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Designing quantum systems in self-assembled quantum dots

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The possibility of designing electronic properties of quantum dots by varying shell spacing, number of confined shells, and number of electrons is investigated. Numerical results demonstrate the creation of magnetic moments beyond Hunds rules.

Designing electronic properties of quantum system requires the capability to design and control single particle energy levels and, more importantly, many-particle configurations. The many-particle aspect is particularly interesting as collective behavior of electrons leads to a number of nontrivial and useful phenomena, from magnetism to superconductivity. The design tools are the number and structure of single particle levels "i", the form of two-body interaction "V" among electrons $\langle i,j| V |k,l\rangle$, and the number of electrons in the dot N_e. The single particle levels can be controlled by shape, size and composition of a self-assembled quantum dot or by arrangment of gates and applied voltages in lateral or vertical quantum dots [1, 2]. The Coulomb interaction matrix elements are related to the single particle wave function and the form of interaction. The form of interaction can be modified by differences in dielectric constants or screening by metallic electrodes. Hence it is fair to assume that interaction matrix elements can be to some extent modified independently of the single particle spectrum. The problem therefore rests in the study of the Hamiltonian of the interacting electron systems:

$$H = \sum_i E_i^e c_i^+ c_i + \tfrac{1}{2} \sum_{ijkl} \langle i,j | V_{ee} | k,l \rangle \, c_i^+ c_j^+ c_k c_l \,. \tag{1}$$

The operators c_i^+ (c_i) create(annihilate) the electron $|i\rangle$ with the single-particle energy E_i, and Coulomb matrix elements $\langle i,j| V |k,l\rangle$ scatter electrons from states $|k,l\rangle$ to states $|i,j\rangle$. The key difficulty is the size of Hilbert space: for N_{sp} single particle states and N_e electrons there are $\binom{2N_{sp}}{N_e}$ configurations. Even for 10 orbitals and 7(10) electrons this number is 77,520 (184,756) and grows exponentially quickly with the system size. The calculations of electronic properties of a quantum dot with given ω_0, N_s, N_e were carried out using total spin resolved numerical diagonalization techniques employing conjugated gradient methods [3]. This means that Hilbert space has been divided into blocks corresponding to total angular momentum L, total spin projection S_z, and total spin S. For given N_e and L the calculation was carried out in each $S = S_z$ Hilbert spaces.

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In this work we focus on the quantum dot with finite parabolic confinement and strictly 2D Coulomb interactions. The single-particle energy levels $E(m,n) = \omega\,(m + n + 1)$ are that of two harmonic oscillators with frequency organized into a finite number N_s of degenerate shells. The electronic properties of such dots were studied and Hund's rules demonstrated theoretically [4] and experimentally [5]. The shell structure is realised in e.g. lens shaped self-assembled quantum dots. Fig. 1 shows the results of 8 band $k \cdot p$ calculation, including strain, of a lens shaped InAs/GaAs self-assembled dot with width 20 nm and height 2.3 nm. Each energy eigenvalue is assigned a profile of probability density corresponding to the eigenstate. The spectrum shows a finite number of energy shells, with the number of shells and their spacing controlled by dot radius and other processes, such as thermal annealing. The net result is the electron spectrum, which is well approximated by the spectrum of two harmonic oscillators.

Given the single particle spectrum characterised by a number N_s of equally spaced shells, the question arises what are the possible ground states of N_e electrons, and how do they depend on N_s and spacing ω_0. Some aspects of this problem were already addressed in Refs. [6, 7]. Let's consider what appears to be the simplest case of filled shells and one electron in an empty shell. The simplest example corresponds to filled s-shell and one electron in a p-shell. The state in question, illustrated in Fig. 2, has angular momentum $L = 1$ and $S = 1/2$. In Rydbergs, the energy of this state is $E(1,1/2) = 4\omega_0 + \sqrt{\pi}\,\sqrt{\omega_0}\,(2.25)$. It consists of kinetic energy $\sim\omega_0$ and the interaction energy $\sim\omega_0^{1/2}$. By changing the confinement energy we can change the ratio of kinetic to interaction energies. By comparison, a state in which all electrons have parallel spin has angular momentum $L = 0$ and total spin $S = 3/2$. Its energy is $E(0,3/2) = 5\omega_0 + \sqrt{\pi}\,\sqrt{\omega_0}\,(1.50)$. It has higher kinetic energy but lower interaction energy. The lowering of interaction energy is due to single occupancy of orbitals (reduced direct repulsion) and spin alignment (increased exchange energy). The design aspect appears when we vary confining energy: at

