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This report results from a contract tasking Ludwig-Maximilian University of Munich as follows: The contractor will investigate quantum circuits and on-chip millimeter wave sources for probing the dynamics of coupled quantum dots. The on-chip sources he will apply are especially designed Josephson oscillators operating in the frequency range of some 10 GHz up to 600 GHz. The goal will be to combine two different low-dimensional electronic systems and use superconducting millimeter wave oscillators to study the intricate electron wave function interaction.

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## **On-chip timing of quantum bits: Final report**

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## Executive Summary

The starting point of this project was to find new avenues of computation by applying quantum dots for developing quantum computational circuits. The primary goal then was to use microwave sources for on-chip timing and spectroscopy of these quantum dots. Containing only few electrons quantum dots represent the ultimate limit of integrating transistors. This finally enables to employ the quantum character of circuits: Apart from the elementary charge  $e$  the electron possesses a spin quantum number. This additional degree of freedom can be brought to use in information processing, i.e. not only the spatial part of the electron wave functions but also the 'magnetic' part have to be considered. A particular advantage of the electron spin is its reduced interaction with the environment as compared to the charge. This is expressed in enhanced relaxation times in excess of 100 secs. The statement naturally extends to coupled quantum dots where orbital and spin wave functions are responsible for the binding and anti-binding states and a rich structure of energy levels is found [Hol02]. Such intricate level schemes are probed by tracing its magnetic field dependence or by applying microwave spectroscopy. The underlying idea in these experiments was to induce excitations in these artificial atoms and molecules for information processing and storage.

## Quantum information manipulation and storage

The key questions in quantum information processing are how to efficiently manipulate and store such information in quantum bits with a sufficient life time. The starting point of our approach was to first investigate the details of the molecular binding mechanisms of two coupled dots, while the focus was then shifted towards manipulating the electron's spin in single quantum dots. The system of choice for achieving this are quantum dots realized in AlGaAs/GaAs-heterojunctions with a high mobility two dimensional electron gas embedded close to the surface of the semiconductor. This allows for patterning the surface by electron beam lithography and subsequent evaporation of a metallic gate structure. In such a way small puddles of only a few electrons can be formed whose coupling to source and drain contacts is widely tunable. This is also possible when two or more dots are connected for the interdot coupling (see Fig. 1). By simply altering the gate voltage in the range of some mV the direct overlap of wave functions can be exactly determined.

Transport spectroscopy is applied as a fundamental means for achieving insight into the nanoscopic systems. We combine this technique with microwave spectroscopy operating in the frequency range of some GHz up to 100 GHz. During the first part of the work on this proposal we investigated the details of the molecular binding mechanisms [Hol02] and were able to show that microwave radiation can be applied for manipulating the phase coherent wave functions [Qin01a,Qin01b,Bra01] as reported earlier. In the second part the focus shifted towards getting a handle on the electron's spin degree of freedom and to manipulate it by microwave pulses. The first step henceforth was to

clearly define a spin state, which we identified by spin blockade of single electron transport through a single dot [Hue02a]. The second step to be taken was to manipulate the  $N$ -electron wave function to put the high spin state to use. This we achieved by monitoring the interaction of the electron-nuclear spin system. The embedding crystal matrix of the semiconductor material obviously consists of the nuclei of Al, Ga, and As with radio frequency transition frequencies. The overwhelming advantage of having control over the electron-nuclear spin interaction are the extremely slow decay times of nuclear spin relaxation. Thus we achieved for the first time to transfer quantum information of an electronic dot state via a spin-flip mechanism to the nuclear spin matrix with a life time easily exceeding 20 minutes. This is especially exciting comparing conventional electron spin life times which are of the order of 10 nsec to mostly 100 nsec. In the course of the measurements as outlined below we finally employed electron spin resonance to induce spin-flips which can be regarded as a switching mechanism varying the life time according to the requirements.

