

## Spin filling of a quantum dot derived from excited-state spectroscopy

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**Abstract.** We study the spin filling of a semiconductor quantum dot using excited-state spectroscopy in a strong magnetic field. The field is oriented in the plane of the two-dimensional electron gas in which the dot is electrostatically defined. By combining the observation of Zeeman splitting with our knowledge of the absolute number of electrons, we are able to determine the ground state spin configuration for one to five electrons occupying the dot. For four electrons, we find a ground state spin configuration with total spin  $S = 1$ , in agreement with Hund's first rule. The electron  $g$ -factor is observed to be independent of magnetic field and electron number.

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**1. Introduction**

A single electron spin confined in a semiconductor quantum dot (QD) is considered a promising candidate for the implementation of a qubit [1, 2]. Also the joint spin state of  $N$  electrons on a single dot may be used for quantum computation schemes. For instance, as long as the  $N$ -electron ground state has spin  $S = 1/2$ , it can be used in a similar way as the spin of a single electron. Furthermore, proposals exist for encoding a qubit in two specific spin states of three electrons in a single dot, and for controlling this qubit fully electrically [3]. Therefore, it is important to understand the interaction of multiple electron spins confined in a QD, and specifically the spin configuration of the ground state. This can be done by studying spin filling, i.e. by determining the spin of successive electrons that are added to the dot, starting from zero electrons.

QDs defined in pillars etched from a GaAs/AlGaAs double-barrier heterostructure (so-called ‘vertical’ QDs) have been studied extensively, showing spin filling obeying Hund’s rule [4], triplet–singlet ground state transitions [5] and a dependence of spin filling on the anisotropy of the confinement potential [6]. Furthermore, zero magnetic field addition spectra have revealed an atomic-like shell structure induced by a two-dimensional harmonic potential. Therefore, these devices are commonly referred to as ‘artificial atoms’. The addition spectrum of few-electron QDs, defined electrostatically within a two-dimensional electron gas (2DEG) by means of surface gates (so-called ‘lateral’ QDs), has also been studied [7]. Although a two-electron singlet–triplet ground state transition has been observed in these systems [8], evidence for a shell structure and spin filling obeying Hund’s rule has not yet been found.

Here we study the spin filling of a few-electron lateral QD by performing excited-state spectroscopy at a fixed magnetic field  $B_{\parallel}$  of 10 T, applied parallel to the 2DEG. Firstly, we explain in detail our general method for determining spin filling. Then the device characteristics and settings are described. Finally, we apply the method in order to determine spin filling for five successive transitions in the electron number, starting from an empty dot.

## 2. Zeeman splitting and spin filling

The method for determining spin filling is based on the facts that any single orbital can be occupied by at most two electrons, and that these electrons must have antiparallel spins, due to the Pauli exclusion principle. Therefore, as we add one electron to a dot containing  $N$  electrons, there are only two scenarios possible: (I) the electron moves into an empty orbital, or (II) it moves into an orbital that already holds one electron. We will now show that (in a high magnetic field) for the transition from the  $N$ -electron ground state,  $GS(N)$ , to the  $(N + 1)$ -electron ground state,  $GS(N + 1)$ , these two scenarios always correspond to the addition of a spin-up electron and a spin-down electron respectively.

We first consider case I where an electron enters an empty orbital. In a strong magnetic field  $B_{||}$ , spin-up electrons have a lower energy than spin-down electrons<sup>2</sup> due to the Zeeman splitting  $\Delta E_Z = |g|\mu_B B_{||}$ , where  $\mu_B = 58 \mu\text{eV T}^{-1}$  is the Bohr magneton. Therefore, if the orbital is empty, addition of a spin-up electron is energetically favoured and thus takes the dot from  $GS(N)$  to  $GS(N + 1)$ . In contrast, addition of a spin-down electron takes the dot from  $GS(N)$  to the  $(N + 1)$ -electron excited state,  $ES(N + 1)$ , which lies  $\Delta E_Z$  higher in energy.

Next we look at case II, where an electron moves into an orbital with already one electron present. The electron that already occupies the orbital has spin-up if the dot is in  $GS(N)$ , as explained above. Therefore, the electron added in the transition from  $GS(N)$  to  $GS(N + 1)$  must have spin-down in order to satisfy the Pauli exclusion principle. A spin-up electron can only be added to the same orbital if the first electron is spin-down, i.e. when the dot starts from  $ES(N)$ ,  $\Delta E_Z$  higher in energy than  $GS(N)$ . Thus, addition of a spin-up electron corresponds to a transition from  $ES(N)$  to  $GS(N + 1)$ .

