The Collapse of the Spin-Singlet Phase in Quantum Dots

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We present experimental and theoretical results on a new regime in quantum dots in which the filling factor 2 singlet state is replaced by new spin polarized phases. We make use of spin blockade spectroscopy to identify the transition to this new regime as a function of the number of electrons. The key experimental observation is a reversal of the phase in the systematic oscillation of the amplitude of Coulomb blockade peaks as the number of electrons is increased above a critical number. It is found theoretically that correlations are crucial to the existence of the new phases.

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During the last decade, Coulomb blockade (CB) spectroscopic techniques have been used to investigate the electronic properties of quantum dots containing a discrete number of electrons 1, 2, 3, 4, 5, 6, 7, 8. CB peaks in the current through the dot are observed whenever the electrochemical potential of the dot is aligned with the source and drain leads. (For lateral dots these leads are the edges of a two dimensional electron gas, 2DEG.) The dot’s ground state and the total spin can be tuned by applying a perpendicular magnetic field. Each new ground state is observed as a cusp in the position of the CB peak 1, 2, 3, 4, 5, 6, 7, 8 leading to the frequent use of this technique for spectroscopy. The amplitude of the peaks also contains information. For irregular quantum dots with hundreds of electrons, fluctuations in CB peak amplitudes were used to investigate chaotic phenomena 9 since the amplitude is reduced whenever the overlap of the dot ground state with the leads is reduced. A similar spatial overlap argument was also introduced to explain drops in the peak amplitude at certain points in the addition spectrum of medium sized quantum dots 10. For smaller quantum dots containing fewer electrons, spectroscopic information was, however, inferred exclusively from the spacing of the CB peaks (the addition spectrum) 1, 2, 3, 4, 5, 6, 7, 8. Recently we discovered an important additional mechanism for amplitude modulation. In our experiments on lateral devices, we found that electrons injected into the dot were partially spin polarized 1, 2, (spin down). The origin of the observed spin-polarized injection lay in the exchange-enhanced spin splitting of the magnetic edge states of the 2DEG leads at the entrance and exit barriers of the dot. The amplitude modulation is determined by the difference in the electronic configuration of ground states with two consecutive electron numbers $N_e$. Whenever this difference involves a spin up electron the current is dramatically reduced due to spin blockade 1, 2, 3, 4, even if the spatial overlap is large. Coulomb blockade spectroscopy of lateral quantum dots is thus accompanied by spin blockade (SB) spectroscopy 1, 2, 3, 4, 5.

We have previously utilized SB spectroscopy to directly investigate singlet-triplet (ST) transitions 3, 4, 5 that occur close to the filling factor $\nu = 2$ regime 1, 2, 3 in quantum dots containing up to 20 electrons. The $\nu = 2$ regime in dots corresponds to a droplet of electrons occupying an equal number of the lowest spin-up and spin-down states of the lowest Landau level. The ST transitions had been predicted theoretically 11 and were first observed in the CB peak spacing of vertical quantum dots by Tarucha et al. 12 and interpreted in terms of direct and exchange interactions of the two electrons involved. In this Letter we report on a new and unexpected effect which is not discernible in the spacing of CB peaks but appears clearly in the pattern of CB amplitude modulation: the complete disappearance or quenching of the spin-singlet phase itself above a critical number of electrons $N_e$. We show that this effect can be understood in terms of a correlated behavior of many electrons.

The SEM picture of a device similar to the ones used in our experiments is shown in the inset of Fig. 1a. The layout of gates in the device allows us to form a slightly deformed parabolic dot 13 in which the number of electrons can be controllably tuned from around 50 electrons down to 1 14. In Fig. 1a we show a typical addition spectrum for the first 30 electrons entering our dot obtained by means of CB spectroscopy. The $\nu = 2$ line, indicated by a series of red dots and the arrow, is a very pronounced feature of the spectrum. Immediately to the right of this feature is the $\nu = 2$ regime. Figure 1b shows results of SB spectroscopy, i.e., the amplitude of the CB peaks obtained from the same set of measurements as the addition spectrum shown in Fig. 1a. The amplitude shows strong oscillations for $B > 0.4$ T where spin-polarized injection and detection takes place. The $\nu = 2$ line, marked by red circles, is clearly visible as a dip in the amplitude starting with five electrons.

