$) calculated for each nanostructure [14,15].

Fig. 2 shows the calculated ground-state energies $E_N$ of correlated N-particle states as a function of the inter-dot distance $d$. The three lowest excited states are also shown for comparison. As expected, when $d = 5$ nm the system behaves as two isolated QDs. With decreasing $d$, some of the many-body excited states, favored by Coulomb interactions, become lower in energy. For $N = 3$ a single quantum phase transition occurs, below which the new ground state is a molecular-like state. For $N > 3$, two successive transitions take place, at $d = d_b$ and $d = d_c$, and an intermediate non-trivial phase is predicted to occur in the range $d < d_c < d_b$. Note that this phase is stable in a relatively large range of $d$ values, which depends on the number of electrons. An accurate determination of $d_b$ and $d_c$ requires the correct inclusion of all inter-dot coupling terms, including the inter-dot Coulomb integrals. If the latter are neglected, all quantum transitions occur for smaller values of the inter-dot distance, and the $d$-range of the intermediate phase is underestimated significantly: had also the other many-body terms been neglected, the intermediate phase would disappear, leaving a simple crossover from a molecular to an atomic-like regime at $d = d_a = d_c$.

To understand the nature of the different phases it is useful to examine the many-body states in terms of the single Slater determinants that contribute to each of them. Here we discuss explicitly the 4-electron case with the help of the insets in Fig. 2, but the same reasoning can be followed for the other cases. Both in the case of very small and very large inter-dot distances the ground state can be essentially described in terms of a single Slater determinant: for large values of $d$ ($d > d_b$), the relevant configurations for the ground state have two electrons in the lowest orbital; in each isolated dot, the $|s\rangle$ and $|a\rangle$ extended ‘molecular’ orbitals derived from the lowest ‘atomic’ states are of course almost degenerate and both filled with two electrons. In the opposite limit ($d < d_c$), by expanding the localized atomic orbitals in terms of molecular orbitals we recognize that the $|s\rangle$ state derived from the lowest atomic state is filled with two electrons, while the corresponding $|a\rangle$ molecular state is empty. The two remaining electrons occupy the next bonding molecular orbitals—derived from the higher $p_z$ and $p_y$ levels—with parallel spin, in such a way that $S$ is maximized. This is the manifestation of Coulomb interaction, which leads to Hund’s rule for molecules.

In these two extreme phases the single particle picture is essentially correct, provided that the appropriate basis set (either localized or extended orbitals) is used. In the intermediate phases $d_a < d < d_c$, this is no longer true and the ground state is a mixture of different Slater determinants in any basis set [16]. In this sense, again the intermediate phase exhibits an intrinsic many-body character. Coulomb direct and exchange terms, responsible for the selection of the global quantum numbers, determine a new ground-state configuration, where both $S$ and $M$ are maximized.

We obtain a clear evidence of the different electronic distribution in the three quantum phases by calculating the spin-dependent electronic pair correlation function, defined as

$$g_{s',\sigma'}(r, \vec{z}, \vec{z}') = \int d\mathbf{R} \langle \hat{\Psi}_{s'}^\dagger(\mathbf{r}') \hat{\Psi}_{s'}(\mathbf{r}') \hat{\Psi}_{\sigma'}(\mathbf{r}) \hat{\Psi}_{\sigma'}^\dagger(\mathbf{r}) \rangle,$$

where $\mathbf{R} = (\rho' + \rho)^{\frac{1}{2}}$ and $\rho = (\rho' - \rho)^{\frac{1}{2}}$. Here $\rho' = (x', y')$, $\rho = (x, y)$ are the in-plane spatial coordinates.
of the electrons in the pair, and $z'$ and $z''$ are their coordinates along $z$, that will be kept fixed at the center of either QD, i.e. in $z_{1}$ or $z_{2}$. For $d = d_{a}$, the wavefunctions extend over both dots, and the pair correlation functions are very different depending on the location of both electrons in the same or in different dots. Note that transitions between different electronic configurations vs. $d$ were recently identified theoretically also for classical coupled dots [17]: we find that, for the small values of $N$ considered here, the number of distinct phases and the spatial distribution of electrons (as reflected in their correlation functions) is drastically modified by quantum effects.

The above findings are expected to be observable experimentally. First, the calculated magnetic-field dependent addition spectra $A(N)$ present clear signatures of the phase transitions described above, as illustrated in Fig. 4. Here $A(N) = E(N) - E(N - 1)$ is obtained from the many-body ground-state energies of the $N$- and $(N - 1)$-electron systems, on the basis of single-particle states calculated in the presence of the external magnetic field $B$. The behavior shown in the left, central, and right panels is representative of the three phases. Secondly, the changes in the magnetization induced by one electron addition—also accessible experimentally [18]—are expected to follow a different pattern in each phase (see the different sequence of quantum numbers in Fig. 2).

A large experimental effort is presently devoted to transport experiments in double QDs. In most cases, the dots are obtained by gating a two-dimensional electron gas (lateral confinement), and their coupling can be tuned through a gate voltage [18–22]. Indeed, a set of experiments has recently demonstrated a clear transition to a ‘coherent’ state with increasing coupling between dots [20,22]. Outside this strong coupling regime, however, experiments performed in the lateral geometry have so far evidenced classical inter-dot capacitance effects, probably owing to the size of dots. On the other hand, transport experiments are now available on DQD structures with strong lateral confinement fabricated by combined growth and etching techniques. The advantage is that the number of electrons in the structure is limited, while an accurate control on the inter-dot coupling is still possible by designing samples with appropriate barrier thickness. Both aspects are important to enhance many-body effects and to explore intermediate coupling regimes. Recent experimental work on these structures has focused on the weak-coupling regimes [12]. We hope that further investigations will be stimulated by the present work, since the relevant transitions are now predicted quantitatively and should occur in a range of parameters that is accessible to state-of-the-art experiments. We expect that such studies will bring new insight into electron–electron interaction effects in coupled quantum nanostructures.

Acknowledgements

We acknowledge helpful discussions with C. Calandra and G. Goldoni. This work was supported in part by INFM PRA99-SSQI, by the EC under the TMR Network ‘Ultrafast Quantum Optoelectronics’, and by the MURST-40% program ‘Physics of Nanostructures’.