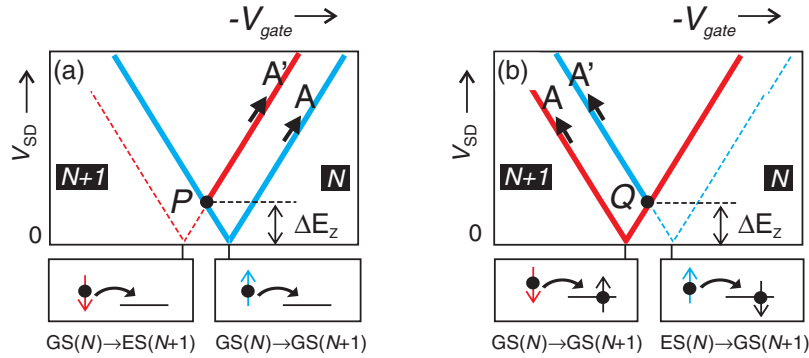
Comparing the two cases, we see that in case I, where a spin-up electron is added, there is an  $(N + 1)$ -electron ES separated from  $GS(N + 1)$  by  $\Delta E_Z$ , while in case II, where a spin-down electron is added, there is an  $N$ -electron ES, separated from  $GS(N)$  by  $\Delta E_Z$ . Thus, the spin filling has a one-to-one correspondence with the excited-state spectrum [9].

We can discriminate between cases I and II by looking at electron transport through the dot as a function of the voltage bias ( $V_{SD}$ ) applied between source and drain contacts, and gate voltage. Figures 1(a) and (b) schematically show the expected result of such a measurement for cases I and II respectively. Lines in the differential conductance  $dI/dV_{SD}$  indicate where electron transitions involving ground and excited states become energetically accessible. The transition from  $GS(N)$  to  $GS(N + 1)$  is only allowed in the V-shaped region spanned by the two solid lines in  $dI/dV_{SD}$  that intersect at  $V_{SD} = 0$ . These lines thus form the edges of the Coulomb blocked (CB) region. The onset of the transition from  $GS(N)$  to  $ES(N + 1)$ , as in case I, appears as a line terminating at the edge of the  $(N + 1)$ -electron CB region, at point  $P$  in figure 1(a). In contrast, the onset of the transition from  $ES(N)$  to  $GS(N + 1)$ , as in case II, appears as a line terminating at the edge of the  $N$ -electron CB region, at point  $Q$  in figure 1(b).

Thus, if we see a line at a distance  $\Delta E_Z$  from the edge of the CB region and terminating at the  $(N + 1)$ -electron CB region, we have case I. Here a spin-up electron is added to the dot. In contrast, if there is a line at a distance  $\Delta E_Z$  from the CB region that terminates at the  $N$ -electron CB region, we have case II, where a spin-down electron is added to the dot.

The main requirement for this method is the ability to identify the Zeeman splitting in the excited-state spectrum. In GaAs/AlGaAs lateral QDs Zeeman splitting has already been observed

<sup>2</sup> Since the  $g$ -factor in GaAs is negative, the electron spin is aligned with the magnetic moment of the electron, and spin-up (aligned with the magnetic field) has a lower energy than spin-down.



