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Singlet–triplet transitions in a few-electron quantum dot

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Abstract

We have measured spin-singlet–spin–triplet (ST) transitions in a vertical quantum dot containing up to four electrons. Current through the dot is measured as a function of gate voltage and magnetic field (0–9 T) at both small and large source drain voltages. The ST transitions cannot be explained within the framework of single-particle states in combination with a constant Coulomb interaction. Taking into account exchange interaction and a magnetic field dependent direct Coulomb interaction is essential for describing the observed ST transitions.

Keywords: Singlet–triplet transition; Quantum dots; Mesoscopic physics

Quantum dots are often referred to as artificial atoms, because of the many similarities with real atoms [1]. The relatively large dimensions of quantum dots (~100 nm) make that accessible magnetic field regimes (up to ~20 T) correspond to regimes of the order 10^6 T for real atoms. The constant interaction (CI) model has been successful to describe transport through dots [2,3]. The CI model assumes that the Coulomb interactions between electrons are independent of, for instance, the magnetic field, so that changes in the observed ground state energies are fully asscribed to changes in the single-particle energies. While the CI model is very useful at small magnetic fields ($B \lesssim 1$ T) [2], at larger B it is essential to include a varying Coulomb interaction [4]. Here, we discuss this non-constant interaction regime for dots with one to four electrons and B between 0 and 9 T. In particular, we describe the singlet–triplet (ST) transition induced by a magnetic field for a dot with two electrons.

Our vertical quantum dot is a sub-micrometer pillar fabricated in an In/Al/GaAs double barrier heterostructure [5,6]. Source and drain wires are connected to the doped top and substrate contacts. Previous measurements have shown that the lateral confinement potential has the form of a circular-symmetric, harmonic potential [4,5]. The diameter of the dot can be reduced with a gate voltage, V_g, from a few hundred nanometers to zero, thereby decreasing the electron number, N, from ~70 to 0. The measurements are done in a dilution refrigerator at 100 mK.

We first discuss the energy spectrum of a few-electron 2D quantum dot. The Darwin–Fock (DF) energy spectrum for non-interacting electrons in a quantum dot with a parabolic confinement potential $V(r) = \frac{1}{2}m^*\omega_0^2 r^2$ is given by [7]

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$$E_{n,l} = (2n + |l| + 1)\hbar\sqrt{\frac{1}{4}\omega_c^2 + \omega_0^2} - \frac{1}{2}l\hbar\omega_c, \qquad (1)$$

where $m^* = 0.06\,m_0$ is the effective mass in GaAs, ω_0 the harmonic oscillator frequency, $\omega_c = eB/m^*$ the cyclotron frequency, n the radial quantum number, and l the angular momentum number. For a two-electron dot, we only consider the two lowest single-electron states, $E_{0,0}$ and $E_{0,1}$, which are relevant to the discussion here. For any value of B, $E_{0,0} < E_{0,1}$ and hence the two electrons both occupy the state $E_{0,0}$ with opposite spin; i.e. a singlet, $S = 0$ state. Including the Zeeman energy, we obtain

$$E_{0,0} = \hbar\sqrt{\frac{1}{4}\omega_c^2 + \omega_0^2} \pm \frac{1}{2}g^*\mu_B B$$

$$E_{0,1} = 2\hbar\sqrt{\frac{1}{4}\omega_c^2 + \omega_0^2} - \frac{1}{2}\hbar\omega_c \pm \frac{1}{2}g^*\mu_B B$$

with g^* the effective Landé factor and μ_B the Bohr magneton. We now expect a crossing between $E_{0,0,\downarrow}$ and $E_{0,1,\uparrow}$ beyond which the two electrons are spin-polarized; i.e. a triplet, $S = 1$ state. For $\hbar\omega_0 = 5.6$ meV and $g^* = -0.44$ in GaAs, we obtain a ST transition driven by the Zeeman energy at $B = 25$ T. Note that this estimate neglects electron–electron interactions. As we now discuss, the Coulomb interactions between the two electrons drive the ST transition to much lower B.

