Spin pairs in a weakly coupled many-electron quantum dot

S. Hellmüller,* D. Bischoff, T. Müller, M. Beck, K. Ensslin, and T. Ihn
Department of Physics, Solid State Physics Laboratory, ETH Zurich, 8093 Zurich, Switzerland
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We report the observation of an unusually large number of consecutive spin pairs in a weakly coupled many-electron GaAs/AlGaAs quantum dot. The pairs are identified due to pairwise parallel shifts of Coulomb resonances in a perpendicular magnetic field. Using a nearby quantum point contact for time-resolved charge detection, the tunneling rates are investigated as a function of gate voltage and magnetic field. We compare our experimental data to a single-level transport model and discuss possible reasons for deviations.

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I. INTRODUCTION

Quantum dots (QDs) can be regarded as artificial atoms [1–3] with an addition spectrum that depends on the single-particle state defined by the confinement [4,5] as well as on the Coulomb interaction and the exchange interaction between the electrons [6]. A successive filling of a spin-up electron and a spin-down electron into the same orbital state is called a spin pair. The addition spectra of quantum dots have been investigated in various materials and for different geometries [7–21]. While for QDs formed in carbon nanotubes a successive filling with spin pairs is normally observed [17,18], this is usually not the case for QDs in GaAs/AlGaAs heterostructures [7,12–16]. Indeed, the exchange interaction in GaAs/AlGaAs heterostructures is typically large enough to prevent a successive filling of spin-up and spin-down electrons into a QD by favoring parallel spins, and hence, spin pairs are observed only occasionally [12,16]. Signatures of spin pairing [15] were also observed in a statistical analysis [22–25] of even and odd nearest-neighbor-peak spacings [26,27].

In this paper we present the measurement of many consecutive spin-pair candidates in a lateral GaAs/AlGaAs quantum dot similar to the QDs investigated in previous studies [12–16,26]. Unexpectedly, pairwise parallel shifts in magnetic field of 20 or more consecutive Coulomb peak pairs [28] are observed. Tunneling rates are recorded as a function of magnetic field and detuning of the chemical potential of the QD using time-resolved charge detection techniques.

II. DEVICE AND SETUP

Our device, shown in Fig. 1(a), contains a quantum dot with a nearby quantum point contact (QPC) formed with metal electrodes. They are fabricated by electron-beam lithography on a GaAs/AlGaAs heterostructure with a two-dimensional electron gas (2DEG) located 34 nm below the surface. The 2DEG has a mobility of 33 m2/Vs and an electron density of $4.8 \times 10^{15}$ m$^{-2}$ at a temperature of 4 K. The device is measured in a dilution refrigerator at an electron temperature $T_e \approx 90$ mK, which corresponds to 7.7 $\mu$eV.

The QD is investigated in a regime where it is populated by at least 160–200 electrons and where the current through the dot is equal to or less than 0.25 fA. Accurate measurements of such small currents are challenging. We therefore use a QPC charge detector capacitively coupled to the QD in order to detect the tunneling of single electrons in a time-resolved manner. A bias voltage of 200 $\mu$V is applied to the QPC. The resulting time-resolved current is recorded with a sampling rate in the range of 500 kHz to 1 MHz. The time traces contain a random telegraph signal in which the steps correspond to single electrons leaving or entering the QD.

For the data evaluation it is crucial that the signal-to-noise ratio (SNR) is large enough to avoid false counts. Therefore, after sampling, the data are digitally filtered (5 or 15 kHz, eighth-order Bessel filter) and resampled before we extract the number of electrons which entered and left the QD during a certain time. From the average times the QD is occupied (unoccupied) by an excess electron, the rate for tunneling out (in) can be determined accurately. Part of a time trace is exemplarily shown in Fig. 1(b). It corresponds to a point inside the white diamond in Fig. 1(d).