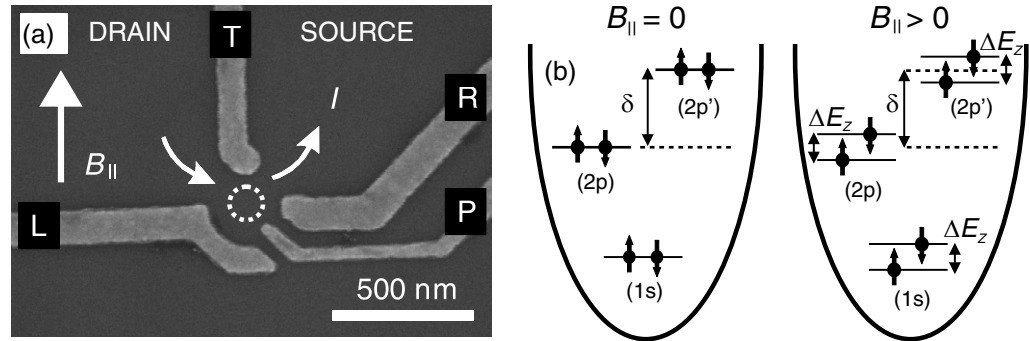
**Figure 1.** Schematic representation of excited-state spectra for case I (a) and case II (b). Shown is the differential conductance  $dI/dV_{SD}$  as a function of  $V_{SD}$  and gate voltage  $V_{gate}$  in the presence of a strong in-plane magnetic field. The lines indicate where the transitions depicted in the corresponding diagrams become energetically accessible. Current can only flow in the V-shaped region defined by the  $GS(N)$  to  $GS(N+1)$  transition (line A); outside this region the dot is in Coulomb blockade and the number of electrons on the dot is fixed. When a transition involving an ES becomes accessible, the current changes, leading to an extra line in  $dI/dV_{SD}$  (line A'). In case I, this line terminates at the  $(N+1)$ -electron CB region (point  $P$  in (a)), whereas in case II the line terminates at the  $N$ -electron CB region (point  $Q$  in (b)). Each of the excited-state spectra is symmetric with respect to  $V_{SD}$ . Therefore, the spectra for  $V_{SD} < 0$  can be obtained by rotating the shown spectra about the  $V_{gate}$  axis.

in several experiments [10]–[13]. We emphasize that the method is valid regardless of the spin  $S$  of the ground states involved, as long as the addition of one electron changes the spin of the ground state by  $|\Delta S| = 1/2$ .<sup>3</sup> We now utilize this method to determine the change in spin at subsequent electron transitions in a lateral GaAs QD containing zero to five electrons.

### 3. Device characteristics

The lateral QD is defined by Ti/Au gate electrodes patterned on top of a Si modulation doped GaAs/ $Al_xGa_{1-x}As$  heterostructure ( $x = 0.265$ ), containing a high mobility 2DEG 60 nm below the surface, see figure 2(a). Annealing of Ni/AuGe/Ni contacts provides electrical contact to the source and drain reservoirs in the 2DEG. The 2DEG has an electron density  $n_s = 4.0 \times 10^{15} \text{ m}^{-2}$ . This sample was cooled down with +266 mV on each of the surface gates in order to reduce background charge fluctuations [15]. The voltage on gate electrode T,  $V_T$ , is used to vary the electrochemical potential of the dot in each of the excited-state spectra. A magnetic field is applied parallel to the 2DEG to minimize additional lateral confinement and to exclude Landau level formation. All measurements are performed in a dilution refrigerator at a base temperature  $T = 15 \text{ mK}$ .

<sup>3</sup> If  $|\Delta S| > 1/2$ , the  $(N+1)$ -electron GS cannot be reached from the  $N$ -electron GS by adding a single electron, leading to a spin blockade of electron transport through the dot, see [14].

