Chapter 4

Electronic correlations as a function of the confinement energy

In Chapter 2 I have found the single-particle spectra of the most important classes of QD potentials, and in Chapter 3 I have formulated the problem of many interacting particles confined in QDs. In order to solve this problem, I have introduced the exact diagonalisation method with optimised many-particle basis set. I have demonstrated that this method treats all aspect of particle-particle interactions on equal footing, and, within the assumed cutoff of the single-particle basis, delivers the exact eigenenergies of the system. In the following chapters I shall use the theoretical tools thus constructed to analyse the properties of systems of many interacting particles confined by nanostructures with various geometry, with special attention devoted to the manifestations of particle-particle correlation effects.

My presentation starts with the description of properties of N electrons confined in a parabolic quantum dot at zero magnetic field. This description is given in the paper "Designing quantum systems in self-assembled quantum dots", by M. Korkusiński, W. Sheng, and P. Hawrylak, published in Physica Status Solidi (b), vol. 238, page 246

(2003). This publication is an integral part of this thesis and is appended to the presented material. Below I shall highlight the most important points of this work.

Let us start by comparing the energy scale of the single-particle energy quantisation with that of Coulomb interactions. For a parabolic dot the fundamental scale of the single-particle energy quantisation is introduced by the characteristic oscillator energy $\Omega_0 = \hbar\omega_0/\mathcal{R}$ (see Section 2.1; $\mathcal{R}$ is the effective Rydberg). As for the interactions, their strength can be measured, e.g., by the magnitude of the fundamental Coulomb matrix element described in Section 3.1.2:

$$\langle 00, 00|v|00, 00\rangle = E_0 = \frac{\sqrt{\pi}}{\ell}, \tag{4.1}$$

where the oscillator length ℓ at zero magnetic field, when expressed in the effective Bohr radii, is simply $\ell = 1/\sqrt{\Omega_0}$. Therefore, the characteristic energy $E_0 = \sqrt{\pi\Omega_0}$.

Let us now compare these two energy scales. The single-particle energy quantisation scales linearly with the oscillator energy, while the interactions scale as square root of Ω_0. Then, as I tune Ω_0 - by changing the dot size in the case of SADs, or by tuning the gate voltage in the case of gated devices - I can change the ratio of these two energies. The characteristic single-particle energy becomes equal to that of interactions when $\Omega_0 = \pi$. If the confinement is stronger (Ω_0 larger), then the single-particle energy quantisation is larger than interactions, and if the confinement is softer ($\Omega_0 < \pi$), the situation is reversed.

Thus, when considering the system of many interacting particles one expects to find two regimes. In QDs with strong confinement the single-particle energy quantisation dominates, and one can construct the ground state of the system just by distributing the particles on the single-particle levels with the lowest energies, seeing only that the Pauli exclusion principle is satisfied. The total energy of the system can be then calculated perturbatively. On the other hand, for QDs with sufficiently soft confinement the interactions will dominate the energy landscape of the system. One expects strong configuration mixing, and so one can no longer anticipate the ground-state configuration reliably: it

has to be calculated. It is possible that the ground states of the systems in each of the regimes will be different.

To demonstrate this, I shall now consider a system of three electrons in a parabolic quantum dot with two single-particle shells. As I demonstrated in Section 2.1, the lowest, s shell in the parabolic potential consists of one doubly-spin-degenerate state with single-particle angular momentum $l = 0$. The second, p shell, on the other hand, consists of two doubly-spin-degenerate states, one with angular momentum $l = -1$, and one with angular momentum $l = +1$. This simple structure of the single-particle energy levels is presented in Figure 4.1 (a) and (b).

Let us now distribute my three electrons on the three orbitals. If I were to assume that the particles do not interact, the natural choice of the candidate for the ground state of the system would be that shown in Fig. 4.1 (a), since it has the lowest total energy. This configuration is built out of two electrons with opposite spins distributed on the s shell, and the third electron on the p shell. The total angular momentum of this state is $0 + 0 + (-1) = -1$, the total spin $S = 1/2$, and the projection of the total spin $S_z = -1/2$. One can construct another state with the same energy by putting the third electron on the other orbital of the p shell. This configuration would have the same total spin and projection as the previous one, but the total angular momentum of $+1$. Since the angular momenta of the two configurations are different, the Hamiltonian does not couple them, and they can be considered independently. Moreover, due to the symmetry of the system at zero magnetic field, the energy levels corresponding to these two configurations are degenerate, so it is sufficient to consider just one of them. The configuration $|L, S_z\rangle = |-1, -1/2\rangle$ shown in Fig. 4.1 (a) is not the only configuration that can be generated with this particular suite of quantum numbers. Another one would involve leaving the s shell completely empty, and putting one electron on the orbital with $l = 1$, and two electrons - with opposite spins - on the orbital $l = -1$. Therefore, the configuration I have chosen is not the exact many-body state of the system since

