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 two-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 past decade, Coulomb blockade (CB) spectroscopic techniques have been used to investigate the electronic properties of quantum dots containing a discrete number of electrons [1–13]. Cusps in the position of CB peaks in the current through the dot are directly related to transitions in the dot's ground state tuned by, e.g., applying a perpendicular magnetic field. The amplitude of the CB peaks also contains information. For large but irregular quantum dots, fluctuations in CB peaks amplitude were used to investigate chaotic phenomena [14] since the amplitude was lowered whenever the overlap of the dot ground state with the leads was reduced. A similar spatial overlap argument was introduced to explain drops in the peak amplitude at certain points in the addition spectrum of medium sized quantum dots [1]. For smaller quantum dots containing few electrons, spectroscopic information was, however, inferred exclusively from the position of the Coulomb blockade peaks (the addition spectrum) [1–13,15]. Recently, CB spectroscopy was combined with spin polarized injection/detection (spin-down electrons) into spin blockade (SB) spectroscopy [10–13]. In SB spectroscopy, the amplitude of CB current is determined by the difference in the spin of electronic configurations 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 [10–13] even if the spatial overlap is large. Hence, SB is a very sensitive tool in probing electron spin in quantum dots. We have previously utilized SB spectroscopy to directly investigate singlet-triplet (ST) transitions [10,12,13] that occur close to the filling factor $\nu = 2$ regime [1,3–5,7,8] 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 [16] and were first observed in the CB peak spacing of vertical quantum dots by Tarucha *et al.* [15] who interpreted them in terms of direct and exchange interactions of the only 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_c. 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 [17] in which the number of electrons can be controllably tuned from around 50 down to 1 [13]. 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 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 take place. The $\nu = 2$ line, marked by black 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 squares for even electron numbers and with diamonds for odd electron numbers. The square 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 square and diamond features correspond to the first spin flip for each N_e as a function of magnetic field, an event which is invisible in the position of the CB peak.

First, we concentrate on the previously understood regime ($N_e < 25$) [12]. The line described by us as the $\nu = 2$ line is more accurately defined as the low-field boundary of the $\nu = 2$ singlet phase. To the right of the

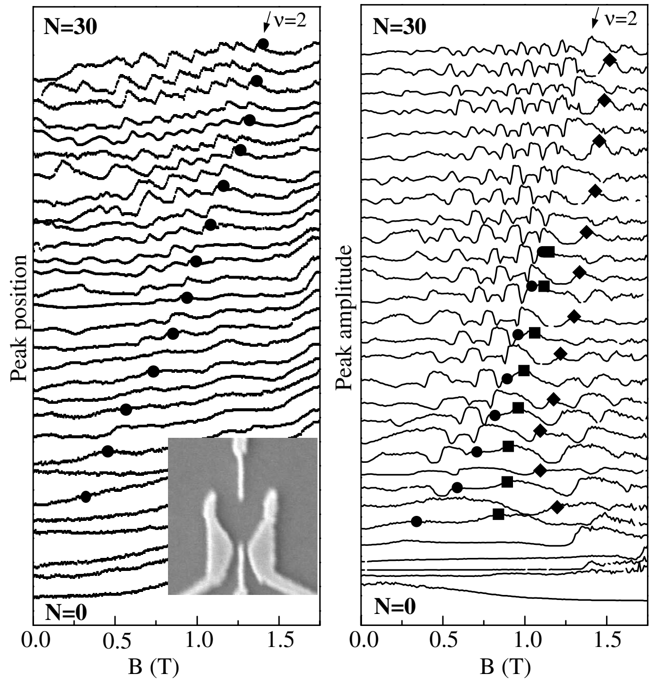

FIG. 1. The addition spectrum (a) and amplitude spectrum (b) of the first 30 electrons with the charging energy manually removed. The arrow points to the $\nu = 2$ droplet, where the spin of the dot oscillates between zero for even electron numbers and $1/2$ for odd electron numbers. The inset shows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

