

Fate of Spin Doublets in Quantum Dot with Many Interacting Electrons.

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Abstract:

Using the Single Electron Capacitance Spectroscopy, we study the energies required to add electrons to a quantum dot in a broad range of electron occupancy N . Following evolution of these energies as a function of magnetic field, we unveil the nature of the states occupied by electrons. For small N electrons fill states, which resemble one-electron energy levels in a parabolic confinement. When N is large these states are drastically altered by increased Coulomb repulsion between electrons. We present a new and unified picture of the evolution from small to large N regimes..

The behavior of a single electron in a potential well is rather simple. It can be described in basic terms of single electron energy levels. However, for many electrons confined together the strong Coulomb repulsion between them makes this new many body problem enormously complicated. The rich physics in this regime is governed by the interplay of the two energies scales: the single particle level spacing and Coulomb interaction. The two scales depend on the size of the dot and the electron density, and typically do not vary significantly for small changes in the number of electrons N .

For a small N in a sufficiently steep potential well (or small quantum dot) the confinement energy dominates the interaction energy. Single particle energies in this case resemble spatial quantization levels of *one* electron in the potential well. Noticeably, two electrons with opposite spin consecutively occupy the same spatial state. For that reason the energies required to add an odd $\mu(2k-1)$ and a consecutive even $\mu(2k)$ electron show similar behavior in magnetic field, specific to that spatial state. First, this behavior was predicted (1) and observed in several experiments in small vertical (2-4) and more recently small lateral dots (5-7) containing just a few electrons. While most features in the small dot spectra can be roughly described within the framework of the one electron levels picture (8), some experiments indicate on importance of electron interactions at least under certain conditions. Several interaction-driven singlet-triplet transition were observed in small dots (2,4,9).

Coulomb interactions become comparable to single particle spacing for larger dots with substantial N . In these dots consecutive electrons do not necessarily fill the same spatial state with anti-parallel spins (10,11). Instead the small single particle level spacing allows for a ground state with a significant spin build-up (12-14). At high magnetic field the addition energies $\mu(B)$ demonstrate a saw-tooth behavior, as the electrons swing between the two lowest orbital Landau levels to minimize their repulsion energy (15,16). From this behavior, no similarity between energies $\mu(B)$ for adding odd and even electrons can be discerned. Recently, it has been shown that when electrons in a quantum dot completely fill the two lowest Landau levels, the interactions cause a significant spin build-up above some critical N (17).

Our investigation bridges the gap between the studies of small and large dots. The experimental technique – Single Electron Capacitance Spectroscopy (18) permits examination of dots' addition spectra while tuning the electron occupancy N from one to hundreds in *one* sample. To probe the nature of the states occupied by electrons, we follow the evolution of the addition spectrum with perpendicular magnetic field B . First few electrons fill well-known Darwin-Fock (DF) single-particle states as is typical for a small dot with parabolic confinement (3,8). These levels exhibit multiple crossing as B grows. We find that in the vicinity of *every* crossing, the electron interaction becomes the dominant energy scale, even in a small dot for a small number of electrons. Thus, *four* successively added electrons redistribute between *two* different crossing spatial states. We describe the observed resulting patterns by simple Hartree-Fock arguments. By gradually filling the dot with electrons we decrease the single particle level separation, hence increasing the relative importance of the Coulomb repulsion. As the result, the regions of the spectrum strongly affected by the interactions expand, until at high enough N no signature of DF states remains observable. Our study presents a unifying picture for the two limits: the small N regime with dominant confinement and the large N regime with prevailing interactions.

The dots were fabricated within an AlGaAs/GaAs heterostructure as described in previous work (18). The essential layers (from bottom to top) are a conducting layer of GaAs serving as the only contact to the dots, a shallow AlGaAs tunnel barrier, a GaAs active layer that contains the dots, and an AlGaAs blocking layer. On the top surface, we produce a small AuCr top gate using electron beam lithography. This top gate was used as a mask for reactive ion etching that completely depletes the active GaAs layer in the regions away from the AuCr gate. A larger overlapping metal electrode then provides electrical connection to the gate (a schematic of our samples is shown in the Fig.1A).

The measurements are carried out using on-chip bridge circuit described in (18,19). To register electron additions, we monitor the a.c. capacitive response to a small ($<80\mu V$) a.c. excitation applied between the top gate and the contact layer. At gate voltage values corresponding to the electron additions, an electron oscillates between the dot and the contact increasing the measured a.c. capacitance and resulting in a peak (18) in our measurements.