The material system of choice are AlGaAs/GaAs-heterostructures containing a high-mobility two-dimensional electron gas with a phase coherence length of typically 10 – 30 microns at low temperatures. Definition of quantum dots is usually achieved by electron beam lithography and adjacent deposition of Schottky field-effect gates. The quantum dots contain roughly 10 - 100 electrons and are attached to metallic leads by tunneling barriers. The main advantage of heterostructures is the high degree of perfection with which its electronic, photonic and phononic properties can be tailored. These materials already enabled ground breaking work which demonstrated that quantum dots in the few electron limit show not only charge quantization, but reveal a discrete energy spectrum similar to real atoms. The major appeal of quantum dots or artificial atoms and molecules in the context of quantum computing of course is the promise that semiconductor technology eventually will enable efficient up-scaling of fundamental circuits necessary for quantum data processing. It has to be noted that the low temperatures required for the measurements enable clear demonstration of operation of the circuits, so that these concepts finally can be implemented into data processing automatons running at room temperature.

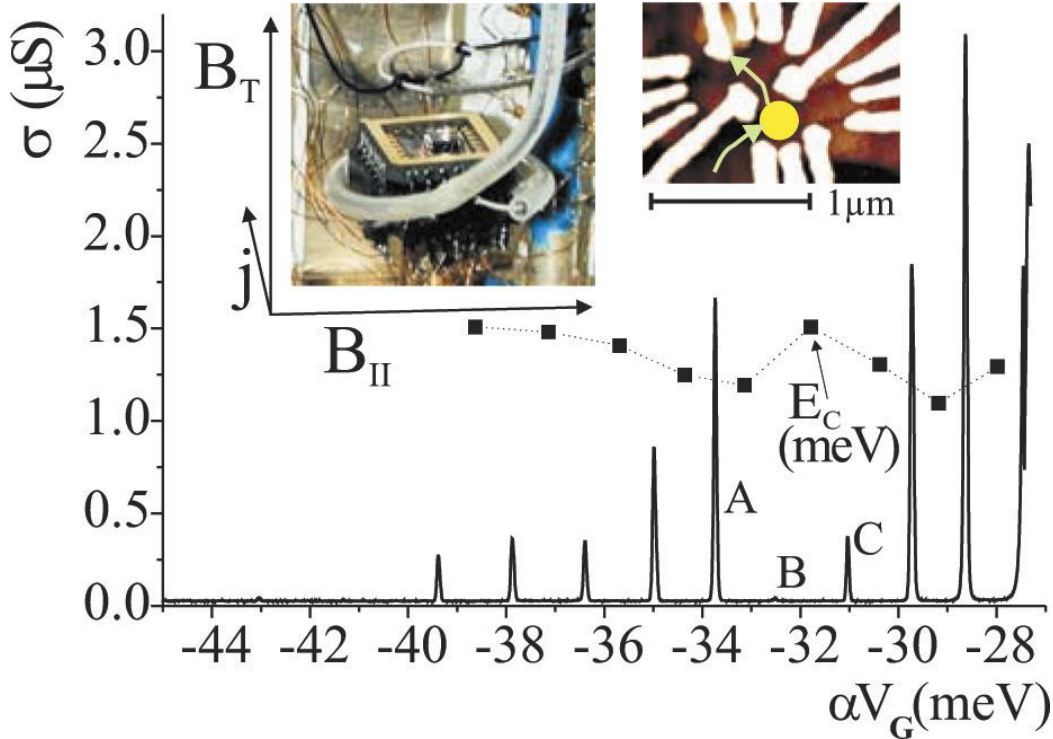


Fig.1: Measurement on a single quantum dot showing spin blockade of transport at the peak marked B. The resonances left and right indicate single electron tunneling (A) and a quantum dot state with a large angular momentum (C). Left inset shows sample stage with the antenna coupling radiation from radio to microwaves. Right inset gives an AFM picture of the quantum dot geometry, allowing definition of up to four connected dots. For the current investigation we focus on a single dot only (yellow circle). This setup has the specific advantage to be equipped with a vector magnetic field, i.e. allowing 6 T perpendicular to the sample's plane and 3 T in plane. This is an essential ingredient when spin effects are to be considered.

