The Coulomb-interaction-induced breaking of the Aufbau principle for the hole charging of InGaAs/GaAs quantum dots

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Abstract

The so called ‘incomplete hole shell filling’ phenomenon, that is the breaking of the Aufbau principle was reported by Reuter et al (Phys. Rev. Lett. 94 026808) in the hole charging spectra of a quantum dot when the results were interpreted in the context of the s, p, d shell system—typical for electrons. We report an example of an inter-particle-interaction induced Aufbau principle violation even if it is applied to the one-particle Kohn–Luttinger eigenstates. We present a $\mathbf{k} \cdot \mathbf{p}$ configuration-interaction study that concerns multiple holes confined in an InGaAs/GaAs self-assembled cylindrical quantum dot. The eigenenergies and eigenvectors of up to six hole ground states were obtained—along with the corresponding one-hole orbital occupations—and discussed in the context of the Aufbau principle.

Keywords: quantum dots, valence band mixing, orbital occupation

(Some figures may appear in colour only in the online journal)

1. Introduction

Quantum dots are the structures of size of nanometres to micrometres that can confine charge carriers (conduction band electrons and/or valence band holes) in all three directions. This kind of confinement leads to the energy quantization and gives a discrete spectrum of energy levels. This is an analog of the discrete spectrum of natural atoms and thus the quantum dots are often called artificial atoms [1, 2]. Quantum dots of an especially small size are made by applying an electrostatic potential (see [3–5]) and by the self-organization (see [6, 7]). In these very small structures that confine the conduction band electrons, the single-particle energy levels separation is so large that the observation of the occupation of the individual orbitals in many-electron states was possible. The sequence of the occupation for electrons in small quantum dots is generally governed by the Aufbau principle and the Hund’s rule outside the energy-level crossings induced by an external magnetic field [3, 4, 6–8].

In [9] the hole charging spectra of self-assembled InAs quantum dots in perpendicular magnetic fields were studied by the capacitance-voltage spectroscopy. The authors of that work interpreted the results in the terms of the typical results obtained for electrons i.e. the s, p and d shell system for the envelope functions. From the magnetic-field dependence of the individual peaks it is concluded that the s-like ground state is completely filled with two holes but that the fourfold degenerate p shell is only half filled with two holes before the filling of the d shell starts. This so called ‘incomplete shell filling’ is attributed by them to a large influence of the Coulomb interaction in this system.

Climente et al [10] suggested that by using a model that takes into account the valence band mixing via the Kohn–Luttinger (KL) Hamiltonian the behaviour of the system can be interpreted as abiding the Aufbau principle in the context of the one-particle KL shells instead of the electron-type ones. However, we report an example of an inter-particle-interaction induced Aufbau principle violation even if it is applied to the one-particle KL eigenstates.

In this work we present a $\mathbf{k} \cdot \mathbf{p}$ configuration-interaction study of multiple-hole ground states of an InGaAs/GaAs self-assembled cylindrical quantum dot. The model that was used
allows for the valence band mixing via the 6-band Luttinger-Kohn Hamiltonian. The dot is embedded in an external magnetic field applied in the growth direction (i.e. along the symmetry axis). The lattice constant mismatch-induced strain is taken into account by the Pikus–Bir Hamiltonian.

The ground state energies and eigenstates of up to six holes confined in the system were obtained for two dot sizes: the strong and weak confinement cases. The occupation of the one-particle orbitals for those wavefunctions was also calculated. We present a few cases when the Aufbau principle is evidently broken due to the Coulomb interaction and a relatively small difference in energy of the related one-particle levels (a ‘strong’ violation). We also describe the so called ‘weak’ violations of the Aufbau principle. They consist of a shift of the value of magnetic field for which a change in the orbital character of a multi-hole state takes place and the value for which a corresponding level crossing occurs in the single hole spectrum.

2. Theory

2.1. KL Hamiltonian

We work within the envelope ansatz using the 6-band axial Luttinger (KL) Hamiltonian. It is written in the Bloch basis of

\[
\begin{bmatrix}
J^B_{\text{R}} = \frac{3}{2}, J_z^B = \frac{3}{2} \\
J^B_{\text{R}} = \frac{3}{2}, J_z^B = -\frac{1}{2} \\
J^B_{\text{R}} = \frac{1}{2}, J_z^B = \frac{1}{2}
\end{bmatrix},
\]

where \( J^B_{\text{R}} \) is the total angular momentum of the Bloch function and \( J_z^B \) is its component along the symmetry (z) axis. \( J^B_{\text{R}} = \frac{3}{2} \) corresponds to the heavy hole bands, \( J^B_{\text{R}} = \frac{3}{2} \) and \( J_z^B = -\frac{1}{2} \) corresponds to the light hole components and \( J^B_{\text{R}} = \frac{1}{2} \) corresponds to the spin–orbit split-off bands. The Hamiltonian has the form of:

\[
\hat{H}_{\text{KL}} = \begin{bmatrix}
-\hat{S} & \hat{R} & 0 & -\hat{S} & \sqrt{3}\hat{R} \\
-\hat{S} & 0 & \hat{R} & -\sqrt{3}\hat{Q} & \sqrt{3}\hat{S} \\
\hat{R} & 0 & \hat{S} & \sqrt{3}\hat{Q} & \sqrt{3}\hat{S} \\
0 & \hat{R} & \hat{S} & -\sqrt{3}\hat{R} & -\hat{S} \\
-\hat{S} & -\sqrt{3}\hat{Q} & \sqrt{3}\hat{S} & -\sqrt{3}\hat{R} & \hat{R}, m_{\text{z}} + \Delta_{\text{SO}} \\
\end{bmatrix}
\]

where \( \hat{R} = -\frac{\sqrt{3}}{2} \gamma_2 \rho_2 - \frac{\sqrt{3}}{2} \gamma_3 \rho_3, \hat{S} = \sqrt{3} \gamma_3 \rho_3, \hat{Q} = -\frac{\sqrt{3}}{2} (\rho_1^2 - \rho_2^2) \), \( \hat{R} = \hat{R}_z - i \hat{P}_z, \hat{Q} = \sqrt{-1} \) and the band Hamiltonians are given by:

\[
\hat{T}_{J^B_{\text{R}} = \frac{3}{2}, J_z^B = \frac{3}{2}} = -\frac{1}{2} [(\gamma_1 + \gamma_2) \rho_2^2 + (\gamma_1 - 2\gamma_2) \rho_2^2] + V_{\text{pot}}(\hat{r}),
\]

\[
\hat{T}_{J^B_{\text{R}} = \frac{3}{2}, J_z^B = -\frac{1}{2}} = -\frac{1}{2} [(\gamma_1 - \gamma_2) \rho_2^2 + (\gamma_1 + 2\gamma_2) \rho_2^2] + V_{\text{pot}}(\hat{r}),
\]

\[
\hat{T}_{m_{\text{z}} = \frac{1}{2}} = -\frac{1}{2} (\rho_1^2 + \rho_2^2) + V_{\text{pot}}(\hat{r}),
\]

where the in-plane envelope momentum operator is: \( \hat{p}_z = (\hat{p}_x, \hat{p}_y, 0) \) [11]. \( \gamma_1, \gamma_2 \) and \( \gamma_3 \) are the Luttinger parameters—the values for the dot material are adopted for the whole system for simplicity.

2.2. Basic informations on the system

We consider a system of up to six holes confined in a cylindrical quantum dot (made of InGaAs/GaAs) in the presence of an external magnetic field. The z-axis is the direction of the growth and the symmetry axis. Two sizes of the dot are considered: the smaller one corresponds to a strong confinement of particles, while the bigger one—to a weak confinement. In the former case the radius of the dot \( R_{\text{dot}} \) is assumed to be 10 nm and the height of the dot \( 2Z_{\text{dot}} \) is 2 nm. The values for the latter case are: \( R_{\text{dot}} = 20 \) nm and \( 2Z_{\text{dot}} = 6 \) nm, respectively.

The band parameters are taken from [12] and correspond to Ga_{1-x}In_{x}As for \( x = 0.53 \). The Luttinger parameters for InGaAs are taken as: \( \gamma_1 = 11.01, \gamma_2 = 4.18, \gamma_3 = 4.84 \). The spin–orbit splitting is assumed to be \( \Delta_{\text{SO}} = 329.6 \) meV. The valence band offset of dot material in respect to barrier material is equal to \( V_0 = -206 \) meV. The deformation potential of the dot material \( a_{\text{GaInAs}}^{\text{GaAs}} = 679 \) meV and for the barrier material \( a_{\text{GaAs}}^{\text{GaInAs}} = 700 \) meV, and the deformation potential \( b_{\text{GaInAs}}^{\text{GaAs}} = -1894 \) meV while for the barrier material \( b_{\text{GaAs}}^{\text{GaInAs}} = -2000 \) meV. The interpolation between the values for InAs and GaAs takes into account bowing parameters, where appropriate (see [12]).