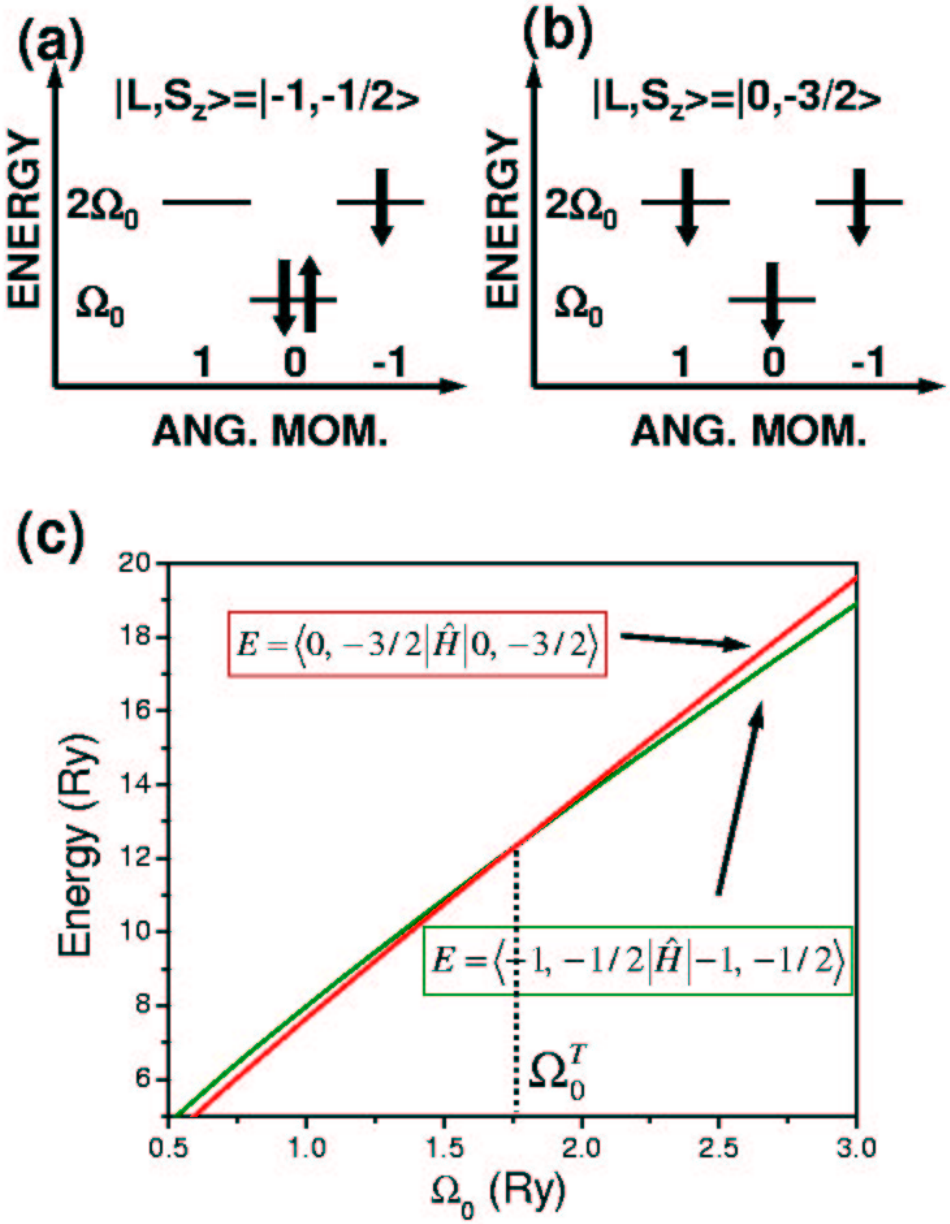

Figure 4.1: The configuration of the three-electron system with quantum numbers $|L, S_z\rangle = |-1, -1/2\rangle$ (a) and $|0, -3/2\rangle$ (b). The graph (c) shows the expectation values of the total energy of the system as a function of the confinement energy Ω_0 for the low-spin state (green line) and the high-spin state (red line)

it is mixed with the other one by Coulomb interactions. I shall include the effects of configuration mixing later on.