References


4 Interesting deviations from predictions of simple capacitance models are however reported in Refs. [18,19].

5 DQD with different geometries, obtained by cleaved-edge or self-organized growth, have recently become available but were so far investigated mostly by optical experiments. See, e.g. Refs. [23–25].
THEORY OF ADDITION SPECTRA IN DOUBLE QUANTUM DOTS: SINGLE-PARTICLE TUNNELING VS COULOMB INTERACTIONS

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ABSTRACT

We study coupled semiconductor quantum dots theoretically using a generalized Hubbard approach, where intra- and inter-dot Coulomb correlation, as well as tunneling effects are described on the basis of realistic electron wavefunctions. We find that the ground-state configuration of vertically coupled double dots undergoes non-trivial quantum transitions as the inter-dot distance $d$ changes; at intermediate values of $d$ we predict a new phase that should be observable in the addition spectra and in the magnetization changes.

INTRODUCTION

Semiconductor quantum dots (QDs) are nano- or mesoscopic structures that can be regarded as ‘artificial atoms’ because of the three-dimensional carrier confinement and the resulting discrete energy spectrum. They have currently received much attention because they can be designed to study and exploit new physical phenomena: On the one hand, the nature and scale of electronic confinement allows the exploration of regimes inaccessible in conventional atomic physics; on the other hand, they lead to novel devices where single- or few-electron effects are important[1].

One of the important challenges at this point is to understand the fundamental properties of coupled quantum dots, the simplest structures that manifest the interactions controlling potential quantum-computing devices. Here also the interdot coupling can be tuned through external parameters, far from the regimes known in ‘natural molecules’ where the ground-state interatomic distance is dictated by the bonding. We expect to see new phenomena in these ‘artificial molecules’ (AMs) when the relative importance of Coulomb interaction and single-particle tunneling is varied. Through the study of such phenomena, coupled dots may become a unique laboratory enabling one to understand the role of correlations better.

In this paper we analyze ground and excited few-particle states of realistic double quantum dots (DQDs) using a theoretical scheme that fully takes into account intra- and inter-dot many-body interactions. We focus on strongly confined vertically-coupled DQDs in the zero- or low-magnetic field regime; this subject is relatively unexplored[2]. We show that, for a given number of electrons, $N$ (≤ 6), the ground state configuration undergoes non-trivial quantum transitions as the inter-dot distance $d$ changes; we identify specific ranges of the DQD parameters characterizing a ‘coherent’ molecular phase, where tunneling effects dominate, and a phase where the inter-dot interaction is purely electrostatic. At intermediate values of $d$ we predict a new phase that should be observable by performing transport experiments and observing changes in the addition spectra and in the magnetization.
THEORY

In our calculations, we cannot make use of perturbative or single-particle schemes in the calculation of the DQD many-body ground state, since we want to investigate all inter-dot coupling regimes. We thus write the many-body Hamiltonian \( \hat{H} \) within a generalized Hubbard (GHH) approach [3], and choose a basis set formed by suitable single-particle wavefunctions localized on either dot \((i = 1, 2)\), and characterized by orbital quantum numbers, \( \alpha \) (to be specified below), and by the spin quantum number, \( \sigma \). In this basis the many-body Hamiltonian is

\[
\hat{H} = \sum_{ia} \epsilon_i \hat{n}_{ia} - t \sum_{a(ij)} \hat{c}_{ia} \hat{c}_{ja}^\dagger + \frac{1}{2} \sum_{ia \beta} U_{\alpha \beta} \hat{n}_{ia} \hat{n}_{i\beta} + \frac{1}{2} \sum_{ia \beta} \hat{U}_{\alpha \beta} \hat{n}_{ia} \hat{n}_{i\beta}^\dagger + \frac{1}{2} \sum_{ia \beta} (U_{\alpha \beta} - J_{\alpha \beta}) \hat{n}_{ia} \hat{n}_{i\beta} + \sum_{a(ij)} \hat{U}_{\alpha \beta} \hat{n}_{ia} \hat{n}_{i\beta}.
\]

The first two terms are the single-particle on-site and hopping term respectively: \( \epsilon_i \) are the single-particle energies, \( t \) the tunneling parameter; \( \hat{n} \) are the occupation numbers and \( \hat{c} \) (\( \hat{c}^\dagger \)) the creation (destruction) operators. The third and fourth terms account for intra-dot Coulomb interaction between electrons with antiparallel and parallel spins, respectively: \( U \) and \( J \) are the intra-dot Coulomb and exchange integrals. Finally, the last term represents the inter-dot Coulomb coupling, \( \hat{U} \) being the inter-dot Coulomb integrals. These integrals are in turn expressed in terms of single-particle states:

\[
U_{\alpha \beta} = \int \frac{e^2 |\phi_{\alpha}(\mathbf{r})|^2 |\phi_{\beta}(\mathbf{r'})|^2}{\kappa_r |\mathbf{r} - \mathbf{r'}|} d\mathbf{r} d\mathbf{r'};
\]

\[
\hat{U}_{\alpha \beta} = \int \frac{e^2 |\phi_{\alpha}(\mathbf{r})|^2 |\phi_{\beta}(\mathbf{r'})|^2}{\kappa_r |\mathbf{r} - \mathbf{r'}|} d\mathbf{r} d\mathbf{r'};
\]

\[
J_{\alpha \beta} = \int \frac{e^2 \phi_{\alpha}^*(\mathbf{r}) \phi_{\beta}(\mathbf{r'}) \phi_{\alpha}(\mathbf{r}) \phi_{\beta}(\mathbf{r'})}{\kappa_r |\mathbf{r} - \mathbf{r'}|} d\mathbf{r} d\mathbf{r'};
\]

where \( \phi_{\alpha}(\mathbf{r}) \) are the full three-dimensional single-particle wavefunctions (obtained within the usual envelope-function approximation), and \( \kappa_r \) is the relative dielectric constant.

