Singlet-triplet transitions in highly correlated nanowire quantum dots

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Abstract

We consider a quantum dot embedded in a three-dimensional nanowire with tunable aspect ratio \(a\). A configuration interaction theory is developed to calculate the energy spectra of the finite 1D quantum dot systems charged with two electrons in the presence of magnetic fields \(B\) along the wire axis. Fruitful singlet-triplet transition behaviors are revealed and explained in terms of the competing exchange interaction, correlation interaction, and spin Zeeman energy. In the high aspect ratio regime, the singlet-triplet transitions are shown desirable by tuning the parameters \(a\) and \(B\). The transitions also manifest the highly correlated nature of long nanowire quantum dots.

Key words: Nanowire quantum dot, Exchange, Correlation, Singlet-triplet transition

1. Introduction

For years, few electron charged quantum dots have attracted extensive attention due to the controllable electronic and spin properties \([1]\). However, only few attempts have so far been made for studies of finite 1D nanowire quantum dots (NWQDs). More recently, it was shown that the NWQDs formed in the heterostructures in nanowires can be fabricated as single electron transistors and successively charged with controlled number of electrons \([2]\). The successful experimental works motivate us to explore the possible geometric effects of NWQDs characterized by their aspect ratios, \(a\), on the electronic and spin properties of two-electron charged NWQDs.

In this work, we focus on the study of singlet-triplet (ST) transitions in two-electron charged NWQDs \([3]\), conducted by using a developed configuration interaction (CI) theory in combination with the exact diagonalization techniques based on a 3D asymmetric parabolic model. It will be illustrated that the ST transitions in InAs-based NWQDs driven by an appropriate magnetic field are associated with the competing effects of large spin-Zeeman energies as well as the exchange and correlation energies. The correlation-dominated nature of a long NWQD (i.e. with high aspect ratio) will be identified by the spin phase diagram with respect to the applied magnetic fields and the tunable aspect ratio.

2. Theoretical Model

2.1. Single-electron spectrum

We begin with the single electron problem of a NWQD with axial magnetic field \(B = (0, 0, B)\), described by the Hamiltonian \([4]\)

\[
H_0 = \frac{1}{2m^*}(p + e\mathbf{A})^2 + V(x, y, z) + H_Z, \tag{1}
\]

where \(\mathbf{A} = (B/2)(y, -x, 0)\) denotes the vector potential and \(m^*\) stands for the effective mass of an electron with charge \(-e\). The spin-Zeeman Hamiltonian \(H_Z = g^*\mu_B B z\) is in terms of the z-component of electron spin \(s\), and the effective Lande g-factor of electron \(g^*\) and the Bohr magneton \(\mu_B\). In addition, the confining potential \(V(x, y, z) = m^* [\omega_0^2 (x^2 + y^2) + \omega_z^2 z^2]/2\) is assumed of the parabolic form with \(\omega_0\) and \(\omega_z\) parametrizing, respectively, the transverse and the longitudinal confining strength.

In this work, we assume a constant \(g^*\) and take the \(g^* = -8.0\) for InAs \([3, 6]\). The single electron Hamiltonian \([1]\) leads to the extended Fock-Darwin single-particle spectrum

\[
\epsilon_{n,m,q,s_z} = \hbar\omega_z \left( n + \frac{1}{2} \right) + \hbar\omega_c \left( m + \frac{1}{2} \right) + \hbar\omega_0 \left( q + \frac{1}{2} \right) + E_Z, \tag{2}
\]

where \(n, m, q = 0, 1, 2, \ldots\) denote oscillator quantum numbers, \(s_z = +\frac{1}{2} (s_z = -\frac{1}{2})\) the projection of electron spin \(\uparrow (\downarrow)\) \(E_Z = g^*\mu_B B_z\), the spin Zeeman energy, and \(\omega_0\) and \(\omega_z\) defined in terms of the hybridized frequency \(\omega_h \equiv (\omega_0^2 + \omega_z^2/4)^{1/2}\) and the cyclotron frequency \(\omega_c = eB/m^*\).

The eigenstate \(|n, m, q\rangle\) possesses the orbital angular momentum \(l_z = \hbar(n - m)\) and the parity \(P = 1 (P = -1)\) with respect to \(z\)-axis for the even (odd) \(q\) number. The wave function of the lowest orbital is given by \(\psi_{000}(r) = \exp\left[-((x^2 + y^2)/\beta^2 + z^2/\ell_z^2)/4\right]/(2\pi\beta^2\ell_z^2)^{1/4}\) with the characteristic lengths of the wave function extends \(l_h = \sqrt{\hbar/2m^*\omega_0}\) and \(l_z = \sqrt{\hbar/2m^*\omega_z}\), from which one can generate the wave functions of any other excited states by successively applying
2.2. Interacting NWQD