Let us now consider a different configuration, presented in Fig. 4.1 (b). This is the only way of distributing the three electrons on single-particle states if I require that all of them have the same spin. The total angular momentum of this configuration is zero, the total spin $S = 3/2$, and the projection of the total spin $S_z = -3/2$. This configuration, further referred to as $|L, S_z\rangle = |0, -3/2\rangle$, is the only configuration in its suite of quantum numbers, and is thus an *exact* many-body state in the two-shell approximation.

Let us now compare the energies of these two states as a function of Ω_0. The corresponding total energy in each case is calculated as the expectation value of the full many-body Hamiltonian

$$\hat{H} = \sum_{i\sigma} E(i,\sigma) c^+_{i\sigma} c_{i\sigma} + \frac{1}{2} \sum_{ijkl\sigma\sigma'} \langle i\sigma, j\sigma' | V | k\sigma', l\sigma \rangle c^+_{i\sigma} c^+_{j\sigma'} c_{k\sigma'} c_{l\sigma}. \tag{4.2}$$

Using the energies of single-particle states $E(nm\sigma) = \Omega_0(n+m+1)$ obtained in Section 2.1 and the Coulomb matrix elements calculated in Section 3.1.2, I can express these energies in the following form:

$$E_{-1,-1/2} = 4\Omega_0 + 2.25\sqrt{\pi\Omega_0}; \tag{4.3}$$

$$E_{0,-3/2} = 5\Omega_0 + 1.5\sqrt{\pi\Omega_0}. \tag{4.4}$$

The low-spin state has lower orbital energy, but higher interaction energy as compared to the high-spin state. Therefore, for strong confinements (large Ω_0) one may expect the low-spin state to be lower in energy than the high-spin state, and for small Ω_0 the situation may be reversed. This is indeed the case, as can be seen in Fig. 4.1 (c). In this graph I present the above energies as a function of the confinement energy Ω_0. For strong confinement the low-spin configuration is the ground state of the system. But as Ω_0 is decreased, the two energies cross, and a transition to the high-spin state occurs. This transition takes place at a critical value of confinement energy, further referred to as Ω_0^T. Note that this transition is due entirely to the interplay between the single-particle

(orbital) energy quantisation and direct and exchange Coulomb terms. In this simple model correlations play no role, since I have not considered the interaction mixing effects in the low-spin subspace.

To include the correlation effects in a controllable fashion, let us perform the exact diagonalisation study as a function of the confinement energy Ω_0 and the number of confined shells N_S. This time I fully account for *all* configurations possible within each subspace. Thus, the basis sets of the low-spin and the high-spin subspace no longer contain just one configuration, but the sizes of these sets grow factorially with N_S. This is shown in Fig. 4.2 (a), where I give the number of configurations in the low-spin (green bars) and high-spin subspace (red bars) as a function of N_S; inset to this figure shows the ratio of these numbers. Note that for all values of N_S except for $N_S = 3$ this ratio is approximately $2:1$, while for $N_S = 3$ it is almost $3:1$.

With these basis sets I have performed the exact diagonalisation studies as a function of the number of shells, and in most cases have observed the low-spin-high-spin transition. The characteristic value of confinement energy Ω_0^T as a function of N_S is shown in Fig. 4.2 (b). I see, in general, that as I increase the number of shells, the transition occurs for lower and lower confinements, with the exception of $N_S = 3$, where the transition does not occur at all (the ground state is the low-spin state for all values of Ω_0). This behaviour can be understood in the context of the number of available configurations in each subspace. As can be seen in Fig. 4.2 (a), for all N_S there are more low-spin configurations than the high-spin configurations. This is due to the fact that in the case of low spin one can distribute two electrons on the same orbital, while for spin-polarised configurations this is forbidden by the Pauli principle. The low-spin states, although not favoured by the direct and exchange Coulomb terms, have a correlation advantage over the high-spin states due to larger correlation mixing effects. It is particularly visible in the case of $N_S = 3$, where the low-spin basis set is particularly large as compared to the high-spin set, and the correlation advantage thus gained causes the low-spin state to be the ground state of the