Here we shall consider a specific type of nanostructure, namely vertically coupled cylindrical DQDs [4]; the contributions to the Hamiltonian can therefore be defined explicitly. For simplicity, the single-particle confining potential \( V(\mathbf{r}) \) can be written in the form

\[
V(\mathbf{r}) = \frac{1}{2} m^* \omega (x^2 + y^2) + V_0(z),
\]

where \( V_0(z) \) is a symmetric double quantum well (QW) and \( m^* \) is the effective electronic mass [4]. We write the eigenstates of the two dimensional harmonic oscillator in the usual Fock-Darwin form, \(|m n\rangle = |n, m\rangle\) where \( n, m \) are the radial and angular quantum numbers [1]. Along \( z \), the QW thickness is such that only the lowest eigenstates (symmetric, \(|s\rangle\), and antisymmetric \(|a\rangle\)) are relevant for the low-energy spectrum. From these, we construct a complete set of states that are localized on either dot: \(|\uparrow\rangle = (|s\rangle + |a\rangle)/\sqrt{2}\) and \(|\downarrow\rangle = (|s\rangle - |a\rangle)/\sqrt{2}\). The basis we use is therefore the direct product \(|i \alpha \sigma\rangle = |i\rangle \otimes |\alpha\rangle \otimes |\sigma\rangle\). The single-particle energies are \( \tilde{\epsilon}_i = \epsilon_s + (\epsilon_s + \epsilon_a)/2 \), and the tunneling parameter is \( t = (\epsilon_s - \epsilon_a)/2 \), with \( \epsilon_s, \epsilon_a \) double-well eigenenergies, and \( \epsilon_a \) oscillator energies. Using the above expressions, the three-dimensional Coulomb integrals and the tunneling parameter are computed directly from the single-particle states for each sample.
To obtain the many-body energies and eigenstates, the Hamiltonian $H$ is then diagonalized exactly for each value of $N$ (total number of electrons) on each configuration subspace labeled by the quantum numbers $S$ (z-component of the total spin) and $M$ (z-component of the total angular momentum).

This approach has two main advantages: First, it allows us to solve the many-body problem consistently in the different coupling regimes, from the limit where $t$ is much greater than the Coulomb integrals to the opposite limit where $t$ is negligible. Second, it can provide quantitative predictions for given DQD structures, since it contains no free parameters and uses realistic values of $t$, $U_{\alpha\beta}$, $\tilde{U}_{\alpha\beta}$, and $J_{\alpha\beta}$ calculated for each nanostructure[5].

RESULTS AND DISCUSSION

Fig. 1(b) shows the calculated ground-state energies of correlated $N$-particle states as a function of the inter-dot distance (barrier width) $d$. The three lowest excited states are also shown for comparison. As expected, when $d$ is large, the system behaves as two isolated QDs. With decreasing $d$, some of the many-body excited states, favored by Coulomb interactions, become lower in energy. For $N \leq 3$ a single quantum phase transition occurs, below which the new ground state is a molecular-like state. For $N > 3$, two successive transitions take place, at $d = d_b$ and $d = d_a$, and an intermediate non-trivial phase is predicted to occur in the range $d_a < d < d_b$. Note that this phase is stable in a relatively large range of $d$ values, which depends on the number of electrons. An accurate determination of $d_a$ and $d_b$ requires the correct inclusion of all inter-dot coupling terms, including the inter-dot Coulomb integrals. If the latter are neglected, all quantum transitions occur for smaller values of the inter-dot distance, and the $d$-range of the intermediate phase is underestimated significantly. If we completely neglect all the many-body terms in eq. (1), we obtain the ground state configurations shown in fig. 1(a). Here the intermediate phase disappears, and for $3 \leq N \leq 6$ we have a simple crossover between single Slater determinants, at $d = d_a = d_b$. This proves that the appearance of the intermediate phase is an intrinsic correlation phenomenon.

To understand the nature of the different phases it is useful to examine the many-body states in terms of the single Slater determinants that contribute to each of them. However, one should remember that, even if a single configuration is a good approximation for the ground state wavefunction when $d$ is very small (LCAO-like) or large (Heitler-London-like), this is not the case when $d$ has an intermediate value[6]. Here we discuss explicitly the 4-electron case with the help of Fig. 2, but the same reasoning can be used for the other cases. For large values of $d$ ($d > d_b$), the relevant configurations for the ground state have two electrons in the lowest level of each isolated dot; the $|s\rangle$ and $|a\rangle$ extended ‘molecular’ orbitals derived from the lowest ‘atomic’ states are of course almost degenerate and both filled with two electrons. In the opposite limit ($d < d_a$), by expanding the localized atomic orbitals in terms of molecular orbitals we recognize that the $|s\rangle$ state derived from the lowest atomic state is filled with two electrons, while the corresponding $|a\rangle$ molecular state is empty. The two remaining electrons occupy the next bonding molecular orbitals — derived from the higher $p_x$ and $p_y$ levels— with parallel spin, in such a way that $S$ is maximized. This is the manifestation of Coulomb interaction which leads to Hund’s rule for molecules. In the intermediate phase, $d_a < d < d_b$, the $|a\rangle$ molecular state of the lowest shell is nearly degenerate with the $|s\rangle$ molecular state of the second shell, so the available states are different from the previous case. Coulomb direct and exchange terms,
Figure 1: Energies of the ground state (thick line) and three lowest excited states (thin lines) as a function of the inter-dot distance, \( d \), for a double QD occupied by \( N \) electrons. The total spin \( S \) and orbital angular momentum \( M \) are shown for every configuration. The confinement energy \( \hbar \omega \) is 10 meV, the inter-dot barrier height is 200 meV (the width of both wells is 4 nm), \( \kappa_r = 12.98 \), and \( m^* = 0.065m_e \). Panel (a): ground states of the single-particle Hamiltonian. Panel (b): ground states of the full GHH.