On closer inspection, however, it is clear that there are certain features visible only in the SB spectra. These
are marked with yellow for even electron numbers and with blue for odd electron numbers. The yellow marks approach and eventually cross the line in Fig. 1b, at a critical number of electrons $N_c$, an effect observed in all of our samples (this feature pictorially marks the transition to the new phases described in this paper). The yellow and blue features correspond to the first spin flip for each $N_c$ as a function of magnetic field. It is well established that as the field is raised and the filling factor in the quantum dot changes from $\nu = 2$ to $\nu = 1$ the quantum dot spin polarizes through a sequence of spin flips. It is difficult, however, to experimentally resolve the first few spin flips using CB spectroscopy. The amplitude magneto-fingerprint of a spin flip event (a drop in the peak amplitude due to both spatial and spin blockades) is found, however, to be observable even for the first spin flip in Fig. 2a. For an odd number of electrons, one unpaired electron occupies a level at the edge of the droplet and the total spin of the in this case is $-1/2$. As the magnetic field is lowered, the singlet phase becomes unstable against the transfer of an electron from the edge of the dot to the second Landau level orbital (0,1) in the center of the dot. The spatial charge distribution corresponding to the center and a representative edge orbital are shown at the bottom of Fig. 2b. For an even number of electrons, decreasing the magnetic field transfers an electron from an edge orbital to a center orbital with angular momentum $-1$ while simultaneously flipping its spin. The dot is then in a triplet state formed by one electron in the edge and one in the center of the droplet. This configuration does not, of course, just consist of two electrons, but is a many-body state. For an odd number of electrons, an unpaired electron at the edge of the droplet is also transferred to the center but without flipping its spin so the total spin of the droplet in this case remains unchanged. As seen in Fig. 1b, there is no discernible experimental difference in the magnetic field at which transitions for even and odd total electron numbers take place. At higher magnetic fields there is a second boundary. The $\nu = 2$ droplet becomes unstable against spin flips at the edge of the quantum dot. This boundary is dependent, however, on whether the dot contains an even or odd number of electrons. For both odd and even electron numbers $N_c$, a spin up elec-

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**FIG. 1:** The addition spectrum (a) and amplitude spectrum (b) of the first 30 electrons with the charging energy manually removed. The arrow points to the $\nu = 2$ droplet, where the spin of the dot oscillates between zero for even electron numbers and 1/2 for odd electron numbers. The inset shows the gate layout of the experimental device.

**FIG. 2:** Electronic configurations of the ground state of the $N$-electron droplet in the vicinity of the $\nu = 2$ line for $N < N_c$ (a) and $N > N_c$ (b) (Black: edge orbitals, Gray: center orbitals). Shown schematically is the magnetic field evolution of the CB amplitude related to changing the number of electrons from even to odd. Bottom inset shows spatial probability density of the center ($m = 0, n = 1$) and edge ($m = 9, n = 0$) orbitals. (See text for details.)
electron at the edge of the droplet moves to the first available empty orbital with a higher angular momentum and flips its spin. For even $N_e$, and in the absence of interactions, the electron flips its spin at the edge whenever the cost of kinetic energy is compensated by the gain in Zeeman energy $E_Z$. For odd $N_e$, spin flips cost twice as much kinetic energy. Hence, the magnetic field for spin flips for odd $N_e$ is much higher than that for even $N_e$—the parity, not the magnitude, of $N_e$ is what is important. These features can be seen in the data corresponding to the first electron spin flip. The weak dependence on $N_e$ together with the large shift between the even and odd spin flips, while renormalized by interactions, is still visible in the SB spectrum of quantum dot shown in Fig. 1.