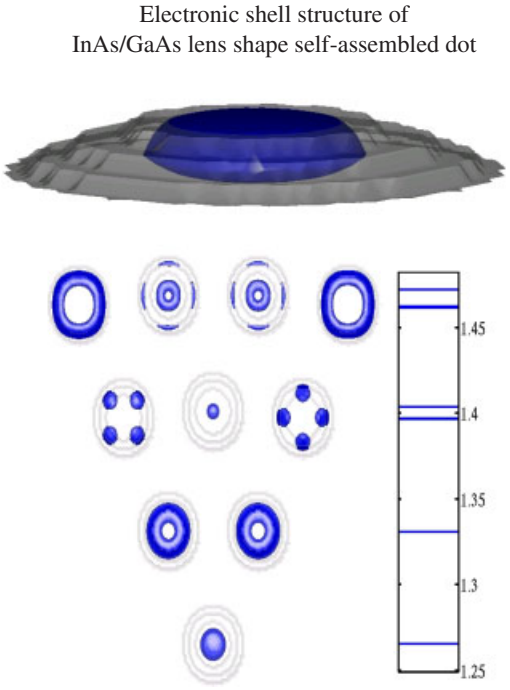

Fig. 1 (online colour at: www.interscience.wiley.com) Single particle energy levels and wavefunctions of a lens shaped InAs/GaAs self-assembled quantum dot.

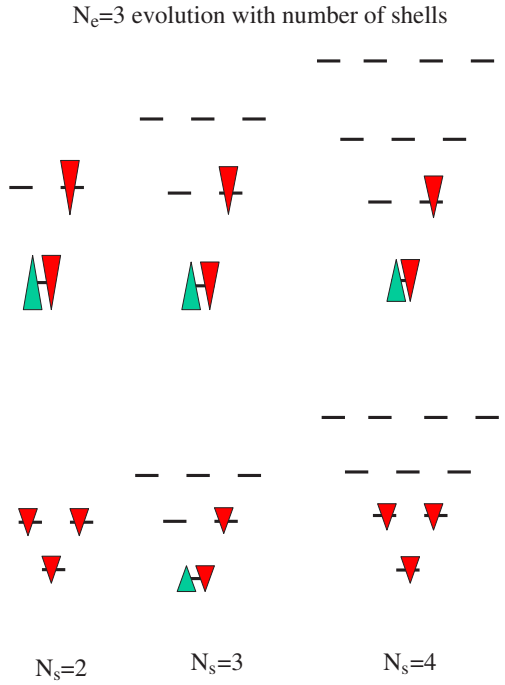

Fig. 2 (online colour at: www.interscience.wiley.com) Schematic representation of $N_e = 3$ electron configurations for $N_s = 2,3$ and 4 shells. Top panel: large shell spacing. Bottom panel: small shell spacing.

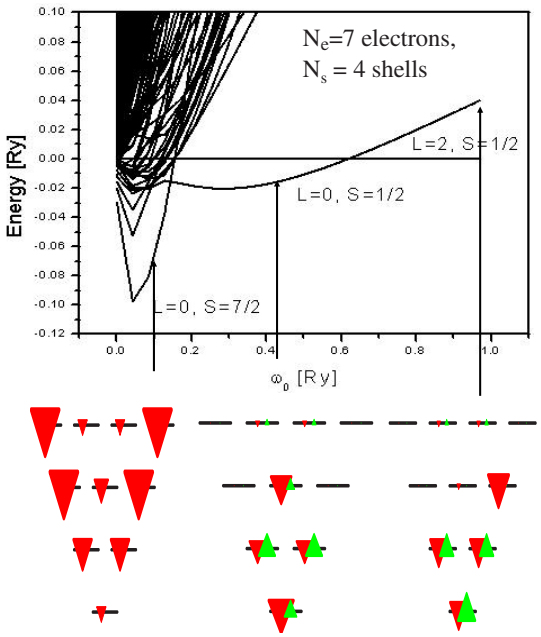

Fig. 3 (online colour at: www.interscience.wiley.com) Energy levels and example of configurations for $N_e = 7$ electrons on $N_s = 4$ shells as a function of shell spacing. The size of symbols reflects of occupation of corresponding orbital in a correlated ground state.