The interdependence of Coulomb interactions and single-particle states becomes important when a magnetic field changes the size of the electron states. For instance, the size of the DF-states shrinks in the radial direction at larger B. To illustrate this, we show the probability functions $|\psi_{n,l}(r,\varphi)|^2$ for the $E_{0,0}$ and $E_{0,\pm1}$ states at $B = 0$ in Fig. 1(a) [3]. Fig. 1(b) shows cross sections of $|\psi_{n,l}(r,\varphi)|^2$ versus B. Increasing B, the distribution function shrinks in the radial direction. When two electrons both occupy the $E_{0,0}$ state, the average

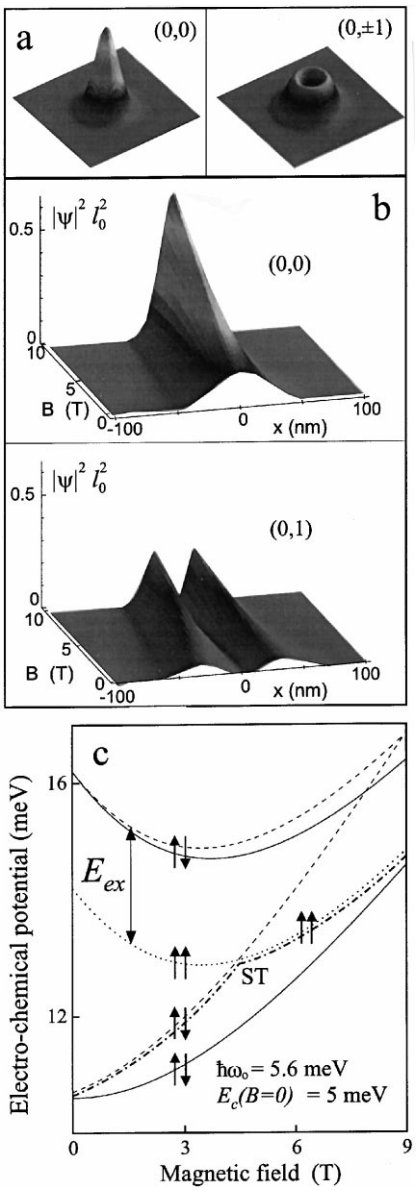

Fig. 1. (a) Calculated spatial distribution functions $|\psi_{n,l}(r,\varphi)|^2$ for $(n,l) = (0,0)$ and $(0,\pm1)$. (b) Magnetic field dependence of the distribution functions of (a) ($\hbar\omega_0 = 3$ meV). The graphs are normalized by $l_0^2 = \hbar/m^*\omega$. (c) Electro-chemical potential $\mu(2)$ of a two-electron dot as function of magnetic field ($\hbar\omega_0 = 5.6$ meV, $E_c(B=0) = 5$ meV). The lower solid curve represents the CI ground-state electro-chemical potential $\mu^{\uparrow\downarrow}(2) = E_{0,0} + E_c$ (spins indicated by arrows), whereas the upper solid curve corresponds to the excited state, $\mu^{\uparrow\downarrow,\mathrm{ES}}(2) = E_{0,1} + E_c$. The dashed lines schematically represent the situation in which a B-dependent Coulomb interaction is taken into account. Note that the dashed lines grow faster than the solid ones. The rise of the lower one is larger, due to the larger overlap of states when both electrons are in the groundstate. The upper dashed curve with subtraction of a constant exchange energy E_{ex} results in the dotted curve $\mu^{\uparrow\uparrow}(2) = E_{0,1} + E_c(B) - E_{ex}$. $\mu^{\uparrow\downarrow}(2)$ and $\mu^{\uparrow\uparrow}(2)$ cross at $B \simeq 4.5$ T. The ground state before and after the ST transition is indicated by a dashed-dotted line.

distance between them decreases with B and hence the Coulomb interaction increases. At some magnetic field it is energetically favorable if one of the two electrons makes a transition to a state with a larger radius (i.e. from $l=0$ to $l=1$), thereby increasing the average distance between the two electrons. This transition occurs when the gain in Coulomb energy exceeds the costs in single-particle energy. So, beside the Zeeman energy, the shrinking of wave functions favors a transition in angular momentum.

Numerical calculations by Wagner et al. [8] have predicted these ST transitions. In our discussion here, we generalize the CI model in order to keep track of the physics that gives rise to the ST transition. The electro-chemical potential of a dot containing N electrons is defined as $\mu(N) \equiv U(N) - U(N-1)$ where $U(N)$ is the total energy of the dot. For an $N=1$ dot, $U^{\uparrow}(1) = E_{0,0}$ is the exact ground state energy (spins are indicated by arrows). In the CI model, $U^{\uparrow\downarrow}(2) = 2E_{0,0} + E_c$, so that the ground-state electro-chemical potential is $\mu^{\uparrow\downarrow}(2) = E_{0,0} + E_c$. Note that the Coulomb interactions are assumed to be described by a constant charging energy E_c. The first excited state is $U^{\uparrow\downarrow,ES}(2) = E_{0,0} + E_{0,1} + E_c$ and $\mu^{\uparrow\downarrow,ES}(2) = E_{0,1} + E_c$. The solid lines in Fig. 1(c) show $\mu^{\uparrow\downarrow}(2)$ and $\mu^{\uparrow\downarrow,ES}(2)$, where we neglect the small contribution of the Zeeman energy.