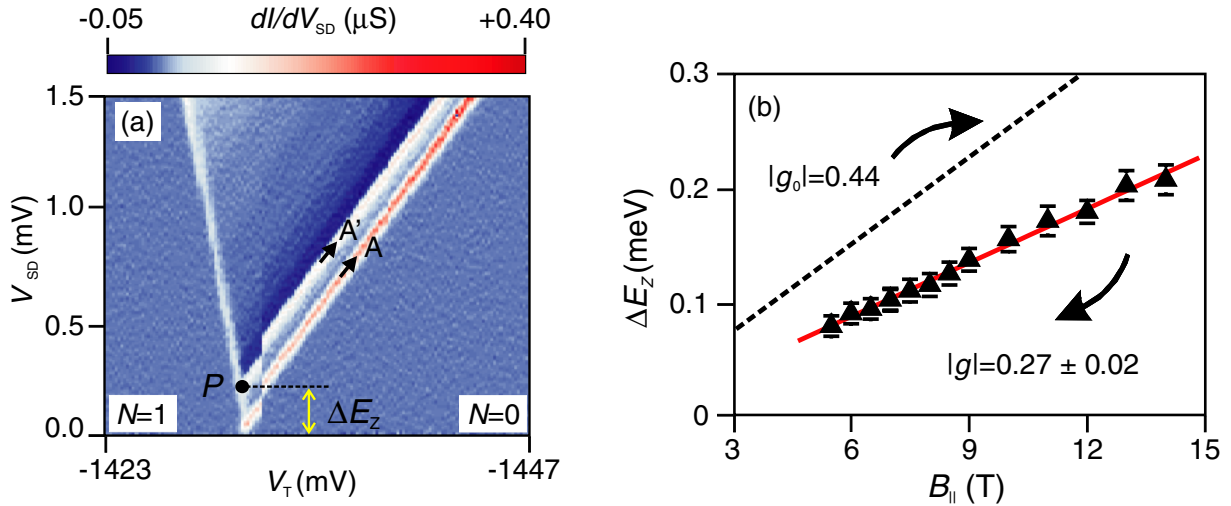


**Figure 2.** (a) Scanning electron micrograph of the device showing Ti/Au gate electrodes lying on top of a GaAs/AlGaAs heterostructure. The gate electrodes can locally deplete the 2DEG when negatively biased. Gate electrodes L, R and T are used to form the QD, indicated by the white dashed circle. Gate electrode P is left floating. (b) Level structure of a QD with anisotropic parabolic confinement in zero and finite magnetic field. Two electrons with opposite spin can occupy the orbital states (1s), (2p) and (2p'). The latter two orbitals are separated in energy by  $\delta$  due to anisotropy of the confinement potential.

We tune the QD to the few-electron regime at  $B_{\parallel} = 0$  T. We identify the  $0 \leftrightarrow 1$  electron transition by the absence of further electron transitions in sweeping the gate voltages to more negative values under large applied source–drain voltage [16]. Then we track the  $0 \leftrightarrow 1$  electron transition as the magnetic field is swept to  $B_{\parallel} = 10$  T.

The charging energy of the dot,  $E_C$ , is 4.8 meV (for adding a second electron to a one-electron dot). For  $N = 1$ , the level spacing from the orbital ground state to the first orbital excited state is 1.7 meV (data not shown). The level spacing between the first and second orbital excited state is considerably smaller, 0.8 meV. This implies that the confinement potential of the dot has no circular symmetry as in [17]. We nevertheless adopt the nomenclature from [16, 17] to denote the lowest orbital states in our QD as (1s), (2p), etc. In fact, we shall see that the data is well explained by assuming an anisotropic confinement potential in the dot, where the twofold orbital degeneracy of the first excited state (2p) is lifted. This gives rise to a level structure as shown in figure 2(b), in which the two (2p)-like orbitals, now denoted (2p) and (2p'), are offset by an amount  $\delta = 0.8$  meV. In lateral QDs, the spacing between successive orbitals is found to be dependent on gate voltage (and thus electron number) [8, 18, 19]; generally, the level spacing decreases as the size of the dot is increased (which is needed to allow more electrons on the dot). Therefore, we also expect the value of  $\delta$  to decrease as we increase the number of electrons on the dot.

We set the tunnel rate of the incoming barrier  $\Gamma_L$  much smaller than the tunnel rate for the outgoing barrier  $\Gamma_R$ . As a result, in all of the excited-state spectra shown, the intensity of the lines involving transitions to or from excited states is enhanced when they run from bottom left to top right. In turn, the intensity of the lines involving excited states and running from top left to bottom right is suppressed [20]. Thus, lines corresponding to transitions from GS( $N$ ) to ES( $N + 1$ ) (line A' in figure 1(a)) are most easily observed for  $V_{SD} > 0$ , while lines corresponding to transitions from ES( $N$ ) to GS( $N + 1$ ) (line A' in figure 1(b)) are most easily seen for  $V_{SD} < 0$ .



**Figure 3.** (a) Colour scale plot of the differential conductance  $dI/dV_{SD}$  as a function of  $V_{SD}$  and gate voltage  $V_T$  near the  $0 \leftrightarrow 1$  electron transition, at  $B_{||} = 10$  T. Zeeman splitting of the orbital ground state is clearly observed (A–A′). The vertical shift in the data near  $V_T = -1430$  mV is caused by a background charge rearrangement in the environment of the dot. (b) Extracted Zeeman splitting  $\Delta E_Z$  at the  $0 \leftrightarrow 1$  electron transition as a function of  $B_{||}$ . A linear fit of  $\Delta E_Z$  (red curve) results in  $|g| = 0.27 \pm 0.02$ . The dashed black line corresponds to the Zeeman splitting in bulk GaAs, where  $g_0 = -0.44$ .