CONCLUSIONS

A large experimental effort is presently devoted to transport experiments in double QDs. In most cases, the dots are obtained by gating a two-dimensional electron gas (lateral confinement), and their coupling can be tuned through a gate voltage [7, 8, 9, 10, 11]. Indeed, a set of experiments has recently shown that there is a clear transition to a ‘coherent’ state with increasing coupling between dots [9, 11]. Outside this strong coupling
Figure 2: For $N = 4$, the prominent Slater determinants contributing to the many-body ground state are shown, for different values of the inter-dot distance $d$. Molecular orbitals are labeled according to the following notation: $\sigma, \pi$ stay for $m = 0, 1$, $u, g$ subscripts denote the symmetry under inversion $(x, y, z) \rightarrow (-x, -y, -z)$. Note that the $\pi$ molecular states are doubly degenerate because of the two-fold degeneracy of the second shell in the single-dots ($p_x, p_y$).

regime, however, experiments performed in the lateral geometry have so far evidenced classical interdot capacitance effects, probably owing to the size of the dots[12]. On the other hand, transport experiments are now available on DQD structures with strong lateral confinement fabricated by combined growth and etching techniques [4, 13]. The advantage is that the number of electrons in the structure is limited, while an accurate control on the inter-dot coupling is still possible by designing samples with appropriate barrier thickness. Both aspects are important for enhancing many-body effects and for exploring intermediate coupling regimes. Recent experimental work on these structures has focused on the weak- or strong- coupling regimes [4]. We hope that the present work will stimulate further investigations, since the precise locations of the relevant transitions are now predicted quantitatively and such transitions should occur in a range of parameters that is accessible to state-of-the-art experiments. We expect that such studies will bring new insight into electron-electron interaction effects in coupled quantum nanostructures.

ACKNOWLEDGMENTS

We acknowledge helpful discussions with C. Calandra and G. Goldoni. This work was supported in part by the EC under the TMR Network "Ultrafast Quantum Optoelectronics", and by the MURST-40% program "Physics of Nanostructures".
REFERENCES


5. The same approach was introduced for the case of isolated QDs giving addition spectra in good agreement with experiments [M. Rontani, F. Rossi, F. Manghi, and E. Molinari, Appl. Phys. Lett. 72, 957 (1998); Phys. Rev. B 59, 10165 (1999)]. In that case, however, the Hubbard hamiltonian reduces to an effective one-body hamiltonian since \( t \) and the inter-dot integrals are zero.


12. Interesting deviations from predictions of simple capacitance models are however reported in Refs. [7, 8].

13. DQDs with different geometries, obtained by cleaved-edge or self-organized growth, have recently become available but were so far investigated mostly by optical experiments. See e.g. G. Schedelbeck, W. Wegscheider, M. Bichler, G. Abstreiter, Science 278, 1792 (1997); N.N. Ledentsov et al., Phys. Rev. B 54, 8743 (1996); R. Cingolani et al. (1999), unpublished.
We considered the electronic structure of semiconductor vertical quantum dots. These state-of-the-art devices are a unique paradigm for a strongly correlated system where practically all parameters can be varied in a controlled way, and therefore display an exceptionally large phase diagram of different kinds of ground states. We limited our investigation to regimes of low magnetic field and spatial confinement strong enough to make the electronic atom-like shell structure experimentally observable.

Our first result was the explanation of some very intriguing features of measured addition spectra like open-shell spontaneous spin polarization, exchange effects, Hund’s rule, and electron term intersections in presence of an external magnetic field. The Generalized Hubbard Hamiltonian allowed for a very clear and physical picture of correlation effects, which are responsible for the global behavior of the system.

We checked also very accurately the role of the device geometry on open-shell effects. A related methodological issue which was investigated is the reliability of different possible approaches with regards to the correct treatment of correlation.

The double-dot system presents an even more rich phenomenology. We gave theoretical predictions of how electron terms depend on the inter-dot coupling and $B$. Non-trivial kinds of ground states were found with no analogue in “natural” molecules, and changes in the spontaneous magnetization at zero field were expected. Some preliminary experimental findings seem to confirm our results.

What emerges from our work is the great variety of different manifestations of the competition between confinement and correlation: if, on one hand, QDs presents many similarities with natural systems, on the other they can realize completely new physical states. In relation to this, the potential for new technological applications is enormous.

Maybe the most exciting aspect of this Thesis is the huge work still to be done in this field, both experimental and theoretical. Some extensions are obvious (more electrons, higher values of $B$), some are not. The phase space still to explore is truly vast.
Appendix A

Second Quantization

In this Appendix we introduce the Second Quantization formalism and according to this we write down a generic many-body Hamiltonian $\mathcal{H}$.

We assume a quite general expression for the Hamiltonian $\mathcal{H}$ of electrons in QDs:

$$\mathcal{H} = H^{(1)} + V^{(2)},$$ \hspace{1cm} (A.1)

where

$$H^{(1)} = \sum_{i=1}^{N_e} h^{(1)}(\mathbf{r}_i, s_i),$$ \hspace{1cm} (A.2)

and

$$V^{(2)} = \frac{1}{2} \sum_{i \neq j} \sum_{K \tau} \frac{e^2}{K r |\mathbf{r}_i - \mathbf{r}_j|}.$$ \hspace{1cm} (A.3)

The operator $h^{(1)}(\mathbf{r}_i, s_i)$ is a general one-particle Hamiltonian, acting on both space and spin coordinates. The meaning of all other symbols is the same that in Chapter 2. Our goal is to change the representation of the Hermitian operator $\mathcal{H}$, from the coordinate representation of Eqs. (A.2) and (A.3), to a new representation in the so called Fock space. In this new space the Hamiltonian is given by products of Fermionic operators and the Hilbert space is built up by successive applications of creation operators on the vacuum (occupation number representation).
To start we introduce a Fermi Field operator $\Psi(r, s)$:

$$\Psi(r, s) = \sum_{\alpha\sigma} \varphi_{\alpha\sigma}(r, s) c_{\alpha\sigma}. \quad (A.4)$$

Here $\{\varphi_{\alpha\sigma}(r, s)\}_{\alpha\sigma}$ is a complete and orthonormal set of single-particle states, i.e. with the property

$$\sum_{s=\pm 1/2} \int \varphi_{\alpha\sigma}^*(r, s) \varphi_{\beta\sigma'}(r, s) \, dr = \delta_{\alpha,\beta} \delta_{\sigma,\sigma'}, \quad (A.5)$$

and $c_{\alpha\sigma}$ ($c_{\alpha\sigma}^\dagger$) is a destruction (creation) Fermi operator associated with the single particle orbital $\alpha\sigma$, obeying Fermi anticommutation relations:

$$c_{\alpha\sigma} c_{\beta\sigma'}^\dagger + c_{\beta\sigma'}^\dagger c_{\alpha\sigma} = \delta_{\alpha,\beta} \delta_{\sigma,\sigma'}, \quad (A.6)$$