The next level of approximation is to include the magnetic field dependence of the charging energy to account for the shrinking wave functions. The dashed curves in Fig. 1(c) rise somewhat faster than the solid curves, reflecting the B-dependence of the charging energy, $E_c(B)$. These dashed lines are schematic curves and do not result from calculations.

When both electrons occupy the $E_{0,0}$ state their spins must be anti-parallel. However, if one electron occupies $E_{0,0}$ and the other $E_{0,1}$, the two electrons can also take on parallel spins; i.e. the total spin $S=1$. In this case, the Coulomb interaction is reduced by an exchange energy E_{ex} and the corresponding electro-chemical potential becomes $\mu^{\uparrow\uparrow}(2) = E_{0,1} + E_c(B) - E_{ex}$; see the dotted line in Fig. 1(c). (The exchange energy is due to a deformation of the distribution functions of Fig. 1(a) for electrons with parallel spin, which yields a reduction of the Coulomb repulsion.) Importantly, $\mu^{\uparrow\downarrow}(2)$ and $\mu^{\uparrow\uparrow}(2)$ cross at $B \simeq 4.5$ T for the parameters chosen in Fig. 1(c). So, while for $B < 4.5$ T the ground state energy corresponds to two electrons with anti-parallel spins in the lowest single-particle state, for $B > 4.5$ T the ground state has contributions from two single-particle states and has total spin $S=1$ (the two-electron ground state is indicated by a dashed-dotted line in Fig. 1(c)). Thus, while the Zeeman-driven transition would occur at 25 T, the electron–electron interactions push the ST transition to 4.5 T [9].

Capacitance [10] and tunneling [11,12] spectroscopy have provided evidence for ST transitions in the two-electron ground state energy. Here, we report the evolution of the ground-state as well as the first excited state versus B. Fig. 2(a) shows the linear response Coulomb blockade peaks for $N=0$–4. The four curves reflect how the ground state electrochemical potentials $\mu(N)$ for $N=1$–4 evolve with B. We emphasize that, based on the DF-spectrum for non-interacting electrons, one does not expect transitions or kinks in the B-dependence of $\mu(N)$ for $N=1$–4. The peak for $N=1$ indeed has a smooth B-dependence. For $N=2$, 3, 4, however, we observe kinks, which are indicated by arrows. These kinks must arise from interactions not included in the CI model. The left arrow in the $N=4$ trace is due to the destruction of a Hund's rule state, which has been discussed previously [4,5]. To blow up the different kinks, we extracted the peak positions and converted their values from gate voltage to energy [3]. The plotted curves in Fig. 2(b) are shifted towards each other and represent the variation of the electrochemical potential with B. The lowest curve for $N=1$ shows a smooth [13] increase in energy in accordance with the expected solid curve for $E_{0,0}$ in Fig. 1(c). The next curve for $N=2$ rises faster with B than the $N=1$ curve, which reflects the magnetic field dependent interaction $E_c(B)$ (see lower dashed curve in Fig. 1(c)). At 4.5 T, indicated by "a", we observe a kink in the $N=2$ curve. This is the expected ST transition. Our choice of shifting the peak position curves for $N=1$ and 2 to zero at $B=0$ in Fig. 2(b), allows a direct comparison, and shows a good agreement with the lower solid curve and the dashed-dotted curve in Fig. 1(c).