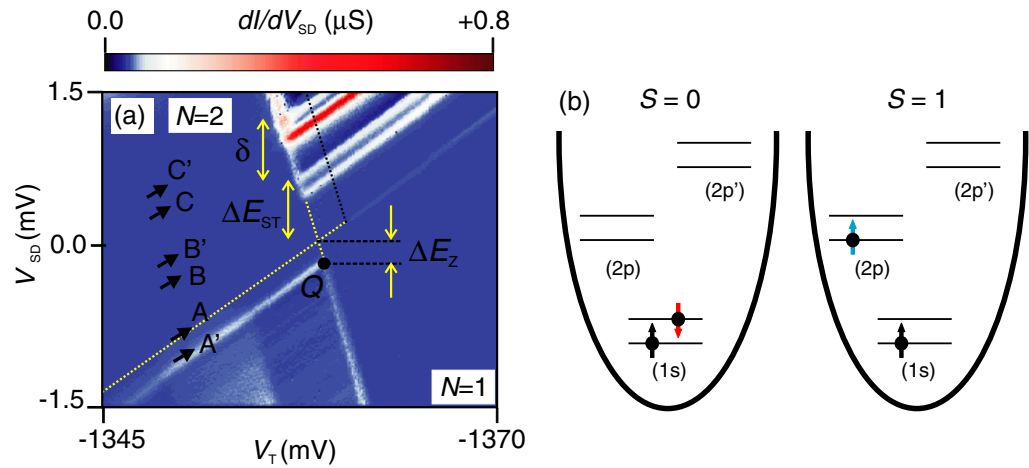
#### 4. $N=0 \leftrightarrow 1$ transition

The excited-state spectrum obtained around the  $0 \leftrightarrow 1$  transition is shown in figure 3(a). Clearly two parallel lines are observed, A and A′. The separation between these lines increases linearly with  $B_{||}$ , and thus corresponds to the Zeeman splitting. From the spacing between lines A and A′ (to be precise, from the value of  $V_{SD}$  at point P), we extract  $\Delta E_Z = 0.16 \pm 0.01$  meV at 10 T. Since A′ terminates in the  $N = 1$  CB region, the electron added to the empty dot to form the  $N = 1$  GS has spin-up (see figure 1(a)), as expected.

In figure 3(b), the Zeeman energy  $\Delta E_Z$  at the  $0 \leftrightarrow 1$  transition is plotted versus applied magnetic field in the range from 5.5 to 14 T. For  $B_{||} < 5.5$  T, we cannot clearly resolve the Zeeman splitting from the spectroscopy data. As a reference, the Zeeman splitting  $\Delta E_Z$  expected for bulk GaAs is plotted with  $|g_0| = 0.44$  [21] (dashed black line). Clearly, the  $g$ -factor we extract from figure 3(b) is independent of field. Linear fitting of the data points results in a  $g$ -factor value of  $0.27 \pm 0.02$ . This value is lower than the bulk value of GaAs. Deviations of the  $g$ -factor from  $g_0$  in QDs and possible explanations for this effect have been reported before [10]–[12].

#### 5. $N=1 \leftrightarrow 2$ transition

Next we tune the dot to the  $1 \leftrightarrow 2$  electron transition. At zero magnetic field, the ground state for a two-electron QD is always a spin-singlet state  $|S\rangle$ , where two electrons with opposite spin occupy the lowest orbital and the total spin  $S = 0$  [22]. We expect that this is still true for an