The greyscale panel in Fig.1B expands a part of the addition spectrum between 9th and 15th electrons. Each successive trace corresponds to the energy for adding an extra electron to the dot. The data shown were taken on an elliptical dot with bare parabolic confinement with $\omega_x=2.78mV$; $\omega_y=8.33mV$. The lateral dimensions of the electron puddle ($L_x\approx 200nm$; $L_y\approx 70nm$ for $N\approx 20$) are much smaller than the lithographic size of the dot. Such a large lateral depletion of the electron gas together with small lithographic dimensions ensure the parabolicity of the confining potential in both x and y . Thus, the overall spectral features can be described qualitatively within the constant interaction model for DF states, as is typical for small elliptical dots. Quantum numbers $[n_x, n_y]$ can be assigned to the orbital DF states (1,20) and some are shown on Fig.1. Addition energies “wobble” with magnetic field as different orbital states become the ground state of the dot. Pairs formed by each odd and consecutive even Coulomb blockade traces (marked by arrows), show mostly similar oscillatory behavior. This similarity arises because of the paired electrons entering the same spatial state but with opposite spins. However, more careful inspection of the data shows features that the simple model does not explain (marked by ovals).

Each oval encases a region on V_g - B plane containing *four* electron additions to *two* crossing orbitals. Consider electron addition traces №11-14 in the magnetic field range of 1.8-2.5T (second lowest oval). The 11th trace forms a peak at $B_0=2.2T$, as the electron jumps from the orbital $[0,1]$ for $B<B_0$ (moving upwards with B) to the orbital $[6,0]$ for $B>B_0$ (moving downwards with B). The 12th trace mostly follows this trend, except for a small vicinity of B_0 , where it forms a small asymmetrical V-notch. Intriguingly, the 13th trace develops an inverse feature at slightly different value of B : Λ -notch at $B_0^*=2T$. Finally, a dip at the 14th trace occurs again at the same $B_0^*=2T$, slightly shifted from the peak at $B_0=2.2T$ in the 11th trace. Neither this misalignment nor the existence of the V and Λ notches is present in the single-electron *non-interacting* picture (21), and therefore both are the result of the Coulomb interactions. Note, that some of these features, namely V-notches at even traces occurring only for some crossing orbitals were reported in (9).

To analyze the observed behavior in greater details we plot the addition energies of the four electrons №11-14 as a function of magnetic field in the range 1.8-2.5T in Fig.1C. Here, constant energies of 98.8; 103.6; 107.8; 111.9 meV were subtracted from

the traces, so that to facilitate their comparison. Observe that the additional features in the 12^{th} and 13^{th} traces are complementary: the Λ -notch in the 12^{th} trace perfectly matches the V-notch in the 13^{th} trace. Moreover, the overall perfect point symmetry of the plot calls for an explanation involving all *four* electrons. So we adopt a simple Hartree-Fock arguments that were put forward in (22) to explain a mechanism for internal spin-flip transitions in armchair carbon nanotubes.

We denote the bare single particle states $[0,1]$ and $[6,0]$ in our elliptical DF level sequence as a and b . At $B_0=2.2T$, the two levels E_a and E_b cross, and correspondingly the 11^{th} electron fills the orbital a for $B<B_0$ and the orbital b for $B>B_0$. The 12^{th} electron follows the 11^{th} away from B_0 . On the contrary, in the very vicinity of B_0 the 12^{th} electron avoids the Coulomb repulsion with the 11^{th} by filling a different higher energy orbital. This becomes possible as the two orbital a and b are nearly degenerate, and thus the reduction of the Coulomb repulsion is greater than the loss in single particle energy $|E_b-E_a|$. In particular, for $B_1<B<B_0$, the 12^{th} electron fills the orbital b . As we will show below the two electrons furthermore lower their energy by forming the triplet spin state. In the Hartree-Fock terms, the addition energy for the 12^{th} electron in this case is $E_b(B)+V_{ab}-J_{ab}$, where V_{ij} and J_{ij} are direct and exchange matrix elements for the electrons on the orbitals i and j . In determining B_1 , one has to equal this expression taken at B_1 : $E_b(B_1)+V_{ab}-J_{ab}$ to $E_a(B_1)+V_{aa}$, - the addition energy required to doubly occupy the orbital a . Similarly, at $B_0<B<B_2$, the 12^{th} electron is added to the orbital a , and B_2 is determined from the condition $E_b(B_2)+V_{bb}=E_a(B_2)+V_{ab}-J_{ab}$. Based on the data we assume that in the considered magnetic field range both V_{ij} and J_{ij} are field independent.