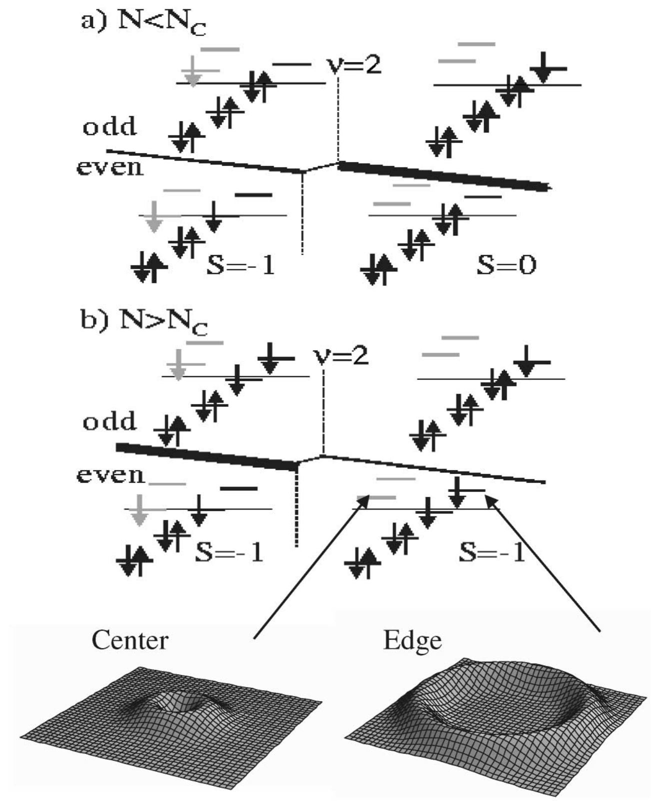

FIG. 2. Electronic configurations of the ground state of the N-electron droplet in the vicinity of the $\nu = 2$ line for $N < N_c$ (a) and $N > N_c$ (b) (black: edge orbitals; gray: center orbitals). Shown schematically is the magnetic field evolution of the CB amplitude related to changing the number of electrons from even to odd. The bottom inset shows the spatial probability density of the center ($m = 0, n = 1$) and edge ($m = 9, n = 0$) orbitals. (See text for details.)

$\nu = 2$ line, electrons occupy spin-split states $(m, 0)$ of the lowest Landau level with positive angular momentum m [10] (the zero indicates the lowest landau level). The states $(m, 0)$ are equally spaced, with an energy spacing decreasing with increasing magnetic field which would eventually converge at very high magnetic fields to a single Landau level. For each state $(m, 0)$ there is a state $(m, 1)$ originating from the second Landau level separated from it by a fixed energy which would approach the cyclotron energy at very high magnetic fields. For an even number of electrons $N_e = 2N$, both spin-down and -up states $(m, 0)$ are filled up to the Fermi level forming the $\nu = 2$ spin-singlet (total spin $S = 0$) state. This state, together with electronic configurations of other states in close proximity to the $\nu = 2$ line, are shown schematically 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 dot 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. 2. For an even number of electrons, decreasing the magnetic field transfers an electron from an edge 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 [12,16,17]. 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 [12,16,17]. As seen in Fig. 1a, 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 due to spin flips at the edge of the quantum dot. This boundary depends, however, on whether the dot contains an even or an odd number of electrons. For both odd and even electron numbers N_e, a spin-up 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 flip costs 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 the odd spin flips, while renormalized by interactions, is still visible in the SB spectrum of quantum dots shown in Fig. 1.

Model calculations were carried out to understand the stability of the $\nu = 2$ singlet phase [17]. In Fig. 3, 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 local spin density approximation (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_e evolves from a center configuration with a spin-down electron at the center and at the edge, to an edge configuration with two spin-down electrons at the edge of the droplet. This is shown schematically in Fig. 2b. A comparison of Figs. 2a, 2b, and 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 [13], 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 the $\nu = 2$ line and low current on the right [13]. 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. 4, we show inverted color 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 $N_e > N_c$. Red and blue shades in the color 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_e. 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.