It is important to stress that single particle states $\varphi_{\alpha\sigma}(r, s)$ do not need to be a solution of a particular eigenvalue problem. The only thing which matters is that the set is complete and Eqs. (A.5) and (A.6) hold, hence in general the $\varphi_{\alpha\sigma}(r, s)$’s are not eigenfunctions of the single-particle Hamiltonian $h^{(1)}(r, s)$. The Fermionic operators act on a vacuum $|0\rangle$ defined by:

$$c_{\alpha\sigma} |0\rangle = 0 \quad \forall \alpha, \sigma. \quad (A.7)$$

From Eqs. (A.6) and from the definition (A.4) anticommutation relations for the Fermi fields follow:

$$\Psi(r, s) \Psi^\dagger(r', s') + \Psi^\dagger(r', s') \Psi(r, s) = \delta(r - r') \delta_{s, s'},$$

$$\Psi(r, s) \Psi(r', s') + \Psi(r', s') \Psi(r, s) = 0. \quad (A.8)$$

These latter relations allow us to interpret the Fermi field $\Psi^\dagger(r, s)$ as the operator which creates an electron with spin up and one with spin down at the position $r$. $\delta(r - r')$ is the Dirac’s delta.

Now we follow Ref. [133] to obtain the Second Quantization form of $H$ in terms of Fermi fields $\Psi$ and $\Psi^\dagger$. The one- and two-body terms of the Hamiltonian $H$ are, respectively:

$$H^{(1)} = \sum_{s=\pm 1/2} \int \Psi^\dagger(r, s) h^{(1)}(r, s) \Psi(r, s) \, dr \quad (A.9)$$
and

\[ V^{(2)} = \frac{1}{2} \sum_{s = \pm 1/2} \sum_{s' = \pm 1/2} \int \int \Psi^\dagger(r, s) \Psi^\dagger(r', s') \frac{e^2}{\kappa_r |r - r'|} \Psi(r', s') \Psi(r, s) \, dr \, dr'. \]  

(A.10)

If we substitute expression (A.4) for \( \Psi \) into (A.9) and (A.10) we obtain:

\[ H^{(1)} = \sum_{\alpha \sigma} \varepsilon^0_{\alpha \sigma} n_{\alpha \sigma} + \sum_{\sigma} \sum_{\alpha, \beta} t^0_{\alpha \sigma, \beta \sigma} c^\dagger_{\alpha \sigma} c_{\beta \sigma}, \]  

(A.11)

where we have separated the diagonal matrix elements \( \varepsilon_{\alpha \sigma} \) from those non-diagonal \( t^0_{\alpha \sigma, \beta \sigma} \), and

\[ V^{(2)} = \frac{1}{2} \sum_{\alpha \sigma} \sum_{\beta \sigma'} \sum_{\gamma} \sum_{\delta} V_{\alpha \sigma, \beta \sigma'; \gamma \sigma', \delta \sigma} c^\dagger_{\alpha \sigma} c_{\beta \sigma'} c_{\gamma \sigma'} c_{\delta \sigma'}, \]  

(A.12)

Here \( n_{\alpha \sigma} = c^\dagger_{\alpha \sigma} c_{\alpha \sigma} \) is the number occupation operator, and the meaning of the parameters entering Eqs. (A.11) and (A.12) is the following:

\[ \varepsilon^0_{\alpha \sigma} = \sum_{s = \pm 1/2} \int \varphi^\dagger_{\alpha \sigma}(r, s) h^{(1)}(r, s) \varphi_{\alpha \sigma}(r, s) \, dr \]  

(A.13)

is an average single-particle energy,

\[ t^0_{\alpha \sigma, \beta \sigma} = \sum_{s = \pm 1/2} \int \varphi^\dagger_{\alpha \sigma}(r, s) h^{(1)}(r, s) \varphi_{\beta \sigma}(r, s) \, dr \]  

(A.14)

is an hopping term, and

\[ V_{\alpha \sigma, \beta \sigma'; \gamma \sigma', \delta \sigma} = \sum_{s, s' = \pm 1/2} e^2 \int \int \frac{\varphi^\dagger_{\alpha \sigma}(r, s) \varphi_{\beta \sigma'}^\dagger(r', s') \varphi_{\gamma \sigma'}(r', s') \varphi_{\delta \sigma}(r, s)}{\kappa_r |r - r'|} \, dr \, dr'. \]  

(A.15)

is a Coulomb Matrix element.
Appendix B

Coulomb Matrix Elements

In this Appendix we derive an analytic expression for the generic Coulomb matrix element in the case of a single two-dimensional QD[134, 135, 136].

We start with the expression (A.15) of the Coulomb matrix element $V_{\alpha\sigma, \beta\sigma'; \gamma\sigma', \delta\sigma}$ written with respect to generic single-particle orbitals $\varphi_{\alpha\sigma}(r, s)$:

$$V_{\alpha\sigma, \beta\sigma'; \gamma\sigma', \delta\sigma} = \sum_{s, s'=\pm\frac{1}{2}} e^2 \int \int \frac{\varphi_{\alpha\sigma}^*(r, s) \varphi_{\beta\sigma}^*(r', s') \varphi_{\gamma\sigma'}(r', s') \varphi_{\delta\sigma}(r, s)}{\kappa_r |r - r'|} \, dr \, dr'.$$

We shall now compute this quantity in the case in which the $\varphi_{\alpha\sigma}(r, s)$’s are the eigenfunctions of the purely two-dimensional Hamiltonian $H_0(r)$ of Eq. (2.16), whose expression (spatial part) is given by Eq. (2.42):

$$\psi_{n,m}(\rho, \varphi) = \lambda^{[|m|+1]/2} \sqrt{\frac{n!}{\pi (n + |m|)!}} \rho^{|m|} e^{-\lambda\rho^2/2} L_n^{|m|} (\lambda \rho^2) e^{-i m \varphi}.$$  