Consider now the behavior of the 13^{th} electron. For $B<B_1$ ($B>B_2$) the orbital a (b) is doubly occupied, and this electron is inevitably added to the lowest empty orbital b (a). In the range of magnetic field $B_1<B<B_2$ the 11^{th} and 12^{th} electrons each singly occupy the orbitals a and b . The 13^{th} electron will join one of them with the addition energy of $E_a+V_{aa}+V_{ab}+J_{ab}$ for $B_1<B<B_0^*$ or $E_b+V_{bb}+V_{ab}+J_{ab}$ for $B_0^*<B<B_2$. Transition of the 13^{th} electron between orbitals a and b occurs at B_0^* , which can be determined from: $E_a(B_0^*)+V_{aa}+V_{ab}+J_{ab}=E_b(B_0^*)+V_{bb}+V_{ab}+J_{ab}$. Since V_{aa} is not equal to V_{bb} , the transition point $B_0^*=2.0T$ does not coincide with $B_0=2.2T$. As a result of the transition the 13^{th} trace exhibits slopes inherent to the orbitals a (b) for $B_1<B<B_0^*$ ($B_0^*<B<B_2$), thus forming the experimentally observed Λ -notch. Finally, the last electron to fill the two orbitals finds the orbital a (b) doubly occupied and the orbital b (a) singly occupied for $B<B_0^*$ ($B>B_0^*$). Hence, the 14^{th} trace has a single dip centered at B_0^* . All the matrix elements V_{ij} and J_{ij} can be directly measured from the data as shown on Fig.1C. Note that $|B_1-B_0^*|=|B_2-B_0|$ ($B_1=1.9T$ $B_2=2.3T$). This equality explains the point symmetry of Fig.1C.

These simple arguments describing rearrangement of *four* successively added electrons between *two* different crossing spatial states capture every feature of the experimentally observed behavior. We stress, that the single particle picture can account for none of these features (21). The traces described above are not the only set exhibiting notches. Several other pairs of complementary “V” and “ Λ ” notches are marked on Fig.1B. Notice the 10^{th} and 11^{th} traces at $B=1.7T$ (note the deviation from 9^{th} trace) and on the 14^{th} and 15^{th} traces at $B=1.4T$ and $2.5T$. In fact, similar pairs of notches appear at every orbital crossing.

To examine the addition spectrum in its entirety we collapse addition traces for electrons from 7^{th} to 23^{rd} by shifting them in the gate voltage (Fig.1D). The subtracted

voltage values are chosen to allow neighboring traces touch. This means that the gaps marked as $(V_{ab} \pm J_{ab})$ on Fig.1C are eliminated. In shifting each trace we aimed to match the rightmost feature on that trace (near $\nu=2$). It appears that all features can be fit with one number. The plot shows a very ordered pattern of single-particle DF orbitals. Each single-particle state can be followed through various pieces of different addition traces. The V-notch and Λ -notch on the traces $\mathcal{N}\{12;13\}$ discussed above form a diamond at $B \approx 2.1T$. More diamonds formed by similar pairs of notches are seen at *every* orbital crossings. Note three pairs of half-notches at zero field comprised by the pairs of traces $\mathcal{N}\{8,9\};\{12,13\};\{18,19\}$. Two deviations from this rule occurs for the pairs of traces $\mathcal{N}\{16,17\}$ and $\mathcal{N}\{20,21\}$ at around $B \approx 0.7T$ and $B \approx 0.85T$ correspondingly. Instead of complementary notches these pairs of traces display smooth anticrossings. Careful examination of the picture actually reveals a duplicated set of DF orbitals. This is because the V and Λ notches prevent us from positioning even addition traces exactly on top of the odd ones when collapsing traces in the gate voltage.

Two important conditions become obvious from the plot: the rearrangement of electrons appear near any *degeneracy* points of any *two* orthogonal orbitals. First, when two states are nearly degenerate the loss in single particle energy $|E_i - E_j|$ is diminishing and become smaller than the gain in the Coulomb repulsion $|V_{ii} - (V_{ij} - J_{ij})|$, which is large for orthogonal states. On the other hand, when the degeneracy is lifted for two mixed orbitals, the two states separate far in energy, while the would-be Coulomb gain approaches zero for mixed states ($\mathcal{N}\{16,17\}$ and $\mathcal{N}\{20,21\}$). The single-particle states in our dot are orthogonal (and, therefore, can be potentially degenerate) either for the strong confinement potential (relatively small N) or sufficiently high magnetic field. Second, in the range shown on the plot only *two* states participate in a formation of notches. The slope in magnetic field allows us to identify precisely a given orbital state. Since, in the vicinity of every diamond structure, we see only two slopes, we conclude that only two orbitals are involved, i.e. *four* electrons rearrange for the optimal filling of only *two* orbitals.