The sample is placed on the mixing chamber of a dilution refrigerator operating in the range of 5 mK to 1.5 K. Part of the sample stage is shown in the left inset of Fig.1. The chip carrier is plugged to a cold finger fabricated from pure silver with an evaporated gold layer for thermal anchoring. In addition the chip is surrounded by two Hertzian loop resonators which couple radio and microwave radiation to the sample. In the center of the figure the chip carrier with the bonded quantum circuit and the support microwave oscillator can be identified. A large number of DC lines are contacting the chip carrier supplying the operating voltages to the electrodes seen in the right inset of Fig. 1. A typical transport measurement on a single dot is seen in Fig. 1: The gate voltage  $V_G$  – calibrated by  $\alpha$  – is ramped adding single electrons one after another to the dot as the conductance peaks indicate. In between Coulomb blockade prevails and electron tunneling is completely suppressed. The remarkable feature we discovered is seen in the sequence of peaks A, B, and C. While A appears to be an ordinary conductance resonance, the amplitude of peak C is already not at the same level as the following peak's. However, the most pronounced difference is found for peak B, which seems to be completely suppressed. In accordance with theoretical predictions [Wei95] this phenomenon is interpreted as spin blockade in the linear response regime (type-II). This

occurs in addition to Coulomb blockade and directly reveals that not only the repulsive charging energy has to be overcome but in addition spin selection rules of electrons tunneling through the dot have to be satisfied. In turn this strict requirement indicates that the spin quantum number can be approximated by  $S_z > 3/2$ , increasing the possible coupling to the nuclear spins via the hyperfine interaction.

The measurements on nuclear spin relaxation probed by a single quantum dot in a high-mobility electron gas are shown below in Figs. 2 – 4: The dot is tuned into a high spin state applying the mechanism of spin blockade. This effectively leads to a spin transfer from the electronic to the nuclear spin system. Applying electron spin resonance the transfer mechanism can directly be tuned. Additionally, the dependence of nuclear spin relaxation on the dot gate voltage is observed. We find electron-nuclear relaxation times to be longer than 10 minutes.

