Spin blockade in ground state resonance of a quantum dot

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Abstract. – We present measurements on spin blockade in a laterally integrated quantum dot. The dot is tuned into the regime of strong Coulomb blockade, confining ~ 50 electrons. At certain electronic states we find an additional mechanism suppressing electron transport. This we identify as spin blockade at zero bias, possibly accompanied by a change in orbital momentum in subsequent dot ground states. We support this by probing the bias, magnetic field and temperature dependence of the transport spectrum. Weak violation of the blockade is modelled by detailed calculations of non-linear transport taking into account forbidden transitions.

Conventional electronics relies on controlling charge in semiconductor transistors. The ultimate limit of integration is reached when these transistors, termed quantum dots, are operated by exchanging single electrons only. The mechanism governing electron transport through dots is known as Coulomb blockade (CB) [1,2]. Apart from this, electrons naturally possess a spin degree of freedom, which recently attracted considerable interest regarding the combination of spintronics and quantum information processing [3]. Hence, studies on the interplay of spin and charge quantum states in quantum dots form an integral contribution for defining and controlling electron spin quantum bits.

One of the key techniques applied to study electronic structure in quantum dots is transport spectroscopy: Defining the dots by locally depleting a two-dimensional electron gas enables this direct monitoring of ground and excited states of the artificial atoms [4]. In contrast to Kondo physics [5] in the limit of transparent tunneling barriers between the quantum dot and the reservoirs, we are focusing on the regime of opaque barriers and Coulomb blockade. In other words the strong hybridization with electronic reservoir states found for the dot electrons in Kondo physics is suppressed and both electron spin and orbital quantum numbers remain well-defined.
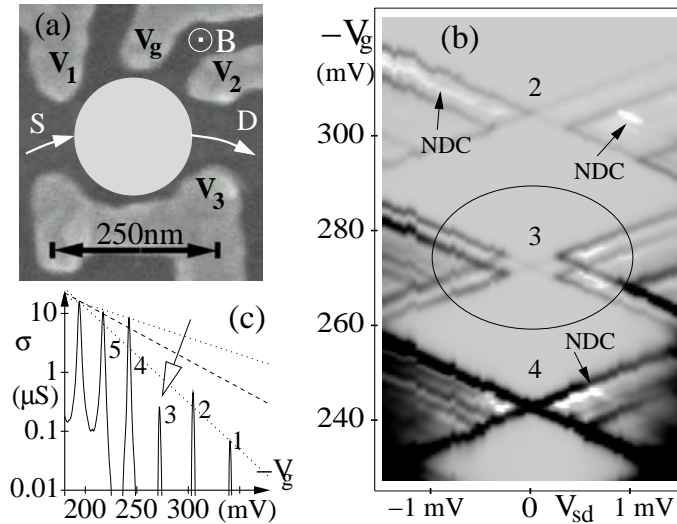
Fig. 1 – (a) SEM micrograph of the gate electrodes defining the quantum dot (top view). Approximate dot area (circle), source (S) and drain (D) contacts are marked schematically. (b) Conductance measurement on the quantum dot ($B = 0$ T; white denotes $-0.1\,\mu$S (NDC), grey (large areas) $0\,\mu$S, black $\geq 2.0\,\mu$S). (c) Conductance trace at $V_{sd} = 0$ V; an exponential fit through the conductance peak maxima has been added (dashed line). Conductance peak three is suppressed by spin blockade.

Inspired by an early experiment of Weis *et al.* [6] it was suggested by Weinmann *et al.* [7] that transport through single quantum dots can be blocked due to spin effects. Spin selection rules particularly prohibit single electron tunneling (SET) transitions between N and $N + 1$ electron ground states of the dot which differ in total spin by $\Delta S > 1/2$. This phenomenon was termed spin blockade (SB) (type-II), occuring in addition to conventional CB and leading to a suppression of the corresponding conductance peak in linear transport [7].