The confinement potential arises from the difference between the energy of the top of the valence band in both materials. We set the potential energy outside of the dot to be equal to zero. In our model the confinement potential is given by a following function:

\[
V_{\text{pot}}(\hat{r}) = V_{\text{pot}}(\rho, \phi, z) = \begin{cases} V_0, & \rho < R_{\text{dot}} \land |z| < Z_{\text{dot}} \\
0, & \rho > R_{\text{dot}} \lor |z| > Z_{\text{dot}} \end{cases}
\]

The external magnetic field is applied in the growth direction: \( \hat{B} = (0, 0, B_z) \).

3 Footnote 1.

4 This approach was used in numerous computational works that concern the systems of a single and multiple holes in quantum dots (see e.g. [15, 17, 19, 21–23]), including the two works that constitute the most relevant context for our study, i.e. [10, 18]. The values of the Luttinger parameters of the holes in the barrier material are smaller than the ones for the dot material, which is equivalent to the larger barrier effective masses. This means that the holes are more likely to penetrate the barrier material in our study that in reality.

5 Please note that the rotational in-plane symmetry of the dot plays an important role in our model. This, study is limited to the disk-like dots, with shapes similar to the shapes of the dots described in [10, 16–19, 21, 22, 30, 31]. The pyramidal quantum dots (see e.g. [32]) are outside the scope of this work.

6 Unless noted otherwise all formulae are written in atomic units.
2.3. Strain effects

The difference between the lattice constants of the dot material and the barrier material ($\epsilon_0 = \frac{\text{GaAs}}{\text{Si}} - 1 = 3.8\%$) is the source of the stress and strain in the system. We use the elastic model of [13] to calculate the strain field. The relative strain is given by:

$$\epsilon_{ij}^{rel}(\mathbf{r}) = -\frac{\epsilon_0}{4\pi(1 - \nu)} \int \frac{(\mathbf{r}' - \mathbf{r}) dS_i}{|\mathbf{r}' - \mathbf{r}|^3},$$

where the Poisson’s ratio $\nu = \frac{1}{3}$ for $i, j \in \{x, y, z\} \otimes \{x, y, z\}$ and the integration is conducted over the surface of the dot. The hydrostatic strain is given by $\epsilon_{ii} = \epsilon_0$ inside the dot and is equal to zero outside.

The effect of this strain on the energy of the system is given by the Pikus–Bir Hamiltonian [11]. However, we restrict ourselves to the biaxial strain only (as in [16]) by assuming $\epsilon_{xx} = \epsilon_{yy} = \epsilon_{zz}$ and $\epsilon_{xy} = \epsilon_{yz} = \epsilon_{zx} = 0$. Written in the Bloch basis of equation (1) the biaxial Pikus–Bir Hamiltonian has a form of:

$$\hat{H}_{\text{PB}} = \begin{pmatrix}
\hat{P} + \hat{Q}_r & 0 & 0 & 0 & 0 & 0 \\
0 & \hat{P} - \hat{Q}_r & 0 & 0 & -\sqrt{2}\hat{Q}_r & 0 \\
0 & 0 & \hat{P} - \hat{Q}_r & 0 & 0 & \sqrt{2}\hat{Q}_r \\
0 & 0 & 0 & \hat{P} + \hat{Q}_r & 0 & 0 \\
0 & 0 & 0 & 0 & \sqrt{2}\hat{Q}_r & 0 \\
0 & 0 & 0 & 0 & 0 & \hat{P}
\end{pmatrix},$$

(6)

where $\hat{P} = -a_i(\epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz})$ and $\hat{Q}_r = \frac{b}{2}(\epsilon_{xx} + \epsilon_{yy} - 2\epsilon_{zz})$.

2.4. Magnetic field

There are several propositions how to include an external magnetic field $B_z$ into the KL model. The impact of the field on the envelope functions can be taken into account by substituting the canonical momentum $\hat{p} = -i\nabla$ by $\hat{p} = -i\nabla - \hat{A}$ in equation (3) like e.g. in [18]. Planelles and Jaskólski suggested reversing the order of the operations: to start with considering magnetic field in the context of each band separately and then subsequently to apply the envelope approximation in the $\hat{k} \cdot \hat{p}$ procedure [15]. This approach leads to different magnetic terms in the KL Hamiltonian. Specifically that all the quadratic terms are diagonal and—in the 4-band model—also the linear terms are. A further work by Climente et al compared the both approaches in the scope of the 4-band KL study of a hole in InAs/GaAs quantum molecules [16]. The authors concluded that the second method gives results that are in agreement with experiment while the first one artificially enhances the HH-LH mixing (by means of the off-diagonal terms) which leads to the not observed bonding-antibonding ground state magnetic switching. Work [16] also refined the second model by putting the relevant effective masses for the direction perpendicular to the growth axis into the magnetic terms instead of the effective masses in direction of that axis. After this change the model correctly retrieves the single-band limit in the case of band-decoupling. Finally, the model was reshaped once more in [17] by including the spin degree of freedom and by defining the Zeeman terms for the Bloch functions via the hole $g$-factors, independently of the relevant Zeeman envelope-dependent ones. The latter work predicts a quadratical increase of the excitonic gap with increasing magnetic field, as observed in photoluminescence experiments of InGaAs QDs.

In our work we follow [17] in including the magnetic field $B_z$ into the model as:

$$\hat{H}_{\text{PB}} = (\gamma_1 + \gamma_2) \left( \frac{B_z^2}{8} + \frac{B_z J_{\text{env}}^{\text{eff}}}{2} \right) + \frac{3}{2} \kappa \mu B_z,$$

$$\hat{H}_{\text{PB}} = (\gamma_1 - \gamma_2) \left( \frac{B_z^2}{8} + \frac{B_z J_{\text{env}}^{\text{eff}}}{2} \right) + \frac{1}{2} \kappa \mu B_z,$$

$$\hat{H}_{\text{PB}} = (\gamma_1 - \gamma_2) \left( \frac{B_z^2}{8} + \frac{B_z J_{\text{env}}^{\text{eff}}}{2} \right) - \frac{1}{2} \kappa \mu B_z,$$

$$\hat{H}_{\text{PB}} = (\gamma_1 + \gamma_2) \left( \frac{B_z^2}{8} + \frac{B_z J_{\text{env}}^{\text{eff}}}{2} \right) - \frac{3}{2} \kappa \mu B_z,$$

$$\hat{H}_{\text{PB}} = \gamma_1 \left( \frac{B_z^2}{8} + \frac{B_z J_{\text{env}}^{\text{eff}}}{2} \right) + \frac{1}{2} \kappa \mu B_z,$$

$$\hat{H}_{\text{PB}} = \gamma_1 \left( \frac{B_z^2}{8} + \frac{B_z J_{\text{env}}^{\text{eff}}}{2} \right) - \frac{1}{2} \kappa \mu B_z,$$

(7)

where $J_{\text{env}}$ is the $z$-component of the total angular momentum of envelope, $\mu_B$ is the Bohr magneton and $\kappa = \frac{4}{3}$, $\kappa' = \frac{2}{3}$ are the effective hole $g$-factors.

2.5. Computation process

The first step in the computation process is to solve the eigenproblem of the $\hat{H}_{\text{KL}} + \hat{H}_{\text{PB}}$ for a single hole. The variational basis for the envelope contains the functions of the following kind:

$$\psi_{k, J_{\text{env}}}^{\text{eff}}(\mathbf{r}) = \frac{\exp(i J_{\text{env}}^{\text{eff}} \phi)}{\sqrt{2\pi}} \beta_{k, J_{\text{env}}}^{\text{eff}} \left( \frac{\theta_{K_{L_{\text{env}}}}^{\text{eff}}}{R_{\text{ef}}} \right) \zeta_n \left( \frac{z}{Z_{\text{ef}}} \right),$$

(8)

where $\beta_{k, J_{\text{env}}}^{\text{eff}}$ is a relevant normalized Bessel $J$ function ($K_{L_{\text{env}}}$ is its $k$th zero) for $\rho < R_{\text{ef}}$ and zero otherwise. The function $\zeta_n$ has the form of:

$$\zeta_n(y) = \frac{1}{\sqrt{Z_{\text{ef}}}} \cos \left( \frac{\pi}{2} [n(y - 1) + 1 + (n - 1)(n \text{ mod } 2)] \right),$$

(9)

for $|z| < Z_{\text{ef}}$ and zero otherwise. The $R_{\text{ef}}$ is an effective radius of the wavefunction, that is assumed to be not less than the radius of the dot confinement potential: $R_{\text{ef}} > R_{\text{dot}}$. It is given

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Note that the sign of $a_i$ and $b_i$ values and/or of $\hat{P}, \hat{Q}_r$ is differently defined in various sources. The orientation of the relative signs should be that—in the case of biaxial strain—the confinement potential is deepened in the case of the heavy holes and shallowed for the light holes (see [14, 16, 30]).
by \( R_{\text{ef}} = \frac{R_{\text{oh}}}{b_R} \), where \( b_R \) is a variational parameter. Similarly, the \( Z_{\text{ef}} \geq Z_{\text{dot}} \) is the effective half-height of the dot, and:

\[
Z_{\text{ef}} = \frac{Z_{\text{dot}}}{a_{\text{HH}}} \text{ for the heavy-hole bands and } Z_{\text{ef}} = \frac{Z_{\text{dot}}}{a_{\text{LHSO}}} \text{ for the light-hole and the split-off bands, where } a_{\text{HH}} \text{ and } a_{\text{LHSO}} \text{ are variational parameters.}
\]