To investigate the few-electron interaction effects in a NWQD, we express the few electron Hamiltonian in second quantization as

\[ H = \sum_{i,j,k,l} \epsilon_{ij} c_{i,j}^\dagger c_{j,k} + \frac{1}{2} \sum_{i,j,k,l} (ij) V[kl] c_{i,j}^\dagger c_{k,l}^\dagger c_{l,i} c_{k,j}, \]

where \( i,j,k,l \) stand for the composite indices of single electron orbitals (e.g. \( | i \rangle = | m_i, m_j, q_i \rangle \)), \( \sigma = \uparrow, \downarrow \) denotes the electron spin with \( s_z = +\frac{1}{2}, -\frac{1}{2} \), and \( c_{i,j}^\dagger (c_{i,j}) \) is the electron creation (annihilation) operators. The first (second) term on the right-hand side of Eq. (4) represents the kinetic energy of electrons (the Coulomb interactions between electrons) and the Coulomb matrix elements are defined as \( (ij)V[kl] \equiv e^2/(4\pi\epsilon) \int dx dy dx' \int dy' \psi^*(r) V(r) \psi(r') (|r_1 - r_2|) \psi(r_2) \psi(r_1) \), where \( \epsilon \) is the dielectric constant of dot material (\( \epsilon = 15.15 \epsilon_0 \) is taken for InAs throughout this work). After lengthy derivation, for NWQDs with \( a \geq 1 \), we obtain the following formulation of the Coulomb matrix elements:

\[ \langle n_i m_i q_i ; n_j m_j q_j | V[n_l m_l q_l ; n_l m_l q_l] \rangle = \left( \frac{1}{n_l} \right) \delta_{R_l, R^*} \delta_{q_l + q_j, q_j + q_l, even} \min(n_i, n_j) \min(n_i, n_j) \min(q_i, q_j) \min(q_i, q_j, q_k) \times \prod_{p=0}^{n_1} \prod_{p=2}^{n_1} \prod_{p=3}^{n_1} \prod_{p=4}^{n_1} \prod_{p=5}^{n_1} \times \left( -1 \right)^{n_i + n_j} \times \frac{1}{2} \times \left( \frac{1}{2} \right)^{x+\frac{1}{2}} \Gamma\left( 1 + \frac{x+\frac{1}{2}}{2} \right) \right. \]

where we have defined \( u = m_1 + m_2 + m_3 + m_4 - (p_1 + p_2 + p_3 + p_4) \), \( v = q_1 + q_2 + q_3 + q_4 - 2(p_3 + p_4) \), \( R_L = (m_1 + m_2) - (n_1 + n_2) = -(L_1 + L_2) \), \( R_R = (m_1 + m_3) - (n_1 + n_3) = -(L_1 + L_3) \), \( x \equiv \omega / \omega_{0} \), and \( \Gamma \) is the hypergeometric function. The \( \delta \)-functions in the formulation ensure the conservation of the parity with respect to \( z \)-axis and the \( z \)-component of angular momentum of system \( L_z \), respectively. The formulation of Eq. (5) is reexamined by computing the Coulomb integral numerically.
spectrally. In Figure 4(a), we show the large, while
of two interacting electrons in a NWQD with
3. Results and discussion

Figure 3 presents the FCI result of magneto-energy spectrum
The energy spectrum of an interacting two-electron NWQD
is calculated using the standard numerical exact diagonalization
technique [8]. The numerically exact results are obtained by increas-
ing the numbers of chosen single electron orbital basis and
the corresponding two-electron configurations until a numerical
convergence is achieved. In our full configuration interaction
(FCI) calculations, we chose the typical orbital number from 20
to 26 and the number of corresponding configurations is from
190 to 325. In order to highlight the Coulomb correlations, we
also carry out the partial CI (PCI) calculations in which only
the lowest energy $N_e$ configuration is taken and compare the
PCI results with those obtained from the FCI calculations.

Figure 3: Correlated two-electron energy spectrum as a function of magnetic
field in a NWQD with diameter $L_x = 50 \text{ nm}$ and aspect ratio $a = 3$.

2.3. Exact diagonalization

The energy spectrum of an interacting two-electron NWQD
is calculated using the standard numerical exact diagonalization
technique [8]. The numerically exact results are obtained by increas-
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resulting two-electron configurations until a numerical convergence is achieved. In our full configuration interaction
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the lowest energy $N_e$ configuration is taken and compare the
PCI results with those obtained from the FCI calculations.

Figure 4: (a) Singlet-triplet splitting $\Delta_{ST}$ as a function of aspect ratio $a$ with
$B = 1 \text{ T}$. (b) The spin phase diagram for electrons making singlet-triplet transition
with respect to magnetic field $B$ versus aspect ratio $a$.

3. Results and discussion

Figure 3 presents the FCI result of magneto-energy spectrum
of two interacting electrons in a NWQD with $a = 3$. The ST
transition of the two-electron ground state is shown to happen as $B_{ST} \sim 0.9 \text{ T}$. As the applied magnetic field is weak, the spin
Zeeman splitting is small and the two electrons mostly dou-
bly fill the lowest S-orbital. With increasing magnetic field in-
creases, the energy difference between triplet and singlet states
of the two electrons decreases because of increasing spin Zee-
man and exchange energies, both of which energetically fa-
vor the triplet states $|T^+\rangle$. As the applied field is higher than $B_c \sim 0.9 \text{T}$, the ground state of two electrons transit from the
singlet state to the triplet one.