Here, we demonstrate a spin blockade effect in the many-electron ($N \sim 50$) limit in a two-dimensional quantum dot, in contrast to earlier measurements by Rokhinson *et al.* [8] on a small three-dimensional silicon quantum dot with only a few electrons. In particular we present detailed measurements on the bias and magnetic field dependence of the transport spectrum of our laterally gated quantum dot. Weak violation of spin blockade due to spin orbit coupling is found. Modelling the system by numerical calculations taking into account type-II spin blockade and weak spin relaxation, we observe qualitative agreement with the transport spectrum.

The quantum dot we use is shown in fig. 1: A number of split gates is defined by electron beam lithography on top of an AlGaAs/GaAs heterostructure. When applying negative gate voltages V_1, V_2, V_3, and V_g, a single nearly circular quantum dot is formed in the two-dimensional electron system (2DES) 120 nm below the surface. At 4.2 K the carrier density of the 2DES is $n_s = 1.8 \times 10^{15}\,\mathrm{m}^{-2}$ and the electron mobility is $75\,\mathrm{m}^2/\mathrm{Vs}$. For the measurements presented here a dot with an electronic radius of approximately 90 nm was defined. The 2DES was cooled to a bath temperature of 23 mK and an electron temperature of $T_{el} = 95$ mK in a ^{3}He/^{4}He dilution refrigerator. Using an excitation voltage of $12\,\mu$V at 18 Hz, the noise in the CB regime is lower than 50 nS. Electronic radius and mean level spacing both lead to an estimate of $N \sim 50$ electrons on the dot.

In fig. 1(b) the conductance is plotted as a function of gate voltage V_g and source/drain bias V_{sd}. Near $V_g = -290\,\text{mV}$, a capacitance ratio of $\alpha = C_g/C_\Sigma = 0.059$ has been obtained from this measurement, leading to a total dot capacitance of $C_\Sigma = 83\,\text{aF}$. Therefore the Coulomb charging energy $E_C = e^2/C_\Sigma$ of around $1.9\,\text{meV}$, giving the electrostatic energy required to add an electron to the quantum dot, is dominant compared to the spatial quantization energy ϵ. Nevertheless, a rich spectrum of excited state resonances in all SET regions is revealed. In addition, multiple lines of negative differential conductance (NDC) are visible, as predicted in ref. [7]. Hence, we can conlude that in these regions spin polarized excited states lead to a reduction of current via spin blockade type-I.

As a striking feature, the conductance at peak three in fig. 1 is, for small $|V_{\text{sd}}|$, significantly below the expected value as compared to peaks two and four. Only for $|V_{\text{sd}}| \geq 300\,\mu\text{V}$ transport via an excited state becomes possible, and the conductance increases. Such strong suppression of ground state tunneling is caused by spin blockade of type-II. This phenomenon involves ground state transitions only. An electron is blocked from entering the dot, since the transition involves two levels with total spin difference $\Delta S > 1/2$.

Fig. 1(c) shows the conductance trace at $V_{\text{sd}} = 0$ in logarithmic scale. As the width of the tunneling barriers increases at higher $|V_g|$, the maximum current approaches zero in the case of complete pinch-off. In a simple quantum-mechanical picture the maximum peak conductance is assumed to decrease exponentially with $-V_{\text{g}}$. Again, the amplitude of peak three is considerably below the most general maximum/minimum bounds (dotted lines in fig. 1(c)). An exponential fit through the peak maxima (dashed line) allows us to estimate a lifetime of the spin state causing spin blockade. We assume that the tunneling rates $\Gamma_{L/R}/h$ of the left and right barrier defining the dot are equal – which appears reasonable because of the high degree of symmetry shown in the diamond conductance structure of fig. 1(b).

In our case of weak coupling to the reservoirs, where $\Gamma \ll k_B T \ll \epsilon < E_C$, the maximum conductance through the quantum dot is given by [9]

$$\sigma_{\text{max}} = \frac{e^2}{h} \frac{1}{4k_B T_{\text{el}}} \frac{\Gamma}{2}.$$

The dwell time for an electron in the system is estimated with $\tau_b = h/\Gamma_b = 9.5\,\text{ns}$ in the case of transport blockade compared to $\tau_e = h/\Gamma_e = 0.8\,\text{ns}$ as extrapolated value without any spin effects [10]. Therefore, coupling of the long-lived state to its environment corresponds to a time scale of $\Delta\tau = \tau_b - \tau_e \approx 8.7\,\text{ns}$, i.e. the high-spin state survives for several nanoseconds.