Because the spatial part of the eigenfunction does not depend on the spin, we can suppress the spin index in the Coulomb matrix element. Moreover, Coulomb interaction preserves the total angular momentum $L_z$ in the scattering of two electrons. Hence, if $(n_1, m_1), (n_2, m_2), (n_3, m_3),$ and $(n_4, m_4)$ are the quantum numbers of incident and scattered electrons, respectively, we a priori know that the only non-zero matrix elements are those with

$$m_1 + m_2 = M = m_3 + m_4.$$

(B.1)
Now we substitute Eq. (2.42) for $\varphi_{\alpha \sigma}(r, s)$ into Eq. (A.15), working with dimensionless coordinates $[(x, y) = \lambda^{1/2}r]$:

$$
V_{n_1m_1,n_2m_2;n_3m_3,n_4m_4} = \frac{e^2 \sqrt{\lambda}}{\kappa_r} \frac{1}{\pi^2} \left( \frac{n_1!n_2!n_3!n_4!}{(n_1 + |m_1|)!(n_2 + |m_2|)!(n_3 + |m_3|)!(n_4 + |m_4|)!} \right)
\times \int dx_1 \int dy_1 \int dx_2 \int dy_2 \frac{\rho_1^{m_1} \rho_2^{m_2} \rho_3^{m_3} \rho_4^{m_4}}{\sqrt{(x_1 - x_2)^2 + (y_1 - y_2)^2}}
\times L_{n_1}^{m_1}(\rho_1^2) L_{n_2}^{m_2}(\rho_2^2) L_{n_3}^{m_3}(\rho_3^2) L_{n_4}^{m_4}(\rho_4^2)
\times e^{-\left(\rho_1^2 + \rho_2^2\right)} e^{im_1 \varphi_1} e^{im_2 \varphi_2} e^{-im_3 \varphi_3} e^{-im_4 \varphi_4}.
$$

(B.2)

Note that in Eq. (B.2) the only dimensional parameter is the prefactor

$$
\frac{e^2 \sqrt{\lambda}}{\kappa_r} = \sqrt{\frac{\varepsilon^4 m^s}{\kappa_r^2 \hbar^2}} \frac{1}{\Omega} = \frac{\sqrt{2\mathcal{R}^*(\hbar \Omega)}}{\sqrt{2\mathcal{R}^*(\hbar \Omega)}},
$$

(B.3)

where $\mathcal{R}^* = \varepsilon^4 m^s / (2\hbar^2 \kappa_r^2)$ is the effective Rydberg constant of the host material.

We carry on our derivation recovering the expansion (2.40) of the Generalized Laguerre Polynomial

$$
L_{n}^{m}(\rho^2) = \sum_{j=0}^{n} (-1)^j \left( \begin{array}{c} n + |m| \\ n - j \end{array} \right) \frac{\rho^{2j}}{j!}
$$

and substituting it for its compact expression in (B.2):

$$
V_{n_1m_1,n_2m_2;n_3m_3,n_4m_4} = \frac{e^2 \sqrt{\lambda}}{\kappa_r} \frac{1}{\pi^2} \left[ \prod_{i=1}^{4} \frac{n_i!}{(n_i + |m_i|)!} \right]^{1/2}
\times \sum_{(4j=0)} \frac{(-1)^{j_1+j_2+j_3+j_4}}{j_1!j_2!j_3!j_4!} \prod_{l=1}^{4} \left( n_l + |m_l| \right) \int dx_1 \int dy_1 \int dx_2 \int dy_2
\times \frac{\rho_1^{m_1} \rho_2^{m_2} \rho_3^{m_3} \rho_4^{m_4}}{\sqrt{(x_1 - x_2)^2 + (y_1 - y_2)^2}}
\times e^{-\left(\rho_1^2 + \rho_2^2\right)} e^{im_1 \varphi_1} e^{im_2 \varphi_2} e^{-im_3 \varphi_3} e^{-im_4 \varphi_4}.
$$

(B.4)

with

$$
\sum_{(4j=0)} \prod_{i=1}^{4} \frac{n_i!}{(n_i + |m_i|)!} = \prod_{i=1}^{4} \frac{n_i!}{(n_i + |m_i|)!}.
$$
At this point, introducing the definitions

\[\gamma_1 = j_1 + j_4 + (|m_1| + m_1) / 2 + (|m_4| - m_4) / 2,\]
\[\gamma_2 = j_2 + j_3 + (|m_2| + m_2) / 2 + (|m_3| - m_3) / 2,\]
\[\gamma_3 = j_2 + j_3 + (|m_2| - m_2) / 2 + (|m_3| + m_3) / 2,\]
\[\gamma_4 = j_1 + j_4 + (|m_1| - m_1) / 2 + (|m_4| + m_4) / 2,\]

we may write (B.4) as

\[V_{n_1m_1n_2m_2n_3m_3n_4m_4} = \frac{e^2 \sqrt{\lambda}}{\kappa_r \pi^2} \left[ \prod_{i=1}^{4} \left( \frac{n_i!}{(n_i + |m_i|)!} \right) \right]^{1/2} \]
\[\times \sum_{(j_1j_2j_3j_4)_j=0}^{n} \frac{(-1)^{j_1+j_2+j_3+j_4}}{j_1!j_2!j_3!j_4!} \prod_{i=1}^{4} \left( \frac{n_i + |m_i|}{n_i - j_i} \right) \int dx_1 \int dy_1 \int dx_2 \int dy_2 \]
\[\times \frac{e^{-\left(\rho_1^2 + \rho_2^2\right)}}{\sqrt{(x_1 - x_2)^2 + (y_1 - y_2)^2}} \left( \rho_1 e^{i\varphi_1} \right)^{\gamma_1} \left( \rho_2 e^{i\varphi_2} \right)^{\gamma_2} \left( \rho_2 e^{-i\varphi_2} \right)^{\gamma_3} \left( \rho_1 e^{-i\varphi_1} \right)^{\gamma_4}. \]

(B.6)

It is convenient to define the multiple integral \(I\):

\[I = \int dx_1 \int dy_1 \int dx_2 \int dy_2 \frac{e^{-\left(\rho_1^2 + \rho_2^2\right)}}{\sqrt{(x_1 - x_2)^2 + (y_1 - y_2)^2}} \]
\[\times \left( \rho_1 e^{i\varphi_1} \right)^{\gamma_1} \left( \rho_2 e^{i\varphi_2} \right)^{\gamma_2} \left( \rho_2 e^{-i\varphi_2} \right)^{\gamma_3} \left( \rho_1 e^{-i\varphi_1} \right)^{\gamma_4}. \]

(B.7)

