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The transport properties of a quantum dot that is weakly coupled to leads are investigated by using the exact quantum states of a finite number of interacting electrons. It is shown that, in addition to the Coulomb blockade, spin selection rules strongly influence the low temperature transport and lead to experimentally observable effects. Transition probabilities between states that correspond to successive electron numbers vanish if the total spins differ by $| \Delta S | > 1/2$. In nonlinear transport, this can lead to negative differential conductances. The linear conductance peaks are suppressed if transitions between successive ground states are forbidden.

Periodic oscillations of the linear conductance of quantum dots in AlGaAs/GaAs heterostructures are well established consequences of the Coulomb repulsion between the electrons at sufficiently low temperatures [1,2]. They are observed as a function of the carrier density which can be tuned via an external gate voltage. When the quantum dot and the reservoirs are weakly coupled, transport is dominated by the quantum mechanical properties of the isolated dot. For small transport voltages and at low temperatures the current is blocked (Coulomb blockade) if $\mu_L \approx \mu_R \neq E_0(n) - E_0(n - 1)$. $\mu_{L/R}$ is the chemical potential of the left (right) reservoir, $E_0(n)$ the ground state energy of n electrons in the dot. On the other hand, if $\mu_L \approx \mu_R = E_0(n) - E_0(n - 1)$, the number of electrons inside the dot can oscillate between the two values n and $n - 1$ [single-electron tunneling (SET) oscillations], and the current is nonzero.

For finite transport voltage $V \equiv (\mu_L - \mu_R)/e > 0$, transitions between excited states of the confined electrons can also contribute to the electronic transport [3–6]. Additional steps in the current occur when V is enlarged. However, the current is not necessarily increased when, by raising V, the number of transitions between n and $n - 1$ electron states is increased. Spin selection rules can suppress certain transitions and thus *reduce* the current. This happens, for instance, if the electrons in the dot are spin polarized and the total spin $S = n/2$. Then, the electron number can only be decreased by simultaneously reducing the total spin [7]. The basic mechanism of this *spin blockade effect* was discovered by using a quasi-one-dimensional (1D) model for the spectrum of the quantum dot. Negative differential conductances were observed in some of the nonlinear transport experiments [3–6]. In other experiments, it was observed that certain peaks in the linear conductance, which were almost vanishing at very low temperature T, could be recovered by increasing T [8].

In this Letter, we present strong evidence for the spin blockade effect as an explanation of the experimental findings. We consider n-electron states in *square quantum dots* and present novel results which are associated with low-lying excited states that do not necessarily have maximum spin. We show that even in linear transport the current may be suppressed by spin effects, namely, if the total spins of the ground states of successive electron numbers differ by more than $1/2$. The corresponding characteristic and counterintuitive dependencies on temperature and transport voltage of the conductance peaks are observed in experiments [8,9]. Finally, we demonstrate that negative differential conductances can occur close to a conductance peak already at very low transport voltages as observed experimentally. This happens if an excited state with large total spin lies energetically close to the ground state.

As previously, we use the double barrier Hamiltonian $H = H_L + H_R + H_D + H_L^T + H_R^T$ [7]. Here, $H_{L/R}$ describes free electrons with spin in the left/right lead. The electrons in the dot are described by $H_D = H_0 + H_I$. $H_0 = \sum_{l,\sigma}(\varepsilon_l - e\Phi)c_{l,\sigma}^\dagger c_{l,\sigma}$ corresponds to the noninteracting electrons with energies ε_l. The gate voltage V_G and the capacitive influence of the voltages applied to the leads are assumed to be incorporated into an electrostatic potential Φ. H_I is the Coulomb interaction between electrons including spin.

