Spin Blockade in Nonlinear Transport through Quantum Dots

D. Weinmann\textsuperscript{a,b}, W. Häusler\textsuperscript{b}, W. Pfaff\textsuperscript{a}, B. Kramer\textsuperscript{b}, U. Weiss\textsuperscript{a}

\textsuperscript{a}) Universität Stuttgart, II. Inst. f. Theoret. Physik, Pfaffenwaldring 57, 70550 Stuttgart, F. R. G.
\textsuperscript{b}) Universität Hamburg, I. Inst. f. Theoret. Physik, Jungiusstr. 9, 20355 Hamburg, F. R. G.

and PTB Braunschweig, Bundesallee 100, 38116 Braunschweig, F. R. G.

The influence of excited levels on the nonlinear transport properties of a quantum dot weakly coupled to ideal leads is studied using a master–equation approach. A quantum mechanical model for interacting electrons is used to determine the spectrum of the dot. The current–voltage characteristic shows Coulomb blockade and additional fine structure that is related to the excited states of the correlated electrons. Asymmetric coupling to the leads causes asymmetric conductance peaks. It is demonstrated that spin selection rules can lead to regions of negative differential conductance.

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Periodic oscillations of the conductance through quantum dots that are weakly coupled to leads\textsuperscript{4} are well established consequences of the charging energy of single electrons entering or leaving the dot at sufficiently low temperatures. They are observed in linear transport as a function of the carrier density. At bias voltages larger than the differences between discrete excitation energies within the dot, a characteristic splitting of the conductance peaks is observed\textsuperscript{2–5}. We will demonstrate unambiguously that this is related to transport involving the excited states of \( n \) correlated electrons and that the shape of the peaks depends on the coupling between the quantum dot and the leads. Regions of negative differential conductance have occurred in some of the experiments\textsuperscript{2,5}. It was suggested that the existence of excited states with weaker coupling to the leads than groundstates can lead to such an effect\textsuperscript{6}. Only in the context of the fractional quantum Hall effect (FQHE) states such a behaviour was found\textsuperscript{6}. Our paper presents a novel physical mechanism, that is due to spin selection rules, which leads to such a situation. This ‘spin blockade’ is a consequence of the existence of excited states with different total spin.

As a model, we consider the double barrier Hamiltonian

\[ H = H_L + H_R + H_D + H^T_L + H^T_R, \]

(1)

where \( H_{L/R} = \sum_{k, \sigma} \epsilon_k^L/R \phi_{k, \sigma}^L/R \phi_{L/R, k, \sigma} \) describes free electrons in the left/right lead and

\[ H_D = \sum_{m, \sigma} (\epsilon_m - eV_G) \phi_m^{+} \phi_m \sigma + \sum_{m_1, m_2, m_3, m_4} V_{m_1 m_2 m_3 m_4} \phi_{m_1 \sigma_1}^{+} \phi_{m_2 \sigma_2}^{+} \phi_{m_3 \sigma_3} \phi_{m_4 \sigma_4} \]

(2)

the interacting electrons within the dot. The energies of the noninteracting electrons are \( \epsilon_m \) and \( V_{m_1 m_2 m_3 m_4} \) the matrix–elements of the Coulomb–interaction. The voltage \( V_G \) is the potential change in the dot due to an externally applied voltage and serves to change the electron density in the well.