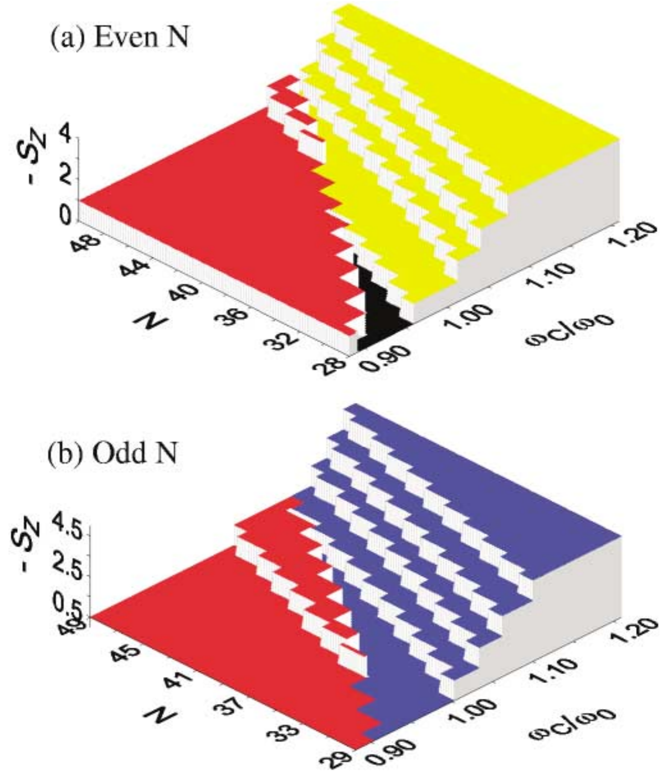

FIG. 3 (color). Calculated ground-state spin configurations of electron droplets with (a) even and (b) odd N near the collapse of the spin-singlet phase.

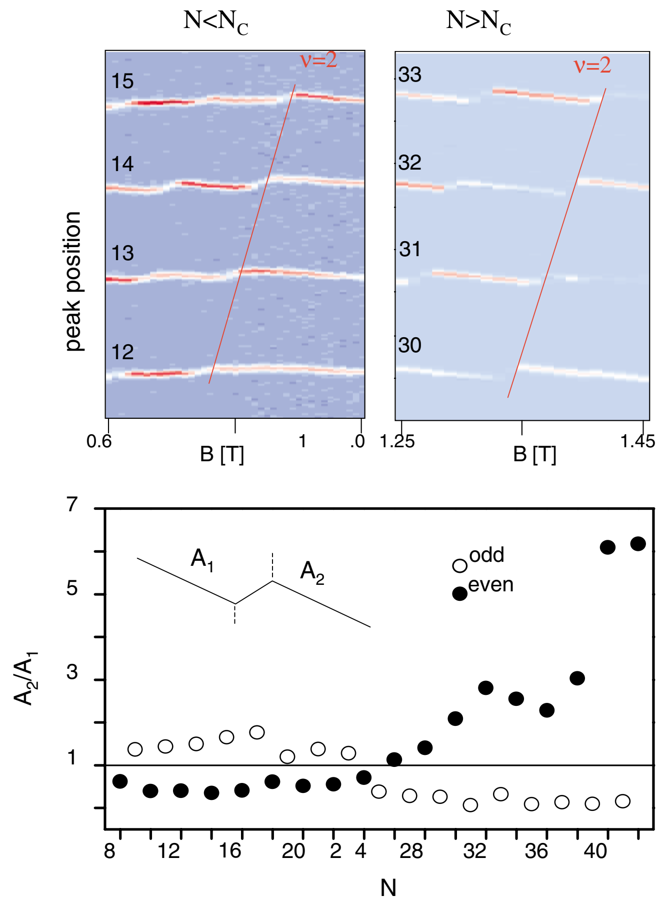

FIG. 4 (color). Upper panel: inverted color scale showing magnetic field evolution of four CB peaks in the vicinity of the $\nu = 2$ line for $N < N_c$ and $N > N_c$. Lower panel: ratio of CB peak amplitude A_2 on the right of the $\nu = 2$ line to the CB peak amplitude A_1 on the left as a function of electron number.

The pattern reverses around $N_c = 25$. This number is different from the calculated one which points to perhaps 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 the experiment were reproduced by Hartree-Fock calculations for a number of confining energies. However, only LSDA 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.

In summary, we have studied the stability of the $\nu = 2$ spin-singlet phase of a quantum dot as a function of electron number N_e 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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