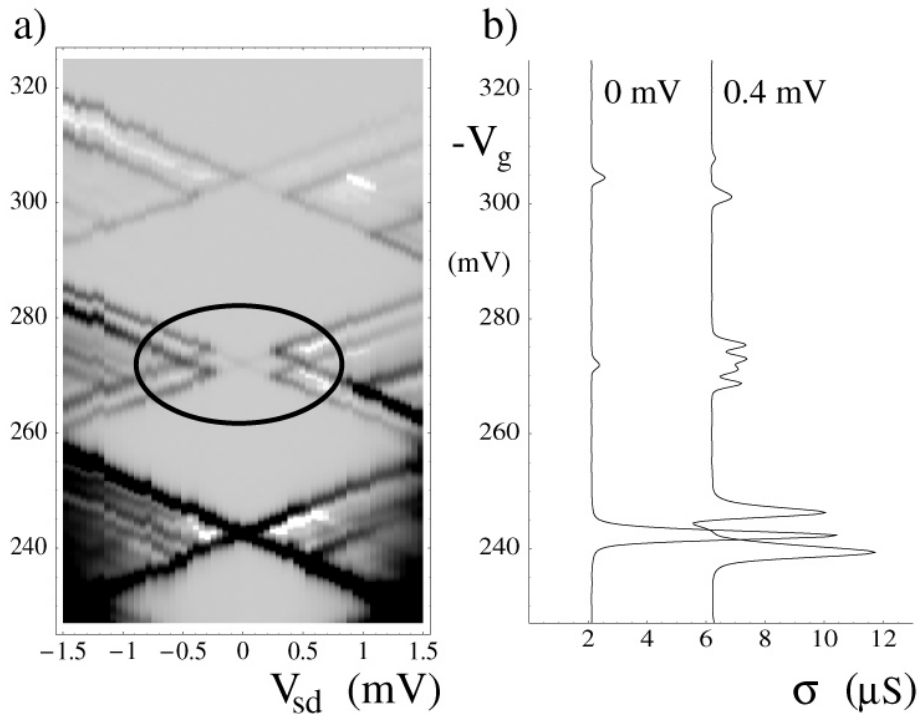


Fig. 2: (a) A typical transport spectrum of a single quantum dot: The bias across the sample  $V_{sd}$  is stepped while a gate voltage  $V_g$  is scanned. The resulting differential conductance  $\sigma$  is depicted in gray scale, where gray corresponds to complete suppression of transport, while for black we calibrated  $G > 7 \mu\text{S}$ . The white lines indicate negative differential conductance. The diamonds in the center give the regions of Coulomb blockade – left and right to these regions of single electron tunneling are found. The distinct pattern of conductance resonances is a fingerprint of the excitation spectrum of an artificial atom. (b) Single line traces taken from (a) for clarity.

In Fig. 2(a) a full scale transport spectrum of a single quantum dot is plotted in a gray scale representation: Black represents a conductance of  $G > 7 \mu\text{S}$ , gray indicates complete suppression, i.e. Coulomb blockade of electron transport and white indicates negative differential conductance (NDC). The diamond like structure in Fig. 2(a) gives

information about the capacitance of the quantum dot and the charging energy. The resonance lines on the left and right to the Coulomb diamonds on the other hand indicate single electron tunneling and reveal discrete excited states of the artificial atom. At the resonance position encircled we found strong suppression of electron tunneling due to spin blockade (type-II) with a total number of 50 electrons in the quantum dot. By adjusting the gate voltages properly the different ground and excited states can be addressed individually and spin blockade can be overcome. Interestingly at non-zero drain/source bias we also find regions of NDC being interpreted as spin blockade of type-I according to [Wei95]. In Fig. 2(b) single line plots from the gray scale pattern are taken out for clarity: traces are given for zero drain/source and a bias of 0.4 mV. The high degree of symmetry of the diamond pattern suggest that the tunneling barriers of the quantum dot are reasonably symmetrical, i.e. we can be more than confident that conductance suppression is indeed caused by a spin state in the quantum dot.