Additional information on spin blockade is given by the temperature dependence of the current peaks at the SET resonances, taken from a measurement at $V_{\text{sd}} = 20\,\mu\text{V}$ (dc) and displayed in fig. 2(a). In the present case of CB ($E_C > \epsilon \gg k_B T$), a decrease of the current at higher temperatures is expected [9], while peak three shows an increase above $T_e \sim 400\,\text{mK}$. This gives rise to the assumption that via the broadening of the Fermi-Dirac distribution in the leads a non-blocked transport channel becomes accessible at an energy scale comparable to $k_B T_e \approx 35\,\mu eV$ above eV_{sd} [7], by order of magnitude consistent with a change in spin configuration at $B = 0\,\text{T}$ [11].

In fig. 2(b), we observe the change in electron addition energy $\Delta E = E_C + \Delta\epsilon$ with increasing magnetic field, which is proportional to the gate voltage SET peak spacing (see inset of fig. 2(b)). Whereas a magnetic field parallel to the 2DES couples primarily to the electron spin via the Zeeman energy term, the perpendicular field applied here strongly influences the orbital states as well. Most observed peaks display a weak field dependence. Strikingly, the spin-blocked resonance is shifting strongly at low magnetic fields, as can bee seen at hand of the high slopes of charging energy 2-3 and 3-4 in fig. 2(b).
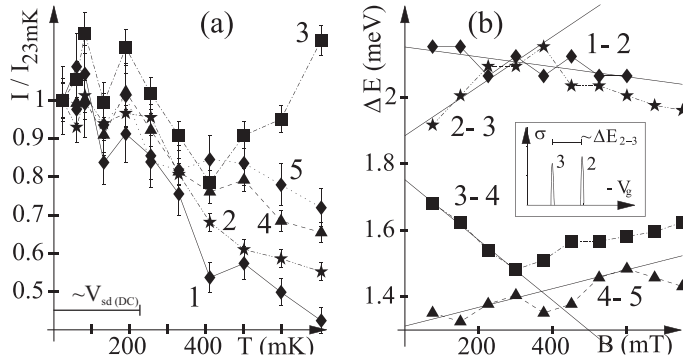
Fig. 2 – (a) Relative temperature dependence of the CB peak height (dc measurement at $V_{sd} = 20 \, \mu$V), including fit errors; only peak three increases with temperature above $T_e \sim 400$ mK, in every other case the decay characteristic for CB ist apparent. (b) B-dependence of the single electron addition energy $\Delta E_{N-(N+1)} = \alpha(V_g) \left| V_{g,N} - V_{g,(N+1)} \right|$. For small B, peak spacings around peak three vary rapidly with respect to a magnetic field perpendicular to the 2DES.

Different explanations for this phenomenon are possible. On one hand, recent measurements by Pallecchi *et al.* [12] indicate the possibility of greatly enhanced g-factors in 2D quantum dots; values of up to $g \sim 20$ have been observed in AlGaAs/GaAs quantum wires. As an example, at $g \approx 5.4$ line shifts can be approximated solely by the Zeeman shift caused by a spin difference $\Delta S = 5/2$ of subsequent electron number ground states (cf. ref. [13]), thus explaining spin blockade. At nearby, non-blocked resonances, the peak shift is then consistent with a spin change in the dot of $\Delta S = 1/2$, i.e. the addition of a single electron spin. Theoretical predictions [14] support a g-factor deviation in case of unusually large spin-orbit interaction.

However, orbital states are modified by the magnetic field perpendicular to the 2DES as well. Hence, on the other hand a change in orbital quantum numbers, particularly ground state angular momentum, is an alternative explanation for the peak shift. Since momentum and angular momentum of an electron are not preserved when traversing the quantum point contacts [15], a misalignment of spatial quantum numbers alone will not lead to a total blocking of transport. Therefore, a change in both L and S between subsequent electron number ground states leads to a scenario explaining the data.