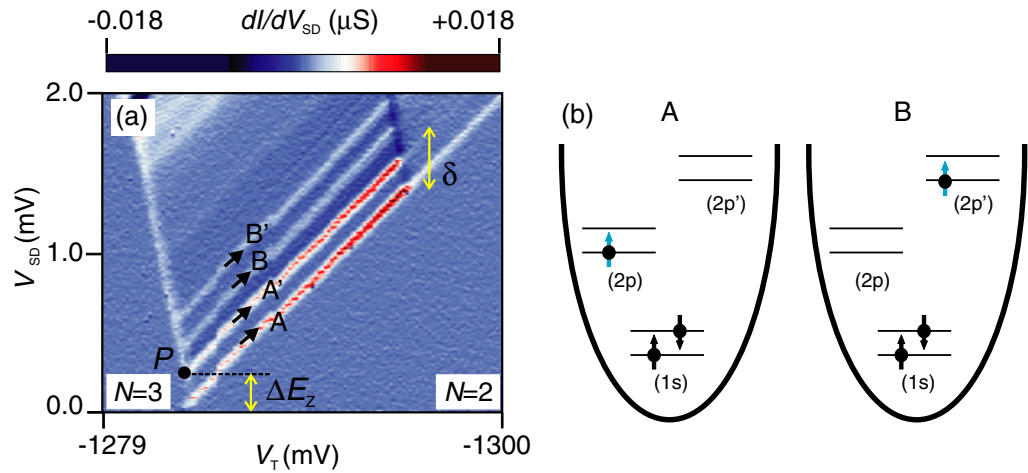
**Figure 4.** (a) Colour scale plot of the differential conductance  $dI/dV_{SD}$  as a function of  $V_{SD}$  and gate voltage  $V_T$  near the  $1 \leftrightarrow 2$  electron transition, at  $B_{\parallel} = 10$  T. (b) Left: ground state spin configuration for a two-electron dot. A spin-down electron (red) is added and pairs with the spin-up electron already present, to form a singlet state  $|S\rangle$  with total spin  $S = 0$ . Right: spin configuration of an excited state. The second spin-up electron occupies the  $(2p)$  orbital to form a two-electron triplet state  $|T\rangle$  with total spin  $S = 1$ . When the second spin-up electron occupies the  $(2p')$  orbital an excited triplet state is formed.

in-plane field of 10 T, as the Zeeman energy is much smaller than the zero-field singlet–triplet energy separation [13].

The  $dI/dV_{SD}$  data obtained for the  $1 \leftrightarrow 2$  electron transition is shown in figure 4(a). Two lines A and A' separated by  $0.16 \pm 0.01$  meV are visible for  $V_{SD} < 0$ . This is exactly the same energy scale of the Zeeman splitting found for the  $0 \leftrightarrow 1$  transition. Because A' terminates in Q at the edge of the  $N = 1$  CB region (and not at the edge of the  $N = 2$  CB region), we conclude that the transition from GS(1) to the GS(2) involves adding a spin-down electron. The spin-down electron pairs with the spin-up electron already present in the  $(1s)$  orbital to form a two-electron singlet state  $|S\rangle$ , as illustrated in the left panel of figure 4(b).

For the colour scale chosen, parts of line A are difficult to observe. Therefore, dashed yellow lines are added as a guide to the eye. For  $V_{SD} > 0$ , line A' is also hardly visible. Its position is indicated by a black dashed line. The absence of this latter line is caused by the asymmetry of the tunnel barriers, as explained earlier. For the same reason, the lines B–B' and C–C' are hardly visible for  $V_{SD} < 0$ . Line A is weaker than line A', because in a strong magnetic field spin-up electrons generally couple better to the source and drain reservoirs than spin-down electrons, even if the magnetic field is applied in the plane of 2DEG [23].

The lines B–B' correspond to the onset of transitions involving the three triplet states. These lines too are separated by the Zeeman splitting; B and B' involve transport of spin-up and spin-down electrons respectively [13]. The distance between A and B' gives the singlet–triplet energy splitting  $E_{ST}$ ,  $0.56 \pm 0.02$  meV. The data also shows a third set of parallel lines with the same spacing, C–C'. The fact that the pair of lines C–C' have a different intensity than the pair of lines B–B' suggests that different orbitals are involved. We believe that the lines C–C' correspond to transitions to and from triplet states with one electron in the  $(2p')$  orbital instead of in the  $(2p)$



**Figure 5.** (a) Colour scale plot of the differential conductance  $dI/dV_{SD}$  as a function of  $V_{SD}$  and gate voltage  $V_T$  near the  $2 \leftrightarrow 3$  electron transition, at  $B_{\parallel} = 10$  T. (b) Left: ground state spin configuration for a three-electron dot. Right: spin configuration of the excited state that can be reached starting at line B in (a).

orbital. Here, the offset between the (2p) and (2p') orbitals,  $\delta$ , is 0.52 meV (B'–C'), somewhat smaller than the value at the  $0 \leftrightarrow 1$  electron transition.

## 6. $N = 2 \leftrightarrow 3$ transition

Next we move on to the  $2 \leftrightarrow 3$  electron transition. We have seen that the two-electron ground state is a singlet state  $|S\rangle$ , where two electrons with opposite spin occupy the (1s) orbital. When a third electron is added, it has to occupy a different orbital in order to satisfy the Pauli exclusion principle.