The barriers are represented by tunneling Hamiltonians $H_{L/R}^T = \sum_{k,l,\sigma}(T_{k,l,\sigma}^{L/R}c_{L/R,k,\sigma}^\dagger c_{l,\sigma} + \text{H.c.})$, where $T_{k,l,\sigma}^{L/R}$ are the transmission probability amplitudes. We further assume that the leads are in thermal equilibrium with reservoirs described by the Fermi–Dirac distributions $f_{L/R}(\varepsilon) = \{\exp[\beta(\varepsilon - \mu_{L/R})] + 1\}^{-1}$. The transmittances $t_{L/R} \propto |T_{k,l,\sigma}^{L/R}|^2$ of the two barriers can be different. The current is mainly restricted by the smaller of the $t_{L/R}$. For simplicity, they are assumed to be independent of energy and spin. If they are small as compared to the phase breaking rate τ_φ^{-1}, the (reduced) density matrix for the quantum dot can be assumed to be diagonal. In this limit, the time evolution of the occupation probabilities can be described by a master equation which allows one to consider transport at arbitrary voltages.

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The transition rates between the n-electron states of the dot $| i \rangle$ are calculated in lowest order perturbation theory in H^T. Simultaneous transitions of two or more electrons [10] are of higher order in H^T and neglected. Each of the states is associated with a certain electron number n_i, an energy E_i, a total spin S_i, and a magnetic quantum number M_i. When calculating the energy levels of the electrons by diagonalizing the corresponding n-particle Hamiltonian, the interaction is fully taken into account. This is in contrast to previous investigations [11–13], where transitions between the states were considered within the "charging model."

An electron that leaves or enters the dot through the left (right) tunnel barrier causes transitions between $|i\rangle$ and $|j\rangle$. The corresponding rates are

$$\Gamma_{ji}^{L/R} = \frac{t^{L/R}}{2} |\langle S_i, M_i, \tfrac{1}{2}, \pm\tfrac{1}{2} \mid S_j, M_j\rangle_{\mathrm{CG}}|^2$$

$$\times \left(f_{L/R}(E)\delta_{n_j,n_i+1} + [1 - f_{L/R}(-E)]\delta_{n_j,n_i-1} \right).$$

The electron has to provide the energy difference $E = E_j - E_i$. Most importantly, the rates contain Clebsch–Gordan coefficients $\langle \cdots \mid \cdots \rangle_{\mathrm{CG}}$. They account for the combination of the spin of the incoming or leaving electron with the spin of the initial dot state to the spin of the final dot state and introduce spin selection rules. The quantum numbers S_i and M_i can be changed only by $\pm 1/2$ when one electron enters or leaves. At zero magnetic field when the states with different magnetic quantum numbers M_i are degenerate, an average yields effective spin dependent factors which favor an increase of the total spin [7]. The spin selection rules are a consequence of using the exact correlated n-particle states.

The matrix of the total transition rates between the states of the isolated dot is given by $\Gamma = \Gamma^L + \Gamma^R$. The stationary nonequilibrium populations P_i obey

$$\sum_{j(j\neq i)} (\Gamma_{ij}P_j - \Gamma_{ji}P_i) = 0.$$

They determine the dc current for arbitrary V via

$$I = (+/-)e \sum_{i,j(j\neq i)} P_j\Gamma_{ij}^{L/R}(n_i - n_j). \qquad (1)$$

It equals the number of electrons that pass the left or the right barrier per unit of time.

The spin selection rules influence the transport properties qualitatively. In addition to the Coulomb blockade, further new blocking mechanisms occur. One of them is the "spin blockade" discussed previously [7]. It results in negative differential conductances and is related to the population of states with maximal spin $S = n/2$. They occur as excited states both in 1D [14] and in 2D [15] quantum dots. The transitions

$$n, S = n/2 \longrightarrow n - 1, S' \qquad (2)$$

that reduce the electron number starting from the spin polarized state must also reduce the total spin to $S' =$

$(n - 1)/2$. In contrast, states with $S < n/2$ can either increase or decrease S. Thus, the $S = n/2$ state is stable for relatively long times. As a consequence, the current is reduced when a state with $S = n/2$ can be occupied. This spin blockade appears at transport voltages of the order of the excitation energies of the $S = n/2$ states.