To specify the dot Hamiltonian \( H_D \) we consider \( n \leq 4 \) interacting electrons in a quasi one–dimensional (1D) square well of length \( L \) including the spin degree of freedom\textsuperscript{7}. We calculated numerically the exact eigenvalues \( E_\nu \) and the corresponding \( n \)-electron states \( |\nu\rangle \) for this correlated electron model. The interaction potential \( \propto ((x - x')^2 + \lambda^2)^{-1/2} \) was used, where \( \lambda (\ll L) \) is due to a transversal spread of the electronic wave function. Since the interaction is spin independent, the \( n \)-electron total spin \( S \) is a good quantum number. The properties of the correlated states and the energy spectrum have been discussed in detail previously\textsuperscript{7}. For not too large electron densities tendency towards Wigner crystallization is found\textsuperscript{8}. In this regime, the excitation spectrum consists of well separated multiplets, each containing \( 2^n \) states. The energetic differences between adjacent multiplets decrease algebraically with electron density. They correspond to vibrational excitations. The considerably smaller intra–multiplet energy differences decrease exponentially. The wave functions of individual levels within a given multiplet differ in symmetry and \( S \). The excitation energies in the lowest multiplet can be calculated analytically\textsuperscript{9} and depend only on one tunneling integral \( t_n \) (Table I). In summary, two different energy scales characterize the \( n \)-electron excitations. We will demonstrate that they can in principle be distinguished by a nonlinear transport experiment.

The barriers are represented by the tunneling Hamiltonians

\[ H_{L/R}^T = \sum_{k, m, \sigma} (T_{k, m, \sigma}^{L/R} \phi_{L/R, k, \sigma}^{+} \phi_{m, \sigma} + h.c.), \]

(3)

where \( T_{k, m, \sigma}^{L/R} \) are the transmission probability amplitudes which we assume to be independent of \( m \) and \( \sigma \). We assume that the phase coherence between the eigenstates of \( H \) is destroyed on a time scale \( \tau_\phi \), which is much larger than the
time an electron needs to travel from one barrier to the other. Thus, the motion of the electrons inside the dot is sufficiently coherent to guarantee the existence of quasi–discrete levels. We assume also that the leads are in thermal equilibrium described by the Fermi–Dirac–distributions \( f_{L/R}(\varepsilon) = (\exp[\beta(\varepsilon - \mu_{L/R})] + 1)^{-1} \). The chemical potential in the left/right lead is \( \mu_{L/R} \) and \( \beta = 1/k_B T \) the inverse temperature. We assume the tunneling rates through the barriers \( t^{L/R} = (2\pi/h) \sum_k |T_{k,m,\sigma}^{L/R}|^2 \delta(\varepsilon_k^{L/R} - E) \) to be independent of energy \( E \). If they are small compared to the phase breaking rate \( \tau_\phi^{-1} \), the time–evolution of the occupation probabilities of the many–electron states in the dot can be calculated using a master–equation.

In contrast to \(^{10,11}\), where changes in the occupation probabilities for one–electron levels were considered, we take into account the populations \( P_i \) of all possible Fock states \( |i \rangle \) of \( H_0 \). Transitions between the latter occur when an electron tunnels through a barrier. Our method allows to determine the stationary nonequilibrium state without further restrictions. Deviations from equilibrium linear in the applied voltage have been mentioned in \(^{12}\). In addition the exact many–electron states of the dot including spin can be taken into account without being restricted to the conventional charging model. A similar method was applied in the FQHE regime without spin \(^6\).

For sufficiently small \( H^T \), simultaneous transitions of two or more electrons \(^{13}\) which are processes of higher order in \( H^T \) are suppressed. Each of the states \( |i \rangle \) is associated with a certain electron number \( n_i \), an energy eigenvalue \( E_i \) and the total spin \( S_i \). The transition rates between states \( |i \rangle \) and \( |j \rangle \) with \( n_i = n_j + 1 \) are given by \( \Gamma_{i \rightarrow j}^{L/R,-} \) and \( \Gamma_{i \rightarrow j}^{L/R,+} \), depending on whether an electron is leaving or entering through the left/right barrier, respectively. Here, \( \Gamma_{i \rightarrow j}^{L/R,-} = \gamma_{j,i} t^{L/R}[1 - f_{L/R}(E)] \), \( \Gamma_{i \rightarrow j}^{L/R,+} = \gamma_{i,j} t^{L/R} f_{L/R}(E) \) and the electron provides the energy difference \( E = E_i - E_j \). As an additional, very important selection rule, we take into account that each added or removed electron can change the total spin \( S_i \) of the \( n_i \) electrons in the dot only by \( \pm 1/2 \). The consideration of the vector coupling Clebsch–Gordan coefficients yields the spin dependent factors

\[
\gamma_{j,i} = \frac{S_i + 1}{2S_i + 1} \delta_{S_i+1/2,S_j} + \frac{S_i}{2S_i + 1} \delta_{S_i-1/2,S_j}
\]

in the transition rates.