In our highly symmetrical dots the wavefunctions of different single particle states have very small overlap. Thus the direct Coulomb repulsion between two states i and j is much stronger than the exchange interaction ($V_{ij} > J_{ij}$). Still the exchange contribution to the interaction is measurable in our experiment. In Fig.2A, we plot the gate voltage values that we subtracted from addition traces ($V_{ij} \pm J_{ij}$) to collapse them on Fig.1D. All values are relative to that of the 9th trace. As electrons entering the dot gradually screen the confinement potential, each next electron added sees more relaxed confinement due to this screening. As a result electrons spread over the larger area reducing the Coulomb repulsion V_{ij} . This explains the general decrease seen in the plotted spacings, which can be accounted for by subtraction of the smooth background. Fig.2B plots the spacings after a smooth quadratic background is removed. The spacing for even electrons are $V_{ij} - J_{ij}$ while those for odd numbers are $V_{ij} + J_{ij}$.

The apparent alternation of spacings on the plot indicate on observable nonzero contribution of exchange interaction, allowing us to deduce the spin configuration of the dot for various gate voltages and magnetic fields. The resulting color spin maps in V_g - B space are shown on Fig.2C and 2D. The data shown repeats that presented on Fig.1C and 1D. The spin maps show multiple but isolated regions of $s=1$ spin build-up. Spin never

exceeds one, as it requires rearrangement of electrons on more than two orbitals, which does not happen in the V_g - B range shown.

Finally, Fig.3 illustrates evolution of the addition traces with increasing N . The V-notches on the even and the Λ -notches on the odd traces become more pronounced for higher N , eroding the similarity between even and odd traces. Essentially the notches expand to leave double the number of “wiggles” on addition traces occurring for magnetic fields corresponding to $2 < \nu < 4$. The even and odd traces become completely indistinguishable when N reaches about 60 in our dots. The shown traces №{61-65} appear remarkably similar to those of a highly populated dot in high magnetic field (15,16). And quantum dots in this regime are known to be spin polarized (15,16). We speculate that for N somewhere in between $25 < N < 60$ interplay of diminishing single particle separations and growing Coulomb interactions results into involvement of more than two orbital states in sequential filling of the dot. Thus, more complex spin configurations become possible leading to eventual spin polarization of the dot.

Indirect evidence of this effect can be obtained from the slopes of the addition traces in magnetic field for $\nu < 2$. Note that because of the small effective mass and small Lande g factor in GaAs, the Zeeman splitting is generally an order of magnitude smaller than changes in spatial energies. But for magnetic fields corresponding $\nu < 2$ every spatial state is a smooth function of the field and the effect of Zeeman splitting appears in alternating slopes seen on Fig.1D. The alternating slopes are the sign of consecutive spin-up-spin-down filling of the dot. We observe that in our dots of a similar size a regular pattern of alternating slopes abruptly breaks around $N \approx 35-40$ indicating on a possible spin polarization.

In conclusion, we study the energies required to add electrons to a quantum dot while tuning the electron occupancy N from one to hundreds in *one* sample. To probe the nature of the states occupied by electrons, we follow the evolution of the addition spectrum with perpendicular magnetic field B . We find that for a small N electrons fill states, which resemble one electron energy levels in a parabolic confinement, which exhibit multiple crossing as B grows. We find that in the vicinity of *every* crossing, electron interaction becomes the dominant energy scale. Thus, *four* successively added electrons redistribute between *two* different crossing spatial states. We describe the observed resulting patterns by simple Hartree-Fock arguments. By gradually filling the dot with electrons we decrease the single particle level separation, hence increasing the relative importance of the Coulomb repulsion. As the result, the regions of the spectrum strongly affected by the interactions expand, until at high enough N no signature of DF states remains observable. Our study presents a unifying picture for the two limits: the small N regime with dominant confinement and the large N regime with prevailing interactions.

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21. In non-interacting picture both electrons №11-12 occupy orbital $[0,1]$ for $B < B_0$ and orbital $[6,0]$ for $B > B_0$, while electrons №13-14 complementary occupy $(6,0)$ state for $B < B_0$ and $(0,1)$ state for $B > B_0$. Therefore, the traces №12(14) would be identical to №11(13).
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23. We are grateful to H.U. Baranger, B.L. Halperin, K.A. Matveev and Y. Oreg for illuminating discussions. Expert etching of samples was performed by S.J. Pearton.

Figure captions:

Figure1

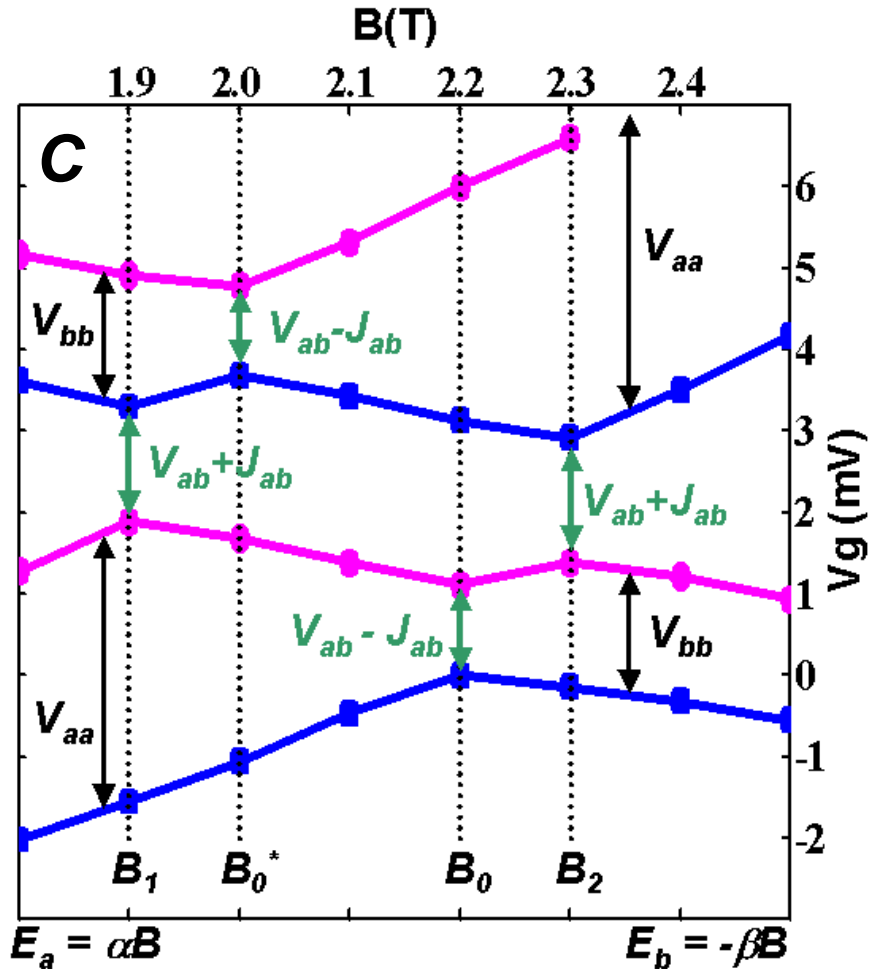
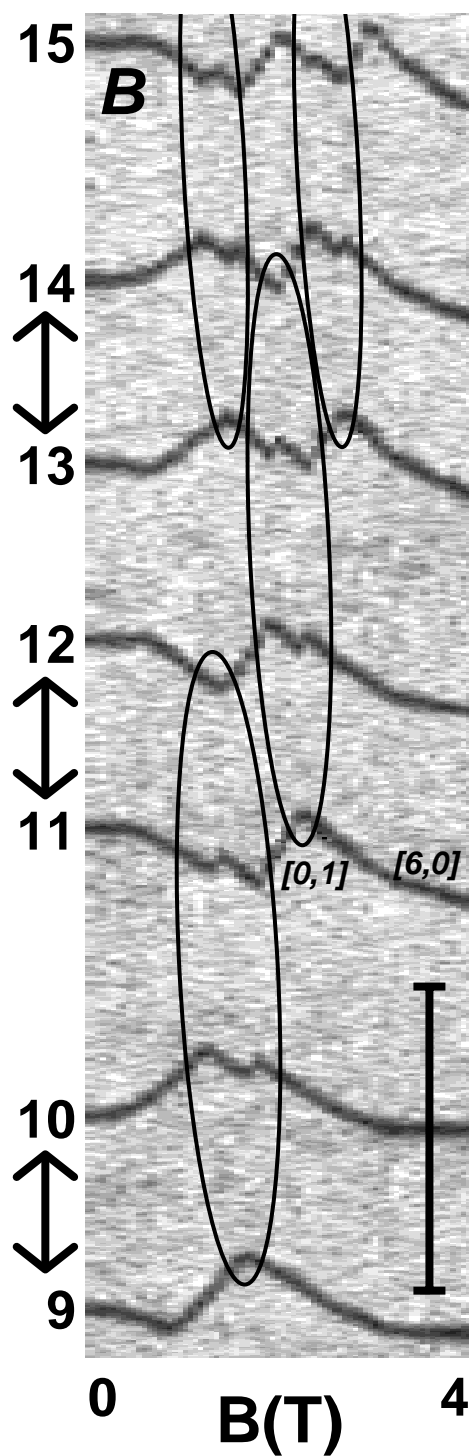
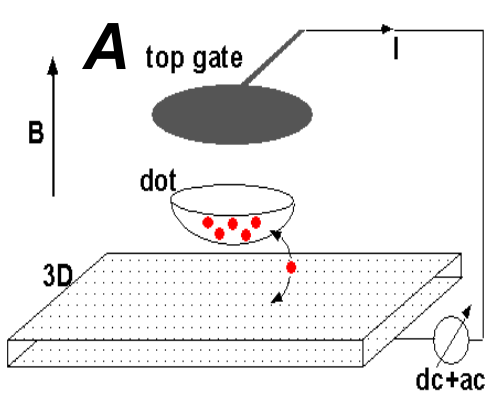
(A) Sample schematic. (B) Greyscale panel of measured capacitance with dark lines denoting capacitance peaks. Each peak corresponds to an electron addition to the dot. Electron addition are numbered. Double-headed arrows mark additions to the same spatial state. Vertical bar at the lower right corner corresponds to $10mV$ in the gate voltage. Gate voltage scale can be converted to the energy scale by dividing by two. Ovals indicate on deviations from the DF states. (C) Addition traces №11;12;13;14, shifted by $98.8;103.6;107.8;111.9mV$ Odd in blue, even in magenta. Symbols are data. Lines are guides to an eye. (D) Collapsed addition traces №7-23. Odd in blue, even in magenta. Gaps marked as $(V_{ab}\pm J_{ab})$ on Fig1.C are eliminated.