The differential conductance versus gate and transport voltage, V_G and V, is shown in Fig. 1 in a grey-scale representation. Along the $V = 0$ axis the peaks in the linear conductance can be observed with the intervals of the Coulomb blockade in between [2]. Lines that intersect at the positions of the peaks in the linear conductance correspond to the ground state to ground state transitions. The regions of the Coulomb blockade are the diamond-shaped areas between these lines. The lines parallel to the edges of the Coulomb blockade areas reflect the dot spectrum [9,16]. Similar features have been observed in experiments [6]. When either V_G or V are changed, the set of the dot states involved in the transport changes. At $T = 0$, this leads to jumps in the current. In Fig. 1 (left) the energy spectrum of a 1D quantum dot [15] has been used. In general, a finite transport voltage V broadens the conductance peaks and leads to fine structure which is characteristic for the dot spectrum and is in general asymmetric [3,17]. The asymmetry is reversed when reversing the voltage [7,18] if the barriers are not equally transparent, in agreement with experimental findings [4]. Bright regions that correspond to negative differential conductances occur preferably when the lower chemical potential is attached to the less transmitting barrier and transitions of the type (2) limit the current.

Another spin blockade effect occurs if the total spins of the ground states that correspond to electron numbers n and $n - 1$ differ by more than $1/2$. It can even be observed in linear transport, namely, when

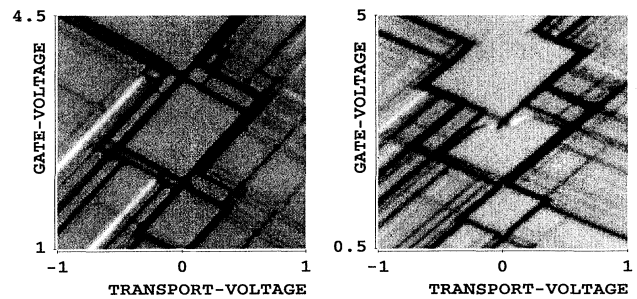$$(E_0(n), S) \longleftrightarrow (E_0(n - 1), S'), \qquad |S - S'| > 1/2.$$

$$(3)$$

FIG. 1. Differential conductance versus gate V_G and transport voltage V in units of E_H/e. Zero conductance inside the diamond-shaped Coulomb blockade regions ($V \approx 0$) corresponds to grey. Dark and bright parts indicate positive and negative differential conductances, respectively. Left: 1D dot. Bright regions are preferably found for $V < 0$ since $t_L = t_R/2$ 7. Right: 2D square dot. Transition between the ground states for $n = 4$ and $n = 5$ is forbidden by spin selection rules.
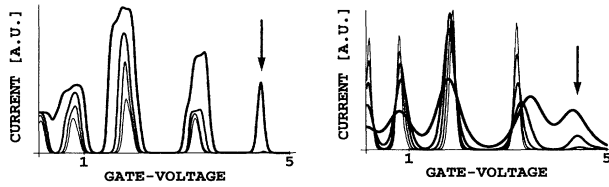
FIG. 2. Current versus gate voltage for a square quantum dot. Left: Transport voltage increases. Curves with growing line thickness correspond to $V = 0.04, 0.06, 0.1, 0.2$ in units of E_H/e. Right: Temperature increases. Increasing line thickness corresponds to $\beta E_H = 100, 80, 60, 40, 20$. The missing peak in linear conductance corresponding to transitions between $n = 4$ and $n = 5$ electrons is recovered (arrow). Unusual behavior of the peak corresponding to the transition between $n = 3$ and $n = 4$ is due to the weakness of the ground state to ground state transition and the population of excited levels with increasing temperature or voltage.

Then the dot is blocked in the n- or the $(n - 1)$-electron ground state, and the corresponding peak in the linear conductance is missing at zero temperature. At finite temperatures and/or transport voltages excited states with appropriate spin values become populated. This can lead to the recovery of a spin-forbidden conductance peak. Such a spin blockade is specific for 2D quantum dots. In 1D the Lieb-Mattis theorem guarantees that the spins of the ground states are always 0 or 1/2 (depending on the parity of n). Thus in a "slim" quantum dot no linear conductance peak should be missing.