The simplest method to describe the wavefunction of a hole in a quantum dot (which is also true in the case of an electron) is to employ the infinite barrier scenario. This approach was used e.g. in [18], where the authors considered a pair of quantum holes (described by the 4-band KL Hamiltonian) in a cylindrical quantum dot with the Coulomb interaction in a magnetic field. If the barrier outside a cylindrical quantum dot is infinite, then the wavefunction is completely localized inside the dot. In this case the eigenfunctions of the individual bands are represented by the sine/cosine functions in the \( z \) (growth) direction and the Bessel functions in the \( x,y \) plane. In [18] it is mentioned that it is possible to consider an effective thickness of the dot—i.e. the cut-off point of the sine/cosine functions—that is not identical to the actual ‘physical’ thickness. On the other hand the work [19] describes a hole in a system of double quantum dots. The authors of that work also adopt the infinite barrier for the \( x,y \) plane. In the growth direction, the specific finite value of the band-offset potential is taken into account. They also use the sine/cosine basis functions for the individual bands but the cut-off point in this case is the boundary of the computation box. The size of the box is not specified but it is probably many times greater than the thickness of the dot. Please note that this approach requires many sine/cosine basis functions (as much as 46 in the case of work [19]) to properly describe the wavefunctions. Due to the many-body character of this work and its great computational complexity the maximal number of the sine/cosine functions was limited in our case. Moreover, using a ‘big box’ scheme is resource-demanding for each computed element per se as it involves the integration and/or differential equations solving in many dimensions over a very large volume. Hence it was not possible for us to employ the mentioned method. However we did not want to restrict our model to the infinite barrier case. So we developed an intermediate method that allows the wavefunction to penetrate the barrier. As the degree of the hole tunnelling outside the dot cannot be known a priori then the effective thickness is a variational parameter. We also decided that we will employ the same procedure for the radius of the dot. While this method does not model the decaying tails of the wave functions it can model the confining potential/kinetic energy/interband mixing interplay that is responsible for what the optimal scope of barrier penetration is.

The \( z \)-component of the total angular momentum is defined for a KL eigenfunction and is equal to the sum of the total angular momentum of the Bloch state and the angular momentum of the envelope \( z \)-components: \( J_z = J_z^{\text{ev}} + J_z^{\text{bl}} \) (i.e. the axial approximation of KL Hamiltonian commutes with the operator of the total angular momentum of the hole). This enables a very significant simplification of the variational basis. The computation can be done for each \( J_z \) separately and only one value of \( J_z^{\text{ev}} = J_z - J_z^{\text{bl}} \) is used for each component of KL state vector in each of these calculations. This fact was taken advantage of in the case of the dots with an axial symmetry in numerous works (see e.g. [10, 18–23], among many others).

We considered \( J_z \) values from the range \( \{\frac{1}{2}, \frac{3}{2}, \frac{5}{2}, ...\} \). The \( k \) and \( n \) quantum numbers put in order the basis functions in \( \rho \) and \( z \) directions, respectively. The set of values that was used in calculation is \( k \in \{1,8\} \) and \( z \in \{1,6\} \).

At this stage \( \{a_{\text{HH}}, a_{\text{LHSO}}, b_R\} \) constitutes the set of the variational parameters. Each element of this set can in principle have any value in the range of \( (0,1] \). We conducted calculations for each case when a parameter is equal to one of the values: \( \frac{1}{2} \text{ef dot} \) for \( j \in \{1,19\} \) (j-s of parameters are independent). For the case of the strong confinement the lowest KL ground-state energy was obtained in the case of the following set of values: \( \{a_{\text{HH}} = 0.25, a_{\text{LHSO}} = 0.40, b_R = 0.55\} \), what is equivalent to: \( R_{\text{ef}} = 1.28R_{\text{dot}}, Z_{\text{ef}} = 3R_{\text{dot}} \) for the heavy hole band and \( Z_{\text{ef}} = 2.5Z_{\text{dot}} \) for the light hole and the split-off bands. By an analogous procedure for the bigger dot the following values were obtained: \( \{a_{\text{HH}} = 0.40, a_{\text{LHSO}} = 0.55, b_R = 0.75\} \) and consequently \( R_{\text{ef}} = 1.33R_{\text{dot}}, Z_{\text{ef}} = 2.5Z_{\text{dot}}/1.82Z_{\text{dot}} \) for the heavy holes / the light and split-off holes, respectively.

The matrix elements of the off-diagonal operators \( \hat{R}, \hat{S} \) of equation (2) are calculated analytically where possible and otherwise by the Legendre–Gauss quadratures with 1000 points. All other matrix elements of this Hamiltonian can be obtained analytically. The calculation of the matrix elements of the Pikus–Bir Hamiltonian (equation (6)) was conducted by the rectangle integration on a mesh with \( d\xi = 0.1 \text{ nm} \) and \( d\rho = 0.2 \text{ nm} \) in the case of the smaller dot, and two times bigger spacings for the weak confinement system. In order to obtain the relevant strain tensor elements we used the same method on a mesh with \( d\phi = \frac{\pi}{1024} \).

The resulting eigenvectors of the envelope have the form of:

\[
\phi_{E_{\text{m},a}}(\vec{r}) = \begin{pmatrix}
\xi_{m}^{n_{1}}(\rho, z)e^{i(k_{9}z+\frac{\pi}{2})} \\
\xi_{m}^{n_{2}}(\rho, z)e^{i(k_{3}z+\frac{\pi}{2})} \\
\xi_{m}^{n_{3}}(\rho, z)e^{i(k_{1}z+\frac{\pi}{2})} \\
\xi_{m}^{n_{4}}(\rho, z)e^{i(k_{2}z+\frac{\pi}{2})}
\end{pmatrix} \frac{1}{2}h_{L}^{\text{so}} \frac{3}{2}(\rho, z)_{\text{so}}^{-\frac{3}{2}}(\rho, z)_{\text{so}}^{-\frac{3}{2}}.
\]

with the definite total angular momentum of \( J_z \), where \( \xi_{m}^{n_{i}} \) is the one-band wavefunction component and \( m \in \{1,2,3,...\} \) numerates states in the order of increasing energy for a given \( J_z \). All pairs of the kind: \( \phi_{E_{\text{m}},a} \) and \( \phi_{E_{\text{m}},a} \) are degenerate for all \( J_z \) and \( m \) when no external magnetic field \( B_{z} \) is applied to the system. These eigenfunctions are used for the construction of the basis states for the many-hole calculation.

In the special case of one hole it is easy to include the magnetic field in the computation directly. The eigenstates of \( \hat{H}_{\text{KL}} + \hat{H}_{\text{PB}} + \hat{H}_{B} \) are obtained in the same basis as described before. The matrix elements of equation (7) are calculated analytically when possible and by the Legendre–Gauss.
quadratures with 1000 points otherwise (some cases of ‘diagramatic’ components). Note that adding the magnetic field in the \( z \) direction does not affect the axial symmetry of the system and \( J = 1 \) is defined. As a consequence the eigenstates of \( \hat{H}_{KL} + \hat{H}_{PA} + \hat{H}_{B} \) can also be written in the form of equation (10).

In the case of the many hole states the variational basis is constructed from the KL eigenfunctions obtained for the case of no magnetic field. After that the full many-body Hamiltonian (that is the single-hole energies plus the magnetic and Coulomb terms) is diagonalized in this basis. The total angular momentum \( z \)-component of a many-hole state is defined:

\[
J_{N}^{z} = \sum_{i=1}^{N} J_{i}^{z},
\]

where the index \( i_{p} \) runs over all particles and \( N_{p} \) is the number of particles in a given case. Thus the construction of the basis for each \( J_{N}^{z} \) (and the diagonalization following it) can be done separately.

There are two stages of the basis construction process. \( N_{\text{first}} \) parameter is the number of basis states that are taken into account in the first stage and \( N_{\text{second}} \) parameter is the number of basis states that are taken into account in the second stage. At the beginning we find all the sets of particular \( J \)-s that sum to \( J_{N}^{z} \). For each sequence \((J_{1}, \ldots, J_{N_{p}})\) and for each particle we take into account \( m = 1, \ldots, N_{\text{first}} \) one-particle eigenfunctions and form the sequences \((\phi_{J_{1},m_{1}}, \ldots, \phi_{J_{N_{p}},m_{N_{p}}})\). Multiple equivalent sequences are reduced to a single instance.\(^7\)

Secondly, we sort all the latter-type sequences by the sum of the respective one-particle energies. Then we take \( N_{\text{second}} \) sequences of the lowest sums and construct the relevant Slater determinants:

\[
\Psi_{J_{N}^{z},M_{j_{N}}} = \hat{A}(\phi_{J_{1},m_{1}}, \ldots, \phi_{J_{N_{p}},m_{N_{p}}}),
\]

where \( \hat{A} \) is the normalized antisymmetrization operator and \( M_{j_{N}} \) numerates the functions by the sum of the one-particle energies. The set of all such Slater determinants is the variational basis for a given \( J_{N}^{z} \).