The matrix of the transition rates is \( \Gamma = \Gamma^{L,+} + \Gamma^{R,+} + \Gamma^{L,-} + \Gamma^{R,-} \). The master equation for the time evolution of the occupation probabilities \( P_i \) is

\[
\frac{d}{dt} P_i = \sum_{j (j \neq i)} (\Gamma_{i,j}P_j - \Gamma_{j,i}P_i), \quad \sum_i P_i = 1.
\]

From the stationary solution \( \{\bar{P}_i\} \), which is obtained by putting \( d\bar{P}_i/dt = 0 \) in (5) one can determine the dc–current

\[
I \equiv I^{L/R} = (-/+e) \sum_{i,j (j \neq i)} \bar{P}_j(\Gamma_{i,j}^{L/R,-} - \Gamma_{i,j}^{L/R,+}).
\]

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

At zero bias voltage the occupation probabilities of the \( n \)–electron states are given by a Gibbs distribution \( P_i^G = (\exp[-\beta(E_i - \mu n_i)])/Z \) with the chemical potential \( \mu = \mu_L = \mu_R \). They solve the rate equation (5) for all \( t^{L/R} \). \( Z \) is the grand canonical partition function. For temperatures lower and voltages higher than the level–spacings, \( \bar{P}_i \) deviate from equilibrium and cannot be described by defining an effective chemical potential for the dot.

Current–voltage characteristics and conductivity peaks calculated by using the excitation energies given in Table I are shown in Fig I for temperatures lower than the excitation energies. We observe finestructure in the Coulomb staircase consistent with recent experiments\(^2,5\) and earlier theoretical predictions using a different approach\(^{10,11}\). Within our model, the Coulomb steps are not of equal length as in the phenomenological charging model used in \(^{10–12}\). The deviation from the classical behavior is related to the inhomogeneity of the quantum mechanical charge density of the ground state\(^6\). The heights of the finestructure steps are more random due to the non–regular sequence of total spins (cf. Table I) and the spin selection rules. In certain cases, finestructure steps in the \( I–V \) characteristic may even be completely suppressed.

Strikingly, regions of negative differential conductance occur (Fig. 1). They are related to the reduced probability for the states with maximal spin, \( S = n/2 \), to decay into states with lower electron number. In contrast to transitions that involve \( S < n/2 \) they are only possible if \( S \) is reduced. The corresponding Clebsch–Gordan coefficients are smaller than those for transitions with increasing \( S \) (cf. eq. 4) which leads to an additional reduction of the current as compared to the situation where \( S < n/2 \). When the voltage is raised to such a value that an \( S = n/2 \) state becomes involved into the transport, this state attracts considerable stationary population at the expense of the better conducting \( S < n/2 \) states, as can be seen in Fig. 2. Both effects together can then add up to a decreasing
current. The decrease in the \( I-V \) curve becomes less pronounced if \( t^L < t^R \), because then the dot is mostly empty and the \((n-1) \to n\) transitions determine the current. On the other hand if \( t^L > t^R \) the “spin blockade” becomes more pronounced, because the \( n \to (n-1) \) transitions limit the current in this case. Both features can be observed in Fig. 1 and also the experimental data of \(^2,^5\) are clearly consistent with an interpretation that the potential barriers are slightly different. Negative differential conductances can in principle be used to construct a mesoscopic oscillator.