We now separate integration variables into center of mass \(\mathbf{R}\) and relative motion \(\rho\) coordinates

\[\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2) / 2, \quad \rho = \mathbf{r}_1 - \mathbf{r}_2, \]

(B.8)

identifying the two-dimensional real space with the complex plane, namely

\[(x_1, y_1) \equiv \rho_1 e^{i\varphi_1} = z_1 = Z + z/2, \quad (x_2, y_2) \equiv \rho_2 e^{i\varphi_2} = z_2 = Z - z/2, \]
\[Z = (z_1 + z_2) / 2, \quad z = z_1 - z_2. \]

Eq. (B.7) turns into

\[I = \int dZ e^{-2|Z|^2} \int dz |z|^{-1} e^{-|z|^2} (Z + z/2)^{\gamma_1} \]
\[\times (Z - z/2)^{\gamma_2} (Z^* - z^*/2)^{\gamma_3} (Z^* + z^*/2)^{\gamma_4}. \]

(B.9)
Making use of the binomial theorem

\[(a + b)^n = \sum_{i=0}^{n} \binom{n}{i} a^i b^{n-i}\]  

Eq. (B.10) is reduced to

\[I = \sum_{(\ell_i) \in \mathbb{Z}} \prod_{i=1}^{4} \frac{\gamma_i}{\ell_i} (-1)^{\gamma_2 + \gamma_3 - \gamma_4} \int d\ell_1 e^{-2|\ell_1|^2} \int d\ell_2 e^{-2|\ell_2|^2} \int d\ell_3 e^{-2|\ell_3|^2} \int d\ell_4 e^{-2|\ell_4|^2}\]

\[\times \int d\ell \int d\ell' |\ell|^\Lambda e^{-2|\ell|^2} \int d\ell \int d\ell' |\ell'|^\Lambda e^{-2|\ell'|^2}\]

with the definitions

\[G = \gamma_1 + \gamma_2 + \gamma_3 + \gamma_4, \quad \Lambda = \ell_1 + \ell_2 + \ell_3 + \ell_4.\]  

Eq. (B.12) becomes:

\[I = \delta_{m_1, m_2, m_3, m_4} 4\pi^2 \sum_{(\ell_i) \in \mathbb{Z}} \prod_{i=1}^{4} \frac{\gamma_i}{\ell_i} (-1)^{\gamma_2 + \gamma_3 - \gamma_4} \int d\ell_1 e^{-2|\ell_1|^2} \int d\ell_2 e^{-2|\ell_2|^2} \int d\ell_3 e^{-2|\ell_3|^2} \int d\ell_4 e^{-2|\ell_4|^2}\]

\[\times \int d\ell \int d\ell' |\ell|^\Lambda e^{-2|\ell|^2} \int d\ell \int d\ell' |\ell'|^\Lambda e^{-2|\ell'|^2} \times \Gamma\left(\Lambda/2 + 1\right) \Gamma\left(|G - \Lambda + 1|/2\right)\]  

After some manipulation, using the definition of Gamma function \(\Gamma(x)\) (note that \(G \geq \Lambda\)), we obtain

\[I = \delta_{m_1, m_2, m_3, m_4} 2^{-G/2} \sum_{(\ell_i) \in \mathbb{Z}} \prod_{i=1}^{4} \frac{\gamma_i}{\ell_i} (-1)^{\gamma_2 + \gamma_3 - \gamma_4} \int d\ell_1 e^{-2|\ell_1|^2} \int d\ell_2 e^{-2|\ell_2|^2} \int d\ell_3 e^{-2|\ell_3|^2} \int d\ell_4 e^{-2|\ell_4|^2}\]

\[\times \Gamma\left(\Lambda/2 + 1\right) \Gamma\left(|G - \Lambda + 1|/2\right)\]  

Performing the integration over azimuthal angles we obtain the two constraints \(\ell_1 + \ell_2 = \ell_3 + \ell_4\) and \(\gamma_1 + \gamma_2 - \gamma_3 - \gamma_4 = 0\). This latter condition is equivalent to angular momentum conservation (B.1), as it is easy to verify from definitions (B.5).
and finally

\[ V_{n_1 m_1, n_2 m_2, n_3 m_3, n_4 m_4} = \delta_{m_1 + m_2, m_3 + m_4} \frac{e^2 \sqrt{\lambda}}{\kappa r} \left[ \prod_{i=1}^{4} \frac{n_i!}{(n_i + |m_i|)!} \right]^{1/2} \]

\[ \times \sum_{(4)j=0}^{n} \frac{(-1)^{j_1 + j_2 + j_3 + j_4}}{j_1! j_2! j_3! j_4!} \prod_{l=1}^{4} \left( \frac{n_l + |m_l|}{n_l - j_l} \right) \]

\[ \times 2^{-G/2 - 1/2} \sum_{(4)\ell=0}^{j} \delta_{\ell_1 + \ell_2, \ell_3 + \ell_4} \prod_{l=1}^{4} \left( \frac{\gamma_l}{\ell_l} \right) \frac{(-1)^{\gamma_2 + \gamma_3 - \ell_2 - \ell_3}}{\Gamma(\Lambda/2 + 1) \Gamma([G - \Lambda + 1]/2)} \].

(B.16)
Appendix C

Wigner-von Neumann Theorem

In this appendix we derive the Wigner-von Neumann theorem presented in Sec. 3.2[133].

Theorem C.1 (E. Wigner and J. von Neumann 1929) If electron terms depend on one parameter, only terms of different symmetry can intersect, while the intersection of terms of like symmetry is impossible.