### 2.6. Coulomb interaction

The Hamiltonian that describes the Coulomb interaction between the particles \( i \) and \( j \) is \( \hat{H}_{\text{int}}^{ij} = \frac{1}{|r_{i} - r_{j}|} \) and the total interaction Hamiltonian is given by: \( \hat{H}_{\text{int}} = \sum_{i>j} \hat{H}_{\text{int}}^{ij} \). We adopted the electric permittivity constant in the whole system equal to the value for dot material: \( \varepsilon = \varepsilon_{e} \varepsilon_{0} = 13.9 \varepsilon_{0} \). The Coulomb matrix element of any two Slater determinants can be expressed as the sum of matrix elements between the products of a pair of the respective wavefunctions multiplied by the relevant coefficients:

\[
\langle \Psi_{J_{N},M_{j}} | \hat{H}_{\text{int}} | \Psi_{J_{N},M_{j}} \rangle = \sum_{i>j} C_{ij} I_{ij}^{B} = \sum_{i>j} C_{ij} \langle \phi_{J_{i}^{1},m_{1}}^{1} \phi_{J_{i}^{2},m_{2}}^{2} | \hat{H}_{\text{int}} | \phi_{J_{i}^{1},m_{1}}^{1} \phi_{J_{i}^{2},m_{2}}^{2} \rangle,
\]

where the value of the coefficient \( C_{ij} \) can be one of \( \{-1, 0, 1\} \), depending on the parity of the relevant antisymmetrization permutations and the values of the relevant Kronecker deltas. The \( A \) and \( B \) indices belong to the set that contains all the considered \( M_{j_{N}} \) for a given \( J_{N}^{z} \). The matrix elements of the right hand side may have non-zero values only if \( |J_{1}^{1} + J_{1}^{2}| = |J_{i}^{1} + J_{i}^{2} \rangle = 0 \) and could be calculated directly, by a six-dimensional integration. However, it is much advantageous to translate the issue in question to a three dimensional integration over an effective potential:

\[
I_{ij}^{A,B} = \int \phi_{J_{i}^{1},m_{1}}^{1} \phi_{J_{i}^{2},m_{2}}^{2} \phi_{J_{i}^{1},m_{3}}^{3} \phi_{J_{i}^{2},m_{4}}^{3} V_{i}^{A,B}(f) df_{0}^{3}.
\]

This potential is obtained by solving the Poisson equation with the complex right hand side [24]:

\[
\nabla^{2} V_{i}^{A,B}(f) = -\frac{4\pi}{\varepsilon_{R}} \phi_{J_{i}^{1},m_{1}}^{1} \phi_{J_{i}^{2},m_{2}}^{2} \phi_{J_{i}^{1},m_{3}}^{3} \phi_{J_{i}^{2},m_{4}}^{3}(f).
\]

In the latter task we employ the multigrid approach and the overrelaxation with the relaxation parameter \( \omega = 1.9 \) and \( dz = \Delta \rho = 0.1 \) nm on the final mesh. For a more detailed description of this computation scheme see e.g. our earlier work [25]. The number of iterations used in the process is adaptive, ensuring a sufficiently small value of inaccuracy parameter:

\[
\int |\nabla^{2} V_{i}^{A,B}(f) + \frac{4\pi}{\varepsilon_{R}} \phi_{J_{i}^{1},m_{1}}^{1} \phi_{J_{i}^{2},m_{2}}^{2} \phi_{J_{i}^{1},m_{3}}^{3} \phi_{J_{i}^{2},m_{4}}^{3}(f) | df_{0}^{3}
\]

after the last iteration.

After the diagonalization of the total Hamiltonian of the system (i.e. kinetic + magnetic + Coulomb), the multi-hole levels are described in the form of \( (J_{N}^{z}, m) \), where \( m \) numerates the levels of a given \( J_{N}^{z} \) in the order of increasing energy.

### 3. Results

#### 3.1. Strong confinement

In this section the results for the weak confinement case i.e. the system with \( R_{\text{dot}} = 10 \) nm and \( 2Z_{\text{dot}} = 2 \) nm are presented.

#### 3.1.1. Single hole

The energy spectrum of a single hole in an external magnetic field \( B \), is presented in figure 1(a). The profile of the effective confinement potential including the strain along the growth axis is shown in figure 1(b). The inset shows the potential profile for \( \varepsilon = 0 \) i.e. in the centre...
of the dot in the growth direction as a function of the radius in the x-y plane. As noted before, the strain essentially reinforces the confinement for the heavy holes (322 meV instead of 206 meV without the strain) and essentially weakens it for the light holes (38 meV in the middle of the dot instead of 206 meV without the strain) while the potential for the split-off bands remains nearly unaffected (180 meV). As one can see in figure 1(a)—for a low magnetic field—the six energy levels of the lowest energy are (in order) the states with: $(J_z = -\frac{3}{2}, m = 1)$, $(J_z = \frac{3}{2}, m = 1)$, $(J_z = -\frac{1}{2}, m = 1)$, $(J_z = \frac{1}{2}, m = 1)$, $(J_z = -\frac{5}{2}, m = 1)$, $(J_z = \frac{5}{2}, m = 1)$. The above sequence changes in the scope of the crossing that takes place at $B_0$ and for stronger magnetic fields the $(J_z = -\frac{5}{2}, m = 1)$ is the fourth level and $(J_z = \frac{1}{2}, m = 1)$ is the fifth level in the order of increasing energy.

When no magnetic field is present in the system then the two corresponding levels with $+J_z$ and $-J_z$ are Kramers degenerate. In this case the energy separation between the ground state energy level $(|J_z| = \frac{3}{2})$ and the states with $|J_z| = \frac{1}{2}$ is relatively large and equal to 25.5 meV. On the other hand the energy separation between the energy level corresponding to $|J_z| = \frac{1}{2}$ and the states with $|J_z| = \frac{5}{2}$ is 1.5 meV which is a small value. Also, the mentioned six levels are quite far from all other states in the energy scale—the next level (which is $J_z = -\frac{1}{2}, m = 2$) is 22 meV higher. These facts bear essential consequences to the multi-particle spectra that will be discussed in a later part of our work.

If a weak magnetic field is introduced into the system then the response of the energy levels is mainly linear, as the diamagnetic term is significantly smaller than the orbital/spin Zeeman ones. For all six levels presented in figure 1(a) the heavy hole band of the smallest envelope angular momentum is strongly dominating. For the states with positive $J_z$, it is the band with $J_{\text{env}}^{B1} = \frac{3}{2}$ and for the states with negative $J_z$ it is the band with $J_{\text{env}}^{B1} = \frac{3}{2}$. The corresponding spin Zeeman terms are: $\frac{3}{2} \mu_B B_z$ and $-\frac{3}{2} \mu_B B_z$, which is the reason why the state with negative $J_z$ is lower in energy in each $|J_z|$ pair. Moreover, the absolute value of the spin Zeeman terms of the six states are all nearly the same. However, the envelope angular momentum of the dominating band $J_{\text{env}}^{B1} = J_z - J_{\text{env}}^{B1}$ is different for each state, as presented in table 1. In the case of states with $|J_z| = \frac{3}{2}$ the dominating $J_{\text{env}}^{B1}$ is zero and the corresponding orbital Zeeman term is also zero. For the states with $|J_z| = \frac{5}{2}$ the dominating $J_{\text{env}}^{B1}$ has the sign opposite to the $J_{\text{env}}^{B1}$ so the orbital Zeeman term partially suppress the larger spin one. Conversely, the states with $|J_z| = \frac{5}{2}$ have their respective dominating $J_{\text{env}}^{B1}$ sign parallel to the $J_{\text{env}}^{B1}$ and these two Zeeman terms add together. As a result the splitting between the energy levels in a $|J_z|$ pair is the smallest in the case of the $|J_z| = \frac{5}{2}$, the medium one for the $|J_z| = \frac{3}{2}$ and the largest in the $|J_z| = \frac{3}{2}$ case—see the values in table 1.

In systems of this kind the occupation of the single-particle orbitals in the multi-electron wavefunctions obey the Aufbau principle. This principle states that particles occupy the one-particle orbitals in the order of the increasing energy. If our hole system was governed by this rule, the ground state of the two-hole case would be well described by assuming that the ground state and the first excited state of figure 1(a) are occupied. In the case of three holes, it would mean that additionally the orbital of the second excited state is occupied. In the case of such a system we can infer the total angular momentum of a multi-hole ground state from the one-particle spectrum as:

$$J_z^B = \sum_i J_i,$$

where $(J_i)$ is the total angular momentum of the one hole state that is the $i$th lowest in energy. For our system we would have: $J_{z^B}^0=0$, $J_{z^B}^1=\pm\frac{3}{2}$, $J_{z^B}^2=0$, $J_{z^B}^3=\pm\frac{5}{2}$ and $J_{z^B}^4=0$. The Aufbau principle is trivially fulfilled when no particle-particle interaction is present.