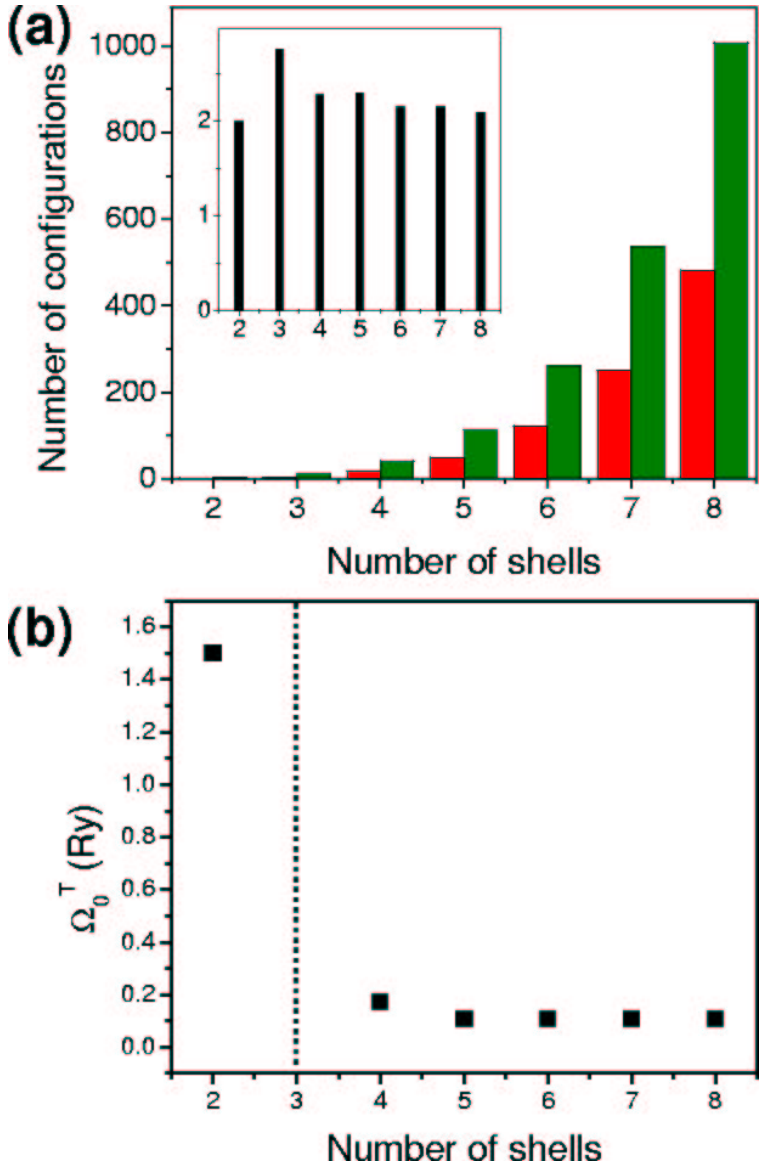

Figure 4.2: (a) Number of configurations in the low-spin subspace ($L = -1$, $S_z = -1/2$ - green bars) and in the high-spin subspace ($L = 0$, $S_z = -3/2$ - red bars) for a three-electron system as a function of the number of single-particle shells N_S. Inset shows ratios between these numbers. (b) Critical confinement energies Ω_0^T marking the low-spin - high-spin transition as a function of the number of shells N_S. For $N_S = 3$ there is no transition

system for all confinement energies. The progressive increase in basis sizes also causes the critical value Ω_0^T to decrease with the increase of N_S, and saturate around $N_S = 5$, where the convergence of the low-energy configurations is already achieved (at this stage further increase of N_S supplements the basis sets only with high-energy configurations, having a negligible effect on the lowest-energy states). As can be seen, by changing the number of available single-particle shells one can tune the electronic correlations. This tuning can be realized in the case of self-assembled quantum dots by engineering these nanostructures to contain only a desired number of single-particle orbitals.

The paper invoked in the beginning of this Chapter describes similar evolution of the system with the number of shells and confinement energy for up to eight confined electrons. The most important aspect of this work is the fact that one can create magnetic moments in quantum dots by appropriately engineering its single-particle and many-particle properties: the shell spacing, the number of confined shells, and the number of electrons distributed on them. For instance, in the case of $N_S = 4$ shells the electrons tend to align their spins as the confinement energy Ω_0 is lowered, which means that the Hund's rules and the magnetic moments associated with half-filled shells are not valid in the regime of strong interactions.