Let’s take as parameter on which electron terms depend the inter-dot coupling $t$. They can be represented graphically by plotting the energy as a function of $t$. Let $U_1(t), U_2(t)$ be two different electronic terms. If they intersect at some point, then the functions $U_1$ and $U_2$ will have neighboring values near this point. To decide whether such an intersection can occur, it is convenient to put the problem as follows. Let us consider a point $t_0$ where the functions $U_1(t), U_2(t)$ have very close but not equal values (which we denote by $E_1, E_2$), and examine whether or not we can make $U_1$ and $U_2$ equal by displacing the point a short distance $\delta t$. The energies $E_1$ and $E_2$ are eigenvalues of the Hamiltonian $\mathcal{H}^{GHH}$ of the dots with a coupling strength $t_0$. If we add to the hopping coefficient $t_0$ an increment $\delta t$, the Hamiltonian becomes $\mathcal{H}^{GHH} + V$, where $V = \delta t (\partial \mathcal{H}^{GHH}/\partial t)$ is a small correction; the values of the functions $U_1, U_2$ at the point $t_0 + \delta t$ can be regarded as eigenvalues of the new Hamiltonian. This enables us to determine the values of the terms $U_1(t), U_2(t)$ at the point $t_0 + \delta t$ by means of perturbation theory, $V$ being regarded as a perturbation to the operator $\mathcal{H}^{GHH}$. 
The ordinary method of perturbation theory is here inapplicable, however, since the eigenvalues $E_1, E_2$ of the energy in the unperturbed problem are very close to each other, and their difference is in general small compared with the magnitude of the perturbation. Since, in the limit where the difference $E_2 - E_1$ tends to zero, we have the case of degenerate eigenvalues, it is natural to attempt to apply to the case of close eigenvalues the degenerate perturbation theory.

Let $|\psi_1\rangle, |\psi_2\rangle$ the eigenstates of the unperturbed operator $H^{\text{GHH}}$ which correspond to the energies $E_1, E_2$. As an initial zero-order approximation we take, instead of $|\psi_1\rangle$ and $|\psi_2\rangle$ themselves, linear combinations of them of the form

$$|\psi\rangle = c_1|\psi_1\rangle + c_2|\psi_2\rangle.$$  \hfill (C.1)

Substituting this expression in the perturbed equation

$$\left(H^{\text{GHH}} + V\right)|\psi\rangle = E|\psi\rangle,$$  \hfill (C.2)

we obtain

$$c_1 (E_1 + V - E) |\psi_1\rangle + c_2 (E_2 + V - E) |\psi_2\rangle = 0.$$  \hfill (C.3)

Multiplying this equation on the left by $\langle \psi_1 |$ and $\langle \psi_2 |$ in turn, we have two algebraic equations:

$$c_1 (E_1 + V_{11} - E) + c_2 V_{12} = 0,$$

$$c_1 V_{21} + c_2 (E_2 + V_{22} - E) = 0,$$  \hfill (C.4)

where $V_{ik} = \langle \psi_i | V | \psi_k \rangle$, since the operator $V$ is Hermitian, the quantities $V_{11}$ and $V_{22}$ are real while $V_{12} = V_{21}^*$. The compatibility conditions for these equation is

$$\begin{vmatrix}
E_1 + V_{11} - E & V_{12} \\
V_{21} & E_2 + V_{22} - E
\end{vmatrix} = 0,$$  \hfill (C.5)

whence we obtain after some calculation

$$E = (E_1 + E_2 + V_{11} + V_{22})/2 \pm \sqrt{(E_1 - E_2 + V_{11} - V_{22})^2 + |V_{12}|^2}/2.$$  \hfill (C.6)

This formula gives the required eigenvalues of the energy in the first approximation.
If the energy values of the two terms become equal at the point $t_0 + \delta t$ (i.e. the terms intersect), this means that the two values of $E$ given by formula (C.6) are the same. For this to happen, the expression under the radical in (C.6) must vanish. Since it is the sum of two squares, we obtain, as the condition for there to be points of intersection of the terms, the equations

$$E_1 - E_2 + V_{11} - V_{22} = 0, \quad V_{12} = 0.$$  \hspace{1cm} (C.7)

However, we have at our disposal only one arbitrary parameter giving the perturbation $V$, namely the magnitude $\delta t$ of the displacement. Hence the two equations (C.7) cannot in general be simultaneously satisfied (we suppose that the functions $|\psi_1\rangle, |\psi_2\rangle$ are chosen to be real, so that $V_{12}$ also is real).

It may happen, however, that the matrix element $V_{12}$ vanishes identically; there then remains only one equation (C.7), which can be satisfied by a suitable choice of $\delta t$. This happens in all cases where the two terms considered are of different symmetry. This statement holds because the operator $V$ (like the Hamiltonian $\mathcal{H}_{\text{GH}}$) commutes with all the symmetry operators for the system. Q.E.D.

If, as a result of some approximate calculation, we obtain two intersecting terms of the same symmetry, they are found to move apart on calculating the next approximation, as shown by the continuous lines in Fig. C.1.
Figure C.1: Diagram of electron terms $U(t)$ vs the hopping coefficient $t$. If, as a result of some approximate calculation, we obtain two intersecting terms of the same symmetry, they are found to move apart on calculating the next approximation, as shown by the continuous lines.
Acknowledgements

I want to thank my supervisors, Franca and Elisa, for all the support, encouragement and stimulation they always gave me. It was very nice to work with them. It was a pleasure also to work with Prof. Fausto Rossi, who is now in Torino. I am in debt with Professor Sham at UCSD with whom I worked for one year. It was a very exciting experience, and I think I learned a lot about Physics. Thanks also to Prof. S. G. Louie and M. L. Cohen at UCB for their very kind hospitality in Berkeley. I acknowledge very useful discussions about quantum dots with Prof. Daniela Pfannkuche, Prof. Pawel Hawrylak, Dr. Axel Lorke, Dr. J. J. Palacios, Dr. Davide Vanossi, and last but not least, Dr. Guido Goldoni. Thanks for technical support to Gianpaolo Prampolini, Cristina Murari, and Roberta Cantaroni of Computer Center at University of Modena, and to G. Casalini for his kind help in nasty bureaucracy matters.

It is really an impossible task to thank all friends and colleagues in Modena and around the world with whom I worked and had fun: I would not have resisted without them. Let me just say thank to my family: babbo, mamma, zia and fratellino. Last word is for Emanuela.
Bibliography


[83] Eyal Goldmann and Scot R. Renn, cond/mat/9909071.


If the Hartree-Fock potential has not spherical (or cylindrical, for a vertical QD) symmetry, as it is the case e.g. in Atomic Physics for atoms with the outer valence shell not completely full (the open-shell regime in which the ground state configuration is predicted by Hund’s rules), then this statement is only approximately true. However it is believed that non-diagonal $\lambda_{ij}$ are negligible. For this point see J. C. Slater, *Quantum Theory of Atomic Structure*, vol. II, (McGraw-Hill, New York, 1960), sec. 17-1.