Figure2

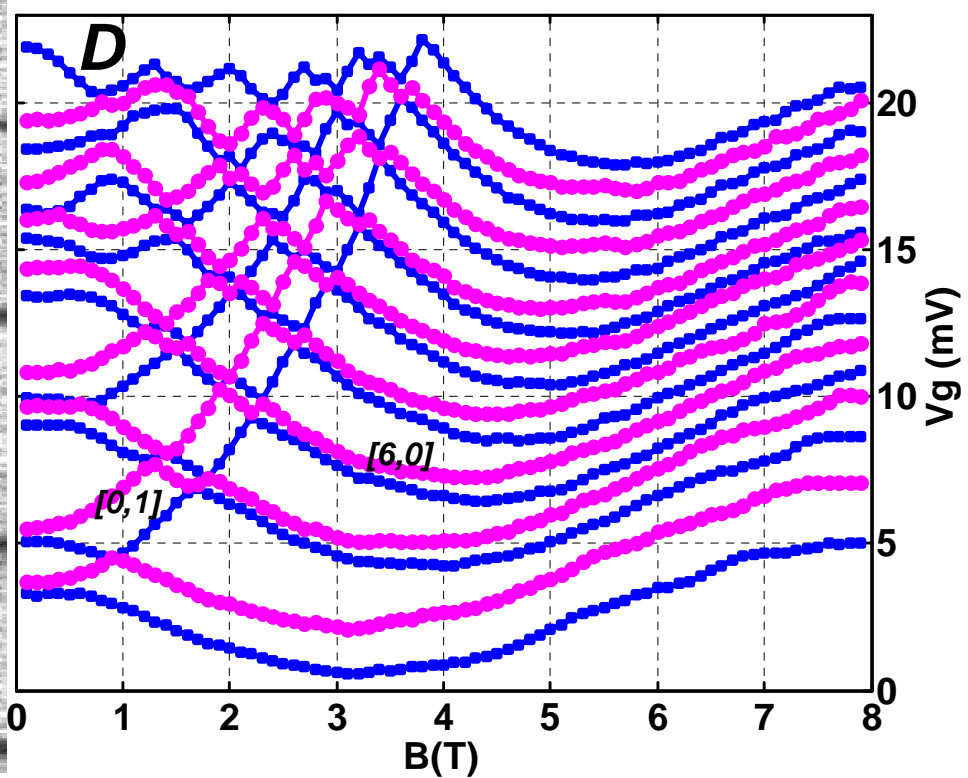
(A) Gate voltage values subtracted from the addition traces to build the plot on Fig.1D $(V_{ab}\pm J_{ab})$. (B) The same with a smooth quadratic background removed. C Schematic of the data shown on Fig.1C. Spin configurations are indicated for all five regions in magnetic field. Left (right) square represents orbital a (b). Total spin is marked by color: green for $s=0$, yellow for $s=1/2$, red for $s=1$. (D) The same color scheme used to mark the spin states of the entire spectrum (the same data as on Fig.1D)