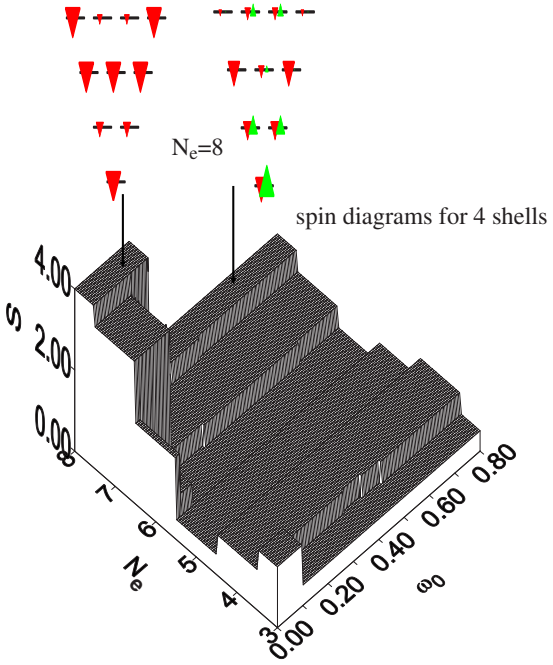

Fig. 4 (online colour at: www.interscience.wiley.com) Total spin of the ground state as a function of the number of electrons N_e and shell spacing.

high $\omega_0 > 1.767$ low spin state $L = 1$, $S = 1/2$ is the ground state, at low $\omega_0 < 1.767$ high spin state is the ground state. If direct and exchange energy were the only contributions there would always be a spin polarised state. However, there is correlation energy. The contribution from correlations can be tuned by changing the number of confined shells. This changes the number of possible configurations corresponding to different total spin states in a different manner. For example, adding a third shell stabilizes the $L = 1$, $S = 1/2$ state, and it is a ground state for all ω_0. Adding 4-th shell brings back transition to spin polarised state but at $\omega_0 = 0.18$. Hence by changing shell spacing and a number of shells one can design ground state of the three-electron system with different degree of spin polarisation. The next example of one electron in empty shell with filled core shells is the $N_e = 7$ electron droplet. Here 6 electrons doubly occupy the s- and p-shells, and one electron occupies orbitals of the d-shell. Fig. 3 shows results of spin resolved diagonalisation of the $N_e = 7$ electron Hamiltonian as a function of ω_0 for $N_s = 4$. For large ω_0 the electronic configuration is dominated by a single configuration of filled s and p shells and one electron in the $m = 2$, $n = 0$ state ($L = 2$, $S = 1/2$). When ω_0 is lowered to $\omega_0 = 0.4$, the d-electron transfers from the edge to the center of the dot ($L = 2$ $S = 1/2$) to ($L = 0$, $S = 1/2$). This angular momentum transition is followed by spin transition where all three spin up electrons flip their spin. The tendency of electrons to align spin when ω_0 is lowered, i.e., interactions dominate, is illustrated in Fig. 4. Fig. 4 shows total spin of the ground state of $N_e = 2$–8 electrons on $N_s = 4$ shells as a function of ω_0. We see that electrons in all droplets align their spin when shell spacing is lowered. Hence Hunds rules and magnetic moments associated with partially filled shells are valid only in weakly interacting systems.

In summary, we investigated the possibility of designing electronic properties of quantum dots by varying shell spacing, number of confined shells, and number of electrons. We showed that by lowering shell spacing all electron spins can be aligned and magnetic moments created.

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