III. EXPERIMENTAL RESULTS

A. Observation of parallel shift of neighboring Coulomb peaks at finite magnetic fields

The evolution of the Coulomb peaks in a magnetic field $B$ of up to 1.5 T is shown in Fig. 1(c) for $V_{bias, QD} = 0$ V. Their positions fluctuate in magnetic fields up to 1.5 T by $\Delta V_{PG} = 1.5–3$ mV, which is about 15%–30% of their spacing [29]. A pairwise correlation is clearly visible, suggesting the occurrence of spin pairs. In our sample the perpendicular magnetic field dependence is determined by the orbital wave functions rather than by the spin. Therefore, two parallel shifting peaks indicate the same orbital wave function and therefore opposite spins due to the Pauli principle [30]. Differences in the fluctuations between the first and the second peak of a spin pair [31,32] are observed occasionally but are not the focus of this paper. Two examples of such deviations are visible for peaks X and X' at $\approx 0.57$ T and at $\approx 0.27$ T, marked by blue arrows in Fig. 1(c).

In total, 46 consecutive Coulomb resonances are investigated, and the pairwise correlation for all of them is as clear as for those shown in Fig. 1(c). In five cases, however, triples are observed instead of pairs. The triples are most likely in which one Coulomb resonance is recorded twice due to corresponding charge rearrangements [33]. These charge rearrangements can be clearly identified in Coulomb-blockade

*hesarah@phys.ethz.ch
FIG. 1. (Color online) (a) SEM image of the measured GaAs/AlGaAs device including a QD and a nearby QPC charge detector. (b) Part of a time trace (5 kHz, eighth-order Bessel filter), corresponding to a point inside the white diamond in (d). (c) Evolution of some of the Coulomb peaks (numbers VIII–X′) in a perpendicular magnetic field $B$ at zero $V_{\text{bias}}$. Plotted is the number of electrons entering and leaving the QD per second as a function of the plunger-gate voltage $V_{\text{PG}}$. The parallel shift of pairs of neighboring peaks suggests that spin-up and spin-down electrons are filled pairwise into the QD in this regime. (d) Coulomb diamonds corresponding to the peaks in (c). Plotted is the number of electrons passing the QD per second extracted from 200-ms-long time traces similar to the one shown in (b).

They are caused by single-charge traps close to the quantum dot and occur at a certain plunger-gate voltage. Additionally, several times over the total measurement period of about 4 months, spontaneous small charge rearrangements happened. For example, between the measurements in Figs. 1(c) and 1(d), a shift of 2 mV occurred. The fluctuations and the order of the peaks did not change due to the rearrangements. For small rearrangements no triples were observed, and the positions of all peaks shifted only slightly. However, for large rearrangements, which happened at a certain plunger-gate voltage, triples were measured, and all peaks occurring at larger plunger gate voltages are shifted with respect to the others.

C. Investigation of tunneling rates at zero magnetic field

In Fig. 2(a) two other spin pairs (II/II′ and III/III′) are shown as a function of plunger-gate voltage $V_{\text{PG}}$ and perpendicular magnetic field $B$. The color code corresponds to the number of electrons entering and leaving the QD per second extracted from the QPC current measured as a function of time.

D. Tunneling rates as a function of energy-level detuning and magnetic field

For further investigation, the tunneling rates are determined also as a function of perpendicular magnetic field $B$. The
results for pair III/III′ [shown in Figs. 3(f) and 3(g)] are presented in Figs. 3(a)–3(e). In Fig. 3(a) the tunneling-out rate \( \Gamma_{\text{out}} \) is plotted, and clear steplike transitions are visible (marked by arrows). A cut of Figs. 3(a) and 3(d) is presented in Fig. 3(c) to emphasize these steplike transitions in the tunneling-out rate of peak III. The data corresponding to peak III are plotted in Figs. 3(b), 3(e), and 3(g). The steplike features can be explained by a crossing of two energy levels [36]. Additionally, four cuts at various magnetic fields are plotted in Figs. 3(h)–3(k) and are discussed later on in the paper. In the following section we focus on the investigation of the individual tunneling rates of single levels.