3.1.2. Energy spectra of multiple holes. The energy spectra of the two-hole to six-hole strongly confined systems are presented in figure 2. As we are primarily interested in the identification of the ground state of the system, we only include the states with $m = 1$ for each $J_{z^B}^N$, that is the lowest-energy level in each sub-space defined by the multi-hole total angular momentum$^8$. The variational parameters of each computation are presented in table 2.

If two holes are confined in the dot, the state with $J_{z^B}^2=0$ is the ground state as shown in figure 2(a). There is not any state with a different $J_{z^B}^2$ within a close vicinity of this state on the energy scale. The level that is next in order is the one corresponding to $J_{z^B}^2=\pm\frac{3}{2}$, separated by 18.6 meV for $B_z = 0$. In the case of three holes, the levels that correspond to the states with $|J_{z^B}^3|=\frac{3}{2}$ and $|J_{z^B}^3|=\frac{5}{2}$ are close in terms of energy (see figure 2(b))—the difference being equal to 0.9 meV in the absence of a magnetic field. For $B_z < 7.2$ T the $J_{z^B}^3=\pm\frac{3}{2}$ state is the ground state but as the intensity of the field rises, the $J_{z^B}^3=\pm\frac{3}{2}$ level approaches the $J_{z^B}^3=\pm\frac{5}{2}$ one and eventually above 7.2 T it becomes the ground state. When one more hole is added to the quantum dot ($N_p = 4$; figure 2(c)) then for $B_z = 0$ the $J_{z^B}^4=\pm\frac{3}{2}$ and $J_{z^B}^4=\pm\frac{5}{2}$ states have the same energy. In the presence of a non-zero magnetic field this degeneracy is lifted and the

$^8$ The omission of the states with $m > 1$ causes strictly zero error in the process of identification of the ground state. The states with a given $J_{z^B}^N$ are ordered by their energy with the quantum number $m$. Hence no state with $m > 1$ can be the ground state at any point, only the $J_{z^B}^N$ of the ground state can change. Please note that it is still possible that the character of the ground state changes without any change to its quantum numbers. This is the case when the two levels with the same $J_{z^B}^N$ and with $m = 1$ and $m = 2$, respectively, have a crossing. In all cases where it does happen the relevant $m > 1$ states are included in the figures and described in the text—see the results for the weak coupling.
and one. If one
ones. A
state is the ground state. When the dot is charged
\( J_z = \pm \frac{1}{2} \) levels are relatively close to the \( |J_z = \mp \frac{5}{2}\) \) ones, with their
energy only higher by 1.2 meV. Although the eigenenergy of the
\( J_z = -\frac{11}{2} \) and \( J_z = -\frac{7}{2} \) states strongly decreases with the increasing magnetic field and the \( J_z = -\frac{5}{2} \) level
energy increases, the latter one has the lowest energy in the
spectrum for as much as 10 T. For six-holes the \( J_z = 0 \)
level energy is by far the lowest (18.3 meV) in the low
magnetic field regime. Although many other levels approach it with
the increasing magnetic field, it remains the ground state in
the whole presented range of the magnetic spectrum—as shown in figure 2(e).

To sum up, the actual sequence of the \( J_z \) values of the
ground states for \( N_P \in \{2,...,6\} \) is \( \left(0, -\frac{1}{2}, -3, -\frac{5}{2}, 0\right) \). If one
compares this sequence to the one that was deduced using the
Aufbau principle from the single-hole calculations, it may be
noticed that the principle correctly predicts \( J_z \) for all cases
apart from the four-hole one.

Let us consider how the magnitudes of energy separations between the respective single-hole states influence the behaviour of a multiple-hole system. Firstly, the energy separation between the \( J_z = \frac{3}{2} \) and \( J_z = -\frac{1}{2} \) levels is large (see figure 1(a)). Hence it is easy to predict that the dot charged with two holes will adhere to the Aufbau principle as the one-particle energy is the dominant factor. The \( \sim 25 \) meV energy difference is an effective barrier to occupation of any other orbitals apart from the \( |J_z| = \frac{3}{2} \) ones. A
similar case is encountered for the six-hole system. The first
six one-hole levels are strongly separated from the seventh
and the next ones (22 meV for \( B_z = 0 \)). This allows us to predict that in the six-hole wavefunction one will be facing
occupation of the orbitals corresponding to the mentioned
states. In both cases the two large energy separations in the
single-hole spectrum also translate to large separations in the
\( N_P = 2 \) and \( N_P = 6 \) ones—see figures 2(a) and (e),
respectively. Conversely, the relative vicinity of the \( |J_z| = \frac{1}{2} \) and
\( |J_z| = \frac{5}{2} \) states in the single-hole spectrum of figure 1(a)
opens the possibility of the occupation of the orbitals other than the
lowest \( N_P \) ones in the case of three, four and five holes. This
possibility is actually realized only for \( N_P = 3 \) and \( N_P = 4 \).

Let us now focus on the direct cause of the breaking of the
Aufbau principle in these two cases—that is the Coulomb
interaction. For \( N_P = 3 \) in the range where the violation
occurs, the \( J_z = -\frac{5}{2} \) orbital is preferred to the \( J_z = -\frac{1}{2} \) one.
This allows us to deduce that the Coulomb repulsion between the
\( J_z = -\frac{5}{2} \) and \( J_z = \pm \frac{3}{2} \) orbitals is weaker that between the
\( J_z = -\frac{1}{2} \) and \( J_z = \pm \frac{3}{2} \) orbitals. For \( N_P = 4 \) analogously the
\( J_z = -\frac{5}{2} \) orbital is preferred to the \( J_z = \frac{1}{2} \) one in respect of the
repulsion between it and the other occupied states (\( J_z = \pm \frac{3}{2} \)
and \( J_z = \frac{1}{2} \)).

In order to directly show that the Coulomb interaction is
a necessary factor for a violation of the Aufbau principle, the
spectrum figure 2(f) is included. This figure shows the results for a \( N_P = 4 \) model with no particle-particle interaction.
As expected, the \( J_z = \frac{3}{2} \) level is the ground state for the

| State          | \( J_z \)   | Dominating \( J_z^{\text{HI}} \) | Dominating \( J_z^{\text{env}} \) | Energy splitting (meV T\(^{-1}\)) |
|---------------|------------|-------------------------------|-----------------------------|-----------------------------|
| Ground        | \(-\frac{3}{2}\) | \(-\frac{3}{2}\) | 0                           | 0.234                       |
| 1 st excited  | \( \frac{3}{2} \)  | \( \frac{3}{2} \)  | 1                           | 0.211                       |
| 2 nd excited  | \(-\frac{1}{2}\) | \(-\frac{3}{2}\) | -1                          | 0.286                       |
| 3 rd excited  | \( \frac{1}{2} \)  | \( \frac{3}{2} \)  |                            |                            |
| 4 th excited  | \(-\frac{5}{2}\) | \(-\frac{3}{2}\) |                            |                            |
| 5 th excited  | \( \frac{5}{2} \)  | \( \frac{3}{2} \)  |                            |                            |

Figure 1. (a) The energy spectrum of one hole in an external magnetic field \( B_z \); strong confinement. (b) The profile of the confining potential along the \( z \)-axis including the biaxial strain for the different valence bands. Inset: the profile for \( z = 0 \) along the \( x-y \) plane radius \( \rho \).
magnetic field up to 8.25 T, which is the same $B_z$ value as for the crossing of the $J_z = \frac{1}{2}$ and $J_z = -\frac{5}{2}$ levels in figure 1(a).

A detailed study of the system in regard to the Aufbau principle is enabled by obtaining the occupation coefficients of the six lowest-in-energy orbitals for the multi-hole ground states. As the multi-hole basis is constructed for no magnetic field, and it is taken into account at the same stage as the Coulomb interaction then it is much more practical to use the one-particle orbitals at a $B_z = 0$ for obtaining these coefficients instead of the actual orbitals at given $B_z > 0$. This is justified as (i) we want only to discern the occupied orbitals from the unoccupied ones, and will not draw any conclusions from the details of the dependence of the coefficients on $B_z$ (ii) the single-hole states may only mix in the scope of the same $J_z$ subspace but for the whole concerned range of the magnetic field the $m = 1$ states are energetically separated from the $m \geq 2$ ones for each $J_z \in \{\pm\frac{1}{2}, \pm\frac{3}{2}, \pm\frac{5}{2}\}$—please note that this situation will differ for the weak confinement case.

