Hund's rule, spin blockade, and the Aufbau principle in strongly confined semiconductor quantum dots

2000 Europhys. Lett. 50 243

(http://iopscience.iop.org/0295-5075/50/2/243)

EUROPHYSICS LETTERS 15 April 2000

Europhys. Lett., **50** (2), pp. 243–249 (2000)

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(received 24 November 1999; accepted in final form 2 February 2000)

PACS. 73.20.Dx – Electron states in low-dimensional structures (superlattices, quantum well structures and multilayers).

Abstract. – The ground-state configuration of a system of N electrons or holes ($N = 1 \cdots 8$) in strongly confined InAs, InP, and Si quantum dots (diameter ~ 30 Å) is calculated using pseudopotential single-particle energies and wave functions as input to the many-body expansion of the total energy. The validity of generally accepted "rules of level occupation" (Hund's rule, Aufbau principle, and single spin-flip rule) is examined. We find that while Hund's rule is generally obeyed, deviations from the Aufbau principle are common when single-particle energy levels are separated by a few meV. We also find a few instances where the single spin-flip rule is violated, leading to "spin blockade" in linear conductance.

When a quantum dot is weakly coupled to an electron reservoir, either via a gate structure [1–4] or an STM tip [5,6], one can load, one by one, N electrons or holes into the dot by changing the gate voltage or the tip-substrate bias. The ability to successively load carriers into the dot enables one to study the "rules of level occupation" determining the sequence in which the energy levels of the dot are occupied. These rules are best illustrated by considering a dot with a pair of spatially degenerate single-particle energy levels e_{a1}, e_{a2} and a non-degenerate energy level e_b such that the single-particle energies obey $\varepsilon(e_{a1}) = \varepsilon(e_{a2}) < \varepsilon(e_b)$.

i) *Hund's rule*: Degenerate single-particle levels are occupied with a maximum number of unpaired electrons. For example, the transition from $N = 1$ to $N = 2$ electrons will prefer

$$e_{a1}^{\uparrow} \xrightarrow{\text{Hund}} e_{a1}^{\uparrow}\, e_{a2}^{\uparrow} \tag{1a}$$

over

$$= e$$

$$e_{a1}^{\uparrow} \quad \text{Non--}$$

$$_{a2}).$$

ii) *The Aufbau principle*: Non-degenerate single-particle levels are occupied in order of increasing single-particle energy. For example, the $N = 2$ to $N = 3$ transition would prefer to

e ective Hamiltonian involving only the conduction band electrons, provided that the electron-

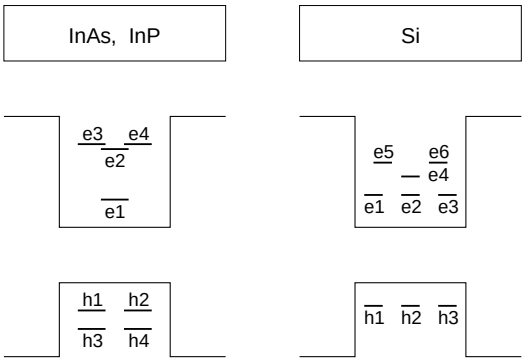

Fig. 1 – Schematic diagram of the near band-edge single-particle energy levels of InAs, InP, and Si spherical quantum dots. The valence band levels are denoted as h_1, h_2, etc. in order of decreasing energy starting from the valence band maximum, while the conduction band levels are denoted as e_1, e_2, etc. in order of increasing energy starting from the conduction band minimum.

degeneracy of the single-particle orbitals. Finally, the ground-state configuration for a given number N of electrons (or holes) is obtained by scanning di erent single configurations and diagonalizing the many-particle Hamiltonian (4) in search of the configuration with the lowest energy.

Our approximations are: i) The single-particle energies and wave functions are obtained by solving eq. (7) for a fixed, non–self-consistently screened potential. This approximation was examined in the case of an exciton [19] by comparing the electron-hole Coulomb integral obtained using non–self-consistent wave functions with the result of a self-consistent calculation. The di erence was less than 5%. ii) The single-configuration approach used to diagonalize the many-particle Hamiltonian of eq. (4) neglects correlation e ects due to configuration mixing. These e ects were examined in ref. [18], and found to be small in strongly confined quantum dots. iii) Surface-polarization e ects, which exist if the dielectric constant of the dot is different from that of the surrounding material, are neglected. This appoximation is valid when $_{\text{out}} \sim _{\text{in}}$.

We consider here InAs, InP and Si spherical quantum dots having the T_d point-group symmetry. The surface dangling bonds are fully passivated. The single-particle energy levels calculated using pseudopotentials [15–17] have the general pattern shown in fig. 1. Each energy level in fig. 1 is doubly degenerate because of time reversal symmetry, and can be occupied by two particles (either electrons or holes). In the case of InAs and InP dots the two degenerate single-particle levels at the top of the valence band (h_1 and h_2) originate from the bulk Γ_8 states, and have an s-like envelope function. The next two degenerate hole levels (h_3 and h_4) are also Γ_8 -derived, but have p-like envelope functions. The electron level at the bottom of the conduction band (e_1) originates from the bulk Γ_{6c} state and has an s-like envelope function. In InAs dots the next three electron levels (e_2, e_3, and e_4) derive from the bulk Γ_{6c} state and have a p-like envelope function [15] (e_2 is split from e_3 and e_4 because of spin=DDTDkfg6T“kromgS=D=TEE”xF=ΔDTxD:=F6FT96F8k(g9Mj“=F“F9xTΔC=EF6acouFfig,S=D=”6

holes are present in the quantum dot each of these two levels is occupied by one hole. In the diagonal approximation of eq. (6), the energy di erence between the Hund configuration $(h_1^1 h_2^1)$ and the non-Hund configuration $(h_1^2 h_2^0)$ is $\Delta E_2 = E_2(h_1^1 h_2^1) - E_2(h_1^2 h_2^0) = J_{h1,h2} - J_{h1,h1} - K_{h1,h2}$. For a 30.3 Å diameter InAs dot we find $\Delta E_2 = 166 - 172 - 6 = -12\,\mathrm{meV}$; for a 36.5 Å diameter InAs dot $\Delta E_2 = 137 - 141 - 4 = -8\,\mathrm{meV}$. Thus, only about half of the stabilization of the Hund configuration comes from interelectronic exchange. The other half comes from the reduction in the Coulomb repulsion achieved by placing the two holes in di erent spatial orbitals. We find only one case where Hund's rule is violated: When six electrons are loaded into a 36.5 Å diameter quantum dot, two of them occupy the e_3 level, while the e_4 level (degenerate with e_3) is empty. This is due to the fact that the coupling between the configurations $(e_1^2 e_2^2 e_3^2)$ and $(e_1^2 e_2^2 e_4^2)$, which are degenerate at the single-particle level, is su ciently strong to overcome the exchange and Coulomb energies gained by placing the electrons in the configuration $(e_1^2 e_2^2 e_3^1 e_4^1)$.

ii) *The Aufbau principle*: We find several instances where the Aufbau principle is not obeyed. For example, in InAs dots for $N_{\mathrm{holes}} = 3 \cdots 7$ the hole levels h_3 and h_4 are occupied

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This work was supported by the U.S. DOE, OER-SC, Division of Materials Science, under Grant No. DE-AC36-98-GO10337.

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