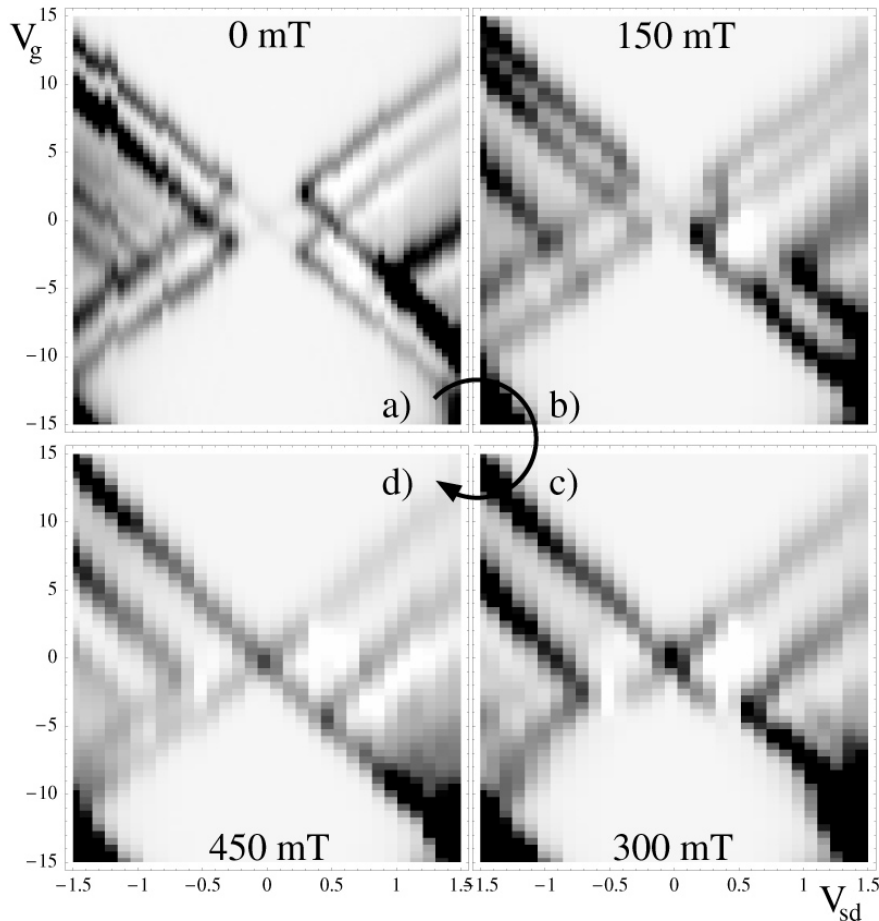


Fig. 3: Part of the transport spectrum shown in Fig. 2 (circled region – plot is in axis of drain/source and gate voltages  $V_{sd}$  and  $V_g$ ): Coulomb blockade is given in white, while a non-zero conductance appears in gray and black. As seen without any perpendicular magnetic field applied conductance at zero bias is suppressed. Increasing the field over 150 mT, to 300 mT and finally 450 mT, conductance is re-established and the conventional single electron tunneling prevails. This obviously demonstrates that the quantum dot's spin degree of freedom is responsible for the blockade of transport at zero bias.

Further evidence for the occurrence of spin blockade we derive from temperature dependent measurements of the spin gap and most notably from quenching this gap in a perpendicular magnetic field as shown in Fig. 3. The spin gap is plotted in the usual gray scale plot at a variety of magnetic field values: At first spin blockade and NDC is well pronounced – with the increase of some 100 mT the gap is considerably quenched and single electron tunneling re-emerges. Apparently we have realized a quantum system with an adjustable spin state which can be applied for quantum information storage as well as manipulation.