Table 2. The variational parameters for the multi-hole systems. For the meaning of $N_P$, $N_{\text{first}}$, $N_{\text{second}}$ and $J_z^{N_P}$ symbols—see the text.

| $N_P$ | $N_{\text{first}}$ | $N_{\text{second}}$ | $J_z^{N_P}$ ∈ |
|-------|-------------------|---------------------|----------------|
| 2     | 100               | 100                 | $\{-9,..,9\}$ |
| 3     | 100               | 200                 | $\{-15/2,..,15/2\}$ |
| 4     | 100               | 300                 | $\{-8,..,8\}$ |
| 5     | 100               | 800                 | $\{-17/2,..,17/2\}$ |
| 6     | 100               | 1000                | $\{-9,..,9\}$ |

Figure 2. ((a)–(e)) The energy spectra of the multi-hole systems in an external magnetic field $B_z$; strong confinement. The eigenenergies for: (a) two holes, (b) three holes, (c) four holes, (d) five holes, (e) six holes. (f) The energy spectrum for the four hole case with no Coulomb interaction included. Only the states with $m = 1$ are shown.
The results for two holes are presented in figure 3(a). As expected, the orbitals corresponding to the ground \( J_z = -3/2 \) and the first excited states \( J_z = 3/2 \) of the single hole are occupied. The relevant coefficients are equal to about 0.97 for the whole range of the magnetic field. The other orbitals are empty, with the occupation coefficients of circa 0.01. The results for \( N_P = 3 \) are presented in figure 3(b). In the low magnetic field range, where the \( J_z^{N_P=3} = -1/2, m = 1 \) state is the ground state of the system, the first three orbitals are occupied: \( J_z = \pm 3/2, J_m = -1/2 \) with the relevant coefficients of 0.94 or larger. The two lowest orbitals are occupied: \( J_z = 1/2, J_m = \pm 5/2 \) with coefficients below 0.03. At the point of the crossing \( (B_z = 7.81 \) T) the character of the ground state changes to \( J_z^{N_P=3} = -5/2, m = 1 \) and— as expected—an electron is transferred from the \( J_z = -1/2 \) orbital to the \( J_z = -5/2 \) one. At this point one should note that this crossing has no corresponding one in the single hole spectrum (see figure 1(a)). The three orbitals that are lowest-in-energy for any value of the magnetic field in the considered range are: \( J_z = -3/2, m = 1 \), \( J_z = -5/2, m = 1 \) and \( J_z = -1/2, m = 1 \). The system follows the Aufbau principle for \( B_z < 7.81 \) T but another instance of its violation is the case of the \( N_P = 4 \)—as presented in figure 3(c). Although the \( J_z = \pm 3/2 \) orbitals are occupied, the \( J_z = \pm 1/2 \) ones are not paired. The \( J_z = -5/2 \) orbital is occupied instead of the \( J_z = 1/2 \) one for the whole considered range of the magnetic field. Please note that in figure 2(c) there is no signature of the \( B_0 \) crossing in figure 1(a). This is an example of a breaking of the Aufbau principle, one even more clear than in the \( N_P = 3 \) case because it happens for any \( 0 < B_z < B_0 \), so it obviously may not be interpreted as caused by a strong magnetic field. Our system returns to the previous behaviour, when an additional, fifth hole is confined in the dot. As shown in figure 3(d) the orbitals that correspond to the one-particle states of the five lowest energies are occupied with their occupation coefficients of about 0.91 to 0.95, depending on the orbital. The \( J_z = 5/2 \) orbital has its occupation coefficient lower than 0.07. All six one-particle orbitals are occupied with the occupation coefficients of over 0.93 when six holes are present in the system, as shown in figure 3(e).

In order to understand the details of the mechanism that leads to the breaking of the Aufbau principle in the case of \( N_P = 4 \) a simple matrix model was constructed. At the beginning we notice that the \( (J_z = \pm 3/2, m = 1) \) orbitals are occupied in the actual results and also should be occupied according to the mentioned principle. The difference between the two pictures is not due to two holes occupying the lowest orbitals. Hence we restrict ourselves only to effectively a \( N_P = 2 \) case.

As we want to know why the state has \( J_z^{N_P=2} = -3 \) instead of \( J_z^{N_P=2} = 0 \) then we should include in our model the \( J_z^{N_P=2} = -3 \) and \( J_z^{N_P=2} = 0 \) ones, excluding the levels that are assumed to be already occupied. In the energy range that we intend to study there are three such Slater determinants, namely:

\[
\Psi_0 = \hat{A}
\begin{pmatrix}
\phi_{3/2,1}
\phi_{3/2,-1}
\end{pmatrix}
, \quad \Psi_0 = \hat{A}
\begin{pmatrix}
\phi_{5/2,1}
\phi_{5/2,-1}
\end{pmatrix}
, \quad \Psi_0 = \hat{A}
\begin{pmatrix}
\phi_{7/2,1}
\phi_{7/2,-1}
\end{pmatrix}
.
\]

(18)

For \( B_z = 0 \) the Hamiltonian—when written in this basis—has the form of:

\[
\hat{H}_{KL} = \begin{pmatrix}
C_a & E_{m} & 0
2\Delta + C_b
0 & 0 & \Delta + C_a + C_b
\end{pmatrix}
\begin{pmatrix}
0
0
\frac{\Delta^2}{2} - E_m
\end{pmatrix}
\]

where \( \Delta = 0.64 \) meV is the effective difference between the energies of the \( \left(J_z = \pm \frac{5}{2}, m = 1\right) \) and \( \left(J_z = \pm \frac{3}{2}, m = 1\right) \) one-particle levels. \( \Delta = 19.32 \) meV is the Coulomb integral for \( \Psi_a \), \( C_a = 19.04 \) meV is the Coulomb integral for \( \Psi_b \) and \( E_m = 6.35 \) meV is the exchange integral for \( \Psi_b \) and also the off-diagonal element between \( \Psi_a \) and \( \Psi_b \) (for the last three values see table 3). The two lowest eigenvalues of this matrix are \( E_0 = \Delta + \frac{C_a + C_b}{2} - E_m \) and \( E_{a-b} = \Delta + \frac{C_a + C_b - \frac{1}{2} \sqrt{2 \left(C_a - C_b - 2 \Delta^2 + 4 E_m^2\right)}}{2} \). It can be seen that the latter one differs from the former only by the first term under the square root, which is small (1 meV²) in relation to the \( 4E_m^2 \) one (161 meV²). Indeed, the eigenvalues are numerically very similar: 13.47 meV and 13.45 meV. Thus the Coulomb-interaction-induced mixing of \( \Psi_a \) and \( \Psi_b \) is responsible for closing the energy gap between the \( J_z^{N_P=2} = -3 \) and \( J_z^{N_P=2} = 0 \) levels in the simplified model—which corresponds to the near-degeneracy of the relevant \( J_z^{N_P=2} = -3 \) and \( J_z^{N_P=2} = 0 \) levels in figure 2(c). At this point let us focus on the impact of a weak magnetic field on the two lowest energies of our simplified model. We will restrict ourselves to the linear terms only. Firstly, it should be noted that the vector corresponding to \( E_{a-b} \) is constructed as a linear combination of \( \Psi_a \) and \( \Psi_b \). Each of these two is a Slater determinant constructed with the states that are ‘mirror images’ in respect of \( J_z^{N_P=2} \) and \( J_z^{N_P=2} \) (see table 1). As a consequence the linear response to the magnetic field is zero for both basis states and therefore for their linear combination. On the other hand the \( \Psi_c \) that corresponds to \( E_c \) is a Slater determinant of the states that are more substantially different which results in a non-zero linear response to the magnetic field. As the value of the relevant coefficient is negative, the energy of \( J_z^{N_P=2} = -3 \) state is lowered while the energy of \( J_z^{N_P=2} = 0 \) state is not affected. This is precisely the situation that we encounter for the \( J_z^{N_P=2} = -3 \) and \( J_z^{N_P=2} = 0 \) levels in the full model (figure 2(c)) for low \( B_z \). To sum up: we have shown that the Coulomb interaction
and the magnetic field are both responsible for the breaking of the Aufbau principle in the system with four holes and we presented mechanisms by which those two factors contribute to the mentioned phenomenon.

3.2. Weak confinement

In this section we present the results for the weak confinement case i.e. the system with $R_{\text{dot}} = 20$ nm and $2Z_{\text{dot}} = 6$ nm. The volume of this dot is twelve times bigger than the volume of the former one. The value of the one-particle energy separation between the $J_{z} = \pm \frac{3}{2}$ levels and $J_{z} = \pm \frac{5}{2}$ ones at $B_{z} = 0$ for the system with the strong confinement is 25.5 meV and the value for the system with the weak confinement is 12.7 meV which constitutes about 50% of the first one. By comparing the values of the corresponding matrix elements in both systems (see tables 3 and 5) one can see that the values for the bigger dot are about 60% of the respective values for the smaller system. So as the dot size increases the characteristic values of both mentioned parameters diminish but the energy separation decreases slightly faster than the Coulomb interaction.