Figure 1 (right) shows the differential conductance through a square, hard wall quantum dot. The low energy spectra have been calculated in the low electron density limit by using the method described in [15]. In contrast to 1D dots, states with high spin occur already at low excitation energies and lead to new effects. One prominent feature is the lack of the conductance peak corresponding to the transition between $n = 4$ and $n = 5$ electrons since the spins of the ground states are $S = 0$ and $S = 3/2$, respectively. Finite transport voltages or finite temperatures cause transport through excited states with spins $S = 1$ ($n = 4$) and $S = 1/2$ ($n = 5$). The voltage- or temperature-induced recovery of the conductance peak is shown in Fig. 2. Such an effect was indeed experimentally observed [8].

States with high spin, not necessarily completely spin polarized and which are energetically close to the ground state, can cause additional blocking phenomena. This is shown for the transition between $n = 3$ and $n = 4$ in Fig. 3. In contrast to the spin blockade (2) discussed previously [7], the new mechanism (3) can lead to negative differential conductances even close to the linear conductance peak. Here the lowest $n = 3$ states with $S = 3/2$ and $S = 1/2$ are almost degenerate (their energetic difference is lower than the temperature we have chosen). Within the Coulomb blockade region all transition rates that increase the electron number are exponentially small. At
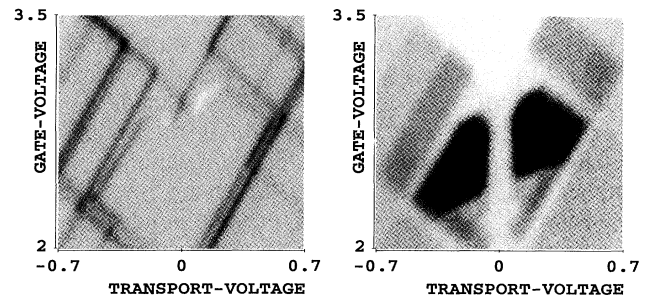

FIG. 3. Left: Region around the transition between $n = 3$ and $n = 4$, magnified and with the energy of the $n = 3$; $S = 3/2$ state being close to the $n = 3$; $S = 1/2$ ground state. At low but finite transport voltage the $S = 3/2$ state becomes populated and transitions to the $n = 4$ ground state are spin forbidden. Negative differential conductances appear. Right: Same region but now showing the population of the $n = 3$; $S = 3/2$ state in dark. If the transport voltages are sufficient to occupy the $S = 3/2$ state it is easily populated, but depopulation is difficult.

$V = 0$ the system is in thermal equilibrium and the three-electron ground state is populated. Already a slightly increased voltage changes the *ratio* between certain rates by orders of magnitude, favoring the occupation of the $S = 3/2$ state (cf. Fig. 3). This is due to a delicate interplay between multiple transitions that connect eventually the lowest $n = 3$ states via at least three intermediate states. Transitions from the $S = 3/2$ state to the $n = 4$ ground state are spin forbidden which causes the pronounced negative differential conductance at low voltages.

In order to simulate the spectrum of a rectangular dot, we enhance slightly the energy of the $n = 3$; $S = 3/2$ state. Figure 4 shows the differential conductance and the stationary occupation probability of this state. Now, the region of negative differential conductance is shifted in V by the excitation energy of the $S = 3/2$ state. When
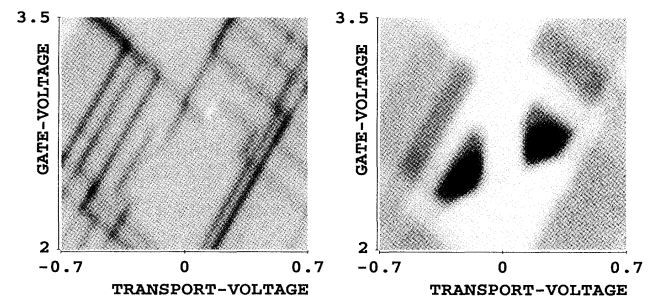

FIG. 4. Left: Region around the transition between $n = 3$ and $n = 4$, magnified and with the energy of the $S = 3/2$ state being slightly increased. At low but finite transport voltage the ground state to ground state transition is blocked and negative differential conductances appear. Right: Same region but now showing the population of the $n = 3$; $S = 3/2$ state in dark.

the $n = 3; S = 3/2$ state starts to contribute to the transport, it attracts a large portion of the population at the expense of the $n = 3; S = 1/2$ ground state. This suppresses the ground state to ground state transition. Only at even higher voltages the $S = 3/2$ state can again be depopulated, and the line corresponding to transport involving the ground states is recovered. Striking features like this can also be detected in the grey–scale representations of the experimental results [6].