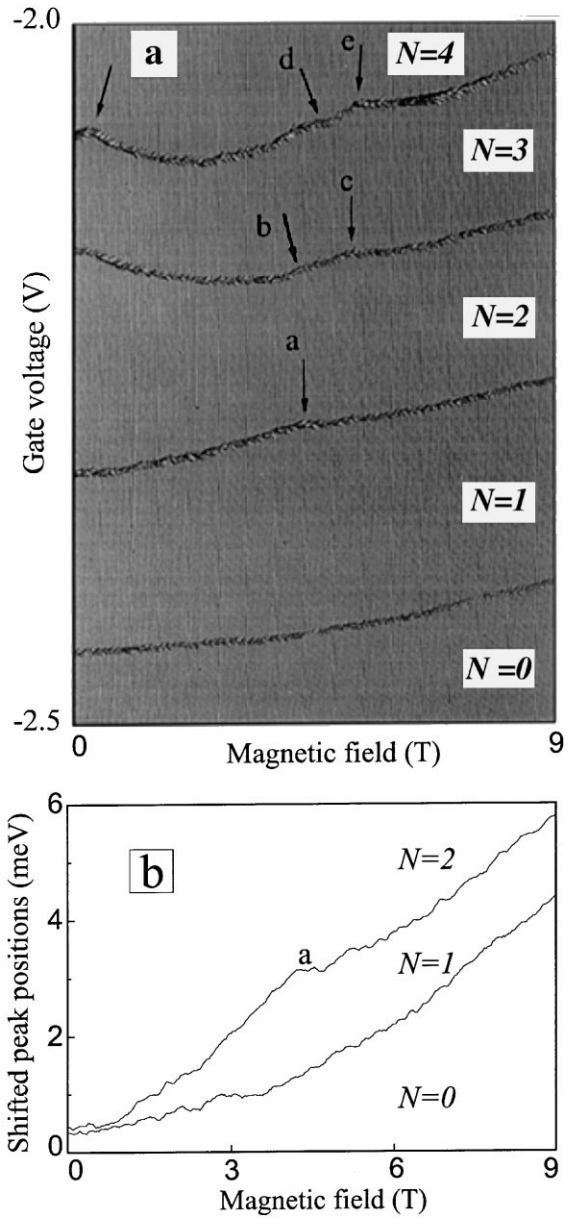

Fig. 2. (a) Current measurement as function of gate voltage and magnetic field (0–9 T in steps of 25 mT) for $N = 1$–4 and $V_{sd} = 30$ μV. The indicated transitions are discussed in the text. (b) Peak positions extracted from the data in (a) and shifted towards each other. Gate voltage is converted to electro-chemical potential.

For larger source-drain voltage, V_{sd}, the current peaks become stripes with a width equal to V_{sd} [4]. In Fig. 3 an $I(V_g, B)$ gray scale plot is given of the

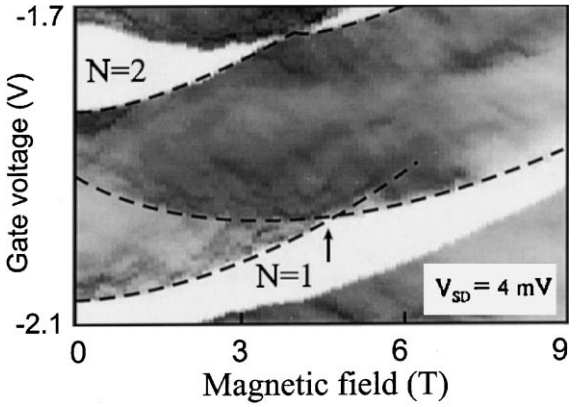

Fig. 3. Gray scale plot of $I(V_g, B)$ for $V_{sd} = 4$ mV. The stripe for the second electron entering the dot is shown. The ground state and first excited state are accentuated by dashed curves. The crossing corresponds to the ST transition.

$N = 2$ stripe measured for $V_{sd} = 4$ mV. The edges of the stripe represent the ground state electro-chemical potential $\mu(2)$, which clearly contain the ST transition at ~4.5 T. Within the stripe, we clearly observe the first excited state. The down-going B-dependence of the first excited state is similar to $\mu^{\uparrow\uparrow}(2)$ (dotted curve in Fig. 1(c)). The crossing of the dashed curves in Fig. 3 is a direct observation of a crossing between the first excited state and ground state, which is in close agreement with Fig. 1(c).

We briefly discuss the $N = 3$ and 4 curves in Fig. 2(a), which both contain two kinks. The left kink (labeled "b") in $\mu(3) = U(3) - U(2)$ is not due to a transition in the energy $U(3)$ of the three electron system, but is a remnant of the two electron ST transition in $U(2)$. The right kink (labeled "c") corresponds to the transition from $U(3) = E_{0,0}(\uparrow) + E_{0,0}(\downarrow) + E_{0,1}(\uparrow) + 3E_c$ to the spin-polarized case $U(3) = E_{0,0}(\uparrow) + E_{0,1}(\uparrow) + E_{0,2}(\uparrow) + 3E_c$. Detailed analysis shows that also this transition to increasing total angular momentum and total spin is driven largely by interactions. Similar transitions occur for the $N = 4$ system where on the right of the last kink (labeled "e") the system is again in a polarized state with sequential filling of the angular momentum states: $U(4) = E_{0,0}(\uparrow) + E_{0,1}(\uparrow) + E_{0,2}(\uparrow) + E_{0,3}(\uparrow) + 6E_c$.

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