3.2.1. Single hole. The energy spectrum of a single hole in an external magnetic field $B_{z}$ for the weak confinement case is presented in figure 4(a). For a low magnetic field the two energy levels of lowest energy are: $\left(\pm \frac{1}{2}\right)$, $\left(\pm \frac{1}{2}\right)$. The next four levels, i.e. $\left(\pm \frac{3}{2}\right)$, $\left(\pm \frac{1}{2}\right)$, $\left(\pm \frac{5}{2}\right)$, $\left(\pm \frac{5}{2}\right)$ are nearly degenerate at $B_{z} \sim 0$. This is in agreement with a simple intuition that links the larger dimensions of the quantum dot with...
and \( \phi \) should be actually understood as level is strongly preferred to the states are nearly-degenerate for a significant \( \phi \) ones additionally 10. This would lead to an state is the ground level of the system. However, one (these will be called — especially the difference between the \( \phi \) ones while in single-hole spectrum the \( \phi \) level at

\[
J_z = -\frac{1}{2}, m = 1 \quad \text{and} \quad J_z = -\frac{5}{2}, m = 1 \quad \text{one (these will be called lower-energy pair) remains relatively small. Although the latter two levels are indeed slightly separated and their order changes in the crossing at \( B_0 \) (see table 4), they are nearly degenerate.}

For a magnetic field interval of \( B_\parallel \in (7,9) \) there is a series of level crossings that change the character of the levels from the third to the eighth, respectively. This part of the spectrum is presented in more detail in figure 4(b). The details concerning each crossing can be found in table 4.

Here a short digression is necessary. One should notice that in the case of the weak confinement in the single-hole spectrum there is a crossing of the states with the same \( J_z = -\frac{1}{2} \). We want to analyse the occupation coefficients for the multi-hole states in the same way, as it was done for the system with the strong confinement, thus for this purpose these two orbitals will be referred to by their orbital character at \( B_0 = 0 \). The orbital that corresponds to the \( (J_z = -\frac{1}{2}, m = 1) \) level at \( B_0 = 0 \) will be referred to as type I, and the one that corresponds to the \( (J_z = -\frac{1}{2}, m = 2) \) level at \( B_0 = 0 \) as type II. The appropriate marks were put on figure 4(b) and in table 4.

At this point one should note that the fact that the \( J_z = -\frac{1}{2} \) and \( J_z = -\frac{5}{2} \) states are nearly-degenerate for a significant part of the spectrum is a remarkable difference between the strong confinement case (figure 1(a)) and this one. The consequence of this situation is that in the non-interacting picture for \( N_p = 3 \) the energy separation of the ground and the first excited states will be very small. This allows us to suspect that the characteristics of the actual relevant multi-hole spectrum will be very essentially dependent on the Coulomb interaction between the holes. Furthermore, the presence of a series of level crossings in the upper right part of the single-hole case suggests that some signatures of them may be found in the multi-hole spectra.

3.2.2. Energy spectra of multiple holes. The energy spectra of the two-hole to six-hole weakly confined systems are presented in figure 5. The bigger-dot case means there is a multitude of excited states in the multiple-hole spectra and including them all would render the figures unreadable. Hence, for \( N_p \geq 4 \), only a ‘relevant’ subset of all levels is shown, which includes the ground state and all levels that interact with it in any way. The computation was conducted with the variational parameters as presented in table 2.

The analysis of the spectra should be supplemented by an examination of the occupation coefficients of the orbitals that are lowest-in-energy (at \( B_0 = 0 \)) for the multi-hole ground states—figure 6. However, because of the multiple level crossings occur in the one-particle spectrum, we decided to take these eight states into consideration: the same six as in the case of the strong confinement plus \( J_z = -\frac{7}{2}, m = 1 \) and \( J_z = -\frac{7}{2}, m = 2 \) ones additionally 10. This would lead to an appearance of too many lines if the figures were prepared in the format of figure 3, so only the occupied orbitals are shown.

If two holes are confined in the dot, the state with \( J_z = 0 \) is the ground state as shown in figure 5(a). This state is strongly separated from the excited states, which corresponds to the separation between the first two single-hole levels and the ones that are next in energy in figure 4. As expected, the lowest two single-hole orbitals are occupied, which can be seen in figure 6(b). Because of this character of the one-particle spectrum, the \( N_p = 2 \) case obviously follows the Aufbau principle.

In the case of three holes, for a weak magnetic field the \( J_z = -\frac{5}{2} \) state is the ground level of the system. However, at the \( B_\parallel = 7.4 \) there is a crossing and for the larger magnetic fields the \( J_z = -\frac{5}{2} \) state becomes the ground state. In terms of the level occupation (figure 6(b)) the crossing is a transfer of an electron from the \( J_z = -\frac{5}{2} \) orbital to the \( J_z = -\frac{7}{2} \) one. This corresponds to the \( B_0 \) crossing—shown in figure 6(b) and table 4—that takes place between the two single-hole levels in question at a similar magnetic field intensity (8 T). It should be noted that there is no trace in the \( N_p = 3 \) spectrum and/or the orbital occupation of neither the single-hole level crossing at \( B_0 \) nor the near-degeneracy of the relevant levels. Moreover, when no magnetic field is present in the system, the \( J_z = -\frac{5}{2} \) states have a noticeably lower energy than the \( J_z = -\frac{5}{2} \) ones while in single-hole spectrum the \( J_z = \pm \frac{5}{2} \) states have a slightly higher energy than the \( J_z = \pm \frac{5}{2} \) ones for \( B_0 = 0 \). In both the \( J_z = -\frac{5}{2} \) and \( J_z = -\frac{5}{2} \) states the \( J_z = \pm \frac{3}{2} \) orbitals are occupied. Hence, the mentioned facts suggest that the \( J_z = -\frac{5}{2} \) level is strongly preferred to the \( J_z = -\frac{5}{2} \) one in respect to the Coulomb repulsion between it and the

10 (\( J_z = -\frac{7}{2}, m = 2 \)) should be actually understood as \( J_z = -\frac{7}{2} \) type II.
spectra (ii) the shift between the magnetic field one which is a more subtle effect. The equivalent single-

6, 7 value to 6, 7, meV. The difference in the Coulomb interaction 3, 4, type II one meV and meV. Please refer to tables 3 and 5 for and orbital is of the state is clearly lower in T the 4, 5 orbital to the orbital to the 6, 7, meV, preferring is orbital to the 7, 8, value of the ground state does not change for up to 7, 8 level. Note that for the system with the strong confinement and NP = 3 the sequence of these two levels is reversed, as predicted by the Aufbau principle. In that case we have Δ = 1.46 meV and CP = −0.82 meV, hence

Table 4. The details concerning the level crossings of figures 4(a) and (b).

| Jz-s of involved levels | Ordinates of involved levels | Bz at crossing |
|-------------------------|-------------------------------|---------------|
| -5/2, -1/2              | 3, 4                          | B0 = 3.86 T   |
| -7/2, 5/2               | 6, 7                          | B1 = 7.51 T   |
| -7/2, 1/2               | 5, 6                          | B1 = 7.66 T   |
| -1/2, 5/2               | 7, 8                          | B1 = 7.76 T   |
| -1/2, 1/2               | 6, 7                          | B1 = 7.90 T   |
| -7/2, -1/2              | 4, 5                          | B1 = 7.97 T   |
| -7/2, -5/2              | 3, 4                          | B1 = 8.00 T   |
| -1/2, type I/II         | 5, 6                          | B1 = 8.17 T   |
| -5/2, -1/2              | 4, 5                          | B1 = 8.21 T   |

Jz = ± 3/2 orbitals (just like in the case of NP = 3 for the system with the strong confinement—see the description in the section 3.1.2). In the context of the Aufbau principle we should note: (i) the absence of the B0 level crossing clearly marks its violation—it may be thought of as a ‘strong’ kind of violation, the same kind as described earlier for the strong confinement NP ∈ {3, 4} spectra (ii) the shift between the magnetic field value of the Jz = ± 5/2 Jz = - 7/2 level crossing and the Jz = ± 3/2 Jz = - 1/2 one which is a more subtle effect—a ‘weak’ violation of the Aufbau principle.

The reason why the Jz = ± 3/2 state is clearly lower in energy than the Jz = ± 1/2 one for Bz = 0 while the relevant one-hole states have nearly the same energies, can be traced to the Coulomb interaction. The difference between the one-hole energies of (Jz = - 5/2, m = 1) and (Jz = - 1/2, m = 1) is Δ = 0.06 meV. The difference in the Coulomb interaction energy between the (Jz = ± 3/2, m = 1) states and the states in question is equal to CP = -0.28 meV. The effective energy difference is therefore Δ’ = Δ − CP = -0.22 meV, preferring the Jz = - 5/2 level. Note that for the system with the strong confinement and NP = 3 the sequence of these two levels is reversed, as predicted by the Aufbau principle. In that case we have Δ = 1.46 meV and CP = −0.82 meV, hence

Δ = Δ − CP = 0.64 meV. Please refer to tables 3 and 5 for the values of the particular matrix elements.