Figure3

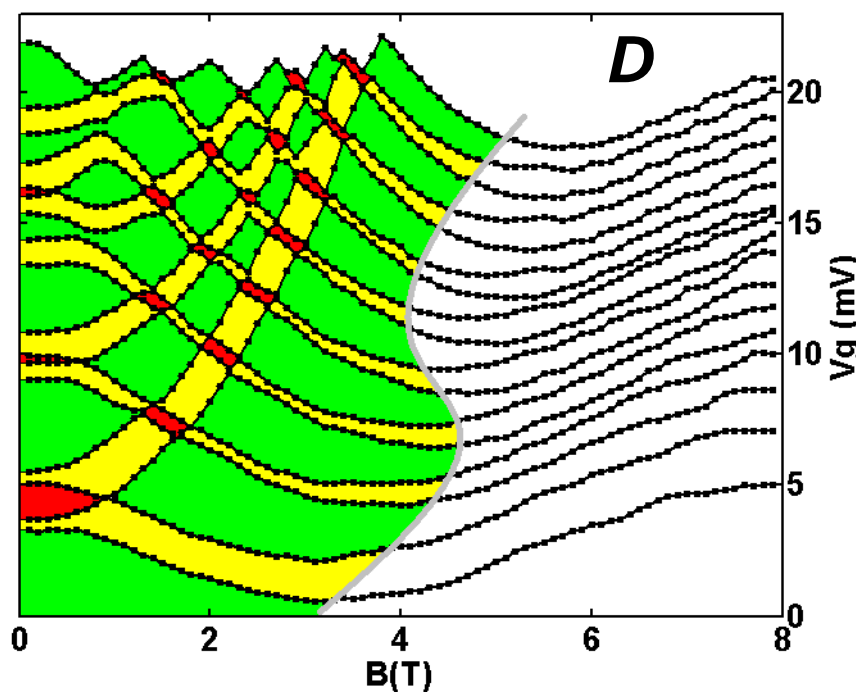
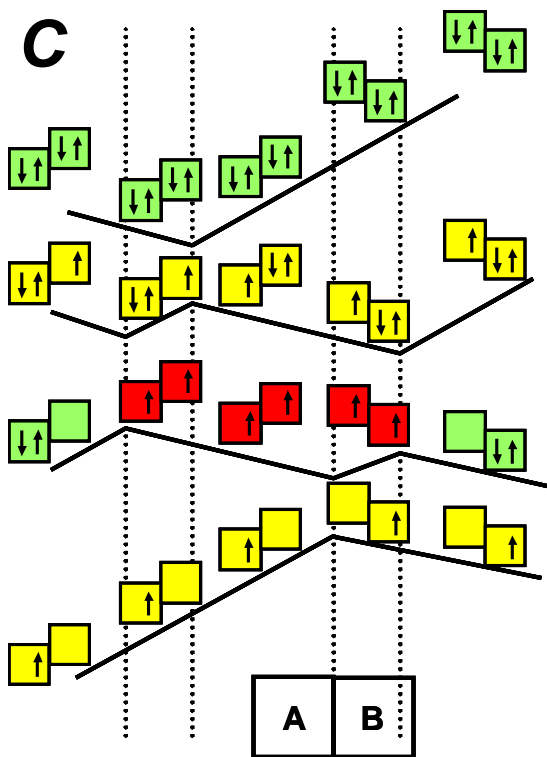
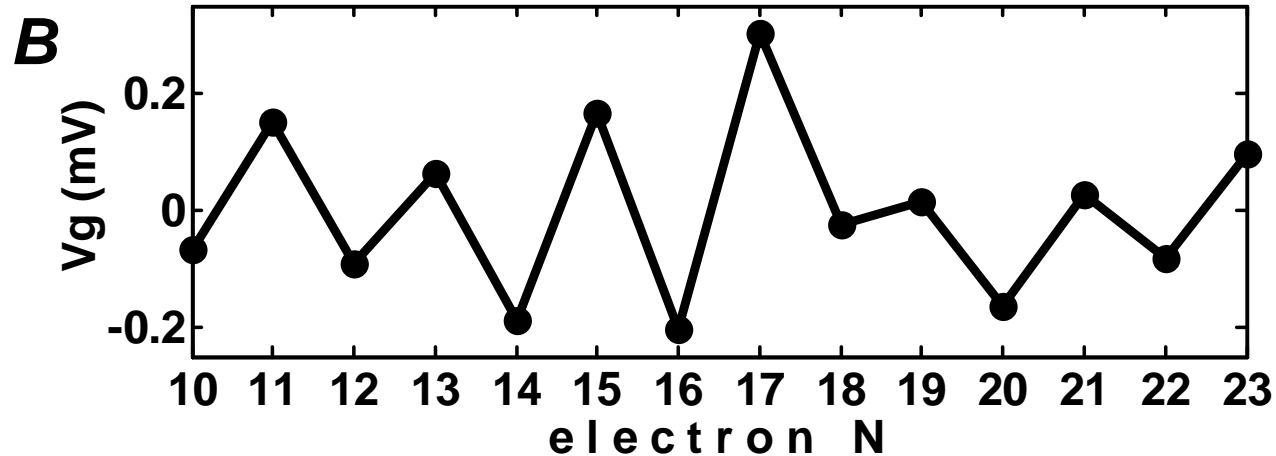
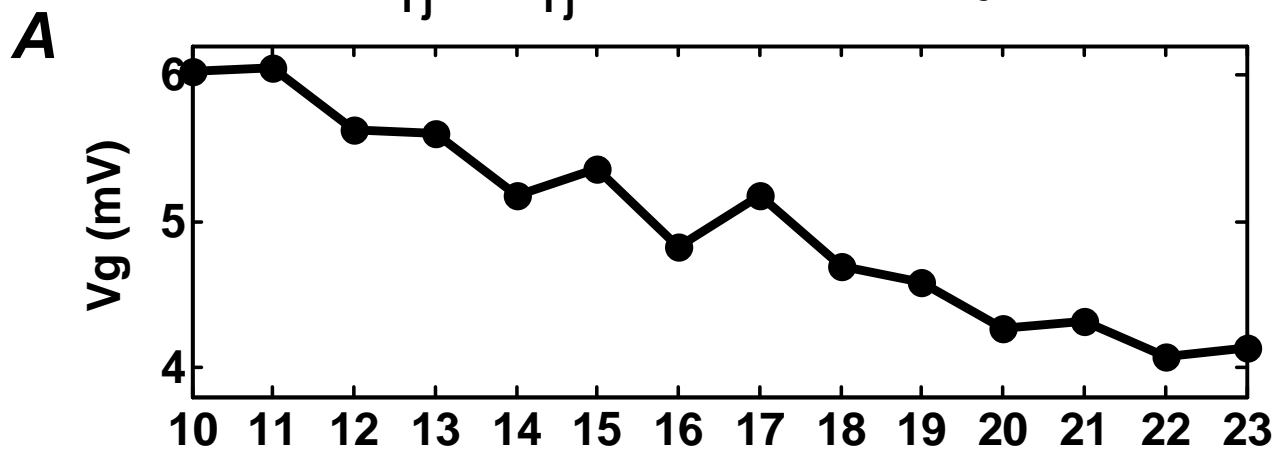
(A) Addition traces №61-65. Odd in blue, even in magenta. Gaps of $3.7mV$ are manually removed. (B) Addition traces №16(17) in magenta (blue) shifted by $127(130.5)mV$. Effects of complementary notches seen at $B\approx 1.95;2.8T$ are highlighted by the dotted lines. No notches but anticrossing at $B\approx 0.7T$.