In addition nonlinear conductance measurements at finite magnetic field perpendicular to the 2DES have been taken. In fig. 3(a) to (c) the transport-blocked SET resonance is shown at 0 mT, 150 mT, and 450 mT. At a field strength of only 300 mT the quantum levels in the dot are already shifted sufficiently to reenable ground state transport. This is also demonstrated in fig. 3(d), which shows the conductance at $V_{sd} = 0$ V and the blockade gap energy E_G as function of B. At $B = 300$ mT a strong increase in conductance is seen. Here, one quantum ground state participating in SET changes because of a level crossing; for higher B, $|\Delta S| = 1/2$ for the N and $N + 1$ electron ground state and SET transport takes place. This assumption is directly supported by the data of fig. 2(b), $B = 300$ mT being the field strength where the B-dependence of addition energies around peak three adapts to the one around nearby peaks. For high B the overall conductance through the quantum dot is decreasing because of a gradual compression of the dot states by the magnetic field [16].

The data indicate strong electronic correlations in the quantum dot involving both spin
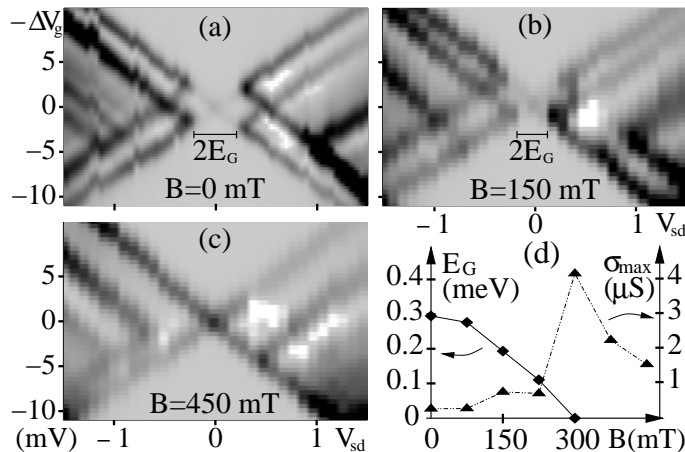
Fig. 3 – (a) to (c): Effect of an external perpendicular magnetic field on the spin blocked SET resonance three. (a) Detail from Fig. 1(b). (b) At $B = 150\,\text{mT}$ the gap has partly closed. Conductance is still suppressed for $|V_{\text{sd}}| < 200\,\mu\text{V}$. (c) Blockade phenomena disappear completely at $450\,\text{mT}$. Simultaneously regions of NDC are evolving. (d) Magnetic field dependence of the transport blockade gap as indicated in (a) and the maximum conductance at $V_{\text{sd}} = 0\,\text{V}$. At approximately $300\,\text{mT}$ the gap is quenched.

and orbit of the wavefunction. The expected impact of correlation effects can be estimated by the conventional parameter $r_s = 1/\sqrt{\pi n_s}\, a_B$ [17] with a_B as the Bohr radius in GaAs. In our sample, it is given by $r_s = 1.2$, thus even assuming a somewhat lower electron density in the dot, it remains still far below critical values of $r_s \sim 8$ to 36. Taking into account the high number of electrons present ($N \approx 50$), the measurement presents an unusually strong deviation from the constant interaction model [2].

A spin blockade level scenario and results from numerical transport calculations are depicted in fig. 4. The calculations were performed using a master equation approach, describing the regime of sequential tunneling, as in ref. [7]. The transition rates between the many-body states of the dot include a Clebsch-Gordon factor which accounts for the spin selection rules. Spin values of $S = 2$ and 3 have been assumed in the model for the ground and excited N-electron states, $7/2$ and $5/2$ for the $(N + 1)$-electron states, respectively. Given the blockade condition, similar spin values may lead to the same qualitative results; the model retains a high degree of generality. If one assumes perfect spin conservation in tunneling processes as well as for the electrons confined in the dot, the theory yields transport spectra which are qualitatively different from the experimental ones (see fig. 4(a)). This comparison of the theory and the measurements indicates that spin blockade is weakly violated, possibly due to spin-orbit coupling. Such a state mixing mechanism in the quantum dot leads to a violation of both spin and angular momentum conservation. The couplings might differ considerably in our case from previous observations [8]; possible reasons include the material system (AlGaAs/GaAs instead of Si), the potential shape of the dot, and their effect on the details of the spin-orbit Hamiltonian.