When one more hole is added to the quantum dot (NP = 4; figure 5(c)) then for Bz ∈ (0, 6.49 T) the Jz = - 3/2 level is the ground state of the system. Within this interval, the Jz = ± 3/2 orbitals are occupied as well as the lower-energy pair. At Bz = 6.49 T this level crosses the Jz = - 1/2 orbital to the Jz = - 7/2 one—as shown in figure 6(c). This intersection is an effect of the crossing of the relevant single-hole levels at Bz (see figure 4(b) and table 4). For the magnetic field of Bz = 8.48 T, there is an another crossing—this time connected to the B0 crossing in figure 4(b) and table 4—as the Jz = - 1/2 of the ground state switches to -4, and an electron is transferred from the Jz = - 5/2 orbital to the Jz = - 1/2 (type II) one.

When the dot is charged with five particles (figure 5(d)), the ground state in the magnetic field interval Bz ∈ [0, 4.25] T has Jz = - 3/2. The occupied orbitals are: Jz = ± 3/2, ± 1/2 and Jz = - 5/2 (figure 6(d)), which are the five lowest-energy ones for a weak magnetic field. At the point of the crossing (4.25 T) the Jz = 1/2 orbital changes to the Jz = - 7/2 one and that switches the Jz = - 5/2 value to - 1/2. The equivalent single-hole crossing takes place at Bz (see figure 4(b) and table 4). This Jz = - 5/2 value of the ground state does not change for up to 10 T, but at Bz = 8.72 T we observe an another crossing in the five-hole spectrum, and by analysing figure 6(d) one can see that it is connected to the crossing of the type I and the type II Jz = -1/2 single-hole states, as seen in figure 4(b) at Bz.

For six holes the Jz = 0 level has the lowest energy up to 5.05 T (figure 5(e)), with the Jz = ± 3/2, ± 1/2 orbitals occupied (figure 6(e)). At that point the Jz = ± 5/2 orbital is exchanged for the Jz = - 7/2 one, which corresponds to the B1 crossing in figure 4(b) and which lowers the Jz = 0 level of the ground state by six. The next crossing, which takes place for the magnetic field of 6.22 T, marks the transfer of an electron from the Jz = ± 5/2 orbital to the Jz = 0 type II one—an analogue to B1 in single-hole system—which sets Jz = 0 to -7.
In the cases of \( N_p \in [4, 5, 6] \) the Aufbau principle allows the set of occupied one-particle orbitals to be predicted for each inter-crossing interval along with the order in which the occupied orbitals change in the case of each crossing. However, the actual values of the magnetic field intensities at which these changes do occur in the multi-hole spectra do not coincide exactly with the \( B_z \) values of the relevant single-hole crossings. In short, the principle is only ‘weakly’ violated in the meaning described above for \( N_p = 3 \). The details on the subject of comparison of the particular crossings are presented in table 6.

The absence of a multi-hole crossing corresponding to the \( B_1 \) one is not a surprise, as the latter one involves the seventh and the eighth single-hole levels (in the order of the increasing energy) and hence the former would appear in the multi-hole spectra of \( N_p < 7 \) only in a case of a ‘strong’ violation of the Aufbau principle.

4. Discussion and conclusion

In [9] experimental data were presented by Reuter et al for the hole charging spectra of self-assembled InAs quantum dots in perpendicular magnetic fields probed by the capacitance-voltage spectroscopy. The authors of that work interpreted the results in the terms of the typical results obtained for electrons i.e. the \( s, p \) and \( d \) shell system for the envelope functions that conforms to the Aufbau principle. They reported the so
called ‘incomplete hole shell filling’ (the $d$ shell starts to be occupied before the $p$ shell is full) and they understood this as the breaking of the Aufbau principle induced by the Coulomb interaction.

Later, Climente et al. presented a work [10] that illustrated the difference between the typical $s$-$p$-$d$ shell system of the heavy hole model (similar as in the case of conduction band electrons) and the results obtained when the valence band mixing is taken into account. Authors of [10] have shown that using the latter model for a single hole—in the form of the four-band KL Hamiltonian—leads to obtaining a set of three twofold Kramers degenerate shells. The shells correspond to the absolute value of the total angular momentum of the holes $|J_z|$ equal to 3/2, 1/2 and 5/2, respectively. In the mentioned work, the authors do not include the Coulomb interaction in the case of the KL model but instead claim that the behaviour of the system can be understood in terms of non-interacting holes when the valence band mixing is included. The non-interacting picture implies that the Aufbau principle is trivially fulfilled.

This work is a continuation of the studies of the group of Planelles & Climente (see [10, 15–17]). It employs a model that is in several ways more precise than the one used by [10]. Firstly, the spin–orbit split-off bands are included with the six-band KL Hamiltonian instead of the four-band one. Furthermore, we include the Coulomb interaction between the holes via the configuration interaction method. Thirdly, the effect induced by the strain on the confinement potential

**Figure 6.** (a)–(e) The occupation of the single-hole orbitals in the multi-hole systems in an external magnetic field $B_z$; weak confinement. The case of: (a) two holes, (b) three holes, (c) four holes, (d) five holes, (e) six holes. Only the occupied orbitals are shown. The vertical dash–dot–dot lines correspond to $B_z$ of the respective level crossings, as marked in figure 5.
The production of the valence band mixing at the one-particle level is directly induced. Furthermore, we show that the sole introduction of a few/several holes confined in a quantum dot of the kind completely omits the Coulomb interaction. On the other hand, the result of [10] could suggest that inter-particle interaction should be included in a more direct manner. Together with their results this implies that the hole Coulomb interaction and smaller quantization energies. The authors of the latter work use a model in a manner. The opposite is true. The latter work uses the Pikus–Bir Hamiltonian. They also say that induced by the inter-particle interactions.

We prove that the Coulomb interaction influences very strongly the conduction band electrons and thus the spatial carrier confinement. The valence band holes have larger effective masses than the conduction band. It is taken into account by using the Pikus–Bir Hamiltonian. Finally, an enhanced model for the magnetic field is used that is known to avoid some problems of the old model, as described in section 2.4.

We show that even if the Aufbau principle would be applied at single-hole crossings, it may still be violated and that violation is induced by the inter-particle interactions. Authors of [9] note that the results in the case of electron charging experiments could be well explained when treating the Coulomb interaction as a perturbation. They also say that the valence band holes have larger effective masses than the conduction band electrons and thus the spatial carrier confinement in the hole systems is stronger and this results in a stronger hole–hole Coulomb interaction and smaller quantization energies. Together with their results this implies that the inter-particle interaction should be included in a more direct manner. On the other hand, the result of [10] could suggest that the opposite is true. The authors of the latter work use a model that completely omits the Coulomb interaction.

We prove that the Coulomb interaction influences very essentially the general behaviour and properties of the system of a few/several holes confined in a quantum dot of the kind considered in the work. Thus, we believe that all further $\vec{k} \cdot \vec{p}$ studies of the systems similar to this one should include this interaction directly. Furthermore, we show that the sole introduction of the valence band mixing at the one-particle level is insufficient to understand the so called ‘incomplete hole shell filling’ phenomenon.

Please note that works [26–29] offer an alternative explanation of the phenomenon that is the topic of this work, from a completely different approach—the atomistic scale one.

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### Table 5.

| Matrix element | Coulomb integral | Exchange integral | Overall value |
|----------------|------------------|------------------|---------------|
| $\langle \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \mid \hat{H}_{\text{int}} \mid \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \rangle$ | 14.50 | 0 | 14.50 |
| $\langle \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \mid \hat{H}_{\text{int}} \mid \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \rangle$ | 12.11 | 3.45 | 8.66 |
| $\langle \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \mid \hat{H}_{\text{int}} \mid \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \rangle$ | 12.11 | 0 | 12.11 |
| $\langle \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \mid \hat{H}_{\text{int}} \mid \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \rangle$ | 12.02 | 3.55 | 8.47 |
| $\langle \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \mid \hat{H}_{\text{int}} \mid \phi_{\downarrow 1}, \phi_{\downarrow 2}, \phi_{\downarrow 3} \rangle$ | 12.02 | 0 | 12.02 |

### Table 6.

| $B_0$ at single-hole crossing | $N_p$ for multi-hole crossing | $B_0$ at multi-hole crossing |
|-----------------------------|-----------------------------|-----------------------------|
| $B_0 = 3.86 \text{ T}$      | 3                           | Does not occur              |
| $B_1 = 7.51 \text{ T}$      | 6                           | 5.05 T                      |
| $B_2 = 7.66 \text{ T}$      | 5                           | 4.25 T                      |
| $B_3 = 7.76 \text{ T}$      | Not applicable              | Does not occur              |
| $B_4 = 7.90 \text{ T}$      | 6                           | 6.22 T                      |
| $B_5 = 7.97 \text{ T}$      | 4                           | 6.49 T                      |
| $B_6 = 8.00 \text{ T}$      | 3                           | 7.4 T                       |
| $B_7 = 8.17 \text{ T}$      | 5                           | 8.72 T                      |
| $B_8 = 8.21 \text{ T}$      | 4                           | 8.48 T                      |
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