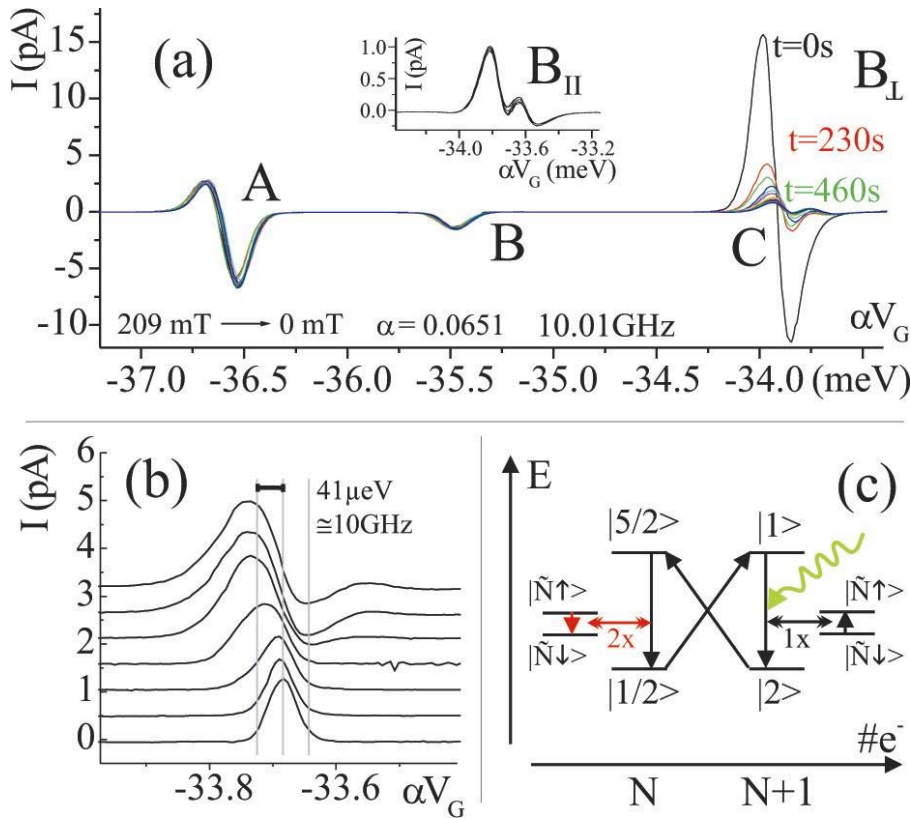


Fig.4: (a) Quantum dot photocurrent peaks A, B, and C under microwave radiation at 10.01 GHz – the particular feature of this measurement is that prior to taking these data the perpendicular magnetic field was ramped from  $B_T = 0$  T to 487 mT in  $t_r \sim 11$  min, maintained at this value for  $t_m \sim 8$  min, and reduced to  $B_T = 0$  T within 11 min. Evidently peak A gives the conventional rectification signal, while the spin blocked transition at B reveals a backward current only. The most prominent feature is found at peak C: relaxation back into the ground state needs over 10 min. This strong hysteresis is attributed to the electron spin flip coupling to the nuclear moments via hyperfine interaction. The inset gives the same measurement for peak C, using a parallel field orientation. Obviously no hysteresis is found, indicating the connection between spin and orbital effects. (b) Focusing on resonance C at low microwave power we find resonant photon absorption. As seen the peak of the current pumped in forward direction is shifted by  $\alpha\delta V_G = h \cdot 10$  GHz (at  $B_T = B_{||} = 0$ , relaxed state). (c) Level diagram for the transition of  $N$  to  $(N+1)$  electrons.

As noted above the conductance spectrum of Fig. 1 gives a sequence of three peaks marked by the letters A, B, C. Peak A displays conventional conductance, whereas peak B is nearly completely blocked at low transport voltage and peak C shows a response smaller than average. In subsequent measurements the suspended loop antenna is emitting microwave radiation onto the sample chip: Fig. 4(a) again displays the three peaks now showing the induced photocurrent under irradiation at 10.01 GHz with moderate input power levels. It is very important to note that prior to taking this data trace the perpendicular magnetic field was ramped from  $B_T = 0$  T up to 487 mT in  $t_r \sim 11$  min, maintained at this value for  $t_m \sim 8$  min, and subsequently reduced to  $B_T = 0$  T within 11 min. As seen peak A gives the conventional rectification signal with a forward and backward pumped electron current. Surprisingly, the spin blockade transition at B reveals a backward current only, which indicates a certain spin texture in the quantum dot. This assumption gains evidence when focusing on resonance C – which is featuring a high spin state, since it is located next to the spin blocked peak: after ramping  $B_T$  the relaxation back into the ground state needs additionally more than 10 min. We found  $B_{T,max} = 40$  mT, as well as ramping times and a waiting period of  $t_{r,max} \sim 11$  min to be sufficient for clearly demonstrating the effect.

This strong hysteresis is attributed to the electron spin flips in the dot coupling to the nuclear spins via the hyperfine interaction. Even at very low magnetic fields as in our case, a straightforward approximation of the induced nuclear magnetic field gives  $(A/N^{1/2}) g^* \mu_B$  where  $A$  is the hyperfine interaction constant,  $N$  the number of nuclei engulfed by the quantum dot,  $g^*$  the effective electron  $g$ -factor and  $\mu_B$  the Bohr magneton. For  $g^* = -0.44$  and a typical size of a quantum dot in an AlGaAs/GaAs-heterostructure with a diameter and height of 126 nm and 10 nm we have  $N \sim 2.2 * 10^7$  nuclei and obtain a resulting field of  $\sim 0.3$  mT [Lya02].