We take this effect into account by including a phenomenological spin-independent transition rate between many-body states in our model, allowing for electron tunneling processes from and into the dot in which the spin selection rules are violated. Using the small value of 1% of the spin-allowed transition rates, and assuming quantum levels as displayed in fig. 4(b),
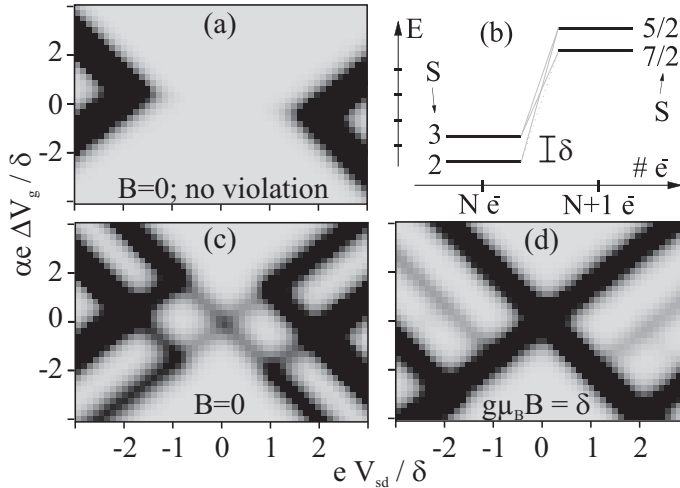
Fig. 4 – Model calculation of nonlinear transport for spin blockade type-II. (a) Transport spectrum in the case of perfect spin conservation, differing considerably from measured data. (b) Schematic representation of the quantum levels assumed for the calculation. (c) When taking into account a weak violation of spin transition rules, the characteristic blockade situation at $B = 0$ T (cf. Fig. 1(b)) emerges. In (d) spin blockade is lifted by a magnetic field $B = \delta/g\mu_B$, with δ being the excitation energy of the N electron system: because of a level crossing, the blockade condition $|\Delta S| > 1/2$ is not fulfilled anymore (cf. Fig. 3).

good qualitative agreement of theory and experiment is found in fig. 4(c) and (d). The magnetic field was introduced as a Zeeman-like shift of the levels, leading to a level crossing in the N-electron spectrum when $g\mu_B B$ equals the N-electron excitation energy δ. The actual mechanism of the level shift observed in the experiment may be more complex, however, our findings explains that the spin blockade is lifted at stronger magnetic field, in agreement with the experimental observations.

In conclusion, we have demonstrated transport blockade in a quantum dot containing approximately 50 electrons, caused by spin selection rules: electronic correlations lead to a discontinuity in ground state spin quantum numbers and possibly in addition in spatial quantum numbers for subsequent electron counts. As a result, ground state transport blockade caused by spin selection rules is found.

We observe the properties predicted for such a system: Spin blockade can be lifted by raising the temperature, or applying a source/drain voltage. Shifting the quantum levels via a magnetic field leads to a change in ground state spin, lifting spin blockade as well. Two alternative mechanisms for the observed level shift are proposed, firstly, a large Zeeman shift via g-factor enhancement, secondly, a combined change of both spin and orbital quantum numbers.

In the measurement, spin blockade is weakly violated. A likely candidate for this conduction process is given by the spin-orbit interaction induced spin state mixing in the quantum dot, being consistent with both proposed mechanisms of level shifting. Taking a corresponding weak violation of selection rules into account, numerical calculations of a straightforward spin blockade model lead to good qualitative agreement of theory and experiment, and the properties of the nonlinear transport spectrum at zero and large B are accurately reproduced.

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