electrons # 7 -23

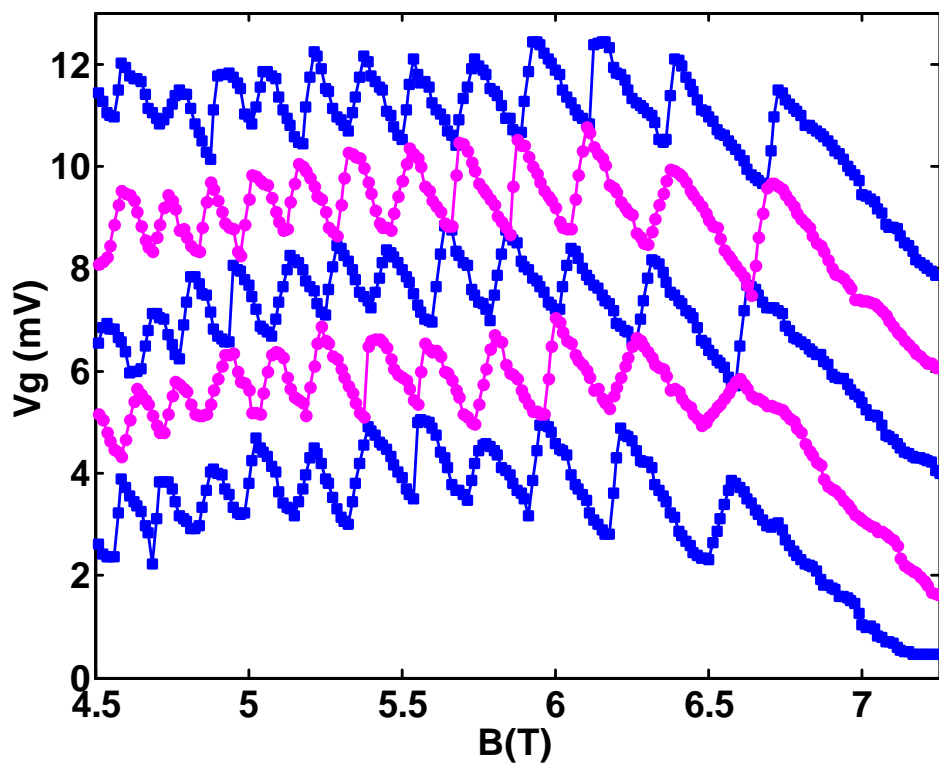


$$V_{ij} \pm J_{ij} \text{ for } i=N-1; j=N$$



electrons # 61 - 65

A



B