IV. MODEL TO EXPLAIN TUNNELING RATES DEPENDING ON THE SPIN-CONFIGURATION OF THE QUANTUM DOT

A. Model to explain tunneling rates considering a single energy level

Spin-up and spin-down electrons are filled alternately into a single spin-degenerate energy level of the QD in a model in which the exchange interaction is neglected. The rates for tunneling into and out of the QD are expected to behave as depicted in Figs. 4(a) and 4(b), assuming zero bias voltage, equal electrochemical potential for \( \uparrow \) and \( \downarrow \) electrons in the QD \( \mu_\uparrow = \mu_\downarrow \equiv \mu \), and equal tunnel coupling for both spin species. For these assumptions the spin-relaxation time has no influence on the resulting tunneling-in and tunneling-out rates. The red arrows mark options for a tunneling-in or -out event, and the gray (white) circles refer to occupied (unoccupied) states. The first electron of the spin pair can either be a spin-up or a spin-down electron [see Fig. 4(a)]. As soon as one electron tunnels into the QD, a second electron can enter only if the spin is opposite to the one of the first electron of the spin pair and if it has a larger energy due to the extra charging energy needed [see Fig. 4(b)]. If the QD is occupied by the first electron of the spin pair, this electron has the chance to tunnel out. If, however, both the spin-up and the spin-down electrons are in the QD, either of them can tunnel out. Therefore, the tunneling-in rate for the first electron of the spin pair is expected to be twice as large as the tunneling-out rate and vice versa for the second electron of the spin pair [37]. Here we used the additional assumption that the tunnel coupling is independent of any extrinsic parameter such as gate voltage and magnetic field.

B. Comparison of model and measured tunneling rates at zero magnetic field

Even though there is evidence that spin pairs are present in the investigated QD regime due to the parallel Coulomb peak evolution at finite magnetic field, the simple model presented in Figs. 4(a) and 4(b) predicts the rates observed in the experiment only approximately.

As an example we consider peaks II/II′ and III/III′ in Fig. 2(b). The first electron of the spin pair is marked with a prime symbol, while the second electron has no symbol. For peaks III (\( \Gamma_{\text{in}} \), \( \Gamma_{\text{out}} \)) and III′ (\( \Gamma_{\text{in}} \), \( \Gamma_{\text{out}} \)) we find \( \Gamma_{\text{in}}/\Gamma_{\text{out}} = 1.65 \pm 0.02 \), \( \Gamma_{\text{out}}/\Gamma_{\text{in}} = 1.76 \pm 0.02 \), \( \Gamma_{\text{in}}/\Gamma_{\text{out}} = 1.41 \pm 0.03 \), \( \Gamma_{\text{out}}/\Gamma_{\text{in}} = 2.07 \pm 0.02 \). Similarly, for the other presented pair (II, II′) we find ratios of \( 3.03 \pm 0.12 \), \( 1.91 \pm 0.06 \), \( 1.89 \pm 0.08 \), and \( 3.07 \pm 0.09 \). Investigating the tunneling rates of 12 pairs/triples at zero magnetic field, we find that most
of them do not behave precisely as predicted by the model, meaning that the ratio $\tilde{\Gamma}_{\text{out}}/\tilde{\Gamma}_{\text{in}}$ of the tunneling rates is often neither exactly 2 nor $1/2$.

Possible reasons why the model does not hold at zero magnetic field are discussed in the following. The wave-function overlap of an electron in the QD with the lead might depend on the spin with the consequence that $\Gamma_{\text{in}}^{\uparrow} = \Gamma_{\text{in}}^{\downarrow}$ is not necessarily true. However, the effect that electrons with a specific spin direction ($\uparrow$ or $\downarrow$) prefer to tunnel would only bring the ratios of tunneling rates closer to 1. Hence, this argument does not explain the measured tunneling rates. The capacitive cross talk of the plunger gate to the source and drain barrier can influence the tunneling rates as well. The barriers.