For fixed \( V \), the conductance shows peaks when \( V_G \) is varied which can be described within thermal equilibrium in the limit of linear response\(^1^4\). Only the (correlated \( N \)-electron) ground states are involved at zero temperature. For finite bias voltages, \( eV = \mu_L - \mu_R \), larger than the level spacing, a varying number of levels contribute to the current when \( V_G \) is changed. The conductance peaks split and show structure as is observed experimentally and explained qualitatively in \(^2^,^3,^1^5\) within the charging model. Asymmetric coupling to the leads changes the shape of the peaks considerably, as can be seen in Fig. 1. We propose to explain the “inclination” of the conductance peaks observed in the experiment\(^^2,^3\) by asymmetric barriers and predict that this inclination will be reversed if the sign of the bias voltage is changed. Our result is consistent with the most recent experimental observation of the dependence of this inclination on the ratio of the barrier transmissions\(^3\). Such asymmetric conductance properties can be used to construct mesoscopic rectifiers. Similar rectifying effects were inferred earlier from the high frequency properties of mesoscopic systems containing asymmetric disorder\(^\text{1}^6\).

In summary, we have investigated nonlinear transport through a double barrier taking into account Coulomb interactions, spin and non-equilibrium effects. Occupation probabilities, current–voltage characteristics and conductances versus gate–voltage at finite bias voltage have been calculated using a master equation approach.

Taking into account the quantum mechanics of Coulombically interacting electrons including their spins leads to considerable corrections to the transport as compared to the charging model. The Coulomb blockade intervals in the \( I-V \) characteristic and the conductance peaks are no longer constant. Because of spin selection rules for the correlated electron system, the heights of the single steps in the finestructure of the Coulomb staircase look more or less random. Most strikingly, regions of negative differential conductance occur because for each electron number the state of maximum spin can only contribute to transport by reducing the total spin, and the transition probability into states with lower spin is reduced by the Clebsch–Gordan coefficient. As a consequence, the transition probability into states with lower electron number is reduced. This general feature of a ‘spin blockade’ is not restricted to the quasi–1D model considered here but should also apply to 2D dots considered in the experiments as long as the number of electrons is sufficiently small. Asymmetric barriers cause pronounced asymmetries of the conductance peaks. We predict the reversal of this asymmetry when the bias voltage is reversed.

All of the theoretically predicted features described above are qualitatively consistent with experiment\(^^2,^3,^5\). Further experiments, in particular using ‘slim quantum dots’, are however necessary to clarify the quantitative aspects.

Preliminary theoretical results taking into account a magnetic field in the direction of the transport show that the negative differential conductance is influenced and suppressed at high fields, when the spin polarized states with \( S = N/2 \) become the groundstates\(^1^7\). This is consistent with most recent experimental findings\(^1^8\). To clarify these questions and to be able to make quantitative comparisons with existing experimental data, generalization of the above correlated electron model to 2D\(^9\) is necessary.

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FIG. 1. Current–voltage characteristic ($\mu_R = 0$) and the splitting of the fourth conductance peak at $\mu_L = 0.3E_H$ and $\mu_R = 0$ (inset) of a dot of length $L = 15a_B$ described by the correlated electron model for $\beta = 200/E_H$ ($E_H \equiv e^2/a_B$ Hartree–energy). Tunneling integrals are $t_2 = 0.03E_H$, $t_3 = 0.07E_H$ and $t_4 = 0.09E_H$, numerically determined ground state energies $E_0(1) = 0.023E_H$, $E_0(2) = 0.30E_H$, $E_0(3) = 0.97E_H$, $E_0(4) = 2.15E_H$. Dashed, dotted and solid lines correspond to $t^L/t^R = 1$, 2 and 0.5, respectively. The current is plotted in units of the total transmission rate $\tilde{t} = t^L t^R / (t^L + t^R)$.

FIG. 2. The most prominent feature in Fig. 1 for $t^L > t^R$ is magnified and the populations of the most relevant dot states $a : n = 2$, $S = 0$, $b : n = 2$, $S = 1$, $c : n = 3$, $S = 1/2$ (groundstate), $d : n = 3$, $S = 1/2$ (first excited state), $e : n = 3$, $S = 3/2$ versus bias Voltage $V$ are shown. The prominent feature of decreasing current (dotted, in units of $e\tilde{t}$) is accompanied by an increase of the population of the spin polarized $n = 3$, $S = 3/2$ state at the expense of the other populations. The populations shown here do not sum up to unity because of the occupation of other states.

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