The inset in Fig. 4(a) gives an identical measurement for peak C in a parallel magnetic field –as seen no hysteresis is observed. This leads us to the conclusion that orbital effects bound to a high spin state are responsible for coupling to the nuclear magnetic moments as assumed by Lyanda-Geller *et al.* [Lya02]. A pure spin flip would obey Zeeman splitting in a parallel magnetic field as well, and the hysteresis should persist in this case. For peak C the quantum dot can be thought to be in a state where a spin current is circulating on its edge. It allows to transfer momentum from the electronic to the nuclear system and operates as a pick-up loop in turn. In order to verify this picture we performed photocurrent measurements on peak C under increased microwave power and found resonant absorption at 10.01 GHz as seen in Fig. 4(b). The forward current peak of the ground state results from minimal forward bias. It eventually develops a side band at the resonance energy of  $hf \sim 41$   $\mu$ eV where a single photon is absorbed switching the total spin by  $\Delta S_z = 1$ .

A possible level scenario of this spin flip operation is given in the diagram of Fig. 4(c): the transition at resonance peak C from  $n$  to  $(n + 1)$ -electrons with the spin flips involved starts from an  $n$  electron system with total spin  $S_z$  and results in a final  $(n + 1)$  electron state with spin  $S'_z$  so that  $\Delta S_z = S_z - S'_z > 1/2$ . Hence, ground state (GS) transitions from  $n$  to  $(n + 1)$ -electrons are prohibited due to spin blockade. Nevertheless, adding an electron with  $S_z = +1/2$  to the  $n$  electron system and assuming a small but finite bias voltage the dot can be brought into an excited state (ES) of the  $(n + 1)$  electron system with a total spin of  $S_z = 1$ .

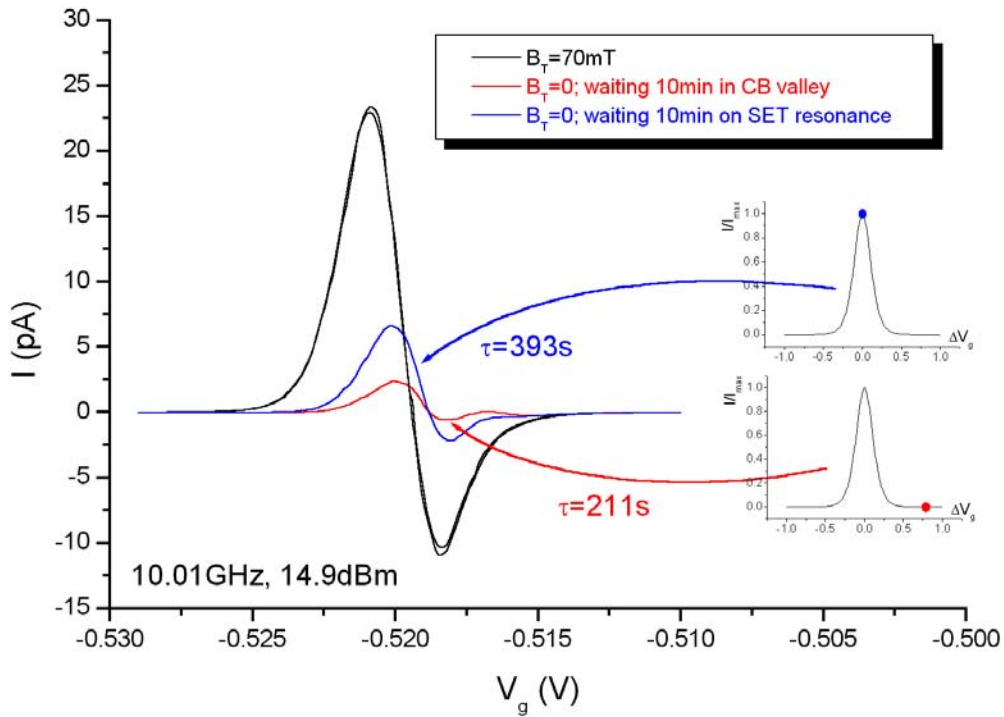


Fig. 5: Time dependence of the hysteresis due to the Overhauser shift (main plot) probed in two different current states of the quantum dot (rhs insets). In resonance (blue position) relaxation into the ground state is slower than in the off-resonance state (red position). This counter intuitive variation of the electron-nuclei relaxation mechanism agrees perfectly with the prediction by Lyanda-Geller *et al.* [Lya02].