Now we turn to study the singlet-triplet triplet splitting as a function of $a$, defined by $\Delta_{ST} \equiv E_{T} - E_{S}$, with $i = -, 0,$
and $+ \text{ corresponding to the } T^-, T^0,$ and $T^+$ triplet states,
respectively. In Figure 4(a), we show the $\Delta_{ST}$ as a function of aspect ratio $a$ under a fixed magnetic field $B = 1 \text{ T}$. In the non-
interacting case, $\Delta_{ST}^{\text{dir}}$ are shown to decrease monotonically with increasing aspect ratio $a$. Since only $T^+$ energy is decreased by spin-Zeeman term, the ST transition could only occur between $S$ and $T^+$ states. That is, only $\Delta_{ST}^+$ crosses zero as $a$ is very large, while $\Delta_{ST}^+$ remains positive always. Thus below we shall
only consider $\Delta_{ST}^+$ for the discussion of ST transition. The non-
interacting ST splitting can be derived as $\Delta_{ST}^{\text{dir}} = \hbar \omega_0 / a^2 - 2E_Z$, explicitly showing the quadratic decrease of $\Delta_{ST}^+$ with respect to $a$. Accordingly, in the non-interacting picture, the critical aspect ratio $a_{ST}^*$ where the ST transition occurs is predicted as $a_{ST}^* = \sqrt{\hbar \omega_0 / g^* \mu_B B}$.

However, the PCI calculation predicts a much smaller value
of critical aspect ratio $a_{ST}^* = 2.5$. In the PCI result, the ST
splitting is substantially reduced by the energy reduction of the
T state due to the reduced direct Coulomb interaction and the
negative exchange interaction between the two electrons in the
state. The PCI calculation shows $a_{ST}^* = 2.9$, as indicated by the
dashed vertical line in Figure 4(a). In fact, the difference in the values of $\Delta_{ST}^+$ obtained from the PCI and FCI calculations
increases as $a$ increases. This indicates that the Coulomb correlation effect tends to increase the ST splitting again and
becomes even more pronounced in long NWQD with high $a$.

Figure 4(b) shows the calculated spin phase diagram of two-
electron NWQDs with respect to the aspect ratio $a$ and applied
magnetic field $B$. The spin singlet and triplet phases, appearing
in the low $a$-B and high $a$-B regimes, respectively, are distin-
guished by the curve of $B_{ST}$ which show a monotonic decrease
with $a$. For noninteracting electrons, the critical magnetic field
can be derived as $B_{ST} = \hbar \omega_0 / \gamma_0 \mu_B a^2$, showing a quadratic decay
with $a$.

In comparison with the non-interacting cases, the PCI cal-
culations obtain the $B_{ST}$ that is significantly reduced and goes
to zero for $a > 2.9$. In the one-configuration approxima-
tion used in the PCI calculation, the ST splitting is given by
$\Delta_{ST} = \Delta_{ST}^{\text{dir}} + \Delta_{ST}^{\text{ex}}$, where $\Delta_{ST}^{\text{ex}}$ is the ST splitting in the
non-interacting cases, $\Delta_{ST}^{\text{dir}} \equiv V_{ST}^{\text{dir}} - V_{ST}^{\text{ferm}} < 0$ is the direct energy
difference between the triplet and the singlet states, and $V_{ST}^{\text{ex}}$ is the exchange energy between electrons in the $T^+$ state.
ingly, we obtain $B_{ST} = (\hbar \omega / a^2 + \Delta_{\text{dir}}^{\text{ST}} - V_{\text{ex}}^a) / g' \mu_B$. In the large aspect ratio regime, the negative $\Delta_{\text{dir}}^{\text{ST}}$ and $V_{\text{ex}}^a$ reduce the $E_T$ and $B_{ST} = 0$ results for $a > 3$. However, the FCI calculation predict larger and always positive $B_{ST}$. In fact, as increasing $a$, the relative strength of electronic Coulomb correlations increases because of reduced $\hbar \omega$, and strong configuration interactions. Such $a$-engineered Coulomb correlations energetically favor the singlet two-electron states and result in the non-zero $B_{ST}$ in the high aspect ratio regime.

4. Summary

In conclusion, a configuration interaction (CI) theory is developed for studying the magneto-energy spectra and the singlet-triplet transitions of two-electron NWQDs with longitudinal magnetic field $B$ and tunable aspect ratio $a$. For short NWQDs of low aspect ratio $a < 3$, the ST transition behaviors are dominated by the spin Zeeman, Coulomb direct and exchange energies, and can be well studied by using PCI calculation. However, our FCI calculations show the increasing importance of Coulomb correlations in long NWQDs with increasing aspect ratio $a$ over 3. The FCI calculation present the spin phase diagram of a two-electron NWQD which are highly dependent on $a$, and suggests the controllability of singlet or triplet spin states by changing the aspect ratio of NWQD.

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