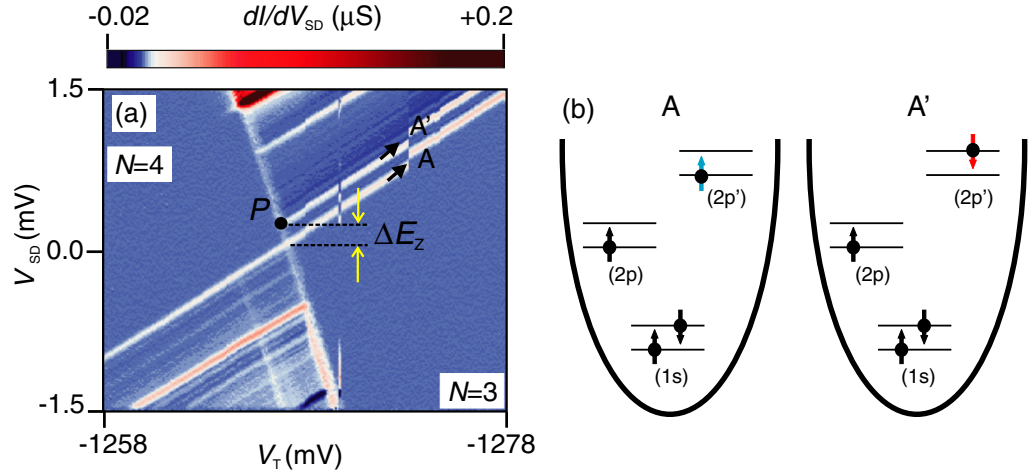
The  $dI/dV_{SD}$  data we find is shown in figure 5(a). We notice several important features. A first pair of lines, A–A', is split by  $0.17 \pm 0.01$  meV, the Zeeman splitting. Since these lines terminate at the edge of the  $N = 3$  CB region, the transition from GS(2) to GS(3) involves adding a spin-up electron. The three-electron ground state then corresponds to the situation where the third electron (with spin-up) occupies the orbital (2p), as shown in the left panel of figure 3(b).

A second set of Zeeman split lines, B–B', runs parallel to the lines A and A', with smaller amplitude. The separation between both sets of transitions,  $\delta$ , is 0.34 meV. In our spin filling picture, the lines B and B' correspond to electron transitions where the third electron occupies the (2p') orbital, as in the right panel of figure 3(b). The value of  $\delta$  we find here is smaller than at the  $1 \leftrightarrow 2$  electron transition, as expected from our earlier considerations.

## 7. $N = 3 \leftrightarrow 4$ transition

In figure 6(a), we show the excited-state spectrum for the  $3 \leftrightarrow 4$  electron transition. Ignoring spin-exchange interactions, one expects a four-electron dot with total spin  $S = 0$ , where two electrons occupy the (1s) orbital and two electrons occupy the (2p) orbital. This implies that a spin-down electron must be added to a three-electron dot in the GS in order to reach the  $N = 4$ ,  $S = 0$  state.





**Figure 6.** (a) Colour scale plot of the differential conductance  $dI/dV_{SD}$  as a function of  $V_{SD}$  and gate voltage  $V_T$  near the  $3 \leftrightarrow 4$  electron transition, at  $B_{\parallel} = 10$  T. (b) Left: ground state spin configuration of the four-electron dot ( $S = 1$ ). Right: spin configuration of one possible spin-excited state, where the  $(2p')$  orbital is occupied by a spin-down electron.