Relaxation into the ground state via hyperfine coupling to the nuclear spin system comprises a change in spin quantum number by  $\Delta S_z = 1$ , the total spin conservation of the hyperfine interaction results in a flip-flop process of electron and nuclear spins. This flips the spin of a nearby nucleus – as before, an electron with  $S_z = -1/2$  can now tunnel off the dot under a finite bias voltage, leaving the system in the resulting excited state  $S_z = 5/2$ . Finally, the cycle is completed by a second hyperfine relaxation, this time involving not one but two spin flips. This important asymmetry leads to a net polarization of the lattice of nuclear spins. This is interpreted that the quantum dot at resonance C is operated as a spin filter and inverter. Thus, a polarization of the nuclear spins even without supporting

microwave radiation is possible, as long as a weak magnetic field perpendicular to the surface provides an orientation. Pulsed radiation on the other hand is the method of choice for manipulating the transfer rates and thus controlling spin flow.

Finally we want to address the change in nuclear relaxation time in dependence of the quantum dot's resonance state as Lyanda-Geller *et al.* [Lya02] investigated in their calculations: Again we focus on resonance C in a perpendicular field orientation with the field cycling as introduced above. The main difference now is that relaxation into the ground state after switching off  $B_T$  is not monitored sweeping continuously over the gate voltage range. The quantum dot is kept either at a gate voltage in resonance  $V_{G,res}$  or off resonance  $V_{G,off}$  as exemplified schematically in the insets of Fig. 5. After waiting 10 min, a trace of the peak is taken. As can be seen, in the case of SET resonance the relaxation slows down considerably. This increase in life time at first seems to be counterintuitive, as already stated by the authors of [Lya02]. However, a non-negligible spin-orbit interaction in combination with the differing nature of coupling processes in separate gate voltage regimes can be shown to act accordingly. In addition, considering that in our case the nuclear polarization is build up by a spin current in the quantum dot, this current 'refreshes' the polarization each time an electron tunnels into the dot. Thus a slowdown of nuclear spin relaxation at a conductance resonance seems reasonable as well.

Specific accomplishments include:

- Development of a new probing mechanism of coupled quantum dots in close analogy to quantum non-demolition (QND) methods [Hol02]
- Found spin blockade in quantum dot [Hue02a]
- Applied spin blockade for storing quantum information in nuclear spin system of the crystal lattice via hyperfine interaction [Hue02b] – as suggested by Kane in 1998 [Kan98]
- Relaxation time exceeds 20 minutes at low temperatures [Hue02b]

## Conclusions

In the first part of the project we build and tested the on-chip microwave source operating at low temperatures. This included setting up the necessary dilution refrigerator, cabling and sample stage. We successfully probed single and coupled dots and were able to show the coherent coupling of two dots by microwave spectroscopy. As it turned out the spin degree of freedom of the electrons captured in the quantum dots is best way of processing phase coherent information. This comprises the complex interaction of two dots, since the coupled wave function strongly relies on its spin and spatial parts. In conclusion the techniques and methods we have developed during the course of this project allow us to control electron charge and spin in single and coupled quantum dots. By making use of the dots' environment we have shown how to realize one of the prime suggestions for quantum computing by Kane [Kan98]. This concept relies on the spin interaction of electrons and nuclei: The mobile electrons ensure quick response to an externally applied switching signal such as electron spin resonance with maximal phase coherence times of the order of 10 – 100 nsec. On the other hand the nuclei offer phase relaxation times in the range of minutes and eventually hours. Hence, the combination of both in a quantum box is the basic element for a quantum information storage cell.

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