A number of model calculations for our system—spin density functional theory in the local spin-density approximation (LSDA) with Landau-level mixing, and Hartree-Fock calculations with and without Landau-level mixing—were performed. In particular, the stability of the $\nu = 2$ singlet phase against transfer of electrons to the center (center configurations) and against spin flips at the edge (edge configurations) was studied. In Fig. 2a we show the calculated spin of the ground state configuration as a function of magnetic field for electron droplets with even and odd $N_e$. Red denotes center configurations, yellow denotes edge configurations for even $N_e$, blue denotes edge configurations for odd $N_e$, and black denotes the $\nu = 2$ spin singlet droplet. The self-consistent calculations employed the LSDA and include mixing of ten Landau levels. The calculations used a confinement energy of $\omega = 1$ meV (extracted from the magnetic-field evolution of the CB peak corresponding to the first electron in the dot), a Zeeman energy of $E_Z = 0.04$ meV/T, and strictly 2D Coulomb interactions. The results of these calculations were already schematically summarized in Fig. 2. The LSDA and Hartree Fock calculations with and without a mixing of Landau levels all give a finite stability range of the spin singlet droplet (black region). At a critical number of electrons $N_{c}$, the spin-singlet $\nu = 2$ phase ceases to be the ground state. As seen in Fig. 3, upon increasing the field, the dot with even $N_{c}$ evolves from a center configuration with a spin-down electron at the center and, to an edge configuration with two spin-down electrons at the edge of the droplet. This is shown schematically in Fig. 3a. A comparison of Figs. 2a, 2b, and Fig. 3 reveals a change in the center configuration of the droplet consisting of an odd number of electrons. The key effect is the triggering of spin polarization at the edge by the spin and charge of an electron at the center. These configurations persist over a finite range of electron numbers, as shown in Fig. 3.

This effect is too weak to be observed in the addition spectrum of Fig. 1. In contrast, the effect can be directly observed with SB spectroscopy. Consider how the predicted changes in the ground-state configurations should affect the current through the dot. The magnetic-field evolution of the CB peaks is schematically shown in Fig. 2. The thickness of the lines indicates the expected current amplitude (thin for low, thick for high). For $N_e < N_c$, the two center configurations for odd and even $N_e$ differ by one spin-up electron at the edge of the dot. The two edge configurations differ by a spin-down electron at the edge. Because of spin-polarized injection, SB-spectroscopy measurements should reveal a relatively small current flowing through the dot whenever a transition occurs between center configurations (left of the $\nu = 2$ line) and a relatively large current whenever a transition occurs between edge configurations (right of the $\nu = 2$ line). A similar analysis for transitions from odd to even $N_e$ would give high current on the left of $\nu = 2$ line and low current on the right.

In the case of $N_e > N_c$, as seen in Fig. 2b, the electronic configurations of the respective ground states have changed. The initial center configuration for even $N_e$ and the final center configuration for odd $N_e$ differ by a spin-down electron at the edge, and so we expect a large current on the left of the $\nu = 2$ line. The initial and final edge configurations differ by a spin-up electron at the edge of the droplet, and so the observed current is expected to be low. Thus, the collapse of the $\nu = 2$ spin-singlet droplet should be seen through SB spectroscopy as a reversal of the amplitude oscillation pattern in the vicinity of the $\nu = 2$ line as the number of electrons is increased. This is indeed observed in our experiments. In Fig. 3, we show inverted gray-scales of the magnetic-field evolution of four CB peaks in the vicinity of the $\nu = 2$ line in the regime of both $N_e < N_c$ and $N_e > N_c$. Dark and light shades in the gray-scale indicate respectively large and small peak amplitude. The amplitude of the CB peaks behaves in the
way predicted in the above discussion of ground-state electronic configurations. In the bottom panel of Fig. 4, we plot the ratio of the peak amplitude $A_2$ on the right side of the $\nu = 2$ line to the amplitude $A_1$ on the left side of the $\nu = 2$ line as a function of electron number $N$. For a low electron number, this ratio is greater than unity when adding an odd electron to the dot and less than unity when adding an even electron. The pattern reverses around $N_c = 25$. This number is different from the calculated one which perhaps points to our overestimation of the strength of Coulomb interactions, and the lack of detailed knowledge of the change of confinement on the number of electrons. The collapse of the $\nu = 2$ spin-singlet droplet and the critical number of electrons $N_c$ observed in experiment was reproduced by Hartree-Fock calculations which include correlations were capable of producing a phase diagram leading to amplitude reversal. Hence amplitude reversal appears to be connected to correlations, and more realistic calculations are in progress to illuminate this connection.

To summarize, we have studied the stability of the $\nu = 2$ spin-singlet phase of a quantum dot as a function of electron number $N$ and magnetic field $B$. We have demonstrated that this phase collapses at a certain electron number $N_c$ in favor of spin-polarized configurations. We were able to observe this effect experimentally with spin-blockade spectroscopy. The experiments and calculations demonstrate new effects uncovered by the control of electron spin in a nanoscale object with a tunable and controlled number of electrons. These findings should have impact on the merging fields of spintronics, nanotechnology, and quantum information, which require the ability to control and manipulate spin and charge at the single-electron level.

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