In summary, we have shown that spin selection rules qualitatively influence the transport properties of semiconducting quantum dots where electron correlations are important. The excitations of the n-electron system cannot be described by the occupation of single-electron states. Instead, the states that describe the n correlated electrons have to be used, with the total spin as a good quantum number. The latter is changed when an electron enters or leaves the dot. Quantum mechanical selection rules explain in a natural way various experimental observations which cannot be accounted for within the "charging model" where excitations are treated within a one-particle model.

We have proposed two different spin blockade effects. One is related to spin-polarized n-electron states. It leads to negative differential conductances in the nonlinear regime. The other is a more general mechanism. It occurs, for example, when the total spins of the ground states of n and $n - 1$ electrons differ by more than $1/2$. This influences the heights of the linear conductance peaks and is of particular importance for 2D quantum dots. The present results do not include the spatial dependence of the wave function. Preliminary investigations show that the blockade effects discussed here are not markedly influenced by the spatial part of the transition matrix elements. Both spin effects are suppressed by a sufficiently high magnetic field, when the spin-polarized states become ground states [16].

The above spin blockades for correlated electrons, together with the well-known Coulomb blockade, are in principle capable of explaining qualitatively all of the presently observed features in linear and nonlinear transport through quantum dots in semiconductor heterostructures.

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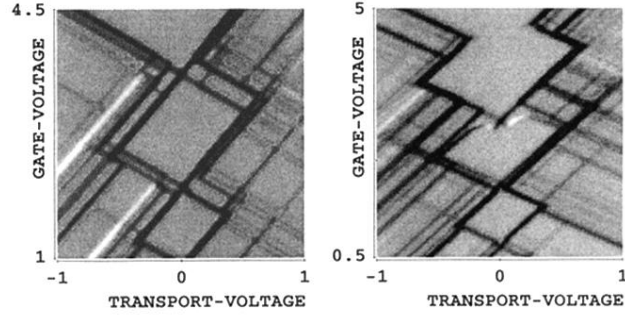[18] W. Pfaff, D. Weinmann, W. Häusler, B. Kramer, and U. Weiss, Z. Phys. B (to be published).

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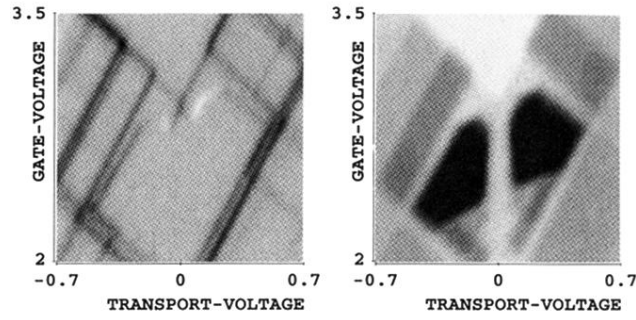
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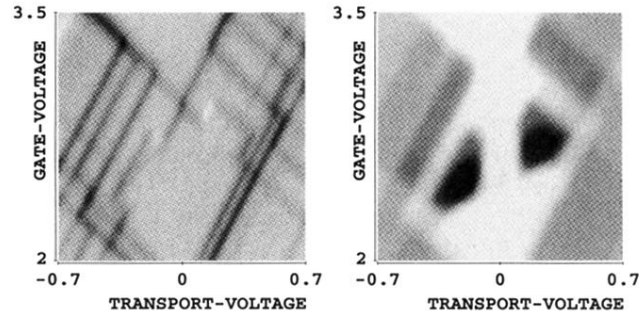
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