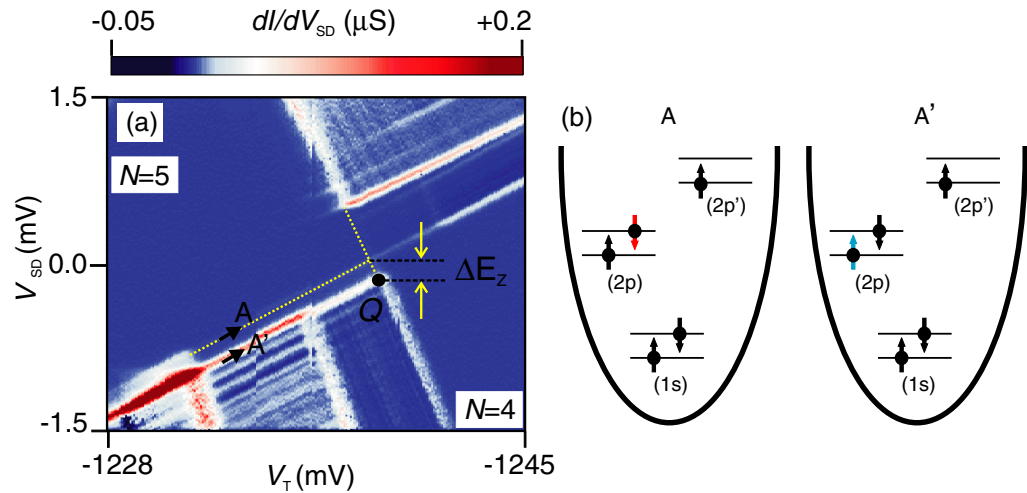
However, the data in figure 6(a) indicates that the fourth electron added to the dot has spin-up, because the line  $A'$ , separated from line  $A$  by the Zeeman splitting ( $\Delta E_Z = 0.17 \pm 0.01$  meV), terminates at the edge of the  $N = 4$  CB region. Since both the third and fourth electron have spin-up, they occupy different orbitals,  $(2p)$  and  $(2p')$ , as shown in the left panel of figure 6(b). The  $N = 4$  ground state thus has  $S = 1$ . This  $N = 4$  ground state can be understood when we take into account the exchange interaction  $K_{ab}$  between the spins in the  $(2p)$  and  $(2p')$  orbital and the terms  $C_{aa}$  and  $C_{ab}$ , representing the direct Coulomb energy when the two spins are in the same or in a different orbital state respectively [16].  $S = 1$  spin filling is favoured when  $K_{ab} + |C_{aa} - C_{ab}| > \delta$ .

This particular spin configuration of the  $N = 4$  ground state is related to Hund's first rule, which states that a shell of degenerate orbitals will, as much as possible, be filled by electrons with parallel spins, up to the point where the shell is half filled. Exchange energy  $K_{ab}$  (causing a lowering of the Coulomb energy when spins align parallel in different orbitals) reduces the total energy and favours the  $S = 1$  state. This state can only exist when the offset between the  $(2p)$  orbitals,  $\delta$ , is small compared to the exchange energy and the difference in direct Coulomb energy terms [6].

## 8. $N = 4 \leftrightarrow 5$ transition

As we have seen, the four-electron ground state has a total spin  $S = 1$ , in agreement with Hund's first rule. When the fifth electron is added, the total spin of the system is expected to change back from  $S = 1$  to  $S = 1/2$  because of spin pairing in the  $(2p)$  orbital. A less likely option for the transition from GS(4) to GS(5) is that the fifth electron tunnels into the next empty orbital.

In figure 7(a), we show the excited-state spectrum for the  $4 \leftrightarrow 5$  electron transition. Indeed, the transition from GS(4) to GS(5) corresponds to adding an electron with spin-down, as the line  $A'$  terminates at the edge of the  $N = 4$  CB region (as before, some of the lines are hardly visible;



**Figure 7.** (a) Colour scale plot of the differential conductance  $dI/dV_{SD}$  as a function of  $V_{SD}$  and gate voltage  $V_T$  near the  $4 \leftrightarrow 5$  electron transition, at  $B_{\parallel} = 10$  T. (b) Left: ground state spin configuration of a five-electron dot. Right: spin configuration of the spin-excited state. In both cases, the total spin is  $S = 1/2$ .

their position is indicated by yellow dashed lines). The resulting spin configuration for five electrons is indicated in the left panel of figure 7(b). The added spin-down electron occupies the (2p) orbital. The Zeeman splitting extracted from the data ( $V_{SD} < 0$ ) is  $\Delta E_Z = 0.17 \pm 0.01$  meV.

## 9. Conclusion

We have determined the spin filling of a few-electron lateral QD containing one up to five electrons in a parallel magnetic field of 10 T. The spin filling was extracted without magnetic field sweeps, by looking at the position of the excited spin state in the spectroscopy data. The Zeeman splitting is equal for all orbitals and independent of the number of electrons on the dot. The ground state of a four-electron dot is a  $S = 1$  Hund state. Field dependence of the Zeeman splitting for the  $0 \leftrightarrow 1$  electron transition yields a field-independent  $g$ -factor value of  $0.27 \pm 0.02$ .

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