FIG. 3. (Color online) (a)–(e) The tunneling rates of the Coulomb peak pair III/III’ [shown in (f) and (g)]. (a) and (b) The tunneling-out rate and (d) and (e) the tunneling-in rate are presented as a function of plunger-gate voltage and perpendicular magnetic field $B$ at zero bias voltage ($V_{\text{bias,0D}} = 0$). (c) A cut along the dotted lines in (a) and (d). The number of energy levels participating in transport decreases stepwise from left to right. The blue and the green arrows mark transitions from one to two energy levels contributing to transport. The energy level marked by blue arrows decreases in energy for increasing magnetic field. The opposite is true for the one marked by green arrows. The curved, finely dotted lines in (a), (b), (d), and (e) are copies of the dotted black lines in (f) and (g) and mark the position of the Coulomb resonances. (h)–(k) $\tilde{\Gamma}_{\text{out}}/\tilde{\Gamma}_{\text{in}}$ as a function of magnetic fields of (h) $B = 0.05$ T (peak III), (i) $B = 0.15$ T (peak III’), (j) $B = 0.29$ T (peak III’), and (k) $B = 0.42$ T (peak III), with tunneling rates of (h) $\tilde{\Gamma}_{\text{in}} = 4.60 \pm 0.06$ kHz, $\tilde{\Gamma}_{\text{out}} = 5.54 \pm 0.07$ kHz, (i) $\tilde{\Gamma}_{\text{in}}' = 6.17 \pm 0.10$ kHz, $\tilde{\Gamma}_{\text{out}}' = 3.53 \pm 0.05$ kHz, (j) $\tilde{\Gamma}_{\text{in}}' = 5.02 \pm 0.08$ kHz, $\tilde{\Gamma}_{\text{out}}' = 3.09 \pm 0.04$ kHz, and (k) $\tilde{\Gamma}_{\text{in}}' = 2.87 \pm 0.03$ kHz, $\tilde{\Gamma}_{\text{out}}' = 6.6 \pm 0.13$ kHz.
orbital degeneracies might be another reason for the measured tunneling rates at zero magnetic field, which is partly shown in Fig. 2.

C. Comparison of model and measured tunneling rates at finite magnetic field

The tunneling rates at small finite magnetic fields are analyzed using the data shown in Fig. 3. For the first electron of the spin pair the extracted ratios of the tunneling rates  $\Gamma_{\text{in}}/\Gamma_{\text{out}}$ are $1.75 \pm 0.05$ [Fig. 3(j)] and $1.62 \pm 0.05$ [Fig. 3(j)], which are also marked in Figs. 3(a) and 3(d) by dash-dotted white lines. For the second electron the ratios of the tunneling rates  $\Gamma_{\text{in}}'/\Gamma_{\text{out}}'$ are $1.21 \pm 0.03$ [Fig. 3(h)] and $2.30 \pm 0.07$ [Fig. 3(k)], also marked in Fig. 3(b,e) by dash-dotted white lines. Even though the measured values are around the expected ratio of  $\Gamma_{\text{in}}'/\Gamma_{\text{out}}' = \Gamma_{\text{in}}/\Gamma_{\text{out}} = 2$, significant deviations can be identified. We therefore conclude that the assumptions for the model described in Sec. IV A and Fig. 4 are not entirely valid for our measurements. Specifically, the dependence of the tunneling rate on the magnetic field and on the energy is not included in the model. In addition we observe signs of resonances in the leads. A potential candidate for such a resonant state in the leads is marked by an orange arrow in Figs. 3(b) and 3(h).

V. CONCLUSION

The presented data reveal many consecutive spin pairs in a regime in which at least 160–200 electrons populate the QD. Surprisingly, all investigated Coulomb peaks appear in pairs (or in triples in the case where one of the peaks is measured twice due to a charge rearrangement). One possible reason why exchange interaction is negligible for these measurements might be the shielding by the top gates located only 34 nm above the 2DEG in this heterostructure. Furthermore, the tunneling rates are investigated as a function of plunger-gate voltage and magnetic field. A comparison of the data with a standard model reveals discrepancies between data and expectations. However, resonances in the leads and energy-dependent tunnel barriers seem to be promising candidates to explain the deviation.

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[30] Parallel shifting peaks can indicate parallel spins if the peak shifts are due to the Zeeman effect. In carbon nanotubes the Zeeman effect has a major impact on the magnetic field dependence, while for our device the Zeeman effect can be neglected in the magnetic field range investigated.

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